NUREG/CP-0086
CONF-860820
Volume 1

PROCEEDINGS of the 19th DOE/NRC NUCLEAR AIR CLEANING CONFERENCE

Held in Seattle, Washington
18–21 August 1986

Sponsors: U.S. Department of Energy
Office of Assistant Secretary for
Environment, Safety, and Health
U.S. Nuclear Regulatory Commission
Office of Nuclear Reactor Regulation
Office of Nuclear Regulatory Research
The Harvard Air Cleaning Laboratory

Editor
Melvin W. First

Published
May 1987

NOTICE

These proceedings have been authored by a contractor of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in these proceedings, or represents that its use by such third party would not infringe privately owned rights. The views expressed in these proceedings are not necessarily those of the U.S. Nuclear Regulatory Commission.

Available from

Superintendent of Documents
U.S. Government Printing Office
P.O. Box 37082
Washington D.C. 20013-7082

and

National Technical Information Service
Springfield , VA 22161

NUREG/CP-0086
CONF-860820
Volume 1

PROCEEDINGS of the 19th DOE/NRC NUCLEAR AIR CLEANING CONFERENCE

Held in Seattle, Washington
18–21 August 1986

Sponsors: U.S. Department of Energy
Office of Assistant Secretary for
Environment, Safety, and Health
U.S. Nuclear Regulatory Commission
Office of Nuclear Reactor Regulation
Office of Nuclear Regulatory Research
The Harvard Air Cleaning Laboratory

Editor
Melvin W. First

Published
May 1987

PROGRAM COMMITTEE

W. L. Anderson
C. B. Bastin
R. R. Bellamy
A. G. Croff
H. J. Ettinger
A. G. Evans
H. Gilbert
R. T. Jubin
M. J. Kabat
J. L. Kovach
R. T. Kratzke
W. H. Miller, Jr.
D. M. Moeller
K. S. Murthy
T. R. Thomas
C. A. Willis
R. E. Yoder

CONFERENCE CHAIRMAN

Melvin W. First
Harvard School of Public Health

FOREWORD

The Nineteenth DOE/NRC Nuclear Air Cleaning Conference was held in Seattle, Washington, on August 18-21, 1986, under the sponsorship of the U.S. Department of Energy (DOE), the U.S. Nuclear Regulatory Commission (NRC), and the Harvard Air Cleaning Laboratory. A total of 247 air-cleaning specialists attended, representing the United States, West Germany, Canada, Japan, England, France, Italy, Austria, Belgium, Israel, Korea, The Netherlands, Republic of China, and Sweden. Those attending were affiliated with government agencies, educational institutions, national laboratories, and numerous private sectors of the nuclear industry.

Those familiar with prior Air Cleaning Conferences will be aware that the U.S. Nuclear Regulatory Commission was, for the first time, a sponsor of an Air Cleaning Conference. Originally sponsored by the U.S. Atomic Energy Commission (through the 13th Conference), responsibility for these Conferences was allocated first, to the Energy Research and Development Administration (14th Conference) and then to the successor agency, the U.S. Department of Energy (15th to 18th Conferences), after the U.S.A.E.C. was terminated. Although the NRC was not an official sponsor of the 14th through 18th Conferences, personnel from this agency continued to participate in all phases of the Conferences, from serving on the Program Committee to recruiting speakers, chairing sessions, and providing a significant portion of the technical presentations. Therefore, the 19th Conference has special significance in that it marks the first time the two successor agencies of the U.S.A.E.C. have joined forces to sponsor a Nuclear Air Cleaning Conference. In celebration of this event, the official title of the Conference has been returned to its origins and is again called the Air Cleaning Conference. Nevertheless, the interests of the DOE in nuclear airborne waste treatment and disposal remained undiminished, and the 19th Conference continues to feature discussions of this topic.

The conference proceedings show that over the past few years air cleaning research has been directed primarily to the analysis and critique of existing systems, with operational assessment and the potential impact of new source terms in the forefront. Significant effort has been devoted to the development and revision of U.S. standards and regulations. In European countries, researchers continue to stress air-cleaning problems associated with spent fuel reprocessing, whereas U.S. concerns are more in areas of fire detection, prevention, and elimination, and in the operation of control room habitability systems.

Meetings of this size and complexity require the dedicated service and wise counsel of many. The Chairman of the 19th DOE/NRC Nuclear Air Cleaning Conference wishes to record his appreciation of the firm support and willing assistance provided by the entire Program Committee throughout the lengthy planning process and during the Conference, itself. The committee names are displayed on page

ii. Special mention is due to Mr. Robert Kratzke, U.S. DOE, and Mr. Charles Willis, U.S. NRC, both members of the Program Committee, who were the designated representatives of their sponsoring agencies and who loyally and effectively served the interests of the Conference without neglecting in any way their responsibility to their own agencies. Administrative support for the Conference and preparation of the Proceedings was ably provided by Mrs. Joan K. Sullivan at the Harvard School of Public Health who has earned the Chairman's heartfelt and sincere thanks for a difficult job cheerfully and very well done.

Melvin W. First
Conference Chairman

TABLE OF CONTENTS

VOLUME 1

SESSION 1

OPENING OF THE CONFERENCE

MONDAY: August 18, 1986
CHAIRMAN: D.M. Moeller
Harvard School of Public Health

SESSION 2

FIRE, EXPLOSION, ACCIDENT ANALYSIS

MONDAY: August 18, 1986
CHAIRMEN: K.S. Murthy
Battelle Pacific Northwest
R.T. Kratzke
U.S. Department of Energy

SESSION 3

ADSORPTION, ADSORBERS, IODINE RETENTION

MONDAY: August 18, 1986
CHAIRMEN: J.L. Kovach
Nuclear Consulting Service
W.L. Anderson
Consultant

SESSION 4

FILTERS, FILTER TESTING

TUESDAY: August 19, 1986
CHAIRMEN: H.J. Ettinger
R.C. Scripsick
Los Alamos Scientific Lab.

SESSION 5

STANDARDS AND REGULATIONS

TUESDAY: August 19, 1986
CHAIRMAN: W.H. Miller, Jr.
Sargent & Lundy Co.

SESSION 6

KRYPTON, RADON, TRITIUM, CARBON-14 TREATMENT

TUESDAY: August 19, 1986
CO-CHAIRMEN: W. Motes
Westinghouse Idaho Nuclear
A.G. Croff
Oak Ridge National Lab.

VOLUME 2

SESSION 7

VENTILATION AND AIR CLEANING IN REACTOR OPERATIONS

TUESDAY: August 19, 1986
CHAIRMEN: R.R. Bellamy
U.S. Nuclear Regulatory Commission
A.G. Evans
E.I. DuPont de Nemours

19th DOE/NRC NUCLEAR AIR CLEANING CONFERENCE

SESSION 8

WEDNESDAY: August 20, 1986
CHAIRMEN: C.B. Bastin
U.S. Department of Energy
R.T. Jubin
Oak Ridge National Lab.

PANEL SESSION 9

ADSORBER FIRES

WEDNESDAY: August 20, 1986
MODERATOR: J.L. Kovach
Nuclear Consulting Service

PANEL MEMBERS:
C. Flis
Indiana & Michigan Electric Co.
H. Deuber
Kernforschungszentrum Karlsruhe
R.R. Bellamy
U.S. Nuclear Regulatory Commission
W. Mathewes
Kraftwerk Union
W. Holmes
American Nuclear Insurers

PANEL SESSION 10

NUCLEAR CARBON TEST PROTOCOLS

WEDNESDAY: August 20, 1986
MODERATOR: C.A. Willis
U.S. Nuclear Regulatory Commission

PANEL MEMBERS: J.D. Ludwick
Environmental Engineering & Testing
C. Scarpellino
E G & G Idaho Co.
M. Paska
American Air Filter Co.
J.R. Pearson
Nuclear Containment Systems
C. Sill
E G & G Idaho Co.

SESSION 11

WORKING LUNCHEON

WEDNESDAY: August 20, 1986
CHAIRMAN: M.W. First
Harvard School of Public Health

SESSION 12

GOVERNMENT-INDUSTRY MEETING ON FILTER, MEDIA AND MEDIA TESTING

WEDNESDAY: August 20, 1986
CHAIRMEN: M.W. First
Harvard School of Public Health
H. Gilbert
Consultant

SESSION 13

SAMPLING, MONITORING, TESTING

THURSDAY: August 21, 1986
CHAIRMEN: M.J. Kabat
Ontario Hydro
C.F. Gill
U.S. Nuclear Regulatory Commission

PANEL SESSION 14

FIELD TEST EXPERIENCES

THURSDAY: August 21, 1986
MODERATOR: W.H. Miller, Jr.
Sargent & Lundy Co.

PANEL MEMBERS: W. Gintor
Pacific Gas & Electric Co.
J.W. Jacox
Jacox Associates
W. Paschal, D.L. McQueen
Sargent & Lundy Co.

PLENARY SESSION 15

OPEN END

THURSDAY: August 21, 1986
CHAIRMAN: M.W. First,
Harvard School of Public Health

SESSION 1

OPENING OF THE CONFERENCE

MONDAY: August 18, 1986
CHAIRMAN: D.M. Moeller
Harvard School of Public Health

WELCOME AND OBJECTIVES OF THE CONFERENCE
Melvin W. First

GREETINGS:

HISTORY OF RADIATION MONITORING
Michael J. Lawrence

KEYNOTE ADDRESSES:

BUILDING CONFIDENCE IN ENVIRONMENTAL, HEALTH AND SAFETY PRORAMS
Mary L. Walker

IODINE FILTRATION - DO WE HAVE A PROBLEM?
Robert M. Bernero

WELCOME AND OBJECTIVES OF THE CONFERENCE

Melvin W. First

Harvard School of Public Health
Department of Environmental Science and Physiology
665 Huntington Avenue
Boston, MA 02115

It is my special duty and honor to open the 19th DOE/NRC Nuclear Air Cleaning Conference by welcoming each of you on behalf of the Conference sponsors (who are the Harvard Air Cleaning Laboratory of the School of Public Health, the U.S. Department of Energy, and The U.S. Nuclear Regulatory Commission) and the Program Committee of the Conference. I especially want to acknowledge the dedicated service of the Program Committee over the past two years for planning the Conference in detail, for seeking out appropriate speakers, for presenting papers of their own and arranging panel sessions, and finally, for chairing the many sessions and panels that will take place this week.

Chernobyl was surely on the mind of each of us as we gathered for this Conference. Although much of what occurred is still ill defined, and the nature of the protective arrangements, if any, that were in place to avert emissions of radioactive materials to the environment remains largely a matter of speculation, it is inevitable at a Conference such as this one, that we reflect soberly and at length on the efficacy, adequacy, and pertinency of nuclear air and gas cleaning systems to the full armamentarium of protective measures intended to avert such disasters.

Some years ago I was taken to task by a nuclear engineer for stating that air and gas cleaning systems are important because they represent the ultimate protective barrier to atmospheric emissions. The contrary explanation offered was that the containment structure is the final barrier and, therefore, that air cleaning equipment does not count for much. Still, not all operations involving nuclear materials are conducted inside containment vessels and even when they are, TMI-II has surely shown that air cleaning systems are an effective barrier to emissions under unusual circumstances. Current interest in vented containment concepts in a number of countries are scarcely thinkable without the use of ultrahigh efficiency air and gas cleaning systems.

It will come as no surprise to this audience that public fears of nuclear energy have been enormously stimulated by the news from Chernobyl and relentlessly reinforced by the professional anti-nuclear establishment. They are expressing concerns about the reliability of existing containment structures of U.S. nuclear power plants. In my own home state, the Pilgrim Nuclear Plant has recently announced a multi-million dollar program to strengthen their structure to help allay the publicly-expressed fears of nearby community groups. The Governor of my state has found it convenient to respond to identical public fears to abet those opposing the development of an evacuation plan as a stratagem to halt the startup of the Seabrook Nuclear Plant. I am sure most of you can cite similar events in your own community.

One of the effective remedies to counter fears of failed containment is rigorous application of air and gas cleaning technology that is installed for the purpose of rapidly returning the internal environment of the containment structure, in the remote event of an accident, to a safe pressure and to a greatly reduced amount of airborne radionuclides capable of affecting human health should they be released.

The Program Committee has structured the topics that will be addressed at this Conference and has selected the papers that are most pertinent to 1) describing innovative improvements in nuclear air and gas cleaning systems and components and 2) explaining how greater on-line reliability and effectiveness of existing systems may be obtained. The program is liberally sprinkled with panel and open end sessions that permit free exchange of practical operating experiences among all those in attendance.

It is an excellent program and a timely one. On behalf of the entire Program Committee, our DOE and NRC sponsors, and myself, I extend a sincere welcome to all, and especially to those of you who have come from other countries to enrich our knowledge and to study, with us, ways to make nuclear activities - not safer, but safe.

HISTORY OF RADIATION MONITORING

by

MICHAEL J. LAWRENCE

Manager
U.S. Department of Energy
Richland Operations Office

Hanford has a long and well-publicized history of involvement with air emissions control. As the nation's first plutonium production facility, we were pioneers in environmental engineering. Because we were dealing with a new and unknown capability in those early years, air cleaning technology, as we know it today, did not exist--nor did the knowledge that must precede it.

When the Hanford Site was acquired in 1943, the sparsely populated desert area was covered with sagebrush, inhabited by deer and blessed with pristine air and water. From the very beginning, there has been a real concern for maintaining the clean air and water of that natural environment. This is emphasized by the ongoing development of engineering techniques to minimize production hazards.

Before the first two nuclear production reactors were completed at Hanford, both onsite and offsite air sampling stations were installed. Sampling of air within the facilities and for stack effluents began shortly after initial start-up of Hanford's reactors.

The tolerable levels of pollutants in air and water were first calculated in early 1944. The radiation standard from 1943 through 1949 war 0.1 R per day to the total body. No distinction was made between workers and the public, and the dose limit was based on the exposure that would not produce an observable effect. Hanford complied with these standards and began monitoring worker radiation exposure at the time of production startup.

Routine sampling and analysis of environmental substances--including air, water, and soil--were initiated in 1945, after the war ended. Prior to that time, operations documents reflect an urgent emphasis on plutonium production for the war effort. During the rush to produce the needed plutonium, extremely large quantities of Iodine-131 were released to the atmosphere.

In 1945, there were 345,000 curies of Iodine-131 released; however in December 1949 the approximate release level was measured at 5,000 curies of Iodine-131. It is believed that the reduction was related to our increased detection capability and that part of the curies released were from the Soviet Union's emerging nuclear weapons program.

By today's standards, those early iodine release levels were very high; but the early standards were different. A tolerance level for Iodine-131 in edible plants was established by the Hanford Medical Department in early 1946. This level was set at 200 pico-curies of Iodine-131 per gram of vegetation.

By 1948, the tolerance concentration level for Iodine-131 on vegetation was lowered to 10 pico-curies per gram. And in 1950, the NCRP changed the radiation protection standards to 0.3 R per week and 3.9 R per 13 weeks to the total body. In 1954, the radiation protection standards were changed again--this time to 0.3 rem per week, or 3 rem per 13 weeks and 15 rem per year.

Between 1958 and 1963, a number of new radiation protection standards were established. Many of these standards have remained in effect since that time.

Estimates of radiation doses to members of the public were first identified by Hanford scientists in 1957. Hanford scientists were also the individuals who sat on the international boards that established radiation limits and were otherwise leaders of the nuclear movement. It is difficult to separate Hanford's history from that of nuclear development.

Hanford has played a key role in the evolution of radiaton monitoring, which will continue to be an ongoing process. And of course, as with any new technology, miscalculations were made along the way, resulting in corrective changes in monitoring procedures. Iodine releases like those that occurred in the early years would never be allowed today because we know a great deal more than we did in the beginning.

All of our early records, including those 1945 sampling analyses, were maintained in environmental reports that form a portion of Hanford's historical documents, which were released in February 1986, to the public by the Department of Energy. Those historical documents are part of our "report card" at DOE. That report card shows how our operations affect both people and the environment.

If you look at the records, you will see a dramatic decrease in radiation releases over the years. In 1957, the maximum dose received by an individual from all sources in the Hanford area, including fallout doses, ranged between 10 to 20 mrem per year. In 1963, the dose peaked at 110 mrem, due to significantly increased international nuclear testing and world-wide fallout.

By 1973, the maximum dose had dropped markedly to 2 mrem. Since 1973, the maximum dose has been in the range of a few mrem per year from Hanford operations.

When you compare these totals with coal- and oil-fired plant operations, you will see that the release rates for those fossil fuel plants is ten times higher than Hanford's. The annual does rate to the average population from coal- and oil-fueled plants ranged from 0.25 to 4 mrem per year, according to the Bier III Report. And those figures represent only air readings, while Hanford's 0.03 mrem per year dose rate for the average individual includes all pathways.

I am proud of the role Hanford has played in the evolution of nuclear energy. Of course, I wish we had the knowledge we have today back in our embryonic era. That knowledge would have prevented mistakes that were made due to a lack of necessary technology.

However, looking back at the limits we have used over the years, we can compare them to the speed limit. In the 1940's, the speed limit was anywhere from 70 mph to unlimited speed. If we were to compare operating standards of that time with the national speed limit, Hanford was operating at 69 mph. Today, the speed limit is set at a much safer limit of 55 mph; however, Hanford now operates at approximately 1 mph, far more efficiently than standards require. We are that committed to comply with environmental regualtions and operate in a manner that keeps releases as low as reasonably achievable.

What were the effects of those early years? Based upon workers, people who both work and live in the area, we see no observable health effects. However, we do not stop here. The DOE is funding a review by both the States of Washington and Oregon inconjunction with the Center for Disease Control to analyze the significance of our vast amount of present data to determine if any further health studies could be of significant benefits.

Hanford has been among the most visible entities in the environmental spotlight, and will probably remain there for some time. And while this hasn't always been easy from a public image standpoint, it has pushed us to improve, to innovate and to ensure that the public and the environment are protected at all costs. Our dedication and commitment to a clean environment has never changed . . . fortunately, our technology has.

BUILDING CONFIDENCE IN ENVIRONMENT, HEALTH AND SAFETY PROGRAMS

Mary L. Walker
Assistant Secretary
Environment, Safety and Health
U.S. Department of Energy
Washington, D.C.

I am very honored for the invitation to speak at this conference. Nuclear Air Cleaning has always been extremely important to the Department of Energy and its predecessor agencies, the Energy Research and Development Administration, and the Atomic Energy Commission. Our sponsorship of these 19 conferences, beginning in 1951, demonstrates our active interest in this subject and the protection that nuclear air cleaning systems provide to the public and the environment.

As you are aware, the nuclear industry in the past several years has been criticized extensively for not having adequate nuclear safety and environmental protection programs in place. Although most of this criticism may have been unfair and based upon perception, some of the criticism was, perhaps, deserved. The 1979 incident at Three Mile Island and, on a different scale, the recent nuclear disaster at Chernobyl, have also heightened public concern about the possibility of accidents in nuclear power plants. This has left us with a severe credibility problem. According to the latest Gallup polls, seventy-three percent of Americans today say they would oppose construction of a nuclear power plant within five miles of where they live. Sixty-six percent of the public favored a cutback in nuclear power operations until stricter safety regulations could be effected. A separate CBS poll found that fifty-five percent of Americans thought an accident similar to that at Chernobyl was likely to occur here. The message is clear. The public lacks confidence in the safety of nuclear power.

Restoring confidence, whether because of real or perceived past actions, requires time and a demonstrated commitment to making safety and environmental considerations the highest priority. I would like to discuss with you today how the Department of Energy is enhancing public confidence in its nuclear safety and environmental protection programs. Two topics that I would like to discuss in relation to this theme are the Secretary of Energy's new initiatives strengthening the environment, safety and health functions within the Department of Energy and the reviews of the safety of DOE reactors we are performing out of an abundance of caution in light of the Chernobyl incident. Clearly related are the public's perception, technical reality and the importance of air cleaning systems as critical protection.

Secretary Herrington's Personal Commitment To Safety

Secretary Herrington has made a personal commitment to operate DOE facilities in an environmentally sound and safe manner. In January of this year, the Secretary issued the Department's Environmental Policy Statement which sets forth the framework for the Department to meet its environmental obligations. The policy states that "... protection of the environment and the public are responsibilities of paramount concern and importance ....", and this extends to workers and their families at each of our sites. The policy firmly commits DOE to assuring incorporation of national environmental protection goals in the formulation and implementation of DOE programs. The policy statement contains four major points:

(1) First, as a matter of public trust, the Department considers it an obligation to conduct its operations in a safe and environmentally sound manner;

(2) Secondly, it is the policy of DOE "to conduct its operations in compliance with the letter and spirit of applicable environmental statutes, regulations, and standards;"

(3) Thirdly, the Department pledges itself to exercise good environmental management by seeking to minimize risks to the environment or public health; and

(4) Fourth, DOE expects its operating contractors to share its commitment to good environmental management and to conduct their operations in an environmentally sound manner. DOE will actively oversee contractors' activities to assure compliance with this policy.

Air quality and the effect of DOE activities on air quality, both in operation and in accident mitigation, are important factors in meeting the Secretary's commitment.

The Secretary's formal policy statement followed equally significant actions he took in 1985. In September of that year, the Secretary announced a set of new initiatives to strengthen the environment, safety and health functions of the Department. These new initiatives are the result of a thorough independent review of those functions conducted at Secretary Herrington's request. As I describe these initiatives, keep in mind they were pre-Chernobyl, and pre-Challenger accidents. The key elements of the initiatives are:

(1) A new Assistant Secretary for Environment, Safety and Health and consolidation of all Departmental environment, safety, and health functions under this official, whose responsibilities are independent from and do not include any programmatic functions;

(2) Technical Safety Appraisals of DOE's major nuclear facilities to determine not only compliance with safety requirements, but also industry lessons learned, and licensed facility requirements;

(3) A baseline Environmental Survey of DOE facilities in order to catalog and prioritize all existing environmental problems and areas of concern; and

(4) A computer assisted tracking system that will allow us to monitor compliance and assist us in identifying and reducing areas of potential risk.

In addition to the environmental policy statement, the Secretary will soon release a safety policy statement. The policy statement re-enforces the responsibilities of line managers to conduct their activities in a manner that assures the safety of our workers and the public. The policy further stresses the Secretary's commitment to a strong, centralized safety over-sight organization, to develop health and safety policy, and to assure the implementation of that policy.

A major objective of this safety policy is to have program personnel at all levels in the organization become knowledgeable and sensitive to health and safety requirements and concerns. Headquarters program managers, whose responsibilities include program development, planning and implementation, are expected to integrate safety and health concerns into their basic requirements and have safety and health considerations be a part of their management responsibilities.

Thus, in order to meet our responsibility to assure safety and the protection of the environment, we have embarked on a multi-faceted plan under my direction with the solid support of all of the Department's senior management. Air cleaning plays an important, technical role in meeting this responsibility.

Centralized Responsibilities for Policy and Oversight

The first of the three elements of the initiatives was to consolidate and upgrade the environment, safety and health organization. That has been accomplished. Environment, Safety and Health Department-wide policy is now set by this new office. Also, my office provides DOE line management with more detailed information about the nature of potential safety and environmental problems, and oversees more directly the corrective action to be taken.

Air cleaning concepts have been factored into our policy development and are an important element in our supported programs. We have reviewed the Department's policies in areas such as design criteria and safety analyses. We have updated industrial ventilation design criteria in the Department's General Design Criteria Manual. We have revised the plutonium glove box design criteria, which included many filter and ventilation High Efficiency Particulate (HEPA) System design and operating considerations. (We plan to incorporate the HEPA filter test standards into the Department's directives when the standards are finalized.)

Other projects in which my office is involved include the update of two documents, the "Nuclear Air Cleaning Handbook" and the "Nonreactor Nuclear Facilities: Standards and Criteria Guide." Both contain important confinement and ventilation design criteria. We are also supporting analytical and experimental work examining the effects of explosions on ventilation systems. And finally, there is our sponsorship of conferences such as this one.

Technical Safety Appraisals

The second component of the Secretary's initiatives was to conduct thorough Technical Safety Appraisals of DOE's high and moderate hazard nuclear facilities by teams of experts led and directed by my senior technical staff. In the course of these safety reviews, we will review more than 50 nuclear facilities in over 11 states. The onsite activity for each appraisal lasts about 2 weeks, preceded by weeks of preparatory work. These comprehensive reviews are in addition to, and are significantly different from our other ongoing safety appraisals. First, the Technical Safety Appraisals are detailed, multi-disciplined reviews of a facility's design and operations. Secondly, the Technical Safety Appraisals will not only determine compliance with DOE requirements and policy; but will also review implementation of industry requirements, standards and guides, industry lessons learned, and good engineering practices. In this manner, these appraisals strive to reach a new standard of safety excellence at our facilities. Lastly, the appraisal teams consist of technical experts from DOE, private industry, universities, and private consultants.

The appraisals began at the Fast Flux Test Facility in February of this year. To date, six appraisals have been conducted. The most publicized has been the Technical Safety Appraisal conducted of the N Reactor in Hanford, Washington, because of the similarity to the Chernobyl reactor in the use of a graphite core. For the Technical Safety Appraisal of the N Reactor, a total of 105 PERFORMANCE OBJECTIVES were addressed in 14 subject areas. (These included examining the ventilation system's ability to prevent the spread of radioactivity into clean areas and checking if the gaseous effluents from the plant were being properly monitored.)

The Environmental Survey of DOE Facilities

Nuclear safety appraisals alone are not sufficient to rebuild public confidence. The public wants to know how we are going to fix our environmental problems and be assured that we have addressed environmental risks. This led to the third element of the Secretary's plan -- the Environmental Survey. We have just begun the baseline Survey of six DOE operating facilities and programs. In the area of airborne emissions, DOE's diverse facilities contain about 1800 air emission stacks, not all, of course, related to nuclear activities. But this gives you an idea of the scope of our task!

The Survey will cover all media: air, water, soil, and all areas of environmental regulation. The purpose is to identify problems and areas of risk, and to prioritize Department-wide, in a single effort, necessary corrective actions. The Survey will be a management tool that will enable effective and wise long-range planning to correct problems, to ensure compliance and to reduce identified areas of risk.

Public Confidence - Chernobyl vs. N Reactor

As I stated earlier, the Soviet Union's nuclear accident at Chernobyl has heightened public concern about the possibility of accidents in nuclear power plants. We are still trying to determine the causes to understand the accident at Chernobyl. As with other events, nuclear and non-nuclear, we will be determining whether any new phenomena occurred that may have been overlooked in prior reviews. Our preliminary analysis shows that U.S. reactors are better designed to prevent a Chernobyl-like event. One of these key design differences is in the systems used to prevent uncontrolled release of fission products. A case in point is the N Reactor at Hanford, Washington.

At Secretary Herrington's request, before the fire was even out at Chernobyl, a series of safety reviews were ordered to be conducted by my office of the N Reactor at the Hanford site. The first, a "Special Safety Review," looked specifically at the fire protection safety of the N Reactor's graphite moderator and the safety of the reactor's confinement system. The second was the "Technical Safety Appraisal" which closely examined all operating aspects of the reactor with regard to safety. (The schedule for this review, already planned, was accelerated seven months.) The third review, recently completed, was a "Design Review" which considered the safety of all design features, the safety analysis on which the operation of the reactor is based. Based on the limited knowledge we have of the Chernobyl incident and the reviews we have conducted at N Reactor, we believe that a Chernobyl-type accident could not occur at N Reactor.

A comparison between the N Reactor and the Chernobyl Reactor shows that the N Reactor confinement completely envelopes the entire reactor primary coolant system and reactor core. This is in direct contrast to the relatively small-volume steam suppression confinement system used in the Chernobyl RBMK design, which surrounds only the part of the coolant system below the top core region.

Our preliminary analysis indicates that at Chernobyl the release path of the radioactive debris was through the top core region, completely by-passing the Soviet-designed confinement system which only partially encloses the core. In contrast, N Reactor's confinement system completely surrounds the core, and the air cleaning system provides the public and the environment protection from a significant radiological release should a severe reactor accident occur.

These are the facts. Yet public confidence is lacking. One of my duties at the Department of Energy is the task of enhancing our credibility with the American public and to make the facts known. Your work plays a vital role in this since the effectiveness and reliability of nuclear air cleaning systems is a strategic element in the assurance of safety and environmental protection. Public confidence in these systems is important. The N Reactor case demonstrates the importance of your role.

Closing Remarks

In closing, I want to leave you with a couple of thoughts. First, to improve public confidence in the safety of nuclear power, the Department of Energy is actively enhancing its programs in the areas of nuclear safety and environmental protection. President Reagan and Secretary Herrington have made this a high priority at the Department of Energy. This has been mirrored, too, by private industry's actions that we fully support. Finally, your work, your exchanging of ideas on the international level, and your active participation in conferences such as this, help strengthen the safety and environmental programs that assure a continued good, sound operation and will, ultimately, lead to a strengthening of the public's confidence in nuclear power.

I appreciate and commend your personal and corporate commitment to assuring the integrity of our systems and to improving the nuclear industry worldwide. As bold, new and exciting technologies carry us into the future, we must move ahead, but always maintain our first commitment to the environment, safety and health of our programs and, most importantly, to the public.

DISCUSSION

BELLAMY: Ms. Walker, I tried very hard to get an upbeat, positive philosophy from your talk, but I wonder how much optimism you really have in light of the expected February mandated cuts in our budget due to the Gramm-Rudman-Hollings bill?

WALKER: I have to say that I am optimistic. I think that what the Department of Energy has done was wanted by the nuclear industry. Our focus on a standard of excellence goes well for us. While we are in a time when the American public does not necessarily feel the need for energy sources other than petroleum, I think the nuclear industry will survive. I think we can be upbeat about what we have to say about it in America because other countries feel the same way we do about safety and environmental concerns. Budgets are budgets. They force us to be more constrained about how we spend our dollars. I have to fight for my own budget up on the Hill and I share with you all the concerns you have about that. Both Mike Lawrence and I would like to spend more dollars than we currently have available for oversight, safety, and for correcting our problems. It may mean that it will take us longer to accomplish our objectives and as a result, it may takes us longer to restore public confidence. But, I am confident that ultimately we will.

HULL: Those of us who are here would, by and large, feel that these were responsible and satisfactory efforts. I am speaking more from what I read in the press. I wonder what you feel are the odds that the public will be satisfied short of somebody else, such as the NRC, looking over DOE's shoulder in applying an NRC-type approach to the safety and environmental problems DOE has?

WALKER: I will tell you first of all that self regulation is a big issue for me and for the agency. Every time I testify, which is often these days, I am hit with self regulation and whether or not it is meaningful; whether or not somebody else shouldn't be looking over our shoulders. The real accurate technical answer, and anyone from NRC may wish to debate me on this, is that NRC itself admits they wouldn't now be ready to oversee us. They don't have the expertise, it is at DOE. They would probably end up hiring one-half of my staff if they had to do it. Therefore, I don't know what purpose it would serve except, perhaps, as you suggest, the appearance of having it occur somewhere other than within the Department. However, I am not even sure that that is a public concern as much as it is a political concern. In terms of some of the rhetoric coming out of Congress, it is more of a social agenda than a concern for safety. These people are really opposed to the nuclear power option. They will use any mechanism that they can find to slow it down, impede it, or whatever. I really think the Department of Energy has a credibility problem born of its past actions. That may be true for the nuclear industry as a whole. It is certainly true of corporate America in the environmental area. We have a problem that we, in part, caused and we have to fix it. Assuming that we can do that in a timely enough manner, I believe that we can demonstrate our commitment in such a way that we won't end up with other oversight, which I think wouldn't necessarily be productive, wouldn't necessarily be very effective, and certainly would take a long time to put into place. In short, I really don't think it would be a good idea, but at the same time, we have a problem we have to overcome if we are going to avoid it.

HULL: I just want to say for the record, as a DOE person, we do go out and supply some of our expertise to NRC.

BASTIN: Shortly after the accident at Three Mile Island, Hugh Sidey (Time Magazine Washington Contributing Editor and Agronsky & Company panelist) made the comment at a seminar at the National Academy of Sciences that the news media don't really understand nuclear power and nuclear systems. No matter what we do to improve our safety, the public will see the 6:30 news, five minutes of irresponsible reporting which distorts everything we do and everything we say, and makes our systems appear unsafe. I watched you on TV a few nights ago, and you had good comments on nuclear safety. Then there followed five minutes of how terrible everything is, how radon is released all over the place and gets into the atmosphere. People are concerned. Is there anything being done, are there any programs to provide a bit of education for the news media? Is there any dialogue with the management of the news media?

WALKER: Let me attempt to answer that and then give Mike Lawrence a chance to comment because he is certainly out there on the firing line. You raised a very important point and it has been recognized both by our friends on the Hill as well as internally within DOE that no matter how good a job we are doing, no matter what we are doing in moving forward to the future to seek a standard of excellence, if we can't communicate that effectively it is lost on the public. You are right. The press and the stories that are on the news, whether they understand it or not, certainly are not designed to convey the other side of the story. I can't always tell whether that is by intent or by accident. I think in some cases we need to be more available to educate them on what is happening. In the case of the one news story you were alluding to, that was not a failure of ours to educate them, it was really a decision on their part to proceed in a certain manner to get an effect that they wanted. We are out there; we are available. I indicated that our reports are public. We usually have press releases on them. What I have seen in the private industry sector is an attempt to educate people through the newspapers regarding the safety systems that are already in place. I think it is hard for an industry that is very technical, and has operated for a lot of years with the public fairly content to allow them to be technical off on their own, to realize that all of a sudden we are in the public arena, and if we aren't, we should be. Maybe it is more evident in Europe than it is in America. Maybe we are more behind, I don't know, but certainly we have an educational task ahead of us and we are always looking for opportunities to do that in a meaningful way. You don't want to overwhelm people with so much technical information that they become lost, but at the same time you have to explain the safety systems, or at least explain that they are there, so they will have confidence in them.

<u>LAWRENCE:</u> I would just add that I think we would have to be both constant and candid. It is amazing, sometimes, to see how much they get right considering that they have so little knowledge coming into an issue. They are told a lot of things in a fifteen minute interview or press conference after something goes wrong and then they must report on it. Consequently, I think things like frequent briefings, press conferences, tours to try to educate are important over time. I think when you blow it you have to say you blew it and you are going to do better next time. You have to be candid that way. But, I think if we are going to rely on the public to get all of their information through television, everything they know about a certain topic through a one minute news blip, we are doomed to failure. It has got to come to them over time, it has got to be an educational process so that the public gets the basic information and develops a basic trust in what is going on. That can only happen, as Mary says, over a long period of time through our actions. That is the best hope we have. I am confident it can happen because I am confident that is the type of leadership people we have involved in the industry and in government right now. They recognize that outreach and candor are important to gaining credibility and, given time, that will work.

IODINE FILTRATION - DO WE HAVE A PROBLEM?

by

Robert M. Bernero*

Abstract

Filtration of radionuclides at nuclear power plants is important for normal operation as well as in the event of an accident. For normal operation, data have been collected which indicate that iodine filtration reduces offsite exposures significantly. For accidents, both during and following the event, the effectiveness of filtration depends on whether the filters are bypassed or overwhelmed.

First, I will summarize our present knowledge and use of accident fission product characteristics in Part I. Then, in Part II, I will review our understanding of iodine releases for normal operation. In Part III, I will discuss the filtration of iodine, including an overview of our understanding of the current technology. Finally, in Part IV, I will identify the kind of fission products we might expect for severe accidents, whether present capabilities may be adequate, and what could be done to improve existing capabilities.

I. Accident Source Terms

Accident source terms have been a consideration in the regulation of reactors since the 1950's. In 1962, 10 CFR Part 100 was published, referencing Technical Information Document TID 14844, "Calculation of Distance Factors for Power and Test Reactor Sites." In this document, a loss-of-coolant accident (LOCA) was assumed to release 100% of the noble gases, 50% of the halogens, and 1% of the solids in the core (equal to about 15% of the gross fission product activity). It was assumed that the event involved a substantial meltdown, and that the release was inside an intact containment. Some 50% of the iodines released inside the containment were assumed to be available for release to the environment. Regulatory Guides 1.3, "Assumptions Used for Evaluating The Potential Radiological Consequences of a Loss of Coolant Accident for Boiling Water Reactors," and 1.4, "Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Pressurized Water Reactors," assumed 91% of the iodine inside the containment to be elemental (I_2), 5% to be particulate, and 4% to be organic iodides. In reference to the particulates, the usual practice has been to assume that they are not released from the containment. The above assumed releases characterize the containment source terms that have been used in calculating the effectiveness of engineered safety features (e.g., containments and filters) as well as site suitability. Such calculations are used to perform design basis evaluations.

---

*Director, Division of Boiling Water Reactor Licensing, Office of Nuclear Reactor Regulation, U.S. Nuclear Regulatory Commission, Washington, D.C., 20555

In October 1975, the Reactor Safety Study (WASH-1400) was published. This study indicated that the bulk of public risk, although low, could be attributed to accidents more severe than design basis events. A major part of that study was the assessment of fission product releases for typical PWRs and BWRs.

After the TMI accident of March 1979, several significant actions related to accident source terms were undertaken. Among these were efforts to reassess accident source terms as well as the existing design and operation of filter systems. The source term effort has resulted in a number of significant findings which are based upon detailed models, taking into account the physical and chemical processes associated with the release of fission products and their attenuation through the reactor vessel, coolant systems, containment and exterior compartments. Calculations using these models have led to source term estimates differing significantly from those in WASH-1400. Furthermore, because of differences in plant designs and accident sequences, the estimates themselves vary significantly. The analytical models have been supplemented and partially verified by a number of small and large-scale experiments. The verification efforts are continuing. As part of the same work, considerable attention has been given to the chemical forms of the various nuclides. For iodine, CsI, HOI and I_2 compounds have been identified, with CsI in aerosol form potentially dominating. These studies indicate significant uncertainty in the chemical forms, and point to potentially important differences between boiling water reactors (BWRs) and pressurized water reactors (PWRs). The differences are attributable to the dissimilar uses of boron at BWRs and PWRs.

The WASH-1400 report, as well as some more recent studies, have not credited the plant operators with the ability to arrest core melt, containment failure, or bypass sequences. Also, potential safety improvements have not been considered. In both cases, significant changes in source term estimates could result.

Table 1 illustrates the range of historical source term estimates that have been made for BWRs.

II. Results of Monitoring Plant Releases In Normal Operation

Now, let me review our understanding of iodine releases and filtration for normal operation.

During the past ten years or so, the Idaho National Engineering Laboratory has conducted an in-plant source term measurement program for the NRC. This program involved the assessment of normal plant releases of radioactive materials at five PWRs and one BWR. The primary objective of the program was to provide operational data that could be used for licensing reviews of liquid and gaseous radioactive waste management systems. EPRI has conducted a similar program at three other BWRs.

As expected, it was observed that airborne iodine releases during normal plant operation, including anticipated plant operational occurrences, are directly related to the reactor coolant iodine concentration. In terms of annual release rates, data for 1983 indicate that a total of approximately 10 curies of iodine were released from 66 operating LWRs

TABLE 1

FRACTION OF FULL POWER CORE RELEASED

SOURCE OF ESTIMATE	Xe-Kr 1	organic I 2	FISSION I 3	PRODUCT Cs Rb 4	GROUP Te Sb Se 5	Ba Sr 6	Sr 6.1	Ru,Rh,Co, Mo,Tc 7	La,Y,Zr, Nb,Ce,Pr, Np,Pu,Am, Cm 8	La,Zr,Nd, Eu,Nb,Pm, Pr,Sm,Y 8.1	Ce,Pu,Np 8.2	Ba 8.3	ESTIMATE CIRCA	ACCIDENT TYPE
TID-14844 FOR RE-LEASE FM CONTAIN	1.0	—)))	.25	(.01 OF SOLIDS ————)))									3/23/62	LARGE BREAK LOCA
			IODINES & NOBLE GASES ONLY RELEASED FROM CONTAINMENT @ .001 OF VOLUME PER DAY											
REG GUIDES			ELEMENTAL+ PARTICULAT										6/74	LARGE BREAK LOCA
1.3 & 1.4	1.0	.01	.24	(.01 OF SOLIDS ————)))SOLIDS NOT RELEASED										
			IODINES & NOBLE GASES ONLY RELEASED FROM CONTAINMENT AT TECH SPEC LEAK RATE											
WASH 1400 (PEACH BOTTOM)													10/75	
BWR1	1.0	.007	.4	.4	.7	.05	(b)	.5	.005	(b)	(b)	b		STEAM EXPLOSION
BWR2	1.0	.007	.9	.5	.3	.1	(b)	.03	.004	(b)	(b)	b		LOSS OF DECAY HEAT REMOVAL
BWR3	1.0	.007	.1	.1	.3	.01	(b)	.02	.004	(b)	(b)	b		ATWS
BWR4	.6	.007	.0008	.005	.004	.0006	(b)	.0006	.0001	(b)	(b)	b		CORE MELT WITH ONLY CONTAINMENT LEAKAGE
BWR5	.0005	.00000002	6 E-11(c)	.000000004	8 E-12(c)	8 E-14(c)	(b)	0	0	(b)	(b)	b		DBA/LOCA
NUREG0956 DRAFT FOR PEACH BOTTOM													7/85	
AE	1.0	(a)	.32	.31	.67	.5	(b)	.005	.021	(b)	(b)	b		LOCA WITH OVERPRESSURE FAILURE-BWR2
TW	1.0	(a)	.049	.052	.2	.19	(b)	.0018	.0053	(b)	(b)	b		LOSS OF DECAY HEAT REMOVAL-BWR2
TC)'	1.0	(a)	.16	.14	.40	.39	(b)	.0019	.010	(b)	(b)	b		ATWS-BWR3
BCL FOR GRAND GULF													2/26/86	
TB1	1.0	(a)	.00078	.00024	5.4 E-7(c)	(b)	.0034	.0033	(b)	.00018	.00031	.0023		STATION BLACKOUT W/LATE CONTAINMENT FAILURE
TB2	1.0	(a)	.016	.013	.11	(b)	.30	3.7 E-7(c)	(b)	.021	.034	.10		STATION BLACKOUT W/EARLY CONTAIN. FAILURE FROM IGNITER FAILU
TC2	1.0	(a)	.0032	.0036	.0025	(b)	.0015	5.2 E-9(c)	(b)	8.6 E-5(c)	.00014	.001		ATWS
BCL FOR PEACH BOTTOM													5/13/86	
TC1	1.0	a	.031	.033	.26	b	.49	.00000035	b	.012	.022	.39		ATWS WITH DEPRESSURIZATION/REPRESSURIZATION FAILURE
TC2	1.0	a	.013	.014	.007	b	.3	.000002	b	.0072	.015	.19		ATWS W/O DEPRESSURIZATION
TC3	1.0	a	.00062	.0096	.0045	b	.0072	.00000041	b	.00032	.00049	.0042		ATWS WITH VENTING THRU WETWELL @ 110 % OF DESIGN PRESSURE
TB1	1.0	a	.012	.014	.22	b	.37	.0000004	b	.031	.048	.28		STATION BLACKOUT WITH LATE CONTAINMENT FAILURE
TB2	1.0	a	.036	.041	.23	b	.46	.0000007	b	.035	.055	.34		STATION BLACKOUT WITH EARLY CONTAINMENT FAILURE
V	1.0	a	.46	.44	.26	b	.24	.0000014	b	.013	.023	.2		CONTAINMENT BYPASS THRU LOW PRESSURE ECCS PIPING

NOTES:

a. organic I combined with I in group 3

b. fission product elements regrouped in 1986

c. 6 E-11= 6 X 10-11 = .00000000006

(or 0.15 curies of iodine per reactor). The average annual iodine release per reactor has been somewhat constant for the past 15 years.

For BWRs, most of the iodine is released through the Turbine Building Ventilation System. Within the Turbine Building, more than 85% of the iodine is released from the ventilation system serving the main condenser area (steam line, feedwater heaters, reheaters, moisture separators, etc.) during normal power operation. The remaining iodine releases came from miscellaneous other areas, such as the steam jet air ejector room, the turbine operating floor, the feedwater pump room, and the mechanical vacuum pump room.

For PWRs, negligible amounts of iodine are released from the Turbine Building Ventilation System. On the average, approximately equal amounts of iodine are released from the Auxiliary Building, the Containment Purge and the Radwaste Building Ventilation System, although the relative contributions tend to vary widely depending on the specific plant and its conditions.

Under normal reactor operating conditions, the forms of radioiodine observed in plant atmospheres and plant gaseous effluents are: (1) particulate, (2) the elemental (I_2), (3) possibly hypoiodous acid (HOI) as a vapor or gas, and (4) organic (usually assumed to be CH_3I).

As shown in Table 2, the predominant iodine chemical forms appearing in BWR releases are elemental and HOI. The exception is in the case of releases from the Radwaste Building, where the principal form is organic iodine.

TABLE 2
BWR Releases In Normal Operation

Iodine Form	Fraction of Total Iodine Species			
	Containment	Auxiliary	Turbine	Radwaste
Particulate	0.11	0.2	0.2	0.002
Elemental	0.32	0.48	0.50	0.28
HOI	0.38	0.24	0.22	0.25
Organic	0.19	0.09	0.08	0.47
	1.00	1.00	1.00	1.00

For PWRs, the predominant iodine species released from the Containment and Auxiliary Buildings is organic, while elemental and HOI are the principal forms released from the Turbine and Fuel Handling Buildings respectively.

TABLE 3
PWR Releases In Normal Operation

Iodine Form	Fraction of Total Iodine Species			
	Containment	Auxiliary	Turbine	Fuel Handling
Particulate	0.09	0.04	*	0.01
Elemental	0.21	0.21	0.78	0.17
HOI	0.21	0.22	*	0.57
Organic	0.49	0.53	*	0.25
	1.00	1.00	*	1.00

*No data on breakdown of other species.

We observed that with time the chemical specie of airborne iodine changes from the reactive forms (elemental and HOI) to the relatively non-reactive organic form. We believe that this phenomenon is due to the reactive forms of iodine depositing on surfaces (walls, charcoal filters, etc.), reacting with organics, and then resuspending in the air.

For example, at Turkey Point and Rancho Seco, we observed that the iodine species distribution changes with time after the Containment Building has been isolated. The initial amount of reactive iodine decreased with time and after about six days after containment isolation, organic iodine exceeded 90%.

III. Filtration of Iodine

Now, let me turn to an overview of our understanding of the current iodine filtration technology and the associated difficulties that we face in reactor licensing.

Charcoal filters have been found to be effective in the removal of iodines in both particulate and gaseous forms from the building ventilation systems of nuclear power plants. Observed decontamination factors for various species of iodine are shown in Table 4.

TABLE 4

	Iodine Decontamination Factors	
Iodine Form	Average	Maximum
Particulate	10	600*
Elemental	200	300
HOI	200	1600
Organic	50	100

*included roughing and HEPA filters

The maximum decontamination factors occur for new charcoal, or when the influent iodine concentration increases significantly. This is relatively important during and following an accident, when the iodine concentration can increase significantly and the more reactive iodine species are expected to predominate. Under these circumstances, the efficiency of a charcoal filter for iodine removal is expected to be higher. Hence, charcoal filters are more important under accident conditions than under normal conditions.

It appears that high efficiency and long service life of charcoal filters is critically dependent on the process of charcoal impregnation. For example, commercially available activated charcoal with triethylene diamine (TEDA) and potassium iodide (KI) exhibits less penetration by methyl iodide than charcoal which is impregnated only with potassium iodide. Other important observed parameters are the degree of atmosphere contaminants (ozones, sulfur dioxide, nitric oxide etc.) and water vapor in the outdoor makeup air.

Now, I have two problem areas in charcoal filter operation, namely charcoal testing and iodine sampling. First, let us discuss charcoal testing.

The activated carbon in a charcoal adsorber bed degrades slowly due to atmospheric contaminants and moisture. Therefore, nuclear power plant technical specifications require a periodic laboratory analysis of a representative carbon sample. This permits periodic verification of the capability of charcoal filter systems to remove iodines.

In late 1982, the Committee on Nuclear Air and Gas Treatment (CONAGT) conducted a round/robin interlaboratory comparison test. The test involved measuring methyl iodide penetration of new and used nuclear grade charcoal. Some 15 laboratories participated in this effort. Seven of the participating laboratories were from the U.S., and one each were from West Germany, Italy, Japan, Canada, U.K., Finland, Netherlands, and France. The test results were disappointing. New charcoal exhibited penetration in the range from less than 0.01% to 9.3%. For used charcoal, however, the test results varied between 0.37% and 84% penetration.

Again, in late 1984, the second set of round/robin interlaboratory comparison tests were conducted. This time, some eight laboratories in this country, two laboratories from Canada, and one each from U.K., West Germany, and Korea participated. The test results were equally disappointing.

The new charcoal test results varied between 0.23% and 8.4% penetration, whereas used charcoal tests yielded penetrations between 17.4% and 60%. Nuclear power plant standard technical specifications typically require no greater than 1% penetration of methyl iodine for a 2-inch thick charcoal bed. The specifications also typically call for a penetration test to be performed at least once every 18 months.

The results of the round/robin tests clearly challenge the reasonableness of the existing charcoal testing requirements for all U.S. reactors.

Some participating laboratories recently received charcoal samples with specific test procedures provided by EG&G for the third round/robin interlaboratory comparison tests. These tests can have a significant impact on the technical specification requirements. Hence, I urge full cooperation with the test efforts, particularly with the test procedures specified by EG&G. Specifically, we are considering a relaxation of the acceptance criteria for methyl iodine penetration requirements for all operating reactors as long as they meet the guidelines of Appendix I to 10 CFR Part 50 for normal iodine releases, as well as for iodine releases during and following a DBA. However, if the results of this third test also fail to show reasonable agreement, then we will have to consider a laboratory accreditation program as a means of certification of qualified laboratories which can reproduce the EG&G test results.

The other problem area associated with the operation of iodine filtration systems has to do with representative iodine sampling. Specifically, I am talking about sampling systems installed to sample airborne iodines after an accident. In many of these systems, there is a potential for sample line loss of iodine. The loss can be so large that the iodine sampling process becomes meaningless, and one is left without a representative sample and analysis of the iodine content of gaseous effluents. Hence, in the event of a nuclear accident, the operator is faced with the alternative of calculating projected offsite doses to the population (which may be based on extremely conservative assumptions), or rapidly obtaining radiation measureements in the field.

The requirements of Item II.F.1, of NUREG-0737, were promulgated to assure that a plant operator would have the capability, under accident conditions, to obtain and analyze sufficiently representative iodine samples. This would permit a realistic assessment of the projected offsite doses on the basis of actual accident discharge conditions. Unfortunately, there are no current regulatory guidelines or acceptance criteria for an acceptable line-loss estimating method, or measurement program. Instead, the NRC has recommended that either the actual iodine sample delivery system, or a full-scale mockup, be tested experimentally to determine the extent of sample line-losses. At this time, we are not prepared to either recommend or endorse any specific test method as being acceptable. However, we are receptive to proposals for technically sound test procedures for determining iodine line losses for both particulates and in gaseous forms.

IV. Do We Have A Problem?

As I have indicated, fission product source terms are typically grouped by chemical and physical form. They are further characterized as those that are released in normal reactor operations, and those that can occur in accidents. Filtration can provide significant attenuation of many of them, but not all. For example, the noble gases cannot be attenuated with the types of filter systems that are presently used. However, a significant portion of the dose associated with normal effluents, as well as accidental releases, is comprised of iodine nuclides.

Hence, present nuclear plant filtration capabilities are aimed at attenuating iodine. The focus is on both particulate and gaseous forms through roughing, HEPA and charcoal beds, respectively. The HEPA designs, however, can be overwhelmed by high concentrations of particulates. Also, charcoal filter designs typically are predicted on the assumption that accidental iodine releases are predominately gaseous elemental iodine. Yet recent findings indicate that other forms of iodine can be released in significant quantities. Furthermore, the ability to predict with confidence the filtration capability of charcoal is in large measure dependent on a good testing standard. Currently, this does not exist.

Is there a match between existing filter system designs and what we know about normal and accidental fission product releases?

There is no doubt that the existing filtration systems will continue to be needed for attenuating normal releases as well as releases as well as releases from design basis accidents. However, as I noted earlier, it is increasingly apparent that the bulk of the risk is due to severe accident sequences. Hence, there may be a shift to a greater emphasis on roughing and HEPA filters to cope with potentially high concentrations of post-accident particulates of biologically more significant fission products, such as Cs, Te, and Sr.

We should not rule out charcoal. We should use it effectively in pursuing the ALARA goals, since experience shows that charcoal is effective in removing particulate and gaseous forms of iodine from plant ventilation systems. We should also continue to rely on charcoal for its potential to mitigate the consequences of accidents, where it would be effective in dealing with the more reactive iodine species. However, we must keep in mind that this potential may be limited under severe accident conditions, if the charcoal filters are overwhelmed. We must have rational test and inspection requirements. These should encompass severe accidents, where temperatures, humidity, and particulates are representative of containment venting or bypass accidents. These considerations should be reflected in our regulatory assessments.

Acknowledgements

I want to acknowledge the suggestions, comments and help of J. Hulman, K. Campe, J. Lee and L. Kriesel of my staff in the preparation of this paper.

DISCUSSION

KOVACH: While I agree with most of your comments, and appreciate the emphasis you laid on methyl iodide testing of adsorbents, I do not believe in the potential existence of HOI in the vapor phase. I feel that we should emphasize the identification of penetrating iodine forms instead of calling everything unknown HOI in vapor phase.

BERNERO: I agree there is uncertainty about HOI. It is a problem that has challenged us for years. I am not sure that it is so simple that merely confronting it will get us an answer in a short time because it changes, it is not stable.

WILHELM: Till today, nobody has verified airborne HOI. I have even heard that $10,000 has been offered by a participant here for confirmation of it's existence as an airborne compound. Previously, we never considered the possible iodine compounds which may result from reactions between organic radicals and iodine. In a core melt down accident, one will have high irradiation fields and lots of airborne organics, for example, from burning cable insulation, paint and oil, ignition by the heat of the molten core. Even during normal operations of a PWR, we found concentrations of airborne organics in the equipment compartment rooms (part of the inner containment) in the range of some mg/m^3, for example 6 mg/m^3. During refueling and repair operations, we found concentrations of up to 60 mg/m^3. In comparison, of the air in the equipment compartment of 10^{-9} to 10^{-11} Ci/m^3 of I-131 corresponds to 10^{-14} to 10^{-16} g/m^3 of I-131. Under the influenced of a high irradiation field, organic compounds will be partly converted into radicals which in turn will react with iodine. In organic chemistry, the reaction of iodine with radicals is a well known confirmation reaction for the presence of radicals. As a result of this reaction, airborne organic iodine compounds will be formed with a different sorption behavior than that of the methyl iodide. Mr. Kovach mentioned ethyl iodide, a simple organic iodine compound with two carbon atoms. Our measurements of the sorption behavior of this compound show clearly an ion exchange with the KI-impregnation of the charcoal, but a much lower reaction rate compared to methyl iodide. This results in a much lower removal efficiency. Considering the whole spectrum of all possible iodine compounds, it would be naive to assume that methyl iodide will be a conservative model substance for all cases. It is true that methyl iodide has a high vapor pressure, but the iodine atom in this compound is also extremely reactive. Over all, this results in a relatively good removal behavior. I may add that in one case during a relatively high iodine contamination of the coolant in a BWR, we have seen sometimes only airborne iodine which penetrated the iodine filter completely. Tests showed at the same time an excellent removal efficiency of the same iodine filter for methyl iodide and the absence of any leaks.

BERNERO: I agree that the use of methyl iodide is a shorthand method in that, in effect, it writes everything off as methyl iodide. I am interested in the result Mr. Wilhelm cites of 100% penetration. I hope the members of my staff are aware of it, because I am surprised to learn of 100% penetration through an acceptable tested bed. That is disconcerting.

HULL: First, a comment on long sampling line runs. If you are convinced that you get mostly penetrating, organic forms, than you have less of a problem with long sampling lines because there wouldn't be a deposition problem. Concerning the matter of sampling in the vent from the plant to establish what is going on, I am somewhat concerned with the NRC position that it isn't as important to get a representative sample from the containment itself as it is from the fraction going out the vent. I have the transcript of a conference, the synopsis of which said we just want a representative sample of the containment atmosphere. As an emergency planner, if you could demonstrate convincingly that whatever iodide had been released was in the water and not airborne, than I wouldn't be anywhere near as worried as I would be if it was in the containment and the containment pressure was building up. Why isn't it as important to get a representative sample of the airborne iodine in the containment, if any, as it is to sample what is going out the vent? The NRC position is that representative sampling is not important unless airborne iodine is used in the core damage assessment.

BERNERO: I am not sure whose conclusion it was that it is not important to get a containment sample. I think we do believe that it is important. There is a clear desire to have a large, dry containment because it is a passive device that can be deliberately vented during normal operations. Therefore, any release would be inadvertent and representative of what is in the containment, so it is quite important. Sampling is intended to provide a reasonable measure of what is in the containment atmosphere of a boiling water reactor. If there were to be a pressure buildup and then venting, the focus would be much more on what was going out the vent. It would vary dramatically with the plant design and I think vent sampling may be what was intended for that reason.

HULL: I don't know who talks to whom down at your shop, but I can say that the transcript that I got of the conference I just mentioned was that, unless you are using the information for core damage, it really isn't important to get a representative sample. We have been doing the reviews and that is what we have been told to use as guidance, i.e., that it is not important unless it is used in the core damage assessment procedure.

BERNERO: I am sorry I cannot explain it further. I am not familiar with it.

SILL: As a chemist, I am a bit horrified at all the talk about HOI being a mysterious material. HOI is a very simple hydrolysis product of elemental iodine. If you write the reaction you have: $I_2 + H_2O \rightleftarrows HI + HOI$. I don't know anything about the thermodynamics of the reaction but I will guarantee you that if you put chlorine into sodium hydroxide you will wind up with a solution of sodium hyperchloride. The same kind of a thing is going to happen if you dissolve iodine in sodium hydroxide. Now, even under the conditions of a reactor operation, all you would need to get HOI, for certain, would be to have elemental iodine in the presence of moisture. How far that reaction would go, thermodynamically, I don't know. But if you put a little bit of alkali in there, it certainly is going to kick it in the direction of more HOI, i.e., $I_2 + OH^- \rightarrow I^- + IO^-$.

GUEST: I would like to make a remark about deposition in sample lines that you talked about. We have seen a lot of speculation in the literature and a few laboratory studies. We have been quite concerned and have made measurements within the Bruce A. Nuclear Power Station on the deposition in the sample lines for elemental iodine and methyl iodide. This was a fairly major study and I will be presenting a paper later in the week on this. We found that there is no deposition of either methyl iodide or molecular iodine in the long sample lines at the Bruce A. Station. I would be very please to hear your thoughts on that paper when I present it.

BERNERO: I have staff here at this conference and we are hoping to get a good deal of information from papers such as yours.

DEITZ: In connection with the HOI topic under discussion, this species may not be present in the gas phase, but it is certainly present in the activated carbons. Large quantities of ozone in the air pass through a carbon adsorber in the months that it is in service and the ozone oxidizes the iodine ion introduced in the impregnation. A number of IO_x^- complexes are formed ranging from IO^- to IO_3^-. The reaction can be reversed by chemical reduction; we have used hydrazine. These hypoiodides are responsible for the irregularities encountered in testing used carbons for the penetration of methyl iodide-131.

VIKIS: One more statement about HOI. The reference to abundant quantities of HOI formed by the hydrolysis of iodine is, indeed, correct. There is no doubt that we do have HOI formed in solution from the hydrolysis of I_2. The problem we are facing is identifying the gaseous form of HOI. This is where the question arises, whether there is HOI in the gas phase. At the Whiteshell Nuclear Research Establishment of Atomic Energy of Canada Ltd, we searched for gaseous HOI using definitive techniques (e.g., mass spectrometry). We were unable to detect gaseous HOI above iodine solutions, although we had no problem in identifying gaseous HOCl or HOBr above the corresponding halide solutions.

CLOSING COMMENTS OF SESSION CHAIRMAN MOELLER

We heard first this morning from Michael Lawrence who reviewed the history of environmental monitoring at the Hanford Works. We see there much of the history of radiation protection, in that we began in the early years with relatively high permissible dose and release rates, and these have gradually reduced down through the years.

At the same time, however, it must be recognized that nuclear operations at Hanford were initiated under wartime conditions during which there was an urgency to accomplish their goals on a priority basis. Today, those responsible for operations at Hanford are funding appropriate agencies within the States of Washington and Oregon, as well as within the Centers for Disease Control, to re-examine these releases and to evaluate any effects that they may have caused in terms of the nearby populations or the environment. Even though there are ready explanations for many of these early releases, they have, in some instances, led to a lack of confidence on the part of the public in the safety of nuclear operations, not only in terms of DOE's facilities but also in terms of the commercial nuclear power industry.

Following the initial presentation, Mary Walker reviewed a range of programs that DOE is pursuing to help restore the confidence of the public in DOE's operations. In particular, she reviewed three initiatives currently underway. The first of these is the preparation and issuance of an environmental policy statement confirming that DOE expects operators of its facilities to go beyond the rules and regulations in seeking to reduce environmental risks. Secondly, DOE is conducting technical safety appraisals of all its facilities, using a multi-disciplinary approach. The results of these appraisals will be used as a basis for the initiation of additional improvements. Lastly, her Office is conducting environmental surveys of all DOE facilities to identify problems in this area. The results of these surveys will subsequently be used to establish priorities for addressing these problems, including the establishment of a computerized tracking system to keep up with these problems, the status of each, and the progress being made in terms of corrective actions.

Lastly, we heard this morning from Bob Bernero on nuclear power plant accident sequences and their implications relative to nuclear air cleaning. Mr. Bernero called upon us not to neglect charcoal--it is still an effective material for retarding various radioactive materials that might be released from a nuclear plant not only under normal operating conditions but also, and most particularly, under accident conditions. Mr. Bernero cautioned, however, that we must keep in mind that our testing procedures for evaluating the effectiveness of charcoal leave much to be desired; the same situation applies to our ability to collect the proper samples for preparing estimates of radionulcide releases within a nuclear power plant under accident conditions. Obviously, there is much work to be done. These and related topics and questions will be addressed in later sessions of this Conference. I know that all of you look froward to hearing the papers to be presented and in taking part in the related discussions.

Finally, let me close this session by formally thanking each of the people who appeared on the program this morning. Their presentations have set a high standard for the speakers who follow.

SESSION 2

FIRE, EXPLOSION, ACCIDENT ANALYSIS

MONDAY: August 18, 1986
CHAIRMEN: K.S. Murthy
Battelle Pacific Northwest
R.T. Kratzke
U.S. Department of Energy

INVESTIGATIONS ON THE INFLUENCE OF COMBUSTION PRODUCTS ON THE RETENTION OF RADIOIODINE BY ACTIVATED CARBONS
H. Deuber, V. Giraud

CONCLUSIONS FROM FIRE TESTS IN ACTIVATED CARBON FILLED ADSORBERS
M. Mathewes

A CURRENT EVALUATION OF FIRE LOSS CONTROL SYSTEMS FOR CHARCOAL MEDIA
W.D. Holmes

REVIEW OF FIRES AND FIRE CONTROL METHODS FOR NUCLEAR AIR CLEANING SYSTEMS
J.L. Kovach

MEANS USED TO MAKE SURE THAT THE CONFINEMENT IS MAINTAINED IN CASE OF FIRE
J. Savornin

RELEASE OF RADIOACTIVITY OUT OF WASTE PACKAGES UNDER THERMAL STRESS
D. Gruendler, H.W. Brenig, W. Wurtinger

SIMULATION OF EXPLOSIONS IN AIR CLEANING SYSTEMS AND COMPARISON OF THE RESULTS WITH COMPUTER CODE PREDICTIONS
W.S. Gregory, B.D. Nichols, R.D. Wade, P.R. Smith, D.L. Fenton

FIRE-ACCIDENT ANALYSIS CODE (FIRAC) VERIFICATION
B.D. Nichols, W.S. Gregory, D.L. Fenton, P.R. Smith

METHODS FOR AIR CLEANING SYSTEM DESIGN AND ACCIDENT ANALYSIS
W.S. Gregory, B.D. Nichols

OPENING COMMENTS OF SESSION CO-CHAIRMAN MURTHY

Air cleaning systems assume special importance in minimizing the adverse environmental impacts of unplanned events such as fires, explosions, and accidents in nuclear facilities, thereby adequately protecting the public from the effects of these events. This session on fire, explosion, and accident analysis will provide you with results of recent investigations in this important field.

The session features a total of ten speakers: three from West Germany, one from France, and six from the United States. All five papers in the first part of the session deal with the results of investigations on confinement-retention in the event of fire. Most of these papers deal with carbon-base adsorbent fires. The opening paper deals with an introductory review of fires and fire control methods in nuclear air cleaning systems, outlining experiences from carbon-base adsorbent fires that have occurred in the history of the nuclear power industry.

The last five papers take on a different flavor in that you will be listening to three papers on computer code development and testing by Los Alamos National Laboratory in cooperation with the New Mexico State University. You will also be listening to West German developments on a procedure for determining source terms from waste packages (such as cemented waste) subjected to thermal stresses from accidental fires. Finally, the session will hear from Brookhaven National Laboratory and the US NRC on the current status of Monitoring, Sampling, and Analysis of Reactor Atmosphere Effluents in Post-Accident Concentrations. All in all, we will have a technical feast, and how well we have digested it all will be summarized in the closing remarks of the Co-Chairman, R. T. Kratzke.

INVESTIGATIONS ON THE INFLUENCE OF COMBUSTION PRODUCTS ON THE RETENTION OF RADIOIODINE BY ACTIVATED CARBONS

H. Deuber, V. Giraud

Kernforschungszentrum Karlsruhe GmbH
Laboratorium für Aerosolphysik und Filtertechnik
Postfach 3640, D-7500 Karlsruhe 1
Federal Republic of Germany

Abstract

Investigations were performed on the influence of gaseous combustion products from four relevant materials used in nuclear power plants on the retention of methyl iodide (CH_3I-131) by four commercial impregnated activated carbons. It is concluded that with iodine filters of large bed depths (residence time: e.g. 0.5 s), when containing a usually employed impregnated activated carbon of good quality, a sufficiently high retention of radioiodine should be achievable even with a high challenge of gaseous combustion products from relevant materials used in nuclear power plants.

I. Introduction

Numerous investigations are described in the literature on the various factors that influence the retention of radioiodine by impregnated activated carbons used in iodine filters of nuclear power plants (NPPs). /1,2,3/ However, as for the influence of combustion products, no corresponding investigations are known /4/, although, in the case of a fire in a NPP, this influence could be of concern. To fill this gap of knowledge, we are performing corresponding investigations.

In the present paper, investigations are covered on the influence of gaseous combustion products from four relevant materials used in NPPs on the retention of methyl iodide (CH_3I-131) by four commercial impregnated activated carbons. It should be kept in mind that organic iodine species other than methyl iodide, which may be more difficult to retain /5/, may be formed in the event of a fire in a NPP, e.g. by reaction of elemental iodine (I_2) with combustion products. As for I_2, its retention should be of no concern in the present context. /6/ Fires in iodine filters are beyond the scope of our investigations.

II. Relevant Materials and Combustion Products

As in many industrial installations there are many combustible materials in NPPs. In the present context only those are of interest which are to be found in rooms that are exhausted with iodine filters. Such rooms are e.g. the containment rooms in pressurized water reactors (PWRs). The materials of interest are cables with insulations, in particular PVC or more modern FRNC materials (flame retardent, non-corrosive). Additionally, lubricants (mineral oils) and paints (epoxy resins) must be considered. /4/

The types of combustion products produced depend on the types of materials combusted and on the combustion conditions. Apart from carbon oxides, hydrocarbons and water, which are generally produced, HCl will be generated in large proportions in the combustion of PVC cables. With FRNC cables organic acids such as CH_3COOH may occur. With mineral oils, in addition to evaporated oil, SO_2 and NO_x may be generated in small proportions. In the case of epoxy resins amines and HCN may occur. These compounds may originate from ancillary compounds (e.g. hardeners).

As for the challenge of iodine filters with combustion products in the case of a fire in a NPP, it is difficult to indicate realistic general figures. The following example, relating to a modern German 1300 MWe PWR, may be useful. It may be assumed that the inventory of PVC cables in the containment rooms is 10 000 kg half of which is combustible. The largest iodine filters, featuring a total throughput of about 130 000 m^3/h at a residence time of 1 s, contain roughly 20 000 kg of activated carbon in total. Thus, if these iodine filters were challenged with the combustion products from all the PVC cables in the containment rooms, the loading would be 0.25 g of combustion products per g of activated carbon or 25 %. Obviously, other figures can be calculated with other assumptions.

III. Experimental

As representatives of the relevant materials used in NPPs four materials were employed to generate the combustion products for the investigations:

a) a PVC cable;
b) a FRNC cable;
c) a mineral oil;
d) a paint.

The materials were combusted in a standard combustion apparatus. /7/

As representatives of the impregnated activated carbons used in iodine filters of NPPs the following four carbons were employed in the investigations:

a) 207B (KI);
b) 207B (TEDA);
c) 208C (KI);
d) 208C (TEDA).

The base material of the first two carbons is coal, that of the other two carbons coconut shell. The grain size of all the carbons was 8 - 12 mesh. /8/ The impregnants, as indicated, were either KI or TEDA.

The retention of CH_3I-131 by the carbons was determined in our standard test apparatus. /3/

The procedure pursued was as follows: First the carbon bed (consisting of 20 sections) was preconditioned over night in the test apparatus. Then it was challenged in the combustion apparatus with the combustion products over a period of generally 0.1 h. The combustion was performed at 700 °C in a stream of air. In front of the carbon bed the air stream was diluted (so that the flow was equivalent to that used in the subsequent CH_3I-131 test) and passed through a glass wool filter. Generally immediately afterwards, when the test conditions were again established, the CH_3I-131 was loaded on the carbon bed in the test apparatus, also over a period of 0.1 h. In some cases, to investigate the influence of the duration of the impact of the combustion products, the CH_3I-131 was injected 20 h after the loading of the combustion products. (The "purging time" was 20 h.) More data of the tests with CH_3I-131 are contained in Table I.

As indicated in Table I, the material combusted was either 0.1, 1 or 10 g. Thus, the maximum loading was nominally about 0.1 g of combustion products per g of carbon and of the same order of magnitude as calculated in the example in Chapter II. Of course, the major fraction of combustion products in non-gaseous form was removed by retention with the glass wool (and by deposition on the wall of the tube). However, this seems to be of minor importance with respect to the retention of CH_3I-131 as was shown by comparison tests with and without glass wool.

For comparison also tests with 1 g of HCl were carried out. As mentioned earlier, this compound is generated in the combustion of PVC. Moreover, the retention of CH_3I-131 by the unchallenged (fresh) carbons was determined.

In addition to the investigations described, some measurements were also performed to determine the organic combustion products on the carbon beds. To this end the CCl_4 extracts were analyzed by mass spectrometry. Moreover, both the pH values /9/ and the Cl^- concentrations of the H_2O extracts of the carbon beds were measured after challenge with combustion products from the PVC cable and after loading with HCl.

IV. Results

In this chapter first the results are presented of the investigations with CH_3I-131 using the combustion products and impregnated activated carbons indicated in Chapter III. The test parameters are contained in Table I. Detailed results are given for the carbons 207B (KI) and 207B (TEDA) only because the results for the carbons 208C (KI) and 208C (TEDA) were not decisively different (see below). The results are generally presented in terms of penetration by CH_3I-131.

Figures 1 to 8 display the penetration of the carbons 207B (KI) and 207B (TEDA) as a function of the bed depth at different amounts of combustion products. Without challenge by combustion products (amount: 0 g) the usual linear decrease in penetration with increasing bed depth was found (semilogarithmic plot). For amounts of combustions products of 0.1 g the penetration curves were also nearly linear. Compared with the unchallenged carbons, the penetration was somewhat higher in some cases, in particular for 207B (TEDA).

For amounts of combustion products of 1 and 10 g, a nonlinear course of the penetration curves was obtained: flat at small bed depths and steep at large bed depths. In general, at large bed depths the penetration curves were nearly parallel to the penetration curve of the unchallenged carbon. This form of the penetration curves, as is well known from aging experiments /10,11/, corresponds to a decrease in impact on the carbon with an increase in bed depth, with a negligibly small impact at a large bed depth for parallel penetration curves.

Figures 1 to 8 show that at the same amount of combustion products, for large bed depths ($\geq$ 12,5 cm) the increase in penetration was similar for all the combustion products, both for 207B (KI) and 207B (TEDA). At the maximum challenge, the increase in penetration was mostly 1 to 2 orders of magnitude, with a tendency for lower values for 207B (KI) and higher values for 207B (TEDA).

As for small bed depths, an overview is given in Figures 9 and 10. These show the penetration of 207B (KI) and 207B (TEDA), respectively, at a bed depth of 5 cm for different amounts and types of combustion products. It is obvious that the combustion products from the PVC cable exerted the most adverse effect on the carbons. For 207B (KI), at the maximum challenge the penetration was between 70 and 80 % with the combustion products from the PVC cable and between about 20 to 40 % with the combustion products from the other materials. For 207B (TEDA) the corresponding values were 60 to 70 % and 30 to 40 %, respectively. (The higher value in the case of the combustion products from the oil may be due to an experimental error.)

The strong adverse effect of the combustion products from the PVC cable at small bed depths is also obvious from Tables II and III, which contain the removal efficiencies of the first five beds of 207B (KI) and 207B (TEDA), respectively, at different types and amounts of combustion products.

From all the results obtained for small and large bed depths it is evident that in general the combustion products from the PVC cable exerted a strong impact over a small bed depth, whereas the other combustion products exerted a weaker influence over a larger bed depth, with a resulting increase in penetration of similar magnitude at large bed depths.

As for the carbons 208C (KI) and 208C (TEDA), the increase in penetration due to the impact of the combustion products was similar to that observed with the other carbons. (The results were closest to those obtained with 207B (TEDA).)

As mentioned earlier, in some tests the CH_3I-131 was not injected immediately after the loading of the combustion products, but after a purging time of 20 h only. Some results in terms of penetration versus bed depth are illustrated in Figures 11 an 12. It is obvious that the duration of the impact of the combustion products from the PVC cable was of minor importance in the range investigated, both for 207B (KI) and 207B (TEDA). Analogous results were obtained for the other combustion products.

The results of the comparison tests with HCl are displayed in Figures 13 and 14, both for no purging and purging over 20 h after the injection of HCl. With no purging, at large bed depths the penetration was similar to that obtained with the same amount of the combustion products (compare Figures 1 to 8.) At small bed depths, however, the penetration was much higher. At a bed depth of 5 cm the penetration was about 90 % for 207B (KI) and about 60 % for 207B (TEDA). Similar values were obtained with much higher amounts of combustion products from the PVC cable only (compare Figures 9 and 10).

Thus, the impact of HCl was strong, but over a small bed depth only, as was in principle the case with the combustion products from the PVC cable. It can be concluded that HCl constituted a major factor in the impact of the combustion products from the PVC cable on the retention of CH_3I-131 by the carbons. This is in agreement with other measurements (see below).

The results of the mass spectrometric analyses of the CCl_4 extracts of carbon beds after challenge with combustion products are summarized in Table IV. The results relate to the combined first and second beds of 207B (KI). It is to be seen that alkyl derivatives of benzene were among the main organic combustion products from all the materials used. From aging experiments it is well known that these compounds (and other organic compounds) contribute to the deterioration of carbons with respect to the retention of CH_3I-131. /12,13,14,15,10,11/ Obviously these compounds played also an important part in the impact of the combustion products. The similarity of the compounds would account for the similarity in impact by the combustion products from various materials.

Some results of the pH value measurements in the H_2O extracts of carbon beds after challenge with the combustion products from the PVC cable and after loading with HCl are shown in Figures 15 and 16. It is to be seen that the extracts of the front beds had a lowered pH value. For the combustion products from the PVC cable the decrease of the pH value was 2 to 3 units, for HCl 6 to 7 units, both for 207B (KI) and 207B (TEDA). The stronger decrease of the pH value for HCl correlates with the stronger impact of this compound on the retention of CH_3I-131.

It is mentioned that a higher Cl^- concentration was also only found in the H_2O extracts of the front beds, both for the combustion products from the PVC cable and for HCl.

The measurements of the pH values and the Cl^- concentrations confirm that HCl constituted a major factor in the impact of the combustion products from the PVC cable on the retention of CH_3I-131 by the carbons. The adverse effect of acidic compounds in this context has been observed earlier. /13,15/

V. Summary and Conclusions

Investigations were performed on the influence of gaseous combustion products from four relevant materials used in nuclear power plants (NPPs) on the retention of methyl iodide (CH_3I-131) by four commercial impregnated activated carbons. The materials used to produce the combustion products were a PVC cable, a modern FRNC cable (flame retardent, non-corrosive), a mineral oil and a paint. The carbons differed in base material (coal or coconut shell) and/or impregnant (KI or TEDA).

It was found that at small bed depths (residence times: ≈ 0.05 s) the increase in total (cumulative) penetration by CH_3I-131 was higher for the combustion products from the PVC cable than for the other combustion products. At larger bed depths (residence times: ≈ 0.25 s) the increase in penetration was similar for all the cumbustion products and nearly independent of the bed depth. The increase in penetration was not decisively different for the different activated carbons. At larger bed depths, in general the increase in penetration was 1 to 2 orders of magnitude at the maximum challenge with combustion products (nominal loading: 0.1 g of combustion products per g of carbon). The combustion products were found to contain large proportions of alkyl derivatives of benzene and, in the case of the PCV cable, hydrochloric acid.

It is concluded that with iodine filters of large bed depths (residence time: e.g. 0.5 s), when containing a usually employed impregnated activated carbon of good quality, a sufficiently high retention of radioiodine should be achievable even with a high challenge of gaseous combustion products from relevant materials used in NPPs.

Acknowledgment

The following coworkers participated in performing and evaluating the tests:
K. Bleier, E. Becker, M. Dehm, S. Engelhardt, A. Ladanyi and S. Winkler.

VI. References

/1/ HOLLADAY, D. W.;
"A Literature Survey: Methods for the Removal of Iodine Species from Off-Gases and Liquid Waste Streams of Nuclear Power and Nuclear Fuel Reprocessing Plants, with Emphasis on Solid Sorbents".
ORNL/TM - 6350 (1979).

/2/ KOVACH, J.L.;
"The Evolution and Current State of Radio-Iodine Control".
CONF 801 038 (1981) p. 417.

/3/ WILHELM, J.G.;
"Iodine Filters in Nuclear Installations".
Commission of the European Communities, V/2110/83 (1982).

/4/ GIRAUD, V.;
"Die Abscheidung von Iod mit Iodfiltern in Kernkraftwerken bei Bränden (Literaturstudie)."
KfK 3867 (1985).

/5/ KOVACH, J.L.;
"Adsorbent Removal Efficiencies for Organic Iodine Compounds".
CEC European Conference on Gaseous Effluent Treatment in Nuclear Installations, Luxembourg, 14.-18.10.1985.

/6/ DEUBER, H.;
"Investigations on the Retention of Elemental Radioiodine by Activated Carbons at High Temperatures".
Nucl. Techn. 72 (1986) 44.

/7/ Normenausschuß Materialprüfung im DIN;
"Erzeugung thermischer Zersetzungsprodukte von Werkstoffen unter Luftzufuhr und ihre toxikologische Prüfung."
DIN 53 436 (1981).

/8/ British Standard Institution;
"Specification for Test Sieves".
BS 410 (1976).

/9/ American Society for Testing and Materials;
"Standard Test Method for pH of Activated Carbon".
ASTM D 3838 (1980).

/10/ DEUBER, H.; GERLACH, K.; KAEMPFFER, R.;
"Investigations on the Aging of Activated Carbons in the Exhaust Air of a Pressurized Water Reactor".
Nucl. Techn. 70 (1985) 161.

/11/ DEUBER, H.; GERLACH, K.; GIRAUD, V.; KAEMPFFER, R.;
"Investigations on the Aging of Various Activated Carbons in the Exhaust Air of a PWR over an Extended Period of Time".
19th DOE/NCR Nuclear Air Cleaning Conference,
Seattle, 18.-21.8.1986.

/12/ FURRER, J.; KAEMPFFER, R.; WILHELM, J.G.;
"Alterung und Vergiftung von Iod-Sorptionsmaterialien in Kernkraftwerken/Aging and Poisoning of Filters in Nuclear Power Plants".
Kerntechnik 18 (1976) 313.

/13/ KOVACH, J.L.; RANKOVIC, L.;
"Combustion and Control of Poisoning of Impregnated Carbons Used for Organic Iodide Removal".
CONF 780 819 (1979) 368.

/14/ WILHELM, J.G.; FURRER, J.; KAEMPFFER, R.;
"Untersuchung zur Ertüchtigung von Iod-Sorptionsfiltern zur Reinigung der Abluft von Kernkraftwerken."
KfK 3194 B (1981).

/15/ DEITZ, V.R.;
"Effects of Weathering on Impregnated Charcoal Performance".
NUREG / CR-2112 (1981).

Table I: Parameters of the tests with CH_3I-131

Parameter	Unit	Value
Temperature	°C	30
Relative humidity	%	70
Throughput	m^3/h	0.88
Face velocity	cm/s	50
Preconditioning time	h	$\geq$ 16
Injection time [a]	h	0.1
Purging time [b]	h	2
Bed depth [c]	cm	50
Residence time	s	1
Combustion products injected [d]	g	0; 0.1; 1; 10
I-131 injected	mCi	0.1
I-127 injected	mg	1

[a] Both for combustion products and CH_3I (injection of combustion products generally immediately prior to that of CH_3I)

[b] after injection of CH_3I

[c] 20 beds of both depth and diameter of 2.5 cm
(weight: ca. 5 g per bed)

[d] material combusted

Table II: Removal efficiency of 207B (KI) for CH_3I-131 at different types and amounts of combustion products (Test parameters: Table I)

Combustion product [a]		Removal efficiency of individual beds (%) [b]				
Type	Amount (g)	Bed 1	Bed 2	Bed 3	Bed 4	Bed 5
None	-	70	67	69	62	63
PVC cable	0.1	63	65	67	67	69
	1.0	21	55	60	51	60
	10.0	5	21	43	55	57
FRNC cable	0.1	63	69	69	72	73
	1.0	47	63	64	66	65
	10.0	43	46	52	55	55
Mineral oil	0.1	66	71	68	62	62
	1.0	51	62	70	69	71
	10.0	48	54	53	55	57
Paint	0.1	65	70	74	74	75
	1.0	52	58	63	63	65
	10.0	32	37	42	46	52

[a] Type and amount of material combusted

[b] Beds counted in direction of flow; residence time per bed: 0.05 s

Table III: Removal efficiency of 207B (TEDA) for CH_3I-131 at different types and amounts of combustion products (Test parameters: Table I)

Combustion product [a]		Removal efficiency of individual beds (%) [b]				
Type	Amount (g)	Bed 1	Bed 2	Bed 3	Bed 4	Bed 5
None	-	82	81	83	84	83
PVC cable	0.1	72	81	83	84	83
	1.0	25	68	75	78	79
	10.0	7	29	70	73	77
FRNC cable	0.1	67	70	82	87	87
	1.0	54	74	78	81	83
	10.0	36	51	62	68	72
Mineral oil	0.1	69	76	83	83	86
	1.0	57	74	80	83	84
	10.0	18	29	45	59	66
Paint	0.1	75	84	83	87	84
	1.0	51	69	75	77	79
	10.0	33	48	70	74	77

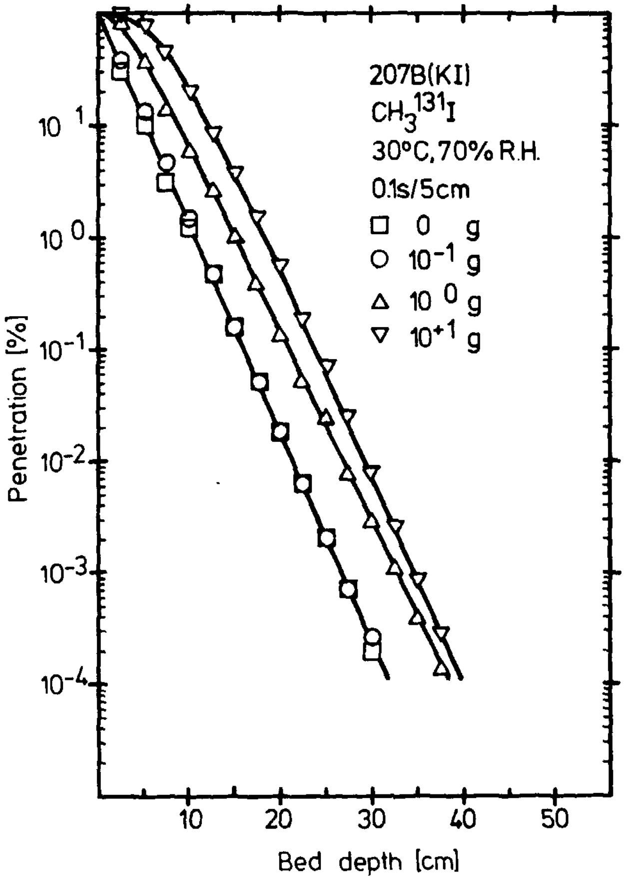
[a] Type and amount of material combusted

[b] Beds counted in direction of flow; residence time per bed: 0.05 s

Table IV: Organic combustion products from the materials used [a]

Material	Main compounds		Additional compounds (selection)
	Aromatic compounds	Non-aromatic compounds	
PVC cable	benzene, methyl-	cycloheptatriene	benzene, chloro-; napthalene
FRNC cable	benzene, methyl-; ethyl-	hydroxylamine, alkyl-; bicyclooctatriene; octane, methyl-; decene, methyl-; napthalene	straight and cyclic unsaturated hydrocarbons (C_6 - C_{14}) and alkyl derivatives thereof
Mineral oil	benzene, methyl-	-	benzene, alkyl-
Paint	benzene, methyl-, ethyl-; benzaldehyde; benzofuran	bicyclooctatriene	cyclohexene, alkyl-; heptane (straight, cyclic), alkyl-

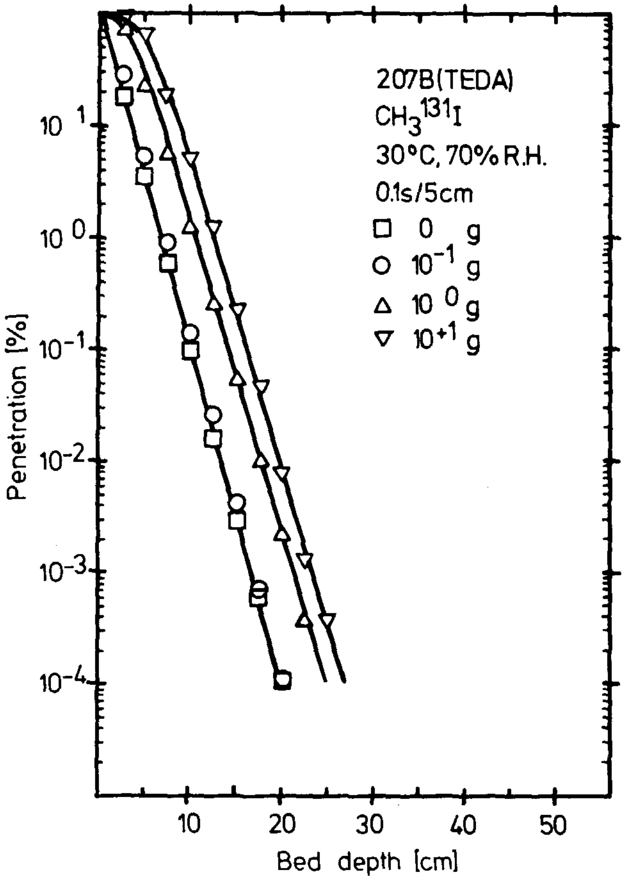
[a] Organic products found in the CCl_4 extracts of the combined first and second beds of 207B (KI) after challenge with combustion products



KfK LAF II D8556E

Penetration as a function of bed depth
at different amounts of combustion products
(PVC cable)

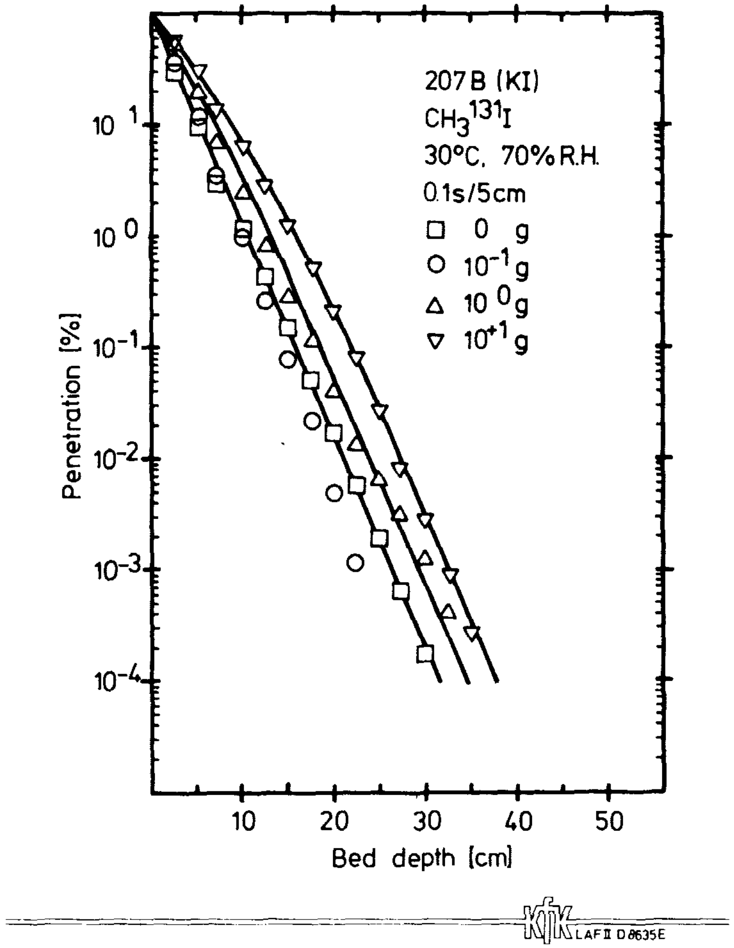
Fig. 1



KfK LAF II D8558E

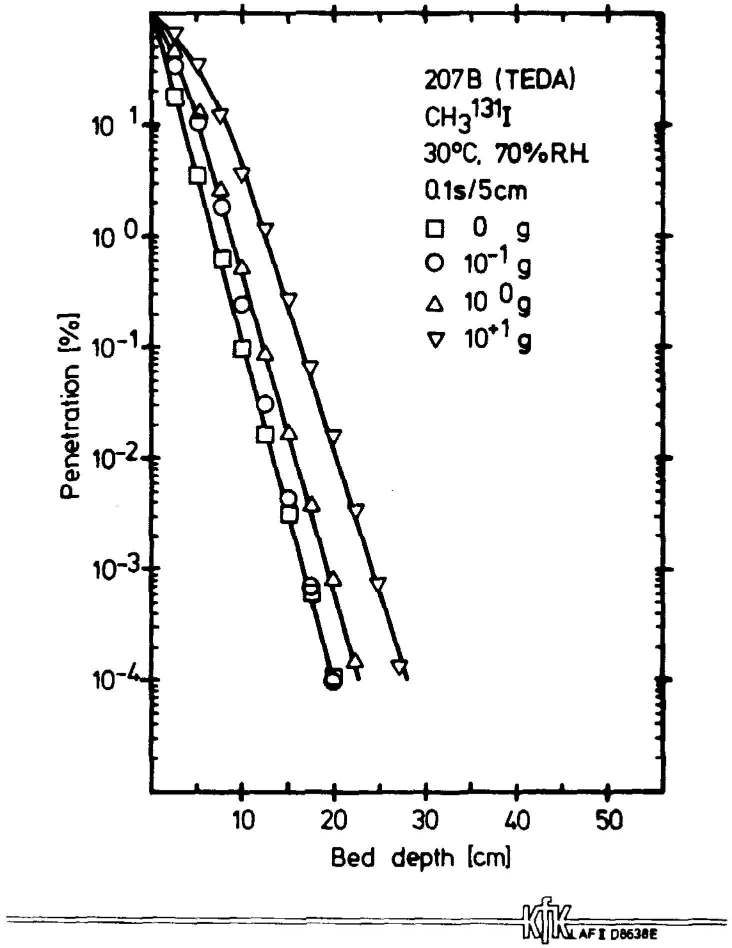
Penetration as a function of bed depth
at different amounts of combustion products
(PVC cable)

Fig. 2



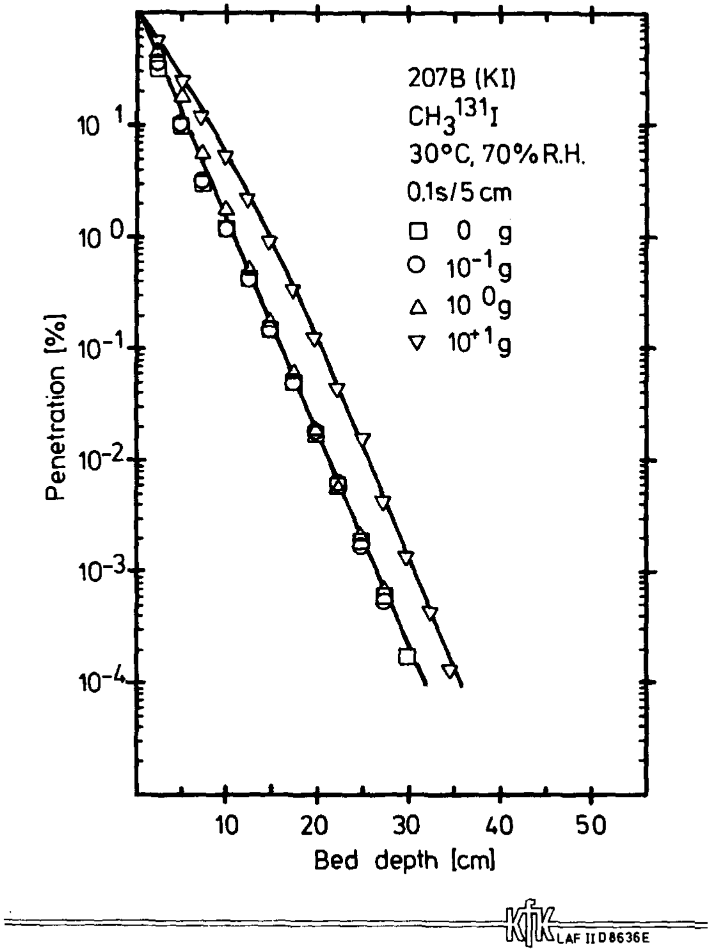
Penetration as a function of bed depth
at different amounts of combustion products
(FRNC cable)

Fig. 3



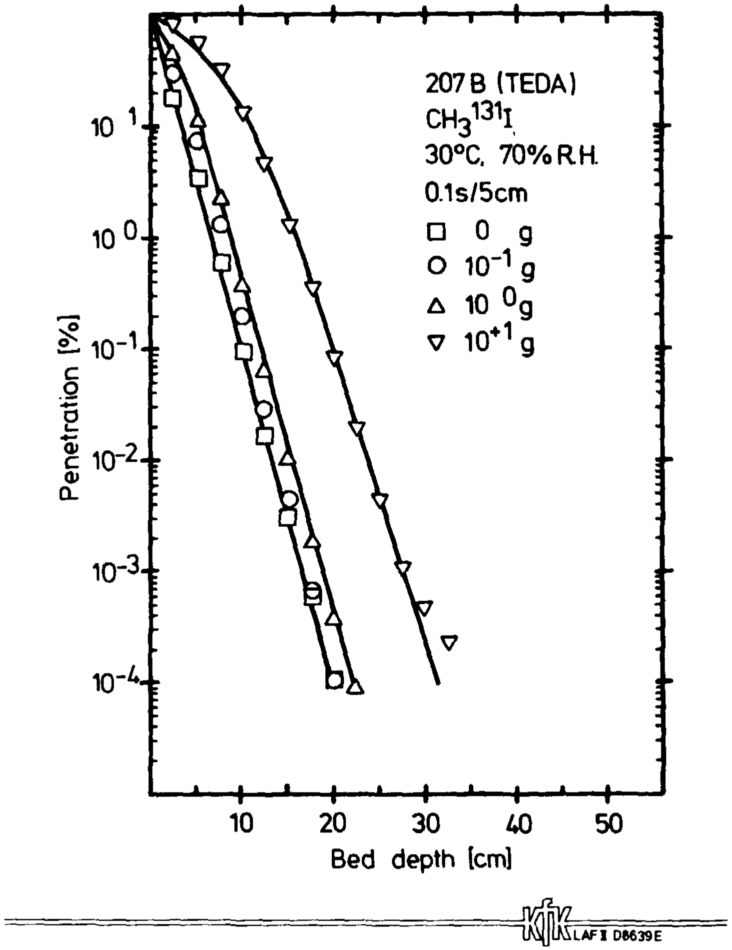
Penetration as a function of bed depth
at different amounts of combustion products
(FRNC cable)

Fig. 4



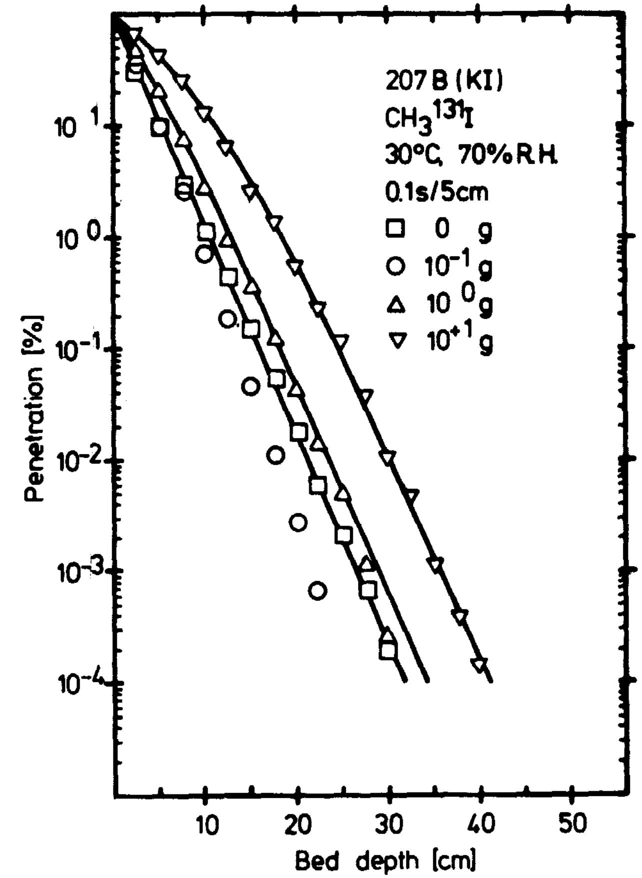
Penetration as a function of bed depth
at different amounts of combustion products
(mineral oil)

Fig. 5



Penetration as a function of bed depth
at different amounts of combustion products
(mineral oil)

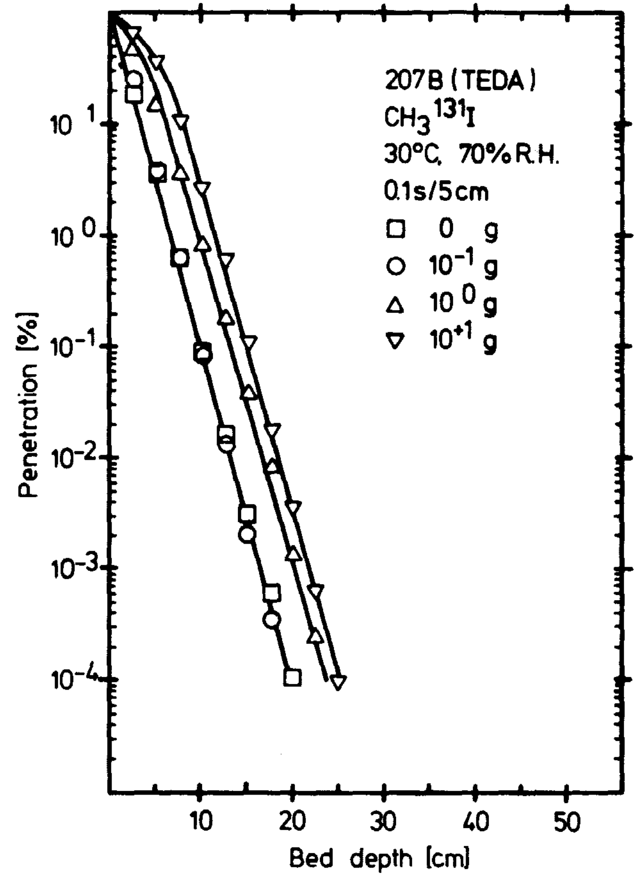
Fig. 6



KfK LAF II D8637E

Penetration as a function of bed depth
at different amounts of combustion products
(paint)

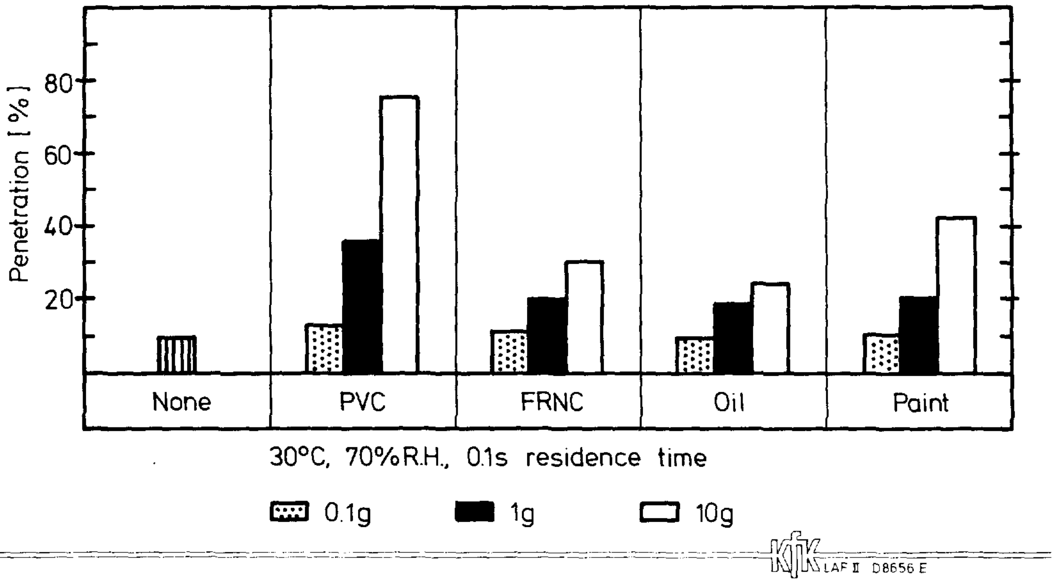
Fig. 7



KfK LAF II D8640E

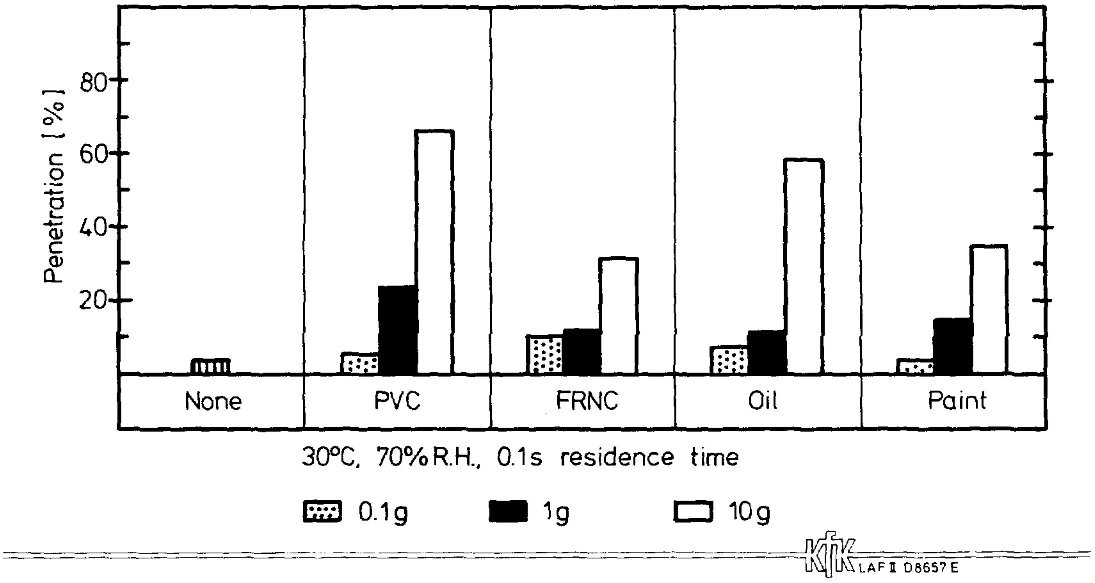
Penetration as a function of bed depth
at different amounts of combustion products
(paint)

Fig. 8



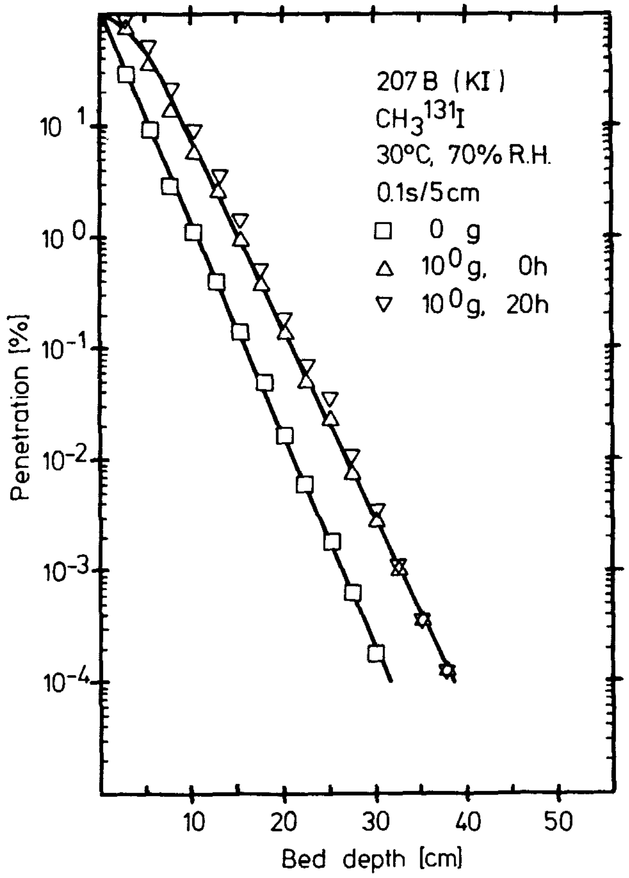
Penetration of 207B(KI) by $CH_3{}^{131}I$ at different amounts of combustion products

Fig. 9



Penetration of 207B(TEDA) by $CH_3{}^{131}I$ at different amounts of combustion products

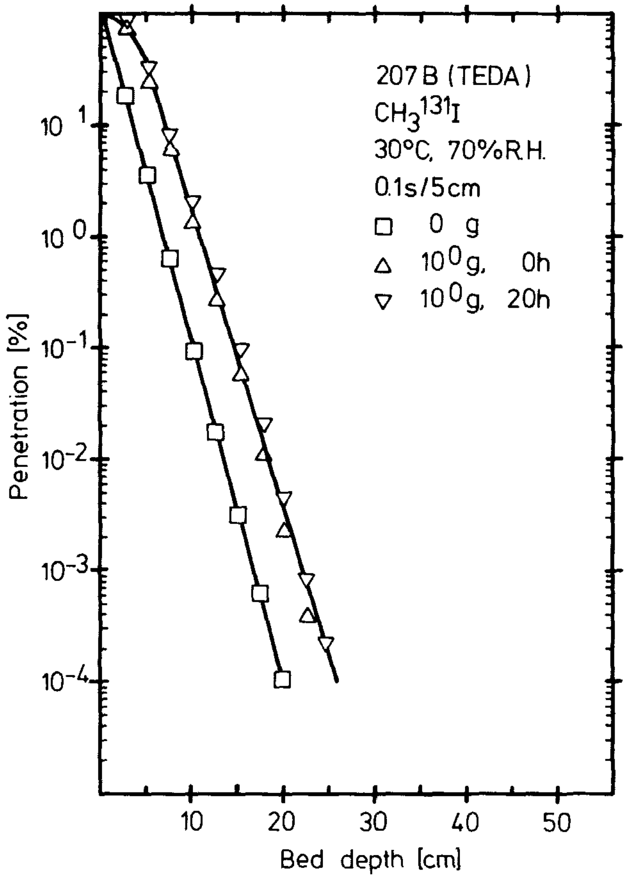
Fig. 10



LAF I D8653 E

Penetration as a function of bed depth at different amounts of combustion products and purging times (PVC cable)

Fig. 11 [a]

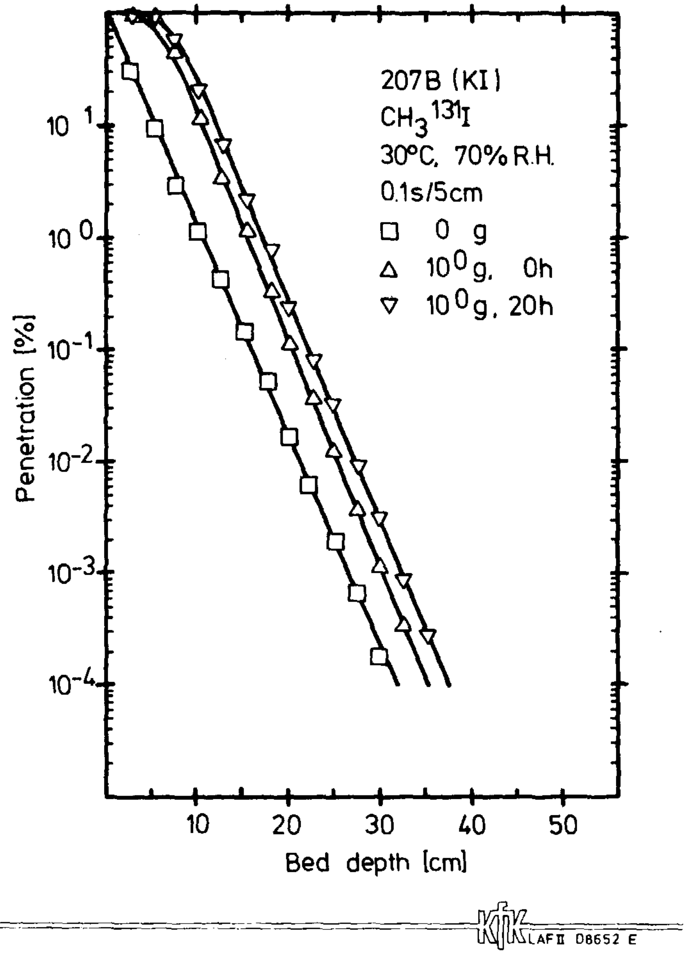


LAF II D8655 E

Penetration as a function of bed depth at different amounts of combustion products and purging times (PVC cable)

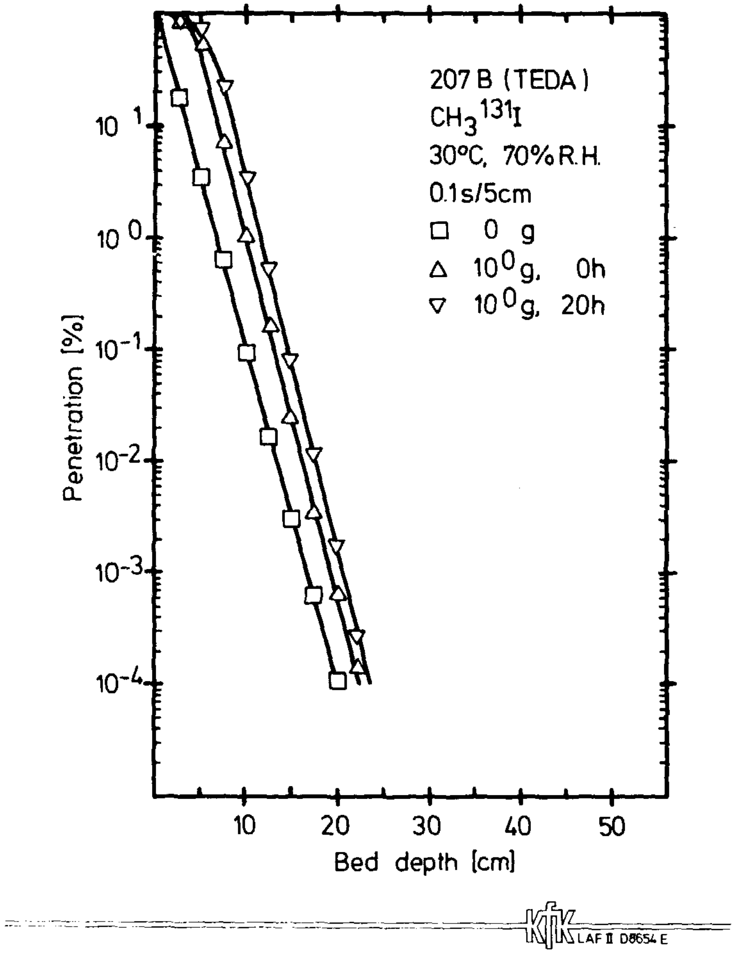
Fig. 12 [a]

[a] Purging, if any, between injection of combustion products and injection of $CH_3{}^{131}I$



Penetration as a function of bed depth
at different amounts of HCl and purging times

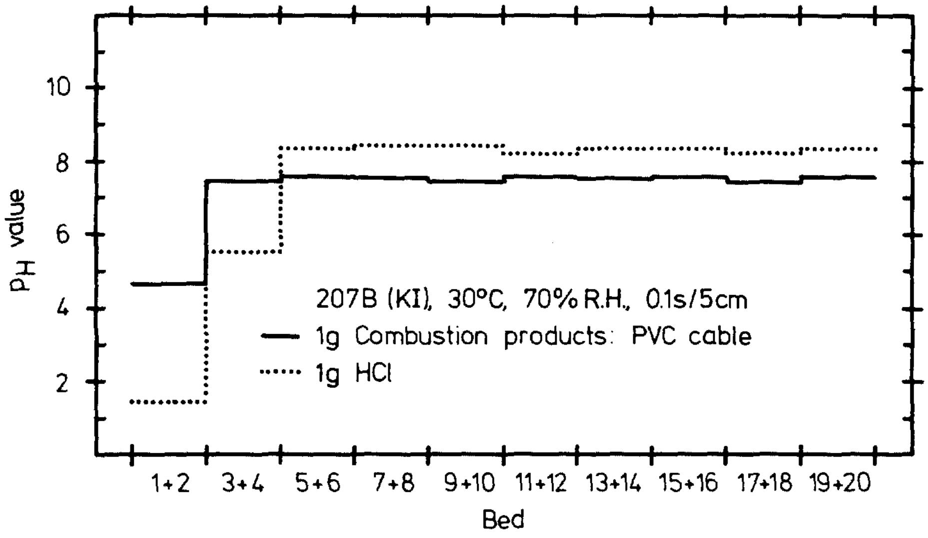
Fig. 13 [a]



Penetration as a function of bed depth
at different amounts of HCl and purging times

Fig. 14 [a]

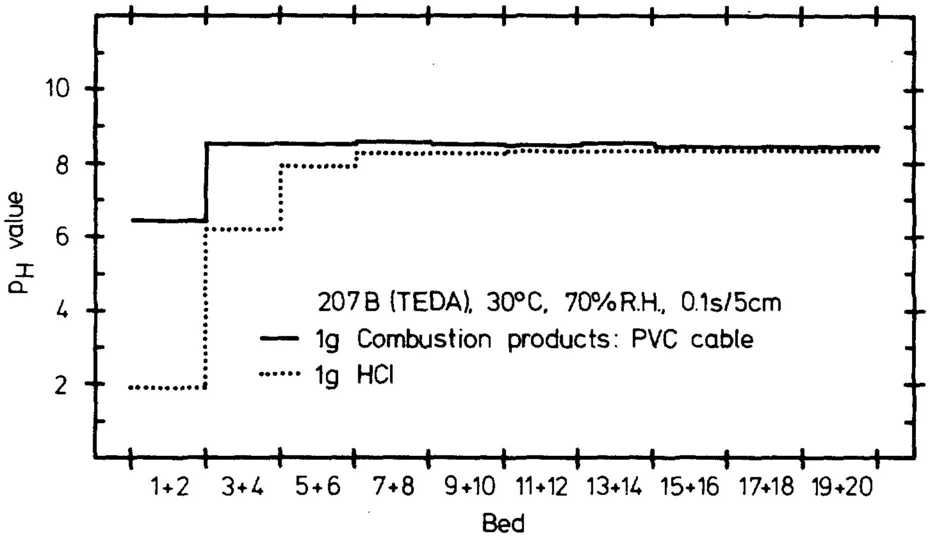
[a] Purging, if any, between injection of HCl and injection of $CH_3{}^{131}I$



LAF II D8659E

P_H value of the aqueous extracts of activated carbon beds after challenge with gaseous combustion products

Fig. 15



LAF II D8660 E

P_H value of the aqueous extracts of activated carbon beds after challenge with gaseous combustion products

Fig. 16

DISCUSSION

VIKIS: Were the combustion products distributed evenly over the charcoal?

DEUBER: No, the distribution was as it would be in the case of a fire: the loading decreased with increasing bed depth.

ALVARES: Why is there a difference in the behavior of TEDA and KI impregnated carbon?

DEUBER: It is generally found that the penetration of new TEDA carbon by methyl iodide is lower than that of new KI carbon. As for the increase in penetration due to the influence of combustion products, there was no decisive difference.

CONCLUSIONS FROM FIRE TESTS IN ACTIVATED CARBON FILLED ADSORBERS

W. Mathewes
Kraftwerk Union Aktiengesellschaft
Offenbach, W-Germany

Abstract

Activated carbons as used in gas-phase adsorption may be subjected to heating, either from heat applied externally to the carbon bed, or heat generated by radioactive contaminants, or by the adsorption process itself.

This report presents results of artificially ignited beds of activated carbon.

This report also considers results concerning the self-ignition of non-contaminated carbon and such of solvent-contaminated carbon subjected to external heating in beds with an air flow and in beds without an air flow.

An estimation is given for the heat generation caused by radioactive contaminants as well as by the adsorption process.

Studies of handling of endangered components and studies of alarm indicating systems give guidance for the contemporary lay-out and design.

Introduction

The nuclear plant ventilation and air exhaust systems, including the air cleaning systems are of some importance for the licensing authorities, the plant operating personnel and the public.

The protection of the environment of controlled areas as well as the releases to the environs have to be kept sufficiently low to assure compliance with relevant guidelines.

Therefore exhaust air systems must be designed, tested and maintained so that they will perform effectively and reliably under normal or emergency conditions to which they might be subjected.

In order to comply with requirements regarding fire hazards, activated carbon and carbon filters were tested under anticipated modes of operation most closely related to actual service conditions.

General considerations

Activated carbon filter systems should be installed in areas isolated by fire-proof walls, doors and dampers, which divide the building into its chief parts. These fire areas serve to maintain an effective barrier for heat and smoke. Contaminants as combustion products should be exhausted by cleanup systems. Smoke detectors in these areas and in the connecting duct work initiate the isolating function of dampers and preclude entrance of combustion products into other areas, or following filter installations.
Easy access for fire guards must be cared for.

Results of Calculations

The calculation of the effect of ignition on activated carbon through heat generation by radioactive contaminants in a normal operation mode operated stand-by unit shows only a negligible temperature rise.
The temperature rise originating from the adsorption process itself is less than 1 °C.

Test Facility

For the investigation of fire hazards in beds of activated carbon a test facility was to be designed and built to supply experimental data.

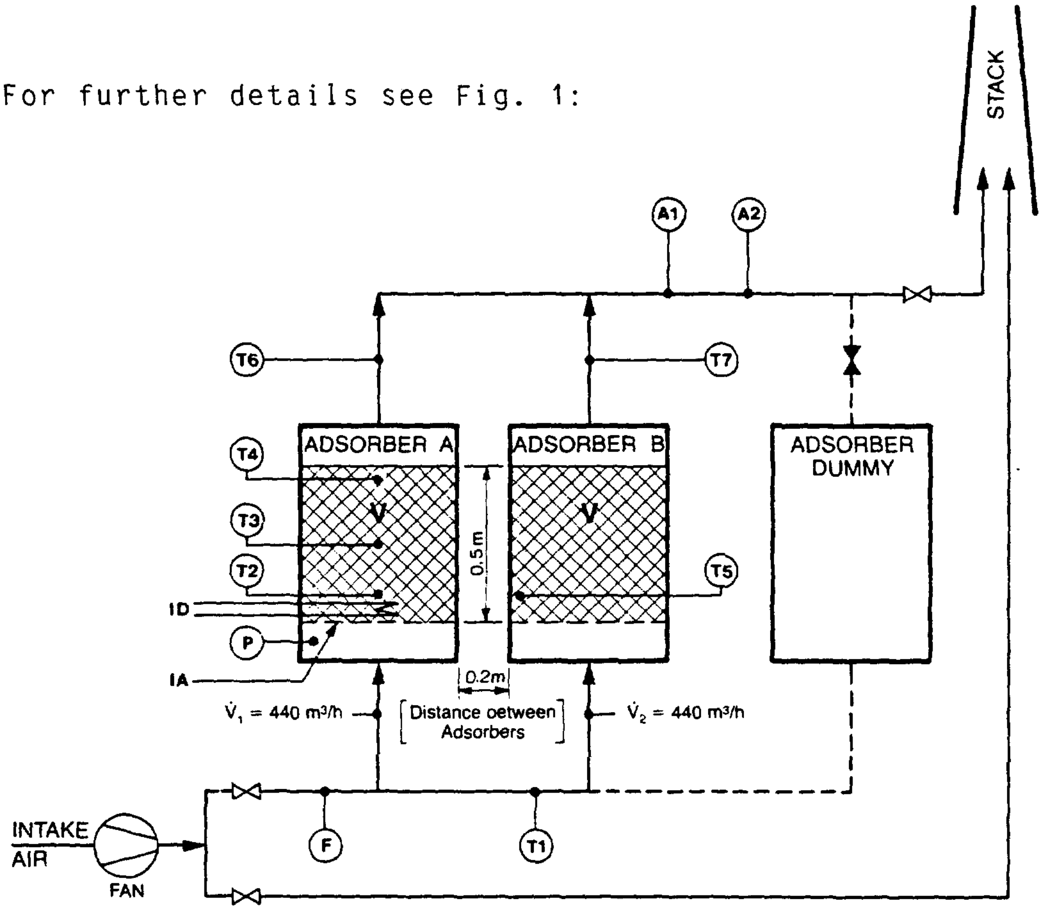
The most essential parts of the equipment were the adsorbers. The inflow surface area (IA = o,35 m^2) corresponded to a quarter of the original adsorber as in the NPP Philippsburg 2. The volumetric air flow rate was calculated to be $\dot{V}$ = 44o m^3/h per test adsorber, the inflow velocity was equal to o.35 m/s based on the inflow area of the adsorber.

The bed depth selected was 5oo mm as in the original adsorber in Philippsburg.

The volume of activated carbon amounted to o.175 m^3 per adsorber.

The carbon was supported by a grate, the air flow was directed from the bottom to the top of the carbon bed. The activated carbon (8-12 mesh) satisfied ANSI/ASME N 5o9.

For further details see Fig. 1:



LEGEND:

A1 = Analysis of Combustion Gas (see Table 3) (intermittent sampling)
A2 = CO-Monitor (0–250 ppm.) – (see Figure 13) (continuous sampling)
F = Flow Meter (m/s)
P = Pressure Drop (mm WG)
T = Temperature Measurements (°C) – (see Figure 5 through 11)
ID = Ignition Device (W)
IA = Inflow Area = 0.35 m²
$\dot{V}$ = Volumetric Air Flow/Adsorber Hour = 440 m³/h
V = Activated Carbon Volume/Adsorber = 0.175 m³
⋈ = Damper open
⧓ = Damper closed

Figure 1 Flow Diagram of Test Facility

Instrumentation

Appropriate instrumentation (fig. 1) was set up to obtain measurements of the following.

1. Ambient pressure and temperature
2. Air temperature evaluated for this report at 7 locations; 27 measuring points at the actual trial installation
3. Average volumetric air flow rate in the duct
4. Filter pressure drop
5. Sampler for intermittent analysis of combustion gas
6. CO-Monitor for continuous sampling
7. Ionization measuring instrument for continuous sampling

Experimental Results

It was necessary to ignite the activated carbon artificially by electrical resistance heating. In general we had a pinpoint resistance ignition device (ID) to generate the electrical heat output.
At an input of 6o W no ignition occurred even when the heating time was extended to more than 24o s. For ignition of activated carbon we had to apply 8o W capacity input for 18o s.

We found the lowest ignition temperature of activated carbon impregnated with calium iodide in a bed with an air flow and with a volume of o.17 m^3 to be at 3oo °C.

The lowest ignition temperature of carbon contaminated with solvent (12 weight % of toluene), ranged up to 25o °C.

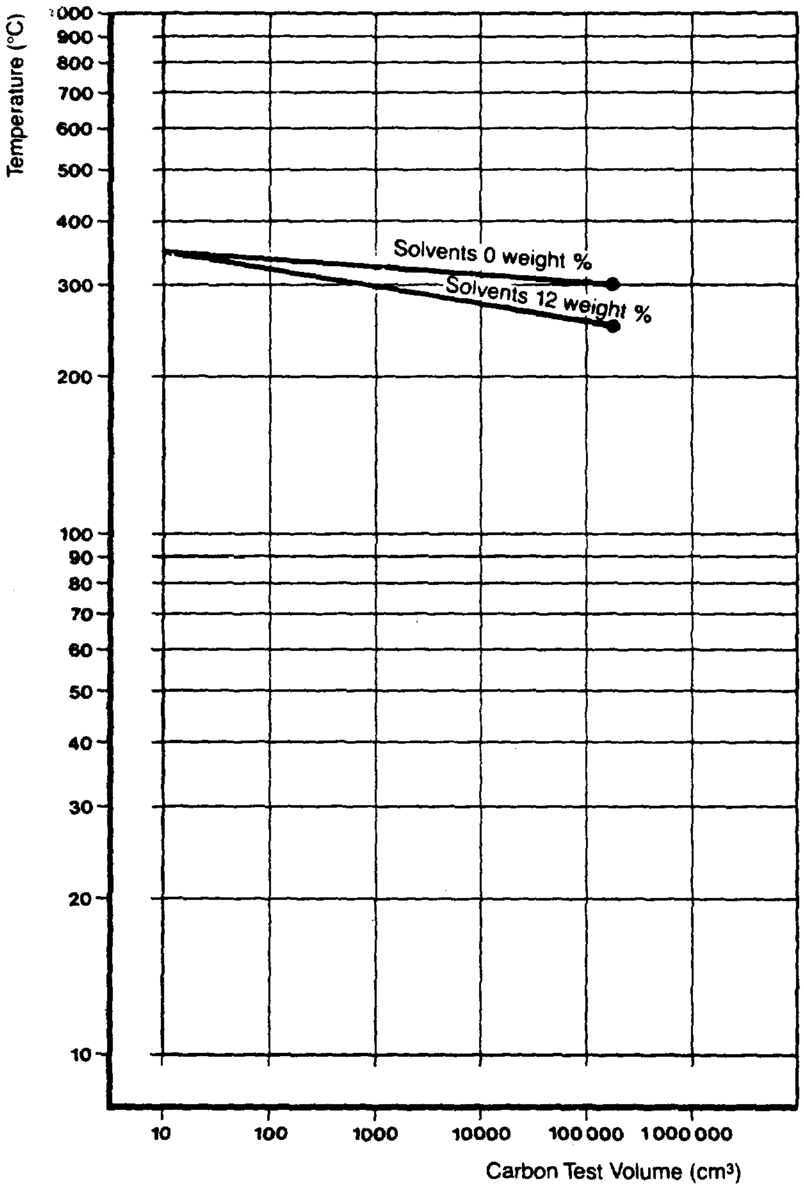


Figure 2 Ignition Temperature of Activated Carbon as a Function of Test Volume of Carbon

Figure 2 shows the relation between ignition temperature and contamination of solvents, and the volume of carbon in beds with an air flow.

The reference ignition temperature of activated carbon in flowing air based on a volume of 1o cm^3 was determined by the Standard Test Method ANSI/ASTM D 3466-76. The report shows, that ignition occurred at 35o °C. No real difference in ignition temperature was found between the unused carbon and the carbon contaminated with solvent (12 weight %), (figures 3 and 4).

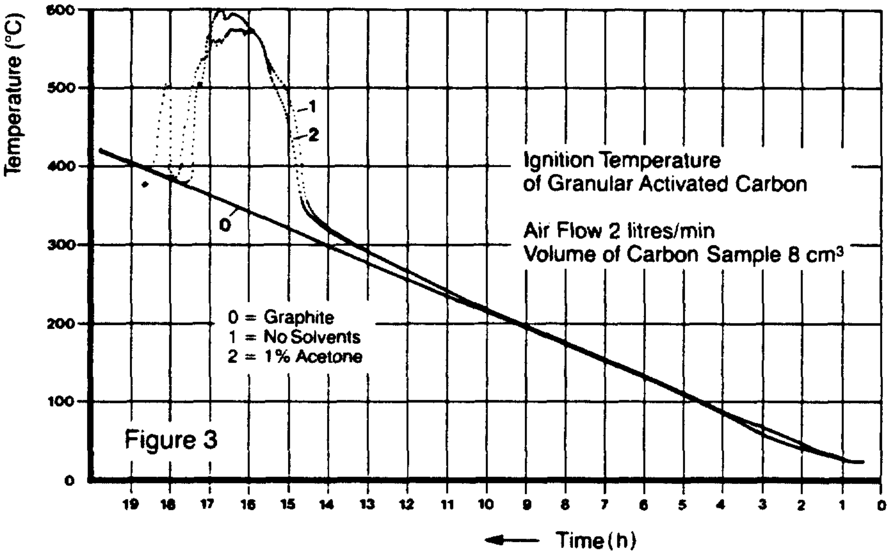


Figure 3

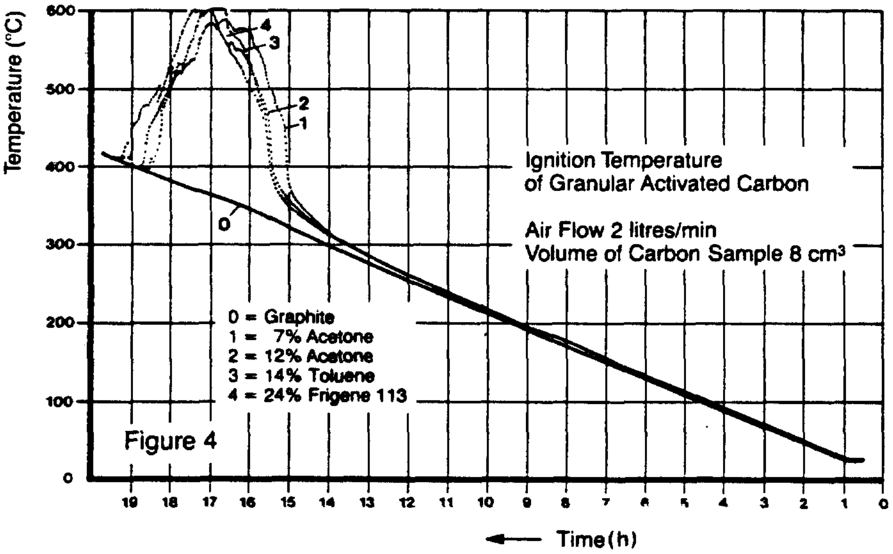


Figure 4

We also considered the possibility of self-heating through the reaction of solvents in beds of activated carbon without air flow subjected to external heating, rendered in Table 1.

We wanted to obtain conservative test results. In this case we decided on acetone instead of other solvents, we also chose 12 weight % as a maximum figure although we knew from experience that the contamination of activated carbon in operational processes does not exceed 8 weight % in general and for acetone in particular does not exceed 2 weight %.

We realised that the test contained two steps as regards the self heating reaction, i. e.:

First step: thermal reaction of solvent through temperature rise to such an extent as to ignite the carbon

Second step: thermal reaction of carbon

TEST	CARBON + ACETON	TEMP. OF BED	SELF-HEATING REACTION	CARBON WEIGHT TEST START	TEST STOP	COMMENTS
No	WEIGHT %	°C		g	g	
1	12	45	t_1 = 4,5 ΔT = 2,9 t_2 = 48,8	323	288	No visible change in material
2	12	70	t_1 = 5,8 ΔT = 0,9 t_2 = 107	322	290	No visible change in material
3	12	90	t_1 = 4,5 ΔT = 1,3 t_2 = 108	324	289	No visible change in material
4	12	140	t_1 = 2,0 T = 348 t_2 = 2,1	323	282	No visible change in material
5	1	140	t_1 = 4,8 ΔT = 10 t_2 = 0,8	370	296	No visible change in material

LEGEND:

t_1 = Heating Time up to ambient Temperature = Temp. of Bed (h)
t_2 = Time up to Maximum Temperature Rise (h)
ΔT = Temperature Rise above Temperature of Bed (°K)
T = Temperature of Sample (°C)

Table 1 Self Heating Reaction of Carbon in Beds with no Air Flow

In test No. 1, 2, 3, 5 there were no indications of any exothermic reaction of aceton or of carbon.

Only test No. 4 showed a rise in temperature responsible for the self heating reaction of solvents (step 1).

In tests Nos. 1-5 we noticed no real loss of carbon weight, so we concluded that no exothermic reaction of the carbon took place (step 2).

After studying the different possibilities of self-ignition of activated carbon, we had to learn to handle this hazard for carbon filter systems.

To ensure that the test procedures simulated as closely as possible our installation conditions, we had to provide an additional volume of air contained in a stand-by vessel in order to account for parallel adsorbers including their connecting duct work (see fig. 1).

To study radiation heating, we installed the parallel adsorbers at a distance of 2oo mm to the test adsorber (fig. 1).

So prepared, we ran 12 tests. For this report the most interesting one for handling the components at risk and for restoring safe conditions, involved the interruption of airflow by shutting the upstream damper.

After ignition as described before and before shutting the damper we reached a top temperature of 14oo °C (fig. 6). At this, a maximum temperature of 42 °C was induced by radiation in the parallel adsorber (fig. 7).

The figures 5 through 11 show the distribution of temperatures we found.

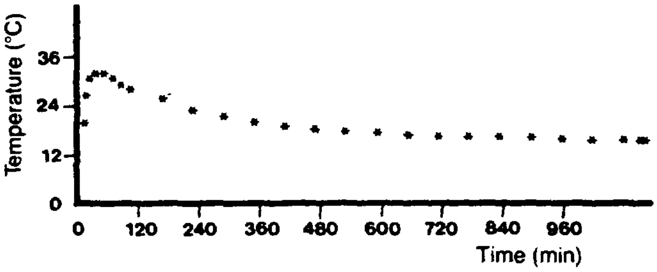


Figure 5 Temperature of Inflow Air T1

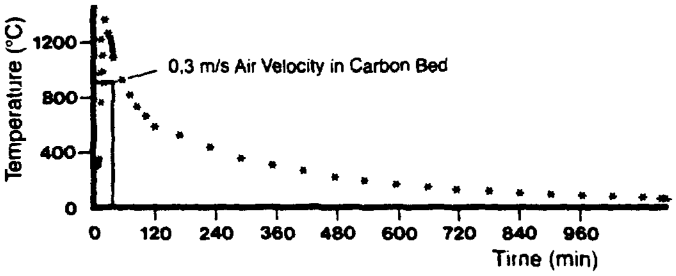


Figure 6 Temperature of Lowest Bed T2

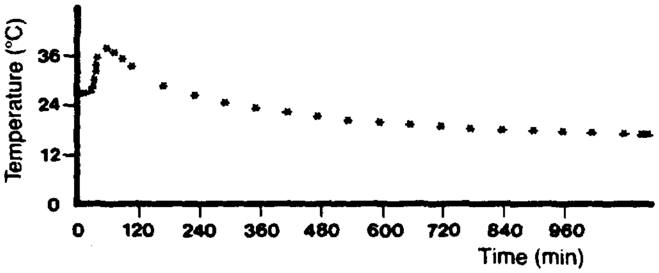


Figure 7 Temperature Near Wall
of Parallel Adsorber T5

All figures show temperature peaks 3o mins after kindling the carbon.

Due to convection the temperature rise above the fire core moved towards the next layer and amounted here to a maximum of about 1000 °C (fig. 8).

At the same time the upper bed layer registered a peak of about 90 °C (fig. 9), and the adsorber outlet temperature reached 44 °C (fig. 1o).

A tendency towards temperature rise can also be noted in the neighbouring adsorber B (fig. 11).

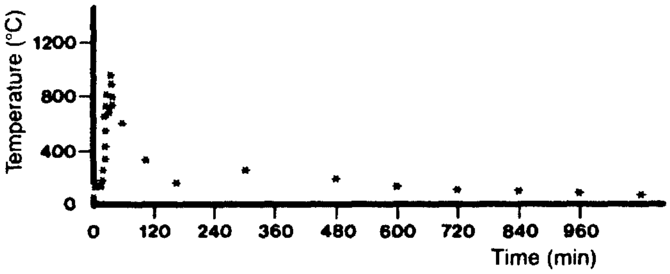


Figure 8 Temperature of Midbed T3

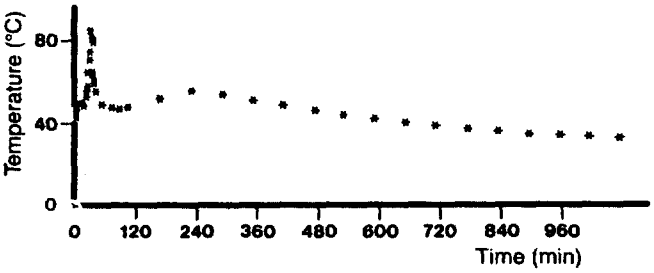


Figure 9 Temperature of Upper Bed T4

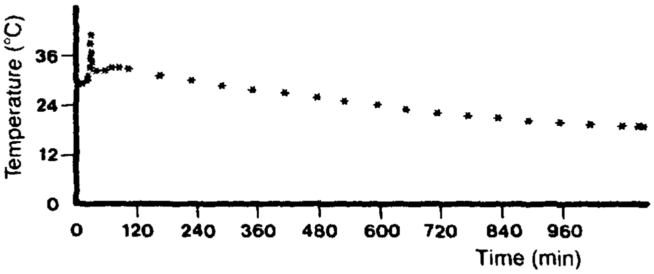


Figure 10 Temperature of Outlet Air
Adsorber A T6

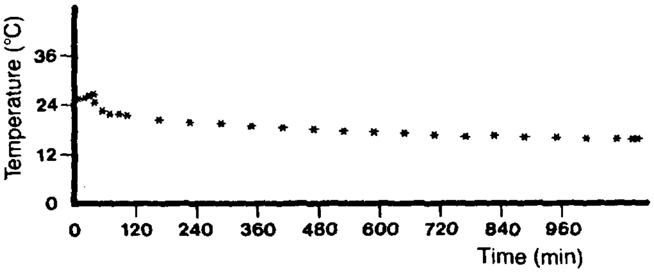


Figure 11 Temperature of Outlet Air
Adsorber B T7

After the shut-down of the upstream damper the measurements listed in Table 2 point to a general decrease in temperatures and to a circulation of air passing the trial installation (fig. 12).

TEST		TIME BETWEEN STEPS	ENTRY TEMP SYSTEM T1	MAX. TEMP. INVOLVED ADS. "A" T2	OUTL.TEMP. INVOLVED ADS "A" T6	MAX. TEMP. PARALLEL ADS. "B" T5	EXIT TEMP. PARALLEL ADS. "B" T7	EVENT
STEP	TIME							
No	(h)	(h)	(°C)	(°C)	(°C)	(°C)	(°C)	
1	2pm		18	20	20	20	20	TEST START
		0,5						
2	230pm		32	>1400	44	40	26	CLOSING UPSTREAM DAMPER
		1,5						
3	4pm		30	550	34	28	22	
		1						
4	5pm		26	500	30	26	28	
		4						
5	9pm		18	200	24	20	18	
		11						
6	8am		18	>100	20	20	18	
		3						
7	11am		18	<90	20	20	18	
		3						
8	2pm		18	<80	18	18	18	TERMINATION OF TEST

Table 2 Thermal effects of circulated air passing the adsorbers A and B and returning via dummy-adsorbers

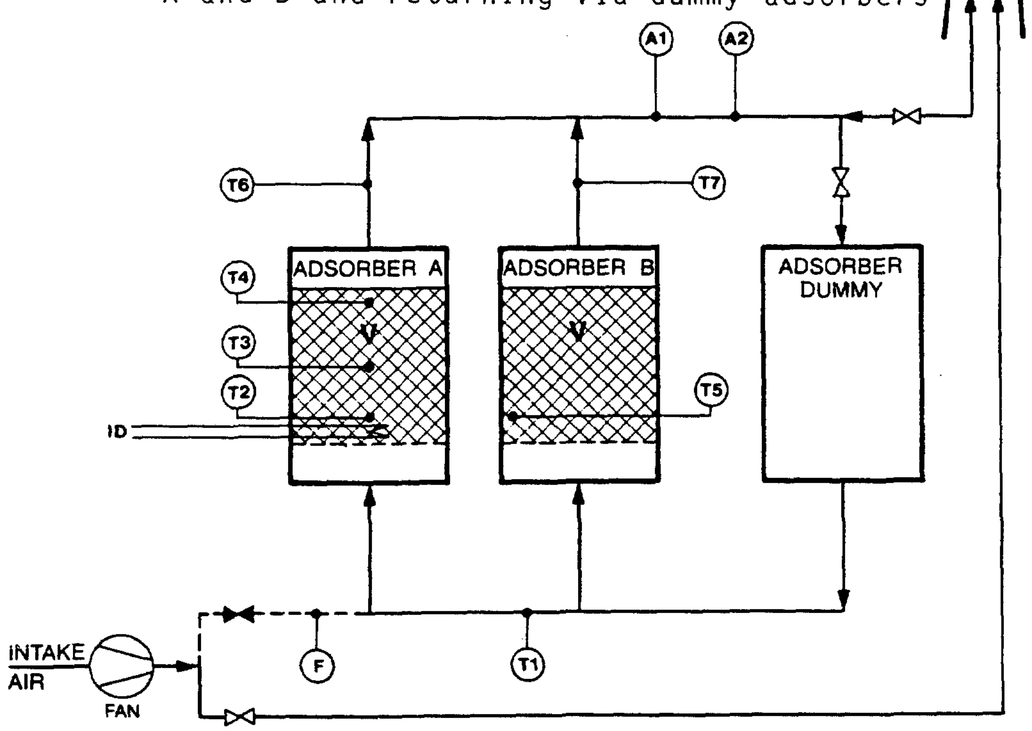


Figure 12 Air passing the pair of Adsorbers and heading back via the Dummy-Adsorber

Table 3 presents the results for the composition of gases found by intermittent sampling in A1.

Wherein test-step 3 shows a fairly good combustion and a near to 2 Vol. % consumption in oxygen. From test-step 4 onwards there is a gradual increase in carbondioxyde and likewise a decrease in oxygen, pointing to an internal current of gas being set at motion by thermal forces, thus conveying additional oxygen originating from the adjacent volumina contained in the second adsorber, the dummy adsorber and the connecting ductwork as well as the oxygen being desorbed by heat.
These sources delivered oxygen for the thermal reaction of the carbon (test steps 4, 6, 7).

It can also be deduced, that the atmosphere of the inflicted adsorber was inertised after closing the upstream damper, in the course of about 2o hours.

TEST		TIME BETWEEN STEPS	MAX. TEMP. INVOLVED ADSORBER T2	GASEOUS COMBUSTION PRODUCTS A1				EVENT
STEP	TIME			CO_2	CO	N_2	O_2 A1	
No	(h)	(h)	(°C)	Vol. %	Vol. %	Vol. %	Vol. %	
1	2pm		20					TEST START
		0,5						
2	230pm		>1400					CLOSING UPSTREAM DAMPER
		1,5						
3	4pm		550	14	5	79	2	1. GAS ANALYSIS
		1						
4	5pm		500	6,6	9,9	79	4,5	2. GAS ANALYSIS
		4						
5	9pm		200					
		11						
6	8am		>100	8	8,8	79	4,2	3. GAS ANALYSIS
		3						
7	11am		<90	10	7,2	79	3,8	4. GAS ANALYSIS
		3						
8	2pm		<80					TERMINATION OF TEST

Table 3 Gaseous Combustion products after Shut down

Temperatures dropped to less than 8o °C after a spell of 23.5 test hours (compare relevant figures and tables), in general the graphs point to a temperature loss in the heated carbon. In all tests, no damage was caused to the structures of the inflicted adsorber or to the other components except in some cases to the thermocouples near the fire core and the grate.

To keep fire accidents under control a quickly acting alarm annunciating system was needed to protect the facilities on the whole. Therefore the time lapse between the start of the fire-induced accident and the actuation of protection equipment resulting from measurements should be as short às possible.

To find out the time within which a distinct rise in temperature occurred, the temperature curve was compared with the CO-curve (fig. 13).

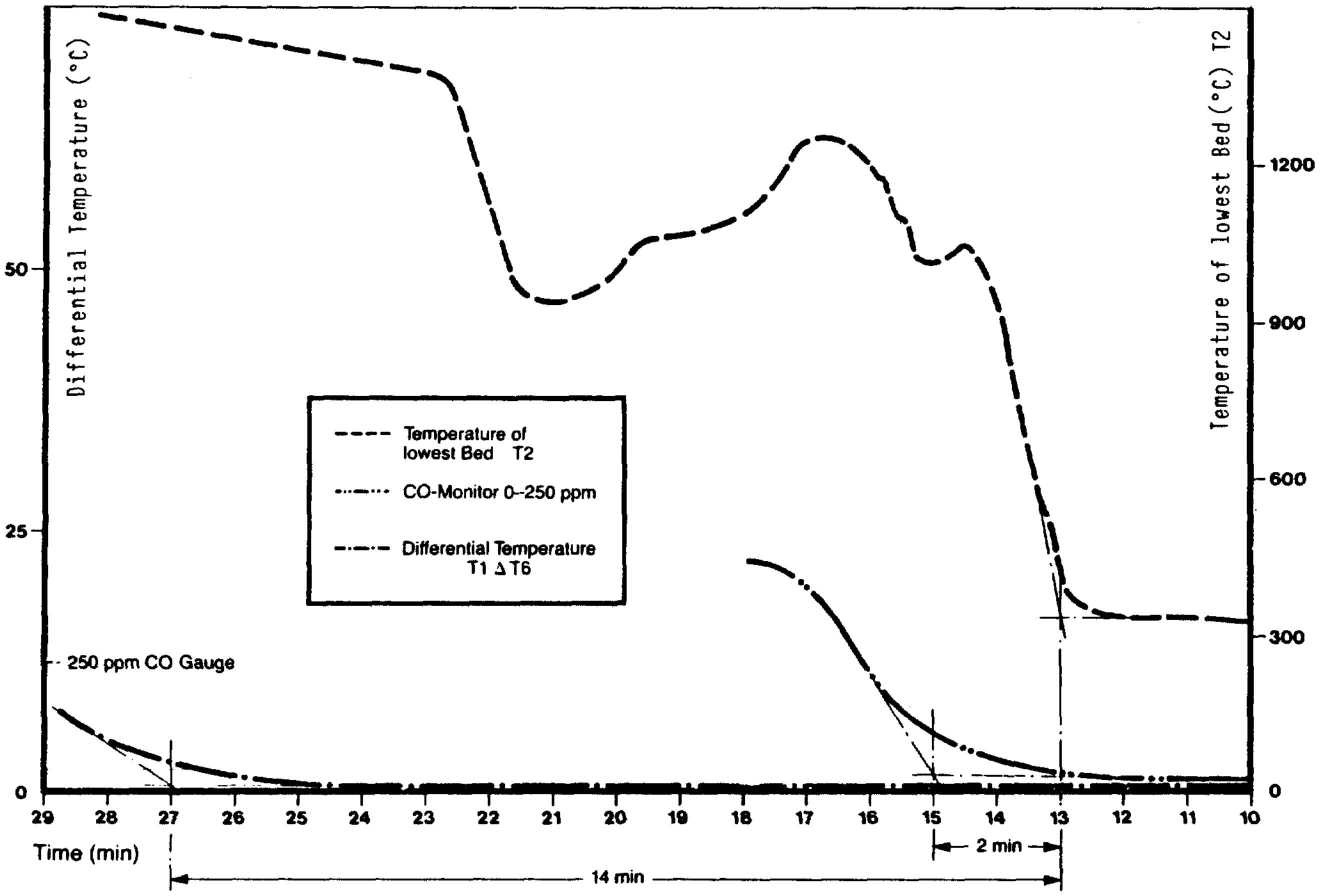


Figure 13 Time Difference between Temperature Rise and CO-Monitoring

We found the time required for a distinct temperature rise in the exhaust duct work resulted in a slow signal. On the base of CO-monitoring in the exhaust duct work (same position as the measuring point for temperature) actuation was obtained after a spell of 2 minutes.

An ionisation control device was tested as well. Although a quick signal was derived from the exhaust, no statement can be given regarding the possibility of dust-pollution of the sensor for its service life, since fine particles of carbon are expected downstream of the filters.

Concluding remarks

Our conclusions regarding these experiments are as follows:

1. A quick-acting and reliable fire detection annunciator must be provided for monitoring the exhaust ductwork, preferably a CO monitoring device.

2. Sufficient protection of carbon filters at risk can be accomplished by closing only one damper of a system thus preventing a pressure rise in the filter and thus reducing further thermal reaction by accumulating the content of gaseous carbon dioxide, therefore effectively combating the fire.

3. Self-ignition temperatures of activated carbon depend on its bulk volume and the content of solvents being adsorbed. In general greater volumes of carbon and a higher solvent content decrease the self-ignition-temperature.

4. No risk of self-ignition was noted for adsorber-filters in the stand-by mode for ambient temperatures up to 9o °C and 12 weight-% of solvents in the carbon.

5. With the presented results of our investigations on the subject of artificially ignited filter beds of activated carbon and the study of different alarm indicating systems, the carbon filter systems will be no risk to the environment or the building structures.

DISCUSSION

VOGAN: Has there been any air cleaning system fire anywhere in Europe that you have been aware of that generated the type of testing that you have undertaken.

MATHEWES: No.

VOGAN: We are looking at the need for fire protection systems for our standby air treatment systems, and we are looking for evidence of self-sustaining fires in air treatment systems. My second question stems from the first, would your testing show that there would be any continuing ignition of a carbon bed type of air treatment system?

MATHEWES: I have presented to you what we tested. It was a deep-bed filter. This type of filter system has been realized in Philipsbourg, but I am happy to say, we haven't had any chance to test its performance in real life.

A CURRENT EVALUATION OF FIRE LOSS CONTROL SYSTEMS FOR CHARCOAL MEDIA

Wayne D. Holmes
American Nuclear Insurers
Farmington, Connecticut

Abstract

Activated charcoal in use in nuclear facilities presents a significant loading of combustible material. There exists within nuclear facilities, numerous sources of ignition of combustible materials. If a fire may occur, it is important to detect the fire and suppress it, thus limiting the consequences of fire.

The arrangement of adsorber units and the mode of combustion of the charcoal media make the extinguishment of charcoal fires quite difficult. Automatic water spray systems have been proven to be an effective, reliable, and inexpensive means of extinguishing charcoal fires. Properly designed, installed, and maintained automatic water spray systems do not fail on a frequent basis and the effects of such failures have been overstated.

I. The Fire Problem Presented by Charcoal Adsorbers

The charcoal media in gas treatment systems in nuclear power plants presents a significant loading of a material which is nearly totally combustible and which has been reduced to a readily burned form. In some areas, many tons of charcoal may be present, representing a fire loading approaching 250 MBTU of potential heat release under fire conditions. Because of the mode by which combustion occurs within charcoal arrays and the means of enclosure of the adsorber systems, charcoal fires in such systems may be extremely difficult to extinguish. In tests performed by the Factory Mutual Research Corporation for the U.S. Navy on the combustibility of charcoal filter units it was noted that the charcoal media burned vigorously and once ignited, will burn to completion (1). Smoke obscuration was moderate to dense during the fire test.

The "Defense-in-Depth" approach to firesafety subscribed by the nuclear power industry is a multi-level approach. The first step in this approach is to prevent fires from occurring. The second step involves detecting fires quickly, suppressing those fires which do occur, and limiting fire damage. These principles apply to charcoal adsorbers as well as to any other plant systems that may contain combustible materials.

II. PREVENTION OF IGNITION

How or when charcoal may be ignited in nuclear plant systems is a subject which has been vigorously debated for many years and which is likely to continue to be debated for many years to come. What is clear is that the charcoal media is a combustible material which may be subject to ignition from numerous sources.

The actual ignition temperature is a function of a number of variables. Among those factors which may have an effect on the ignition temperature are: the ash content of the charcoal, any impurities within the media, the internal surface area of the charcoal, the specific characteristics of the impregnating material, the depth of the charcoal bed, and the air flow rate (2). Moderate heating of bulk activated charcoal may result in a spontaneous exothermic reaction resulting in ignition. Depending upon the depth of the charcoal bed, critical temperatures leading to spontaneous ignitions of activated charcoal have been reported from as low as 60°C. to 125°C. with times to ignition from 1.3 hours to several days (3).

Perhaps too much emphasis has been placed upon the probability of the decay heat of radioiodine as resulting in a cause of ignition of charcoal. It is reasonably clear that desorption of radioiodine is initiated at temperatures below the published standard ignition temperatures for charcoal. The adsorption systems are designed such that the decay heat generated by collected fission products cannot cause ignition of the charcoal nor significant desorption of the collected fission products under normal air flow. However, forced air circulation may provide the only significant cooling method and calculations and laboratory tests have shown that ignition temperatures may be reached under post-accident conditions if the airflow is reduced below 5 FPM (2). Even if decay heat does not, in itself, produce ignition temperatures within the charcoal, the effect of the elevated temperature produced by the decay heat will reduce the amount of additional energy which must be applied to initiate ignition and may also produce temperatures which will initiate self-sustained, spontaneous heating as mentioned previously.

There exists sufficient other possible sources of ignition of charcoal for there to be a concern for protection against fire. External fires may present an exposure to filter units either through direct contact or indirectly through duct systems. Welding and burning operations present another possible source for ignition. Electric heaters, or failures of other electrical systems might serve as a source of ignition. Flammable gases, as might be generated under post-accident conditions, introduces another possible source of ignition.

While the safety concerns associated with charcoal fires are greatest during plant operation, or more specifically during post-accident operations, the highest potential for ignition and resultant direct fire damage occurs during maintenance operations. During maintenance, one might anticipate cutting and welding being performed, solvents being used, portable heaters being utilized, unapproved temporary electrical systems being introduced, or other potentially hazardous operations being performed; all of which increases the potential for ignition of charcoal.

Preventing potential ignition sources is important in overall fire protection programs. The reasonably conservative approach, which is consistent with the fire protection defense-in-depth approach, is to assume that where concentrations of combustible materials are located, fires may occur. It then becomes prudent to design to detect and ultimately suppress those fires which may occur.

III. SUPPRESSION OF CHARCOAL FIRES

The mode of combustion of charcoal is quite different than that of most ordinary combustible materials typically present in nuclear facilities. In the form in which it is used, the charcoal will undergo a glowing combustion rather than a fire burning process. There is a very important and distinct difference between these two processes. Charcoal undergoes a different form of combustion by combining directly with oxygen, as evidenced by glowing. This is an important distinction, since many chemicals which are effective flame inhibitors have little or no effect in retarding flameless combustion (4). Therefore, by the basic mode of combustion of charcoal, fires involving charcoal may be much more difficult to extinguish than free burning fires in ordinary combustible materials.

Aside from basic problems associated with the diffulty in suppressing the glowing combustion process, manual fire fighting, i.e. fire brigades with portable fire extinguishers, may be exceptionally difficult for systems utilizing charcoal. The charcoal is generally located in substantial housings which are not readily accessible, particularly under fire conditions. In addition to physical obstructions to fire fighting, there may be considerable smoke obscuration or radiological conditions which may limit access for manual fire fighting. The use of hoses external to the housing can only help to prevent further damage to the building or adjacent equipment. It cannot extinguish the internal fire nor prevent contamination. By the time ignition occurs, containment has been lost due to desorption.

By the deep-seated nature of charcoal fires, the only reasonable means for manual fire suppression may require dismantling the charcoal array in order to quench the fire at its source. This is likely to be accomplished only after considerable time delay and damage.

With the difficulties presented for manual fire suppression of charcoal fires, it is therefore wise to design charcoal units with fixed, fire extinguishing systems. One must, however, choose the fire suppression medium with care for an effective system.

Dry chemical fire suppression agents applied to the surface of charcoal will not be effective on fires which may be deep within the array. Such systems have not been proposed for protection of charcoal beds.

Gaseous fire suppression agents such as carbon dioxide and Halon 1301 are not considered to be adequate for extinguishing deep-seated fires such as in charcoal. Carbon dioxide would have to be provided in such high concentraions and for such long durations to be effective that it would not be practical and might pose serious life safety problems. (5) Attempts to use carbon dioxide to extinguish charcoal adsorber fires have been unsuccessful. (6) Deep-seated fires are, by definition, not extinguished by a normal, 5%, concentration of Halon 1301 within 20 minutes. (7) To extinguish fires in charcoal beds, an excessive quantity of Halon 1301 would be required.

Liquid nitrogen extinguishing systems have been proposed and their effectiveness has been demonstrated by test. (6) However, under the conditions of the test, a flow of seven liters per minute of liquid nitrogen was required to be maintained for extinguishment of a fire in a 24" X 24" X 12" cell. Such systems would require a constant inventory of a relatively large volume of liquid nitrogen for emergency use.

It may be of interest to note that in the same tests, carbon dioxide was unsuccessfully used to attempt to extinguish the charcoal fire. When the carbon dioxide failed, water was used for final extinguishment.

Water is an optimum fire suppression agent for charcoal fires. It is proven effective on deep seated fires, it is abundantly available, and it is inexpensive.

It has been claimed that water has never been demonstrated effective on charcoal adsorber fires nor have designs for water-based fire extinguishing systems for charcoal fires been proof-tested. These claims are not based upon fact.

An extensive experimental program was conducted to develop an effective water spray system for fire protection of activated carbon adsorber beds. (8) These tests established that the extinguishment by water sprays will vary as a function of the air flow rate, the water injection rate, the nozzle height, the bed thickness, and the preburn time.

The most important factor in extinguishment appears to be the air flow rate. While it is important to maintain airflow for cooling in the pre-burn state, it is equally important to interrupt the air flow once ignition has occurred. Previous attempts by other investigators to extinguish charcoal fires failed because air flow was continued. (6)

The tests established that the water flow rate must be optimized. Too low a water flow rate reduced the saturation time thus reducing the effectiveness of the water spray as a fire suppressant. Too high a water flow rate resulted in an excessive amount of water run-off which would require waste processing without increasing extinguishing efficiency. A water flow rate from nozzles of 3.5 GPM (3.2 gal./cu. ft.) soaked the entire charcoal bed within one minute. The charcoal fire was extinguished within three minutes despite an extended pre-burn period. Repetitive tests clearly demonstrated that water sprays can be used to extinguish charcoal fires rapidly and reliably when properly introduced into the burning medium.

IV. ACCIDENTAL WATER DISCHARGE

As reported by the Department of Energy, automatic, water-based fire suppression systems have an outstanding record of limiting fire damage with an equally outstanding record of a low rate of inadvertent actuations. (9) Fears that automatic systems are frequently subject to unwarranted actuation is unfounded. Nuclear industry data bases for fires and fire systems do not support the position that inadvertent actuations occur frequently. (9)(10)(11)

Within the nuclear industry, INPO and NRC bulletins have perpetuated the perception that serious inadvertent actuations of fire protection systems may be a frequent occurrence. (12)(13)(14) Prompted by the NRC bulletin, a graduate student at the Worcester Polytechnic Institute Center for Firesafety Studies completed a comprehensive study of inadvertent operations of fire protection systems in the nuclear power industry. (15) The resultant thesis included a review of the frequency, causes, and effects of inadvertent actuations.

It was reported in that thesis that the total number of incidents of inadvertent actuations of fire protection systems affecting the safe operation of plants was small. Of those few inadvertent actuations which did occur, less than one-third affected a safety related component in some way. Only one had some potential for affecting more than one redundant, safety related system. Nearly half of the inadvertent actuations resulted from personnel error and, of those, a majority occurred during maintenance and testing.

The thesis cited the low cost of damage resulting from inadvertent actuations as well as the low frequency. EPRI has reported an annual probability of occurrence for the false opening of a deluge valve of 7.85×10^{-4}. (16) The thesis concluded that the low rate of inadvertent actuation could be further reduced by thorough design review of proposed fire protection systems, proper installation and test by qualified personnel, and periodic maintenance by trained technicians. It is further concluded that the problem associated with inadvertent actuations of fire protection systems is not a major one.

V. CONCLUSIONS

The use of activated charcoal in nuclear facilities presents a potential for deep-seated fires.

The defense-in-depth approach to nuclear fire safety requires that if an ignition should occur, fires must be detected quickly and subsequently suppressed.

Deep-seated fires in charcoal beds are difficult to extinguish. Manual fire fighting may be extremely difficult and most common fire suppression agents may be ineffective on these fires.

Automatic water sprays can be used to extinguish fires rapidly and reliably when properly introduced into the burning medium.

The problem associated with inadvertent actuations of fire protection systems is not a major one and it can be further reduced by proper design review, installation, testing, and maintenance. Eliminating automatic fire extinguishing systems for the protection of charcoal adsorbers is not justified.

Removal of automatic fire protection systems due to fear of inadvertent fire protection system operation is a case of treating the effect rather than the cause. On the other hand, properly maintaining automatic fire protection systems will preserve the risk of fire loss at acceptable levels while at the same time reducing the risk of damage presented by inadvertent operation of fire protection systems.

References

(1) Newman, R.M., "Test of Fire Retardency of Charcoal-Filled Gas Filter for Fire Retardency", Laboratory Report Serial No. 16286, Factory Mutual Research Corporation, Norwood, MA, 1966

(2) Burchsted, C.A., Kahn, J.E., Fuller, A.B., Nuclear Air Cleaning Handbook, ERDA 76-21, Oak Ridge National Laboratory, Oak Ridge, TN, p. 57, 1976

(3) Drysdale, D.D., An Introduction to Fire Dynamics, John Wiley and Sons, New York, N.Y., p. 259, 1985

(4) Shaw, J. R., Ph.D., "Fire Retardant and Flame Resistant Treatments of Cellulosic Materials", NFPA Handbook, 15 ed., National Fire Protection Association, Quincy, MA, p. 12-35, 1986

(5) NFPA 12, Standard on Carbon Dioxide Extinguishing Systems, National Fire Protection Association, Quincy, MA

(6) Murrow, J.L., Carbon Adsorber Fire Extinguishment Tests, Lawrence Livermore Laboratory - University of California, Livermore, California

(7) NFPA 12A, Standard on Halogenated Fire Extinguishing Agent Systems-Halon 1301, National Fire Protection Association, Quincy, MA

(8) CVI Technical Note No. 32, A Report on the Extinguishment of Activated Carbon Fires in CVI HECA Adsorber Beds, CVI Corporation, Columbus, OH, 1975

(9) U.S. Department of Energy, Automatic Sprinkler System Performance and Reliability in United States Department of Energy Facilities, 1952-1980, DOE/EP-0052, 1982

(10) Dungan, K.W., Nuclear Power Plant Loss Data, EPRI NP-3179, Electric Power Research Institute, Palo Alto, CA, 1983

(11) ANI Property Loss Data System, American Nuclear Insurers, Farmington, CT

(12) Institute of Nuclear Power Operations, Inadvertent Actuations or Failures of Fire Protection Systems, Draft SOER, INPO, Atlanta, GA, 1983

(13) Institute of Nuclear Power Operations, Inadvertent Openings of Fire Protection Deluge Valves, SER 57-84, INPO, Atlanta, GA, 1984

(14) U.S. Nuclear Regulatory Commission, Actuation of Fire Suppression System Causing Inoperability of Safety-Related Equipment, IE Information Notice 83-41, USNRC, Washington, D.C., 1983

(15) Sawyer, E.A., Failure Analysis of Inadvertent Operations of Fire Protection Systems Including Design Recommendations, Master of Science Thesis, Worcester Polytechnic Institute, Worcester, MA, 1985

(16) Hall, D.T., and Clayton, T.C., Turbine Generator Fire Protection by Sprinkler System, EPRI NP-4144, Electric Power Research Institute, Palo Alto, CA, 1985

DISCUSSION

JACOX: I would like to dwell on two issues. One, you mentioned many times inadvertent actuation but you totally neglected leaks. I know from personal experience over the last twenty years, that leaks are an extreme problem. Admittedly, it does not always happen during operation, but when it does, it is very expensive, given the corrosion that results from the impregnant on the carbon. Therefore, I think any analysis must include leakage through the valves, not just accidental actuation. Second, you addressed in the early part of your paper the ability to halt the ignition source. I would like to differentiate between offgas and air treatment systems. We agree that when there is a failure of a recombiner containing a hydrogen-oxygen mixture, there is a credible ignition source. When we consider nuclear air treatment systems, which represent the vast majority of systems around the world in power plants, I know of no credible way that a fire can start. There is an excellent flame arrester in the perforated stainless steel, there are numerous components upstream that would stop sparks, wherever they may come from. When we consider maintenance caused by welding or flamework, standard OSHA requirements included a fire watch, various manual extinguishers, etc. I admit that there is a very low, but at least an arguable, probability of a fire during maintenance. But during operation, I simply have never seen a credible ignition source over twenty years experience in actual plant systems. Therefore, to me, it is not a realistic approach to require automatic deluge systems because of an infinitesimal threat from an undefined and incredible source of ignition.

HOLMES: I tend to take a more conservative approach by assuming that when you have combustible materials somebody may do something careless that is going to result in a fire. That is a very conservative approach, but I think that is the only way you can expect to have a fire under most reasonable conditions in an occupancy, whether it be a nuclear power plant or a home.

JACOX: I would agree in the abstract, and I certainly agree that it is exceedingly conservative, but I would think that, given the conservatism, one could logically make the same point by backing off from automatic systems and going to manual systems. I think here is where we could get into some statistical and probabilistic countdowns.

HOLMES: I think that statistical and probabilistic studies are worth pursuing further.

KOVACH: I am going to refrain from discussing my disagreements because there is a rather long list and we will try to clear them up during the panel session. But I feel that it is unfair in this context to consider automatic fire control systems throughout an entire nuclear power plant versus strictly looking at the question of air cleaning systems alone. I think that sort of dilutes the issue. If we are evaluating automatic fire control in air cleaning systems, let us look only at air cleaning systems. A number of them have been damaged in about every utility that has automatic water systems. Almost without exception, at least one of multiple units was, in fact, flooded. In some cases, with very serious consequences. I have not read your particular masters thesis, but just on the basis of your abstract I feel that it was an incomplete survey if it shows only a few such cases. The conclusion that somebody reading it draws is that there was little familiarity with what is actually happening in the power industry.

HOLMES: I don't think there is much room for discussion whether the fire suppression systems installed in charcoal should be different than other fire suppression systems. I think fire protection systems for charcoal should be designed just as well as the other fire protection systems in the rest of the plant and should be just as reliable.

GILBERT: Mr. Holmes, I am a bit puzzled by your statement that auto-ignition of a carbon bed from decay heat could occur under low air flow. This might become a matter of how low is "low", or when low becomes nonexistent. During the 1965 to 1970 period, Bob Adams at Oak Ridge National Laboratory, was unable to load sufficient fission products onto a charcoal bed to achieve auto-ignition. Do you know of any more recent results?

HOLMES: I don't think I have anything more recent then that which was quoted in my paper, as previously reported in the Nuclear Air Cleaning Handbook, which goes back to 1976, and includes data from previous Nuclear Air Cleaning Conferences. That is where the data come from. As to how low is "low", I say it is 5 fpm.

ORNBERG: I second Kovach's comments about air cleaning systems and offgas systems. Along that line of thought, do you have any statistics on how many fires or potential fires have taken place, or have been suppressed in air cleaning systems? You talked about the low probability of inadvertent actuation. I think there is a much higher chance of it happening than you do and I have proof that it has been happening and damaging tons and tons of carbon in air cleaning systems, whereas the fires do not take place. I would be interested in knowing what your statistics are.

HOLMES: I could ask the same question in reverse. I hear ghost stories of every utility having twenty or more inadvertent actuations. I hear various sources say there have been over two hundred such actuations, yet I don't see any data that indicate it is factual.

ORNBERG: I will give you one example, Clinton Power Station. Just last week, while getting ready to load the control room carbon for their filter test (right before fuel load) an inadvertent actuation doused all their control room filter carbon.

HOLMES: Perhaps it would be worthwhile, if the information is available, to examine it to see why these events are occurring. Is it because the detection systems are improperly designed? Is it because maintenance procedures are not being followed? Is it because technicians don't seem to know enough to tag out a system or shut a valve when they are going to work on it? I think those are the more important questions.

ORNBERG: Maybe all the complexities we are designing into these systems make them that much harder to operate correctly. If we can reduce the amount of complexities by not having to provide protection for an event that has very little possibility of happening, I think that would be the step we should be taking.

HOLMES: I think we do need to keep it simple, maybe some of these systems have become dinosaurs.

ORNBERG: Second question, on the Factory Mutual tests that you talked about, when were they run and were they on air cleaning systems? What sort of parameters were examined? Did they compare with those typically found in air cleaning systems?

HOLMES: I believe they were 24 x 24 x 12 in. single cells. As to date, I am going to say 1966. Air flow was maintained during the test.

ORNBERG: Other than what the gentleman from Germany discussed, there have been no tests, to your knowledge, on the effect of reducing air-flow on the temperature?

HOLMES: None, other than the water spray tests, which I mentioned, that CVI, Grinnell, and Factory Mutual conducted. They did vary airflow rate, as well as water ejection rate.

REVIEW OF FIRES AND FIRE CONTROL METHODS FOR NUCLEAR AIR CLEANING SYSTEMS

J. Louis Kovach
Nuclear Consulting Services, Inc.
Columbus, Ohio

I. Abstract

The nuclear power industry has experienced four carbon based adsorbent fires in its history, one was of the Monticello Standby Gas Treatment System and other three were in various off-gas delay beds. Although, some of the latter may not be classified as a full fledged fires. There were a number of experiments performed relating to igniting carbon beds and experiments relating to attempts at extinguishing set fires reported in the literature. Review of these experiments indicates that fire resulting from decay heat of adsorbed radioactive iodine is not justified even under the somewhat unrealistic source terms still in effect. At the same time the non-nuclear chemical industry application of carbon base adsorbents for solvent recovery has resulted in numerous fires and significant property losses.

Fire control systems installed in nuclear air cleaning systems in the US consists of water deluge. Accidental initiation of these systems has occurred in excess of one hundred times in US plants, causing safety problems and non-availability of critical engineered safeguards.

The current stance of the US NRC Regulatory Guide 1.52 is that water deluge is not a requirement for nuclear air cleaning units. Although simce the last NRC reorganization, there is confilict within the NRC divisions regarding water deluge for air cleaning systems. Water delute system installation and maintenance has been a requirement from the insurance companies, based mainly on experience in entirely different systems and not based on experience of fire losses in nuclear air cleaning units.

The events leading to the past nuclear air cleaning system fires and several chemical industry adsorption system fires are reviewed.

Experimental data supports that;

a) initiation of water spray in case of radioiodine loaded adsorber rather than cooling adsorber and retaining radioiodine will release already adsorbed radioiodine.

b) many of the current methods used to signal fire would not indicate fire detection early enough.

c) initiation of the water sprays will not necessarily extinguish the fire.

d) there is no justification for the installation of automatic water deluge units for safety class nuclear air cleaning systems.

e) alternate, safer and more reliable methods such as isolation, inerting, manual spray, etc., should be evaluated for the very remote possibility of fires.

f) the current practice of temperature tripped water deluge when evaluated in plant context can be, in fact, a safety hazard.

II. Review of the Nuclear Industry Adsorbent Fires.

Fire 1) Monticello (BWR) air cleaning unit fire (1)

Standard tray type adsorbers were equipped with heating elements to prevent condensation in the carbon (which event would lower methyl iodide removal efficiency). The electric heaters directly attached to the trays heated the carbon above its ignition point. The air flow was stopped and the trays removed from the housing. The fire was extinguished externally using water.

Fire 2) Browns Ferry Off-Gas System Fire (17 July 1977) (2)

From the initial startup in September 1976, until June 1977, the unit 3 off-gas system appeared to operate properly. On June 17, 1977, the unit was shut down for a forced outage. From the time the unit was returned to service on June 20, 1977, until July 8, 1977, off-gas system performance appeared to be slightly abnormal. On July 8, 1977, the unit tripped due to high temperature in the main steam vault and remained shutdown until July 15, 1977, due to high river water temperature, technical specification limits, and cooling tower problems.

The restart of unit 3 on July 15, 1977, was the beginning of a series of events that resulting in the carbon bed temperature excursion. From the time the plant started up on July 15, at 2100 hours until 1200 hours on July 17, the off-gas recombiner was not working properly. The off-gas system received excessive moisture and a combustible mixture of hydrogen and oxygen. At 1200 hours on July 17, with unit 3 operating as 560 MWs, a transfer from the A steam jet air ejector (SJAE) to the B SJAE was executed. At approximately the same time, off-gas flow to the holdup line pegged offscale at greater than 300 scfm (10 times the design value), off-gas reheater inlet temperature spiked from 47 degrees F (normal value) to 81 degrees F, off-gas reheater outlet dewpoint pegged offscale at greater than 100 degrees F (approximately 50 degrees F above normal), and all of the off-gas carbon adsorber bed temperatures spiked in the range of from 4 degrees F to 25 degrees F above the normal bed temperature of 68 degrees F.

From 1202 hours until 1230 hours on July 17, the B SJAE was in service with the off-gas system receiving (1) excessive dilution steam flow due to the malfunctioning third stage SJAE pressure regulator and (2) a combustible mixture of hydrogen and oxygen due to lack of recombination. At approximately 1230 hours a transfer from the B to A SJAE was executed and various system parameters showed spikes or were pegged offscale.

From 1232 hours until 1400 hours on July 17, the A SJAE remained in service, the recombiner continued to operate with low bed temperatures, and the off-gas system continued to receive excessive moisture and a combustible mixture of hydrogen and oxygen. At approximately 1400 hours the drain line on the A recombiner was opened, and by 1420 hours the recombiner bed temperature had increased from 220 degrees F to 430 degrees F indicating that the recombination process had begun again. At approximately this same time (1420 hours) various system parameters spiked and pegged offscale.

System parameters remained essentially constant from 1430 hours on July 17 until 1630 hours. At this time the "B" carbon bed temperature pegged offscale at greater than 150 degrees F. Temporary instrumentation (i.e., potentiometer) installed shortly thereafter indicated the "B" carbon bed temperature was 500 degrees F and increasing steadily. At approximately 1900 hours on July 17, this temperature was still increasing, and steps were initiated to isolate the carbon beds and to continue unit 3 operation with the off-gas passed directly up the stack after holdup. The stack emission rate was still very low, approximately 5 percent of the technical specification limits for all three units. Unit 3 remained in operation at approximately 560 MWs throughout the event.

The "B" carbon bed temperature continued to rise, so at approximately 0430 on July 18 a decision was made to begin a nitrogen (N_2) purge on the carbon beds. At approximately 0520 the site initiated an N_2 purge through the "B" and "C" beds using onsite N_2 bottles connected to the carbon bed drain piping located outside the adsorber vault. The purge was begun at a 5 cfm rate and increased to 10 cfm and subsequently to 20 cfm. By 1300 on July 18 the N_2 purge had reversed the temperature excursion by reducing the "B" bed temperature from 1060 degrees F to 1000 degrees F. At this time, preparations were begun to enter the adsorber vault room. The vault would normally be considered unaccessable due to high radiation levels. However, since the unit had little operating time the fuel was relatively "clean" and fission product, activation and radiolytic gas generation were minimal. Therefore, the radiation levels within the adsorber vault were also minimal. Subsequent entry and inspection revealed the following:

1. The middle 1/3 of the "B" vessel had discolored paint. Few, if any, paint blisters were visible. The "B" vessel in this area was warm to the touch. Contact pyrometer readings indicated the skin temperature was 125 degrees F.

2. The "D" vessel surprisingly had a discolored area around the very bottom of the vessel. The paint in this area was visibly oxidized and charred in the shape of a concentric ring around the bottom of the tank. This area was hot to the touch. Contact pyrometer readings indicated that the temperature was approximately 140 degrees F. None of the other ("A", "C", "E", and "F") beds were warm to the touch. Upon recognizing the condition of the "D" bed, the N_2 purge alignment was shifted to include the "D", "E", and "F" beds. Also, at this time the N_2 supply was shifted from onsite bottles to tube trailers which had been summoned from offsite.

The N_2 purge continued in this alignment and at 20 cfm flow rate until 0900 on July 27. During the period from 1515 on July 19 until 0715 on July 20, temperature readings taken once an hour at various locations on each vessel indicated that a temperature wave was moving through the beds in series due to the 20 cfm purge flow rate. The N_2 purge was terminated July 27 after all internal and external temperatures had stabilized at near normal values.

After termination of the N_2 purge, the charcoal beds were opened and inspected for carbon damage. None was found. Samples were sent to Nuclear

Consulting Services, Incorporated, for analyses. The NUCON analyses received August 8, 1977 showed that carbon samples were within specification on particle size, percent moisture, percent ash, flashpoint, and ignition temperature. Metallurgical inspection of the carbon vessels revealed that the maximum vessel skin temperature attained during the event was approximately 500 degrees F and that no structural damage had been incurred.

Physical damage was observed on a prefilter element and an after filter element.

Fire 3) KKP (Philippsburg) Off Gas System Fire (14 Sept 1984) (3)

On September 14, 1983, the following incident occurred in the Nuclear Plant at Philippsburg 1.

The train A steam jet air ejectors (SJAE), preheater and recombiner were switched on in parallel with the already operating drain B of the off gas system to detect air in-leakage in the condenser. Eight minutes later, the H-concentration set-point (3 v%) was reached as indicated at an alarm in the control room. Subsequently train A was taken out of service. Train A operated less than 20 minutes.

Four hours later, the temperatures in the first charcoal adsorber beds reached the alarm point of 50°C. An inspection of the charcoal vessels was ordered.

After another four hours, bubbles were noticed in the paint on the vessel wall. Plant shut down was initiated. At the beginning of shut down the temperature detector for the first vessel indicated 130°C. During shut down a maximum surface temperature of 480°C was measured for the vessel.

First the vessel was cooled by blowing air on the surface. After plant shut down, SJAEs were shut off, the charcoal beds were isolated and nitrogen was fed into the beds to extinguish the slow burning fire. When this method was not successful, carbon dioxide was fed into the beds to fight the fire and cool the beds. Because of the low thermal conductivity of the charcoal and the low heat-capacity of the carbon dioxide, the temperature in the middle of the vessel decreased very slowly. Therefore, the first vessel was bypassed and its inlet and outlet were sealed.

The malfunction of the off-gas system was caused by a defective condenser drain at the preheater of recombiner A. The condensate in the preheater could not drain and prevented proper heating of the preheater by auxiliary steam.

The mixture of steam, air in-leakage, and radiolytic gases (H_2, O_2) coming from the steam jets was not preheated, so that the recombiner did not ignite because of low temperature and high moisture. As a result the radiolytic gases were not recombined to water.

The SJAEs also provide sufficient steam to dilute the hydrogen to a concentration less than the 4% flammability limit. Since the steam was condensed in the condenser and the undercooler, and the non-condensible gases H and O_2 had not been combined, the ignitable mixture was fed into the off-gas system charcoal beds.

Several possibilities for root cause of the ignition appear possible:

- Ignition of the charcoal because of recombination of the oxygen and hydrogen on the carbon surface. This cause appears most likely. Although the charcoal ignition temperature in air is about 200°C, the charcoal of the first vessel was assumed to have a lower ignition temperature because of impurities accumulated from years of operation. Recombination of hydrogen and oxygen has lead to local overheating and then to a slow burning fire.

- Ignition of the charcoal by detonation or explosion of the ignitable H_2/O_2 mixture in the off-gas system. Evaluation of the trend records and the plant computer output indicated that such an explosion might have happened. The recombiner or the H detector may have provided the ignition source. For this possibility, the flame front from the detonation must penetrate the sand-prefilter to ignite the charcoal.

Fire 4) The Perry NPP Off Gas System Fire (19 June 1986 and 6 July 1986) (4)

The Perry NPP Off-Gas System is a 0°F vault temperature delay bed. The vault refrigeration system acceptance test was completed in May 1985. The test showed that the refrigeration system was capable of achieving temperature pull down from 150°F to 0°F with charcoal filled into the vessels in less than 20 hours. However, a field change request was generated to heat the contents of the vault (i.e. the filled carbon vessels) also to 150°F, simulate process flow through vessels and check the pull down time from 150°F to 0°F.

To accomplish the heat up of contents 4 each 6 KW and 2 each 13.5 KW infrared heaters were lowered into the vault. The testing commenced on 18 June 1986 and the sequence of event was as follows:

SEQUENCE OF EVENTS

EVENT 1

June 18	1100	Initial heatup commenced
June 19	0600	Instrument air flow initiated
	2041	14B center T.E. >250°F
	2110	All heaters de-energized
June 20	0100	Started off-gas vault refrigeration system in pull down mode
	1145	Unusual event declared, Instrument Air Flow Stopped
	1219	N_2 purge initiated
June 20 June 21 > June 22		N_2 purge and vault cooling continued
June 23	1100	All T.E.'s reading <250°F
	1125	Unusual event terminated

June 27	1852	N_2 purge secured with N_2 blanket retained sampled 14A & 14B vessels through top, and vessels resealed. Restarted N_2 purge.
June 28	0958	N_2 purge secured with N_2 blanket retained.
July 2		Sampled 12B & 14B vessels through bottom T.E.'s
July 3		Sampled 12A vessel through bottom T.E.

EVENT 2

July 3		Vessels remaining under N_2 blanket
		Preliminary metallurgical and chemical test results satisfactory
		NRC concurrence obtained to retest from ambient temperature conditions
July 6	1800	Instrument air flow initiated
	1910	Center T.E.'s on vessels 14A & 14B indicate greater than 250°F
	1942	Instrument air flow secured
	2037	Center T.E. on 14A reading 656°F and center T.E. on 14B reading 578°F
	2037	Unusual event declared
	2307	N_2 purge initiated
July 7-8		N_2 purge continued - temperatures dropping
July 8	1645	Unusual event terminated.

ROOT CAUSE EVALUATION

EVENT 1

0 Ignition temperature of 14A & B vessel charcoal determined to be as low as 307°F and 428°F respectively, at the system air flow rate of four (4) lineal feet per minute.

*** Charcoal was overheated during preparation for testing, resulting in combustion. Calculation of the temperature at various distances from the 13.5 KW heater were:

1 foot	1200°F
2 feet	1050°F
3 feet	700°F

EVENT 2

0 Charcoal thought to have been cooled to ambient temperature as indicated by vessel thermocouples.

0 Nitrogen purge/blanket removed and instrument air readmitted.

0 Combustion again occurred within one (1) hour.

*** Vessels contained hot spots that led to reignition upon reintroduction of oxygen (instrument air).

Paint blistering was observed at several vessel locations. Analysis of the data indicates that definite fire existed in vessels 14A and 14B with limited fire in vessels 15A and 15B.

Metallurgical inspection of the vessel indicated no vessel damage. The carbon was removed from all of the vessels and replaced with new carbon.

III. Review of Fire Causes and Events

The evaluation of the events leading to the fire can also be made in the following categories.

1. Original design or procedure related (Fires 1 and 4)
2. Upstream equipment failure initiated (Fires 2 and 3)

In fires 2, 3, and 4, long delays existed between the initial event causing carbon oxidation and the termination of the air flow. Feeding air through an oxidizing or fully burning carbon bed is one of the worst actions. Mankind discovered iron smelting using carbon, thousands of years ago and it is not required to rediscover this event in nuclear power plants. Whenever carbon fire is suspected, the vessel or unit should be isolated and any air entrance to the isolated unit prevented.

Fire 1 was extinguished by adsorber removal and external quenching.

Fire 2 was extinguished by nitrogen purge.

Fire 3 was extinguished by isolation and consumption of available oxygen.

Fire 4 was finally extinguished by nitrogen purge.

None of the nuclear power related fires were caused by organic compound accumulation on the carbon bed. Typical "used" carbon samples removed from air cleaning systems show ASTM (5) ignition temperatures near or in excess of "new" carbon ignition temperatures. However, most of the systems are operated at much lower velocities than those used in the ASTM test and the typical bed depths are also larger.

Both of these deviations from the ASTM test conditions result in lower ignition temperatures (6)(7). When alarm set points are selected and process parameter ranges established, the ignition temperature of the carbon bed should be determined under worst case and not best case conditions.

Due to the low thermal conductivity of the carbon, in the adsorbent bed temperature measurements are not reliable indicators of carbon oxidation or fire. Temperature measuring devices should be placed in the inlet and outlet gas streams of the adsorber units and backed up by either CO or CO_2 monitoring units. (8)(9)

The uniform packing of the carbon bed also influences the uniformity of both heat and mass transfer. Improperly designed adsorbent bed underdrain supports result in "dead spots" in the carbon beds which enhance ignition probability and prevent the effectiveness of adequate extinguishing procedures.

When evaluating the behavior of carbon - particularly in off gas systems - the basic tenets of adsorption processes should be known, i.e. that all gases progress through the carbon bed by adsorption and desorption, and that the carrier gas components are also adsorbed on the carbon.

Thus if in a four bed series system, it fire occurred in bed 1 and 3 and then nitrogen purge was initiated, the CO detection at the system outlet will not be instantaneous because the CO has to penetrate through bed 4 first. After a certain time (depending on flowrate, temperature, bed size, etc.) the CO concentration shall rise and then fall to zero, if, for example, the bed 3 fire was extinguished and some later time it will rise again when the CO generated in bed 1 progresses through beds 2, 3, and 4. Therefore, if CO or CO_2 monitoring is used for fire detection only at the outlet of large carbon beds in series, fires occurring in the first bed may not be detected in time.

As an example, using the dynamic K values common for Kr, Xe delay bed design the following approximate K values can be used for other gases.

Xenon	1200
Krypton	60
Carbon Dioxide	50
Carbon Monoxide	18
Oxygen	14
Nitrogen	12

If under system conditions a 40 hour delay of krypton occurs, a five hour delay of nitrogen can exist when the flow is switched to pure nitrogen. Hot spots generate their local pressure boundary and the normal purge flow condition penetrates the hot spot inadequately to extinguish the fire. Therefore, false "extinguishing" of the fire can occur and upon resumption of the air flow reignition can take place.

The frequent lack of bypass capability of off gas system delay beds also means that the heat generated by fire in an upstream bed can be driven into subsequent beds and in turn igniting these adsorber beds also.

Hydrogen as a fire preventer agent. In several carbon based off-gas systems, hydrogen detonation took place instead of partial hydrogen burn or deflagration. In none of those cases did a carbon fire result because most of the oxygen was consumed by the hydrogen detonation and the residual O amount was insufficient to initiate carbon fire. (10)(11)(12)

Naturally, the lower is the temperature of the operation, the larger is the quantity of oxygen physically adsorbed in the carbon which has to be displaced in case a fire is to be extinguished. (13)

IV. Differences Between Solvent Recovery & Air Cleaning Applications

Activated carbon is used in many diverse air cleaning applications. One of the classification into subcategories can be done by the level of inlet organic concentration to the adsorbent. Inlet concentrations above 100 vppm of flammable solvents, particularly ketones, in regenerative systems has resulted in many fires in the past. Most of these fires are caused by incorrect process and equipment design. Activated carbon fires in low concentration air cleaning systems are very rare. The understanding of the adsorption, the solvent decomposition and the carbon oxidation process has to be understood and the properties of both the adsorbent and the adsorbate (the compounds adsorbing on the carbon) properties known, to permit a correct evaluation of the system fire hazards. Ignoring any of these parameters will result in the installation of either unnecessary or incorrect fire control methods.

Nearly all of the activated carbon based solvent recovery fires occurred with ketones which can be catalytically decomposed by carbon (to a varying degree depending on carbon type). The normal ketone inlet concentrations were in excess of 1000 vppm and the carbon was hot steam stripped periodically to remove the adsorbed ketones. If the adsorber bed design is such that "dead spots" i.e. areas with low air velocity exist, the carbon bed temperature can reach the ignition temperature of the solvent and subsequently that of the carbon. If sufficient oxygen is present without any heat removal means, the result is a runaway ignition. (14)(15)

These conditions are not normally present in nuclear air cleaning systems. First of all, the solvent quantity adsorbed on the shallow (2-4 inches deep) carbon beds never approaches that of solvent recovery system loading, even at the 100 vppm level versus the 1000+ vppm levels of solvent recovery systems. The heat loss of the shallow beds to the air stream is always higher than that of the 2-3 feet deep carbon bed solvent recovery units. (16)(17)(18)

The nuclear air cleaning adsorbers are not periodically "regenerated" which eliminates the temperature cycling of 100+ C temperature rise to the adsorbent.

The typical "dead spot" unperforated areas of nuclear adsorbers is on the order of 1-2 inches versus the 4-16 inches of blanked off areas of solvent recovery units.

The adsorbent used in solvent recovery systems is not damaged by wetting, thus the carbon is left in place and no significant economic or process loss exist if the adsorbent is wetted.

The solvent recovery system does not serve a per se safety function. Failure of the unit or its non-availability does not create a significant safety hazard to the public.

V. Similarities Between Solvent Recovery & Air Cleaning Applications

Numerous solvent recovery systems had fire propagation to significant safety and economic loss level when fire detection method was by temperature rise detection. Fire in a small area of the adsorbent will not generate high enough temperature rise to be detected early enough by temperature alone.

A solvent recovery fire which melted the exhaust duct was not detected by 32 thermocouples installed in the midlevel of a 3 feet deep (30' X 8' area) bed, because the fire started downstream or not in immediate vicinity of any of the installed thermocouples.

Nearly all nuclear air cleaning fire detection methods for carbon beds are temperature based. In some cases, with the thermowells installed in the carbon beds themselves.

Water sprays installed in solvent recovery systems for fire control were found ineffective when incomplete water spray coverage exist, even for a single horizontal (slab shape) carbon beds.

The nuclear air cleaning system fire control water sprays almost always result in incomplete coverage of the individual adsorbent trays.

Solvent recovery systems vessels were found to fail from fire control water pressure due to inadequate drainage and low pressure housing design. The drainage systems and housing pressure design of nuclear air cleaning systems is rarely matched to the pressure of the fire control water. Water penetration through ducts to critical areas can occur resulting in significantly higher safety hazard due to water transfer to unplanned areas, than the presumed fire in the air cleaning system.

VI. Specific Nuclear Air Cleaning Related Problems

1) The adsorbed radioiodine is released from the carbon before it reaches its ignition temperature.

2) Introduction of water to carbon at 130°C results in an initial temperature rise of 50-80°C due to the heat of adsorption of water (~31,000 J/mole).

3) The adsorbed and converted iodides are water soluble and the radioiodine will be transmitted by the water to tanks where, depending on pH conditions, the iodine can be released into uncontrolled areas.

4) Accidental (or incorrectly designed) initiation of the fire control sprays destroys the adsorbent (requiring replacement). The wetted carbon bed per se is not available for its design safety function.

5) Standard ignition test procedures in both the US and Europe where the carbon is forced to an ignition temperature by heating, use nitrogen flow to extinguish the fire. The CO, CO and temperature levels start to drop within 10 seconds of nitrogen flow initiation. (19)(20) Fires which occurred in well isolatable carbon beds have been extinguished by nitrogen flow.

6) Isolation of nuclear air cleaning systems and consumption of only the available oxygen in the housing may not be adequate due to inadequate damper and housing leak criteria.

7) Standard (ASTM) ignition temperature determination is performed on a 1.0 inch deep bed at 100 fpm which results in significantly higher ignition temperature than 2-4 inch bed depth at 40 fpm. (6) Ignition temperature of nuclear adsorbent should be determined at design conditions.

8) No fires in nuclear air cleaning systems extinguished by the installed water deluge systems.

9) There were in excess of 200 nuclear air cleaning units damaged by accidental initiation of water sprays.

10) Regulatory Guide 1.52 Rev 2 does not require water sprays for adsorber beds.

11) The ANI recommends water sprays for adsorber beds (21), verbal communication with ANI indicates that proof tests are needed for alternate fire control methods, even though no proof testing of water sprays was performed.

12) Most moisture separators (if not all) will prevent burning debris from reaching adsorber stages.

13) HEPA filters are more susceptible to fire damage than activated carbon beds.

14) The lowest ignition temperature of carbon beds occurs if they are heated in no air flow and then air (oxygen) is introduced. (22)

15) Uniform packing of adsorbers assures not only uniform mass transfer, but also uniform heat transfer.

REFERENCES

1. Personal Communication from W. Fair to J. L. Kovach (1972)

2. Crisler, H.E. "July 17, 1977 Off Gas System Charcoal Adsorber Bed Fire at Browns Ferru Nuclear Plant". OECD CSNI Report 83, USDOE Report LA-9911-C (1983).

3. Kotthoff, K. and Simon, M. "Presentation of Selected Incidents in German Nuclear Power Plant". NEA/IAEA Meeting for the Exchange of Information on Incidents in Nuclear Power Plants. Paris, July, 1984.

4. Perry Nuclear Power Plant Docket Nos. 50-440; 50-551 "Off Gas System Unusual Events", Final Report. 29 July 1986.

5. ASTM D-3466 Standard Test Method for Ignition Temperature of Granular Activated Carbon.

6. Kovach, J.L. and Green, J.E. "Evaluation of the Ignition Temperature of Activated Charcoals in Dry Air". Nuclear Safety Vol. 8 No. 1 pp. 41-46 (1966).

7. Freeman, W.P. et.al. "Ignition Properties of Nuclear Grade Activated Carbons". OECD CSNI Report No. 83, USDOE Report LS-9911-C (1983).

8. Factory Mutual Technical Advisory Bulletin 7-2 FMELPC Apr. 1979.

9. Kovach, J. L. "Operation and Maintenance of Adsorption Systems". NUCON Report.

10. Vermont Yankee IOG Operation 1973. Personal Observations.

11. Verna, B.J. "Off Gas System Explosions Parts 1, 2, 3". Nuclear News. Nov. 1976, Jan 1977, March 1977.

12. Hydrogen Chemistry Studies at Dresden Plant. 1984.

13. Dubinin, M.M. "Surface Oxides and Adsorption Properties of Activated Charcoals". English Translation in US Report AEC-TR-3750. (1957)

14. Boiston, D.A. "Acetone Oxidation". British Chemical Engineering. Vol. 13 No. 1. pp. 85. (1968)

15. Bratzler, K. "The Autoignition of Carbon Products as a Process Problem". Chem. Ing. Techn. Vol. 28 No. 8/9 pp. 569 (1956)

16. Bernard, E.A. and Zavadoski, R.W. "The Calculation of Charcoal Heating in Air Filtration Systems". USAEC Report CONF 740807. (1974)

17. Shields, R.P. "Ignition of Charcoal Adsorbers by Fission Product Decay Heat". USAEC Report CONF-700816 (1970).

18. Lorenz, R.A. "The Behavior of Highly Radioactive Iodine on Charcoal". USAEC Report CONF-740807 (1974).

19. Chevalier, G. et.al. "Thermal Evaluation of Activated Carbons Used in the Nuclear Industry". French Report CEA-N-2285 (1982).

20. Hardman, J.D. et.al. "Studies of Spontaneous Combustion in Beds of Activated Carbon". Fuel Vol. 59 pp. 151-156 (1980)

21. Personal Communication from American Nuclear Insurers (ANI) to J.L. Kovach (1985)

22. Kovach, J.L. "Evaluation of Activated Carbon Ignition Under System Application Conditions". NUCON Report 240 (1986).

Table 1

Fire Extinguished by

Fire 1	External quenching by water
Fire 2	Nitrogen purge
Fire 3	Isolation
Fire 4	Nitrogen purge

Table 2

Time After First Fire Indication when

		Airflow Stopped	N_2 Purge Start
Fire 1		<0.25 Hrs.	N/A
Fire 2		5+ Hours	15+ Hours
Fire 3		8+ Hours	10+ Hours
Fire 4	Event 1	13 Hours	13.75 Hours
	Event 2	.5 Hours	4.5 Hours

Table 3

First Remedial Action

Fire 1	Remove adsorber
Fire 2	Bypass and isolate
Fire 3	Blowing air on vessel exterior
Fire 4	Vault refrigeration started

Table 4

Assuming All of Air Reacting

Air Flow	Carbon Consumed lb/m	BTU* generated per minute
5 CFM	.0326 lb/min	460 BTU/min
10 CFM	.0652 lb/min	920 BTU/min
20 CFM	.1304 lb/min	1840 BTU/min
40 CFM	.2608 lb/min	3680 BTU/min
80 CFM	.5216 lb/min	7360 BTU/min

*assuming $C + O_2 = CO_2$ reaction

Table 5

Oxygen Present Per Kg of Carbon*

At 25°C	1.75 g
At 0°C	2.35 g
At -20°C	3.65 g
At -185°C	560 g

Typical quantity of carbon in Off Gas Systems 25,000 to 40,000 Kg.

*calculated on basis of standard air passing through 800 m^2/g surface area coconutshell carbon.

Table 6

Heat Transfer Related Carbon Properties
Coconut Carbon

Average Particle Size	0.128 cm
Average Particle Surface	0.10 cm^2
Average Particle Volume	0.00205 cm^3
Average No. of Particles	280/cm
Bed Void Fraction	0.42
Sphericity	0.719
Carbon Thermal Conductivity	3.1×10^{-4} cal/cm sec °C
Carbon Heat Capacity	0.20 cal/g °C

Table 7

Fire Location/100 year reactor operation*

Reactor Building	40
Auxiliary Building	38
Service Building	34
Turbine Building	45
Diesel Gen. Bldg.	3
Other Bldgs.	15
Building HVAC Systems Affected	7

*Canadian Data

DISCUSSION

NESBITT: As stated, the first action in the event of a suspected fire in a charcoal delay system should be to stop the supply of oxygen. Residual oxygen in the system will not support combustion more than a very few hours. Purging with nitrogen will reduce the amount of oxygen in the system and shorten combustion duration as well as aid in cooling. Even with no added heat of combustion, as evidenced at Perry, due to the low charcoal thermal conductivity, the bed could stay hot for weeks.

As an example of the difficulty in cooling following a fire, calculations indicate that a single hot spot in a bed that is being nitrogen purged at 6 ft/min will migrate at a rate of ½ to 1 ft/day. The much higher heat loss from the interconnecting pipes terminates the migration at a connecting pipe with no more than one tank involved, i.e., in the Perry case, a maximum of one twenty foot high tank in each train, or about twenty days.

Experience with three delay tank fires supports my statement that the best charcoal delay system fire detection method is a smoke detector in the charcoal vault exhaust duct.

GILBERT: Mr. Murrow, do you recall if the experiments at Livermore used any "wetter water" as an extinguishing agent?

MURROW: At times we did use a wetting agent in the water. We had a number of different spray patterns and we tried to get water into all parts of the absorber. It didn't help.

PEDERSON: I work for the Office of Inspection and Enforcement and we manned the incident response center for the whole weekend of the initial Browns Ferry fire. Needless to say, there was a lot of concern by my management, because the plant couldn't readily put the fire out. It was pretty tight.

MEANS USED TO MAKE SURE THAT THE CONFINEMENT IS MAINTAINED IN CASE OF FIRE

J. SAVORNIN, head of Fire Protection Section
CEA - IPSN - DAS

Abstract

In case of fire in nuclear facilities, the ventilation and filtration systems have to be designed to exclude any radioactive release to the environnement. For this purpose it's necessary to maintain the extraction ventilation running. As a result, smoke ant heated air will be introduced into the ducts towards the HEPA filters.

Consequently we must know the threshold from which, either filters will stop going their part, either ducts will be partly destroyed.

Tests and experiences are led or provided to give answers to those questions. Informations are provided about tests already drawn and scheduled with the two available apparatus.

I. INTRODUCTION

In a nuclear facility, the units which are very radioactive are separated from the environnement by confinement barriers of which one at least would remain efficient in case of fire. The principles which have been set and the means which have been suggested have often been exposed and espacially in the paper which I have presented at Los Alamos in April 1983 at the CSNI meeting.(1)

The ventilating systems are the weakest parts as regards fire risks and consequently the level at which they would be destroyed must be determined. Experiments are drawn by several services at CEA to try to obtain exact answers to those questions.

I'll present you the chief places where there is a major problem and, for each of them, the experiments which are presently carried out or scheduled.

II THE WEAK PARTS OF A VENTILATION SYSTEM

In the event of an important fire inside a fire zone the exctracting ventilation system of which is organised with the principle of double filtration with dilution (figure n°1), several problems exist due to smoke and very hot gaz circulating in the system. Those problems are presented here under following the stream of air flow (figure n°2).

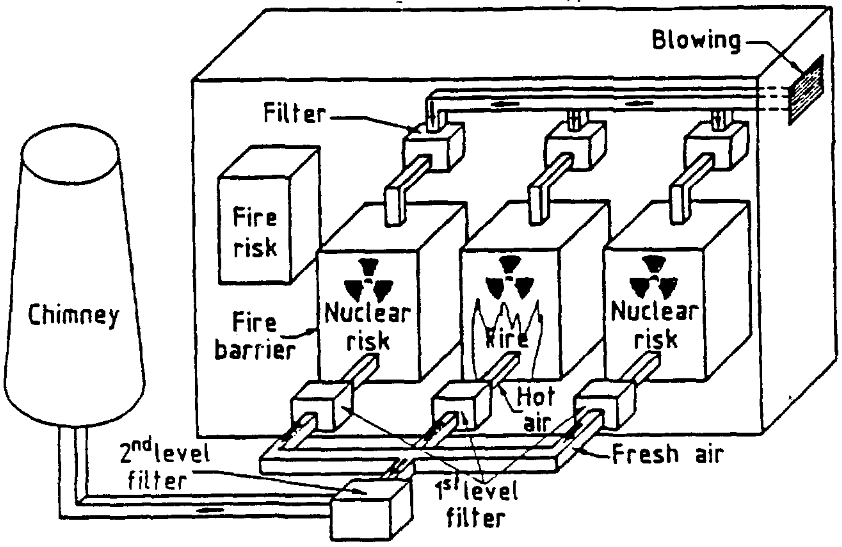


Fig. 1 - DRAWING OF THE VENTILATION SYSTEM OF A NUCLEAR FACILITY WITH FIRE ZONES AND DOUBLE FILTRATION WITH DILUTION

2.1 Duct crossing through a fire barrier

The iron duct crosses the fire barrier through a hole where it supports on the interior face of the fire barrier, a fire whose developpement is defined to follow the program for the temperature growth specified by the ISO norm. It is represented on the figure n°3 which is designed by the relation $T-To = 345 \log (8t + 1)$ where :

T is the temperature at the "t" time
To is the starting temperature

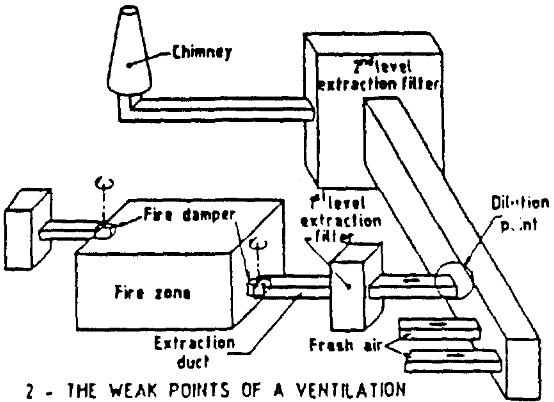
If the thermic insulation given by the material across the fire barrier were perfect, the overheating due to fire would be transmitted in totality, by conduction way, to the part of duct outside of the fire barrier so that the duct may be spoiled or destroyed as soon as the temperature rises to 600°C. This needs a series of tests driven with an oven where they may run the ISO programm. We feel safer since we made a test of this kind about another experiment but we need a global series of experiments about this problem, so as to define the right data to know the permissible level.

2.2 Fire dampers

Fire dampers settled on the extracting air apertures are currently built to shut down at a rather low temperature by the mean of a device fusing towards 70°C.

On the contrary, when the ventilation net is designed with dilution and double filtration system (figure n°1), the fire damper will work only at the moment when the first level filter is just to be spoiled. Since many HEPA filters have a rather good behaviour at high temperatures, and since as the distance between the fire damper and the concerned filter is long, so the fire damper may be worked at very high temperatures which may rise to 500 or 600°C.

We have to make sure that the chosen fire dampers will shut down at the temperature that they may have to undergo. The tests will be made by using an oven designed to draw the temperature programm according to the drawing n°3.



2 - THE WEAK POINTS OF A VENTILATION SYSTEM IN CASE OF FIRE

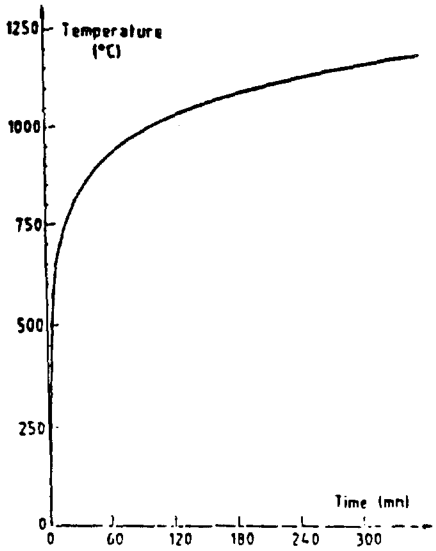


Fig 3 - RISE OF TEMPERATURE IN TERMS OF TIME ACCORDING TO THE ISO NORM

2.3 Ventilation ducts

Fires may initiate inside ventilation ducts where dusts are deposited and often covered by greasy films of oils of many apparatus, among which you find health physics controls. This risk grows together with the age of facilities. Information concerning the way followed by the fire in a ventilation duct, and about the fighting methods have to be collected. The schedule of experiments driven would give good answers.

Fire will be put inside a rig placed at a position depending of the filter, of the dilution point, and of the blowing or extrating fan. The material which is to be burnt will lay on the bottom.

The material which is to be burnt is chosen to comply with the previoused test ; it may deliver either smoke, or heat, or both. Figurative deposit is currently used which has been defined to be similar to the one to be found in a particularly dirty duct. For 65% it is made of three mixed componemts : 50% lucolfex, 25% wood, 25% cotton.

The combustible material will be put in the loop, ventilator working. Fire will be put upstream, then downstream with various air speeds, so as to define wether the fire comes upstream or not. Tests will be made to find the extinguisher appropriate and the mean to apply it.

Other tests of duct fire behaviour will be done by connecting the rig to a little fire room where high temperature will be obtained. The temperature gradient will be measured along the duct and compared with the calculated results.

2.4 Filters

Filter may be submitted either to a hot air stream coming from a room fire, or to a fire of dusts covering it, or both. Experiments concerning those topics are running. Few tests concerning extinguishing means have also been realized.

A covered with dust and alcohol wetted filter is settled in its box whose front part is fitted with two apertures to introduce the extingtuisher, which can be applied in the duct also through several other openings at several distances ahead. The ventilator is running.

Fire is put inside the loop at several different distances upstream of the filter previously covered with the figurative deposit, so as to appreciate the risk of having the fire running from the duct down to the filter. Fire extinguishing tests will be carried on.

A research of convenient fire detectors inside the ducts has to be driven with this opportunity.

2.5 Dilution point

The dilution principle means that the ventilation duct conveys hot air, smokes and unburned gases up to a chief duct in a stream of fresh air. It is an evidence that you find ther a fire or explosion risk due to unburned hot gases melted to a large amount of oxygen.

We have to know if it is an actual risk and in case of an affirmative answer it will be a necessity to point the level where the risk begins. A series of experiments on this topic will be driven.

The piece of loop where the material is burning, will be brought nearer and nearer to the dilution point during several tests while increasing temperature and amount of unburned gases. A pilot flame may be burning at the dilution point.

III AVAILABLE EXPERIMENT DEVICES

3.1 Objectives

Two experiment devices are available in FRANCE for tests of ducts and filters at high temperature. The biggest is the SIMOUN rig (2,3) built on the Saclay plant to test filters in air stream at temperature so high as 400°C. This apparatus cannot be ed with fire or smoke.

The other is the BEATRICE rig which was formerly built on the Marcoule nuclear plant, then recently taken to SACLAY to be close to the facilities of DPT/SPIN which has taken it in charge in 1985. It has been designed to carry out fires in vent ducts. We wanted it to be a flexible tool which will be convenient with every kinds of tests on fire and smoke. Repairing has to be quick and cheap.

3.2 Description of the BEATRICE rig (Figure n°4)

The loop is made of pieces which are all similar. They are all two meters long with a square cross section of 400 x 235 mm fixed together with flats joins. Those pieces are built with blackiron plate 2mm thick. They are covered with a plate fixed with join-pressers so as to be easily taken out. On the covers, controllers and experiment apparatus are disposed. You may have window on it. The elements are cheap ; they may be replaced transfered, inserted, and so on without problem. Ventilators fiter box and dilution point cannot be removed easily.

Ventilation is made by the means of a fan blowing 3000 m3/h so as it will not be harmed by smokes and hot gases. When experiments want that the duct is exhausded the fan can be deplaced at the other end of the rig.

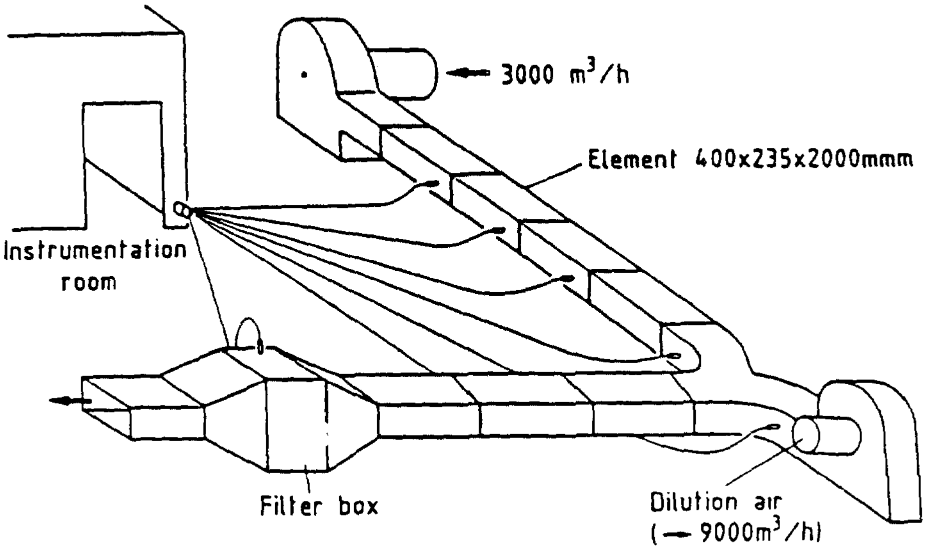


Fig. 4 - THE BEATRICE RIG

An other fan may blow up to 9000 m/3h at a point of the loop to represent the dilution. The speed of the air stream may reach 26,6 m/s.

3.3 Lessons drawn from the first BEATRICE experiments

The first experiments driven with BEATRICE concerned the automatic fire detection systems and the extinguishing means on fire filters.

To test automatic fire detectors, the material to be burned was chosen according to the dust deposit actually found in ducts (see 2.3). This material could never make the detector work and it was necessary to use cartoon boards used currently to test the automatic fire detection systems. So it appears that the detection of fires in ventilation ducts is probably much more difficult than it could be imagined.

Speaking of fire filters, no extinguishing mean was efficient when the fan was on. Though it is often difficult or unpossible to stop the ventilation in nuclear facilities, there is a big problem which have to be solved.

IV CONCLUSION

The experiments which have been driven up to now though very approximate show that fire in ventilation ducts is a serious risk which is rather unknown and against which we have no efficient fighting means.

It appears indeed that the phenomenas concerned are much more complicated than what could be imagined at the beginning.

We have proposed a method to continue the ventilation as long as possible to maintain the confinement of a nuclear facility in case of fire. This supposes that the ventilation system is able to assume his fonction a time long enough to allow to the fire fighters to extinguish definitely the fire. It is the reason why we have tried to define the weak parts of a ventilation system in such circonstances and to develop a serious research program to solve those problems which may be severe in our ancient facilities.

REFERENCES

1. J. SAVORNIN

"General considerations on fire and explosion in a nuclear facility Interaction with ventilation". CSNI specialist meeting on interaction of fire and explosion with ventilation systems in nuclear facilities - Los Alamos - April 1983.

2. A. BRIAND, JC. LABORDE, Ph. MULCEY

"Limits of HEPA Filters operation under high temperature conditions" - 19 th DOE, Nuclear Air Cleaning Conference Seattle - August 1986 - (To be published).

3. J. DUPOUX, Ph. MULCEY, JL. ROUYER, X. TARRAGO

"SIMOUN high temperature dynamic test rig for industrial air filters" - 18 th DOE Nuclear Air Cleaning Conference - Baltimore 1984.

DISCUSSION

ALVARES: I noticed that the ventilation system for the Beatrice test rig is an injection device and not an extraction device. Most of the facilities that we have are balanced, but they do have an extraction fan so that the pressure on the downstream side of the filter can be less than on the upstream side of the filter. Is that something that you are considering adding to your systems?

SAVORNIN: Yes, we are. We designed the test rig the way I showed because we wanted to make tests to determine the direction the fire may take. When we want to test fire extinguishers, we locate the fan so that it is exhausting instead of blowing

RELEASE OF RADIOACTIVITY OUT OF WASTE PACKAGES UNDER THERMAL STRESS

D. Gruendler, H.W. Brenig, W. Wurtinger
Gesellschaft fuer Reaktorsicherheit (GRS) mbH
Schwertnergasse 1, 5000 Köln (Cologne) 1

Abstract

The release of radioactivity out of waste packages under thermal stress is generally determinded by complex processes, which cannot be formulated mathematically. However essential parameters such as mechanisms and heat transfer parameter can be simplified so far, that the determination of the released activity can be carried out analytically. Examples of calculations show release fractions between 10^{-6} and 10^{-3}. Although these results are conservative, the values confirm the thermal behavior of the wastes and thus, the presented models are suitable for the performance of safety analyses.

I. Introduction

In recent years, there has been a worldwide increase in research activities in the field of nuclear waste management. Apart from the analysis of long-term safety, one of the major aspects of research is the investigation of the safe manipulation of these wastes, for example during transport, handling and storage. A comprehensive safety analysis requires a great number of various individual analyses which can all be dealt with along the same lines, cf. Figure 1.

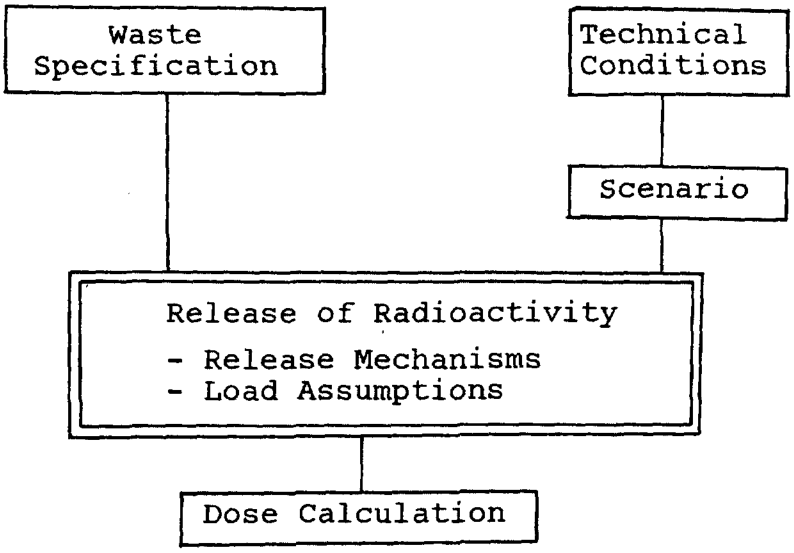


Fig. 1: Scheme of safety analyses

The basic requirements to be met are, on the one hand, the specifications of the radioactive waste and, on the other, the existing technical boundary conditions. These technical boundary conditions are the background of accident scenarios which determine the loads assumed to act upon the radioactive waste. With respect to accidents involving radioactive wastes, the relevant load assumptions are mechanical stresses, thermal stresses or a combination of the two. The load assumption and the specification of the radioactive waste under review determine the activity that is released (source term determination). The last step is the calculation of the dose on the basis of the source term.

The present paper focuses on the determination of source terms. While dose calculations have almost been standardized as a result of various computer codes, there are no comparable algorithms for the determination of source terms.

The paper describes certain elements of a source term determination which may become part of a possible standardized procedure for the determination of source terms.

These are

- release mechanisms,
- the relevance of release mechanisms for certain wastes,
- load assumptions
- temperature fields.

Based on these elements, results of source term determinations are presented as examples with respect to cemented wastes, scrap and compacted wastes in concrete and steel sheet containers.

II. Release Mechanisms

There are four relevant mechanisms for release as a result of thermal stress:

- release due to pyrolysis of the waste product,
- release due to combustion of the waste product,
- release due to the vaporization of water in the waste product,
- release due to sublimation or vaporization of radioactive substances.

Pyrolysis

Pyrolysis is understood as the thermal decomposition of a material. In waste packages, this process takes place if the temperature of the product exceeds a certain threshold, e.g. approx. 300 °C, with a restricted admission of air, and the waste product contains thermally instable constituents. The gaseous decomposition products - the so-called pyrolysis gases - escape from the waste package and burn outside the waste package if they are ignited. With the pyrolysis gases, a certain percentage of activity is released.

In experimental investigations of the percentages of activity released by pyrolysis, results of approx. 5×10^{-3} are obtained for Cs /JOH 85/.

As far as less volatile nuclides are concerned, the released percentage of activity is smaller.

Combustion

In the event of the assumed thermal stress condition, combustible waste products will burn if a sufficient amount of air is supplied to the waste product.

Quite a number of experimental investigations have been carried out to determine release percentages due to combustion /RÄM 79b/, /MIS 70/, /MIS 73/, /SUT 74/, /SEE 82/, /VEJ 82/. In these experiments, the release percentages are between 10^{-2} and 0.4. The great variations are a result of the different experimental boundary conditions.

Added to these released percentages may be the resuspension of the residues of combustion.

Vaporization of Water

Due to co-vaporization, the vaporization of water also causes a transition of radioactive substances into the gaseous phase. In investigations of the evaporization of liquids /MIS 68a/, /MIS 68b/, results of approx. 10^{-5} to 10^{-3} are obtained with respect to the shares released. This release mechanism is particularly relevant for cemented wastes and concentrates. These wastes contain great amounts of vaporizable water in their pore volume, water of crystallization, or the like.

Investigations of cemented wastes in 200 l drums /JOH 85/, /VEJ 83/ confirm the results of the evaporization experiments.

Sublimation of Radioactive Substances

Apart from the release of radioactive substances by means of co-vaporization, radioactive substances are also subject to direct vaprorization or sublimation. In general, this release pathway determines the release of volatile substances such as iodine or tritium. However, it can also be of importance for other elements, in particular Cs. The released percentage of activity depends on the duration and the temperature level of sublimation. The amount of released activity can be theoretically estimated on the basis of the thermodynamic data of the substance and Lewis' law.

III. Relevance of Release Mechanisms for Various Wastes

In the case of cemented wastes, a combination of the release mechanisms of pyrolysis (in hot surface layers) and co-vaporization is possible. For the quantification of the individual contributions it is assumed that pyrolysis takes place in those parts of the volume where the temperature is higher than 300 °C, and that co-vaporization is the main process in those parts of the volume where the temperature is higher than 100 °C.

As far as scrap is concerned, the only relevant process is that of release by sublimation of the radionuclides.

Crude compacted wastes may consist of pure scrap or mixed wastes, for example textiles and paper. In the case of an overwhelmingly metallic composition (scrap), the relevant release process is that of sublimation of the radionuclides, in all other cases it is that of pyrolysis.

Apart from the above-mentioned mechanisms for the release of radionuclides, the release of easily volatile compounds such as organic iodine is determined by sublimation in all kinds of waste flows.

The release by combustion, which was mentioned when describing the possible release mechanisms, will usually be of no effect. This is due to the fact that, in the scenarios investigated, the integrity of packaging is generally preserved to such an extent that only pyrolysis takes place.

IV. Load Assumptions

The released activity mainly depends on the load assumptions for thermal stresses, and in this context, thermal stresses are always assumed to be due to the impact of a fire. For example, the load assumptions determine the volume percentages which are subject to temperatures of more than 300 °C or more than 100 °C, and/or the waste temperature that is relevant for sublimation.

In general, the load assumptions are stated as a temperature-time function of the fire (primary statement). In order to calculate the heating-up of the waste in the package, the heat transfer coefficients for both convection and radiation (secondary statement) have to be quantified as well. Considering a realistic fire load, other factors have to taken into account as well (tertiary statements), for example to what extent the package is at the source of the fire, or whether or not the package is shielded by other objects or whether or not there are also other heat sinks.

The kinetics of the fire is influenced by a number of parameters of which a few are quoted below as examples:

- distribution and kind of the fire loads,
- radiation properties of confining walls,
- cooling of the source of the fire by air draft,
- limitation of the combustion speed by a limited supply of air,
- unsteady temperature field in a flame.

The consideration of all these parameters makes it impossible to furnish a theoretical description of the temperature-time function.

This is the reason why mean model curves are used for a realistic description of the effects of a fire on a waste package, although peak values of the expected realistic temperature-time history are not reached. A well-known example of such a model curve is the load assumption of 800 °C for a period of 30 minutes as contained in the IAEA transport regulations /IAEA 73/. Another curve considering 800 °C for a period of 60 minutes is stated in /ILL 85/ with respect to the accident covering all possibilities in the final disposal of radioactive wastes.

As a result of the numerous influencing factors, even the theoretical determination of the heat transfer coefficient is almost impossible.

The main parameters for the convection transfer coefficient are:

- temperature differential between source of fire and package,
- geometry of the package,
- flow field within the source of the fire.

The following are important for the radiation coefficient:

- soot concentration in the flame,
- chemical composition of the flame,
- geometry of the flame,
- absorption properties of the package.

As in the case of the model curve, the heat transfer coefficients must be described in a model. For example, the transport regulations are based on a radiation coefficient of 0.9 and an absorption coefficient of 0.8 and assume calm environmental air as another boundary condition. The resulting total heat transfer coefficient is approx. 30 ... 110 W/m^2K, depending on the surface temperature of the waste package.

V. Temperature Fields

On the basis of the given temperature-time model curves and the heat transfer coefficients, the calculation of the unsteady temperature fields in the waste package with the available computer codes does not pose any problem. Figure 2 shows a comparison between a calculation using the IAEA model curve and the IAEA heat transfer coefficient and a fire experiment taken from /JOH 85/ for the heating-up of a drum of cemented wastes. This shows that the fixed parameters are sufficient for an adequate description of the effects of a realistic fire.

As examples, two other calculations using the IAEA boundary conditions are presented below.

Figure 3 shows the heating-up of scrap in a drum. The mean product temperature is quoted.

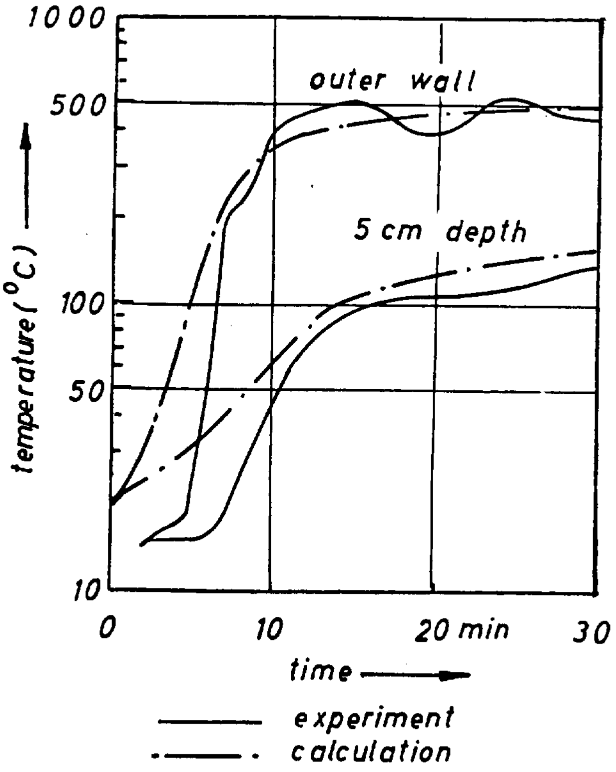


Fig. 2: Comparison between Calculation and Experiment for the heating-up of a drum

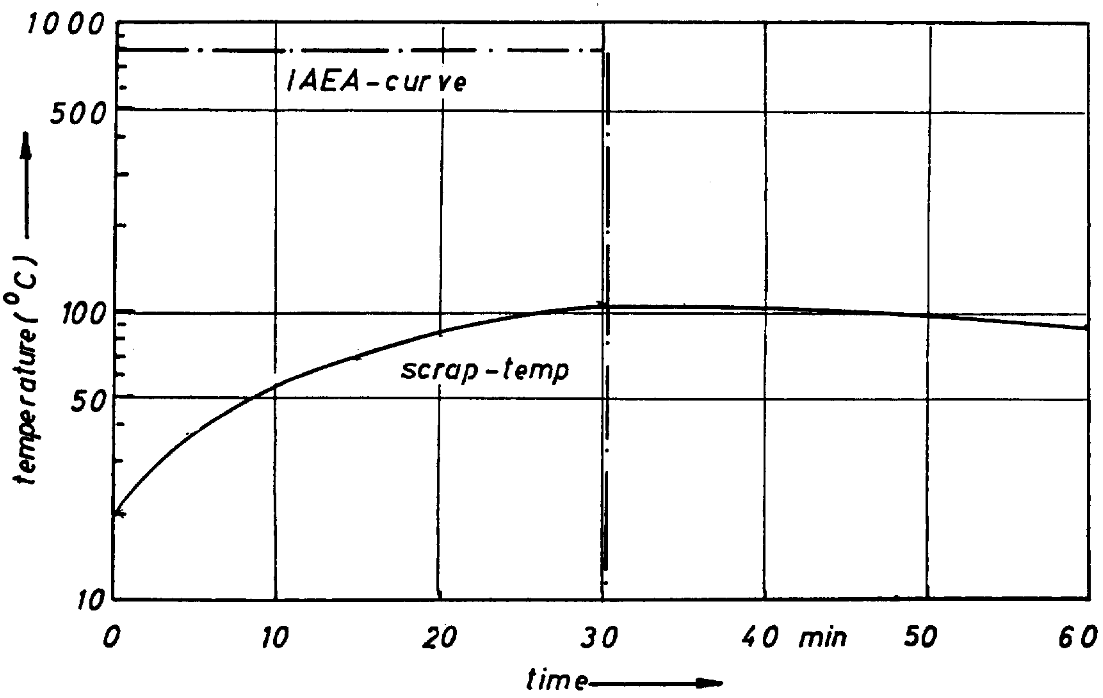


Fig. 3: Heating-up of scrap

Figure 4 shows the wall temperatures of an empty concrete container having a wall thickness of 23 cm. The waste products need not be taken into account, since the course of the inner wall temperature indicates that there can only be an insignificant heating-up of the waste product.

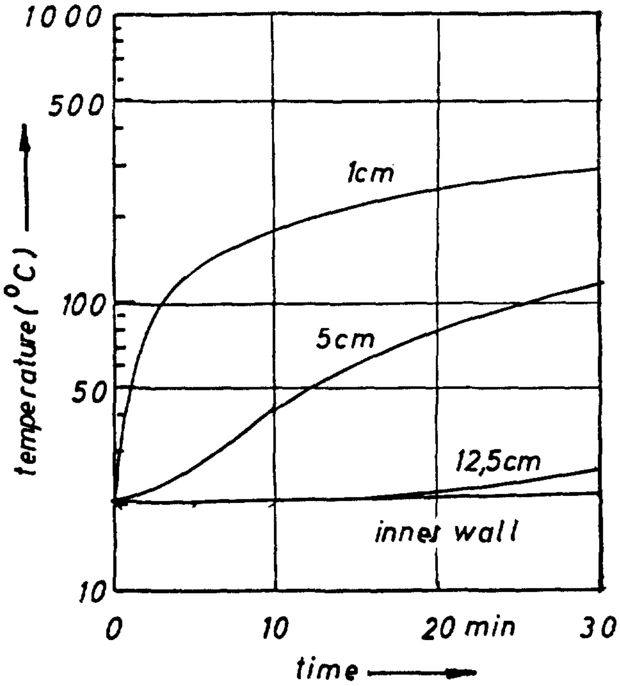


Fig. 4: Temperature in the Wall of a concrete container

VI. Examples of Source Term Determination

The following is an exemplary discussion of the source term determination for

- cemented waste,
- scrap, and
- compacted waste

in

- concrete containers and
- drums.

The simplest case of a source term determination is that of wastes in concrete containers. As can be seen in Figure 4, none of the release mechanisms addressed will apply as a result of the low inner wall temperature and thus, no activity will be released. This applies even if the calculations are based on a model curve of 800 °C and 60 minutes.

Temperature field calculations of cemented wastes in drums show an 8% volume percentage greater than 300 °C and a 20% volume percentage greater than 100 °C and smaller than 300 °C. With the release mechanisms relevant for cemented wastes, a maximum release factor of 5×10^{-4} results.

A calculation of temperature fields in packages containing compacted wastes is impossible due to the heterogeneous structure of the packages. Experiments concerning the release from compacted wastes subjected to thermal stresses show that approx. 30% of the waste is affected by pyrolysis. A value of 1.6×10^{-3} was measured as a release factor for Cs /JOH 85/.

For the sublimation of Cs, a release percentage of less than 2×10^{-6} results from the temperature curve shown in Figure 3 with respect to scrap in drums. For iodine and tritium, the release factor may be higher, depending on the chemical form, while it is smaller by a number of orders of magnitude for all other nuclides.

VII. Evaluation and Summary

The present paper used typical wastes and packages in order to show how the release of activity out of waste packages under thermal stresses can be calculated. Some of the necessary steps were simplified to such an extent that they may form the basis of a general pattern for the calculation of activity releases under thermal stress.

Thus, the complex release mechanisms are reduced to four basic release mechanisms the release behavior of which is known phenomenologically and which cover the entire spectrum of the behavior of all wastes.

Although, in reality, the thermodynamic boundary conditions of heat transfer are a many-sided phenomenon, they can be simplified by model parameters to such an extent that the effects on the packages can be described realistically, and standardized computer codes can be used.

Linking these simplified calculations with the combination of release mechanisms that are relevant for the waste concerned results in the activity percentages that are released.

An evaluation of the examples presented indicates that although the results, compared with experimental investigations, show a conservative tendency, they do confirm the thermal behavior of the wastes as determined in experiments. Thus, the procedure proves to be a useful tool for the performance of safety analyses in connection with radioactive wastes.

VIII. References

/IAEA 73/ IAEA Safety Series No. 6 Regulations for the Safe Transport of Radioactive Materials, Revised Edition, International Atomic Energy Agency, Vienna, 1973.

/ILL 85/ Illi, H.: Sicherheitsaspekte bei der Endlagerung in in "Neuere Entwicklungen zur Sicherheit von Kernkraftwerken und Anlagen des Kernbrennstoffkreislaufs" KTG Meeting in Düsseldorf, Sept. 24/25, 1985.

/JOH 85/ Activity Release from Waste Packages Containing LL and IL Waste Forms under Mechanical and Thermal Stresses, Waste Management 85, Tucson, 1985.

/MIS 68a/ J. Mishima, L.C. Schwendiman, C.A. Radasch: "Plutonium release studies; IV. Fractional release from heating plutonium nitrate solutions in a flowing air stream", BNWL-931 (1968).

/MIS 68b/ J. Mishima, L.C. Schwendiman, C.A. Radasch: "Airborne release of particles in overheating incidents involving plutonium metal and compounds", BNWL-SA-1735 (1968); CONF-680811-1.

/MIS 70/ Mishima, J. et al.: "The amount and characteristics of plutonium made airborne under thermal stresses", BNWL-SA-3379 (1970); Proc. Symp. Health Physics Soc., Idaho Falls, Idaho, Nov. 3-6 (1979).

/MIS 73/ J. Mishima, L.C. Schwendiman: "Interim Report: The Fractional Airborne Release of Dissolved Radioactive Materials During the Combustion of 30 Percent Normal Tributyl Phosphate in a Kerosine Type Diluent" BNWL-B-274, June 1973.

/RAM 79b/ Rämö, E. et al.: "Damage of Reactor Waste Packages in Transport Accidents", NKA, AO (79) 17, 1979.

/SEE 82/ Seehars, H.D., Hochrainer, D.: "Durchführung von Experimenten zur Unterstützung der Annahmen zur Freisetzung von Plutonium bei einem Flugzeugabsturz; Zusatzbericht über Experimente ... zur Freisetzung ... während eines Kerosinbrandes", BMI-Auftrag SR 0205 A, FhG ITA, Grafschaft, 1982.

/SUT 74/ S.L. Sutter, J. Mishima, L.C. Schwendiman: "Fractional Airborne Release of Strontium During the Combustion of 30 Percent Normal Tributyl Phosphate in a Kerosin Type Diluent", BNWL-B-358, June 1974.

/VEJ 82/ Vejmelka, P., et al.: "Endlagerspezifische Charakteterisierung der für eine Endlagerung in der Grube "Konrad" zu betrachtenden Abfallprodukte", KfK-Primärbericht 05.11.0.3. P01A, Karlsruhe, Feb. 1982.

/VEJ 83/ Vejmelka, P., et al.: "Activity Release from Waste Packages Containing ILCW-Cemented Waste Forms Under Mechanical and Thermal Stresses".

SIMULATION OF EXPLOSIONS IN AIR CLEANING SYSTEMS AND COMPARISON OF THE RESULTS WITH COMPUTER CODE PREDICTIONS

W. S. Gregory, B. D. Nichols, and R. D. Wade
Los Alamos National Laboratory
Los Alamos, New Mexico

P. R. Smith and D. L. Fenton
New Mexico State University
Las Cruces, New Mexico

Abstract

Experimental testing and development of computer codes for predicting the effects of explosions in air cleaning systems are being done for the Department of Energy. The work is a combined effort by the Los Alamos National Laboratory and New Mexico State University (NMSU). Los Alamos has lead responsibility in the project and is developing the computer codes; NMSU is doing the experimental testing. Obtaining experimental data to verify the analytical work is the main goal of this effort. Of secondary importance are the experimental data showing the combined effects of explosions within air cleaning systems that contain all of the important air cleaning elements (blowers, dampers, filters, ductwork, and cells). This work will result in tools that safety analysts can use to study the effects of hypothetical explosions in nuclear facility air cleaning systems.

The experimental apparatus is a small version of a large experimental system that was installed at NMSU. The small system is used to obtain gas-dynamic data (temperatures and pressures) throughout the system (such as within the cells, along the ductwork, and before and after dampers and filters). Gas explosions are simulated in the experiments using a unique system of gas-filled balloons. The experiments will yield information on the degree of protection a system offers in attenuating explosive effects within air cleaning systems.

Analytical predictions were made using computer codes that predict gas-dynamic values such as flows, temperatures, and pressures throughout the system. The gas explosions were compared with the predicted results, and good agreement was found for most of the pressure measurements. Future experiments will involve small explosive charges using blasting caps or squibs. Future experiments also will couple material transport with the explosive gas dynamics.

Introduction

There is a potential for accidental explosions within nuclear facilities. (The recent reactor explosion at Chernobyl, Russia, is a case where a catastrophic explosion actually occurred.) In this country, safety analyses for nuclear facilities are required to evaluate the possibility and effects of accidental explosions thoroughly. Therefore, we want to develop computer codes that can be used to evaluate the effect of possible explosion-induced releases from a facility, perform scoping studies involving a multitude of explosion scenarios, and evaluate the effectiveness of various protective designs. To simulate these explosive effects accurately, we must be sure that the computer codes in use will perform as expected. This can be done by comparing the calculated simulations with small-scale experiments.

This paper describes computer codes being developed by the Los Alamos National Laboratory for the Department of Energy to simulate the effects of explosions within nuclear facilities. Particular emphasis is placed on explosive propagation in the plants' nuclear ventilation and air cleaning system. The experimental apparatus and results of tests using gas-filled (hydrogen/air) balloons to create gaseous detonations also are described. Finally, the test data are compared with computer code simulations.

Test Equipment Description

Figure 1 is a plan view of the experimental ventilation system located on the New Mexico State University (NMSU) Campus in Las Cruces. The ventilation system has two steel tanks simulating rooms. One tank is cylindrical and 2.74 m (9 ft) in diameter with a volume of 24.3 m^3 (859 ft^3). The second tank is essentially rectangular and 3.3 by 2.1 m (10.9 by 6.9 ft) on the sides with a volume of approximately 17 m^3 (600 ft^3). The tanks are connected by 0.305-m (1-ft)-diam ducts as shown in Fig. 1. Air is drawn through the system by a 28.4 m^3 (1000-ft^3/min) centrifugal blower attached to the exit duct of the cylindrical tank. Just upstream of the blower is a 30.5- by 30.5-cm (12- by 12-in.) high-efficiency particulate air (HEPA) filter and a 23.5- by 35.2-cm (9.25- by 13.875-in.) parallel-blade damper (fully open).

Shock waves are created in the system by exploding hydrogen/air-filled latex rubber balloons nominally 50.8 cm (20 in.) in diameter. The balloons are filled using a Matheson model 7372T gas proportional flowmeter through which the hydrogen and air flow simultaneously.

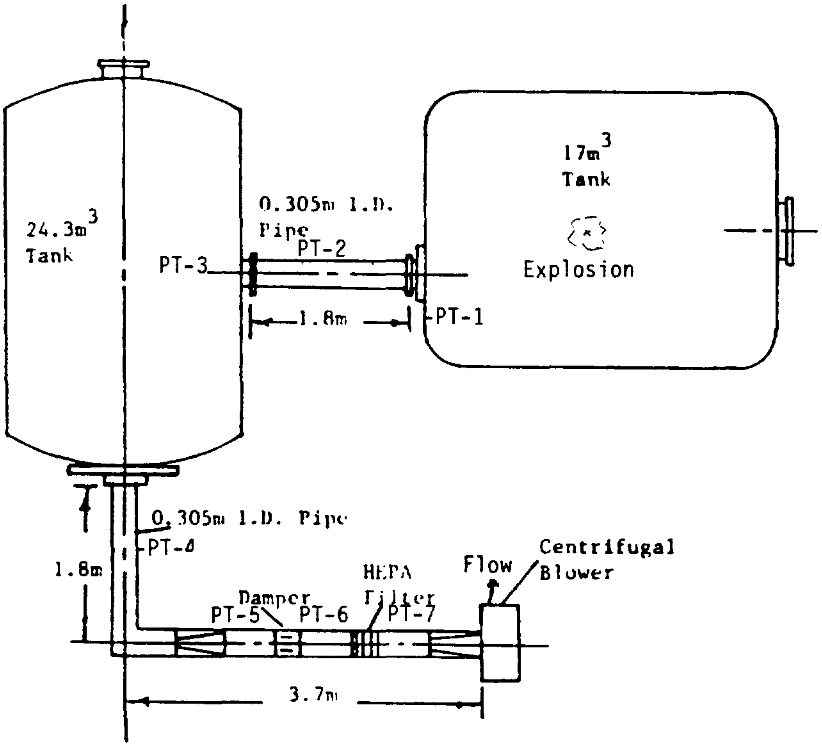


Figure 1. Plan view of the model ventilation system.

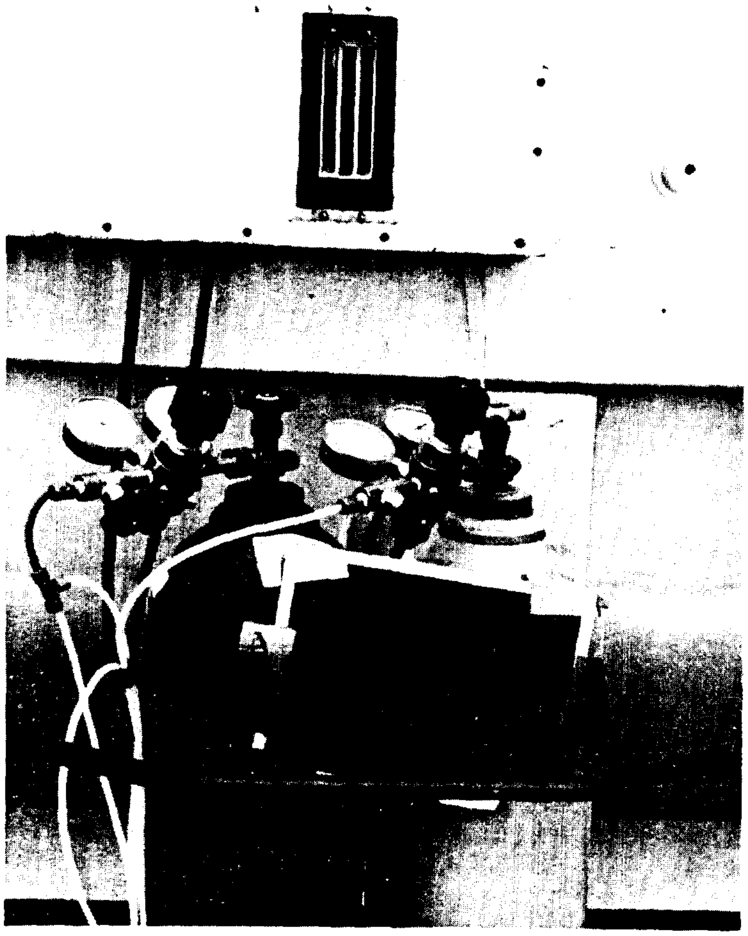


Figure 2. System for filling balloons with gas mixture.

The system for filling the balloons is shown in Fig. 2; Fig. 3 is a photograph of a filled balloon just before an explosion.

The shock wave created by the hydrogen/air explosion was measured by seven Kulite Model XT-190 miniature pressure transducers with a range of 0 kPa to 172 kPa (0 to 25 psia). All pressure measurements except those in the tanks were side-on measurements. The pressure transducer locations are shown by the symbol PT-X in Fig. 1. (X is a digit from 1 to 7.) The data from the pressure transducers were digitized and recorded by a high-speed CAMAC data acquisition system using a DEC PDP11/10 digital computer.

All seven pressure transducers were calibrated against a pressure standard before each experiment. The balloons were filled at a known flow rate for each gaseous component for a measured length of time. A small electrical impulse then caused the hydrogen-air mixture to explode, and the resulting shock wave automatically started the data acquisition system as it encountered a nearby trigger transducer.

Computer Code Description

Two computer codes have been developed at Los Alamos to calculate the gas dynamics associated with confined explosions: EVENT84 and NF85.[1,2] EVENT84 is an updated version of the EVENT computer code and includes an empirical explosion chamber model. Explosions involving TNT, H_2/O_2, acetylene, and red oil are calculated automatically as source terms for the explosion. NF85 is a fully three-dimensional explosion chamber model. It calculates the detailed driver gas dynamics for EVENT84, and its capabilities in simulating shock transmission tests were described earlier.[3]

The emphasis in this paper is modeling the overall system using the EVENT84 computer code. In addition, only gaseous explosions were simulated. Several methods other than NF85 were used to simulate the explosion.

- EVENT84 explosion chamber
- Approximate source-term method
- Pressure-time history

Computer Model for Test Simulation

Certain assumptions are made to model a system using EVENT84: perfect gas (air), compressible flow, momentum balance with friction and inertia, choking, linear and nonlinear filters, certain blower characteristics, and mass and energy addition to the gas phase. The model uses a lumped-parameter formulation; that is, no spatial distribution of parameters within network components is included. In addition, the analytical model must have the same arrangement of components, friction characteristics, capacitance, duct lengths, cross-sectional areas, boundary pressures, and driving forces as the experimental model.

The physical system is described first with a schematic consisting of a network of branches and nodes. Network theory defines system elements that exhibit flow resistance and inertia, or flow potential, as branches. The ventilation system components modeled as branches include dampers, ducts, valves, filters, and blowers. The connection points of branches are network system elements called nodes and always have a finite volume. Nodes include specific network components that have a finite volume such as rooms, gloveboxes, and plenums, or the node may contain only the volume of connecting branches. System boundaries, where the volume is practically infinite, also are specified as nodes.

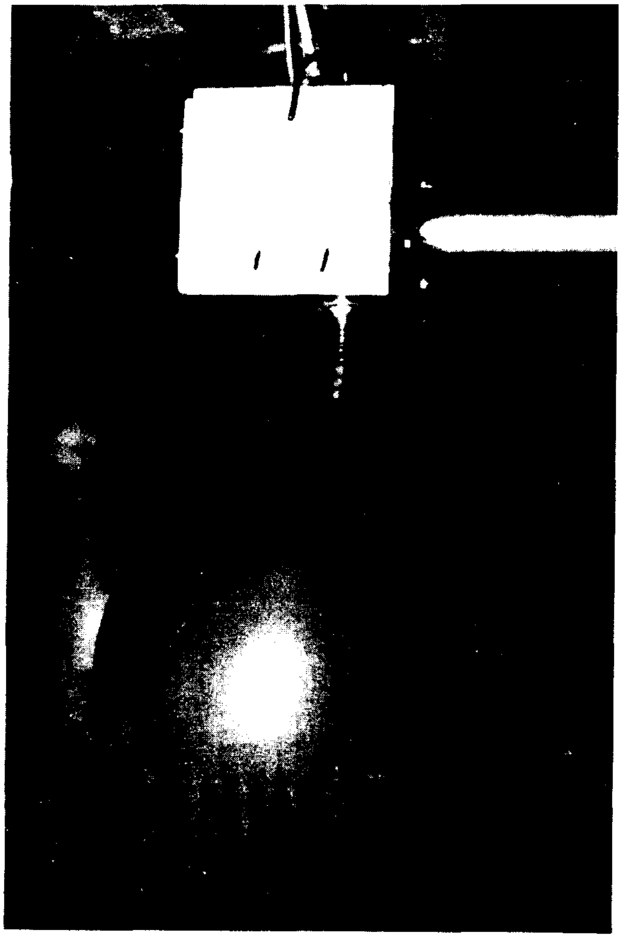


Figure 3. A gas-filled balloon just before an explosion.

The energy conservation equations are applied to internal (capacitance) nodes using a lumped-parameter formulation assuming homogeneous mixture and thermodynamic equilibrium. A momentum equation that includes the effect of wall friction and inertia is used to relate the flow rate to the pressure drop across a duct; choking is imposed on the duct flow if the conditions warrant it. A filter provides only resistance to the flow. A quasi-steady relation is imposed between the pressure head and the flow rate for a blower.

The network system models for EVENT84 are shown in Fig. 4 and Fig. 5. The inlet on the rectangular tank can be open or closed, as can the inlet on the cylindrical tank. Two arrangements were used for modeling the experiments. In Model 1, the rectangular tank inlet is open and in Model 2, the cylindrical tank inlet is open.

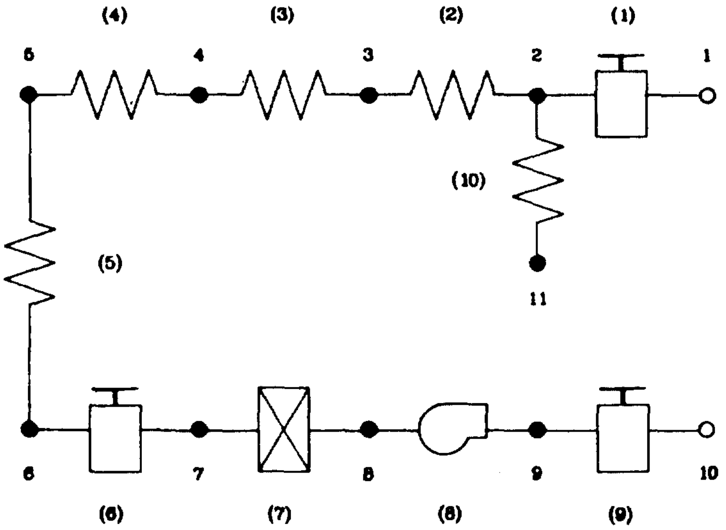


Figure 4. Computer network schematic for Model 1.

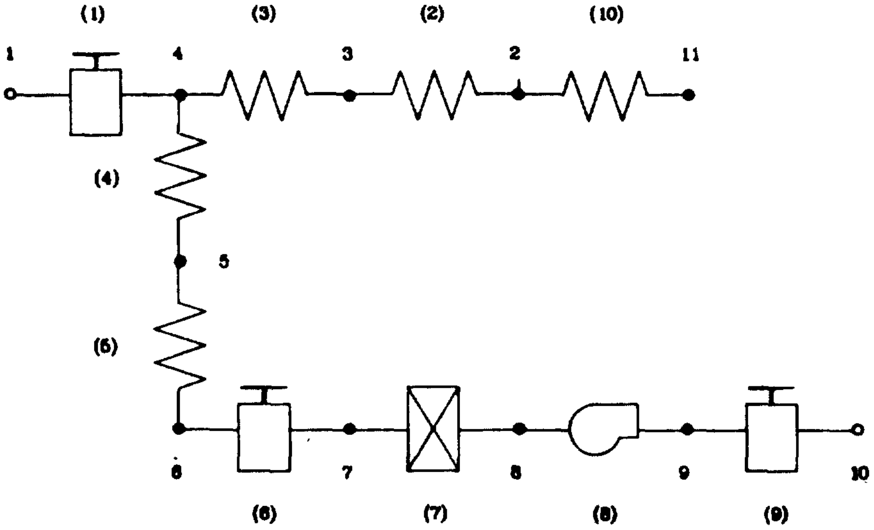


Figure 5. Computer network schematic for Model 2.

Models 1 and 2 consist of 11 nodes, including 2 boundary nodes and 9 internal nodes, and 10 branches. The symbols used on the schematic represent dampers, blowers, duct resistance, filters, and volumes of the ductwork, cylindrical, and rectangular tanks. For example, in Model 1 the numbers enclosed in parentheses represent branches and numbers without parentheses represent nodes. Branches contain blowers, dampers, duct resistance, and filters. The nodes represent points such as the explosion chamber and the cylindrical tank. Pressures and temperatures are calculated at the nodes, whereas flows are calculated for the branches.

To accurately determine the resistance coefficient for each branch, each component (the 90-degree bend, the damper, the filter, and the blower) is modeled as a separate branch. In addition, the entrance and exit for the system are modeled as dampers (branches) to account for the entrance or exit losses. The duct between the two tanks is divided into two branches to accommodate the pressure measurement. The explosions take place within a balloon in the rectangular tank. This balloon is modeled as a separate node, with a flow area into the tank equal to the surface area of the balloon. The explosion is simulated as a mass and energy input into the balloon node, either as user-calculated time functions or through the explosion chamber subroutine.

Experimental and Code Simulation Results

The experimental and code simulation results involve the following.

- Code simulations and comparison with experimental results in the explosion chamber
- Code simulations and comparison with experimental results just before the system filter
- Code simulations of pressures upstream and downstream of the filter
- Experimental results of pressures upstream and downstream of the filter

Both Model 1 and Model 2 were involved in the comparisons.

The first set of experimental and code simulation results is shown in Figs. 6 and 7. Figure 6 shows that the two methods used to simulate the explosion (EXCHAM and SOURCE TERM) over-predict the pressure within the chamber. However, the shape of the pulse is very similar. The peak pressures predicted by the code were 3.174 kPa (0.46 psi), whereas the experimental values were 1.518 kPa (0.22 psi). The peak pressure times were in good agreement. In Fig. 7, Model 2 shows similar results. That is, the peak pressures are approximately twice the values obtained from the experiment. Again, the peak pressure times were in good agreement. Closing the explosion chamber door in Model 2 increases the peak pressure by about 50%. These results were expected using the EVENT84 code. That is, the code is expected to give conservative results in areas where the explosion takes place. That is why the NF85 code has been developed--to more closely simulate explosive effects near the source.

We must point out that the experimental data were smoothed to make the information more presentable. The effect of this process was to take out the highest peaks, which were of the same magnitude as the code results. The high peaks in the experimental data are caused primarily by shock reflections inside the explosion chamber

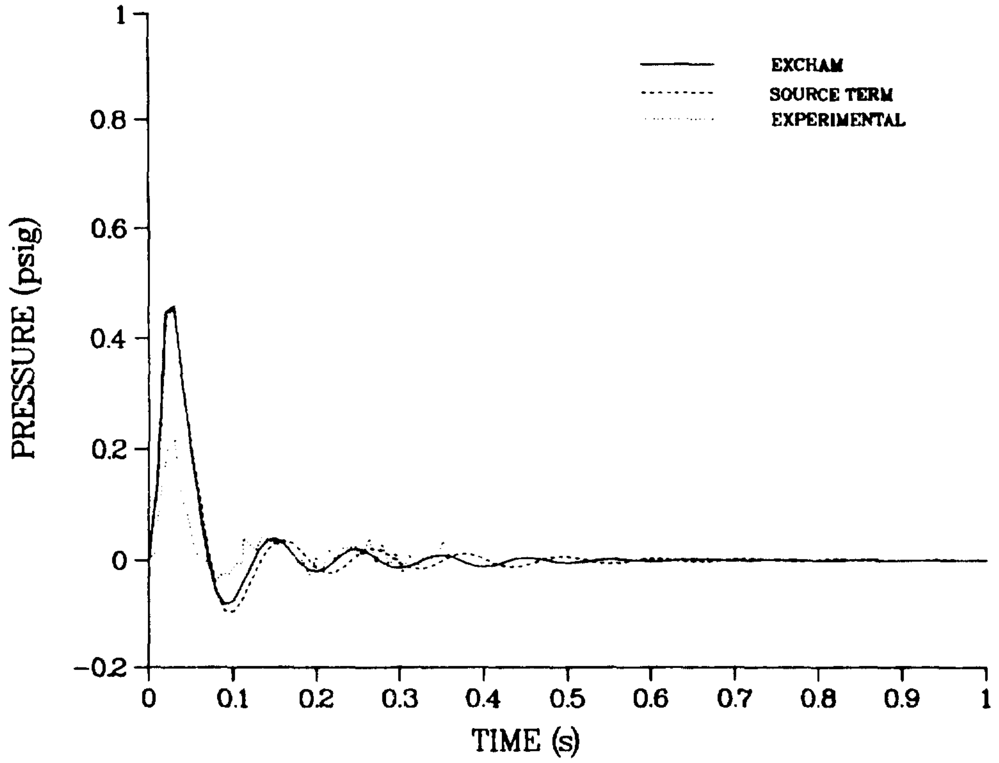


Figure 6. Code simulation of pressure in explosion chamber and comparison with experimental results (Model 1).

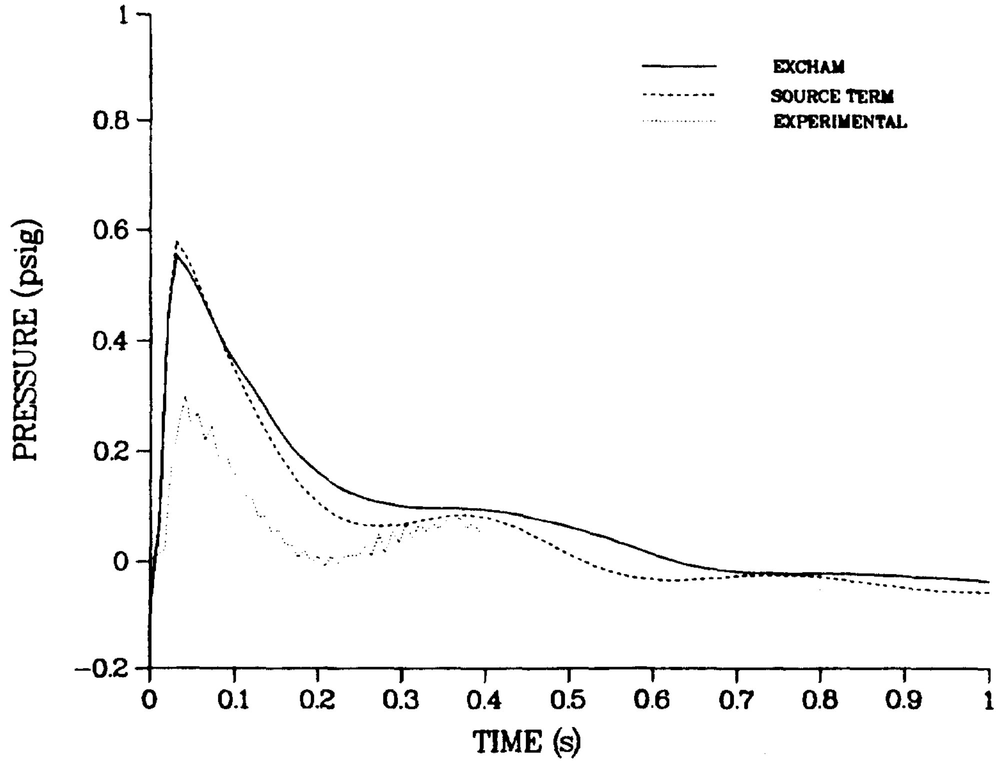


Figure 7. Code simulation of pressure in explosion chamber and comparison with experimental results (Model 2).

Pressure comparisons were made downstream of the explosion (in the chamber right before the filter). This location corresponds to node 7 on Models 1 and 2. As in the results for pressure in the explosion chamber, the code conservatively over-predicts the pressure. However, the analytical and experimental results are much closer. The time at peak pressure is much closer for Model 2 than for Model 1. As shown in Fig. 9, the peak pressure is dissipated to 1.21 kPa (1.208 psi) after passing through the ductwork and the cylindrical tank. The closer EVENT84-predicted pressure transients results supports our claim that the code predicts reasonable pressure levels in regions removed from the explosive source, particularly in areas where the final filters will be located.

Figure 10 is a plot of the pressures upstream and downstream of the filter for Model 2. As shown in Fig. 10, the effect of the filter is to essentially damp out the pressure wave. Figure 11 shows the code simulations of the pressures before and after the filter. These results indicate that the modeling does not indicate a complete dampering of the pressure wave although the peak pressure is reduced from 1.07 kPa (0.155 psi) to 0.345 kPa (0.05 psi).

Summary

Experimental verification of the EVENT84 computer code using hydrogen/air gas mixtures has been performed. The experimental apparatus consisted of two compartments with interconnected ductwork, a damper, a filter, and a blower. EVENT84's methods of simulating explosive events were compared with pressure transients obtained in the explosion chamber. The code predicted results that were conservative by a factor of 2. Comparison of pressure at the system's filter indicate good agreement with the experimental data. Experimental and analytical results show that the effect of the filter is to dampen the pressure wave as it passes through the filter.

Future experiments will use solid explosive and be expanded into a much larger system. These experiments will also use a simulant radioactive aerosol for material transport data.

References

1. Martin, R. A. and Wilson, T. M., "EVENT84 User's Manual--A Computer Code for Analyzing Explosion-Induced Gas-Dynamic Transients in Flow Networks," Los Alamos National Laboratory report LA-10312-M (December 1984).

2. Steinke, R. G., "NF85 Users Manual--A Computer Code to Predict Three-Dimensional Explosion-Induced Gas-Dynamics in Confined Spaces," Los Alamos National Laboratory report in preparation.

3. Smith, P. R., Fenton, D. L., Moore, J. A., and Steinke, R. G., "Experimental and theoretical investigation of shock waves in ventilation systems," in Proc. European Conference on Gaseous Effluent Treatment in Nuclear Installations, Luxembourg (October 1985).

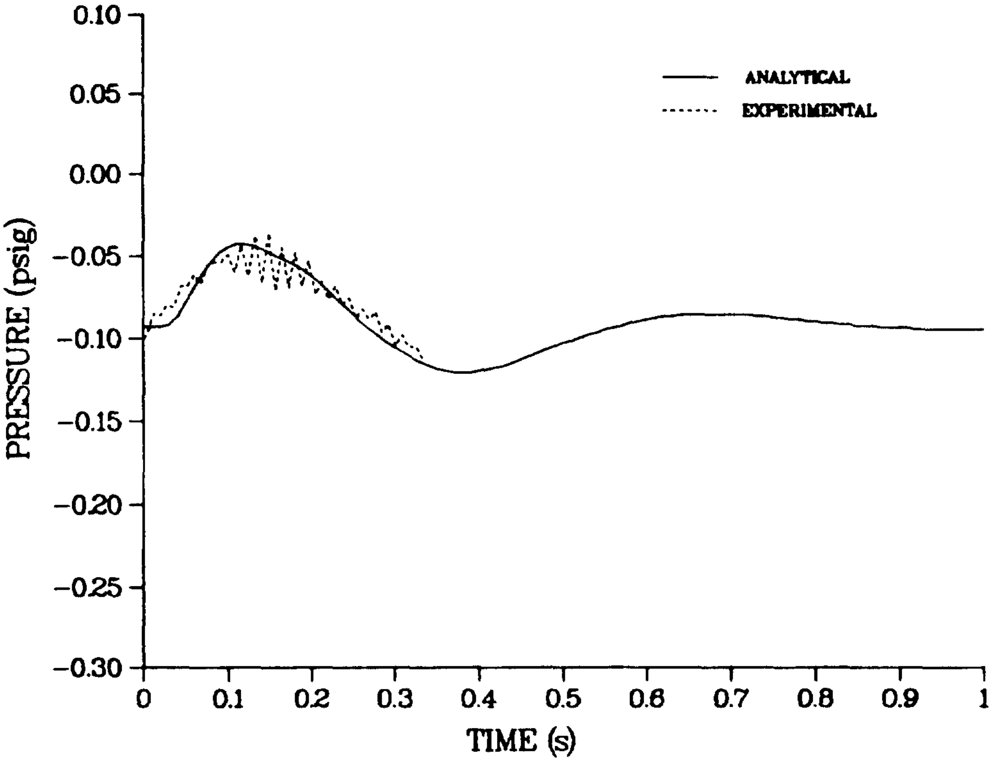


Figure 8. Code simulation and experimenal results of the pressure just before the filter in Model 1.

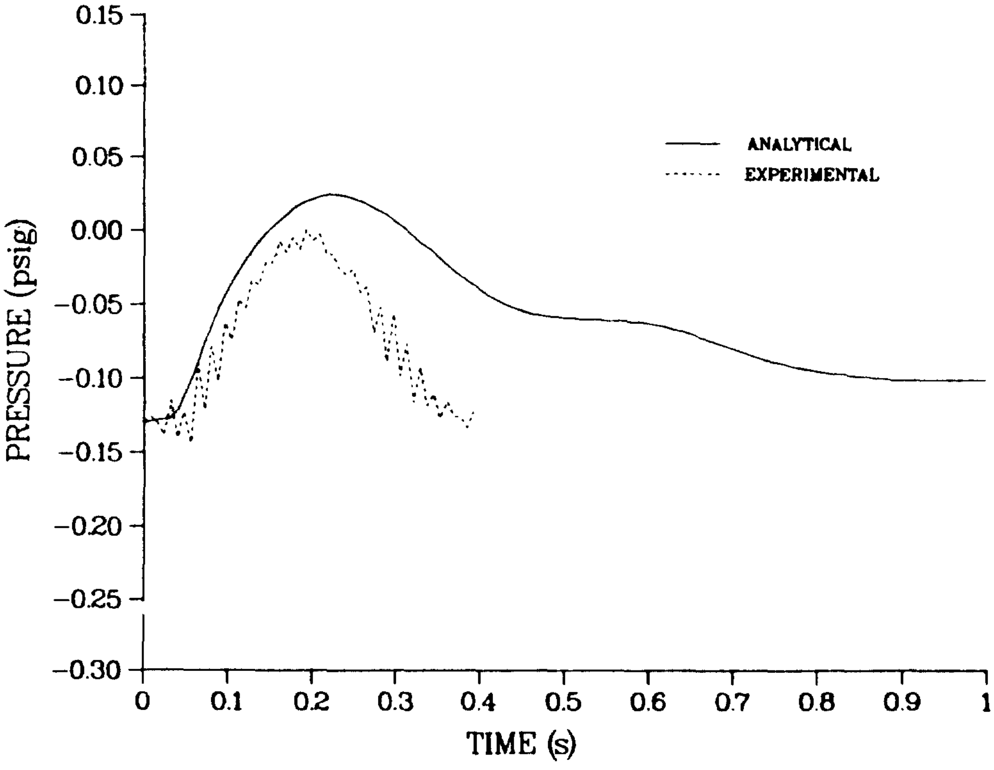


Figure 9. Code simulation and experimental results of the pressure just before the filter in Model 2.

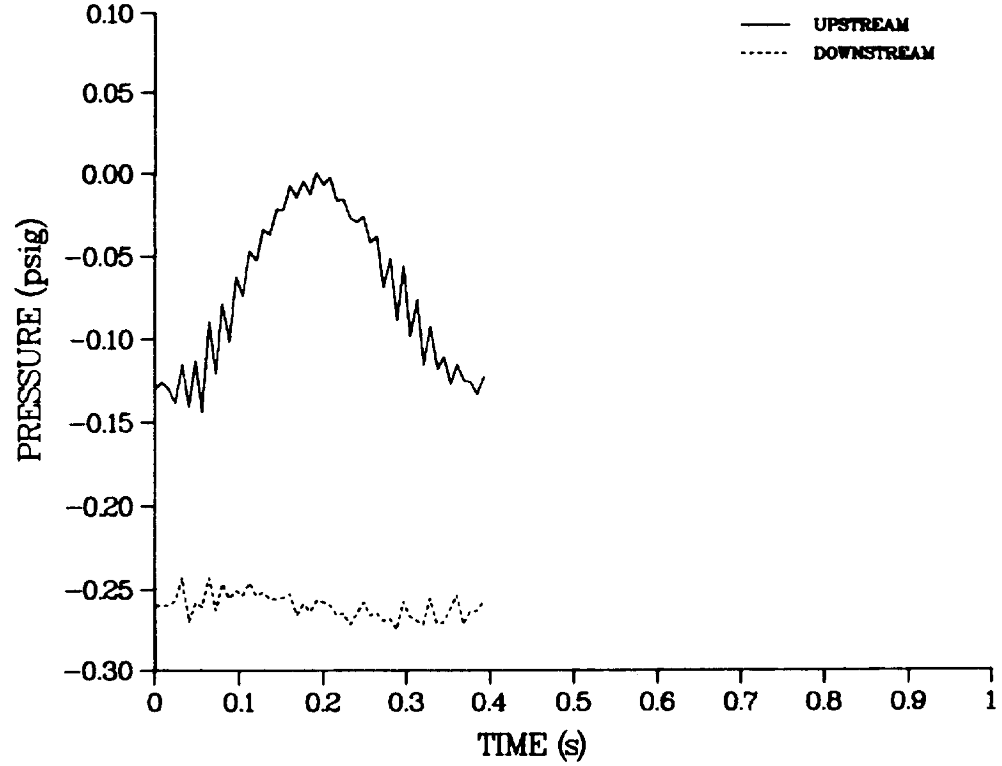


Figure 10. Experimental results for transient pressures upstream and downstream of the filter (Model 2).

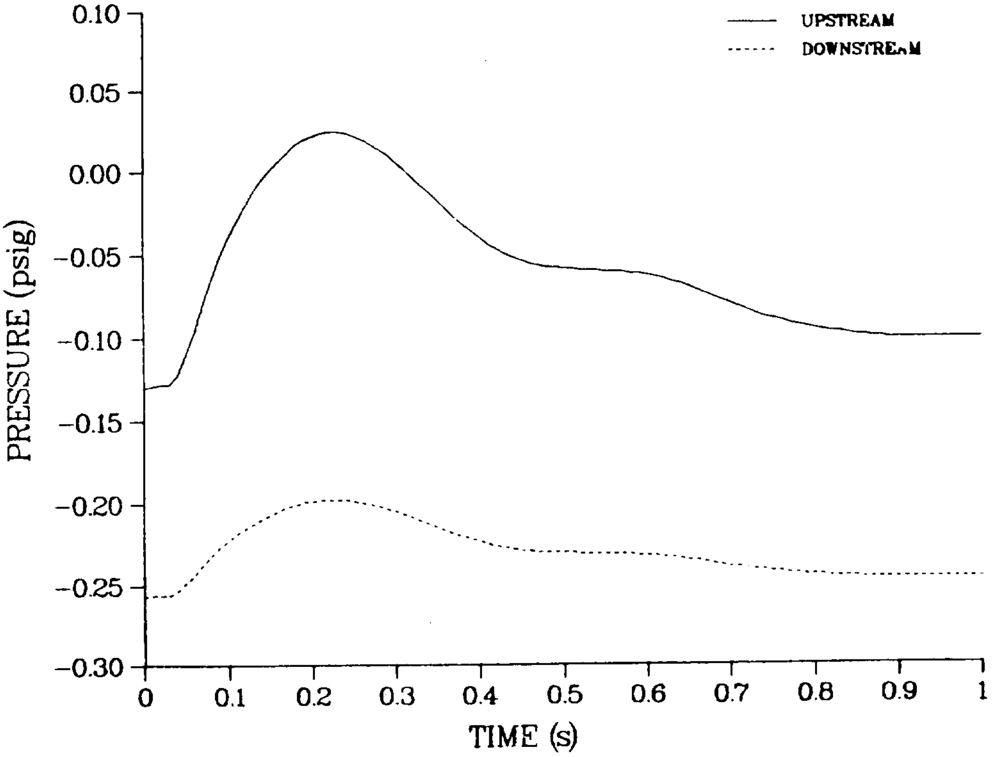


Figure 11. Code simulation results of pressures upstream and downstream of the filter (Model 2).

DISCUSSION

SANDOVAL: Did you measure the performance of the HEPA filters you tested while under shock and environmental loadings? What was the impulse you measured under the pressure loadings?

GREGORY: We didn't do it in these particular tests. We have performed tests like that with a shock tube to look for structural levels where the filter is going to break. We also looked at efficiency whenever we entrained a certain amount of aerosol in the shock pulse itself. Depending upon the filter, we found the filters failed between 0.5 and 2.5 psi across the filter over 0.1 seconds. In the studies discussed here, we were just trying to verify the computer code so we didn't worry about efficiencies or structural levels of the filters.

YAMADA: I am interested in the EVENT code. Is EVENT-84 code the same as the original EVENT code? If not, please tell me what is the advantage of the later version?

GREGORY: As you noted, there is a code called EVENT at Argonne National Laboratory. It is called EVENT-84. The only difference is that EVENT-84 incorporates an explosion chamber model that uses data from the Naval Service Weapons Laboratory. It allows you to create whatever explosive spike you like. Or, you can allow the explosion chamber model to use empirical data that is in the code to calculate the shape and the magnitude of the initiated explosion depending upon the pounds of TNT, or red oil, or whatever you have. That is the only difference.

FIRE-ACCIDENT ANALYSIS CODE (FIRAC) VERIFICATION

B. D. Nichols and W. S. Gregory
Los Alamos National Laboratory

D. L. Fenton and P. R. Smith
New Mexico State University

Abstract

The FIRAC computer code predicts fire-induced transients in nuclear fuel cycle facility ventilation systems. FIRAC calculates simultaneously the gas-dynamic, material transport, and heat transport transients that occur in any arbitrarily connected network system subjected to a fire. The network system may include ventilation components such as filters, dampers, ducts, and blowers. These components are connected to rooms and corridors to complete the network for moving air through the facility.

An experimental ventilation system has been constructed to verify FIRAC and other accident analysis codes. The design emphasizes network system characteristics and includes multiple chambers, ducts, blowers, dampers, and filters. A large industrial heater and a commercial dust feeder are used to inject thermal energy and aerosol mass. The facility is instrumented to measure volumetric flow rate, temperature, pressure, and aerosol concentration throughout the system. Aerosol release rates and mass accumulation on filters also are measured.

We have performed a series of experiments in which a known rate of thermal energy is injected into the system. We then simulated this experiment with the FIRAC code. This paper compares and discusses the gas-dynamic and heat transport data obtained from the ventilation system experiments with those predicted by the FIRAC code. The numerically predicted data generally are within 10% of the experimental data.

Introduction

FIRAC is one of a family of computer codes that has been developed at the Los Alamos National Laboratory to predict the effects of fires, explosions, and tornadoes in nuclear fuel cycle facilities.(1) The FIRAC code was designed to numerically model fire-induced flows, heat transfer, and material transport within ventilation systems and other airflow pathways. Fires may be represented in the code either parametrically or by a fire compartment model, FIRIN1, which was developed at the Pacific Northwest Laboratory(2).

FIRAC and the other accident analysis codes need to be validated. Therefore, a ventilation system model has been constructed at the New Mexico State University (NMSU) to obtain the necessary experimental data. We have performed a series of experiments in which a known rate of thermal energy is injected into the system and have simulated these experiments with FIRAC. In the second part of this paper, we describe the experimental facility and its instrumentation, and in the third part, we present the numerical model and discuss its salient features. In the last part, we discuss the gas-dynamic and heat transport data obtained from the ventilation system experiments and compare them with those predicted by the FIRAC code.

Experimental Facility

The multicompartment ventilation system model is installed in a prestressed concrete building that provides environmental control; the building is at the Mechanical Engineering Test Site on the NMSU campus. The model ventilation system is designed to accommodate thermal, pressure, and aerosol inputs. The thermal inputs are generated by a commercially available duct heater rated at 92 000 kcal/h fired by natural gas and limited to a maximum of 300°C. The duct heater is on casters, which allows thermal input from different locations of the model system. Pressure pulses are limited to a 140-kPa overpressure and must originate in the rectangular volume shown in Fig. 1. The pipe and square ducts are designed to have a mechanical safety factor of 3. The particulate mass input is provided by a commercial dust feeder having variable supply rates between 1 and 40 g/min (particulate material density = 1 g/cm^3). The maximum particulate mass concentration is approximately 1.4 g/m^3 for unit-density particulate material.

The model ventilation system's arrangement of ducts and volumes is shown in Fig. 1. For economy, 30.5-cm-diam Schedule 20 pipe is used for the bypass loop around the two volumes and for the connections between the two volumes. The 0.6-m-square ducts were made from 0.64-cm steel plate and were used for the remainder of the ventilation system. The system's straight length is approximately 24.4 m. The inlet and outlet round duct connections are within the lower third of the rectangular tank and differ in height by 0.69 m vertically. Figure 1 shows one circular duct connecting at the top of the cylindrical volume and the other circular duct connecting at the side of

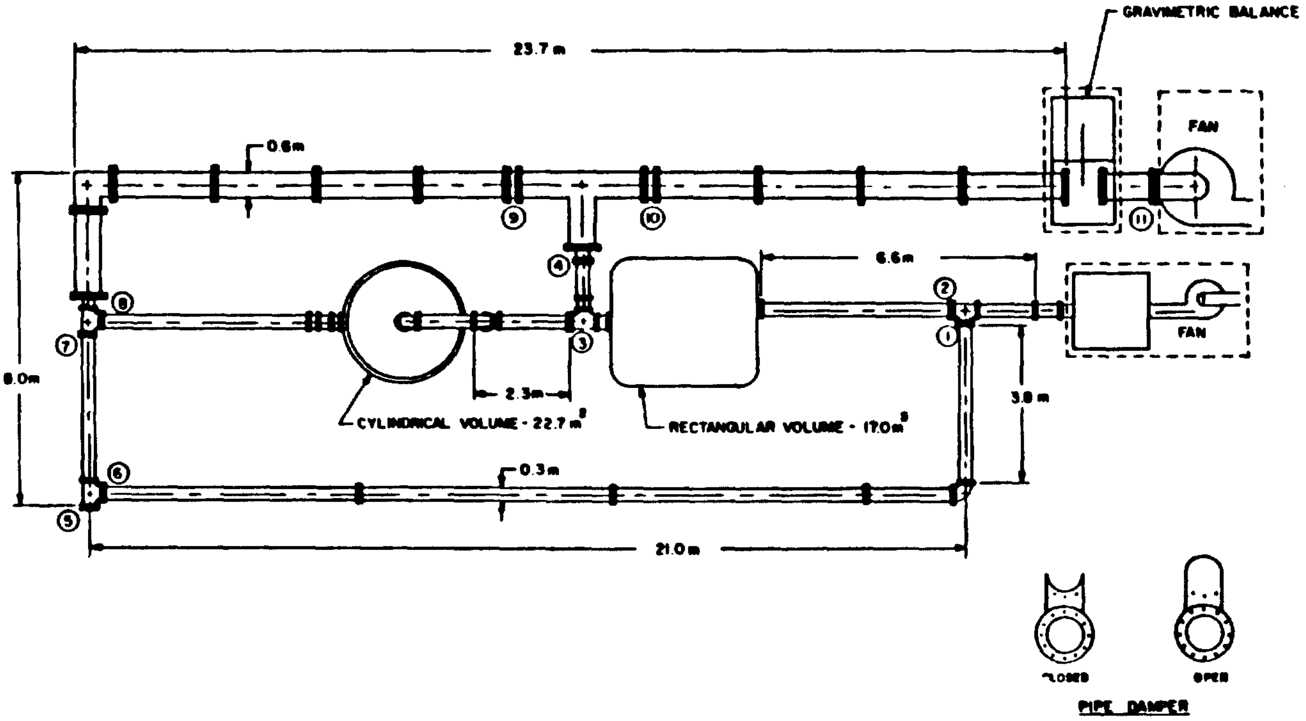


Figure 1. Schematic of the ventilation system model.

the volume (lowest possible location). The centerline distance of both the round and square ducts above the floor is 1.1 m.

The two volumes are steel tanks modified as shown in Fig. 1. The rectangular tank (17.0 m^3 in volume) has 5-cm-thick walls and can withstand the largest pressure pulses. The remainder of the system limits the maximum pressure pulse to about 140 kPa over-pressure. The cylindrical tank (22.6 m^3 in volume and 4.6 m high) is upright to provide the maximum possible stratification and serves as a location for thermal and/or particulate mass input. The two fans in the facility provide positive or negative pressures in the model ventilation system.

The round-duct dampers are numbered in Fig. 1 and are reversible 1.27-cm steel plates secured by the pipe flanges. These dampers are either full open or closed. Additionally, three dampers are located in the square duct between the high-efficiency particulate air (HEPA) filter gravimetric balance and fan, upstream from the square-duct tee, and downstream from the square-duct tee. Dampers 9 and 10 are commercial models and thus are adjustable, but damper 11 is conceptually similar to the round-duct dampers. Numerous model system configurations can be obtained by opening and closing the dampers. The HEPA filter gravimetric balance is designed especially to measure the collected mass on a HEPA filter installed in the system. The balance uses a null technique and an electronic force transducer to achieve a resolution of 2 g.

This model ventilation system is three-dimensional because of the vertical height associated with the cylindrical volume. Thus, thermal loads or "test fires" (possibly in conjunction with a particulate mass) can be input to the base of the cylindrical volume, and the transport of both thermal energy and particulate material can be observed in the model ventilation system. Additionally, the gravimetric balance can determine the collected particulate mass on the HEPA filter. Careful collection of particulate material on the internal surfaces yields information on total deposition.

The thermal tests reported in this paper used only a portion of the ventilation system model shown in Fig. 1. The subsystem used comprises the section of 30.5-cm-diam pipe between dampers 5 and 7 (indicated on Fig. 1), the pipe tee joint adjacent to dampers 7 and 8, and the entire section of the 0.6-m-square duct. The gravimetric balance and the exhaust fan are not included. The commercial duct heater, which includes the supply blower, injects thermal energy into the system at damper 5. Dampers 4, 6, and 8 are closed for these tests.

The internal and external wall temperatures are measured at the longitudinal center of each segment of pipe and duct. The thermocouple generally is placed on the vertical center of one wall. In one section of duct, thermocouples are placed on each vertical wall, in the top, and in the bottom of the duct to measure any thermal gradients in the duct cross section. Pipe and duct wall temperature measurements are made at 15 locations. In addition to the wall temperature measurements, the gas flow temperature is measured at the center of each segment of the 0.6-m-square duct. Twenty gage, type J thermocouple wire was used for these measurements, and the external and internal wall thermocouples were mounted by peening. The internal gas flow thermocouple assemblies were made of a 0.64-cm-diam stainless steel sheath with an aluminum head and with a type J thermocouple. All internal thermocouple wire insulation was replaced by a fiberglass sleeve. The gas flow was obtained by a pitot tube in a five-section traverse of equal concentric areas at the system outlet duct with the pressure readings made with an inclined manometer.

The pressure is measured with a 0.0- to 62.3-Pa manometer at the center of each segment of the 0.6-m-square duct and other selected locations, such as on each side of the 90° elbow and on each side of the commercial dampers. These locations were chosen to provide data spatially compatible with data output by the FIRAC code.

The Numerical Model

The FIRAC computer code initially was designed to predict fire-induced transients in nuclear fuel cycle facility ventilation systems. FIRAC simultaneously calculates the gas-dynamic, material transport, and heat transport transients that occur in any arbitrarily connected network system subjected to a fire. The network system includes ventilation system components such as filters, dampers, ducts, and blowers. These components are connected to the rooms and corridors to complete the network for moving air through the facility.

We used the lumped-parameter method to describe the airflow system. No spatial distribution of parameters within the network components is included in this approach. Network theory defines system elements that exhibit flow resistance and inertia, or flow potential, as branches. The ventilation system components contained in branches include dampers, ducts, valves, filters, and blowers. The connecting points of branches are network system elements called nodes and always have a finite volume. Nodes include specific network components that have finite volumes, such as rooms, gloveboxes, and plenums, or the node may contain only the volume of the connecting branches. In addition, system boundaries, where the volume is practically infinite, are specified as nodes. Fluid mass and energy storage at the internal nodes is taken into account by using the equations for conservation of mass and energy. The conservation equations are applied to the room nodes using the lumped-parameter formulation assuming a homogeneous mixture and a thermodynamic equilibrium. An implicit numerical scheme is used to solve for the pressure and density at each node. In the solution algorithm, the flow rate through branches is modeled as a function of the differential pressure and friction factors.

The material transport model in the code estimates the movement of material through the network of ventilation system components. The code calculates material concentrations and material mass flow rates at any location in the network. This model includes convective transport, depletion by gravitational settling, entrainment from ducts, and filtration. No phase transitions or chemical reactions are modeled.

The code's heat transfer model predicts how the combustion gas in the system cools as it flows through the network ducts. The model predicts the temperature of the gas leaving any section of the duct if the inlet temperature and gas properties are known. The following heat transfer processes are modeled.

- Forced convection between the gas and the inside duct walls
- Radiation between the gas and the side duct wall
- Heat conduction through the duct wall
- Natural convection from the outside duct wall to the surrounding air
- Radiation from the outside duct wall to the atmosphere

The total amount of energy removed from the gas as it flows through the duct is given by the solution of a set of four coupled nonlinear algebraic equations. These equations are solved using an iterative procedure.(1)

The network system model of the experimental system is shown in Fig. 2. The model consists of 25 nodes (including 2 boundary nodes) and 24 branches. We divided the 30.5-cm-diam pipe into 4 branches and the duct into 14 branches (typically 2.44-m segments). We modeled each tee joint and bend as a separate branch because this allows an accurate determination of the resistance coefficients for these branches. The heater is simulated by a temperature-time and mass-time history input at Node 8.

Discussion of Experimental and Computational Results

The experimental sequence began with the duct heater blower running until steady-state flow was reached. with the blower continuing to run, a 30-min heat pulse then was injected into the system at a predetermined thermal rate. The blower then remained on, and the system returned to ambient conditions. Gas flow and wall temperatures typically were recorded by the data acquisition system at 40-s intervals throughout the experiment, and pressures and flow rates were measured manually during each phase of the experiment. The steady-state volumetric flow rate measured before the heat pulse was 0.085 m^3/s, and the pressure drop across the system was about 38 Pa. At the end of the 30-min heat pulse, the flow rate was 0.179 m^3/s with a pressure drop across the system of 100 Pa. As the system cooled, the measured flow rate decreased to 0.080 m^3/s.

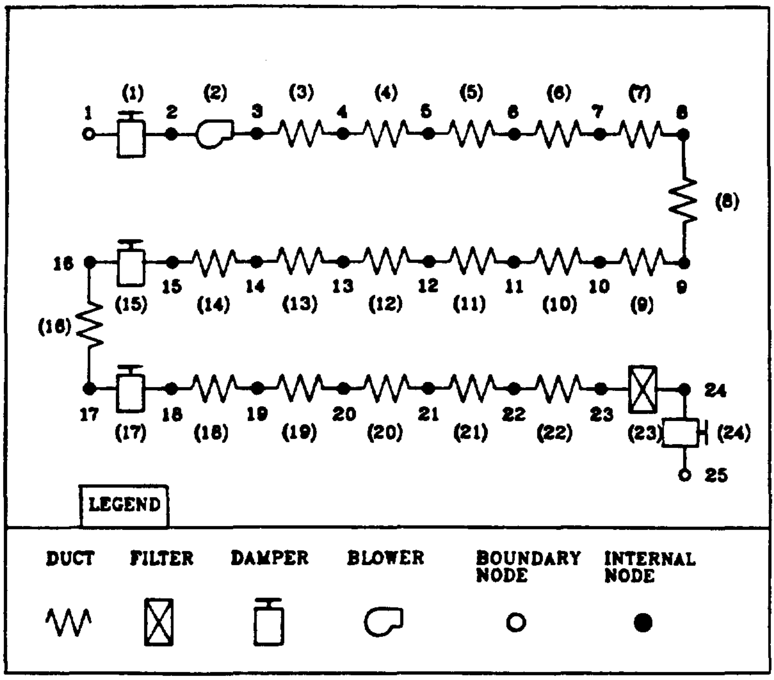


Figure 2. Network system model of the experimental system.

The numerical simulation of the experiment using FIRAC began with the experimentally determined steady-state flow, temperature, and pressure as initial conditions. A temperature-time function representing the measured temperature during and after the heat pulse was input into the network model node 8 (Fig. 2), which corresponds to the temperature at the center of the first section of the 0.6-m-square duct. This is the section of duct connected to the 30.5-cm-diam pipe.

A comparison of the experimental and the FIRAC-generated temperatures as a function of time and space showed generally good agreement. In Fig. 3, the gas flow temperature as a function of time is shown for nodes 11, 14, and 22. (In Fig. 1, node 11 is in the center of the section of duct downstream of the 90° bend, node 14 is in the center of the section of duct upstream of commercial damper 9, and node 22 is in the center of the section of duct just upstream of the filter and gravimetric balance). The code predictions for the temperatures at nodes 11 and 22 are within 5% of the experimental data during the heat pulse and consequent rise in temperature. The node 11 error increases to nearly 12% as the temperature decreases. Because the nodes with the highest and lowest temperatures agree relatively well with the experimental values during the heat pulse, we were surprised that node 14 showed a larger error--about 12% at the peak temperature. We checked the nodes adjacent to node 14, and this indeed is the error pattern, which is shown in Fig. 4. Here the temperature is plotted as a function of duct length at three selected times: at the end of the heat pulse, 150 s after the heat pulse, and 830 s after the heat pulse. The largest errors (up to 12%) are between 9 and 18 m, which corresponds to the locations of nodes 11 through 16.

The FIRAC code duct heat transfer model simulates the amount of thermal energy the gas loses as it passes through a section of duct. This energy loss is a function of radiation and convective processes on the inside and outside of the duct wall and heat conduction through the wall. The computed temperature loss is directly proportional to the energy loss and inversely proportional to the mass flow rate. Heat loss is not modeled at branches other than ducts. The model for heat conduction through the duct wall is based on standard models such as Patankar's.(3) Temperatures at several nodal points through the wall can be calculated with this model. Because the wall is only 0.64 cm thick, the measured temperature differential across the wall is always less than 1.0°C. Therefore, we modeled the wall with only one nodal point; thus, the calculated temperature is the average duct wall temperature.

The measured temperature-time history for the internal duct walls at node 14 is shown in Fig. 5. The general temperature difference at the walls is the pattern expected; however, the magnitude of the temperature difference (23°C) between the top and bottom walls of the duct is larger than expected. The front and back (vertical) walls should have the same value and are within 2°C at all times. The four wall temperatures at node 14 were averaged to compare them with the computed average. The experimental and computed temperatures as a function of time are shown in Fig. 6. The slope of the computed curve is less than that of the experimental curve both during and after the heat pulse. A maximum error of about 10% is seen at late times.

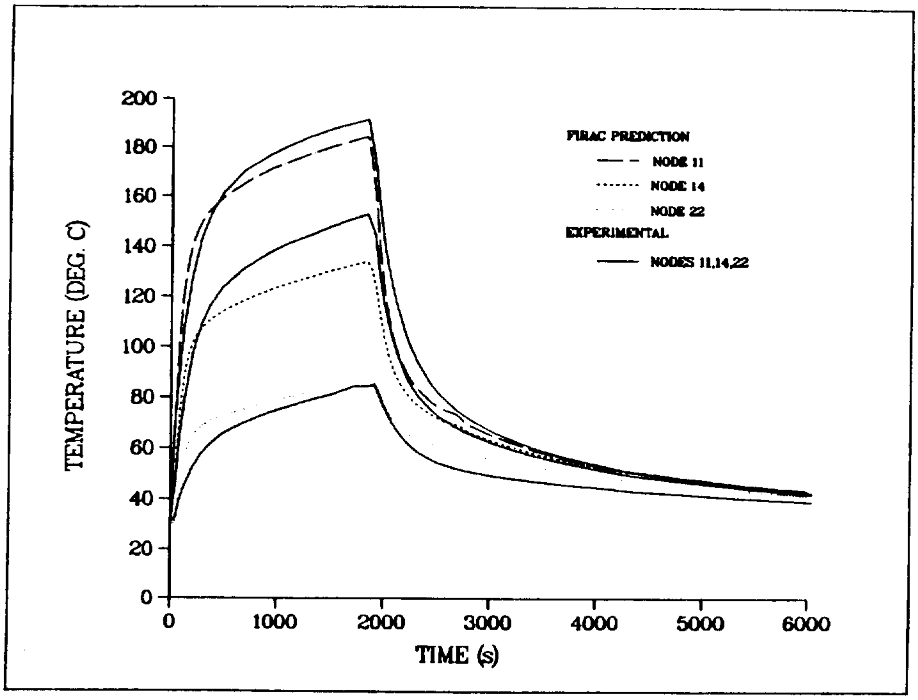


Figure 3. Experimental and FIRAC generated gas flow temperature-time histories for network system nodes 11, 14, and 22.

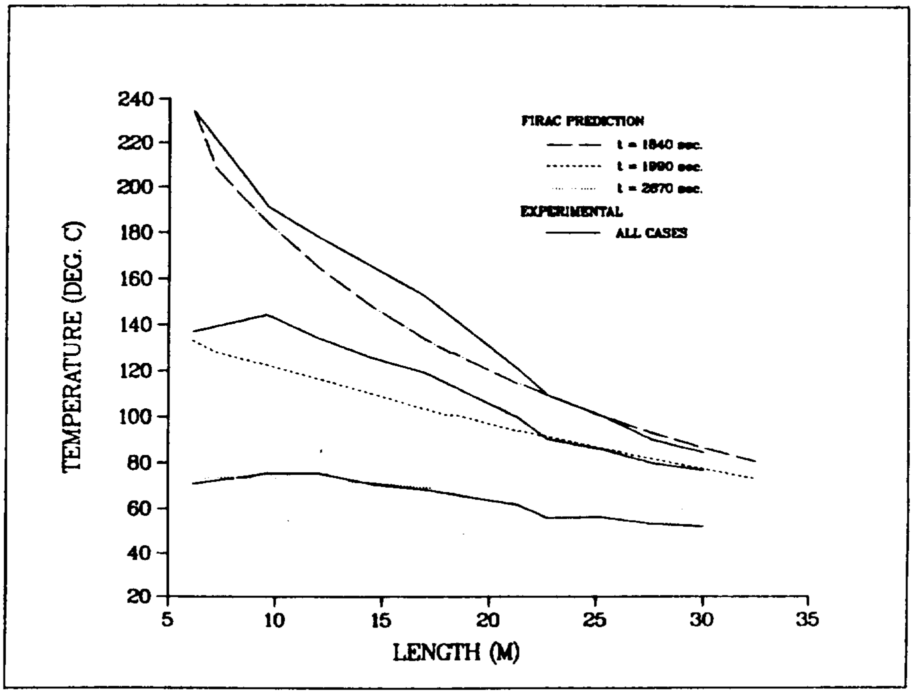


Figure 4. Experimental and FIRAC generated gas flow temperatures as a function of duct length at times 1840, 1990, and 2670 seconds after beginning of heat pulse.

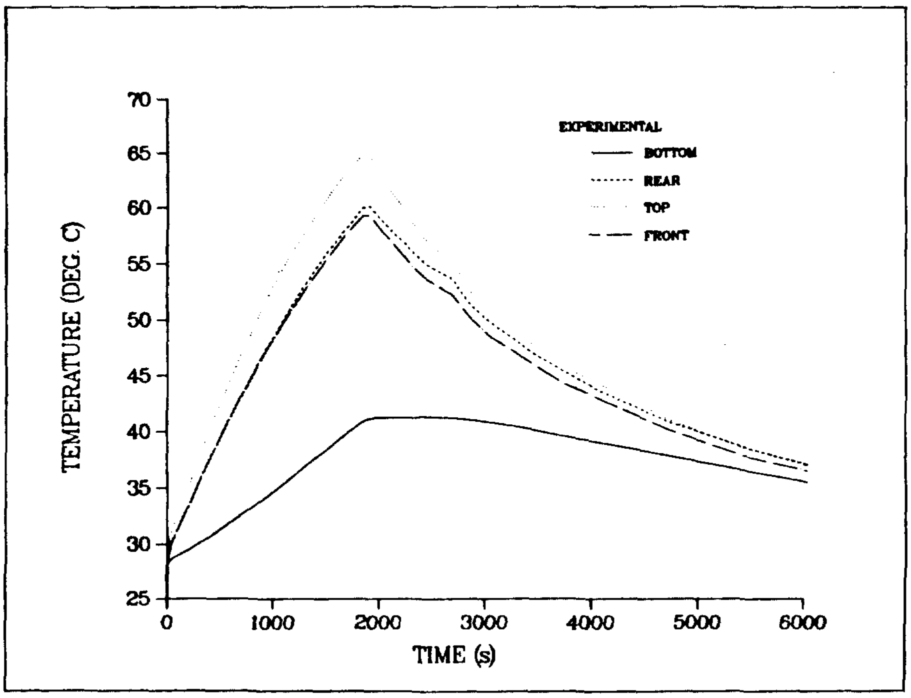


Figure 5. Measured temperature-time histories for the internal duct walls at network system.

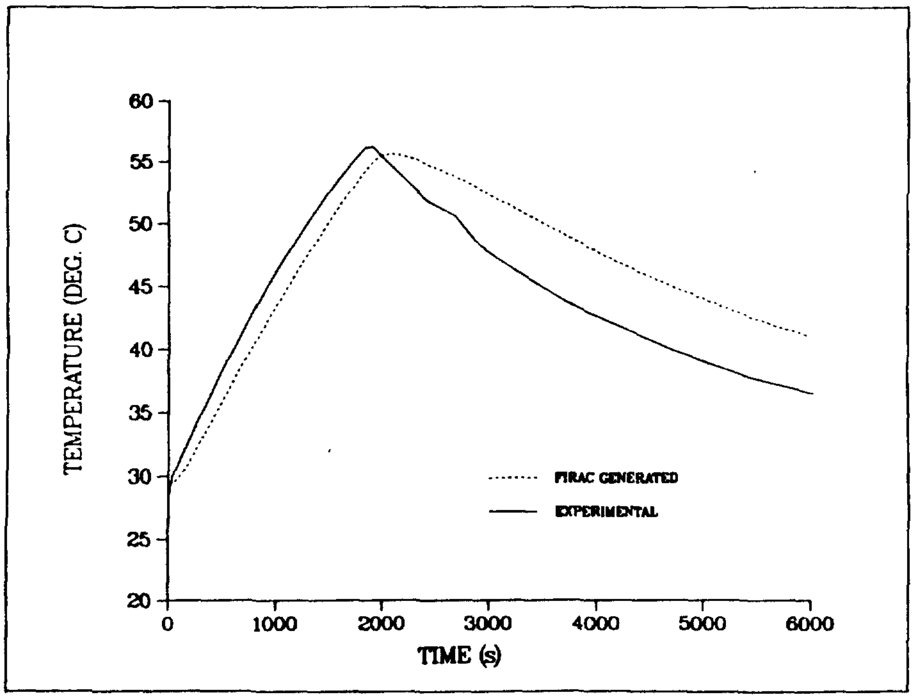


Figure 6. Experimental and FIRAC generated average wall temperature-time history at network system node 14.

Conclusion

The FIRAC computer code has successfully simulated the heat transfer measured in a single experiment with the ventilation system model at NMSU. This suggests that the heat transport model in FIRAC is a reasonable one; however, we need to perform additional experiments at different flow rates and thermal injection rates to further test this model. We then need to verify the other code capabilities, specifically, the material transport model.

References

1. Nichols, B. D. and Gregory, W. S., "FIRAC User's Manual: A Computer Code to Simulate Fire Accidents in Nuclear Facilities," Los Alamos National Laboratory report LA-10678-M (April 1986)

2. Chan, M. K., Ballinger, M. Y., Owczarski, P. C., and Sutter, S. L. "User's Manual for FIRIN: A Computer Code to Estimate Accidental Fire and Radioactive Source Term Releases in Nuclear Fuel Cycle Facilities," Pacific Northwest Laboratory report NPNL-4532 in preparation.

3. Patankar, S. V., Numerical Heat Transfer and Fluid Flow, McGraw-Hill Book Company, Inc., New York, Chap. 4, 1980.

DISCUSSION

PARTHASARATRHY: In the beginning of the paper you referred to material transport, does the code have a capability to predict size distribution and mass concentration of aerosols generated in a fire?

NICHOLS: The FIRIN code, developed at Pacific Northwest Laboratories, predicts the mass generation rate and the size distribution of the particles, and then the information is used as input into the FIRAC code. The FIRAC code does calculate the transport of multiple species of particles of different densities and different sizes, up to about 20 or so different species.

PARTHASARATHY: Will further experiments be concluded to verify this aspect?

NICHOLS: Yes. As soon as we conclude these experiments, we will try in the next series to test the particle flow model.

METHODS FOR AIR CLEANING SYSTEM DESIGN AND ACCIDENT ANALYSIS

W. S. Gregory and B. D. Nichols
Los Alamos National Laboratory
Los Alamos, New Mexico

Abstract

This paper describes methods, in the form of a handbook and five computer codes, that can be used for air cleaning system design and accident analysis. Four of the codes were developed primarily at the Los Alamos National Laboratory, and one was developed in France. Tools such as these are used to design ventilation systems in the mining industry but do not seem to be commonly used in the nuclear industry. For example, the Nuclear Air Cleaning Handbook is an excellent design reference, but it fails to include information on computer codes that can be used to aid in the design process. These computer codes allow the analyst to use the handbook information to form all the elements of a complete system design.

Because these analysis methods are in the form of computer codes, they allow the analyst to investigate many alternative designs. In addition, the effects of many accident scenarios on the operation of the air cleaning system can be evaluated. These tools originally were intended for accident analysis, but they have been used mostly as design tools by several architect-engineering firms.

The Cray, VAX, and personal computer versions of the codes, an accident analysis handbook, and the codes' availability will be discussed. The application of these codes to several design operations of nuclear facilities will be illustrated, and their use to analyze the effect of several accident scenarios also will be described.

Introduction

Ventilation and air cleaning systems can be highly complicated and involve many interconnected flow pathways, rooms, flow controllers, filters, and blowers. The flow arrangement may use parallel and series systems, separate supply and exhaust systems, recirculation, and makeup air. Heating, ventilating, and air conditioning (HVAC) analysts and designers are capable of designing a highly complicated ventilation system. However, the ventilation and air cleaning systems for nuclear facilities require special design considerations and must be analyzed for the effects of hypothetical accident situations.

The "Nuclear Air Cleaning Handbook" provides information of special interest to designers of nuclear air cleaning systems.(1) The possibility of and concern for accidents such as earthquakes, tornados, fires, and explosions are discussed, but no methods are offered to analyze the entire ventilation system for these accident conditions. We believe that there are analytical tools in the form of handbooks and computer codes that will allow ventilation system designers and safety analysts to perform the necessary accident effect calculations. Further, there is no reason why the steady-state portions of the computer codes could not be used to aid the designer in determining the pressures, flows, and temperatures throughout the ventilation system. In addition, these codes would allow the designer to examine many alternative arrangements and subsystems. Perhaps the best part of these methods is that the analyst then can use the same codes to evaluate the potential effects of hypothetical accidents on the system design. In other words, both the system design and the safety analysis could be performed in parallel.

We will describe several computer codes that are available to perform both design and safety analyses. Several examples of applications of these codes will be illustrated.

Computer Code Descriptions

Computer codes that have been developed at the Los Alamos National Laboratory and at the Commissariate a l'Energie Atomique (CEA) in France will be described. All of these codes require a steady-state or normal operating condition calculation. This part of the analysis can be used by designers to verify designs or examine alternative ones. The transient calculations performed by the codes are optional and are especially developed to simulate large pressure surges, explosions, and fires. The codes are presented according to which accidents they are designed to model.

Tornado Modeling, the TORAC Code(2)

TORAC is an improved version of the TVENT computer code, which was developed at Los Alamos.(3) There are three basic differences between TVENT and TORAC:

- material transport capability,
- blowers can be turned on and off, and
- dampers can be controlled with an arbitrary time function.

The TORAC code can model large pressure surges or simulate the effects of a tornado depressurization at the inlet and exhaust points of a ventilation system. The capability to modify the effects of blowers and dampers within the system gives the designer a tool to examine alternative system designs, effects of equipment failure, and multiple damper control points.

The material transport aspects of the code are very basic. That is, only the convection, gravitational settling, entrainment, and filter depletion mechanisms are modeled. The complex interactions between material species and within material species are not modeled. TORAC and TVENT are available from the National Energy Software Center (NESC). CDC 7600 and CRAY versions are available.

Explosion Modeling, EVENT84(4) and EXPAC[5]

Two Los Alamos codes can be used to simulate explosions within air cleaning systems. EVENT84 simulates the gas dynamics of an explosion; EXPAC adds material transport to the calculation. The explosion codes are more complicated than the TORAC code because more detailed data are needed to complete the calculation. Like TORAC, these codes also obtain the flows, pressures, and temperatures in a normal operating condition before any transient calculations are performed. These codes can calculate the propagating effects of solid or gaseous explosions within an air cleaning system or through any air pathway. EVENT84 and EXPAC currently are being verified by selected experimental studies. EVENT84 is available from the NESC, but the EXPAC code is still under development. CDC 7600 and CRAY versions are available.

Fire Modeling, PIAF(6) and FIRAC(7)

Two computer codes have been developed to simulate the effects of fires within nuclear facility air cleaning systems. One was developed at CEA and is named PIAF. It was developed to study the responses of ventilation networks to mechanical or thermal perturbations.

Using the PIAF code to model mechanical systems in facilities has proven useful because it can identify events that are of safety concern and quantify the possible damage within the facility. A steady-state analysis of a large ventilation network is described in Ref. 6. A separate paper describing the PIAF modeling of the ventilation network operation is to be presented at this conference. This code's availability is unknown.

A second code to simulate fire conditions was developed by Los Alamos. This code is named FIRAC and soon will be available from NESC. It has been modified to run on a personal computer. CRAY, VAX, and CDC 7600 versions are available. FIRAC includes material transport and uses a burn room model developed by Battelle Pacific Northwest Laboratories. The code uses the basic gas dynamics available in EVENT84 but adds heat transfer from the system ductwork.

Handbook

An accident analysis handbook has been developed to assist safety analysts in modeling and analyzing nuclear facilities.(8) The emphasis has been placed on fuel cycle facilities, but other facilities also can be modeled. The handbook contains information on fuel cycle operations and processes as well as methods to develop radioactive source terms for fire and explosion events. Modeling techniques, many sample problems, and information on how to use particular computer codes are also included. This handbook is available in draft form from the Nuclear Regulatory Commission and currently is being revised for final form. (A completion date of October 1986 is planned.)

Applications

Design Simulation

An example of a design simulation using one of the TVENT codes involves determining the effect of a recirculation blower shutting off and the effect on room key pressures when the main supply blower slows down.(9) A study of these situations and their effects on part of the Los Alamos Plutonium Processing Facility are reported in Ref. 9. The results of this study showed that loss of the recirculation blower did not have a significant effect on the overall system pressures. (A 10% slowdown in the main supply blower caused a sharp reduction in corridor and compartment pressures.) This information allowed the analyst to identify certain control dampers to control large pressure fluctuations. These calculations could aid in the design of the system and the placement of system controllers.

A second design simulation involves using the FIRAC code to simulate the operation of a proposed radioactive waste incinerator. In this case, the FIRAC results were not the same as the design (because of a lack of extensive design data) but were within a reasonable range. Using FIRAC in parallel with the incinerator design can prevent design flaws that could appear when a safety analysis is performed after the incinerator is constructed.

Accident Analysis

The design simulation of the incinerator discussed above can be used to illustrate how an accident analysis can be performed in parallel with the design phase. In this case, we examined the possibility of an inadvertent injection of an explosive mixture into the pyrolysis chamber of the incinerator. In this accident scenario, we have assumed that a container of 100 cm^3 of toluene is inserted into the process line. After performing the analysis to determine the effects of this scenario, we found that a positive pressure is created in the pyrolysis and combustion chambers. This accident analysis scenario shows that a positive pressure in the pyrolysis and combustion chambers causes a flow reversal through the pyrolysis and combustion inlet air paths. Therefore, protective devices would have to be designed into the system to prevent a radioactive release to the atmosphere when the incinerator chambers overpressurize. A second accident scenario also was examined. This involved failure of the exhaust blower for the system. Again, this caused a pressure increase in the incinerator chambers.

In Ref. 6, an accident analysis using the PIAF computer code is described. The report discusses evaluations of the effects of a 9-kg and a 26-kg fire in gloveboxes. The effects of cell temperature, pressure, exhaust flow rate, and filter pressure drop are determined in these calculations. Future modifications to the code will include aerosol release calculations.

Summary

We have described analysis methods that can be used in the design and accident evaluation of nuclear ventilation and air cleaning systems. Applications of these analytical methods, in the form of computer codes, are given for both design and accident analysis. Four of these computer codes have been developed by Los Alamos, but one code developed in France also is discussed. We feel that future editions of the Nuclear Air Cleaning Handbook could benefit greatly if a chapter is devoted to use of analytical methods that bring the entire nuclear air cleaning system together for analysis. Evaluation of the system as a whole for both design effectiveness and accident analysis could lead to more efficient and safer designs.

References

1. Burchstead, C. A., Fuller, A. B., and Kahn, J. E., "Nuclear Air Cleaning Handbook," Oak Ridge National Laboratory report ERDA 76-21 (March 1976).

2. Andrae, R. W., Tang, P. K., Martin, R. A., and Gregory, W. S., "TORAC User's Manual--A Computer Code for Analyzing Tornado-Induced Flow and Material Transport in Nuclear Facilities," Los Alamos National Laboratory report LA-10435-M, NUREG/CR-4260 (May 1985).

3. Duerre, K. H., Andrae, R. W., and Gregory, W. S., "TVENT--A Computer Program for Analysis of Tornado-Induced Transients in Ventilation Systems," Los Alamos National Laboratory report LA-7397-M (December 1984).

4. Martin, R. A. and Wilson, T. L., "EVENT84 User's Manual--A Computer Code for Analyzing Explosion-Induced Gas-Dynamic Transients in Flow Networks," Los Alamos National Laboratory report LA-10312-M (December 1984).

5. Nichols, B. D. and Gregory, W. S., "EXPAC User's Manual--A Computer Code to Predict Gas-Dynamics and Material Transport in Flow Networks," Los Alamos National Laboratory report in preparation.

6. Perdriau, Ph., Pourprix, M., Rabain, S., Rouger, J. L., and Tarrago, X., "Safety Evaluation of Ventilation Networks in Case of Fire," Proceedings of the CSNI Specialist Meeting on Interaction of Fire and Explosion with Ventilation Systems in Nuclear Facilities, Los Alamos National Laboratory report LA-9911-C (April 1985), Vol. 1.

7. Nichols, B. D. and Gregory, W. S., "FIRAC User's Manual: A Computer Code to Simulate Fire Accidents in Nuclear Facilities," Los Alamos National Laboratory report LA-10678-M, NUREG/CR-4561 (April 1986).

8. "Fuel Cycle Facility Accident Analysis Handbook," US Nuclear Regulatory Commission report NUREG/CR-2508 (Draft) (March 1982).

9. Andrae, R. W. and Rojas, V. A., "Analytical Study of Ventilation System Behavior Using TVENT1 Under Simulated Blower Malfunctions," Los Alamos National Laboratory report LA-9653-MS (February 1983).

CLOSING COMMENTS OF SESSION CO-CHAIRMAN KRATZKE

There were four papers in the first part dealing with iodine and iodine retention in a fire situation. We learned about various points of view. One is held by those engaged in design. They cited tests conducted to look at the characteristics of the releases from a burning carbon bed. From the insurers point of view, once a fire starts, there will be serious problems. Therefore, every effort must be made to contain and reduce the problem. It is important to inquire whether basic theoretical studies have been conducted to examine these phenomena theoretically and then to extrapolate the findings broadly rather than merely looking at individual systems from a purely operational perspective. In other words, do we thoroughly understand the mechanisms that are associated with the phenomena? That, I believe, covers the first four papers.

The next five papers are closely related. They seek to model accidents and thereby to assist analysis and design. The first paper, from France, looked at confinement and the maintenance of confinement. They examined ventilation systems and pointed out weak points. Next, we heard three papers from Los Alamos that dealt with computer simulation and the use of that technology for design and accident analysis.

I want to conclude by saying something about the paper by Dr. Gruendler from West Germany. He looked at the release of radioactivity from a waste package, a different topic than air cleaning systems. However, he is looking at the generation of a source term that could challenge air cleaning systems. I thought the paper was interesting because it looked at some of the basic factors and consolidated them into simplified mechanisms that could become useful calculational tools.

19th DOE/NRC NUCLEAR AIR CLEANING CONFERENCE

SESSION 3

ADSORPTION, ADSORBERS, IODINE RETENTION

MONDAY: August 18, 1986
CHAIRMEN: J.L. Kovach
Nuclear Consulting Service
W.L. Anderson
Consultant

REMOVAL OF RADIOACTIVE GASES BY ZEOLITE
Y.C. Fan, T.Y. Lee, C.S. Tan, C.M. Hsai

IN SITU MEASUREMENTS OF EFFICIENCY OF SILVER ZEOLITE FOR REMOVAL OF AIRBORNE RADIOIODINE SPECIES
J.W. Mandler, J.W. Tkachyk, J.T. Case, J.H. McDaniel, H.K. Peterson

SUMMARY AND RECOMMENDATIONS OF THE NRC/INEL ACTIVATED CARBON TESTING PROGRAM
C.D. Scarpellino, C.W. Sill

NUCLEAR-GRADE, GAS-PHASE ADSORBENT IODINE RETENTION TEST
J.A. Slade, H.M. Philippi, M.J. Kabat

THE U. V. SPECTROMETRIC METHOD OF ANALYSING CHEMICAL IMPREGNANTS OF A NUCLEAR GRADE ACTIVE CARBON AND ITS APPLICATIONS FOR MANAGING THE CARBON FILTER AT A NUCLEAR REGULATORY LAB. AND PLANTS
Y.S. Kim

REMOVAL OF ORGANIC IODINE COMPOUNDS FROM FLOWING AIR STREAMS
W.P. Freeman, M.P. King, J.L. Kovach

INVESTIGATIONS OF THE AGING OF VARIOUS ACTIVATED CARBONS IN THE EXHAUST OF A PWR OVER AN EXTENDED PERIOD OF TIME
H. Deuber, K. Gerlach, V. Giraud, R. Kaempffer

DEPTH PROFILE OF METHYL IODIDE-131 IN THE TESTING OF USED AND REGENERATED ACTIVATED CARBONS
V.R. Deitz

MULTIPLE USE FILTER CELL - COMPARATIVE MEASUREMENTS
H.-G-Dillmann, H.Pasler

PERFORMANCE STUDY ON ACTIVATED CARBON FILTER SYSTEMS AT THE U.S. ARMY CHEMICAL AGENT MUNITIONS DISPOSAL SYSTEM (CAMDS)
D.J. Gladden, Capt. M. Post,

REMOVAL OF RADIOACTIVE GASES BY ZEOLITE

Fan, Y.C.*, T.Y. Lee, C.S. Tan
Department of Chemical Engineering
National Tsing Hua University
Hsinchu, Taiwan

and

C.M. Hsai
Atomic Energy Council
Radwaste Administration
Taipei, Taiwan
R.O.C.

Abstract

Zeolite is proposed for the treatment of radioactive gases emitted from an atomic power plant. Two types of zeolites, 4A and mordenite were chosen for the adsorption study of simulated gases Ar, Kr and CH3I. A DuPont electron-microbalance was employed for the sorption rate and equilibrium measurements. Diffusion study of gas into zeolite was carried out in low pressure. Temperature ranges were 40°-70°C for zeolite 4A, 50°-70°C for mordenite. Isotherms at different temperatures were used to calculate the heat of adsorption. Fick's law decribing the mass transfer of gas into porous solid was employed to obtain the diffusivity from weight-uptake rate curves. Subsequently, the activation energy of diffusion could be evaluated. The order of effective diffusivity and activation energy of diffusion were found to be 10^{-10}-10^{-14} cm^2/sec and 5-10 Kcal/mole respectively in this study. An adsorption column model which took into consideration of the residence time and half-life of the radioactive gas was formulated and the column breakthrough curve was calculated. Consequently, the equilibrium and rate data obtained in the present study may be applied in the actual design of an adsorber for control of the radioactive gases from a nuclear power plant.

I. Introduction

Waste gases from nuclear power plant usually contain certain amount of radioactive components, such as Ar, Kr and CH_3I, etc. In the case that the level of radiation is above safety, treatment of these radioactive wastes is required before releasing them into the atmosphere. One of the applicable method is to encapsulate these radioactive components by solid adsorbents. Among all adsorbents, zeolites were found to possess the properties of high sorption capacity, special selectivity, good temperature stability and strong acid resistance. In addition, they can be easily regenerated to undertake long service cycles. Therefore, zeolites were proposed to be the potential sorbents in treating radioactive gases.

The study of adsorption and diffusion in zeolites was done by Barrer and his coworkers initially. Barrer and Ibbitson (1944) studied the sorption of various gases on chabazite and analcime. They reported that in order of increasing affinity for the zeolite lattice, one may write for a series of non-polar gas as $H_2<He<Ar$ or $O_2<N_2$. Barrer and Brook (1953) studied the sorption kinetics of C_3H_8, $n\text{-}C_4H_{10}$, CH_2Cl_2 and $(CH_3)_2NH$ in chabiazite, mordenite and levynite at room temperature, -78°C, and -185°C. They reported the diffusivities of the gases and the activation energy for diffusion increases as the chain length of the sorbate molecules increases. Iodine occlusion by chabazite was measured by Barrer and Wasilewski (1961 a,b) in the temperature range of 120°-300°C. The partial molal heat of occulsion $\Delta\bar{H}$ was determined from the adsorption isotherms. Very large value of $\Delta\bar{H}$ was reported when the amount of iodine sorbed was low. Barrer and Coughlan (1968) investigated the effects of removal of cations and anions framework charge from clinoptilolite upon the sorption characteristics of polar guest molecule CO_2 and non-polar molecule Kr. The sorbent became progressively more energetically homogeneous towards carbon dioxide as a result of the removal of the charge from the zeolite. On the other hand, all the acid-treated zeolites showed energetic homogeneity towards Kr. Barrer and Vaughan (1971) investigated the sorption and diffusion characteristics of phillipsite along with zeolite K-M and othr silicates for the trapping and storage of inert gases. After outgassing, the zeolite was exposed to high pressures of inert gas at high temperature and then quenched while still under pressure. Both sorption isotherm and diffusion coefficient were measured. Barrer and Papadopoulos (1972) obtained the equilibrium isotherms of Kr and Xe in chabazite in the temperature range of 150-450°C and at pressures up to 100 atm. Barrer and Robins (1953) studied the single gases H_2, Ne, O_2 and N_2 adsorption and diffusion in mordenite, O_2, Ar and N_2 in chabazite and binary gases H_2+N_2, H_2+Ne, N_2+O_2 sorption in mordenite, and $Ar+O_2$, $Ar+N_2$ in chabazite.

Elberly (1969) employed the chromatographic technique to study the sorption and diffusion properties of Ar, Kr in 5A and SF_6 in 13 . Sarma and Hagnes (1974) also used moment technique to investigate the Ar sorption and diffusion characteristics in 4A. Ruthven and Derrah (1972) studied the diffusion properties of Ar, Kr in 4A and Ar, Kr, Xe in 5A.

In this study, two types of zeolites, 4A and mordenite were chosen for sorption of simulated radioactive Ar, Kr and CH3I gases. A Du Pont electronmicrobalance was employed for sorption rate and equilibrium measurements. Diffusion study of gas into zeolites was carried out in low pressure (below atmospheric). Temperature ranges were 40°-70°C for zeolite 4A, 50°-70°C for mordenite. The equilibrium adsorption and diffusion data were fed into a mathematical model of adsorption column which took into consideration of the half-life and level of radiation of the gas. Consequently, the break through radiation counts can be adjusted to meet the pollution control standard by simulation of the column adsorption model.

II. Experimental Section

The adsorbent, chemicals, apparatus and procedure used in the experiment will be stated below.

1) Adsorbents and chemicals

Zeolite 4A and mordenite were purchased from Davison Chemical, W.R. Grace & Co. and Norton Co. respectively. The 4A zeolite is three-dimensional channels of approximately 0.4 μm openning and the mordenite is one-dimensional channels of 0.67x0.7 μm interconnecting with another 0.29x0.57 μm one-dimensional openning.

Research grade of Ar, Kr and CH3I gases of purity 99.9% were purchased from Matheson Chemicals.

2) Apparatus

The weighing instrument is a modified DuPont TGA with improved sealing. The set-up consists of nine parts: 1-fast response recorder , 2-microbalance, 3-gas container with heater and insulation, 4-low pressure gauge, 5-gas cylinder, 6-vacuum gauge, 7-vacuum pump, 8 and 9-variable resistors. The complete set-up is shown in Fig. 1.

3) Procedure

Before a run, the careful calibration of the micro-balance is essential. Approximately 10 mg adsorbent was placed in the sample holder. The vacuum pump was started until the system reached 0.01 mmHg then the sample was heated to 400°C for about 10 hours, meanwhile, the system was maintained at low vacuum and degased. The dry weight of the sample was recorded. After thoroughly degased, the sample was cooled to the desired temperature and maintained at that temperature for about half of an hour. Then, the hydrocarbon gas was injected into the adsorption chamber for the sorption and diffusion study.

At a given injection pressure, the rate of adsorption is determined by the increase in weight of the sample with time. The total amount of gas adsorbed is plotted against equilibrium pressure as the adsorption isotherm. At a given low injection pressure the sorption rate curve was determined by the weight increase of the sample vs. time.

In all the runs, the amount of adsorbent was relatively small in comparison to the volume of the adsorbate. There was no appreciable change in the pressure of the gas chamber. Consequently, all the diffusion runs could be considered as under constant pressure-constant volume condition. All the diffusion runs were in the low pressure region in order to ensure the validity of the Henry's law for adsorption.

III. Correlation Equations

1) Equilibrium

Langmuir isotherm

$$\frac{S}{S_\infty} = \frac{KP}{1+KP}$$

was employed to correlate the concentration in the solid with the partial pressure in the gas phase. The S_∞ and K were obtained by least square fitting of the experimental isotherm.

2) Transient Diffusion

The rate of sorbate entering the sorbent was correlated by the second Fick's law under constant volume and constant pressure condition. The assumptions involved in the mass balance equations are

i) Solid adsorbent is spherical.

ii) Fick's law is applicable.

iii) Solid-gas film resistance can be neglected.

iv) The mobile phase and immobile phase in adsorption is related by Henry's law.

v) The effective diffusivity including pore volume and surface diffusion is constant.

vi) Adsorption equilibrium is instantaneously achieved.

The mass balance in the spherical adsorbent is

$$\varepsilon_p \frac{\partial C_r}{\partial t} + \rho_p \frac{\partial S}{\partial t} = \frac{\varepsilon_p}{r^2} \frac{\partial}{\partial r}\left(r^2 D_p \frac{\partial C_r}{\partial r}\right) \tag{1-1}$$

the initial and boundary conditions are

$$t=0,\ C_r=0 \tag{1-2}$$

$$r=0,\ \frac{\partial C_r}{\partial r}= 0 \tag{1-3}$$

$$r=R,\ C_r=C_b^o \tag{1-4}$$

and the relationship between C_r and S is

$$S=(KS_\infty RT)C_r= K'C_r \tag{1-5}$$

The solution for the system equation (1) was obtained by Crank (11)

$$\frac{M_t}{M} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-D_e n^2 \pi^2 t/R^2}$$

where

$$D_e = \frac{\varepsilon_p D_p}{\varepsilon_p + \rho_p \bar{K}'}$$

$$M_t = C_b^o - C(t)$$

$$M_\infty = C_b^o - C_\infty$$

A least square procedure which minimized the $\Sigma[(\frac{M_t}{M_\infty})_{model} - (\frac{M_t}{M})_{experiment}]^2$ was employed in order to obtain the best De.

3) Heat of Adsorption and Activation Energy of Diffusion

The isosteric heat of adsorption can be obtained by the following equation

$$\left(\frac{\partial \ln P}{\partial T}\right)_\pi = \frac{\Delta H}{RT^2}$$

where

π = at a certain coverage or spreading pressure

Assuming the diffusivity is dependent on temperature in the Arrhenius equation, then

$$D_e = D_e^o \exp(-E_a/RT)$$

The D_e^o and E_a can be obtained from the intercept and slope of the logarithmic plot of D_e vs. 1/T respectively.

4) Radiation Breakthrough Curve

The normal practice of measuring the radiation of a radioactive substance is by γ spectroscopy. The intensity of the radiation is correspondent to the peak area of the spectrum. After corrections of the geometric configuration and the efficiency of the γ-spectroscopy, the radiation activity ϕ is related to concentration by

$$\lambda \phi = C$$

where $$\lambda = \frac{\ln 2}{t_{1/2}}$$

Consequently, the quantity $\phi = \frac{C}{\lambda}$ is used for the column break through study instead of C alone. Hence the system of equations of ϕ balance in the column was

In the zeolite particle

$$\frac{\partial \phi_r}{\partial t} = D_e \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \frac{\partial \phi_r}{\partial r}) \qquad (2\text{-}1)$$

Initial and boundary conditions are

$$t = 0, \quad \phi_r = 0 \qquad (2\text{-}2)$$

$$r = 0, \quad \frac{\partial \phi_r}{\partial r} = 0 \qquad (2\text{-}3)$$

$$r = R, \quad D_e \frac{\partial \phi_r}{\partial r} = k_f (\phi - \phi_s) \qquad (2\text{-}4)$$

In the gas phase

$$\varepsilon_B \frac{\partial \phi}{\partial t} + \varepsilon_B U \frac{\partial \phi}{\partial z} = \varepsilon_B D_L \frac{\partial^2 \phi}{\partial z^2} - \frac{3k_f}{R} (1-\varepsilon_B)(\phi - \phi_s) \qquad (3\text{-}1)$$

Initial and boundary conditions

$$t = 0, \quad \phi = 0 \qquad (3\text{-}2)$$

$$z = 0, \quad U\phi = U\phi_o - D_L \frac{\partial \phi}{\partial z} \qquad (3\text{-}3)$$

$$z = L, \quad \frac{\partial \phi}{\partial z} = 0 \qquad (3\text{-}4)$$

Linear relationship between gas and solid phase

$$\phi_r \Big|_{r=R} = K S_\infty R T \phi = K' \phi_s \qquad (4)$$

If writen in dimensionless form and simplified, the above system of equations become

$$\frac{\partial Q}{\partial \tau} = \frac{1}{X^2} \frac{\partial}{\partial X} (X^2 \frac{\partial Q}{\partial X})$$

$$\frac{\partial f}{\partial \tau} + \psi\theta \frac{\partial f}{\partial z} = \frac{\psi\theta}{Pe} \frac{\partial^2 f}{\partial z^2} - 3\psi\xi \left(f - \frac{Q|_{r=R}}{K'}\right) \qquad (1\text{-}2)$$

Initial and boundary conditions

$$\tau = 0,\ f = Q = 0 \qquad (1\text{-}3)'$$

$$X = 0,\ \frac{\partial Q}{\partial z} = 0 \qquad (1\text{-}4)'$$

$$X = 1,\ \frac{\partial Q}{\partial X} = 0 \qquad (1\text{-}5)'$$

$$z = 0,\ 1 = f - \frac{1}{Pe}\,\frac{\partial f}{\partial z} \qquad (1\text{-}6)'$$

$$z = 1,\ \frac{\partial f}{\partial z} = 0 \qquad (1\text{-}7)'$$

This system of equations was solved by orthogonal collocation method using four collocation points inside the particle and eight collocation points along the column. A semi-implicit Runge-Kutta method was used to solve the resulting ordinary differential equations.

IV. Results and Discussion

The isotherms of A_r and K_r on zeolite 4A is shown in Fig. 2. in the temperature range of 40^o to 60^oC. The maximum adsorption capacities for A_r and K_r are slightly more than 3. and 1 mmoles per gram respectively. The isotherms of A_r and K_r on mordenite are shown in Fig. 3. They are of roughly the same capacity as in 4A. The adsorption capacity of CH_3I on 4A are shown in Fig 4. In the regeneration step, great difficulty was encountered, the amount of CH_3I adsorbed was extremely difficult to remove. Subsequently, new zeolite was placed in the balance for each run. The adsorption capacity is slightly under 3 mmoles per gram at 46^oC and close to 2 mmoles per gram at 72^oC. The values of heat of adsorption calculated from the equilibrium constants of the isotherms were listed in Table I.

The effective diffusivity calculated from the experimental transient curve was also listed in Table I. The order of magnitude of the effective diffusivity is in the range of $10^{-10}\sim 10^{-14}$ cm^2/sec in this study. The activation energy of diffusion is in the range of 5-10 Kcal/mole. Previous study by Walker et al. (1966) for Ar on KA and K_r on CaA zeolites was approximately 10^{-16}-10^{-18} cm^2/sec and by Ruthven and Derrah (1975) for A_r and K_r on 4A was approximately 10^{-10}-10^{-13} cm^2/sec which were extrapolated respectively to the temperature range in this study. Our results appear to be pretty close to those of Ruthven and Derrah. The order of magnitude in the zeolites can be arranged in the following sequence: $K_r/MN > CH_3I/4A > A_r/MN > A_r/4A > K_r/4A$. It is of interest to know that the pore diffusivity defined as $D_p = D_e \frac{\varepsilon_p + \rho_p K'}{\varepsilon_p}$ is strongly influenced by the equilibrium constant K' and does not show an exponential dependence on temperature. The values of D_p are also listed in Table I. The Arrhenius plot of effective diffusivity is illustrated in Fig. 5. The experimental data and model curves of diffusion are shown in Fig. 6 to 7 representatively. Reasonably good agreement between experiment and diffusion model has been attained.

In the simulated column breakthrough model, the required input information are the gas-sorbent equilibrium isotherm and the diffusion coefficient. Other information on the bulk phase can be estimated from the existing coorelations. The input data for the mathematical model are summarized in Table II. For simplicity, we will treated the radioactive gases one by one. The bulk phase property was considered as the property of the air because of the dilute concentration of the radioactive gases. The breakthrough radiations of Ar, K_r, and CH_3I are shown in Fig. 8 to 11. The column was assumed to have a dimension of 17.1 m height and 1.52 m diameter with a gas flow rate of 150 m^3/hr. Approximately, 5000 Kg of zeolite with particle size of 0.16 cm are packed in the column. Suppose the radioactive gas A_r of 1.903×10^{-5} μCi/ml entering the column packed with 4A zeolite, the beginning breakthrough time is about 4 min. but needs a long time to reach 80% of the initial radiation as indicated in Fig. 8. Samilarly, for A_r in mordenite, K_r in 4A and CH_3I in 4A the beginning breakthrough time are also about 4 mins, but they need even longer time to reach 60% of the initial radiation in the column outlet as indicated in Fig. 9-11. The extremely slow rising to the initial radiation may be due to the small diffusivity of the gas in zeolite and to the relatively fast gas flow rate which resulting early breakout in the column. For illustration, if no more than 10% of the initial radiation is allowed to release from the column, then 4 to 5 min. is allowed for A_r and CH_3I in 4A and several days for A_r in mordenite and K_r in 4A column.

The affinity of K_r and CH_3I toward zeolites is much higher than air. They are preferentially adsorbed and the existence of air can be considered as no interference on the adsorption of the radioactive gases. However, the affinity of A_r toward zeolite is compatible to the N_2 and O_2. Study of the mixture adsorption should be done in order to fully understand the influence of air on the sorption of A_r quantitatively. A longer column may be needed in order to accommodate the N_2 and O_2 adsorptive effects on the A_r gas.

In the mathematical model of the column breakthrough curve, Henry's law constant was used which could be unrealistically large. Hence model calculation of using Langmuir isotherm is under-way and will be reported in the future. Also experimental work of employing a small size column with temperature and flow controller should be carried out in order to obtain experimental data for justification and validation of the theoretical results.

Nomenclature

C	bulk phase concentration
C_b^o	inlet concentration
C_r	pore concentration
C_∞	bulk phase concentration for $t \to \infty$
D_e	effective diffusivity
D_e^o	pre-exponential factor in $D_e = D_e^o \exp(-E_a/RT)$
D_L	axial dispersion coefficient
D_p	pore diffusivity
E_a	diffusional activation energy
f	dimensionless bulk phase concentration, C/C_b^o
ΔH	heat of adsorption
$\Delta \bar{H}$	partial molal heat of occulsion
K	adsorption constant
K'	Henry's law adsorption equilibrium constant
k_f	mass transfer coefficient
L	adsorber length
P_e	Peclet Number, UL/D_L
Q	dimensionless pore concentration, C_r/C_b^o
R	radius of zeolite crystal
r	radius coordinate of zeolite crystal
S	solid phase adsorbate concentration
S_∞	S for $t \to \infty$
T	temperature
t	time
$t_{1/2}$	half life of radioative specie
U	superficial velocity
X	dimensionless radial coordinate, r/R
z	axial coordinate
Z	dimensionless axial coordinate, z/L

Greek symbols

ε_B	void fraction in adsorber
ε_P	particle porosity
θ	dimensioless group, $UR^2\varepsilon_B/LD_eK(1-\varepsilon_B)$
λ	radiation constant
ξ	dimensionless group, $k_f R/D_eK$
π	coverage

ρ_p	particle density
τ	dimensionless group, $D_e t/R^2$
ϕ	radiation activity
ϕ_o	ϕ at z=0
ϕ_r	ϕ at radius r
ϕ_s	ϕ at particle surface

References

[1] Barrer, R.M., and D.A. Ibbitson (1944), "Occulusion of hydrocarbons by Chabazite and Analcite", Trans. Farad. Soc. 40, 195.

[2] Barrer, R.M., and D.W. Brook (1953), "Molecular Diffusion in Chabazite, Mordenite, and Levynite", Trans. Farad. Soc. 19, 1049.

[3] Barrer, R.M., and S. Wasilewski (1961), "Occlusion of Iodine in Some Porous Crystals, Part 2. Entropy and Free Energy Relationship", Trans. Farad. Soc. 57, 1153.

[4] Barrer, R.M., and B. Coughlan (1968), "Molecular Sieves Derived from Clinoptilolite", Molecular Sieves, Soc. of Chem. Ind., London PP 141.

[5] Barrer, R.M. and D.E.W. Vaughan (1969), "Sorption and Diffusion of Rare Gases in Heulandite and Stilbite", Surface Science 14, 77.

[6] Barrer, R.M., and R. Papadopoulis (1972), "The Sorption of Krypton and Xenon in Zeolites at High Pressures and Temperatures: I Chabazite", Proc. Roy. Soc. Lond. A, 326, 315.

[7] Barrer, R.M. and A.B. Robins, (1953), "Sorption of Mixtures: Part 2, Equilibrium between Binary Gas Mixtures and Some Zeolites": Trans. Farad. Soc. 49, 929.

[8] Elberly, P.E., (1969) I/EC. Fund., 8, 25.

[9] Sarma, P.N. and Hagnes, H.W., Adv. in Chem. (1974) 133, 205.

[10] Ruthven, D.M. and R.I. Derrah, (1975) J. Chem. Soc. Farad. Trans. I, 71 2031.

[11] Crank, J. (1957) Mathematics of Diffusion, Oxford, England.

[12] Walker, P.L., Jr., L.G. Austin and S.P. Nandi (1966) Chem. Phys. Carbon 2 257.

[13] Ruthven, D.M. and Derrah, R.I., (1975) J. Chem. Soc. Farad. Trans. I, 71 2031.

Table I Summary Results of Equilibrium and Transient Diffusion

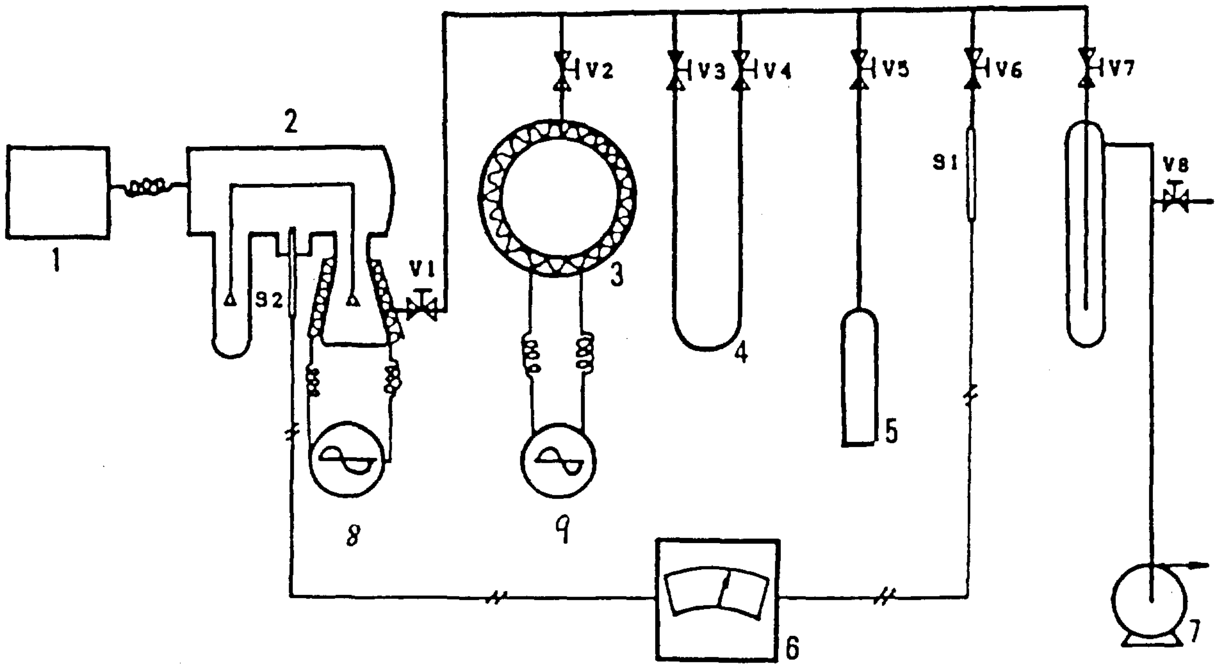
Sorbate	Sorbent	T(^{o}K)	K(1/mmHg)	$S_\infty(\frac{\text{m-mol}}{\text{g}})$	K'(cc/g)	De(cm^2/S)	D_p(cm^2/S)	H (Kca/mol)	Ea (Kcal/mol)
Ar	4A	333	41.55	3.29	2.84×10^{6}	4.07×10^{-14}	3.12×10^{-7}	16.83	7.68
Ar	4A	322	51.65	3.36	3.48×10^{6}	2.87×10^{-14}	2.70×10^{-7}		
Ar	4A	316	185.70	3.43	1.25×10^{7}	2.48×10^{-14}	8.38×10^{-7}		
Kr	4A	333	7.71	1.35	2.16×10^{5}	2.79×10^{-12}	1.63×10^{-6}	12.64	10.70
Kr	4A	323	25.90	1.44	7.52×10^{5}	1.89×10^{-12}	3.84×10^{-6}		
Kr	4A	313	26.46	1.62	8.34×10^{5}	9.92×10^{-13}	2.24×10^{-6}		
Ar	mordenite	334	24.74	3.44	1.77×10^{6}	9.14×10^{-13}	9.82×10^{-6}	15.28	5.47
Ar	mordenite	323	67.25	3.49	4.72×10^{6}	6.70×10^{-13}	1.93×10^{-5}		
Ar	mordenite	311	137.31	3.61	9.60×10^{6}	4.57×10^{-13}	2.67×10^{-5}		
Kr	mordenite	343	24.39	1.30	6.80×10^{5}	1.03×10^{-10}	4.25×10^{-4}	6.66	8.30
Kr	mordenite	323	85.96	1.36	2.89×10^{6}	5.79×10^{-11}	1.02×10^{-3}		
Kr	mordenite	303	91.03	1.50	2.58×10^{6}	2.67×10^{-11}	4.18×10^{-4}		
CH_3I	4A	345	3.55	1.97	1.50×10^{5}	2.04×10^{-12}	8.30×10^{-7}	11.37	4.13
CH_3I	4A	319	13.72	2.92	7.96×10^{5}	1.25×10^{-12}	2.69×10^{-6}		

Table II, Parameters used in model simulation*

System	De(cm^2/sec)	K(cc/cc)	k_f(cm/sec)	D_L(cm^2/sec)
A_r-4A	2.87×10^{-14}	4.42×10^{6}	4.29	0.969
K_r-4A	1.89×10^{-12}	9.55×10^{5}	3.57	0.956
A_r-MN	6.70×10^{-13}	8.02×10^{6}	4.29	0.969
K_r-MN	5.79×10^{-11}	4.91×10^{6}	3.57	0.956
CH_3I-4A	1.25×10^{-12}	1.01×10^{6}	2.81	0.942

* Common parameters:

$R=0.16$ cm, $\varepsilon_B=0.4$, $U=11.42$ cm/sec, $L=17.1$ m



1. Recorder
2. Electrobalance Assembly
3. Gas Reservoir
4. Manometer Assembly
5. Sorbate Gas Cylinder
6. Vacuum Gauge
7. Vacuum Pump
8&9. Resistor

Figure 1. Experimental Apparatus.

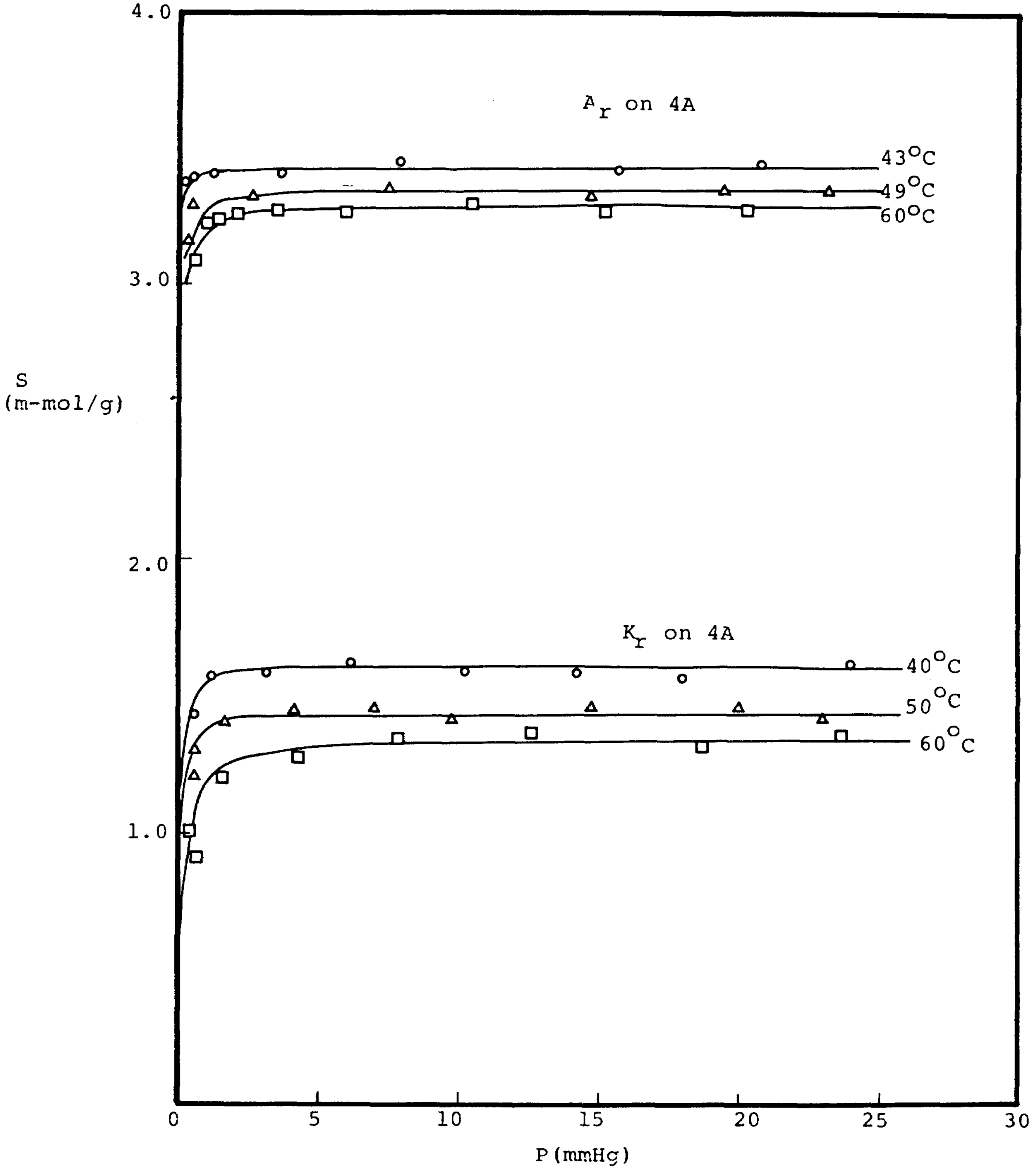


Fig. 2 Equilibrium Isotherms of A_r and K_r on 4A

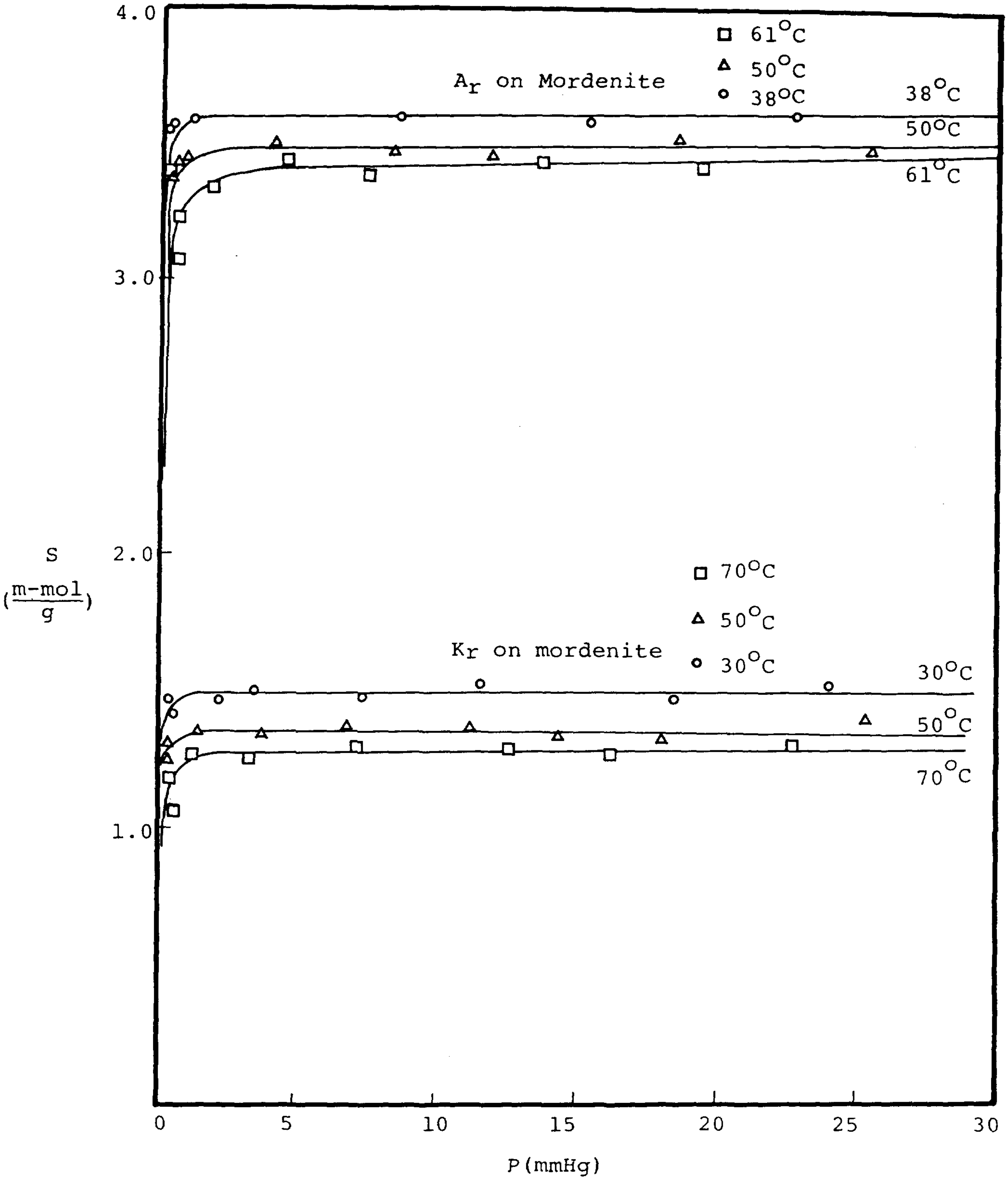


Fig. 3 Equilibrium Isotherms of Ar and Kr on Mordenite

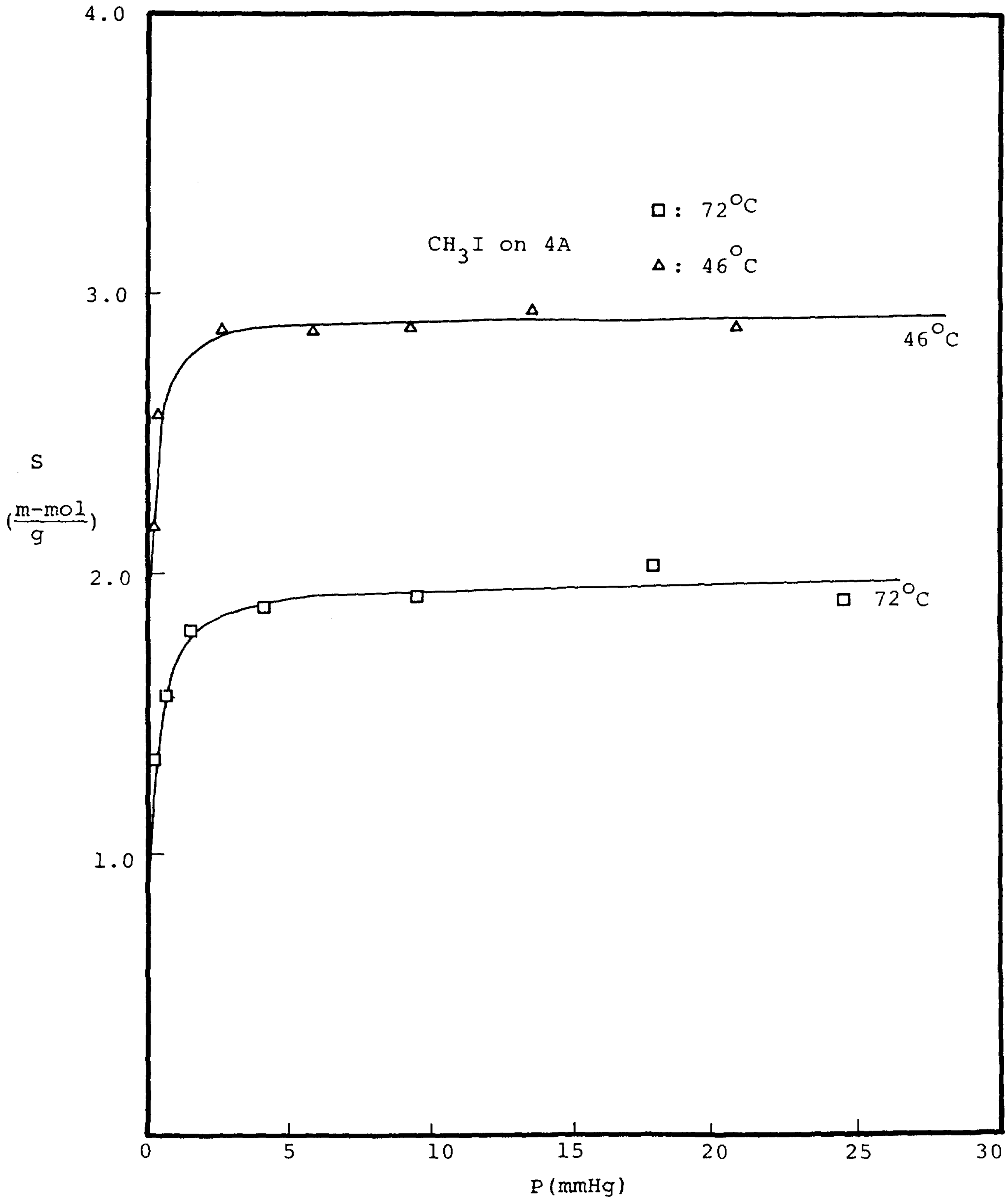


Fig. 4 Equilibrium Isotherms of CH_3I on 4A

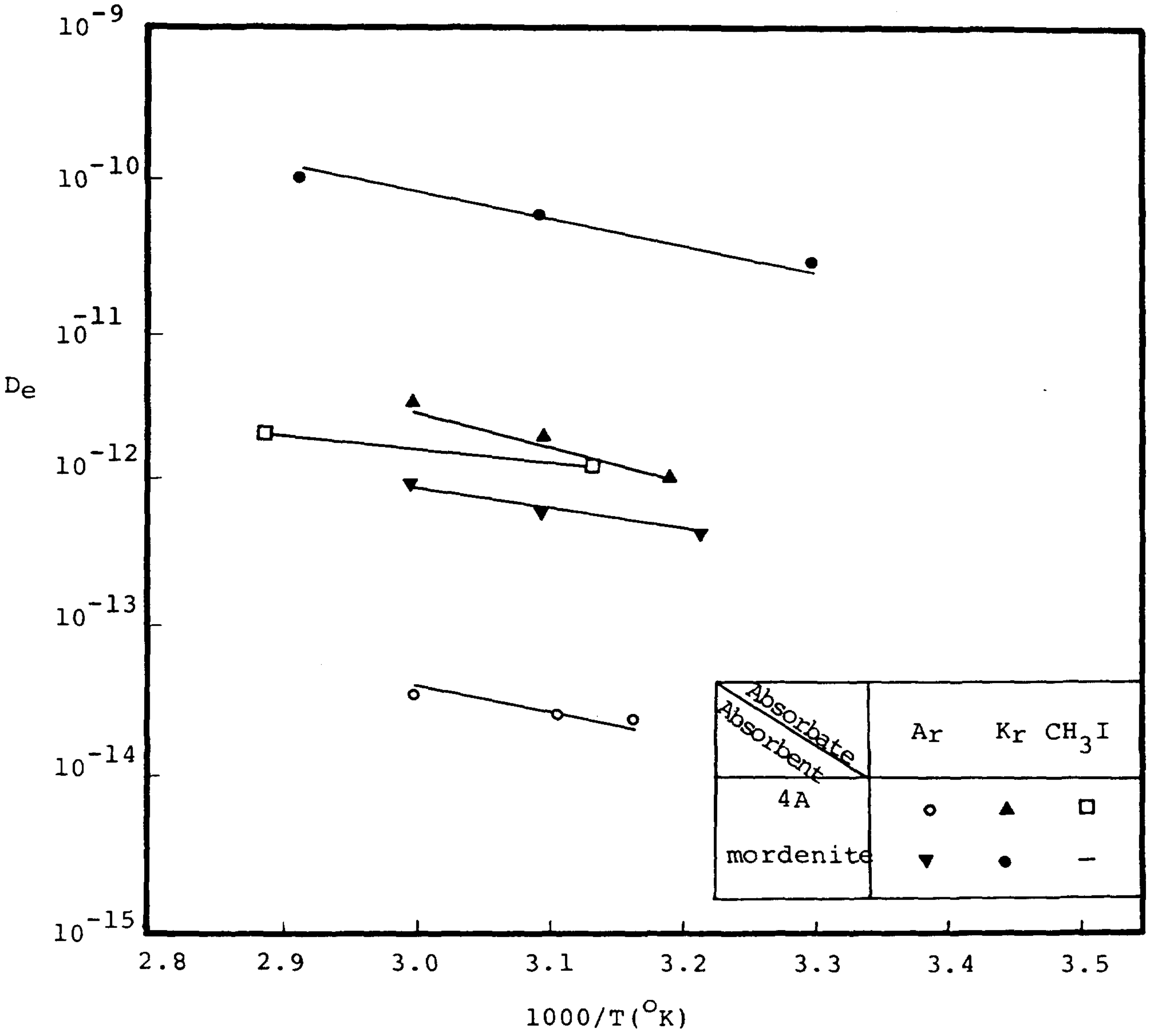


Fig. 5 Arrhenius plot of diffusivities

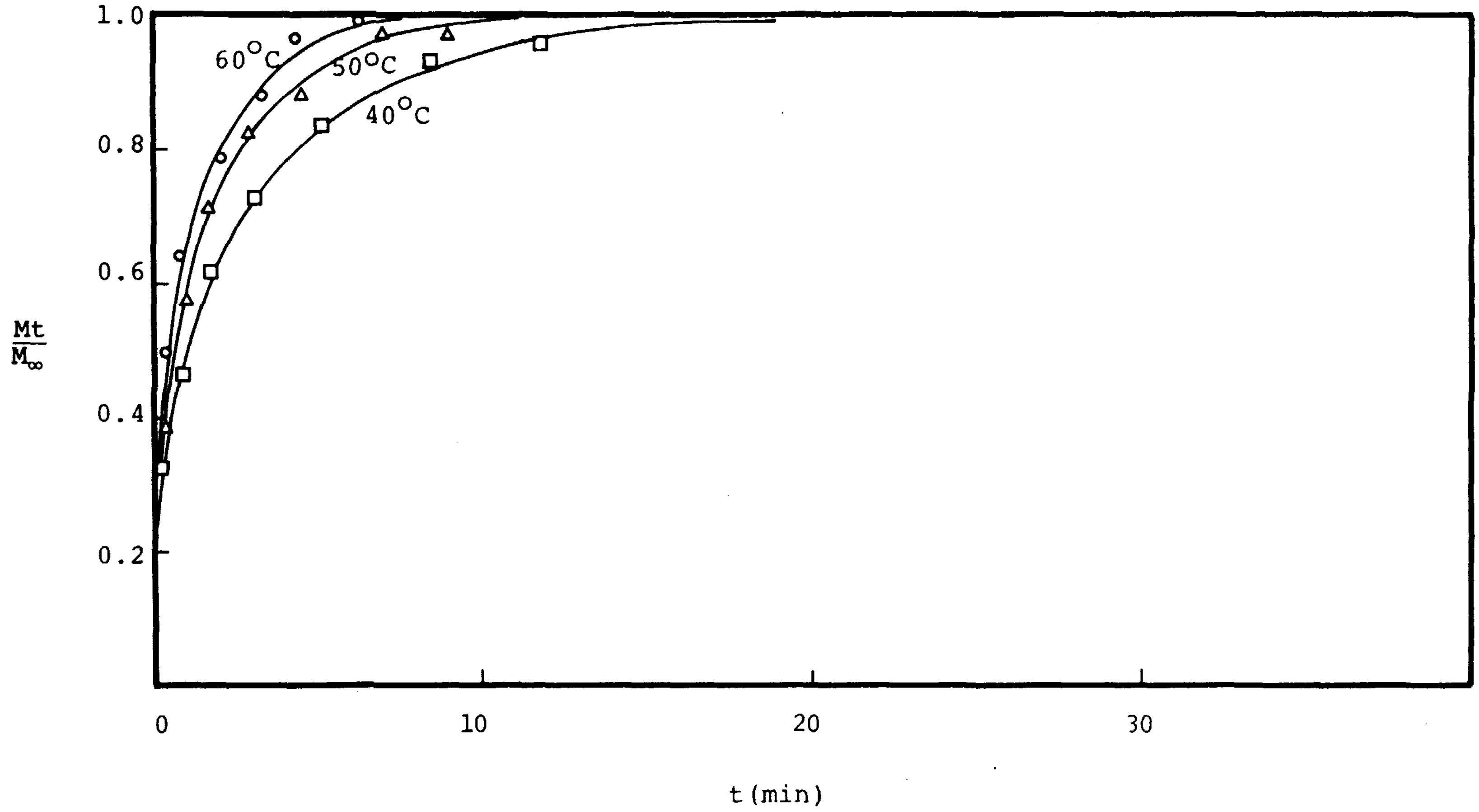


Fig. 6 Transient Diffusion of K_r in 4A Zeolite

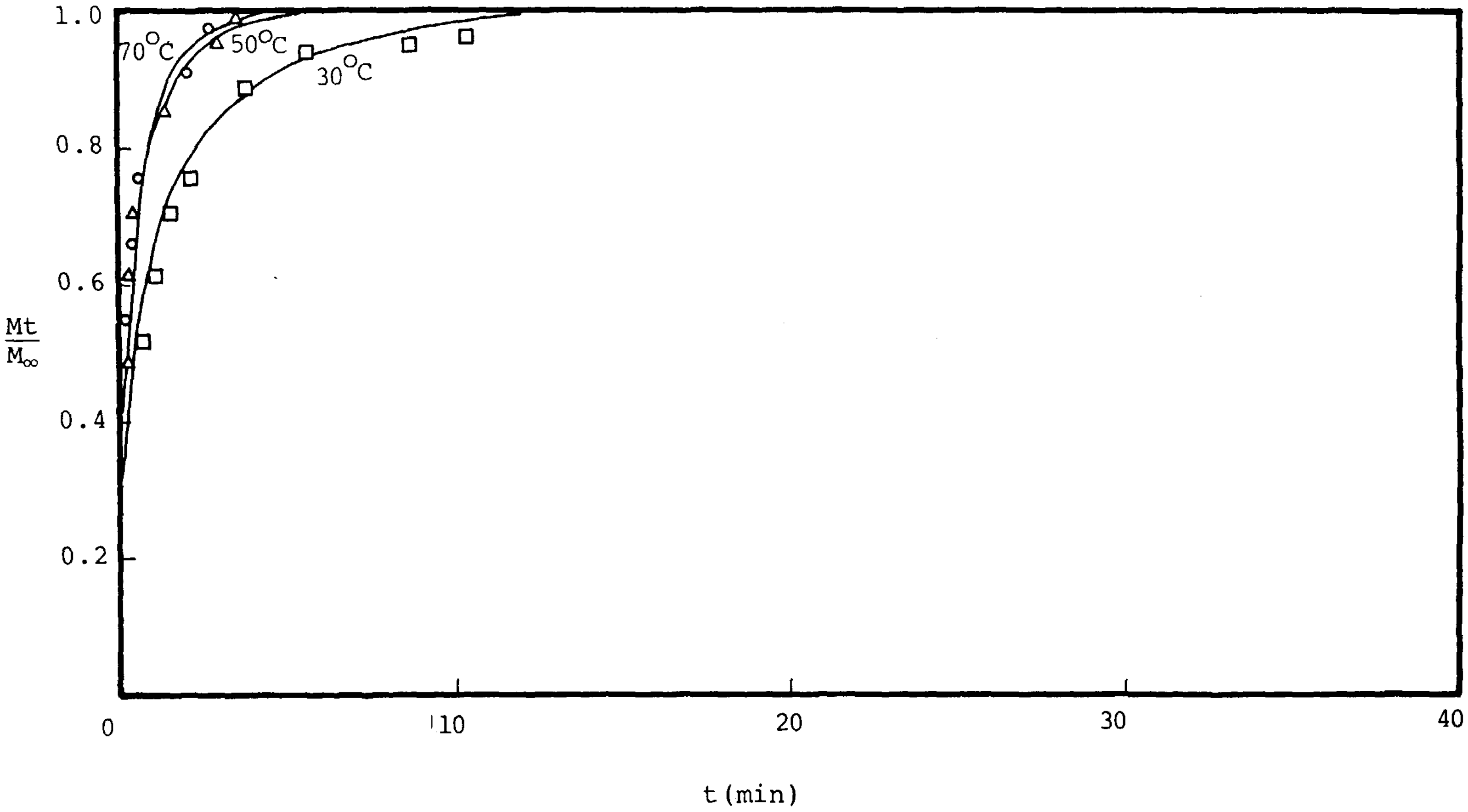


Fig. 7 Transient Diffusion of K_r in Mordenite

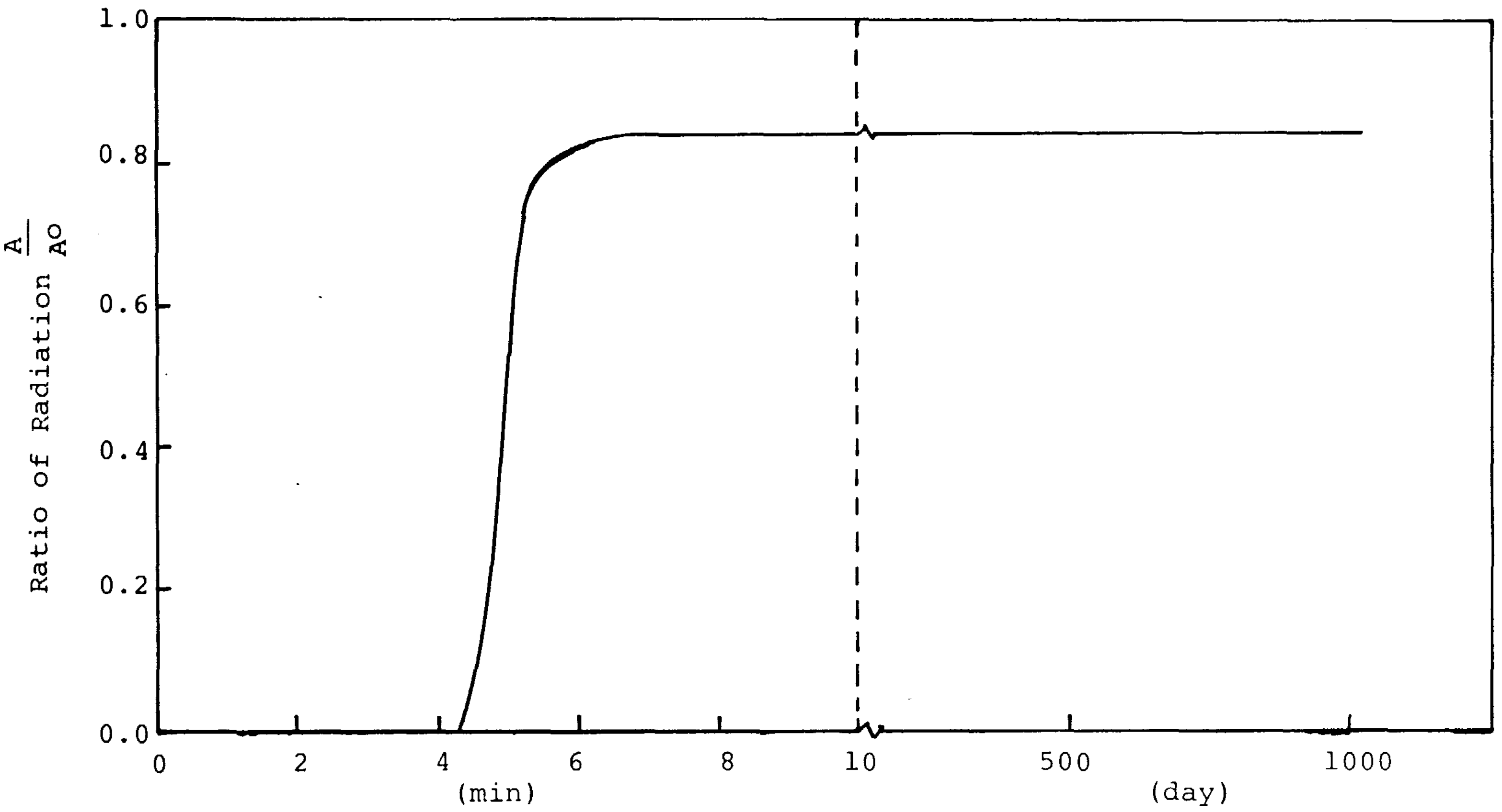


Fig. 8 Breakthrough Curve of A_r in 4A Column at 323°K

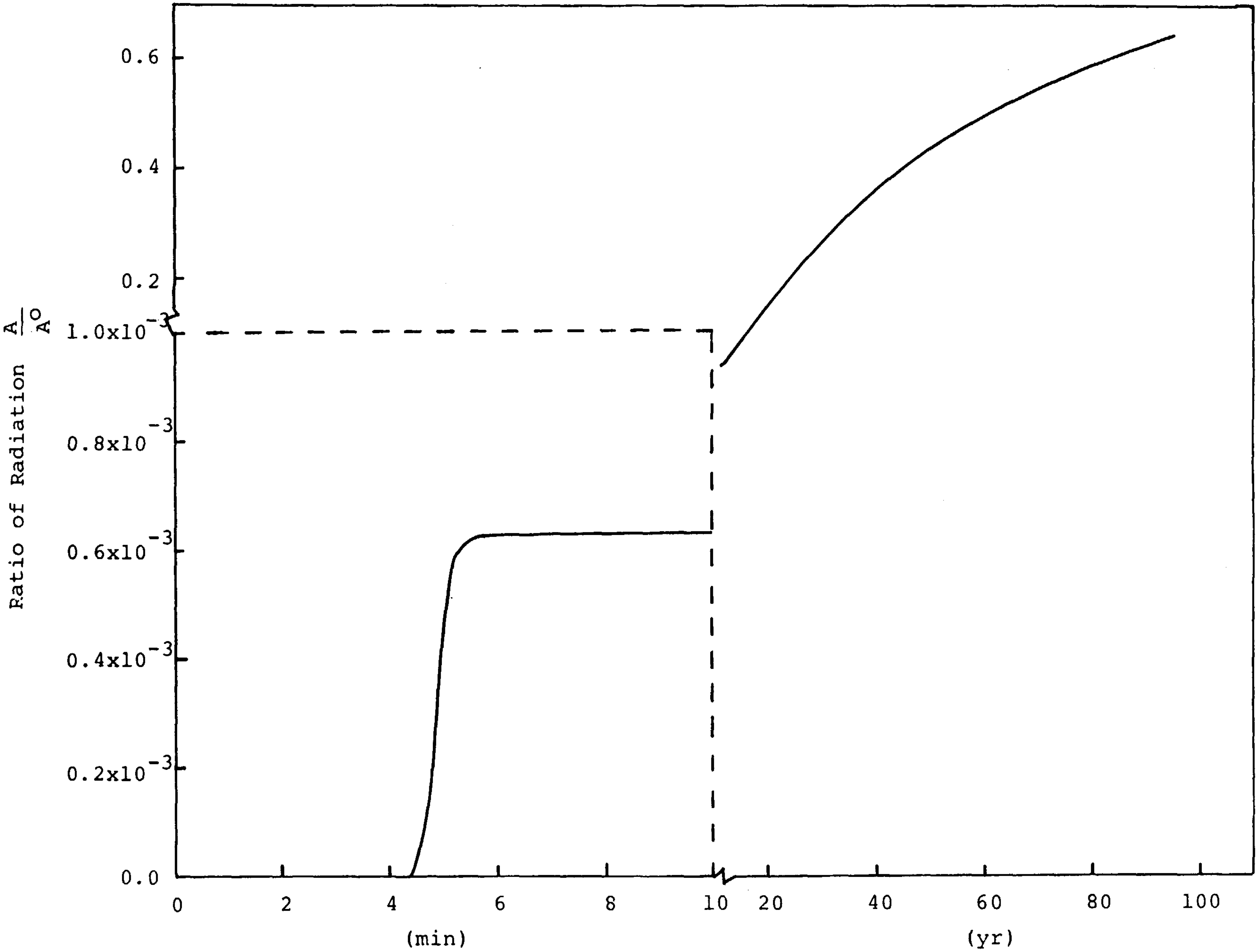


Fig. 9 Breakthrough Curve of A_r in Mordenite Column at 323^oK

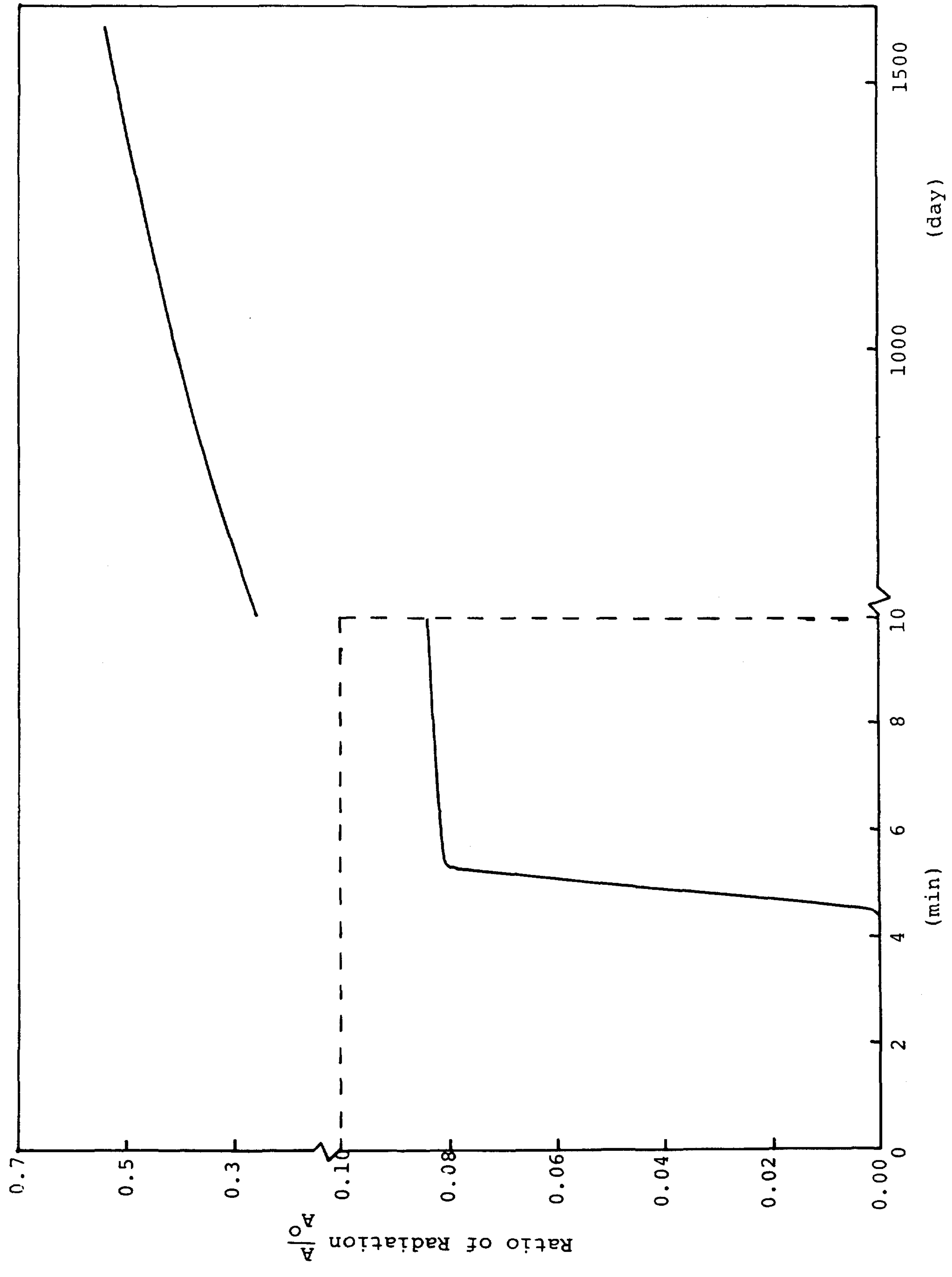


Fig. 10 Break through of Kr in 4A Column at 323°K

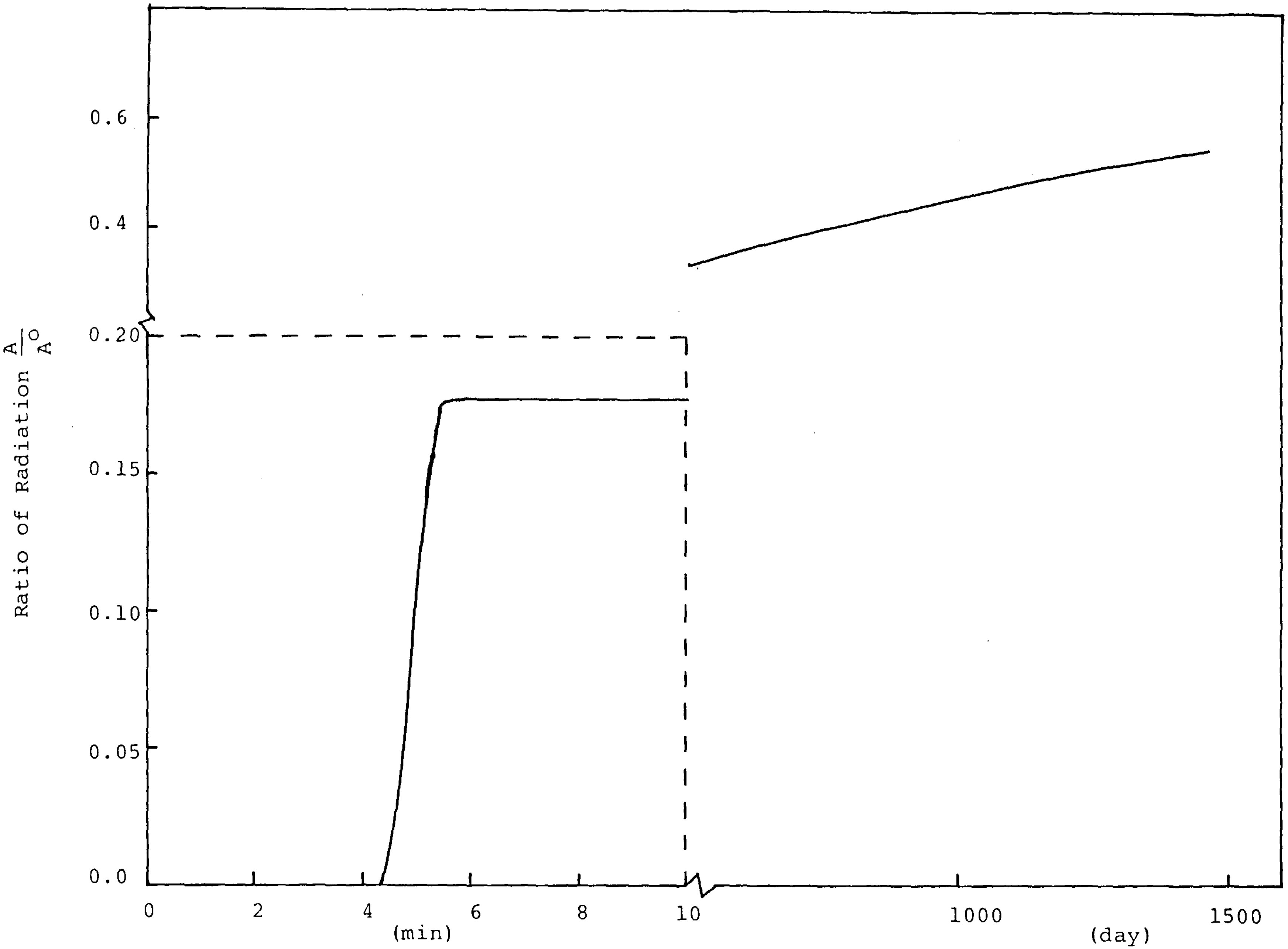


Fig. 11 Breakthrough Curve of CH_3I in 4A Column at 323°K

IN SITU MEASUREMENTS OF EFFICIENCY OF SILVER ZEOLITE FOR REMOVAL OF AIRBORNE RADIOIODINE SPECIES

J. W. Mandler, J. W. Tkachyk, J. T. Case,
J. H. McDaniel, and H. K. Peterson
Idaho National Engineering Laboratory
EG&G Idaho, Inc.
Idaho Falls, Idaho 83415

Abstract

In situ measurements of the decontamination factors (DFs) for the various species of radioiodines were performed for silver zeolite (AgX) filters used to clean up the Loss of Fluid Test (LOFT) containment air exhaust. Results indicated that the DF for elemental radioiodine was the highest, with the DFs for the particulate, hypoiodous acid (HOI), and organic (e.g. CH_3I) species being lower. Measured DFs ranged from 50 to greater than 100,000, depending on filter bank and radioiodine species.

I. Introduction

In situ measurements were made of the radioiodine in the air being exhausted from the containment building during the Fission Product 2 (FP-2) Test conducted during July 1985 at the Loss of Fluid Test (LOFT) facility located at the Idaho National Engineering Laboratory (INEL). Primary objectives of the measurements were to (1) determine the radioiodine species concentrations in the containment air, (2) observe changes with time of the radioiodine species mixture, and (3) measure the decontamination factors (DFs) realized by silver zeolite (AgX) cleanup filters for the various radioiodine species. Results of the measurements aimed at attaining the third objective are discussed in this paper.

The LOFT FP-2 Test was designed to provide data on the release and transport of radiologically significant fission products (e.g., I, Te, Cs) during accident conditions. The test simulated an accident in which a low-pressure injection system line ruptures and the emergency core cooling is delayed until severe fuel damage has occurred. Some of the radioiodines released from the fuel during the test were transported to the containment (reactor building) atmosphere. These radioiodines were subsequently passed through a silver zeolite cleanup filter and then vented to the environment. Measurements were made of the radioiodine concentrations upstream and downstream of the cleanup filter and DFs were determined for three silver zeolite filter banks.

II. Description of Experiment

A. Heating and Ventilating Systems

The LOFT containment (reactor building) has two heating and ventilating (H&V) systems that contain silver zeolite cleanup filters and which are designated as H&V 8 and H&V 9. Both H&V systems were used after the FP-2 test. H&V 9 consists of a 61-cm (24-inch) diameter line which runs from containment to a silver zeolite filter and then either back to the containment (recirculation mode) or to the environment (purge mode). A 5-cm (2-inch) diameter line runs from containment to a 25-cm (10-inch) diameter line in H&V 8. This 25-cm diameter line runs to a silver zeolite cleanup filter and then to the environment. H&V 8 contains two parallel silver zeolite filter beds (designated filters A and B for purposes of this paper), only one of which can be valved in at a time, while H&V 9 contains only one silver zeolite filter (designated filter C in this paper). H&V 8 is used to slowly vent the containment atmosphere while H&V 9 is used for high volume purge of the containment air.

Both H&V 8 and H&V 9 have sample points just upstream and just downstream of their cleanup filters. These sample points were designed for use in testing the cleanup filters. Prior to use, each sample point was shown to yield valid samples. A helium dilution technique was used in the sample point validation tests. This technique consists of injecting helium into each H&V system at a known rate upstream of the sample point and concurrently measuring the helium concentration at the sample point being validated.

B. Iodine Species Samplers

An iodine species and particulate sampler was used to collect samples for analysis.(1,2) The sampler consists of the following five media (in order encountered by flow):

1. Particulate Filter (Flanders F-700). This particulate filter is a HEPA type filter paper that has been waterproofed. It is used to retain particulate material which is pulled into the sampler.

2. Cadmium Iodide (CdI_2) on Chromosorb P (60/80 mesh supplied by Applied Science). This medium is used to adsorb elemental iodine (I_2).

3. 4-Iodophenol (IPH) on Alumina (30/60 mesh F-1 activated). This medium is used to adsorb hypoiodous acid (HOI) or, if HOI does not exist in the gaseous state, whatever iodine chemical species that deposits on the IPH but not on the CdI_2.

4. Silver Zeolite (AgX) (20/40 mesh). This medium is used to adsorb organic iodide species such as methyl iodide (CH_3I).

5. Charcoal (Barnebey-Cheney 151) or Silver Zeolite (20/40 mesh). This medium is used as a backup to adsorb any iodine species possibly escaping through the other media. For most samples, AgX was used because of its lower efficiency for adsorbing noble gases.

The media are loaded into aluminum media cups (9.5 mm high cup for the particulate filter; 35 mm high cup for the CdI_2, IPH, and AgX; and 25 mm high cup for the backup charcoal or AgX). These media cups are loaded into a cylindrical aluminum sampler body in the order given above. The sampler body is 19 cm long and 5 cm outside diameter and has an open and a closed end. The open end is threaded to accept a seal cap. Viton 'O' rings are used to insure a seal between the cups themselves and between the cups and the sampler body.

C. Experimental Setup

Figure 1 shows a schematic drawing of the experimental setup.

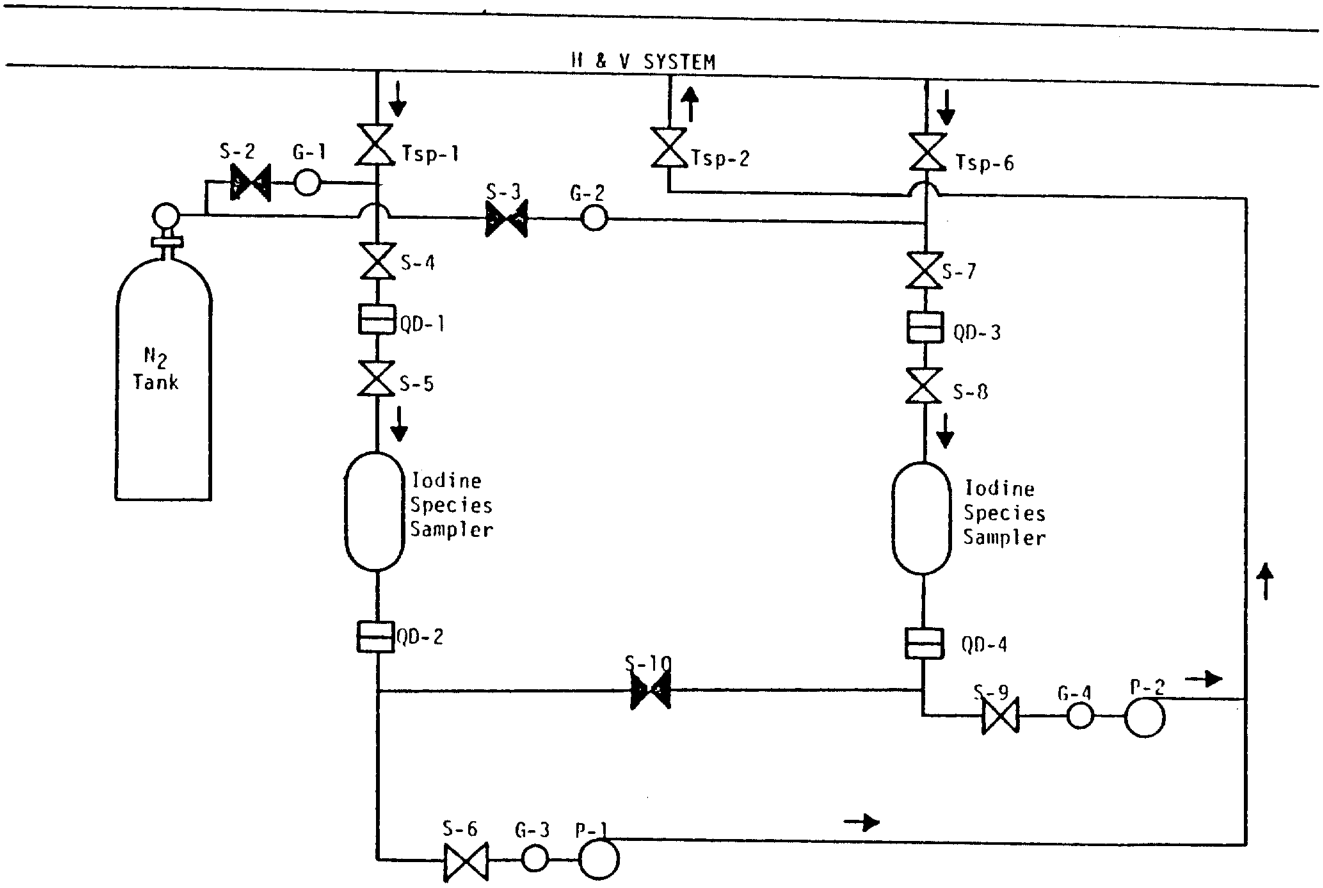


FIGURE 1
SCHEMATIC DRAWING OF SAMPLING SYSTEM

The sample upstream (downstream) of the cleanup filter was obtained from sample point TSP-1 (TSP-6). The flow went through valve S-4 (S-7), quick disconnect QD-1 (QD-3), valve S-5 (S-8), the iodine species sampler, isolating quick disconnect QD-2 (QD-4), valve S-6 (S-9), pump P-1 (P-2), and then returned to the H&V system through TSP-2. Flow rate was measured by G-3 (G-4). The N_2 tank was used to supply gas for leak testing of the system and to flush radioactive gases out of the system prior to removal of the samplers.

D. Sample Counting

After removal of an iodine species sampler from the sampling system, the sample cups were removed and placed in individual vials. These vials were then gamma counted using a hyperpure germanium detector-based spectrometer. The spectrometer was calibrated using National Bureau of Standards (NBS) traceable solutions which were loaded onto the various media.

III. Results of LOFT Measurements

One measurement of the DFs for the various radioiodine species was made on each of the three filters. Filter A was measured at 7.55 hours after the FP-2 test, Filter B at 31.3 hours after the test, and filter C at 75.8 hours after the test. Because the measurements were made at different times and the radioiodine species mixture in the LOFT containment air changed with time, each filter saw a different radioiodine species mixture at its inlet. Table 1 gives the inlet ^{131}I concentration and species mixture (i.e., fractions of the particulate, elemental, HOI, and organic species) during each measurement. The radioiodine species mixtures seen by the filters are the same as the mixtures in the LOFT containment, but the concentrations are lower than those in the containment due to addition of dilution air.

Table 1. ^{131}I Species Mixture at Filter Inlet

Filter	^{131}I Conc. (μCi/cc)	Fractional ^{131}I Species Mixture Part.	I_2	HOI	Organic
A	1.55 E-5	0.02	0.57	0.21	0.20
B	2.84 E-5	0.01	0.45	0.17	0.37
C	6.33 E-5	0.006	0.23	0.10	0.66

Tables 2, 3, and 4 give the inlet and outlet species concentrations and the resulting DFs for the radioiodines (^{131}I, ^{132}I, ^{133}I, and ^{135}I) observed in the inlets to the three AgX filters. The results for ^{132}I are not given in Table 3 because only elemental ^{132}I (the major species component) was seen in the inlet air stream. The concentrations of the other species were below detection limits. Results for ^{132}I and ^{135}I are not given in Table 4 because they were not seen in the samples due to a long delay between sampling and analysis of the samples. Results from other samples obtained of the inlet to the AgX filter several hours earlier, however, indicate that the total ^{132}I and ^{135}I concentrations were both about 2E-8 μCi/cc and that the ^{132}I was predominately elemental and the ^{135}I was predominately organic.

The results of the measurements indicate that the DF for elemental ^{131}I was the highest, with the DFs for the particulate, HOI, and organic species being lower. The ^{131}I DFs were relatively low for cleanup filters A and B (360 to 1200 for particulate, about 3000 for elemental, 46 for HOI, about 60 for organic, and 100 to 120 for the overall species mixture). The ^{131}I DFs were much higher for cleanup filter C (11,000 for particulate, 210,000 for elemental, 3800 for HOI, 1100 for organic, and 1500 for the overall species mixture). For all three cleanup filters, since each contains a HEPA filter upstream of the AgX, the particulate DF is due to the filtration of the combined components.

The results of DF measurements based on the other radioiodines (^{132}I, ^{133}I, and ^{135}I) exhibit good agreement with the results obtained using ^{131}I.

Table 2. Decontamination Factors for LOFT AgX Filter A

Nuclide	Species	Concentration (μCi/cc) Inlet	Outlet	Decontamination Factor
^{131}I	Part.	3.4E-07	2.9E-10	1.2E+03
	I_2	8.8E-06	3.2E-09	2.7E+03
	HOI	3.2E-06	7.1E-08	4.6E+01
	Organic	3.1E-06	5.1E-08	6.1E+01
	Total	1.5E-05	1.3E-07	1.2E+02
^{132}I	Part.	7.1E-08	<2.0E-10	>3.5E+02
	I_2	2.5E-06	9.1E-10	2.8E+03
	HOI	7.9E-07	1.4E-08	5.6E+01
	Organic	6.4E-07	1.1E-08	5.8E+01
	Total	4.0E-06	2.6E-08	1.5E+02
^{133}I	Part.	1.1E-06	8.9E-10	1.3E+03
	I_2	3.3E-05	1.0E-08	3.2E+03
	HOI	1.1E-05	2.4E-07	4.6E+01
	Organic	1.1E-05	1.7E-07	6.2E+01
	Total	5.5E-05	4.2E-07	1.3E+02
^{135}I	Part.	6.2E-07	<9.1E-10	>6.8E+02
	I_2	1.8E-05	5.8E-09	3.1E+03
	HOI	6.0E-06	1.3E-07	4.6E+01
	Organic	5.6E-06	9.5E-08	5.9E+01
	Total	3.0E-05	2.3E-07	1.3E+02

Table 3. Decontamination Factors for LOFT AgX Filter B

Nuclide	Species	Concentration (μCi/cc) Inlet	Outlet	Decontamination Factor
^{131}I	Part.	3.0E-07	8.3E-10	3.6E+02
	I_2	1.3E-05	4.3E-09	3.0E+03
	HOI	4.7E-06	1.0E-07	4.6E+01
	Organic	1.1E-05	1.7E-07	6.4E+01
	Total	2.8E-05	2.7E-07	1.0E+02
^{133}I	Part.	4.8E-07	1.4E-09	3.4E+02
	I_2	2.2E-05	6.9E-09	3.1E+03
	HOI	7.8E-06	1.7E-07	4.6E+01
	Organic	1.8E-05	2.7E-07	6.6E+01
	Total	4.8E-05	4.5E-07	1.1E+02
^{135}I	Part.	5.1E-08	<8.4E-10	>6.1E+01
	I_2	2.2E-06	<1.6E-09	>1.4E+03
	HOI	7.8E-07	1.9E-08	4.1E+01
	Organic	1.8E-06	2.9E-08	6.1E+01
	Total	4.8E-06	5.0E-08	9.5E+01

Table 4. Decontamination Factors for LOFT AgX Filter C

Nuclide	Species	Concentration (μCi/cc) Inlet	Outlet	Decontamination Factor
^{131}I	Part.	4.0E-07	3.5E-11	1.1E+04
	I_2	1.5E-05	<7.0E-11	>2.1E+05
	HOI	6.2E-06	1.6E-09	3.8E+03
	Organic	4.2E-05	3.9E-08	1.1E+03
	Total	6.3E-05	4.1E-08	1.5E+03
^{133}I	Part.	1.8E-07	<1.6E-10	>1.1E+03
	I_2	6.7E-06	<4.8E-10	>1.4E+04
	HOI	2.7E-06	7.6E-10	3.6E+03
	Organic	1.8E-05	1.7E-08	1.1E+03
	Total	2.8E-05	1.8E-08	1.5E+03

In the course of the DF analysis an anomaly was observed. The iodine species sampler contains a backup cartridge (charcoal or silver zeolite) to collect any iodine that may get through the other filter media. The amount of radioiodine on this backup cartridge is always examined to insure that the data from the various media are not compromised due to radioiodine breakthrough of these media. During examination of the data from the backup cartridge, it was noticed that the amount of radioiodine present was approximately the same for samples obtained both upstream and downstream of the cleanup filter being studied (even though the radioiodine concentration in the inlet stream was 2 to 3 orders of magnitude higher than in the outlet stream).

In addition, the ratio of the radioiodine on the backup cartridge medium to that on the medium intended for collection of organic iodine was several hundreths of a percent for samples obtained upstream of the cleanup filter but was several percent for samples obtained downstream of the cleanup filter. The above indicate that that there may be an iodine species (possibly a higher order organic than methyl iodide) that is partially adsorbed on charcoal and AgX but is not effectively removed from the gas stream. Discussions with other investigators have indicated that they have seen similar phenomena.

IV. Conclusions

In situ measurements of the DF for the LOFT H&V systems' AgX cleanup filters indicated that AgX is effective for removing radiodines from an air stream. The actual DF realized, however, depends on the condition of the filter and the radioiodine species mixture.

Measured DFs were in the range 360 to 11,000 for particulate ^{131}I, 2700 to >210,000 for elemental, 46 to 3800 for HOI, 61 to 1100 for organic, and 100 to 1500 for the overall ^{131}I species mixture. Cleanup filters A and B exhibited similar but low DFs, while cleanup filter C had much higher DFs. For all three filters, most of the efficiency for particles is probably due to the HEPA filter which is located just upstream of each AgX unit.

A comparison of the DFs measured for the LOFT AgX filters with DFs measured for charcoal filters at operating nuclear power plants indicates that the DFs for filters A and B are in the range of the values obtained for the charcoal filters.(3) The DFs measured for filter C, however, are much higher than were seen for charcoal filters at power plants. Both the LOFT AgX filters and power plant charcoal filters exhibited similar DF behavior for the various radioiodine species. The DFs for the LOFT AgX filters were highest for elemental and lowest for organic radioiodine, and the power plant charcoal filter DFs were highest for HOI and elemental and lowest for organic radioiodine.

All three of the LOFT cleanup filters exhibited lower DFs than were expected. Filter C had been tested about one month before the FP-2 experiment and had exhibited a DF of 39,000 for methyl iodide, compared to 1100 measured during the FP-2 experiment. Its DF, therefore, decreased by a factor of about 35 in only one month. Filters A and B exhibited DFs in the range 1200 to 1500 for methyl iodide about 9 months before the FP-2 experiment, but only about 60 during the FP-2 experiment. This represents a decrease of about a factor of 20 to 25 in a 9 month period. These large decreases in filter efficiency have not yet been explained.

The measurements on the LOFT AgX cleanup filters also indicated a result that has been seen at operating power plants.(3) Examination of the species mixture at the inlet of the LOFT filters indicates that (1) the radioiodine initially released from the reactor system during the FP-2 experiment was primarily elemental and (2) with time, the elemental fraction decreased while the organic fraction increased. At operating power plants, it was observed that radioiodine freshly released from the reactor coolant system had a high fraction of the more reactive species (elemental and/or HOI). In addition, the conversion with time of the more reactive species to the lesser reactive organic species was also observed.

References

1. Keller, J. H. et al., "A Selective Adsorbent Sampling System for Differentiating Airborne Iodine Species," Proceedings of the 11th USAEC Air Cleaning Conference (1970).

2. Emel, W. A. et al., "An Airborne Species Sampler and Its Application for Measuring Removal Efficiencies of Large Charcoal Adsorbers for Ventilation Exhaust Air," Proceedings of the 14th ERDA Air Cleaning Conference (1976).

3. Mandler, J. W. et al., "Measuring Gaseous Radioiodine at Power Reactors," Proceeding of the 19th DOE/NRC Nuclear Air Cleaning Conference (1986).

DISCUSSION

HULL: As I understand your presentation, it appears that Ag-Zeolite may not be the medium of choice for sampling the HOI and organic species of radioiodines under accident conditions, even though it is widely employed and even required by the NRC. Could you comment on this?

MANDLER: The DFs for HOI and organic radioiodine that we measured for the LOFT AgX filters A and B compare well with results we obtained on charcoal filters at operating power plants. You would expect these DFs to be lower than those for AgX and charcoal used for sampling - mainly because media used for sampling are fresh. I believe that AgX is indeed a good medium for sampling HOI and organic radioiodine. Recall that filter C (a filter that had been in use for at least a month prior to the LOFT test) exhibited DFs of 3,800 and 1,100 for HOI and organic radioiodine, respectively. These DFs correspond to retention efficiencies of >99.9%.

SUMMARY AND RECOMMENDATIONS OF THE NRC/INEL ACTIVATED CARBON TESTING PROGRAM*

C.D. Scarpellino and C.W. Sill
Idaho National Engineering Laboratory
EG&G Idaho, Inc.
P.O. Box 1625
Idaho Falls, Idaho 83415

Abstract

The Committee on Nuclear Air and Gas Treatment (CONAGT) of the American Society of Mechanical Engineers (ASME) sponsored an interlaboratory testing program of nuclear-grade activated carbon. The results of this round-robin revealed gross differences in penetration of radio-labeled methyl iodide as measured by the various laboratories when using Method A of the ASTM D-3803-79 standard. These differences prompted the Nuclear Regulatory Commission (NRC) to establish an activated carbon testing program at the Idaho National Engineering Laboratory (INEL) to determine the causes of these discrepancies and to provide recommendations that could lead to an accurate and reliable testing procedure for assessing the capability of activated carbon to remove radioiodine from gas streams within commercial nuclear power plants.

Formal and informal interlaboratory comparisons have been carried out under the NRC/INEL Activated Carbon Testing Program to identify problems with the test method and its application and to assess the effectiveness of changes to procedures and equipment voluntarily implemented by commercial laboratories to mitigate the disparity of test results. The results of the first formal NRC/INEL Interlaboratory Comparison (IC) essentially confirmed the variability obtained in the CONAGT round-robin results despite the use of a detailed test protocol. The IC protocol required the completion of a standard report form for each test which provided information regarding critical test parameters. This data indicated that many of the participating laboratories probably had been operating outside the ASTM specifications for relative humidity (RH) and flow. Several workshops were held to discuss and attempt to rectify problems associated with activated carbon testing. Informal intercomparisons were used to guide individual laboratories in the iterative process of test system trouble-shooting and modification. In addition, this process provided information which was used to modify the testing protocol employed for the second NRC/INEL Interlaboratory Comparison (IC-2) to make it more rugged and reliable. These changes to the protocol together with the results of INEL sensitivity testing are the basis for the recommendations presented.

Introduction

The NRC/INEL Activated Testing Program has consisted of: 1) a review of the ASTM D-3803-79 Standard to determine if it adequately specifies test parameters and procedures so that consistent and accurate results should be obtained; 2) a review of the CONAGT round-robin testing in an attempt to explain the wide variation of results obtained by the laboratories; 3) visits to the commercial laboratories to review the methods and equipment used to determine if

---

* This work was supported by the U.S. Nuclear Regulatory Commission, FIN No. A6807, under DOE Contract No. DE-AC07-76ID01570.

they adequately conformed to the ASTM D-3803 Standard; 4) fabrication of a test facility at the INEL and the performance of tests to determine the sensitivity of penetration measurements to variations of test parameters and procedures; 5) an initial workshop in Idaho on August 27-29, 1984 with representatives from manufacturers of activated carbon, testing laboratories, the NRC and EG&G Idaho to discuss activated carbon testing and the results of INEL sensitivity tests completed by that date; 6) an Interlaboratory Comparison (IC) with test laboratories performing analyses on identical samples using the procedures of ASTM D-3803 as modified by a written protocol to determine the precision with which penetration of radio-methyl iodide could be measured; 7) a second workshop held in Annapolis, Maryland on June 5 and 6, 1985 to discuss the results of the IC and pertinent INEL sensitivity test results; 8) a workshop held at the INEL on September 10-12, 1985 supervised by technical experts from the National Bureau of Standards (NBS) to calibrate and intercompare devices used to measure flow and RH at the INEL and at several commercial laboratories; 9) a program of informal one-on-one comparisons to identify remaining differences in the results obtained; and 10) a second Interlaboratory Comparison (IC-2) with laboratories required to follow the INEL Standard Test Method for Nuclear-Grade Activated Carbon, which is a substantial revision of ASTM D-3803-79, Method A.

Several important tasks were completed prior to the first NRC/INEL IC which was completed in May 1985. Details of these tasks are not included in this paper and have been reported elsewhere.(1,2) One previous task which has been critical to understanding the problems associated with carbon testing was the design, fabrication, operation and maintenance of the INEL test system. This has been an evolutionary process beginning with a system modeled after the schematic presented in the ASTM Standard and ending with the current system which is a composite of designs and instrumentation used throughout the world. This type of evolutionary system design is extremely common within the industry and is ongoing. Recent advances in off-the-shelf microprocessor controlled monitoring and control devices have contributed significantly to the ease and accuracy of parameter recording and control. The evolution of the INEL test system has been more rapid than most in the private sector because of the observations, experiences and criticisms generously contributed by the commercial carbon testing laboratories. This type of information has not previously been freely exchanged within the industry and has been extremely useful in the identification of problem areas in commercial carbon testing.

NRC/INEL Interlaboratory Comparison

The IC was conducted to determine the precision with which the penetration of radio-methyl iodide through activated carbon was being measured in different laboratories. Eight laboratories from the U.S., two laboratories from Canada, and one each from the United Kingdom, the Federal Republic of Germany, and Korea agreed to participate. The study involved measurements of penetration for a single new carbon at 30°C and 95% RH and for a single used carbon at 30°C and 70, 90, and 95% RH. The procedure used was Method A of the ASTM D-3803 Standard as modified by specific written changes. The new carbon was tested with equilibration, i.e. exposed to the specified temperature, air flow, RH, etc. for a period of 16 hours before being challenged by the methyl iodide. The used carbon was tested without equilibration. Triplicate analyses were performed at each experimental condition to distinguish between differences in results caused by interlaboratory imprecision of measurement and those caused by intralaboratory imprecision of control of the experimental conditions.

To ensure that all carbon samples tested by all laboratories were identical, a large batch of new carbon was hand screened to give a +1mm to -2mm fraction which was then homogenized by thorough blending as a single batch in a twin-shell blender. The batch was then dedusted with clean cylinder compressed air and manual mixing. A 200-ml aliquot was removed batchwise without skimming using a plastic beaker to avoid fractionation, transferred to a polyester bag, heat-sealed immediately, and labeled for distribution to the individual laboratories. The entire process was then repeated with a batch of used carbon. Samples were distributed to participating laboratories by November 23, 1984.

Of the thirteen laboratories that initially agreed to participate in the IC, ten submitted all data requested, two withdrew prior to testing, and one laboratory withdrew their results during the Second Carbon Testing Workshop in Annapolis, Maryland because of their own dissatisfaction with their results. Similarly, another laboratory withdrew their initial results because of their own dissatisfaction with the technical quality of the work. Subsequently, they submitted a second set of test results which has been included in this paper. Two fundamental conclusions were derived from the IC data obtained.

First, the precision within each laboratory was generally good to outstanding. Nine of the ten laboratories reporting triplicate runs on each of the three experimental conditions for used carbon obtained relative standard deviations of the individual measurements about their own mean of 1 to 6%. The precision for the new carbon tests was also generally good but lower than that for the used carbon because of its low values for penetration and its extreme susceptability to minor variations in relative humidity. It was clear that within each laboratory the critical experimental parameters were being controlled adequately.

Second, the precision obtained among laboratories was extremely poor. As shown by the results presented in Table 1, the new carbon results were spread uniformly from 0.23 to 8.4%. No two U.S. laboratories agreed with each other within two standard deviations of their respective means. The results from laboratory 12 are not strictly comparable with the others because of the different equilibration period used. The results for the used carbon generally fell into two distinctly different levels. One group of three laboratories obtained values of 17 to 19%, the other group of six laboratories obtained values in the range of 24 to 26%. Laboratory 8 equilibrated the samples at the 95% RH condition and these data are therefore excluded from comparisons. Although these results, 60% penetration, are not comparable to other IC data, two other laboratories informally reported results for equilibrated used carbon tests which were in close agreement, 59 and 60.2% penetration. Laboratory 10 made measurements at 22 to 23°C and are not directly comparable to the other results, although they clearly show the effect of temperature on penetration. The generally poor agreement among laboratories must be attributed to reproducible systematic differences in both test system operation and performance.

The results of sensitivity testing performed at the INEL on additional samples of the IC new carbon indicated a very strong correlation, $r^2 = 0.996$, between the final moisture content (FMC), % water, of the carbon and the log percent penetration. The log-linear regression of average penetration versus the average FMC reported for new carbon by each of the participating laboratories yielded a poor correlation, $r^2 = 0.46$. This lack of good agreement with the sensitivity data must have been due to the influence of variables other than RH

Table 1. Results of the NRC/INEL Interlaboratory Comparison (IC) of Penetration of Radio-Methyl Iodide Through Activated Carbon.

NEW CARBON AT 95% RELATIVE HUMIDITY

LABORATORY NO.	LABORATORY INITIALS	PENETRATION ± EXP. STD. DEV., %	MOISTURE CONTENT, wt. % H_2O INITIAL	MOISTURE CONTENT, wt. % H_2O FINAL	TEST BED DENSITY, g/cm^3
5	SAIC	DATA WITHDRAWN			
3	NUCON	0.233 ± 0.018 (8%)	4.0 ± 0.4 (10%)	26.8 ± 0.4 (2%)	0.492 ± 0.003 (1%)
7	MSA	0.71 ± 0.03 (4%)	3.77 ± 0.19 (5%)	27.0 ± 0.5 (2%)	0.496 ± 0.004 (1%)
9	BC	0.91 ± 0.02 (2%)	2.87 ± 0.14 (5%)	24.4 ± 0.5 (2%)	0.498 ± 0.004 (1%)
11	OH	1.06 ± 0.09 (9%)	3.04 ± 0.07 (2%)	27.0 ± 0.4 (2%)	0.4751 ± 0.0007 (1%)
8	KFK	1.8 ± 0.4 (22%)	3.80 ± 0.05 (1%)	28.5 ± 0.3 (1%)	0.469 ± 0.004 (1%)
1	EG&G	2.31 ± 0.3 (13%)	3.125 ± 0.018 (1%)	28.1 ± 0.4 (2%)	0.501 ± 0.005 (1%)
10	WNL	[a]2.88 ± 0.14 (5%)	4.9 ± 1.2 (20%)	NA	NA
13	EET	3.65 ± 0.19 (5%)	4.03 ± 0.06 (2%)	32.5 ± 0.4 (1%)	0.504 ± 0.004 (1%)
6	NCS	7.18 ± 0.10 (2%)	4.27 ± 0.06 (2%)	29.7 ± 0.5 (2%)	0.517 ± 0.008 (2%)
12	KAERI	[b]8.4 ± 2.9 (35%)	2.86 ± 0.02 (1%)	24.9 ± 1.5 (6%)	0.499 ± 0.009 (2%)

USED CARBON

LABORATORY NO.	RELATIVE HUMIDITY, %	PENETRATION ± EXP. STD. DEV., %	MOISTURE CONTENT, wt. % H_2O INITIAL	MOISTURE CONTENT, wt. % H_2O FINAL	TEST BED DENSITY, g/cm^3
3	95	18.90 ± 0.15 (1%)	20.1 ± 0.4 (2%)	29.8 ± 1.2 (4%)	0.567 ± 0.007 (1%)
	90	17.9 ± 0.7 (4%)	20.03 ± 0.15 (1%)	28.0 ± 0.3 (2%)	0.565 ± 0.004 (1%)
	70	19.0 ± 0.3 (2%)	20.3 ± 0.4 (2%)	23.6 ± 0.5 (3%)	0.572 ± 0.002 (1%)
6	95	18.6 ± 1.4 (8%)	20.5 ± 0.6 (3%)	31.4 ± 1.4 (5%)	0.599 ± 0.004 (1%)
	90	17.4 ± 0.7 (4%)	20.8 ± 0.6 (3%)	30.1 ± 0.5 (2%)	0.588 ± 0.008 (2%)
	70	19.1 ± 0.6 (3%)	21.0 ± 0.6 (3%)	26.1 ± 0.7 (3%)	0.600 ± 0.006 (1%)
13	95	18.21 ± 0.16 (1%)	20.3 ± 0.2 (1%)	33.14 ± 0.16 (5%)	0.589 ± 0.008 (2%)
	90	17.9 ± 0.5 (3%)	20.2 ± 0.3 (2%)	32.53 ± 0.06 (2%)	0.597 ± 0.006 (1%)
	70	16.6 ± 0.5 (3%)	20.5 ± 0.5 (2%)	28.6 ± 0.8 (3%)	0.590 ± 0.005 (1%)
10	[a]95	33.8 ± 1.1 (3%)	20.8 ± 0.4 (2%)	NA	NA
	[a]90	32.3 ± 0.6 (2%)	20.5 ± 0.8 (4%)	NA	NA
	[a]70	33.3 ± 0.7 (2%)	20.68 ± 0.18 (1%)	NA	NA
5		DATA WITHDRAWN			
9	95	25.1 ± 0.3 (1%)	19.4 ± 0.2 (1%)	29.4 ± 0.2 (1%)	0.574 ± 0.004 (1%)
	90	20.5 ± 0.7 (4%)	19.1 ± 0.5 (3%)	28.6 ± 0.5 (2%)	0.57 ± 0.02 (3%)
	70	22.8 ± 1.1 (5%)	19.18 ± 0.09 (1%)	24.7 ± 0.6 (2%)	0.571 ± 0.005 (1%)
1	95	25.6 ± 0.9 (4%)	20.490 ± 0.004 (1%)	32.6 ± 0.4 (1%)	0.568 ± 0.008 (2%)
	90	26.5 ± 1.7 (6%)		32.0 ± 0.5 (2%)	0.563 ± 0.014 (3%)
	70	24.5 ± 1.0 (4%)		26.5 ± 0.4 (2%)	0.573 ± 0.011 (2%)
7	95	26.4 ± 0.3 (1%)	19.82 ± 0.13 (1%)	31.5 ± 0.6 (2%)	0.561 ± 0.011 (2%)
	90	24.5 ± 0.2 (1%)	19.4 ± 0.2 (1%)	28.9 ± 0.4 (2%)	0.547 ± 0.006 (1%)
	70	24.1 ± 0.6 (3%)	19.8 ± 0.3 (2%)	21.26 ± 0.08 (1%)	0.570 ± 0.017 (3%)
8	[c]95	60.0 ± 1.7 (3%)	24.37 ± 0.18 (1%)	39.76 ± 0.08 (1%)	0.548 ± 0.002 (1%)
	90	25.4 ± 0.8 (3%)	26.2 ± 0.3 (1%)	39.8 ± 0.4 (1%)	0.557 ± 0.008 (2%)
	70	26.4 ± 0.8 (3%)	24.9 ± 0.2 (1%)	30.8 ± 1.2 (4%)	0.546 ± 0.002 (1%)
11	95	24.3 ± 0.4 (2%)	19.9 ± 0.4 (2%)	31.7 ± 1.0 (4%)	0.544 ± 0.005 (1%)
	90	25.2 ± 0.3 (1%)	19.8 ± 0.3 (2%)	30.13 ± 0.14 (1%)	0.551 ± 0.006 (1%)
	70	24.9 ± 0.5 (2%)	19.7 ± 0.7 (4%)	24.6 ± 0.8 (4%)	0.556 ± 0.003 (1%)
12	95	25.4 ± 2.0 (8%)	19.6 ± 0.3 (1%)	32.8 ± 1.2 (4%)	0.573 ± 0.003 (1%)
	90	25.3 ± 2.2 (9%)	19.32 ± 0.14 (1%)	31.6 ± 0.5 (2%)	0.566 ± 0.003 (1%)
	70	22.0 ± 3.0 (15%)	19.7 ± 0.4 (2%)	23.9 ± 0.6 (2%)	0.564 ± 0.004 (1%)

[a] Temperature Used, 22°-23°C.

[b] Equilibration Period of 1 hour.

[c] Pre-equilibrated for 16 hours.

NA - Not available.

THE UNCERTAINTIES GIVEN ARE THE EXPERIMENTAL STANDARD DEVIATIONS OF EACH OF THE THREE INDIVIDUAL RESULTS ABOUT THEIR OWN MEAN. THE VALUES IN PARENTHESES ARE THE RELATIVE STANDARD DEVIATIONS.

which determines the FMC of any particular carbon. Two variables which could easily be evaluated for their contribution to this lack of agreement were the initial moisture content (IMC) determinations made by each laboratory that were used in the calculation of their reported FMC and the initial weight of the carbon used by each laboratory. To eliminate any bias in the IC data introduced by these variables, the moisture gained by the test bed was calculated in terms of grams of water gained per initial gram of carbon. The log-linear regression of the average of this quantity versus the average percent penetration for each laboratory yielded a better correlation, $r^2 = 0.64$, than was obtained simply using the reported FMC and more closely approximates the moisture versus penetration relationship determined by INEL sensitivity testing. This improvement in correlation of moisture with penetration is primarily due to eliminating the variability in the data introduced by interlaboratory differences in the determination of the IMC. This indicates the presence of significant errors in the relatively simple determination of IMC, i.e. determining the initial and final weights of a sample of carbon dried at 150°C for a period of 16 hours. The errors are now believed to have been associated with radiative heating of the carbons and inadequate control of temperature.

Six of the U.S. laboratories which participated in the NRC/INEL IC also participated in the CONAGT round-robin. Information supplied by these laboratories indicated that the test results of each laboratory relative to all others was the same for the new carbon tested in the IC as it was for the new carbons tested in the CONAGT round-robin. In other words, when the laboratories are listed in order of penetration results for the new carbons tested in the IC and the CONAGT round-robin, the order is the same. This indicates that these laboratories have been extremely consistent in their test results and discrepancies.

The penetration results for the used carbon samples showed no effect of relative humidity by any of the reporting laboratories under the conditions prescribed, particularly without equilibration and a one-hour challenge time. However, every laboratory reported an average FMC which increased with increasing RH. To clarify this apparent lack of affect of RH on penetration, duplicate tests tests were performed at the INEL on samples of the IC used carbon using a two-hour challenge time at each prescribed RH. These results showed increasing penetration with increasing RH. Although the difference in penetration results between the 90 and 95% RH tests was not significant, the difference between these results and those from the 70% RH tests were statistically significant. This data indicates that, if a two-hour challenge time had been used for the IC, some differences in test results at different humidities would have been observed. Further increases in the length of challenge or testing the samples with equilibration would undoubtedly increase the differences observed in penetration results as a function of RH.

The lack of any effect of RH on the IC used carbon penetration results under the test conditions prescribed allowed an independent evaluation of the effects of other parameters on the test results. The interlaboratory differences observed must have been due to variations in parameters other than RH. INEL sensitivity testing indicated that the parameter most likely to have caused these differences was flow, or superficial face velocity. This testing indicated that a 15% reduction in the specified flow was required to obtain penetration values near the 18% values reported by three laboratories. Although measurement errors of this magnitude seemed unlikely, other explanations for the observed discrepancies were not obvious.

Second Workshop on Activated Carbon Testing

An NRC/INEL Workshop on Activated Carbon Testing was held in Annapolis, Maryland on June 5-6, 1985. The purpose of this workshop was to report the results of the IC to the participating laboratories and other interested parties and to discuss problems with the testing and potential solutions in light of the INEL sensitivity testing and commercial laboratory experience. INEL testing indicated that many of the laboratories might have been operating outside the ASTM specifications for RH and flow. The general conclusion reached was that most of the interlaboratory variance observed might have been caused by differences in calibration of critical parameter measuring devices.

Each of the laboratories represented at the workshop was asked to present any data, results or opinions obtained from their own testing experience which could possibly help to explain the large interlaboratory variance of IC results. These presentations and associated discussions suggested additional factors which may have affected the IC results or their interpretation. The most significant factors noted included: 1) changes in RH during equilibration which would affect the penetration results and might not correlate well with the FMC of the carbon (RH instability and hysteresis); 2) the misapplication or miscalculation of correction factors using incomplete or erroneous information; 3) the presence of water aerosols in the gas stream which would not be detected by a dew point hygrometer but would significantly change the moisture content of the carbon; 4) the chemical purity of the methyl iodide; 5) the lack of uniformity of face velocity or flow at the test bed; 6) the lack of uniformity of the methods used to determine the IMC of the carbon; 7) differences in test bed density and packing methods; 8) differences in the pretreatment of air used to remove potentially important contaminants; and 9) inhomogeneous mixing of the methyl iodide with the main air stream ahead of the test bed.

Each of these factors was subsequently addressed by sensitivity testing at the INEL. Those factors which were shown to exibit significant influence on the penetration results have been addressed by including specific recommendations or requirements in the INEL Standard Test Method for Nuclear-Grade Activated Carbon which has been included in this paper as Appendix A.

NBS Calibration Workshop

The results of the IC and the INEL sensitivity testing indicated that one of the most important causes of the lack of agreement among the participating laboratories was likely the inaccurate measurement of the critical test parameters. To facilitate measurement accuracy and thus promote interlaboratory agreement, a Calibration Workshop was held at the INEL on September 10-12, 1985. All U.S. carbon testing laboratories were invited to observe and participate in the calibration exercise by bringing their own flow and RH measuring devices to the INEL laboratory for in-line comparisons with secondary standards calibrated by the NBS. Two technical experts from the NBS were present to observe and guide these comparisons and to discuss individual problems or comments with the laboratory representatives present.

Comparisons of the commercial laboratory RH measuring devices were directed and observed by Mr. Saburo Hasegawa, the technical specialist in charge of RH measurements at the NBS.(3) These comparisons were performed by placing each commercial laboratory instrument individually in series with the NBS calibrated unit in the INEL test system. These calibrations served to explain some, but not all, of the variations observed in the IC penetration and FMC results.

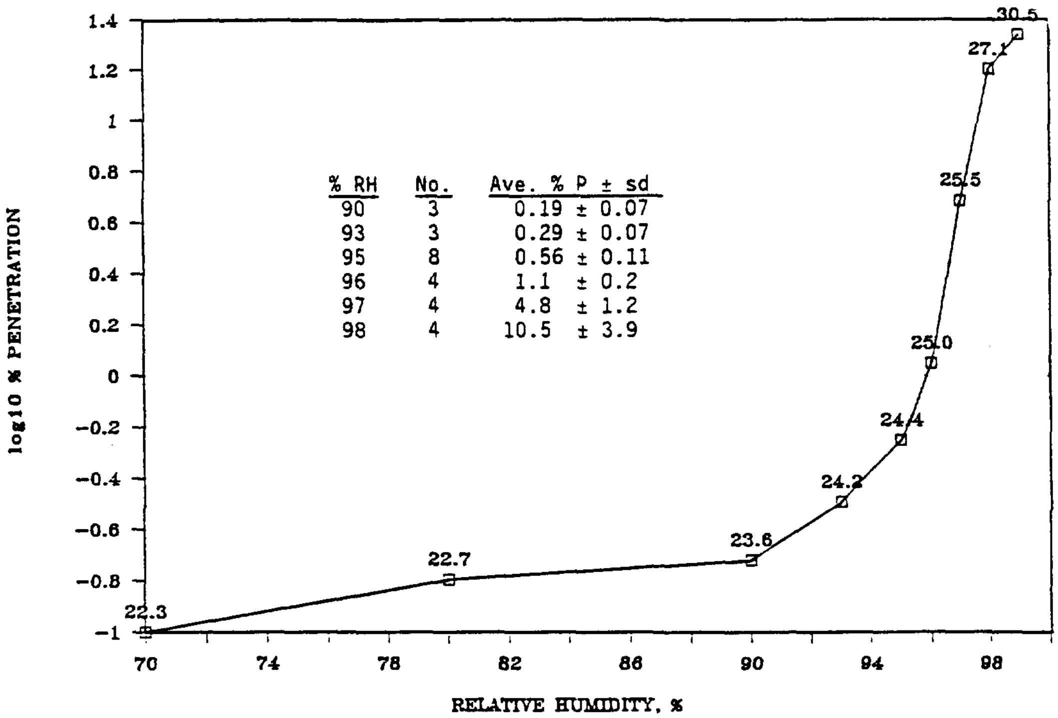
The calibration of flow measuring devices used by the commercial laboratories was accomplished by connecting all the devices in series in the INEL test system. Mr. George Mattingly, the NBS technical expert in the measurement of gas flow, brought an NBS owned and calibrated flowmeter which provided the reference measurements of actual standard flow. The results showed variations in flow calibrations from -4 to +8% of the NBS certified standard flow. This 12% relative difference in flow could account for a 30% difference in penetration results for the IC new carbon based on INEL sensitivity testing. Other problems with the application of flow measurements became obvious through discussions with commercial laboratory personnel. These included: 1) measurement of the exact diameter of the test bed which is used to calculate the flow required to maintain the specified face velocity; 2) the miscalculation of correction factors because of inaccurate measurements of temperature and pressure at the measuring device; 3) the miscalculation or ommission of correction factors required for the addition of gaseous water to the gas stream when "dry" air is actually being measured; and 4) incorrect placement of flow measuring devices behind the test canisters thus subjecting them to variable temperature and RH conditions caused by adsoptive heating of the carbon beds. These problems have been addressed on an individual basis with the commercial laboratories and in a more generic manner in the INEL Standard Test Method, found in Appendix A.

One-on-One Comparisons

Samples of a new carbon, designated 2N, were prepared using the same method as was used for the IC carbons. Samples of this carbon were distributed after the Annapolis workshop to each of the laboratories that participated in the IC. Following the NBS Calibration Workshop, the laboratories were requested to provide IC-type data to INEL personnel from at least one test of this carbon. It was hoped that the calibration of RH and flow measuring devices and the increased awareness by the laboratories of the problem areas identified would improve the agreement of test results. It was also hoped that tests of this carbon would aid the laboratories in attempts to improve their test system designs, instrumentation and operation.

A total of 21 tests were reported from five commercial U.S. laboratories. Penetration results ranged from 0.09 to 4.9%. Reported IMC ranged from 4.1 to 8% and reported FMC ranged from 21.6 to 27.9%. Much of this information was received by phone and is incomplete. Eight tests of the 2N carbon have been performed at standard conditions at the INEL. These tests yielded penetrations from 0.42 to 0.69 with a mean of 0.56 and a standard deviation of 0.11. The mean IMC was determined to be 4.12 ± 0.14 and the mean FMC was 24.4 ± 0.4. Additional INEL sensitivity testing of this carbon has produced a penetration versus relative humidity curve which clearly shows the extreme sensitivity of this carbon to changes in RH, Figure 1. This curve together with additional samples of this carbon may be used by the commercial laboratories as a reference for changes in performance.

The results of these one-on-one comparisons have indicated fairly good performance by some of the laboratories, while others have shown increasing agreement with INEL results. However, some laboratories have presented widely varying results which suggest a lack of parameter control which was not obvious



% RH	No.	Ave. % P ± sd
90	3	0.19 ± 0.07
93	3	0.29 ± 0.07
95	8	0.56 ± 0.11
96	4	1.1 ± 0.2
97	4	4.8 ± 1.2
98	4	10.5 ± 3.9

Figure 1. Results of sensitivity testing of new, co-impregnated, 2N carbon using the IC protocol and varying only RH. Numbers above points on plot indicate final moisture content as % water.

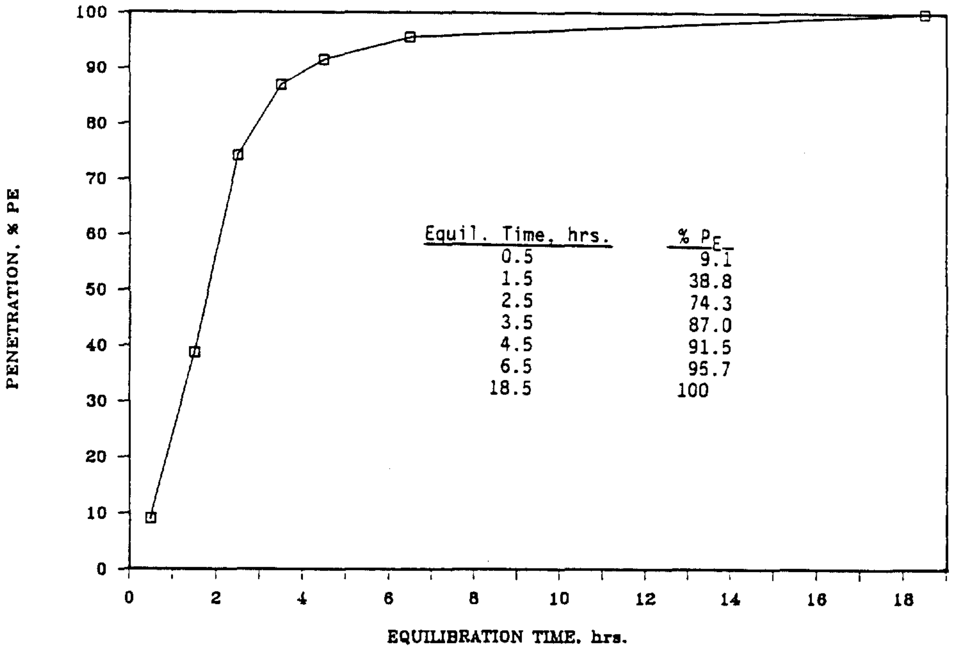
from the IC results, and several commercial laboratories have not participated in this program at all. The acceptability of the current state of U.S. carbon testing is, therefore, still very much in doubt.

Second NRC/INEL Interlaboratory Comparison

Eight U.S. laboratories, including the INEL, and eight foreign labs, one each from Canada, Spain, France, Germany(FRG), Italy, Netherlands, Finland, and Japan, have agreed to participate in the second NRC/INEL Interlaboratory Comparison (IC-2). Samples of one new and one used carbon, designated 3N and 2U respectively, were prepared in the same manner as the IC and 2N carbon samples. These samples were distributed to participating laboratories on June 6, 1986. A minimum of six equilibrated tests and two non-equilibrated tests are to be performed by each laboratory. All tests are to be conducted in accordance with the INEL Standard Test Method for Nuclear-Grade Activated Carbon, included here as Appendix A. Duplicate analyses of each carbon at each condition are required, but triplicate analyses are strongly encouraged. The new carbon is to be tested only at the standard condition, i.e. with equilibration, 95% RH, etc. The used carbon is to be tested at three conditions: 1) standard condition, i.e. with equilibration, 95% RH, etc.; 2) at 90% RH, with equilibration and all other parameters as specified in the standard; and 3) without equilibration, 95% RH, and all other parameters as specified in the standard. Final results of these tests are to be reported in the format given in Annex 1 of the test method by September 1, 1986. The results obtained from the IC-2 should provide a clear indication of: 1) the progress made toward interlaboratory reproducibility of results through this voluntary program; 2) the acceptability of commercial U.S. carbon testing; and 3) the status of carbon testing throughout the world.

Additional Sensitivity Testing

Sensitivity testing was conducted on samples of a fairly good used carbon, designated 2U, by varying the length of equilibration. All tests were conducted at standard conditions, i.e. 30.0°C, 95% RH, 12.2 m/min. velocity, etc., using a 1-hour challenge duration. The equilibration time of the non-equilibrated test was considered to be one-half the challenge time or 0.5 hour. Likewise, the total equilibration time of equilibrated tests was considered to be 0.5 hour plus the actual equilibration duration. The penetration values for these tests are expressed as a percentage of the equilibrated value, P_E, because this carbon is still being evaluated in the IC-2. The results of these tests are shown in Figure 2. It may be observed that this curve is approximately exponential, as will be discussed in the recommendations section following.



Equil. Time, hrs.	% P_E
0.5	9.1
1.5	38.8
2.5	74.3
3.5	87.0
4.5	91.5
6.5	95.7
18.5	100

Figure 2. Penetration of a used carbon, 2U, with equilibration time at the standard IC-2 conditions, i.e. 30°C, 95% RH, etc. The penetration is given as a percentage of that obtained with an 18-hour equilibration.

The Naval Research Laboratory (NRL) has proposed a depth profile method for evaluating the retention characteristics of activated carbon.(4) This method involves dividing the standard 2-inch test bed into four 1/2-inch sections. Each of the sections is counted separately after the standard ASTM test and a linear regression is performed on ln counts per section versus depth. The slope of this regression line, K_L, is then used to compare and evaluate the capability of the carbon. Three different carbons have been tested at the INEL using the IC protocol for new carbon to evaluate the validity and utility of this test method. The results of these tests are presented in Figure 3. The test bed penetration values obtained were 0.54%, 7.45%, and 60.2%, for the 2N, California used carbon, and IC used carbon, respectively. It was hoped that the correlation coefficient for the ln counts versus depth regression could be used as a criterion for the stability and acceptability of the test. Unfortunately, this is not the case.

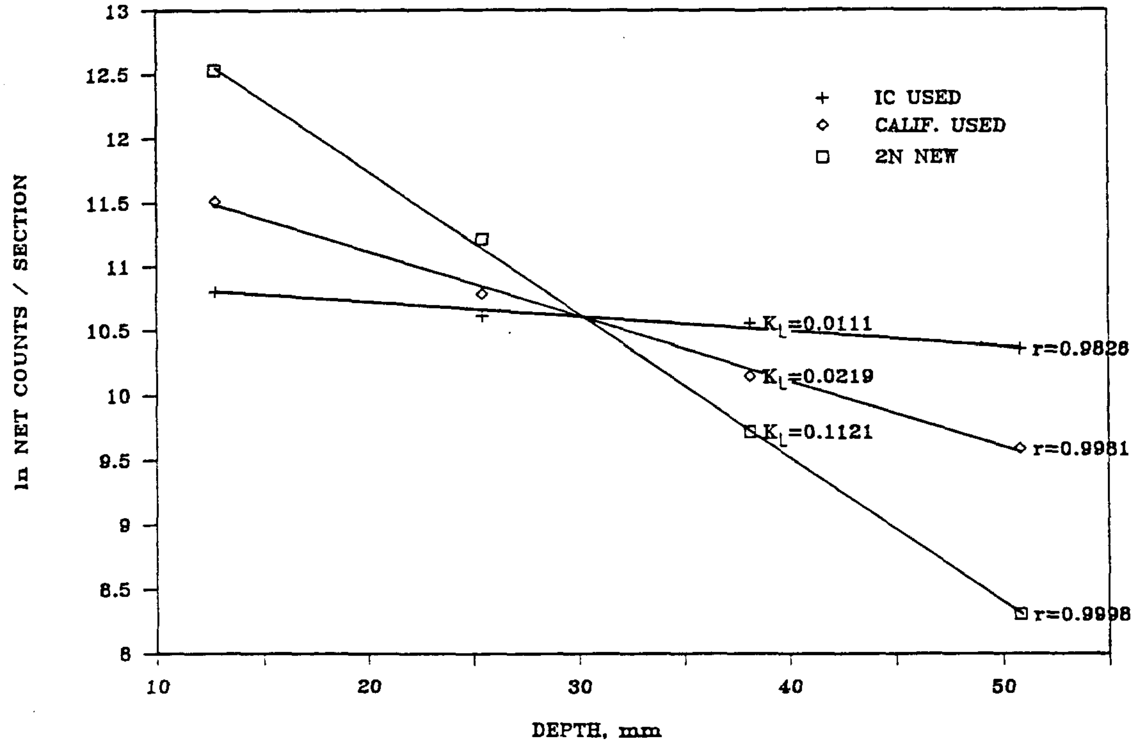


Figure 3. Results of depth profile evaluations of a new, 2N, and two used carbons, California and IC used, tested using the IC test protocol for new carbon.

During the equilibration phase of the test of the California used carbon, the temperature of the constant temperature cabinet dropped 0.6°C in response to a more drastic change in the laboratory. This decrease in cabinet temperature is known to result in a higher test bed RH even though the test gas temperature and RH just prior to the test bed remained at 30.0°C and 95.0% RH. This change in internal test bed RH was reflected in the FMC determinations made for each test bed section, which ranged from 27.4 to 29.6% water, and roughly indicated the time history of the cabinet temperature changes. This variation of a critical test parameter was not indicated by a significant reduction in the K_L correlation coefficient, although it was reflected in a somewhat higher penetration value than was obtained for a stable test under the same conditions. The depth profile method appears to offer no additional information on the activated carbon being tested because each K_L is associated with a unique penetration value measured at 2 in. depth and the general range of the correlation coefficient appears to be inversely related to the penetration value. This depth profile test method is, therefore, subject to all of the parameter stability and measurement problems associated with conventional ASTM carbon testing.

Recommendations

The recommendations presented represent the conclusions of nearly 3 years of laboratory research and extensive interaction with the NRC and members of the commercial carbon manufacturing and testing industry. These recommendations should be utilized by several separate but interrelated groups, including the NRC, commercial carbon testing laboratories, the ASTM D-28.04 committee, and the nuclear power industry. There is much overlap in the application and consequences of the basic findings of this program by the various concerned groups. The recommendations of this program are divided into three basic groups directed toward: 1) a standard test method; 2) the carbon testing laboratories; and 3) the NRC.

A Standard Test Method

A fundamental recommendation of this program must be the elimination of what is essentially "research" required from the commercial laboratories in order to meet the variety of testing requirements found in the nuclear power industry. A standard test procedure should be just that, a procedure which specifies standard test conditions which do not necessarily cover the entire range of power plant operating and accident conditions. If a power plant can show that the test conditions and acceptance criteria prescribed are too stringent for their applications, then research should be conducted to establish an acceptance criteria which is reasonable based on the results of the standard carbon test. In other words, if in-plant conditions do not match the test conditions, change the acceptance criteria rather than the test conditions. Quantitative evaluations of the effects of such things as temperature, flow rate, and carbon bed depth are available in the literature, and are probably more reliable than any single test at non-standard conditions. The reliable and accurate measurement of the penetration of methyl iodide through activated carbon is, obviously, a difficult enough task for the commercial laboratories given a single set of standard conditions.

Recommendations for changes to the ASTM D-3803-79 Standard, Method A have been incorporated into the INEL Standard Test Method for Nuclear-Grade Activated Carbon, included in this paper as Appendix A. These changes include a sweeping revision of the radio-analytical methods prescribed as well as fairly substantial changes to requirements for test procedures and instrumentation. The necessity of revision of the radio-analytical methods was generally accepted and these revised methods have been employed in most laboratories since the first IC. These methods are much less permissive than previous methods and will undoubtedly result in adequate precision in the radio-analytical portion of the test. The significant changes to the experimental portion of the test method are presented following with a brief justification for each.

Equilibration of Used Carbon Samples. Reactor accident scenarios vary widely because of differences in plant design and the exact sequence of events. It is difficult to postulate a scenario in which the occurrence of high humidity conditions and methyl iodide contamination are synchronous and of short duration. However, these are the conditions which are tested by ASTM D-3803, Method A. Methyl iodide insult to the carbon filters will likely follow a period of high humidity which might persist for an extended period of time, as might the iodide contamination. Even if methyl iodide and humidity occur simultaneously, their coexistance in the air challenging the carbon filters may extend for periods of days or even weeks. The resultant cumulative penetration of iodine through these filters approaches the value obtained for equilibrated used carbon.

Parametric studies performed at the INEL indicate that the non-equilibrated penetration results will asymptotically approach the equilibrated carbon result with time. For a fairly good used carbon, the penetration increases rapidly with increasing equilibration time or challenge time without equilibration. This increase in penetration correlates well with the adsorption of water by the carbon for approximately the first six hours, after which, other factors apparently cause the observed minor increase in penetration. Thus, if the carbon is tested without equilibration with the feed period extended to 18 hours, the instantaneous penetration, P_I, at 18 hours will equal the value obtained with equilibration, P_E, but the measured penetration will be the cumulative average of all instantaneous penetrations. The function of instantaneous penetration with time is illustrated in Figure 2, and is easily approximated by a simple exponential function in which the penetration rises rapidly and then levels out as it approaches the equilibrated penetration value. This exponential function has the form

$$P_I = P_E\,[1-e^{-0.5(t)}]$$

where: P_I = the instantaneous penetration, %,
P_E = the equilibrated penetration, %;
t = time since start of test, hours;
and 0.5 = an emperically derived time constant.

The penetration results for the first two hours of the real data are somewhat less than is predicted by this equation, probably due to the increase in test bed temperature and commensurate decrease in test bed relative humidity due to adsorptive heating of the carbon. However, this deviation does not significantly affect the conclusions which may be drawn from this equation. Given the function above, the cumulative penetration is simply the integral of the exponential with respect to t from 0 to 18, which results in a cumulative penetration for an 18-hour challenge test of approximately 89% of P_E. If the test is continued for an additional 18 hours, the resulting penetration would be approximately 94.3% of P_E. Obviously, as the duration of the unequilibrated test increases the resulting penetration value will asymptotically approach the equilibrated test penetration value. The equilibrated used carbon result should therefore be regarded as the best estimate of the long-term average iodine penetration.

Note that these calculations are based on stable 95% RH conditions, which are highly unlikely in a real reactor accident. Testing has shown that relative humidity excursions above 96% dramatically increase the methyl iodide penetration and require a great deal of time to return to the stable 95% RH value. This implies that the equilibrated used carbon test result might still be an underestimate of the true penetration of methyl iodide through activated carbon under accident conditions, but it is probably as good as can be achieved given the uncertainties in predicting actual reactor conditions. In addition, it will probably prove to be a much more accurate, reliable and equitable method for assessing the capability of used carbon.

The equilibration of used carbons prior to challenge can also be justified and indeed is required for the comparison of carbons exposed to different antecedent humidity conditions. Two identical samples of the IC-1 used carbon were each placed in desiccators. One desiccator contained anhydrous calcium sulfate, the other contained a saturated solution of potassium sulfate. The RH in the calcium sulfate desiccator was below 5% and the RH in the potassium sulfate desiccator was above 90%. After slightly more than a month of static equilibration, these carbon samples were tested on successive days using the IC-1 protocol for used carbon. The carbon exposed to low RH yielded a penetration result of 8.4%, while the carbon exposed to high RH yielded a penetration result of 54%. These tests show that identical carbons exposed to different RH conditions prior to testing give significantly different results. It is therefore not legitimate to compare non-equilibrated used carbon test results against a single standard change-out criteria. For example, carbon taken from a plant located on the gulf coast will generally give higher penetration results than carbon taken from the desert Southwest. In order to establish a valid standard test procedure and acceptance criteria, it is necessary to require equilibration for used carbon samples.

An additional benefit of equilibration of used carbon will be to expand the range of penetration results and increase the sensitivity of the method to the degradation of carbon performance. For example, a carbon which yielded an unequilibrated penetration of 25% yielded an equilibrated penetration of 60%. Although both values are well outside any reasonable acceptance criteria, a new standard change-out criteria will need to be established based on more extensive comparative testing. This new acceptance criteria, which should probably be between 10 and 20% penetration, will reduce the experimental and/or interlaboratory uncertainty currently associated with marginal samples.

The primary objection to the equilibration of used carbons has been that "exposure of used carbons to gas flow before challenge with radioactive contaminants may remove some adsorbed material and in effect 'regenerate' the carbon. The test could then show a higher performance than the carbon is actually able to provide." To date, there has not been a single carbon reported to show this enhanced performance with equilibration at 30°C. In fact, all carbons tested have shown significantly increased penetration with equilibration. If this "regeneration" phenomenon exists at all, it is obviously much less significant than the competitive exclusion of methyl iodide by the water added during equilibration. Indeed, the current ASTM test shows a higher performance than the carbon is actually able to provide on a long-term basis.

Equilibrated used carbon testing will increase the continuity of tracking the aging process of carbon. Because of the present specifications for testing, the initial used carbon test results obtained after installation often show an increase in efficiency over the results reported for the new material. This discontinuity has been a source of confusion for some utilities and will be alleviated by equilibration of used carbon samples.

Changes in Duration of Equilibration, Feed, and Elution Periods. Parametric studies performed at the INEL and in Germany indicate no significant differences in penetration results for equilibration durations of 16 and 18 hours, feed durations of 1 and 2 hours, or elution durations of 1 and 4 hours.(5) The changes in feed and elution durations were incorporated into the testing protocol for the first NRC/INEL Interlaboratory Comparison, which was agreed upon by all participating laboratories prior to testing.

The change of equilibration duration from 16 to 18 hours was made as a result of a survey of industry practices and in recognition of the fact that unattended control of critical parameters in the test systems is generally not as good as under close supervision. In the past, the most critical concern in running an equilibrated test was the possibility of test bed flooding during the night which would invalidate the test and cost the laboratory in terms of time and money. To ensure flooding did not occur, the relative humidity level might have been lowered slightly and then readjusted in the morning just prior to the feed period. This practice does not allow sufficient time for the carbon to adjust and results in significantly lower penetration results than those obtained with constant 95% relative humidity equilibration and feed. This change in equilibration time merely allows two additional hours for the carbon to reach a new equilibrium with the adjusted relative humidity. The additional two hour time period appears to be sufficient to reach a new equilibrium if the change in relative humidity is not too drastic.

Changes in Gas Velocity Variation and Absolute Pressure. The change in the permissible variation of gas velocity from 0.5 to 0.3 meters/minute was made to more closely agree with the specification for accuracy of flow measurement of 2%. This change was discussed at some length and agreed upon by all in attendance at the NRC Workshop on Activated Carbon Testing held in Annapolis, Maryland, June 5-6, 1985.

The change in absolute pressure from 104 kPa to 101 kPa was made in recognition of the existence of vacuum induced flow systems which at best can operate at standard atmospheric pressure (101 kPa). INEL testing has shown that over the range of 85 to 115 kPa no significant differences in penetration results so long as the pressure is held relatively constant. Variations in pressure in this range during a single test, however, will affect the water loading of the carbon and often result in increased penetration values.

Change in Maximum Permissible Temperature Variation. The change in the maximum variation of gas stream and constant temperature cabinet/sample bed assembly reflects the importance of temperature in controlling relative humidity and a reasonable judgement of the degree of control easily achievable with current off-the-shelf equipment. Temperature is not only a very important element in overall system control, it is also one of the most easily measured and controlled. Current state-of-the-art PID-type temperature controllers are in fact capable of much finer control than is specified. The INEL test system currently controls the cabinet temperature to ±0.1°C and the test bed inlet gas stream temperature to ±0.05°C. Tests using this system have indicated that even if the test bed inlet gas stream temperature and RH are constant at 30.0°C and 95%, measurable differences in penetration will result if the cabinet temperature and consequently the canister temperature varies by as little as 0.3°C. ASTM D-3803 allows a maximum temperature variation of 0.5°C which in fact exceeds its own prescribed limits for relative humidity if a constant 29.1°C dew point is assumed. Obviously, with simultaneous variations in flow and dew point, and errors associated with dew point hygrometer measurements, the variation due to temperature needs to be limited as much as possible to ensure compliance with the specifications for relative humidity.

The Carbon Testing Laboratories

Nearly all analytical laboratories require some type of internal quality assurance program as a means of maintaining acceptable performance. Even laboratories whose results are not "certified" must use standards as references to interpret their own analytical work. This is not true for carbon testing laboratories. Until the first IC, the common cry was "there is no such thing as a standard carbon". However, the laboratories have proven that a standard carbon can be produced by the intralaboratory reproducibility exhibited in the IC results. Nonetheless, while the rest of the analytical world generally requires a minimum of 10% QA/QC samples, each carbon testing laboratory somehow believes that their answers are the right ones, independent of anyone else. Obviously, a quality assurance program which consists of having their instruments calibrated every six months to NBS traceble standards is not a sufficient guarantee of the accuracy of their test results. It would be in their own best interest to set up an intra-industry quality assurance program with standard samples distributed to each laboratory on a routine basis. Lacking this, such a program will have to be set up by an outside agency. Given the lack of credibility of current carbon testing and the cost to the utilities of mistakes by a laboratory, it might be in the utilities best interest to approach an agency such as the Institute for Nuclear Power Operations (INPO) to implement a laboratory certification program. To date, the NRC has shown extreme patience and concern for the carbon testing laboratories and has given them every opportunity to improve their performance. It is unlikely that the NRC or any other agency involved will show this type of consideration in the future if the past poor performance is allowed to continue.

The NRC

It is recommended that the NRC: 1) adopt a new standard test method similar to the INEL Standard Test Method; 2) implement a mandatory quality control program for the carbon testing laboratories; and 3) re-evaluate and revise the nuclear power plant standard technical specifications for activated carbon performance to reflect the change in testing requirements.

The utilization of the INEL Standard Test Method, or something similar, would not only provide the laboratories with the guidelines they need to perform adequate testing, but would provide results together with supporting data to the utilities which could be evaluated. Allowing licencees to shop-around for a penetration value which meets their technical specification requirements only provides an incentive to those laboratories which tend to bend the testing requirements. The use of a single standard test method would also eliminate confusion by the power plants as to what conditions are to be tested. Indeed, it is entirely possible that carbon which will "pass" the 130°C test specified in Method C of ASTM D-3803 cannot provide the iodine retention capability which might be required of it under real plant accident conditions.

The NRC, as stated previously, has shown extreme patience with the carbon testing laboratories. Hopefully, the results and conclusions of this program will provide sufficient information to provoke some action. It appears that allowing the laboratories to pick and choose those portions of the standard or recommendations which they will implement on a voluntary basis will not solve the problems associated with carbon testing. The NRC cannot continue to allow licencees to fulfill their technical specification requirements by obtaining penetration values with no supporting data and simply trusting the laboratories to follow the insufficient guidelines of the ASTM Standard. A mandatory quality control program should be implemented with satisfactory performance rewarded by certification to test carbon samples for the utilities.

The technical specification requirements for activated carbon performance must be adjusted to reflect the results obtained from a new standard test method. The standard specifications may be set fairly soon if comparative testing proceeds without delay. However, a transition period will be required for implementation of the new requirements and should include reviews for plants whose requirements for performance of the carbon might be less stringent than the standard specifications because of plant design and/or operation.

Acknowledgments

The authors would like to acknowledge the support and encouragement provided by Mr. William P. Gammill, recently retired from the NRC, and Mr. Charles A. Willis, NRC, and the invaluable technical assistance provided by Mr. Fred White in the modification and operation of the INEL test system.

References

1. Sill, C.W., C.D. Scarpellino, et al., "Interim Technical Evaluation Report of Testing Procedures for Activated Carbon Adsorbers in Ventillation Filter Assemblies in Nuclear Power Plants", EG&G Idaho, Inc., Report EGG-PBS-6850 (May 1985).

2. Scarpellino, C.D., C.W. Sill, et al., "Technical Evaluation Report of Testing Procedures for Activated Carbon Adsorbers in Ventillation Filter Assemblies in Nuclear Power Plants", EG&G Idaho, Inc., Report EGG-CS-7131 (December 1985).

3. Hasegawa, S. and J.W. Little, "The NBS Two-Pressure Humidity Generator, Mark 2", J. Res. Nat. Bur. Stand. (U.S.), 81A, No. 1, pp. 81-88 (January - February 1977).

4. Deitz, V.R., "Residual Depth Profile in the Evaluation of Granular Adsorbents", Nucl. Technol., Vol. 73, No. 1, pp. 96-101 (April 1986).

5. Deuber, H. and K. Gerlach, "Parametric Investigations on the Retention of Methyl Iodide by a KI-Impregnated Activated Carbon", Nucl. Saftey, Vol. 26, No. 2, pp. 157-169 (March - April 1985).

APPENDIX A

INEL STANDARD TEST METHOD FOR NUCLEAR-GRADE ACTIVATED CARBON

A Revision of ASTM D 3803-79, Method A.

1. Scope

1.1 This standard covers the most critical test method for establishing the capability of new and used activated carbon to remove radio-labeled methyl iodide from air and gas streams. A single test procedure is described for application to both new and used carbons. The conditions employed were selected to approximate operating or accident conditions of a nuclear reactor which would severely reduce the performance of activated carbons. Increased temperature or decreased relative humidity generally act to increase the efficiency of methyl iodide removal by activated carbon. This test method is a revision of the most stringent ASTM test of nuclear-grade activated carbon and, therefore, is recommended for the qualification of new carbons and the detection of degradation of used carbons.

2. Applicable Documents

2.1 ASTM Standards.

D 2652 Definitions of Terms Relating to Activated Carbon
D 2854 Test for Apparent Density of Activated Carbon
D 2862 Test for Particle Size Distribution of Granular Activated Carbon
D 2867 Test for Moisture in Activated Carbon
D 3195 Recommended Practice for Rotameter Calibration
D 3803 Radioiodine Testing of Nuclear-Grade Gas-Phase Adsorbents
E 300 Recommended Practice for Sampling Industrial Chemicals

2.2 Code of Federal Regulations.

CFR Title 49, Section 173.34, "Qualification, Maintainance, and Use of Cylinders"
CFR Title 49, Part 178, Subpart C, "Specifications for Cylinders"

2.3 Military Standards.

MIL-F-51068D Filter, Particulate High Efficiency, Fire Resistant
MIL-F-51079A Filter, Medium Fire Resistant, High Efficiency

3. Summary of Method

3.1 This test simulates normal containment operating conditions, accident conditions for some systems outside containment, and all conditions for control room atmosphere applications. Botn new and used carbons are first exposed to humid air (pressure, approximately 1 atm.; temperature, 30.0°C; relative humidity, 95%) for an equilibration period of 18 hours. The first 16 hours of this equilibration period may be run unattended with the required parameter monitoring and control devices, but the final two hours must be closely monitored and controlled by qualified laboratory personnel. During the challenge or feed period, radio-labeled methyl iodide at a mass concentration of 1.75 mg/m^3 of humid air flow is passed through the beds for a period of 60 minutes. Following the feed period, humid air flow without test adsorbate is continued at the same conditions for a 60 minute elution period. Throughout the entire test, the effluent from the sample bed passes through two backup beds containing carbon having a known high efficiency for methyl iodide. The two backup beds trap essentially all the radio-labeled methyl iodide that passes the test bed, and provide a differential indication of their efficiency. At the end of the elution period, the gamma activity of ^{131}I in the test and backup beds is measured by a gamma counter, and the percent of adsorbate penetrating the test bed is determined.

4. Definitions

4.1 The following terms have special meanings for the purposes of this standard. Additional terms relating to this standard are defined in ASTM D 2652.

4.1.1 Penetration (P) - The percentage of the contaminant (CH_3I) which passes through the equilibrated test bed of standard depth, and is collected on the backup beds during the feed and elution periods under specified conditions.

4.1.2 Efficiency (E) - The percentage of the contaminant removed from a gas stream by an adsorption bed: E,% = 100 - P%.

4.1.3 Relative humidity (RH) - For the purpose of this test relative humidity is defined as the ratio of the partial pressure of water in the gas to the saturation vapor pressure of water at the gas temperature and pressure. At temperatures below 100°C, this is the normal definition and relative humidity can range from 0 to 100%.

4.1.4 Counter Efficiency (CE) - The fraction of the actual number of disintegrations of a radioactive sample that is recorded by a nuclear counter.

5. Apparatus and Materials

5.1 Sample Preparation Apparatus.

5.1.1 Riffle Sampler, in accordance with 30.5.2 of ASTM E 300, Recommended Practice.

5.1.2 Feed Funnel and Vibrator, in accordance with 2.2 and 2.3, respectively, of ASTM D 2854.

5.1.3 Balance, capacity > 200g, accurate to ±0.01g.

5.2 Sample and Backup Bed Assemblies.

5.2.1 The sample bed canister and backup bed canisters must each be either a single unit capable of containing carbon to a depth of 50mm or they may be assembled from two separate units each capable of containing carbon to a depth of 25mm. Two backup canisters, each of 50mm total depth, are required. Canisters may be reused after being decontaminated to remove residual radioactivity.

5.2.2 Clamping assemblies are needed for sample and backup beds. The only requirements for these assemblies are that they provide a smooth sealing face, uniform alignment of bed canisters, and sufficient clamping force so that the leak test in 8.2 can be met.

5.3 Test System - A single test system cannot be used for all the test conditions currently required by commercial nuclear power plants. Because of the range of conditions and accuracy of parameter measurement and control required, a minimum of two test systems are necessary, one for tests conducted below 100°C and near standard atmospheric pressure, and one for the higher temperature and pressure tests. As an example of a test system which can meet the parameter requirements of this test method as listed in Table A-1, a schematic of the INEL test system is shown as Figure A-1. This system is designed to operate at approximately 30°C and 95% RH, with a gas flow of 24.7 l/min at atmospheric pressure. If test conditions which differ significantly from these are required from the system, then separate calibrations and/or instrumentation may be required for use in these parameter ranges.

5.3.1 Saturator System - The system may be a controlled temperature saturator (bubbler) or spray chamber (environmental condition generator) or any other device of sufficient stability and capacity to supply the required mass flow of water vapor at test conditions.

5.3.2 Flow Generator - This system may be an air compressor upstream of the test system or a vacuum pump downstream of the test system. A dryer, carbon adsorber, and HEPA filter are required for either system to condition the inlet air. Flow measurement and control should be accurate and stable to within ±2%. System capacity shall meet or exceed the volumetric flow requirements as calculated from the specified face velocity. A surge tank and pressure control valve should be employed in either type of system to ensure stable and accurate flow measurement and control. For safety, it is important that the pressure system be equipped with a pressure relief valve. It is important that the pipe diameter and inlet air filters for a vacuum system be designed and maintained to minimize the pressure drop from ambient to ensure that the specifications for pressure at the test bed are met.

5.3.3 Moisture Separator - should be used to protect the HEPA filter by removing large quantities of entrained particulate water, if present, after humidification. A HEPA filter or equivalent is required to function as a final droplet trap to remove small amounts of fine particulate water from the carrier gas ahead of the test bed.

5.3.4 Test Adsorbate Supply - The system shall consist of a stainless steel cylinder, pressure gage, pressure regulator, and a flow regulator capable of providing a steady flow of the challenge gas, i.e. radio-labeled methyl iodide in dry nitrogen, for the duration of the test feed period. The point of injection into the main gas flow of the system must be such that the cross-sectional distribution of the adsorbate at the face of the test bed can be ensured to be homogeneous. A mixing chamber, baffles, glass beads, etc. should be used to acheive adequate mixing.

5.3.5 Constant Temperature Cabinet/Sample Bed Assembly - An enclosure and associated thermoregulatory system must be used that is capable of maintaining temperature to ±0.2°C. All tubing downstream of the moisture separator, the carbon bed canisters and holders, temperature and pressure ports and measurement devices upstream and downstream of the test bed, and an upstream port and tubing to the dew point sensor all must be included within the temperature controlled enclosure. In addition, it is highly recommended that a bypass line be included around the sample bed assembly to avoid exposing the sample to start-up conditions possibly outside those specified.

5.3.6 Air Flow Measurement and Control - Includes mass flow controllers, control valve and oriface meter, rotameter or any device with adequate stability and a demonstrated measurement accuracy of ±2% over the range of test conditions. All flow measuring devices must use correction factors for interpretation and application to actual test conditions. These factors must be carefully predetermined and documented. No flow measuring device should be located directly downstream of the test bed such that it is subject to variable temperature and humidity conditions during a test as a result of water adsorption by the carbon.

5.3.7 Interconnecting Tubing - Tubing beyond the point of methyl iodide injection shall be glass or stainless steel. No rubber, polyethylene, or other organic tubing should be used. Interconnections should be kept as short as possible to reduce the possibility of water condensation.

5.3.8 Temperature Measurement Devices - with certified accuracy and calibration to ±0.1°C are required, three or four wire platinum resistance thermometers (RTD's) are strongly recommended.

5.3.9 Pressure Measurement Devices - Absolute pressure measuring devices must be accurate to within ±2% at standard atmospheric pressure and be capable of digital or analog output to meet the specified recording requirements. The differential pressure device required for measurements across the test bed must be capable of detecting a 0.25 kPa pressure difference and be accurate to within ±2% at the normal operating ΔP.

5.3.10 Heaters and Temperature Controllers - Must be capable of maintaining the inlet gas stream temperature from the point of humidity control to the test bed, and the surface temperature of all carbon canisters within ±0.2°C.

5.3.11 Humidity Measurement - An optical dew point hygrometer with demonstrated accuracy and calibration to ±2% relative humidity at 95% RH and 30°C is required for measurement of RH of the gas stream immediately upstream of the test bed. A secondary check on this measurement device is required to ensure that calibration offset has not occurred. This secondary device may be another optical dew point hygrometer, wet bulb/dry bulb, or any other device capable of ±3% accuracy.

5.3.12 Data Recording - To meet the reporting requirements of 12.1.8-11, the use of potentiometric recorders or a data logger capable of recording temperatures, pressures, flow, and relative humidity data at a maximum of 5 minute intervals is required.

5.4 Gamma Detection System - Any reliable and efficient detection system for gamma rays of 365 keV energy is permissible, provided it produces actual counts of gamma photons and not an analog rate output, and provides adequate elimination of any interferences that might be present. Systems equipped with internal computers that make calculations or corrections for such things as dead time, counting efficiency, decay rates, etc. are also permissible provided they give accuracy equal to that required in this standard. In many cases, either thallium-activated sodium iodide well counters or single- or multi-channel gamma spectrometers that use thallium-activated sodium iodide, lithium-drifted germanium, or intrinsic germanium detectors can be used with appropriate professional guidance, proper shielding of at least 4 inches of lead, and preferably graded adsorbers of cadmium and copper to reduce the production of X-rays in the shielding. When significant gamma-emmitting interferences are absent and penetration of ^{131}I through the test bed is greater than a few tenths of percent, either the principal ^{131}I photopeak at 364.46 keV or the entire spectrum including the Compton continuum can be used. However, when the penetration is low, a multi-channel spectrometer with a germanium detector will be required for the most accurate measurements. This is necessary to identify the ^{131}I in the presence of the ^{214}Pb daughter of ^{226}Ra generally present in carbon, and to permit Compton correction for gamma-emitters such as ^{40}K and daughters of ^{226}Ra. The test bed, backup beds, and carbon backgrounds must all be counted under identical geometrical conditions. This requires the use of a jig on the detector to hold each counting bottle in identically the same position.

5.5 Materials

5.5.1 Air - Compressor, used for pressure systems, shall be of the oil-free type to minimize injection of hydrocarbons into the system. Line filters shall consist of a dryer, activated carbon and HEPA filters and shall be adequately sized and maintained.

5.5.2 Water - Deionized and/or distilled water must be used for water-vapor or steam generation.

5.5.3 Radio-Labeled Methyl Iodide - Shall be prepared and/or diluted with analytical grade chemicals and/or purchased from a reputable vendor. The methyl iodide solution should be stored in the dark below 0°C to slow its decomposition to I_2. The activity of the ^{131}I should be such that the total activity in the entire spectrum from the test bed is between 10^3 and 5×10^5 counts per minute.

5.5.4 Backup Bed Carbon - Shall have a penetration of no more than 2% when tested by this standard method. The calculation of the efficiency of the first backup bed is required for each test.

5.5.5 HEPA Filter Media - In accordance with MIL-F-51079. If a pleated filter is used in place of a flat sheet, it shall be constructed in accordance with MIL-F-51068.

6. Safety Precautions

6.1 Overpressure - The contaminant feed system makes use of dry nitrogen from standard high-pressure gas cylinders, a contaminant feed cylinder which is pressurized, and associated regulators and tubing for transport of the contaminant gas. This system must be designed with adequate saftey factors. Standards for the fabrication of such pressure vessels and associated fittings are contained in 49 CFR 173.34. Elastomeric seals must be replaced on a regular basis or if damaged to ensure system integrity.

6.2 Radioactivity - The radiotoxicity of ^{131}I is well documented. The species used in this test is very volatile and easily inhaled. Rigorous health physics procedures must be followed whenever handling the radioisotope and routine thyroid counting must be provided for laboratory personnel. The system must be adequately vented through a filter system capable of handling the maximum possible contaminant release. Radiation shielding and dosimetry must be provided to limit and monitor worker exposures in compliance with federal and state nuclear regulations. Personnel access to the system should be strictly limited and workers should be trained in health physics procedures.

7. Sampling

7.1 Guidance in sampling granular activated carbon is given in ASTM E 300, Recommended Practice.

7.2 Occasionally, samples received for laboratory analysis are not of sufficient quantity to fill the test canister to the standard depth of 5.08 cm (2 inches). If possible, another sample should be obtained. However, this is not always possible because of critical time constraints. If a substandard quantity of carbon must be tested, the resulting actual penetration value must be converted to the predicted penetration at the standard depth and noted as such on the report form. This conversion is based on the log-linear function of penetration with depth and has the form

$$P_s,\% = 100 \exp\{[\ln(P_a/100)](5.08/d)\}$$

where: P_s = the predicted penetration, in percent, at the standard depth;
P_a = the actual penetration, in percent, at the substandard depth;
and d = the substandard depth, in centimeters.

8. Preparation of Apparatus

8.1 Assemble the test apparatus making sure that the placement of RTD's is such that they accurately reflect the temperature of the gas stream and the temperature controlled cabinet or test canister, and are not subject to radiative heating.

8.2 Leak testing of the system designed to test carbon at standard atmospheric pressure should be performed on a routine basis, recommended prior to each test. This test should be a pressure decay test for pressure induced flow systems or a vacuum decay test for vacuum induced flow systems. The system should be pressurized to approximately 125 kPa or depressurized to approximately 75 kPa with filled test and backup cannisters in place. The system should then be isolated, i.e. sealed at all atmospheric connections, and the pressure change with time recorded. This rate of change should not exceed 5 kPa in 30 minutes to ensure safety and the accuracy of flow measurement.

8.3 To ensure the accuracy of relative humidity measurement, a check of the differential pressure between the test bed and the sensor of the optical dew point hygrometer should be performed initially and whenever the system is modified or semi-annually. This check should be performed with the test and backup canisters filled with carbon and with the system operating at the standard conditions specified, i.e. temperature, flow, RH, pressure, etc. This differential pressure should not exceed 1 kPa or must be corrected for either in the calculation of relative humidity, or preferably by modification of the test system to reduce the ΔP.

8.4 Correction factors for flow measurement devices, esp. rotameters, must be predetermined by the comparison of accurate pressure and temperature measurements made at the device and at the test bed under normal operating conditions. Correction factors to convert standard flow to actual flow at specified conditions, which may include the addition of a significant amount of water vapor, should be carefully determined for all measurement devices. No flow measuring device should be located directly downstream of the test bed such that it is subject to variable temperature and humidity conditions during a test as a result of water adsorption by the carbon.

8.5 From the determination of the "as received" apparent density (ASTM D 2854) and a knowledge of the exact volume of the test canister, determine the weight of carbon required to fill the test bed. Weigh this amount of carbon to the nearest 0.01g and record this weight and its final weight after the test on the report form as shown in Annex A-1. Determine the dry weight of carbon used in the test bed from the initial moisture content as determined using ASTM D 2867 Oven Drying Method, except using a 16 hour drying time. Record the initial and final sample and sub-sample weights and calculate the initial, adsorbed, and final moisture content, %, as described in section 11.1.

9. Calibration

9.1 RTD's, optical dew point hygrometers and other humidity measuring devices, flowmeters, pressure transducers, balances, radiation survey meters, and gamma detection systems shall be calibrated at intervals of not more than six months, or whenever a detectable change in performance occurs, with NBS traceable standards. Devices which are required to output data for recording should be calibrated together with the readout device to the accuracy specified.

10. Procedure

10.1 Stabilization Period - Install the filled test and backup canisters in the system. Perform the leak test described in 8.2 to ensure system integrity. Bring the system up to operating conditions, see Table A-1, and allow it to stabilize while bypassing flow around the test and backup beds. The duration of this stabilization period is recommended to be a minimum of two hours, during which the canisters and carbon must come to thermal equilibrium at the specified test temperature.

10.2 Equilibration Period (for new and used carbons) - Air with 95 ±2% relative humidity at a temperature of 30.0 ±0.2°C is run through the beds for 18.0 ±0.2 hours. This length of time ensures that the test bed carbon will reach equilibrium with the flow of humid air. There will be a sudden change in relative humidity at the start of equilibration that will produce a rapid temperature rise in the carbon caused by the heat of adsorption of water. The extent of this temperature rise cannot be controlled and depends upon the condition of the carbon. The conditions at the test bed inlet must be held at the specified conditions, Table A-1.

10.3 Feed Period (Challenge) - Humid air flow is already at the prescribed conditions, Table A-1, at the start of the feed period. Flow is maintained at 30.0 ±0.2°C at 95 ±2% relative humidity for 60 ±1 minutes with 1.75 ±0.25 mg/m^3 of radio-labeled CH_3I in the total system gas flow provided by the addition of a small and continuous flow of the challenge gas during the feed period.

10.4 Elution Period - To evaluate the ability of the carbon to hold the adsorbate once it is captured, flow is continued at the end of the feed period without change of the flow rate, relative humidity or temperature for a period of 60 ±1 minute. (See Table A-1.)

10.5 Monitor and record gas stream temperatures upstream and downstream of the test bed. A decrease in the downstream temperature is indicative of bed "flooding", the condensation of free water in the sample bed, and the test should be aborted. Monitor temperatures, pressures, humidity, and air flow at least every 5 minutes or continuously by means of a data logger or recorder. Also monitor the pressure drop across the bed. Erratic readings or a substantial increase in this differential pressure is an additional indication of test bed flooding.

10.6 At the end of the elution period, switch the system to bypass mode and shut down the system. Remove and disassemble test and backup beds. Transfer the carbon from the canister to a jar with a volume at least twice that of the carbon. Roll and tumble the jar gently until the carbon is thoroughly homogenized then transfer the blended carbon to a pre-tared plastic counting bottle sufficiently large to accomodate all of the carbon and tamp it down to some reproducible height. The counting bottle must be immediately sealed and reweighed to the nearest 0.01g. These weights should then be recorded on the report form.

10.7 Counting Conditions - It is never permissible to count the ^{131}I activity in the test and backup canisters directly as obtained from the test. The carbon from each canister must be counted in a plastic counting bottle having rigid vertical sides and uniform wall thickness and internal diameter, and then tamped down to a standard and reproducible height. The packing density is not particularly important for gamma counting within the range of densities likely to occur, but the geometrical angle subtended between the sample activity and the detector is of great importance if accurate results are to be obtained. Because penetration is simply a ratio of counting rates, absolute counting efficiencies are not necessary unless an independant determination of the total quantity of radio-iodine is desired. The carefully filled and weighed counting bottles should be placed on the detector in a jig that will guarantee reproducible positioning. Count for whatever period of time is necessary to obtain the desired sensitivity and precision. Calculate the results and propagate the statistical uncertainties as described below.

10.8 Gamma Count Corrections - If each test and backup carbon is homogenized and counted under identically the same conditions of height and geometry in identical counting bottles, no corrections are necessary for attenuation of the gamma rays by either the carbon or the counting bottle, or for geometry or counting efficiency. Corrections for dead time in the counter system are avoided by simply controlling the quantity of radioiodine used in each test. This simple and expedient method also minimizes costs of tracer, both internal and external dose to those operating the test system, and waste disposal. The principal corrections required are those for decay of the ^{131}I activity and for the carbon background, including the Compton contribution from higher energies when such interferences are present and a spectrometer must be used. When counting times can be kept short and all samples are counted with dispatch, even the decay correction can be made negligible, although this is an unnecessary limitation on the procedure.

10.9 Counting Efficiency - Determination of the counting efficiency is unnecessary as far as the measurement of penetration is concerned, and is undesirable because of the extra time and the standard ^{131}I solution that are required. However, if a separate determination of the quantity of ^{131}I used is desired, the counting efficiency can be determined rather simply. Fill a standard counting bottle with carbon to the standard height used in the test procedure. Determine the volume of water required to fill the interstitial voids just to the top of the carbon. Count this sample under the standard counting conditions to determine the blank. Measure an exact volume of a standard solution of ^{131}I of such activity that dead time effects are kept below about 1%. Dilute with water in a non-wetting plastic beaker to the volume determined previously to fill the carbon voids. Repack another counting bottle with carbon to the standard height and add the diluted iodine solution. Count under the identical conditions being used for the test samples, and as were used for the blank. The slight difference in attenuation of the gamma rays due to the water added will certainly be much less than the errors due to inhomogeneous absorption of small volumes of tracer in the carbon without water present. The counting efficiency, CE, is then

$$CE = (R_s - R_b)\ (\exp 0.003592\ t)/A_s$$

where: R_s = counting rate of ^{131}I standard, counts per minute;
R_b = counting rate of background, counts per minute;
A_s = activity of ^{131}I standard taken, as of time of standardization of original solution, disintegrations per minute;
t = length of time between standardization of original solution and counting, in hours;
and 0.003592 is disintegration rate per hour for 8.041-day ^{131}I.
for CE = net counts-per-minute per disintegration-per-minute of ^{131}I at the same time.

10.10 Decay Correction - If the carbon from different canisters from a given test are counted at significantly different times, they must each be corrected for decay to some common base time in order that the counting rates obtained be comparable. Although other times can be used for zero time, it is convenient to correct all counts back to midnight of the first day in which counting for a particular test was done. Using the 24-hour clock, times can be read directly from a watch to the nearest quarter hour, and the various beds can be counted in any order. For ^{131}I compounds, the correction is

$$R_0 = R_t \exp(0.003592\ t)$$

where: R_0 = equivalent counting rate at time zero (midnight);
R_t = counting rate at time, t;
and t = elapsed time between zero time and counting time, hours.

Generally, the counting interval will be small compared to the decay time so that the beginning of the count can be used to calculate the elapsed time. However, the midpoint of the counting interval gives better accuracy and is just as convenient to use. It should be emphasized that the decay correction should be applied to the net counting rate after correction for background, i.e. obviously the background does not decay with the halflife of ^{131}I.

10.11 Radioactivity and Counting Times - Corrections for dead-time losses of counting rate due to overloading the counting system by using too much activity can never be made as accurately or conveniently as avoiding such losses from the beginning. Such losses are particularly undesirable when the penetration is low and very large errors are incurred for the test bed with virtually no error from this source for the backup beds. Locating the test bed counting bottle some distance from the detector and counting only a small fraction of the total flux emitted to bring it within the proper range is neither desirable nor prudent. Consequently, the activity of the ^{131}I used in each test should be such that the test bed will not contain more than about 5 x 10^5 counts per minute of total activity incident upon the detector and associated electronics to avoid the increased uncertainties of making large corrections for dead-time effects. When gamma spectrometry is used, this applies to the total events being processed by the ADC for the entire spectrum, not just those of interest in the 365 keV photopeak. On the other extreme, the activity used should be kept sufficiently high to give 10^3 to 10^5 counts per minute in the test bed to keep the sensitivity and precision of the measurement high without requiring prolonged counting times, particularly when using just the photopeak in gamma spectrometry. Thus, the activity on the test bed can be measured with a relative standard deviation of a few tenths percent with counting times of a very few minutes. For carbon backgrounds and backup beds containing low activity, the counting times should be 30 minutes to 1 hour with gross counters or 1 to 2 hours with spectrometers using just the iodine photopeak. This will permit the iodine activity in the backup beds to be detected above the carbon background and the Compton continuum with reasonable statistical certainty.

10.12 Determination of Contaminant Mass - The efficiency factor can provide an independant means for determining the mass of the contaminant. The equation is

$$M = [\Sigma(R_t - R_b)]/(2.22 \times 10^6 E A_s)$$

where: M = mass fed during test, g;
R_t = count rate for test or backup bed, corrected to base time, counts per minute;
R_b = background count for canister corresponding to R_t, counts per minute;
E = efficiency factor for gamma counter.
and A_s = contaminant specific activity at base time, μCi/g;

11. Calculations

11.1 Initial, Adsorbed, and Final Moisture Content - Initial moisture content (IMC) as weight percent water is simply 100 times the difference in weight of the subsample of carbon before (W_b) and after (W_a) drying for 16 hours at 150°C divided by its initial weight, i.e.,

$$\mathrm{IMC},\ \% = 100(W_b - W_a)/W_b.$$

The percentage moisture absorbed during the test (MADT) is similarly 100 times the difference between the final weight (W_f) of carbon in the test canister after the test and the initial weight (W_i) of carbon in the canister before the test divided by the final weight, i.e.,

$$\mathrm{MADT},\ \% = 100(W_f - W_i)/W_f.$$

The final moisture content (FMC) is

$$\mathrm{FMC},\ \% = (\mathrm{IMC},\%)W_i/W_f + (\mathrm{MADT},\%).$$

11.2 Penetration - All counting must be corrected for the corresponding background counting rates before other corrections are applied. The net activities are then corrected for decay from counting time to some common time zero before calculation of penetration. The halflife and disintegration constant of ^{131}I are 8.041 days and 0.003592/hour, respectively. Because counting efficiencies are not required when counting conditions are kept the same for all fractions, calculate percent penetration as follows:

$$P,\% = 100\ (B + C)/(A + B + C)$$

where: A = net counting rate of the ^{131}I activity collected in the test bed, cpm;
B = net counting rate of the ^{131}I activity collected in the first backup bed, cpm;
and C = net counting rate of the ^{131}I activity collected in the second backup bed;
for beds of equal depth,counted under identical conditions, and corrected for decay.

Obviously, efficiency, E, of the test bed in percent is

$$E,\% = 100 - P = 100(A)/(A+B+C).$$

The efficiency with which the ^{131}I activity passing the test bed was retained by the first backup bed is similarly

$$E_{bu},\% = 100\ B/(B + C)$$

This calculation is important in showing whether or not all the activity passing the test bed was collected, and whether or not the proper blank corrections are being made. When penetration is low and corrections for blanks and/or the Compton continuum are not made, "C" can be larger than "B" and the results will be grossly inaccurate. Specific equations are given in Annexes A-2 and A-3 for calculating both penetration of the test bed and efficiency of the first backup bed from the raw data obtained in a gross counter or in a gamma spectrometer, respectively.

11.3 Error Propagation - The uncertainty with which the measurement was made, expressed as one standard deviation, must be calculated for each measured value of penetration of the test bed and efficiency of the first backup bed. The uncertainty must include every statistical uncertainty incurred anywhere in the entire measurement process, all propagated to the final result by the well-known

Law of Propagation of Error. Thus, the standard deviation of percent penetration defined above is

$$S_P = 100 \frac{\{(B+C)^2(S_A)^2 + (A)^2[(S_B)^2 + (S_C)^2]\}^{0.5}}{(A+B+C)^2}$$

where A, B, and C have the same meanings as above and S is the estimate of the standard deviation of the corresponding quantities. It should also be noted that the standard deviation of efficiency by the test bed has the same absolute value as that of penetration of the test bed, i.e. $S_E = S_P$. Similarly, the standard deviation for percent efficiency of the first backup bed is

$$S_{E(bu)} = 100\ [C^2(S_B)^2 + B^2(S_C)^2]^{0.5}/(B + C)^2.$$

Specific equations are also given in Annexes A-2 and A-3 for calculating the standard deviations of both penetration of the test bed and efficiency of the first backup bed from the raw data obtained in a gross counter and in a spectrometer, respectively.

12. Report

12.1 Report the following information on a standard report form similar to that shown in Annex A-1 for each measurement of penetration:

12.1.1 Name, address, and phone number of laboratory making the test.

12.1.2 Name, signature and months of experience of technician performing test, and name and signature of supervisor approving test.

12.1.3 Date of test.

12.1.4 Source of sample including, for used samples, the original log number of the sample, the filter system and time for which it was exposed, if available.

12.1.5 Type of carbon, supplier name, grade designation, base material, impregnant(s), lot and batch numbers, if available.

12.1.6 Particle size distribution (new carbon only) in accordance with ASTM D 2862 method.

12.1.7 Apparent density for new and used carbons in accordance with ASTM D 2854 method, except determined "as received", i.e. not dried.

12.1.8 Maximum, minimum, average, and standard deviation for gas temperature immediately upstream of the test bed for each of the test periods.

12.1.9 Maximum, minimum, average, and standard deviation for pressure at the test bed for each of the test periods.

12.1.10 Maximum, minimum, average, and standard deviation for relative humidity as measured just prior to the test bed for each of the test periods.

12.1.11 Maximum, minimum, average, and standard deviation for the actual gas flow for each of the test periods.

12.1.12 Report the initial moisture content, %, of the test carbon together with the initial and final weights of the sample used for the moisture test. Also report the initial and final weights of the test bed carbon along with its adsorbed and final moisture content, %.

12.1.13 The duration of each of the test periods.

12.1.14 The penetration of the test bed and the efficiency of the first backup bed will be calculated for each separate measurement made. Every measured value will be reported as a finite number to the proper number of significant figures as indicated by the value of the standard deviation, including negative signs if obtained. No subjective judgements are permitted such as rounding negative results to zero or reporting results as "less than" some arbitrary figure. All raw data obtained will also be reported along with the calculated result, including total counts, counting times and decay times of the test bed, all backup beds, carbon backgrounds, etc., as illustrated in Annex A-1.

12.1.15 Every measurement of penetration of the test bed and efficiency of the first backup bed will be accompanied by an estimate of the statistical uncertainty with which each measurement was made, reported as one standard deviation of all random uncertainties incurred in the entire measurement process, not merely the standard deviation of sample counts.

Table A-1. Parameter Specifications for the INEL Standard Test Method for Nuclear-Grade Activated Carbon.

Parameter, unit	Specification ± Maximum Variation
Temperature, °C	30.0 ± 0.2
Relative Humidity, %	95 ± 2
Absolute Pressure, kPa	101 ± 5
Gas Velocity, m/min.	12.2 ± 0.3
Gas volumetric flow, l/min. (assuming 50.8 mm bed dia.)	24.7 ± 0.7
Bed Diameter & Depth, mm	50 ± 1
Equilibration Period Duration, hr.	18.0 ± 0.2
Feed Period Duration, min.	60 ± 1
Adsorbate Concentration, mg/m^3	1.75 ± 0.25
Elution Period Duration, min.	60 ± 1

Note: Temperature, relative humidity, pressure, and gas velocity are to remain constant within the specified maximum variations throughout the entire test, i.e. for each test period. Parameter excursions outside the specified limits during the last four hours of the test, i.e. the last two hours of equilibration and the feed and elution periods, will invalidate the test results. Parameter excursions during the first 16 hours of equilibration which are more than twice the specified variation shall invalidate the test results.

It must also be noted that testing has shown that unattended equilibration (16 hr.) of new carbon at ≥ 96% RH followed by two hours of attended equilibration at 95% RH results in significantly higher penetration values than those obtained with 18 hours of constant 95% RH equilibration. It is, therefore, very important to maintain the relative humidity below 96% during the 16 hours of unattended equilibration.

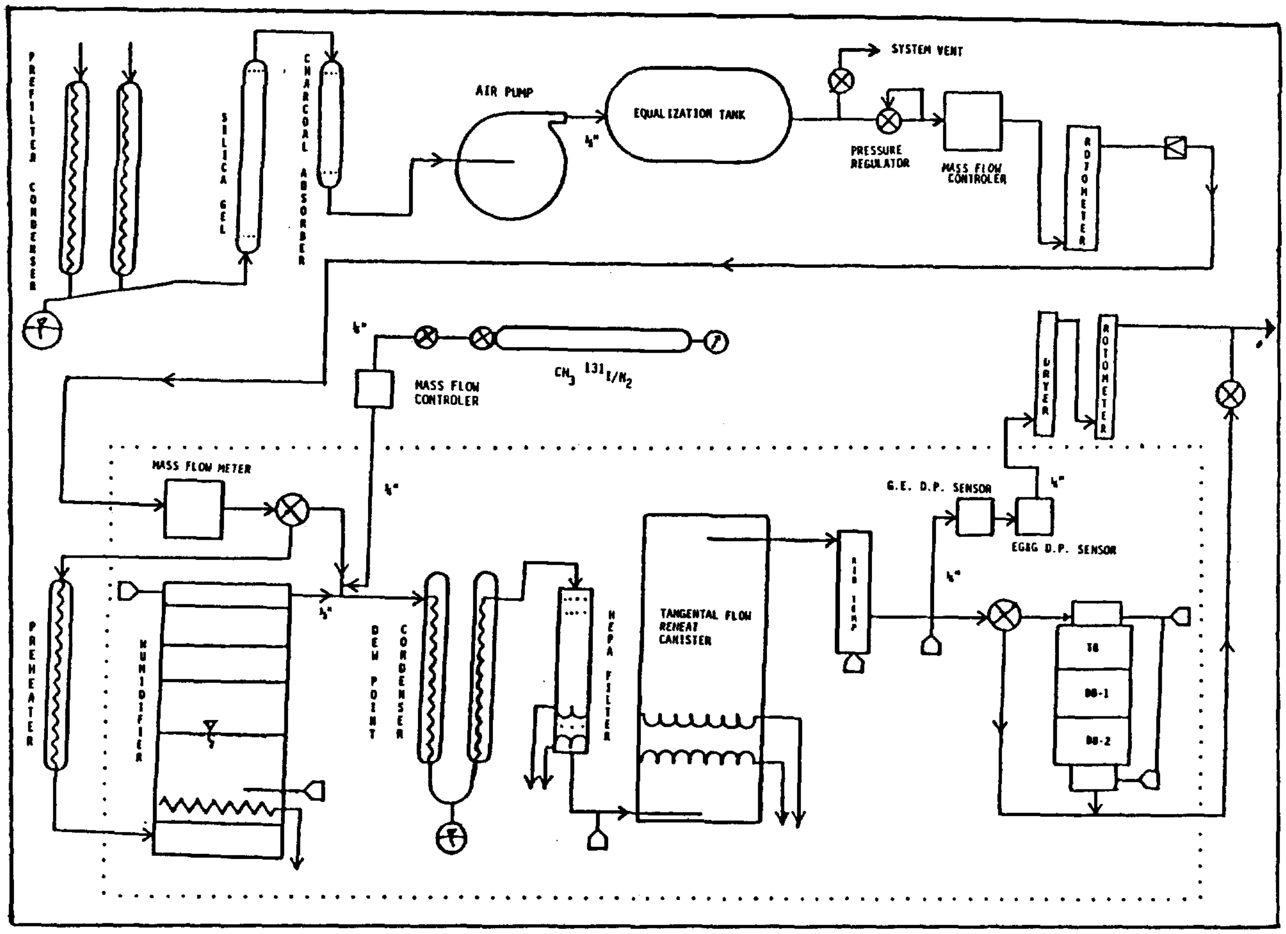


Figure A-1. A Schematic of the INEL Activated Carbon Test System, presented here as an example of a system design which will meet all of the specifications of this standard.

Annex A-1. Report Form for Results of INEL Modified Test Method.

Test Laboratory (12.1.1)

Name:________________________
Address:______________________
____
Telephone:______________________

(12.1.2 & 3) Test Date:__________

Technician
Name:________________________
Signature:________________________
Months operating test system:______.

Supervisor Name:________________________

Signature:________________________

Source of Sample (12.1.4) : New ___ or Used ___ ;
if used, Original Log Number:____________; Filter System:____________;
Time In Service:____________.

Type of Carbon (12.1.5) : Supplier Name:____________________________
Grade Designation:__________________________
Base Material:____________________________
Impregnant(s):____________________________
Lot Number:____________ Batch Number:__________

Particle Size Distribution (12.1.6, new carbon only) :

____________________	____________________
____________________	____________________
____________________	____________________
____________________	____________________
____________________	____________________

Apparent density (12.1.7) : Standard Volume:__________________cc
Weight of Carbon:__________________g
Density:_________________________g/cc

Moisture Content Determinations (12.1.12) :

<u>Initial Moisture Content</u> (ASTM D 2854 Oven Drying Method, t = 16 hrs.)

(1) Weight of weighing bottle alone . . . . .__________g
(2) Weight of weighing bottle plus subsample of carbon . . .__________g
(3) Weight of subsample of carbon (2) - (1) . . . . .__________g
(4) Weight of weighing bottle plus carbon after drying . . .__________g
(5) Weight of moisture removed (2) - (4) . . . . .__________g
(6) Initial Moisture Content, 100 (5)/(3) . . . . .__________%

<u>Moisture Absorbed During Test</u>

(7) Weight of weighing bottle alone . . . . .__________g
(8) Weight of weighing bottle plus sample of carbon . . . .__________g
(9) Weight of sample of carbon (8) - (7) . . . . .__________g
(10) Weight of weighing bottle alone . . . . .__________g

(11) Weight of weighing bottle plus carbon after test . . . .__________g
(12) Weight of sample of carbon after test (11) - (10) . . .__________g
3) Moisture Absorbed During Test, 100 [(12) - (9)]/(12) . .__________%

Final Moisture Content (see section 11.1 for calculations)
(14) Final Moisture Content, (6)(9)/(12) + (13) . . . . . . .__________%

Test Canister Information: Diameter, ±0.1 mm ________mm
Depth, ±1 mm ________mm
Volume, ±0.1 cc ________cc

Summary of Test Parameters (12.1.8-11)
Equilibration Period:

First 16 hours of equilibration:
Duration ________hrs., Monitoring Frequency ________/hr.

Parameter	Minimum	Maximum	Average	Std.Dev.
Temperature, °C				
Relative Humidity, %				
Absolute Pressure, kPa				
Actual Gas Flow, l/min.				

Last 2 hours of equilibration:
Duration ________hrs., Monitoring Frequency ________/hr.

Parameter	Minimum	Maximum	Average	Std.Dev.
Temperature, °C				
Relative Humidity, %				
Absolute Pressure, kPa				
Actual Gas Flow, l/min.				

Feed Period:
Duration ________min., Monitoring Frequency ________/hr.

Parameter	Minimum	Maximum	Average	Std.Dev.
Temperature, °C				
Relative Humidity, %				
Absolute Pressure, kPa				
Actual Gas Flow, l/min.				

Elution Period:
Duration ________min., Monitoring Frequency ________/hr.

Parameter	Minimum	Maximum	Average	Std.Dev.
Temperature, °C				
Relative Humidity, %				
Absolute Pressure, kPa				
Actual Gas Flow, l/min.				

Note: The actual gas flows indicated are at the test conditions, i.e. corrected for temperature, pressure, and water vapor. These values may be converted to superficial face velocity by dividing by the test bed cross-sectional area, and making the proper unit conversions.

Counting Information (12.1.14-15)

Type of Instrument ____________________
(If gamma spectrometer, give energy region used)

Type and Size of Detector ____________________

Results:

Test Bed, Total Counts.............____________________

Test Bed, Counting Time............____________________min.

Test Bed, Time Since Midnight.......____________________hours

1st Backup Bed, Total Counts........____________________

1st Backup Bed, Counting Time.......____________________min.

1st Backup Bed, Time Since Midnight.____________________hours

2nd Backup Bed, Total Counts........____________________

2nd Backup Bed, Counting Time.......____________________min.

2nd Backup Bed, Time Since Midnight.____________________hours

Background Test Bed, Total Counts...____________________

Background Test Bed, Counting Time..____________________min.

Background BU Beds, Total Counts....____________________

Background BU Beds, Counting Time...____________________min.

Penetration ± 1sd, Test Bed.........__________±__________%.

Efficiency ± 1sd, First Backup Bed..__________±__________%.

Note: When a gross gamma counter is used, there will be only a single figure to be entered in the above blanks for total counts. However, when a spectrometer is used, three numbers are required for each blank asking for total counts. The first number in each "count" blank is the total count obtained in the given energy interval. The second number, which will be subtracted from the first, is the background count expected in that interval from extrapolation of the Compton continuum. Both numbers are required for proper error propagation. The third number is the difference between the first and second, or the Compton-corrected net counts that will be used in the calculation of penetration. This method of reporting data from spectrometric measurements is shown in the following example.

Example of Counting Information

Type of Instrument ND-66 Gamma Ray Spectrometer, 360-370 keV
(If gamma spectrometer, give energy region used)

Type and Size of Detector Ge(Li), 75ml

Results:

Test Bed, Total Counts.............. 475626 - 14890= 460736
Test Bed, Counting Time............. 10 min.
Test Bed, Time Since Midnight....... 8.5 hours
1st Backup Bed, Total Counts........ 140685 - 8682 = 132003
1st Backup Bed, Counting Time....... 60 min.
1st Backup Bed, Time Since Midnight. 9.5 hours
2nd Backup Bed, Total Counts........ 3195 - 487 = 2708
2nd Backup Bed, Counting Time....... 120 min.
2nd Backup Bed, Time Since Midnight. 13 hours
Background Test Bed, Total Counts... 492 - 451 = 41
Background Test Bed, Counting Time.. 120 min.
Background BU Beds, Total Counts.... 485 - 492 = -7
Background BU Beds, Counting Time... 120 min.

In this example, the penetration of the test bed and its standard deviation obtained using the Compton-corrected values are 4.619 ±0.014%, and for the efficiency of the first backup bed are 98.969 ± 0.020%.

Annex A-2. Calculation of the Penetration of the Test Bed and Efficiency of the First Backup Bed and their Associated Uncertainties from Data Obtained Using a Gross Gamma Counter without Energy Discrimination.

Defining A, B, and C to be the net counting rates of the single test bed and two consecutive backup beds, respectively, at the same time and under identical counting conditions:

$$\text{Penetration, } P\% = 100\ (B+C)/(A+B+C);$$

$$\text{Efficiency, } E\% = 100\ A/(A+B+C) = 100 - P\%;$$

$$\text{Standard Deviation} = S_E\% = S_P\%$$

$$= 100\ \frac{\{[(B+C)^2(S_A)^2] + A^2[(S_B)^2+(S_C)^2]\}^{0.5}}{(A+B+C)^2}$$

where: $A = (X/t_x - BG1/t_{bg1})\exp(0.003592\ T_x);$

$B = (Y/t_y - BG2/t_{bg2})\exp(0.003592\ T_y);$

$C = (Z/t_z - BG2/t_{bg2})\exp(0.003592\ T_z);$

$$(S_A)^2 = [X/(t_x)^2 + BG1/(t_{bg1})^2] \exp(0.007184\ T_x);$$

$$(S_B)^2 = [Y/(t_y)^2 + BG2/(t_{bg2})^2] \exp(0.007184\ T_y);$$

$$(S_C)^2 = [Z/(t_z)^2 + BG2/(t_{bg2})^2] \exp(0.007184\ T_z);$$

and X, Y, Z, BG1, BG2, t_x, t_y, t_z, t_{bg1}, t_{bg2}, T_x, T_y, and T_z are the total counts, counting times, and decay times of the test bed, backup beds and backgrounds, respectively. The constant 0.003592 is the disintegration constant per hour for ^{131}I using a halflife of 8.041 days. The constant 0.007184 is the disintegration constant multiplied by two to square the exponential. "S" is the estimate of the standard deviation of a single measurement from counting statistics corrected for decay and background. For the distribution between the two backup beds, efficiency of the first backup bed in percent is 100 B/(B+C), and the standard deviation in percent is

$$100\ [C^2(S_B)^2 + B^2(S_C)^2]^{0.5}/(B + C)^2$$

Annex A-3. Calculation of the Penetration of the Test Bed and Efficiency of the First Backup Bed and their Associated Uncertainties from Data Obtained Using a Gamma Spectrometer with Energy Discrimination.

Using the same definitions given in Annex A-2, and defining X_c, Y_c and Z_c to be the Compton corrections for X, Y and Z, respectively, the following equations should be used to obtain the penetration of the test bed, the efficiency of the first backup bed and their respective uncertainties.

$$A = \{[(X-X_c)/t_x] - [(BG1-BG1_c)/t_{bg1}]\} \exp(0.003592\ T_x);$$

$$B = \{[(Y-Y_c)/t_y] - [(BG2-BG2_c)/t_{bg2}]\} \exp(0.003592\ T_y);$$

$$C = \{[(Z-Z_c)/t_z] - [(BG2-BG2_c)/t_{bg2}]\} \exp(0.003592\ T_z);$$

$$(S_A)^2 = \{[(X+X_c)/(t_x)^2]+[(BG1+BG1_c)/(t_{bg1})^2]\} \exp(0.007184\ T_x);$$

$$(S_B)^2 = \{[(Y+Y_c)/(t_y)^2]+[(BG2+BG2_c)/(t_{bg2})^2]\} \exp(0.007184\ T_y);$$

$$(S_C)^2 = \{[(Z+Z_c)/(t_z)^2]+[(BG2+BG2_c)/(t_{bg2})^2]\} \exp(0.007184\ T_z).$$

Note: All counts must be made under identical counting conditions or corrections must be made for the different conditions. The Compton correction must be estimated from the average of at least five channels each above and below the peak. The ^{131}I activity must be distributed homogeneously throughout the carbon and a standard height used. Used carbon might have a different background than the new carbon in the backup beds and should be determined separately. Both backgrounds must be constant. Air backgrounds are unnecessary unless the activity in the carbon is to be evaluated.

DISCUSSION

HOLUB: How did you measure humidity and did you make intercomparisons between various humidity meters?

SCARPELLINO: We used two dew-point hygrometers. One was calibrated within two weeks of our tests at the National Bureau of Standards. We also used a dew point condensing device for which the temperature is controlled by a monitored circulating water bath.

JACOX: I have a philosophic question. You made reference to the fact that the commercial labs were carrying out research functions rather than simply performing a standardized test. It appears to me that either I was very confused as to the initial intent of the INEL contract and round robin laboratory comparison, or else you have been doing a lot of research, too. It seems to me that the real problem is simply reproducibility. We agree that there are flaws, and, perhaps, overly loose tolerances in the ASTM method and that some labs are probably not able to meet strict tolerance requirements. Therefore, I am very curious why, instead of simply trying to find the systematic errors that are present and suggesting tighter tolerances to the ASTM, you are developing a new standard that seems to me to involve the same research effort you are decrying when it is performed by the commercial labs. This seems to me to be a contradiction.

SCARPELLINO: We are not a commercial lab and we are employed by the NRC to do some research work. I think there is a need for a standard test. I think we need to standardize the most critical test, the 30°C test, and then establish criteria based on it. If the laboratories have such a difficult time performing the 30°C test, who knows what happens at 80°C, and, especially, 130°C. The latter tests do not evaluate carbon satisfactorily from the research I have seen. I have done no testing at the several temperatures myself. However, some of the data suggest that the carbon performs better as you increase temperature. We want the final standard test procedure to place the greatest demands on the carbon. Using that test, it will be possible to set whatever criteria you want for your carbon.

JACOX: I agree with what you just said. I also think most people would agree that 30°C is the most critical test when conducted at 95% RH with methyl iodide. But why has there not been more effort to find the systematic errors that obviously exist under D-3803, rather than asking for additional test parameters related to loading, post-sweep, and pre-equilibration? All could be analyzed in an R&D mode, but at the moment, the industry needs to have everyone get the same answer on a single test. It seems we have just added more variables rather than trying to reduce them.

SCARPELLINO: We have only tightened restrictions on some of the variables. The major changes were changes in duration of feed and equilibration periods, which our parametric investigations and those of Deuber, in W. Germany, indicate should make no difference in the equilibrated test result. Whether you challenge for an hour or two hours or whether you equilibrate the sample for an hour or four hours, the measurable differences are insignificant. The changes are stated in the paper. Justifications for some reflect current practices within the commercial industry.

RIGBY: I have an observation and a question. For anyone interested, I have with me the figures of a roundrobin exercise conducted in the United Kingdom. I was very interested in your discussion about equilibrating used carbon at 95% RH before testing. I think it is a very, very good idea. I think it would also be very sensible to measure the level of moisture on the used carbon before equilibration, because the adsorption isotherms when water is on the carbon are a bit knotty anyway. I don't say it would show anything by the end of the day but I think it would be useful to know the initial level of moisture on the carbon before pre-equilibration.

SCARPELLINO: What you recommend would be a useful exercise. However, it is probably more important for research purposes than for routine analysis. I think the commercial labs will say that they simply will not have enough sample for the standard test if they take a subsample to measure the initial moisture content. That is not their fault, it is a fact of life.

RIGBY: Get them to get larger samples.

SCARPELLINO: Yes, I agree. It was put into our test method.

BELLAMY: One point I would like to make is that I do not agree that the technical specification should be based on the most critical test. I think the thing that we should all be concerned with here can be summed up in one word, reproducibility. I think the purpose of a test specification, in fact, the purpose of any test on used carbon, is to verify that, should we need that carbon to perform its intended function in the event of an accident, it will do so. Personally, I don't care whether the used carbon test is equilibrated or not equilibrated. I think it is more important that we get reproducibility into the system. So I guess I agree with what Mr. Jacox said.

The other point I want to make is that I think we have to be very careful about public perception. Some of Dr. First's comments this morning on Pilgrim are very appropriate because I have been at Pilgrim recently in front of some public meetings and, yes, we are getting to the point where we need those "flack jackets" regularly, believe me. If we come out now with a proposal to change the standard test and lower acceptance criteria for carbon, I think we will be in trouble because when you propose changing the standard test specification you are exposing yourself to a public hearing. I, for one, do not want to stand up in front of a public audience and say, "Yesterday we tested the carbon to 95% but today the acceptance criterion is only 90% or 80%." Even though I could explain technically why there was a change, and you could explain technically why there was a change, the people in the audience would have already made up their minds. My point is that I think reproducibility is the key to the solution.

SCARPELLINO: I am not sure it would require much of a technical explanation. I think the general public would be willing to except the fact that you made the test tougher. That is all you really need to say. The test is tougher, and our results are more reliable, so therefore we are going to change it. This does not mean any more iodine is ever going to go through the carbon. I know I wouldn't want to do it personally either.

Let me answer your first comment. I agree that a carbon test should be reproducible. However, we cannot sacrifice the ability to discriminate between good and bad carbon for the sake of reproducibility. If we were to set the standard carbon test conditions at 130°C, 95% RH, and challenge the carbon with elemental iodine, we would, undoubtedly, obtain interlaboratory reproducibility. Those tests would probably all result in penetration values of 0.01 to 0.1%. These results would also be good P.R. for the nuclear industry. In actuality, however, we would have lost the ability to detect poor carbons. A test which produces higher values of penetration can better discriminate between carbons and can reduce the uncertainties associated with determining whether a carbon is 99.0% efficient or 98.9% efficient, which often is the difference between acceptance and failure.

SCHOLTEN: I agree that we need a standard method for testing carbon. However, the actual conditions in a plant will differ from those of the standard test. Therefore, we have to transform the laboratory results to plant conditions. Has any parametric study been done, or planned, to make this transformation possible?

SCARPELLINO: Yes, INEL has performed some parametric investigations, and will, I hope, be able to continue and expand these investigations. I am aware of two other parametric studies. One published by Deuber and Gerlach in Nuclear Safety in 1985, which is cited in my paper, and another study by the Japanese, which was published much earlier.

NUCLEAR-GRADE, GAS-PHASE ADSORBENT IODINE RETENTION TEST

J.A. Slade and H.M. Philippi
Atomic Energy of Canada Limited
Chalk River Nuclear Laboratories
Chalk River, Ontario, Canada

and

M.J. Kabat
Toronto, Ontario Canada

Abstract

This paper describes the Nuclear-Grade Adsorbent Test Laboratory recently established by CRNL. Adsorbents are tested with various radioiodine species under closely controlled environmental conditions. This testing is required as part of the quality assurance testing program of the new LOCA emergency stand-by filter-adsorber installation, for the NRU Research Reactor. The primary feature of the laboratory is the use of aqueous sulphuric acid solutions to achieve and maintain 95% relative humidity. This test equipment has proven to be both simple to use and capable of producing consistent and repeatable test results.

I. Introduction

The Chalk River Nuclear Laboratories (CRNL) has recently established a Nuclear-Grade Adsorbent Test Laboratory. In this laboratory, the efficiency of adsorbents for the removal of various radioiodine species (CH_3I and I_2) can be evaluated under defined, closely controlled conditions.

The laboratory was set up as part of the quality assurance testing program for the new 8 $m^3.s^{-1}$ (16000 CFM) LOCA emergency stand-by filter-adsorber installation, built for the NRU Research Reactor. As well as providing exhaust air filtering for the NRU Reactor in the event of a LOCA emergency, the new filter installation will also be used for reactor operations during special experiments. Adsorbers had always been leak tested in-situ with I-131 tagged elemental iodine; however, it was always realized that the actual iodine retention characteristics of the carbon were not proven by this test. The routine replacement of adsorbent material practiced on small adsorber installations with less than 100 kg of carbon could not be considered economical for a system with more than 1600 kg of carbon. Therefore, laboratory testing of the carbon adsorbent, in addition to in-situ leak testing with methyl iodide-131, was considered necessary for economic reasons as well as assuring the carbon's performance over time.

In order to obtain representative carbon samples for laboratory analysis, bypass lines were installed in parallel with the carbon beds on six of the sixteen parallel filter trains.

Each bypass line contains one 50 mm deep adsorbent test bed canister. The bypass system is shown in Figure 1. A mock-up of the bypass system was tested in the laboratory to ensure leak tightness and proper face velocity through the canister. Measurements confirmed that the air flow through the canisters can be controlled to yield the design face velocity of 20 $cm.s^{-1}$, equal to that in the actual carbon adsorber. These representative adsorbent samples will then be removed at pre-determined intervals and the radioiodine adsorption performance determined over the anticipated life of the carbon.

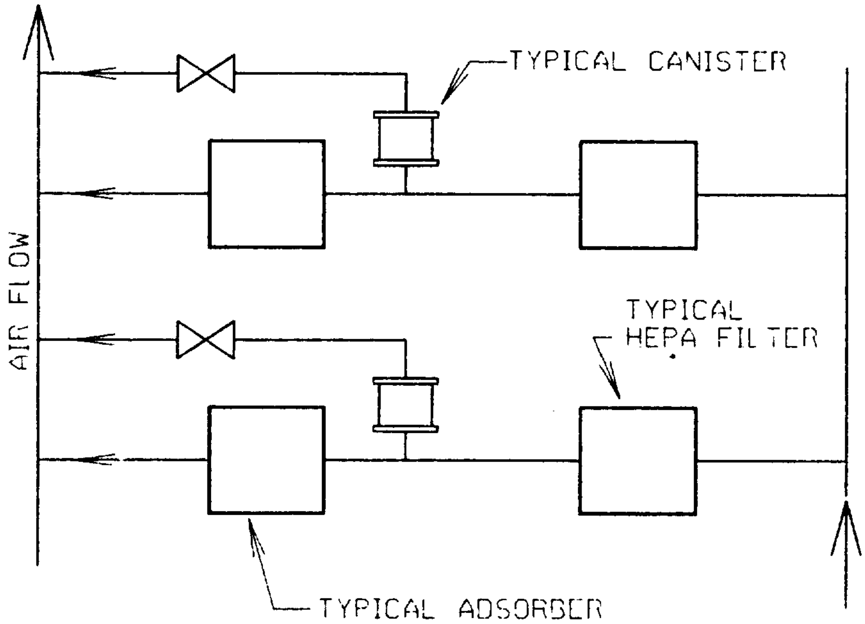


FIGURE 1
Bypass Canister System for 50 mm Deep Pleated Bed Adsorber Cell

II. Test Specifications

Test Principle

Carbon testing is performed in accordance with Test Method A, ASTM D3803, 1979, "Standard Test Methods for Radioiodine Testing of Nuclear Grade Gas Phase Adsorbents", and CSA N288.3.2 Standard, "High Efficiency Nuclear Air Cleaning Systems for Normal Operations".

A representative sample of adsorbent is equilibrated with air (equilibration period), at the design face velocity, to temperature and relative humidity conditions for 16 hours. The equilibrated adsorbent is then challenged with a radioiodine (CH_3I or I_2) and humid air mixture (feed period) for 120 minutes. Following the feed period the carbon sample is purged with the humid air (elution period) for 240 minutes. Radioiodine, retained in both the test and back-up bed adsorbent, is radiometrically evaluated and the iodine removal performance of the tested adsorbent is then calculated for the specified depth values.

Test Conditions

In accordance with CSA Standard N288.3.2, the environmental conditions for the testing of adsorbents are as follows:

Temperature	-	25 ± 1°C
Relative Humidity	-	95 ± 2%
Face Velocity	-	20 ± 1 $cm.s^{-1}$

Approximate adsorbate concentrations in the challenge gas stream are as follows:

CH_3I - 1.75 $mg.m^{-3}$
I_2 - 1.0 $mg.m^{-3}$

III. Test Equipment

A functional diagram of the test assembly is shown in Figure 2. The system is composed of the following functional components:

- Environmental chamber with temperature control
- Constant humidity control
- Adsorbate gas generator and injection line
- Adsorber column
- Challenge gas recirculation line
- Radiometric analysis system
- Computing unit.

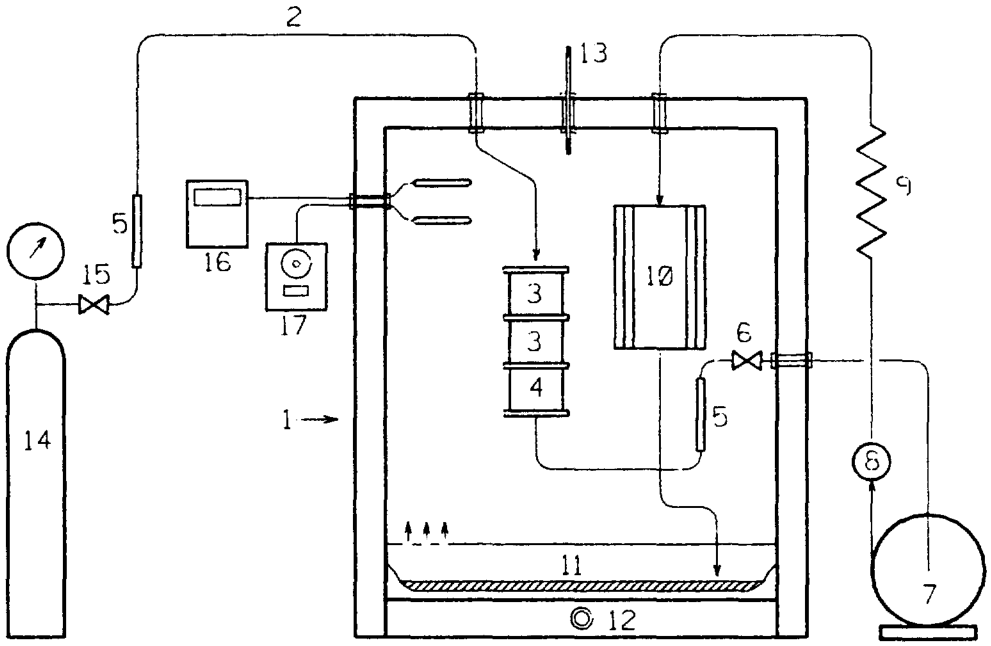
Environmental Chamber

The environmental chamber is a biological type incubator. The internal temperature of the test chamber is controlled, within 0.5°C of the temperature setpoint, with a direct acting thermostat.

Constant Humidity Control

The control of relative humidity within the test chamber is based on the standard method ASTM E 104-51 (1971), "Standard Recommended Practice for Maintaining Constant Relative Humidity by Means of Aqueous Solutions". This standard describes three possible solutions to control humidity; namely, sulphuric acid, glycerin and salt solutions. Sulphuric acid solution is the only suitable alternative for maintaining 95% RH, as both glycerin and salt solutions are not sufficiently stable and are messy. This method has two distinct advantages: 1) the resulting humidity level is not significantly affected by moderate fluctuations of temperature, and 2) any required relative humidity can be established and maintained simply by preparing the appropriate concentration of sulphuric acid for the tray in the chamber. It is estimated in the ASTM Standard E 104 that the relative humidity levels in the test chamber can be maintained constant within ± 0.1%, under proper environmental conditions.

The relative humidity over aqueous solutions of sulphuric acid are given in Table 1 (excerpt from ASTM E 104 Standard).



1. Environmental Test Chamber
2. Injection Line
3. Stainless Steel Test Bed Canisters
4. Stainless Steel Back-Up Bed Canisters
5. Flowmeter
6. Flow Control Valve
7. Oil-Less Air Pump
8. Line Filter
9. Air-Cooled Discharge Line
10. Heat Exchanger
11. Sulphuric Acid Bath
12. Temperature Controller
13. Thermometer
14. CH_3I Challenge Cylinder
15. CH_3I Feed Control Valve
16. Cole Parmer Digital Hygrometer
17. Honeywell Relative Humidity Readout Instrument

FIGURE 2
Carbon Test Assembly Functional Diagram

Table 1. Relative Humidity Over Sulphuric Acid Solutions.

Density (25°C) g.cm^{-3}	Relative Humidity (%) At 0°C	25°C	50°C	75°C
1.0300	98.4	98.5	98.5	98.6
1.0641	95.9	96.1	96.3	96.5
1.0997	92.4	92.9	93.4	93.8
1.1368	87.8	88.5	89.3	90.0

From this table it can be seen that, when the solution and air temperature in the chamber are the same, the equilibrium value of RH is not sensitive to temperature fluctuations at higher humidity values. By interpolation of the above data the density of the aqueous sulphuric acid solution required to maintain a relative humidity of 95% is 1.0765 g.mL^{-1}. Because about 2% of the water will evaporate from the solution, to equilibrate both the chamber air and the carbon to 95% RH, 2 L of solution with an initial density of 1.075 g.mL^{-1} is used.

The temperature and relative humidity of the air in the chamber are monitored by two independent temperature/relative humidity electronic measuring devices. These instruments are regularly checked to ensure they are reading correctly. The method of checking involves drawing a known volume of the humid air in the chamber through a freezing coil cooled with dry ice. Knowing the volume of air sampled, the mass of the water collected, atmospheric pressure, air stream temperature, and the relative humidity and temperature downstream of the freezing coil, the relative humidity in the chamber can be accurately calculated. The computer program and algorithms used for these calculations are shown in the appendix. The calculated value can then be compared to the measured value.

Air Circulation Loop

The air circulation system is a closed loop system. The air pump draws conditioned air (during equilibration and elution period) and a challenge gas (during feed period) from the environmental chamber, through the test and back-up adsorbent beds, and recirculates it back into the chamber.

The challenge gas is a mixture of conditioned air (25°C, 95% RH), with I-131 labeled I_2 vapor or CH_3I. This adsorbate gas is carried by nitrogen from the generating system through an injection line to the test bed inlet, mixed with air and passed through the test bed and back-up bed (which removes all radioiodine), rotameter, flow control valve and air pump. A line filter is installed at the pump outlet for particulate removal. Air temperature, which is increased due to the work done on the air by the pump, is reduced within the air cooled discharge line to the ambient temperature of the room. The air stream is then passed through a heat exchanger in the test chamber to equilibrate it again to 25°C. Finally, the air is discharged into the tray and passes over the surface of the H_2SO_4 solution, and is equilibrated to 95% RH before being returned to the chamber.

The flow capacity of the recirculation loop and air pump is such that two carbon tests can be performed simultaneously, simply by installing an additional test column, flow meter and rotameter in parallel with the existing set.

Test Bed Assembly

The test bed assembly consists of a set of 50 mm deep stainless steel canister beds manufactured to the ASTM D3803 standard specifications. The test rings are individually sealed with rubber O-rings, and held in the test position with bolted stainless steel flanges. This set allows the installation of up to three, 50 mm deep rings with tested charcoal in series. For the compliance testing of 50 mm deep adsorber beds and new carbon, only one 50 mm deep test bed canister and one back-up bed canister are used in series.

Adsorbate Gas Generator and Injection Line

Either CH_3I or I_2 vapor can be used in the challenge gas stream. The methods of generation of these two species are as follows:

CH_3I tagged with I-131 is produced by an isotopic exchange process using stable CH_3I and aqueous, NaI-131 in NaOH/0.02M Na_2So_4 solution. The generation method is designed to yield 1 µCi* of I-131 per mg of stable CH_3I. The I-131 labeled CH_3I is then placed in a 2.25 L stainless steel cylinder and pressurized with N_2. The N_2/CH_3I adsorbate gas is then metered to the inlet of the test column to yield a mass concentration of 1.75 $mg.m^{-3}$ in the challenge humid air stream.

Elemental iodine, I_2, is generated in the environmental chamber, close to the test column inlet, in order to minimize its loss due to surface deposition. The I_2 vapor, labeled with I-131, is generated at a controlled rate and purged from a slightly acidic solution of elemental I-127 and NaI-131 with N_2.

Radiometric Analysis System

The radioiodine counting system is located in a low background area. The system consists of a Ge(Li) spectrometric detector in a shielded castle and a multichannel analyzer with automatic background subtraction. For this application, a counting geometry which keeps detector dead time loss below 10% is chosen.

Computer Unit

Penetration, statistical accuracy valves, and calculations to check the accuracy of the humidity reading instruments are performed with a microcomputer and printed with an attached printer.

Technical Specifications of Test Equipment

The range and accuracy of the operational parameters, their control and monitoring and the test requirements from the CSA N288.3.2 standard are shown in Table 2.

Table 2. Technical Specifications of Test Equipment.

	Temp. (°C)	RH (%)	Test Flow ($L.min^{-1}$)**
Control Range	25 - 65	40 - 98	15 - 50
Monitoring Range	0 - 100	0 - 100	2 - 50
Control of System Parameters	25 ± 0.5	95 ± 1.9	24.3 ± 1.0***
Monitoring of System Parameters	25 ± 0.25	95 ± 1.0	24.3 ± 0.75
CSA Standard Requirements	25 ± 1.0	95 ± 2.0	24.3 ± 1.2

---

* 1 µCi = 37 GBq

** Standard Pressure

*** Corresponds to a face velocity of 20 ± 1 $cm.s^{-1}$

IV. Operational Experience

Not a great deal of carbon testing experience has been gained to date due to the short time the laboratory has been fully operational. The test results obtained thus far, however, are encouraging. Also the problems overcome and the experience gained while achieving and maintaining a 95% RH level may be interesting to someone considering this method for this, or similar applications.

Constant Humidity Control

The use of aqueous sulphuric acid solutions to control relative humidity at 95% has been found to be both simple and accurate. Once the relative humidity level in the chamber reaches the equilibrium value, as determined by the sulphuric acid concentration, it remains very stable for the duration of the test. Figure 3 shows a typical response curve of chamber relative humidity versus time, with time zero representing the moment the air circulation pump is switched on. From this curve it can be seen that approximately 80 minutes are required for the chamber to reach 95% RH. This lag period may be viewed as a disadvantage due to the additional time added to an already lengthy procedure. Experience has shown, however, that the time required for the chamber to reach test relative humidity conditions can be included in the 16 hour equilibration period without compromising the test. This is verified in Figure 4, which shows temperature differential across the test and back-up carbon beds (measured by thermocouples placed in the inlet and outlet airstreams of the canister assembly) versus time. From this figure it is seen that even with the initial lag in relative humidity the adsorption of water on the carbon reaches equilibrium with the 95% RH airstream in 6-7 hours.

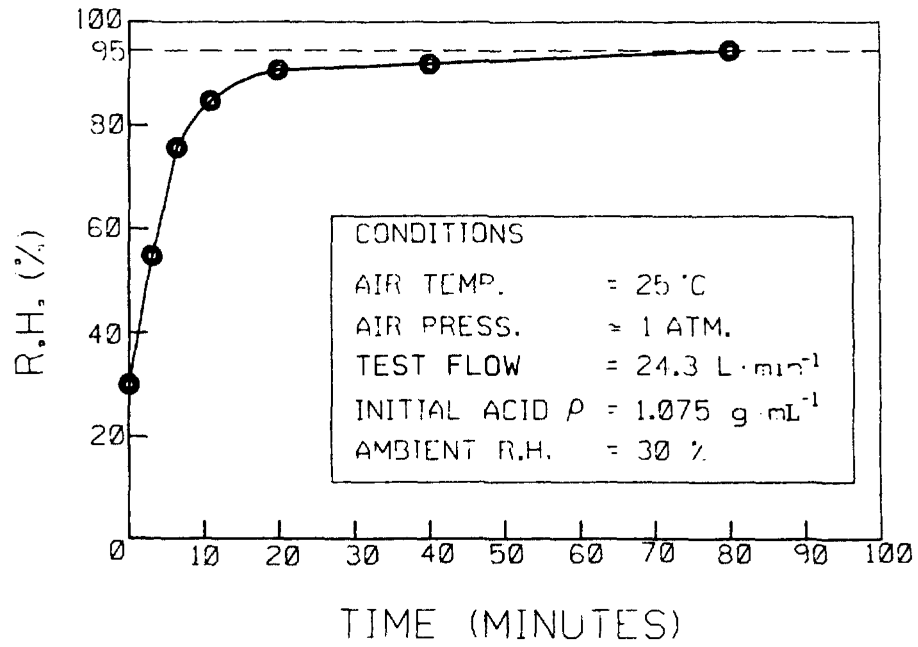


FIGURE 3
Typical Response Curve of Chamber Relative Humidity Versus Time

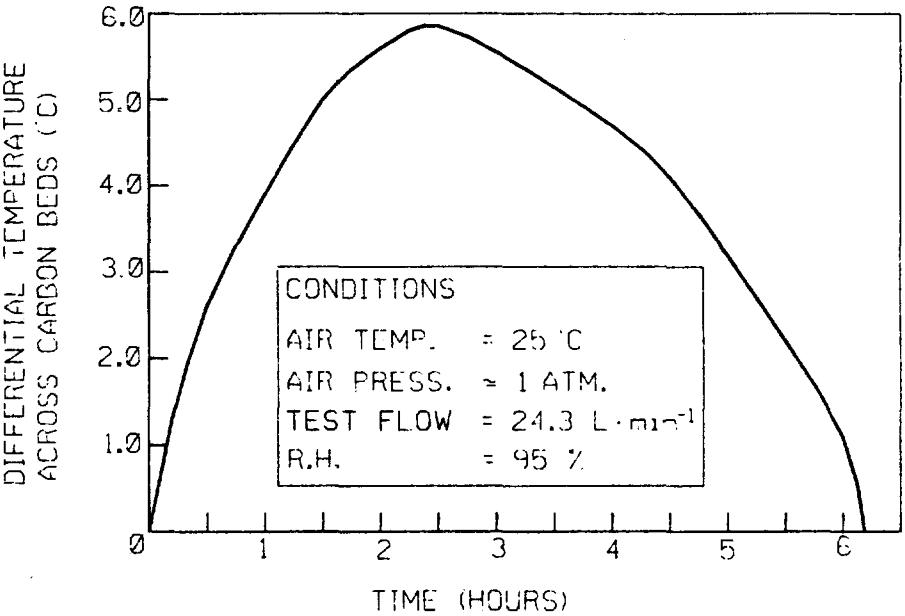


FIGURE 4
Typical Time Required for Adsorption of Water on Carbon to Reach Equilibrium with 25°C, 95% RH Airstream

A potentially serious problem with this method of humidity control is condensation within the chamber. Even a small amount of condensation will result in a loss of humidity control. The air temperature in the room in which the chamber is kept must be carefully controlled, and drafts in the room must be minimized to avoid condensation problems. To maintain temperature control, room temperature was initially kept at 20-22°C, to provide an ample temperature gradient for the dissipation of pump heat in the air cooled discharge line. At these temperatures condensation was forming on the walls and heat exchanger in the test chamber. This problem was completely solved by adding some additional insulation between the inner glass door and outer insulated door of the test chamber, and raising the room temperature. The room temperature now cycles between 22.5-24°C. This temperature range appears optimum to maintain temperature control and avoid condensation problems.

It is also important to keep the environmental chamber essentially airtight to avoid moisture loss. This is simply accomplished by sealing any through holes in the chamber with plasticine and ensuring that the inner glass door is adequately sealed.

Experience also has shown that careful attention must be paid to the temperature of the acid solution. Acid solutions of the proper density are normally prepared in advance and stored in the laboratory. The solutions of course achieve temperature equilibrium with the air (approximately 23°C). It was found that if the acid solution was not warmed to test temperature prior to starting the test, the temperature of the solution would take an unacceptably long time to reach test temperature in the chamber, and 95% RH would not be reached. This is attributed to inefficient heat transfer between the solution and chamber air and the heat loss through evaporation from the solution. To accommodate the heating of the solution a variable speed pump is used to circulate the acid solution external to the chamber through a glass coil immersed in a warm water bath. Circulation rate and bath temperature are adjusted to maintain bath temperature at 25°C. The acid solution is circulated for the first 20 minutes of the equilibration period, or when the relative humidity in the chamber reaches about 90%. The pump has also proved useful for loading and unloading the sulphuric acid solution into and out of the tray.

Carbon Testing

As mentioned earlier, only a limited amount of carbon testing experience has been gained to date, all for new 5% TEDA impregnated, activated, coconut based carbon. Test results are presented in Table 3. The carbon samples tested were from the same manufacturer and the same production lot.

Table 3. Results of Laboratory Testing of Carbon Adsorbent with CH_3I-131 in Air at 25°C and 95% RH*.

Sample No.	Retention and Accuracy (%)
1**	97.23 ± 0.05
2	98.42 ± 0.04
3	98.88 ± 0.04
4	98.31 ± 0.06
5	99.16 ± 0.04
6	98.92 ± 0.02
7	99.31 ± 0.02
Carbon Manufacturer's rating	99.00 minimum

---

* Tests performed in accordance with ASTM standard D3803-Test Bed Depth = 50 mm.

** Conditions for this test were 25°C, 100% RH.

The retention results range from 97.23% to 99.31%. Sample No. 1 was rejected because the relative humidity in this test reached 100% due to a condensation problem in the chamber. Measures described previously were taken to eliminate this problem in subsequent tests. The actual range of retention results are thus 98.31% to 99.31%. The mean value of the retention and its precision is 98.82% ± 0.34%. The accuracy and repeatability of the results are considered very good and are in agreement with the carbon manufacturer's rating. The calculated accuracy (0.34%) is the result of random errors caused by slight differences in many variables, including: the mass of carbon in the test samples, the mass of CH_3I introduced, the amount of TEDA.

impregnate on the carbon, and the relative humidity and temperature of the chamber air. Counting errors make only a minor contribution (0.04% avg.) to the above accuracy value.

V. Conclusions

Even with the limited testing experience and data gained thus far, the following conclusions and comments can be made on the testing equipment in use at CRNL.

- The method using aqueous solutions of sulphuric acid to control relative humidity has proved to be quite simple and capable of maintaining 95% RH in the environmental chamber for the duration of the test.

- The test assembly is simple in design with a limited amount of required instrumentation and hardware, and replacement costs of the components are low.

- When the procedures outlined in ASTM Standard D3803, Method A, are closely followed (only deviation is 25°C test temperature in place of 30°C) the test equipment is capable of producing repeatable results with good accuracy.

VI. Acknowledgements

The authors would like to thank Mr. Brian Latouf (summer student) for writing the software to perform the relative humidity calculations, and Mr. Wayne Keuhl for his valuable contributions to the commissioning of the laboratory and for performing the actual carbon testing.

Appendix

Programmed Psychrometric Calculation

This program provides the psychrometric calculation required for the calibration of the humidity meter used in the adsorbent iodine retention test. All moist air property algorithms are from ASHRAE Handbook of Fundamentals, 1981, Chapter 5.

Program Operation

The program is written in BASIC for a TRS-80 Radio Shack pocket computer. It has approximately 1.5 kilobytes of memory, an attached printer and a cassette storage device. To use the programs simultaneously they must both be loaded onto the computer's current memory banks (see TRS-80 Manual for loading procedure). Definition mode "Shift A" will start the program. All input is manual and echo printed. The "Enter" bar must be hit after each input to keep the program running.

Program Nomenclature

The following variables are used in the program. The TRS-80 pocket computer has a memory limitation of 26 elements; thus, a definition of each variable is needed.

A: Mass Flow Rate of Air in Kilograms Per Minute
D: The Air Density in Kilograms Per Cubic Metres
E: The Mass of Water in the "Freeze-Out" Air in Kilograms
G: The Relative Humidity of the Air Accurate to Two Decimal Points
I: The Relative Humidity of the Dry Air After the "Freeze-Out" in Percent
K: The Relative Humidity of Air, Unrounded
L: New Total Partial Water Pressure
M: Time of Flow in Minutes
O: Accuracy to Goal of the Rounded-Off Relative Humidity
P: Atmospheric Pressure in Kilopascals
Q: The Water Mass in Kilograms
R: The Humidity Ratio of the Air in kg Water/kg Air
S: The Saturation Pressure of the Air in Pascals
T: The Dry-Bulb Temperature in Celsius
U: The Humidity Goal for Error Comparison
V: The Air Flow Rate in Cubic Centimetres Per Minute
W: The Water Mass "Freeze-Out" in Grams
X: Intermediate Calculation for the Saturation Pressure
Y: Intermediate Calculation for the Saturation Pressure
Z: The Dry-Bulb Temperature in Kelvin

Psychrometric Calculation Program List

```
 10  REM EXPERIMENTAL ANALYSIS TO CALCULATE RELATIVE HUMIDITY
 20  REM DEFINE THE PROGRAM AS "A"
 30  "A"
 40  REM INPUT AND ECHO PRINT ALL OF THE VARIABLES NEEDED
```

```
 50   INPUT "TEMPERATURE?(CEL)";Z
 60   PRINT "TEMP(CEL)=";Z
 70   INPUT "ATMOS.PRESS?(KPa)";P
 80   PRINT "PRESS(KPa)=";P
 90   INPUT "WATER MASS?(GRAMS)";W
100   PRINT "WATER(GRAMS)=";W
110   INPUT "AIRFLOW?(CC/MIN)";V
120   PRINT "FLOW(CC/MIN)=";V
130   INPUT "TIME FOR FLOW?(MIN)";M
140   PRINT "TIME?(MIN)=";M
150   INPUT "R.H.DRY AIR?(%)";I
160   PRINT "R.H.DRY(%)=";I
170   INPUT "HUMIDITY GOAL?(%)";U
180   PRINT "GOAL(%)=";U
190   REM CALL UPON THE SATURATION PRESSURE SUBROUTINE
200   GOSUB 800
210   REM CALCULATE THE AIR DENSITY USING IDEAL GAS EQN.
220   D=P/((8.3144/28.97)*T)
230   REM CONVERT WATER MASS IN GRAMS TO KILOGRAMS
240   Q=W/1000
250   REM FIND THE MASS FLOW RATE OF THE AIR IN KG/MIN
260   A=V/100^3*D
270   REM CALCULATE THE AMOUNT OF WATER IN THE CAPTURED DRY AIR
280   E=I/100*S*.62198/(P*1000-S)*A*M
280   REM ADD THE CAPTURE DRY AIR WATER VAPOR TO THE WATER COLLECTED
300   REM AND CALCULATE THE HUMIDITY RATIO
310   R=(Q+E)/(A*M)
320   REM FIND THE NEW PARTIAL WATER PRESSURE
330   L=P/(1+.62198/R)
340   REM FIND THE ACTUAL RELATIVE HUMIDITY FROM FREEZE OUT
350   K=L*1000/S*100
360   REM ROUND OFF BOTH THE R.H. AND FIND ITS % OFF FROM GOAL
370   O=(INT(ABS(U-K)*100))/100
380   G=(INT(K*100))/100
390   REM PRINT OUT THE RESULTS AND PROVIDE A RERUN ALTERNATIVE
400   BEEP3:  PRINT "R.HUM=";G;"%+-";O;"%"
410   INPUT "START OVER?(Y/N)";X$
420   IF X$ = "Y" THEN 50
430   INPUT "CONTINUE?(Y/N)";G$
450   END
800   REM SUBROUTINE FOR CALCULATING SATURATION PRESSURE
810   REM FOR A GIVEN TEMPERATURE
820   T = Z + 273.15
830   X = (-5800.2206/T) + 1.3914993 - (.048640239*T)
840   Y = X + (.4176478 E - 4) * T^2 - (.14452093 E - 7)*T^3 +
6.5459673*LN(T)
850   S = EXP(Y)
860   RETURN
```

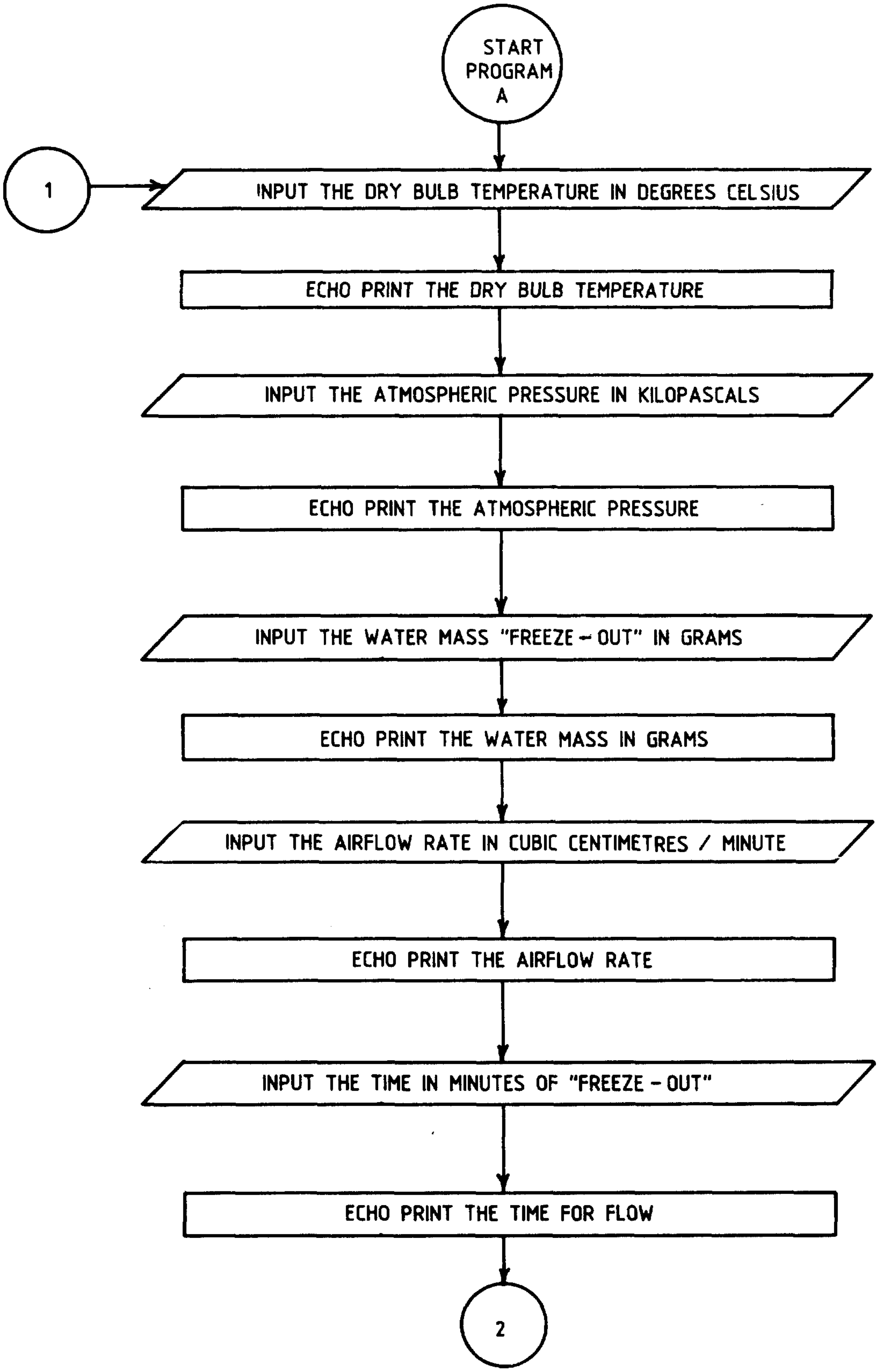


Figure A1. Program Flowsheet

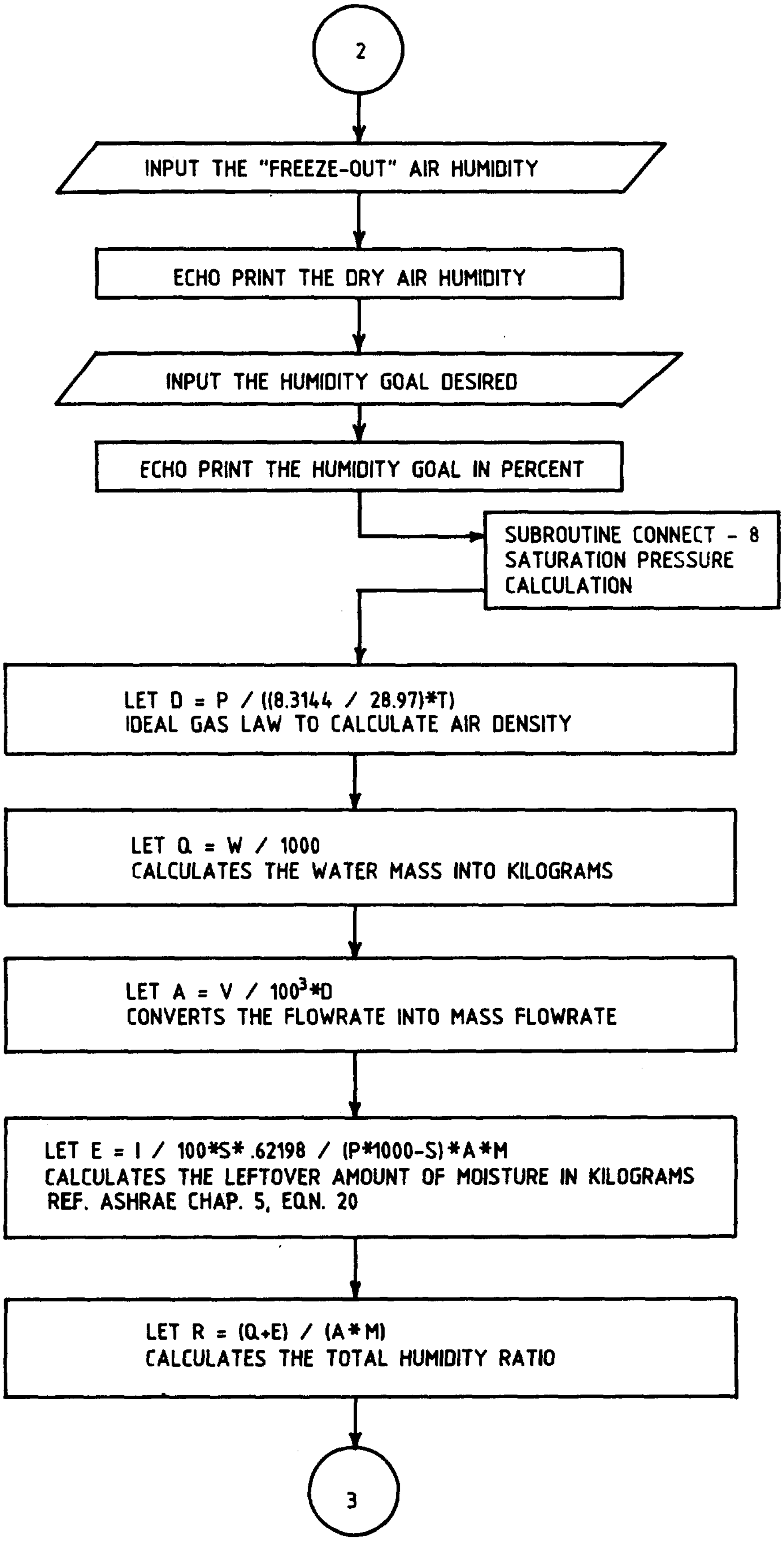


Figure Al Con't. Program Flowsheet

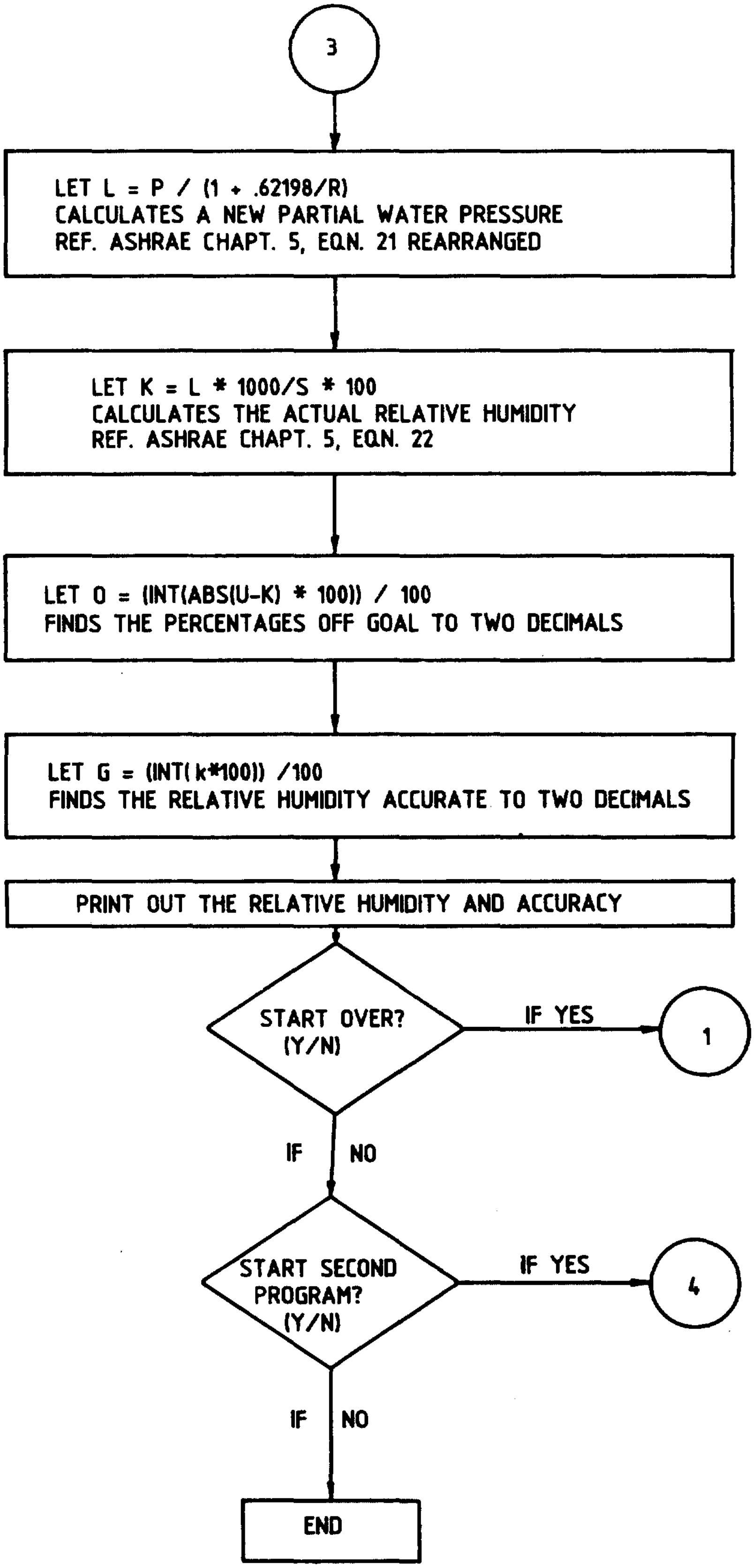


Figure A1 Con't. Program Flowsheet

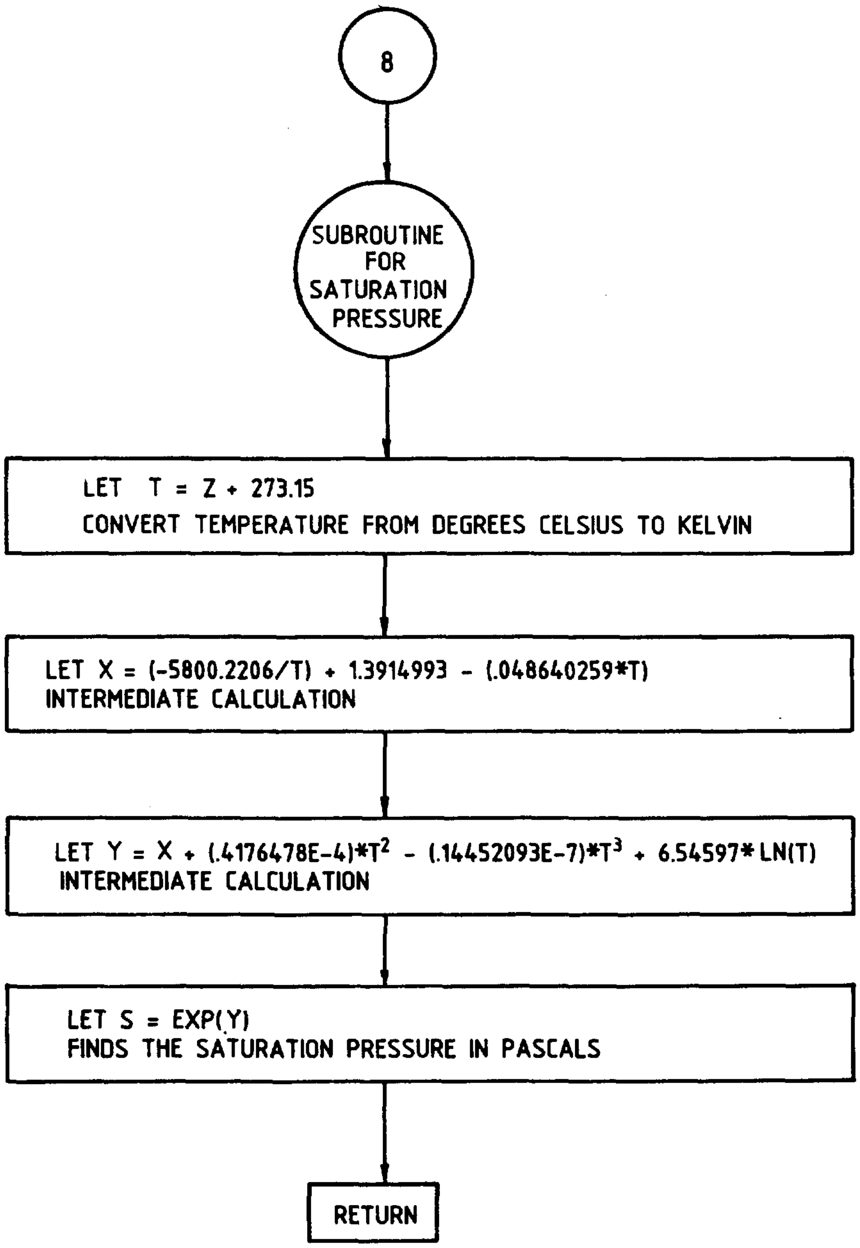


Figure A1 Con't. Program Flowsheet

THE U. V. SPECTROMETRIC METHOD OF ANALYSING CHEMICAL IMPREGNANTS OF A NUCLEAR GRADE ACTIVE CARBON AND ITS APPLICATIONS FOR MANAGING THE CARBON FILTER AT A NUCLEAR REGULATORY LAB. AND PLANTS.

You Sun Kim
Korea Advanced Energy Research Institute,
P.O.Box 7, Daeduk Danji, Chungnam, Korea.

Abstract

A simple and convenient method of analysing chemical impregnants of a nuclear grade active carbon (i.e. TEDA, TEDA + KI, and KI_x) is described by means of ultraviolet spectrometry. The absorption maxima of each impregnant (TEDA, KI, and I_2) were determined in acetonitrile solvent and absorbance was plotted against concentration to afford a good linearity, which could well be applied as an analytical curve. Special considerations were exerted to confirm the influence of the coexistent element or compound to be analyzed such as TEDA in case of TEDA + KI and iodine in case of KI_x. The method was found useful to be practiced at a regulatory lab. or a nuclear power plant to identify or monitor the quality of an impregnated active carbon. Some typical examples of applying this analytical method at the regulatory lab. in this country (This lab.) were shown. The deterioration of TEDA in the used sample collected at the power plant of this country was observed by checking the variation of λ max against the concentation of acetonitrile extract of the sample and this data are referred to monitor the quality of the carbon in practical use. The replacing sample of the carbon sample in use at the domestic power plant was collected before installation in the filter system and the sample was analyzed for its impregnants. This data are the basic standard to follow the carbon during the course of active use at the power plant. This method could also be applied for monitoring the impregnation pattern of chemical species impregnated in a base active carbon sample for a research purpose. In this laboratory, the adsorption efficiency of both used and new carbon samples collected at the domestic power plants were tested for the regulatory purposes. In order to assure the testing results, samples were analyzed for their chemical compositions in advance of testing. Examples of the analysis and testing results were described. It is concluded that the method is of very simple and practical, which may open various applications besides described hereto for a nuclear regulatory lab. or power plants for managing their carbon filter systemes.

1. Introduction

In Korea, 4 PWRs and one CANDU power plants are now under routine operation and another 2 PWRs will be in routine operation within a year. Additional 4 or 10 nuclear power plants are going to be constructed by the end of this century. These power plants are operated by the Korean Electric Power Co., and this institute, as the National Laboratory, has been concerned with regulatory and R & D works to support the safe operation of these power plants.

Impregnants of a nuclear grade active carbon for power plants or hot facilities are generally known as KI + I_2, TEDA, TEDA + KI, and tertiary amine + iodine salts. The amount of the impregnants applied for a nuclear grade active carbon is usually indicated by the manufacturer on its technical data grossly without showing any definite analytical data. It was, therefore, necessary to confirm the definite quantity of chemical impregnants contained in the commercial samples in active use at the power plant to assure their quality. Since new carbon samples are sometimes non uniformly impregnated, which may influence the adsorption efficiency of that sample(1), the analysis of chemical impregnants of a nuclear grade active carbon was considered to be essential to guarantee the adsorption quality of the product. Furthermore, the chemical impregnants in a nuclear grade active carbon are liable to be deteriorated during the period of routine operation. According to recent literatures, iodate was formed in the deteriorated sample which decrease the adsorption efficiency(9) and hydroxide of TEDA, which may be formed by the deterioration of TEDA under a serious moisture, showed less adsorption efficiency than those of the corresponding iodide and TEDA itself(2). These findings warrant to trace chemical composition of the nuclear grade active carbon during the period of its routine use to assure the quality of chemical status of the impregnants, if it is possible to be traced by means of a simple and convenient analytical method.

A simple and convenient method of analysing chemical impregnants of an activated carbon was, therefore, developed by this laboratory. Microquantities of iodine dissolved in 5% aqueous potassium iodide could be analyzed by means of spectrometric method at the ultraviolet range(3), but the method of analysing chemical impregnants of a nuclear grade carbon sample had not been reported in literatures. The absorption maxima for each impregnant was determined in acetonitrile solvent, which was the best suitable solvent in the present cases, by ultraviolet spectrometric method and the influence of coexistent elements or compounds in the acetonitrile extract of a sample carbon was closely studied. The absorbance at λ max. of each impregnate was plotted against concentration to afford a good linearity which may well be applied as an analytical curve. Samples collected from domestic power plants were analysed by this method to monitor the quality of the samples in a chemical sense. The method was found useful to be practiced at a regulatory lab. or nuclear power plant. In this paper, details of the analytical procedure were presented and the feasibility of this method was discussed with special regards to qualify the chemical impregnated active carbon at a nuclear power plant.

During the course of this investigation, interlaboratory comparison of adsorption efficiencies of IC samples sponsored by US NRC had been conducted and this lab. participated to this comparison works. Results obtained in analysing IC samples and their absorption efficiencies were also reported in this paper to review the correlation between chemical uniformity of IC samples and their absorption efficiencies, if there are any. Some research results obtained during the period of impregnating base active carbon with TEDA, TEDA + KI, and KI_x were reported to introduce the use of this analytical method in this field.

2. Experimental Procedure

2.1 Moisture Determination of the Sample

In order to determine the moisture content of sample, ASTM Method D 2867, oven drying method #5 for determining moisture content of a nuclear grade active carbon, was adopted by use of a forced circulated drying oven. The results were summarized in table 12.

2.2 Extraction of the Sample

In a 200 ml., Erlenmeyer flask, a definite amount of the sample carbon weighed nearest 0.001g, 100 ml. of acetonitrile (ACS(GR) Grade, Tedia Co. USA) and a magnetic strring bar were placed and the flask was stoppered by means of a tappered glass joint, and it was stirred magnetically at a definite temperature and time. The mixture was filtered and the filtrate was reserved for further analysis. The filtered carbon was dried in a forced circulated oven and it was reserved for further testing as described in the later paragraph.

2.3 TLC Testing of the Acetonitrile Extract of the Sample

The acetonitrile extract of the sample was examined by means of a TLC for checking its major chemical compositions and purity as follows.

Plate : DC-Pastikfolien, Kiselgel 60 F_{245}, MERCK Art 5735.
Developing Temperature : 24-25°C
Solvent Front : 10.0cm.

A. Developing Solvent : 95% Ethyl Alcohol

The TLC studied by means of 95% ethyl alcohol as the developing solvent showed R_f values as follows.

R_f Values / Chemical Species	R_f Values *Coloring Agent	Spots	**Coloring Agent	Spots
TEDA	(1)	0.00	(2)	0.00
TEDA + KI	(1)	0.00, 0.75(Faint)	(2)	0.00, 0.75
KI_x	(1)	0.75(Faint)	(2)	0.75

* Coloring Agent : 0.2g. of ninhydrin were dissolved in the mixture of 95ml. of n-butanol and 5ml. of 10% aq. solution of acetic acid.
Coloring Method : The sprayed plate was heated for 20 minutes at 105°C.
** Coloring agent : 2.0% alcoholic solution of silver nitrate.
Coloring Method : The sprayed plate was heated to a dryness and it was exposed under sunlight.

B. Developing Solvent : 1-propanol + 33% ammonia water (4:1 V/V)

The TLC studied by means of the predescribed solvent showed R_f values as follows.

2.4 Ultraviolet Spectrometric Analysis of the Sample

A. Instrument

The following instrument and conditions were adopted through the entire experiments of the present study.

(1) Instrument : Safas, 170 D/DF, Monaco.
(2) Absorption Cell : 1cm width, Rectangular Cell.
(3) Conditions : Lamp Source ; H-Lamp
Slit Width ; 10cm. nm
Scan Speed ; 40nm/MN.
Chart Speed ; 25nm/MN.
Enr ; x1

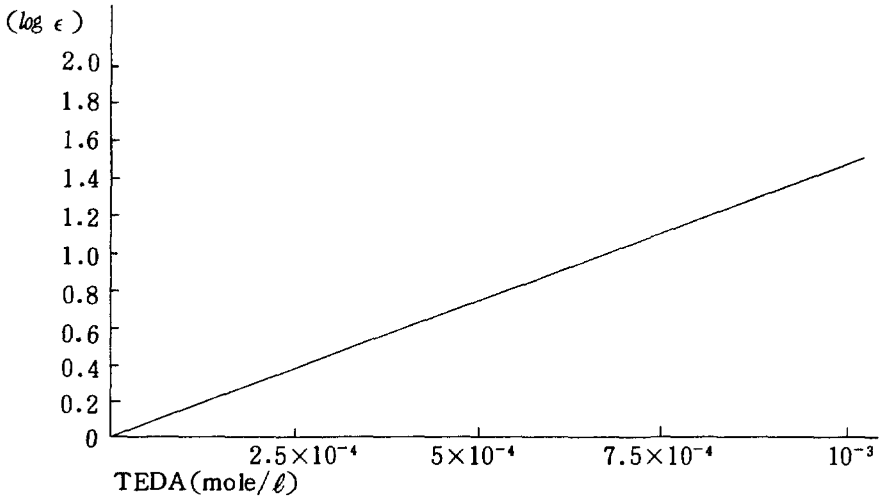
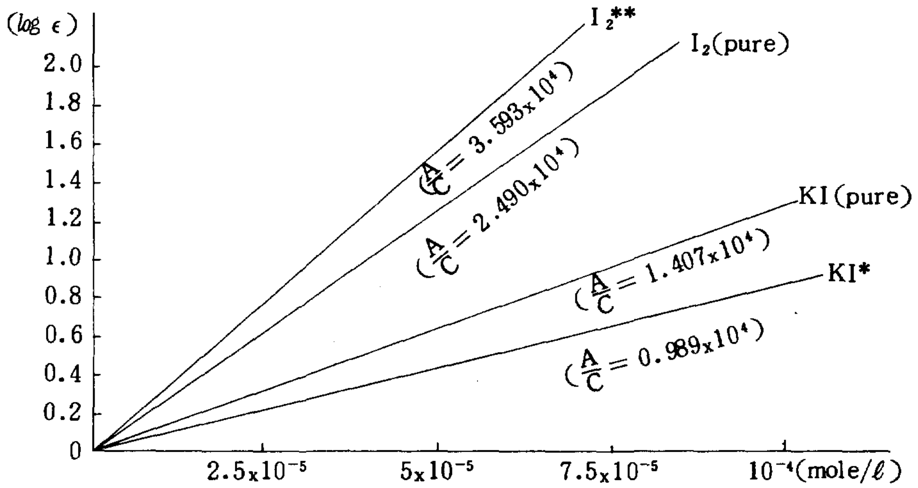


Fig.1. U.V. Spectrometric Calibration Curve of TEDA in Acetonitrile.



* In presence of I_2. ** In presence of KI.

Fig.2. U.V. Spectrometric Calibration Curve of KI_x in Acetonitrile.

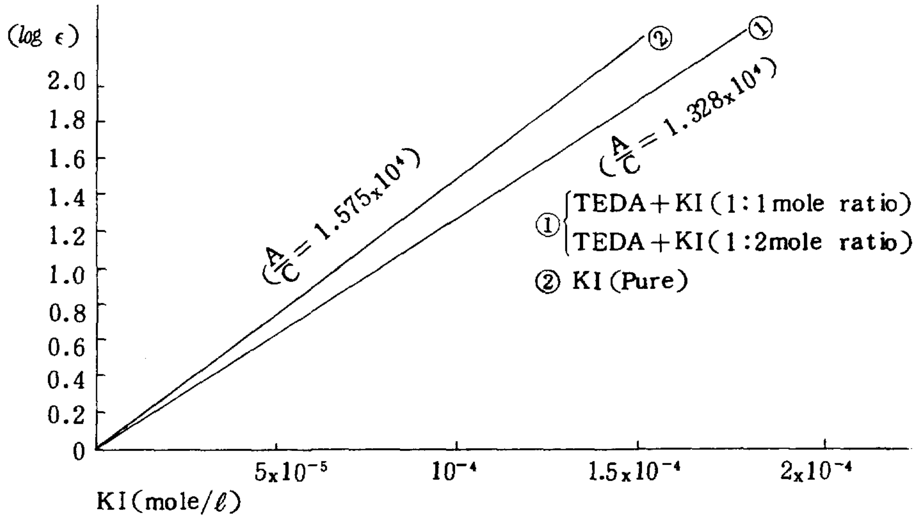


Fig.3. U.V. Spectrometric Calibration Curve of TEDA + KI in Acetonitrile.

R_f Values / Chemical Species	R_f Values			
	*Coloring Agent	Spots	**Coloring Agent	Spots
TEDA	(1)	7.0	(2)	7.0
TEDA + KI	(1)	7.0, 6.5(Faint)	(2)	5.5, 7.0
KI_x	(1)	6.5(Faint)	(2)	5.5

* Coloring agent and method were the same as above (1).
** Coloring agent and method were the same as above (2).

B. Measurement of λmax. of Each Component in Presence of Coexistend Element or Compounds

The acetonitrie extract of the sample was examined for its λmax. at ultraviolet region. The results were summarized in the table 1. The chemical compositions of the extract were identified by means of TLC testing as described in 2.3. The measured λ max. was confirmed as compared to those of authentic samples of each system in acetonitrile solvent.

Table 1. Absorption Maxima of the Major Components of the Acetonitrile Extract of the Sample.

Impregnant's System	λ max.	ε max.
	nm	
KI	256	1360
	302	990
TEDA	237	1426
TEDA + KI	*256	1080

* The λ max. of TEDA could not be observed due partly to the side peaks of KI at 230-240nm.

C. Plotting the Curve of Absorbance v.s. Concentration

In order to confirm the influence of coexistent element or compound on the spectrometric absorption pattern of a system of the acetonitrile extract of the sample, authentic sample solutions of TEDA, KI, and I_2 in acetonitrile were mixed in various molar ratio and the absorbance were plotted against concentrations as shown in figures 1, 2, and 3, respectively.

D. Typical Examples of Analysing the Sample

(1) KI_x Impregnated Sample

Sample Source	:	Kori No.1 Plant (PWR, Replacing sample in 1985)
Extraction	:	0.504g./100ml. of acetonitrile, 5 hrs. at 22-25°C. and left overnight at that temperature.
TLC Data	:	KI and I_2 were identified.
Trade Name	:	5 KI_3, 208 C

KI_3 % : 8.62
I_2 (Free) % : 0.15

(2) TEDA + KI Impregnated Sample

Sample Source : Kori No.2 Plant (PWR, 16 June, 1984)
Trade Name :
Extraction : 0.250g, 100ml. of acetonitrile, 5 hrs. at 20-22°C and left overnight at that temperature.
TLC Data : TEDA and KI were identified.
U.V. Data : λ max. 256nm (No distinct peak was identified at 230-240nm.)
KI % : 1.45%
TEDA % : Since there observed no distinct absorption maxima at 235-240nm region due to the side peaks of iodide, TEDA % was measured as follows : The observed KI molar concentration was half divided to result the reacted TEDA molar concentration and excess amount of KI solution was added dropwise to the sample solution until the absorbance of the solution was corresponded to the concentration of KI on the calibration curve. This value of excess adding KI molar concentration was half divided to result unreacted TEDA molar concentration. The total molar concentrations of reacted and unreacted TEDA were converted to weight % to give 1.45% TEDA (%) in this case.

2.5 Checking the Chemical Status of TEDA Impregnated Active Carbon in Active Use

In order to trace the chemical status of TEDA impregnated active carbon, samples used for a year were collected from KORI POWER PLANT (PWR) and checked their U.V. λ max. variation depending on their concentrations in actonirile solvent. It was confirmed that variation of λ max. was observed in cases of hydrophilic solvents such as methanol, ethanol, and water, whereas there are none variation in case of n-hexane and acetonitrile solvents, if the compound is pure enough. The results were summarized in the table 2. In cases of the samples collected at KORI POWER PLANT a considerable variation of λ max. were observed depending on the dilution factors as shown in the table 3.

Table 2. UV Absorption Maxima of Authentic Sample of TEDA in Various Solvents

Solvent / λ max nm / Sample	MeOH	95% EtOH	n-Hexane	Acetonitrile	H_2O	Remarks
					(Conc. mol/ℓ)	
TEDA	247	240	240	237	240(10^{-2})	
	245	236			236(6.7×10^{-2})	
	243	233			234(5×10^{-3})	
	242	232			288(2.5×10^{-3})	
	239	225			220(1.25×10^{-3})	
	233				216(6.25×10^{-4})	
	229				215(5×10^{-4})	
	223				210(2.5×10^{-4})	
	222					

Table 3. Variation of λ max. Against the Concentration of Acetonitrile Extract of the Active Carbon Used at the Kori Power Plant for a Year.

Source of Sample	Initial λ max.	Variation of λ max.	
	nm	Dilution Factor	nm
Kori Plants Sample			
# 1.	232	2	216
# 2.	231	2	220
# 3.	230	2	223
#4.	228	3	215

2.6 Examination of Samples Impregnated for Research Use

In order to confirm the feasibility of the present method of analysing chemical impregnants and to examine the impregnation pattern of the sample prepared for research use, base carbon (Physical Properties; ASTM D 2862,3802,3467,2854, 2866,3466,2867, and 3838, respectively.) was impregnated in aqueous solution of TEDA, TEDA + KI, and KI + I_2, respectively. The detailed profile of the impregnation process will be reported elsewhere, but the analytical process of checking the impregnation pattern was only reported here as follows.

A. TEDA Impregnation

A definite quantity of base carbon was placed in a 200ml. Erlenmeyer flask and a definite amount of TEDA aqueous solution was mixed with carbon by stirring it for a time and at a temperature. The resulting mixture was filtered and the filtered carbon was dried by menas of a drying oven. An aliquot of carbon was extracted as shown in 2.2, and the extract was analyzed for its chemical composition as shown in 2.4. The results were summarized in the table 4. Emphasis was laid on checking the effective extraction time.

Table 4. Impregnated Active Carbon in Aqueous Solution of TEDA.

*Extracted Time (Hrs.)	TEDA Content in the Sample Analysed (%)	**Recovery (%)
2	4.67	93.40
3	4.83	96.60
4	4.97	99.40
5	4.94	98.80

* 0.5g level of the carbon in 100ml. of acetonitrile was used for extraction. Extraction Temp. : 20-22°C.

** The weight increase of impregnated carbon after being dried was compared to the original weight of the base carbon. The weight increase was averaged to 5.00% in these cases.

B. TEDA and KI Impregnation

The impregnation process and analytical procedure were the same as above. Emphasis was laid on the correlation between the amount of carbon extracted and recovery rate. The results were summarized in the table 5.

Table 5. Impregnated Active Carbon in Aqueous Solution of TEDA and KI.

Exp. No.	Weight Increase of Impregnated Carbon. (%)	*Extracted Carbon g./100ml. of Acetonitrile	Analytical Value KI (%)	TEDA (%)	Recovery %
1	12.29	1.001	4.91	4.98	80.47
2	1.40	0.503	0.58	0.59	83.57
3	6.15	0 253	2.70	2.73	88.29

* Extracted for 5 hrs. and left overnight at 22-25°C.

C. KI and I_2 Impregnation

The impregnation process and analytical procedure were the same as stated above. Emphasis was laid on checking the effective extracted temperature and confirming the correlation between the amount of carbon extracted and recovery rate. The results were summarized in the table 6.

2.7 Analytical Study of IC Samples and Testings

IC samples distributed by EG & G Idaho Inc. were analyzed for their chemical impregnants according to the procedure described as above. In these cases, the moisture content of each sample was measured as described in 2.1 and the analytical values were calibrated for moisture contents. The results were summarized in tables 7 and 8.

Table 6. Impregnated Active Carbon in Aqueous Solution of KI and I_2.

Exp. No.	Weight Increase of Impregnated Carbon. (%)	*Extracted Carbon g./100ml. of Acetonitrile	Analytical Value KI(%)	I_2(%) (Free)	**I_2(%) (Bound)	Recovery %
1	10.65	1.003	0.78	0.07	1.19	37.93
2	2.88	0.505	0.67	0.57	1.02	78.47***
3	2.64	0.503	0.83	0.54	1.26	99.62

* Extracted for 5 hrs. and left overnight at 20-22°C.
** Calculated on basis of the molecular formula, KI_3. Detailed was explained in discussion.
*** Extraction temp. was accidently above 25°C.

Table 7. Analytical Data of Impregnants Contained in IC Samples. (New Cabon).

Sample No.	Iodide* (wt%)	TEDA (wt%)	Remarks
N3-14	2.75	2.78	
N3-15	2.30	2.33	
N3-16	1.76	1.78	
N3-17	1.93	1.95	
N3-18	2.29	2.32	
N3-19	2.03	1.99	

* λ max. 256nm (KI).

Table 8. Analytical Data of Impregnants Contained in Samples. (Used Carbon)

Sample No.	Iodide (wt.%)*	Iodine (wt.%)**	Indicated Testing Method	Remarks
U7-1	0.243	1.083	30°C RH 95%	
U7-4	0.188	1.113		
U7-7	0.216	0.885		
U7-10	0.206	0.897		
U7-13	0.205	0.893		
U7-2	0.246	1.042	30°C RH 90%	
U7-5	0.189	1.425		
U7-8	0.194	0.914		
U7-11	0.208	0.988		
U7-14	0.213	9.830		
U7-3	0.226	0.959	30°C RH 70%	
U7-6	0.200	0.662		
U7-9	0.201	0.951		
U7-12	0.209	1.000		
U7-15	0.196	0.995		

* λ max. 256nm (KI)

** λ max. 302nm (I_2)

The standard deviation of analytical values were calculated and the uniformity of samples were evaluated as summarized in the table 9.

Table 9. Evaluation of the Uniformity of Impregnants in IC Samples.

Indicated Testing Methodes	Iodide Mean ± Std. Dev. %	Iodine Mean ± Std. Dev. %	TEAD Mean ± Std. Dev. %
A. Used Carbon*			
RH 70%, 30°C	0.202 ± 0.005(2.6%)	0.902 ± 0.13(14.4%)	
RH 90%, 30°C	0.201 ± 0.010(5.0%)	1.04 ± 0.23(24.0%)	
RH 95%, 30°C	0.204 ± 0.014(6.8%)	0.947 ± 0.10(10.2%)	
B. New Carbon			
RH 95%, 30°C	2.16 ± 0.66(25.3%)		2.19 ± 33(15.1%)

* U7-1, U7-2 and U7-3 were not included in this evaluation because of their deviated values. (ref. table 8.)

According to the testing protocol of EG & G Idaho Inc., the new and used carbon samples were tested for their adsorption efficiencies as summarized in tables 10 and 11.

Table 10. A Study on Penetration* of Radio-methyl Iodide through Activated Carbon (New Carbon).

Sample No.	Relative Humidity, %	Penetration Test Bed, %	Test Bed Density g/cm^3
N3-14	95	5.36	0.5060
N3-15	95	8.79	0.5021
N3-16	95	11.14	0.4882
Mean ± Exp. Std. Dev.		8.43 ± 2.38(28%)	0.499 ± 0.008(2%)

* Equilibration time was set as one hour for study purpose.

3. Results and Discussion

3.1 Moisture Determination

In table 12. the results of determining moisture of IC samples were summarized for reference. According to these results, moistures in new carbon samples were usually below than 3.0%, which may not be significant enough to effect the analytical results, since there are some more moisture adsorption during the course of sample handling process. However, in case of used carbon, the moisture contents were usually around 20% level, which may have a serious influence on the analytical results, though there may also be some moisture adsorption during the course of sample handling process. These points were seriously dicussed in the last NRC workshop for interlaboratory comparison of

Table 11. Interlaboratory Comparison Penetration of Radio-methyl Iodide through Activated Carbon (Used Carbon).

Sample No.	Relative Humidity, %	Penetration Test Bed, %	Test Bed Density g/cm^3
U7-1	95	24.18	0.5749
U7-4	95	*(27.70)	0.5690
U7-10	95	24.34	0.5737
Mean ± Exp. Std. Dev.		24.26 ± 0.08(0.3%)	0.573 ± 0.002(1%)
U-2	90	25.00	0.5629
U7-5	90	*(27.55)	0.5677
U7-11	90	23.22	0.5671
Mean ± Exp. Std. Dev.		24.11 ± 0.9(4%)	0.566 ± 0.002(1%)
U7-3	70	*(18.00)	0.5682
U7-6	70	24.10	0.5601
U7-12	70	22.87	0.5634
Mean ± Exp. Std. Dev.		23.49 ± 0.6(3%)	0.564 ± 0.003(1%)

* Deviated values were partly due to high or low contents of impregnants contained in samples (ref. tables 8 and 9.)

adsorption efficiencies, which showed a significant difference depending the initial moisture content of the IC sample. The adopted method of drying to measure the moisture content was ASTM-Method D 2867, in which an effective forced circulated oven was recommended for drying instrument. Some local excess drying seemed to cause some volatilization of impregnant, if an effective drying oven is not available or if there are some deviations on drying temperature. It is recommended that preliminary investigation on the moisture content of used sample may be necessary in case of analysing used sample providing an effective forced circulated oven, whereas the moisture determination process may not be essential to analyse the new sample.

Table 12. Moisture Content of IC samples.

Sample	Moisture Content (%)	Sample	Moisture Content (%)
* U-1	19.60	U-10	19.30
U-2	19.46	U-11	19.18
U-3	19.44	U-12	20.09
U-4	19.79	U-13	19.42
U-5	19.31	U-14	19.46
U-6	19.51	U-15	20.22
U-7	19.84	* N-14	2.85
U-8	19.73	N-15	2.88
U-9	19.15	N-16	2.86

* U : Used samples
N : New samples

3.2 Extraction Process

In extraction process, the choice of an effective solvent for extraction was very important to be considered. 10 solvents including water are known to be useful which are transparent in near ultraviolet region.(i.e. 200-360nm) Preliminary investigation to check the solubility of TEDA, TEDA + KI, and KI + I_2 showed that acetonitrile was the most useful for the present study. Water and alcoholic solvents showed better solubility, but these solvents could not reveal the constant λ max, which were varied depending on the dilution factor as shown in table 2 for TEDA case. Next problemes were extraction time and temperature. In case of TEDA, the extractibility was excellent after being stirred for 5 hrs. at 20-22°C, as shown in Table 4. In a TEDA + KI system, the extraction was greatly promoted when the concentration of base carbon was decreased to 0.25g/ 100ml acetonitrile level as shown in Table 5. In a KI + I_2 system, extraction temperature should be kept at a constant level as shown in Table 6. Detailed discussion on extraction time and temperature would better be described in the later paragraph (3. 4.) in conjunction with the chemical behaviors of these systems in the acetonitrile solvent.

3.3 TLC Study

Basically, TLC study on the acetonitrile extract of the sample to be analysed may not be necessary to identify its major chemical composition, if the specification of the sample is clear enough to indicate the composition. However, maker's specifications are sometimes not distinct enough to identify the composition. U.V. spectrometric examination of absorption maxima of the extract may also be useful for this purpose, but there are unclear case such as TEA + KI, TEDA absorption maxima of which could not be identified due to the side peaks of KI. TLC study was, therefore, conducted to confirm the composition in advance of analysis process. As summarized in 2.3, TEDA and KI could clearly be identified. It is recommended that both developing solvents listed in 2.3. would better be adopted to check spots closely. TLC study may also be very useful to check impurity caused by the deterioration of the sample, if any. There may be some other effective instrumental methodes of identifying the composition such as I.R., N.M.R., and others, but TLC was very simple and convenient to be used in an ordinary laboratory practice.

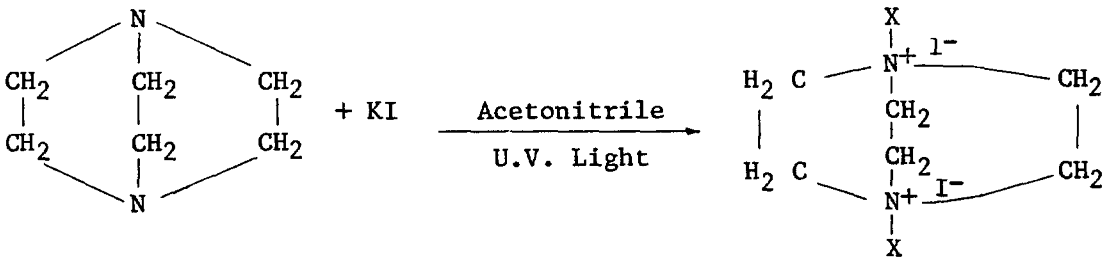
3.4 U.V. Spectrometric Analysis of the Sample

A. TEDA in the Sample

As summarized in table 1. and Fig.1, TEDA in the acetonitrile extract revealed the clear λ max at 237nm and the plot of λ max v.s. concentration showed a good linearity, which was useful as the analytical calibration curve. The extractibility of TEDA impregnated in a carbon was excellent and the recovery rate in the extracted acetonitrile solvent was close to the saturation, when the extraction mixture was stirred for 5 minutes at 20-22°C as summarized in the table 4. However, TEDA in the extract showed a variation of λmax depending on the dilution factor, if TEDA in the sample is deteriorted chemically as indicated in table 3. It is, therefore, recommended that the preliminary examination of the variation of λmax should be conducted in advance of the analytical process. If there are some variations of λmax depending on the dilution factor, the TEDA in the sample had already been deteriorated and hence it should be replaced as discussed in the later paragraph. Judging from the data of table 4, analytical accuracy of this method was 98-99%. The standard deviation of the analytical data was less than 2.0%.

B. TEDA and KI in the Sample

It was observed that the gradient of the curve of KI in this system was varied depending on the concentration of the coexistent TEDA in the system. Thus, gradients of curves of KI were colse together and they were lower than that of the pure KI, when the molar ratio of TEDA and KI adjusted to 1:1 and 1:2, as shown in figure 3. When the molar ratio of TEDA and KI was adjusted to 1:3 or more, gradients of curves were close to that of the pure KI. When the molar ratio was adjusted to 2:1 or more, gradients of curves were colse to that of the curve of the molar ratio of 1:1 and not varied much. On the other hand the λ max of TEDA in this system could not be observed at 237nm region, though there are some side peaks near at this region. These observations suggested that there may be the formation of a compound of TEDA and KI in acetonitrile under U.V. irradiation. It had been known for quite long time that alkyl iodies(5) or iodine(6) could form a salt quantitatively in a polar solvent with TEDA. These salts were prepared by the author to confirm their λmax in acetonitrile solvent, which showed a single λ max without showing TEDA peak at 237nm region. This finding led the author to expect



......(1)

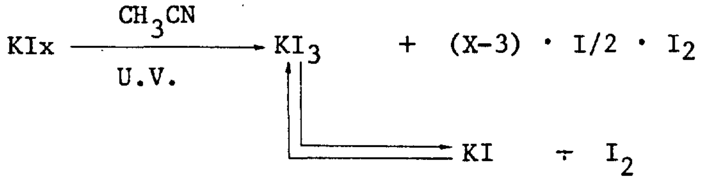
the above compound in the system under U.V. irradiation. For the chemical structure of X in the above chemical formula, further extensive investigations may be necessary to identify it, but this assumption did not incur a serious analytical error as discussed in the following.

On basis of the above observations, the analytical procedure was developed as described in 2.4.D. Thus, the amount of KI in this system was analyzed on basis of the calibration curve of KI of the molar ratio of 1:1 in fig.3. For TEDA, the amount contained in the system was classified in two category such as reacted and unreacted TEDA with KI under U.V. irradiation. The reacted TEDA was calculated by dividing in a half of the molarity of KI analyzed. The unreacted TEDA was analyzed by adding the excess amount of KI solution of the known concentration until the final molar ratio of the TEDA and KI became close to 1:2 as described in 2.4.D. To confirm the feasibility of this method, the impregnated carbon sample was examined and the recovery rate could be amounted to 88.29% at present. This value will, however, be able to be promoted by improving the extraction process.

This method may be said to be too complicate and the analytical accuracy is not high at present for TEDA content, but it can show the KI amount accurately. For used carbon, TEDA may be liable to be evaporated or deteriorated chemically during the course of routine operation. If the amount of KI analyzed by this method is unreasonably high as compared to that of the original sample unused, the evaporation or deterioration of TEDA may be very serious in the sample. Further investigation along this line is now under progressing. In conclusion, this method is of very complicate to check the amount of TEDA, but it may still be worthwhile to be practiced in regulatory lab. to confirm the quality of the carbon sample.

C. KI and I_2 in the Sample

As shown in fig.3, the gradient of the curve of KI in presence of I_2 was lower than that of pure KI solution, whereas it was higher in case of I_2. In order to confirm the influence of the coexistent I_2 or KI on the gradient of the curves, the molar ratio of KI and I_2 in the sample solution was adjusted to 1:1, 1:2, and 2:1. In cases of the molar ratio of 1:1 and 1:2 or more, the curves of KI and I_2 are close to the curves shown in fig.2 as the analytical curves. In case of the molar ratio of 2:1 or more, the curves of KI and I_2 were close to those of pure KI and I_2. These observations led the author to postulate the following sequence of chemical reactions in the analytical solution under U.V. irradiation. Thus, KI_3 and I_2 may be formed initially and the resulting KI_3 may further be decomposed to KI and I_2 depending on the analytical condition. Since anhydrous acetonitrile was used and temperature was kept below than 25°C, the decomposition of KI_3 in the analytical system may be not serious enough to liberate the significant quantity of I_2 in the analytical solution.



$$\begin{tikzcd} KIx \arrow[r, "CH_3CN", "U.V."'] & KI_3 \arrow[d, shift left] & + (X-3) \cdot I/2 \cdot I_2 \\ & KI + I_2 \arrow[u, shift left] & \end{tikzcd}$$

Further extensive studies on these aspects of chemical reactions may be necessary, but the analytical procedure was developed on basis of this postulation at present as described in 2.4. The measured concentration of KI in the sample solution may indicate the presence of the equivalent amount of I_2 bound to KI and that of I_2 may also indicate the presence of free iodine present in the system. To confirm this approach of analysis, impregnated carbon with the mixed aq. solution of KI and I_2 were examined as summarized in the table 6 and 99.62% of recovery was obtained, when 0.503g of the sample carbon was extracted in 100ml. of acetonitrile at 20-22°C. The extraction temperature was very important in this case. If the temperature is higher than 25°C, lower recovery rate was observed. The method was of simple and convenient to be practiced in an ordinary laboratory. According to the recent literature,(7) impregnation on charcoal that contain KI_x as part of the formula could have the following reaction and liberated iodine may further be volatilized during the stage of the routine purge of the carbon filter system by moist air.

$$K^+ I_3^- + H_2O \longrightarrow K^+ I^- + I_2 \cdot H_2O$$

When this analytical method is applied to examine the compositions of this type of partly deteriorated carbon, the measured KI value may indicate unreasonably higher value than that of the original sample of this carbon unused, since the coexistent I_2 and bound I_2 are partly volatilized to increase the gradient of the calibration curve shown in the fig.2. Research along this line may result an efficient way of checking the deterioration pattern of this type of nuclear grade carbon in active use.

3.5 Checking the Chemical Status of TEDA Impregnated

The samples collected at Kori No.1 plant, which had been used for a year and had been experienced some water floggings in the filter system, were dried and examined for their compositions as summarized in table 3. The λ max. observed was lower than that of pure TEDA in acetonitrile solution. The sample solutions were diluted and the diluted solutions were examined again to indicate lower λ max than those of the previous cases. In parallel with these observations, λ max of TEDA in various solvents were examined to select the best solvent suitable for the analysis and it was found that hydrophilic solvents showed a solvent effect varying λ max, whereas acetonitrile solvent gave a definite λ max at 237nm. in acetonitrile solvent, when it was well dried. These observations suggested that TEDA in Kori No.1 samples deteriorated and hence their U.V. λ max were varied depending on dilution factor. Presumably, TEDA deteriorated to its hydroxide or equivalent and this caused the observed λ max variation. It was considered that the examination of TEDA impregnated carbon by this method may be one of the effective method to check the chemical status of the carbon in active use.

3.6 Examination of Samples Impregnated for Research Use

Examination of samples inpregnated in the present study was mainly focussed to confirm the feasibility of the present analytical method as discussed in 3.4. Detailed features of the impregnation processes will be reported elsewhere. It can be said that the present study of examining the chemical composition of an impregnated carbon was very useful to trace the impregnation pattern, which could indicate the optimum condition for preparing a carbon sample of the desired specification.

3.7 Analytical Study of IC Samples and Testings

Since new carbon samples are sometimes non uniformly impregnated(7) due to the inadequate mixing process or deterioration during the course of storage, IC samples received from EG & G were analysed for their impregnants. The results were summarized in table 7. For used carbon samples, the same analytical study was conducted and results were summarized in table 8. These analytical data were further evaluated to confirm their uniformity of impregnants contained.

As shown in table 9. used carbon samples were generally uniform in regards to their impregnants of iodide, which may be main active composition for adsorption of radio-methyl iodide, except several cases of U7-1, U7-2, and U7-3. Experimental standard deviations (relative) for groups of samples classified by the testing conditions were 2.6%, 5.0%, and 6.8%, respectively. For new carbon samples, standard deviations (relative) of each active component of IC samples were amounted to 25.3% for iodide and 15.1% for tertiary amine (TEDA), respectively. Being aware of these analytical data, this laboratory conducted testing works as summarized in tables 10 and 11. Testing works were conducted according to the Standard Method, D-3803-79 and IC testing protocol of INEL. Exceptionally deviated penetrations were observed in cases of U7-3 and U7-5, which may possibly be due to the high or low contents of the impregnant in the sample as compared with the rest of samples in the group. It was, therefore, considered to be deviated testing data and hence they were neglected of the evaluation of the reproductibility. Since experimental standard deviations (relative) of impregnants contents of new carbon samples as summarized in table 7 were too high to be penetration tested, it was condidered worthwhile to study the relative error propagation of penetrations on basis of data in table 9. Results shown in table 10 had indicated that there seemed to exist a correlation between the impregnant's contents and penetrations. (i.e. exp. standard deviation (relative) for penetration, 28% and those for impregnants, 25% and 15%, respectively) Further experimental data would better be accumulated to confirm this relation, but it may be said at present that the preliminary analytical study on the chemical impregnant of a testing sample would be worthwhile to be conducted to assure the testing result itself.

4. Conclusion

The present study on analysing chemical impregnants of a nuclear grade active carbon could result a simple and convenient method to be practiced at an ordinary laboratory by means of ultraviolet spectrometric method. This method may be applied to trace chemical behavior of a carbon sample in active use. The method is also applicable to check the impregnation pattern of a nuclear grade carbon during the course of impregnation process, which will be able to check the optimum condition of preparing a sample of the desired specification. The postulations described in discussion may further be investigated to confirm their detailed insides, but the analytical accuracies were very high except that of TEDA and KI, which was around 88% at present. It is expected that some more applications for managing the carbon filter may be possible in the near future.

Acknowledgement

The author express his sincere appreciation for U.V. spectrometric analytical works conducted by Mrs. S.W. Kim throughout this study.

References

1. Deitz, V.R. and Joans, L.A., "Catalytical Trapping of Methylradioiodide by Bed of Impregnated Charcoal.", Radioisotopes. Vol.37, 59 (1978).

2. Freeman, W.P., Mohacsi, T.G., and Kovach, J.L., "Evaluation of Quaternary Ammonium Halides for Removal of Methyl Iodide from Flowing Air Streams.", Proc. 18th Air Cleaning Confr. Vol.1, 93-97 (1985).

3. Custer, J.J. and Natelso. S., "Spectrophotometric Determination of Microquantities of Iodine.", Anal. Chem. 8 1005 (1949).

4. EG & G Idaho Inc., "NRC Workshop.", Vol.1, 2, Annapolis, Maryland., USA., June 6-7, 1985.

5. Oae, S., et al., "Quarternary Ammonium Salts of 1,4-diazobicyclo-(2,2,2,) Octane.", J. Org. Chem. 24 1348 (1959).

6. Halpern, A.M. and Weiss, K., "Intramolecular Perturbation Effects in Diamine-Iodine Charge Transfer Complexes.", J. Am. Chem. Soc. 90 6297 (1968).

7. Romans, J.B. and Deitz, V.R., "A Non-radioactive Determination of the Penetration of Methyl Iodide through Impregnated Charcoals during Dosing and Purging.", 15th DOE Nuclear Air Cleaning Confr. 1 313 (1978).

8. Kim, Y.S. and Kim, S.W., "Study on Adsorption Characteristics of Filtering Materials.", Annual Research Report in 1985, KAERI/RR-464/85, Korea Advanced Energy Research Institute, Korea.

9. Deitz, V.R., "Regeneration of the Iodine Isotope-Exchange Efficiency for Nuclear-Grade Activated Corbons.", Proc. 18th DOE Nuclear Airborne Waste Management and Air Cleaning Confr., Vol.1, 33 (1985).

REMOVAL OF ORGANIC IODINE COMPOUNDS FROM FLOWING AIR STREAMS

W.P. Freeman, M.P. King and J.L. Kovach
Nuclear Consulting Services, Inc.

Organic radioiodides other than methyl iodide have been identified by several investigators in the past. However, only limited parametric studies have been performed to evaluate the challenge of these organic compounds toward the currently installed adsorbent beds. Initial evaluation of adsorbents has indicated that the removal properties of commercial adsorbents is different for organic compounds other than methyl iodide. Tests were performed using di-iodomethane, iodoethane, iodopropane and iodo benzene.

Adsorbents evaluated included stable iodine alone, amine alone and coimpregnated carbons. The results indicated that there are significant differences (i.e., lower removal efficiencies) for these compounds compared to methyl iodide and also for the individual species depending on the impregnant.

An explanation for these differences is offered based on isotope exchange rates for the various species and the fact that one or several of these species may be responsible for the "penetrating iodine species" suspected to be present by others in the past.

I. Introduction

Current official source term calculations assume that gas phase elemental iodine and organic iodide exist in addition to the solid (particulate) phase cesium iodide in reactor containment gases. The origin of organic iodides under reactor accident conditions has not been clearly established. However, without exception, the organic form of iodine presumed by all agencies is represented by methyl iodide. This also applies to adsorbents used to control radioiodine where removal efficiency is established using methyl iodide as the most penetrating species of organic iodide. This is contrary to results reported by others (1)(2) where a more penetrating form of (organic) iodide was suspected but not identified. While it is well established that in the case of a nuclear accident, fission product iodine is released mainly as cesium iodide (3)(4), various other chemical mechanisms are possible, including reactions at surfaces, reactions in the aqueous phase, reactions in the gas phase and radiation induced reactions in both the aqueous and gaseous phase leading to production of organic iodides. The potential and actual presence of various organic iodine species is well known (5)(6). However, since the establishment that one of these species was methyl iodide, practically all research work focused on evaluating adsorbents using methyl iodide. Based on a knowledge of the removal mechanism using activated carbon adsorbents, commonly used impregnated carbons were evaluated using I-131 tagged organic compounds other than methyl iodide.

II. Organic Iodide Isotope Exchange with Organic Iodides

The major removal mechanism for radioactive organic halides is isotope exchange with stable iodides on activated carbon carriers (7)(8). This is also the major removal mechanism for tertiary amine impregnated carbon which form stable iodine complexes which then further exchange with the radioactive organic iodides (9)(10).

The isotope exchange of organic halides with homologeous inorganic iodides or other organic halides has been extensively studied in the last fifty years. (11)(12)(13)(14)(15)(16)(17)(18). While these studies were not based on data obtained on systems using high surface area carbon supported components, their conclusions are important in the evaluation of the ease or difficulty of isotope exchange between the components involved.

Reaction rate and activation energy data from several investigators is shown on Tables 1 through 3.

The data from these Tables indicate that the isotope exchange of I^- with RI for normal aliphatic iodides decreases with increasing number of carbon atoms in the radical, at least up to four carbon atoms. One paper indicates that $n\text{-}C_4H_{11}$ I has a slightly higher isotope exchange rate than $n\text{-}C_4H_9I$. (12) At least for the isopropyl and butyl groups it is also shown that isotope exchange is significantly slower for the iso than for the normal iodide (13)(14)(15)(16).

There is also a significant decrease in the rate of isotope exchange with increasing substitution of halogens for hydrogen at least up to the point where at least one hydrogen atom is present (14)(18)(19).

One reference was found where isotope exchange between inorganic halides and CI_4 showed higher rate constants than CH_2I_2 or CHI_3 and the corresponding halide (18)

These exchange rate constant differences cannot be explained solely by the differences in activation energies of the isotope exchange process. It is suspected ΔS, the entropy of formation of the intermediate complex, is also an important parameter influencing the resulting rate constant (19).

There are indications in the literature also that olefinic equivalents exchange at a higher rate than the same carbon number parafins (18)(20).

Based on this literature review, it was decided that the use of methyl iodide--on an arbitrary basis--for the determination of the decontamination efficiency of currently used impregnated carbons may not be the most conservative method.

III. Experimental Procedures

The I-131 tagged organic iodides were obtained commercially from ICN.

ASTM D3803 method A was used with the exception that the loading time was one hour. The base carbon in all cases was US 8X16 mesh, 60 carbon tetrachloride capacity coconutshell carbon. The following impregnations were made:

1. KI, 5%
2. TEDA, 5%
3. KI, 2% + TEDA, 2%
4. Tetra Butyl Ammonium Iodide, 2.5%

The tetrabutyl ammonium iodide impregnated carbon was also used as the backup or guard bed carbon during the tests. The results of the tests are presented in Table 4.

Table 1

Isotopic Exchange Rate Constants for Alkyl Iodides with Iodide Ions at 100°C

R	Rate Constant $kX10^5$ liter $mole^{-1}$ sec^{-1}	Activation Energy kcal/mole.
CH_3I	120,000	15.4
C_2H_5I	17,500	19.0
C_3H_7I	10,000	19.3
C_3H_7I	1,000	--
CH_2I_2	230	21.7
CHI_3	120	22.3

Data from Ref. 16

Table 2

Rate Constants and Activation Energies for the Isotope Exchange of Alkyl Iodides with Iodide Ions at 25°C

	Solvent	Rate Constant $kX10^5$ liter $mole^{-1}sec^{-1}$	Activation Energy kcal/mole.
CH_3I	Ethanol	400	16.5
C_2H_5I	Ethanol	17.4	22.3
n-C_3H_7I	Ethanol	14.0	18.4
i-C_3H_7I	Ethanol	0.62	21.2
n-C_4H_9I	Ethanol	9.8	18.4
i-C_4H_9I	Ethanol	1.28	18.8
sec-C_4H_9I	Ethanol	0.71	21.8

Data from Ref. 12

Table 3

Isotopic Exchange Rate Constants at 20°C for Alkyl Iodides with Iodide Ions ($kX10^5$ liter $mole^{-1}$ sec^{-1})

CH_3I	630
C_2H_5I	13
n C_3H_7I	10

Data from Ref. 13

Table 4

Organic Iodide-131 Removal Efficiency of Various Impregnated Carbons

Impregnant	MeI	EtI	PrI	MeI_2	IB
KI	97.89	91.70	97.59	99.83	99.91
TEDA	98.82	95.31	99.60	99.95	99.99
KI + TEDA	99.19	93.25	99.11	99.90	99.91
TBNI	99.10	95.92	99.58	99.99	99.81

IV. Evaluation and Discussion of Results

The removal efficiencies indicate (see Table 4) a drop in removal efficiency for the iodoethane (compared to methyl iodoide) with a gradual increase in removal efficiency going from iodopropane to iodobenzene. This confirms preliminary results obtained by this laboratory and reported elsewhere (21).

This seems to somewhat contradict what would be expected based on isotope exchange rates. However, when the contribution of physical adsorption of these species is taken into account, the results can be explained. The boiling point for the iodomethane, iodoethane, iodopropane, diiodomethane and iodobenzene are 42, 71, 101, 181 and 188 C respectively. Thus we have a high removal efficiency for iodomethane, where the relatively low physical adsorption is offset by the fast isotopic exchange rate. A minimum is obtained in the removal efficiency with iodoethane where the increased physical adsorption cannot offset the slower exchange rate. This trend reverses with iodopropane where physical adsorption begins to dominate compared to the exchange rate. The net effect of the increased physical adsorption is to increase the residence time on the carbon and thus to allow enough time for isotopic exchange to take place even though the rate of exchange is considerably slower.

Work is continuing in this laboratory to look at the effects of loading and postsweep time on the removal of these organic iodides and also to look at their branched chain analogs.

REFERENCES

1. Mandler, J.W. et.al., "19th DOE Nuclear Air Cleaning Conf.", paper 3-2 (1986).

2. Deuber, H. et.al., DOE CONF-840806, p.98-114 (1984).

3. Malinauskas, A.P., DOE CONF-840806, p.644-646 (1984).

4. Gieseki, F.A., DOE CONF-840806, p 64-654 (1984).

5. Eggleton, A.E.F & Atkins, D.H., Trans. Am. Nucl. Soc. 6, 129 (1963).

6. Deuber, H., KfK 2435, p127-139, KfK 2500, p132-145 (1977).

7. Collins, D.A. et.al., AEC CONF-660904, p.159-198 (1966).

8. Parker, T.W. et.al., AEC CONF-660904, p.199-229 (1966).

9. Kovach, J.L. et.al., DOE CONF-820833, p.652-663 (1982).

10. Freeman, W.P. et.al., DOE CONF-840806, p.93-97 (1984).

11. Juliusberger, F. et.al., J. Chem. Phys. 3, 437 (1935).

12. Hull, D.E. et.al., J. Am. Chem. Soc. 58, 535 (1936).

13. Myers, O. & Prestwood, R., Radioactivity Applied to Chemistry, p.11 (1954).

14. Neiman, M.B. et.al., J. Phys. Chem. 28, 1243 (1954).

15. McKay, H., J. Am. Chem. Soc. C5, 702 (1943).

16. Seelig, H. &Hull, D., J. Am. Chem. Soc. 64, 940 (1942).

17. Neiman, M.B. et.al. Dokl., Akad Nauk USSR, 92, 611 (1953).

18. Neiman, M.B. et,al., Symposium on "Questions of Chemical Kinetics, Catalysis, And Reactivity", Moscow Akad Nauk, USSR p.722 (1955).

19. Roginski, S.Z., USAEC-TR-2873 (translation from revision), p.211 (1956).

20. Brezhneva, N.D. & Roginski, S.Z., Progr. Chem. 7 1528 (1938).

21. Kovach, J.L., "Conference on Gaseous Effluent Treatment in Nuclear Installations", Luxembourg, Oct. 1985.

INVESTIGATIONS ON THE AGING OF VARIOUS ACTIVATED CARBONS IN THE EXHAUST AIR OF A PWR OVER AN EXTENDED PERIOD OF TIME

H. Deuber, K. Gerlach, V. Giraud, R. Kaempffer

Kernforschungszentrum Karlsruhe GmbH
Laboratorium für Aerosolphysik und Filtertechnik
Postfach 3640, D-7500 Karlsruhe 1
Federal Republic of Germany

Abstract

With 15 different commercial impregnated activated carbons aged in the containment exhaust air of a PWR over periods of up to 9 months, a tendency for a relatively low penetration by CH_3I-131 was observed for the carbons impregnated additionally or exclusively with a tertiary amine (e.g. TEDA or derivative). It is concluded that in particular for iodine filters of large bed depths comparatively long stay times should be obtained with amine containing carbons under the conditions investigated.

I. Introduction

Nuclear power plants are equipped with iodine filters containing impregnated activated carbons to keep the release of airborne radioiodine as low as reasonably achievable.

Impregnated activated carbons are known to age, i.e. to deteriorate in performance with respect to the retention of radioiodine, particularly in iodine filters that are continuously operated (e.g. containment exhaust air filters of pressurized water reactors). The aging of carbons in iodine filters is essentially due to the adsorption of organic compounds (e.g. solvents) and of inorganic compounds (e.g. O_x, SO_2, NO_x). In the first case the effective surface of the carbon is lowered. In the second case reactions with the carbon (and the impurities such as sulphur) as well as with the impregnant can occur. The alkalinity of the carbon can be reduced in this case.

In iodine filters various impregnated activated carbons are used differing e.g. in base material, grain size and impregnant. In recent years new impregnated activated carbons were developed (see below). Therefore we have performed investigations to provide comparative data on the aging of a variety of commercial impregnated activated carbons from several countries under relevant conditions. The data relate to the retention of methyl iodide (CH_3I-131).

In the present paper, investigations are covered on the aging of 15 different impregnated activated carbons in the containment exhaust air of a typical German pressurized water reactor (PWR 3) over periods of up to 9 months. Data on the aging of 5 carbons in the containment exhaust air of a similar pressurized water reactor (PWR 4) over periods of up to 3 months have already been published. /1/

II. Former Investigations

Numerous investigations have been performed on the aging of impregnated activated carbons in the exhaust air of nuclear power stations. /2,3,4/ It is difficult, however, to judge from these investigations the aging of different carbons in the exhaust air of other nuclear power stations. This is particularly due to the complexity of the aging process and to the differences in type and concentration of the pollutants in the exhaust air.

It must also be mentioned that mostly carbons of small bed depths (residence times (t): less than 0.5 s) were used in these investigations. However, it is not possible to reliably extrapolate to large bed depths (t : > 0.5 s) which, to obtain a long stay time, are used for iodine filters in some countries.

Because of these reasons it is only mentioned here that in studies performed under various conditions, often a good aging behavior with respect to the retention of CH_3I-131 was found with carbons impregnated additionally or exclusively with TEDA (triethylene-diamine) or another tertiary amine. /5,6,7/ Also during the storage of carbons (in closed containers), in which the aging is relatively small, the best aging behavior was observed with a TEDA-impregnated carbon. /8,9/

In our investigations on the aging of 5 carbons in the containment exhaust air of PWR 4 over periods of up to 3 months, also comparatively low penetrations by CH_3I-131 were observed for carbons impregnated additionally or exclusively with a tertiary amine. /1/ However, it was also found that the change in penetration due to aging can be stronger for amine-impregnated carbons than for e.g. KI-impregnated carbons.

Earlier aging investigations in German nuclear power plants were almost exclusively conducted with KI-impregnated carbons, essentially in order to identify the pollutants and optimize the layout of iodine filters. /10,11/

There are some doubts on the suitability of TEDA-impregnated carbons for use in iodine filters of nuclear power plants because of the low ignition temperature and high vapor pressure of TEDA. By using derivatives of TEDA ("heavy TEDA"), possibly together with antioxidants, fire hazards can be reduced. /12,13/ It is mentioned here that in recent comparison tests with a KI- und a TEDA-impregnated carbon at 180 °C, no difference with respect to the retention of CH_3I-131 was observed. /14/

III. Experimental

Data regarding the activated carbons investigated are contained in Table I. Three main groups of impregnants can be distinguished:

(a) KI or KI + I_2;
(b) KI + tertiary amine;
(c) tertiary amine.

Moreover, a carbon impregnated with an organic iodide was investigated.

Most of these carbons were developed in recent years. Efforts were made to include more recently developed carbons in order to cover even a broader range of carbons. However, the companies contacted were not able to supply these carbons in time.

The investigations covered in this paper were essentially performed as those conducted earlier. /1/ The carbons, mounted in sectioned beds, were challenged with the containment exhaust air of PWR 3 over periods of 3, 6 and 9 months, respectively, during power operation of the reactor. The periods did not overlap. The operating conditions of the carbon beds corresponded largely to the operating conditions of the iodine filters in German nuclear power stations in normal situations and were essentially identical with those in the subsequent laboratory tests with CH_3I-131. The parameters of the laboratory tests are given in Table II.

The tests with CH_3I-131 were run with both fresh and aged carbons. In the second case, the original arrangement of the carbons was maintained. Thus, the geometric course of the aging within the sectioned beds could be ascertained. As indicated in Table II, the aged carbons were preconditioned over a period of 1 h only to minimize the desorption of pollutants. (This duration should be sufficient due to the largely identical conditions during aging and testing.)

More details on the performance of the tests with CH_3I-131 are to be found in the literature. /4/ The reproducibility in these tests is excellent in general. /15/ The minimum detectable penetration, determined with NaI detectors, was 10^{-5} to 10^{-4} %.

As in our previous investigations, an additional bed of the carbon 207B (KI) was simultaneously challenged with the exhaust air to determine both the loading of the bed with organic compounds (mainly by gas chromatographic analysis of the CCl_4 extract) and its alkalinity (by pH value measurement of the H_2O extract).

IV. Results

In this chapter first the results are presented of the investigations on the retention of CH_3I-131 by the 15 carbons contained in Table I under the conditions indicated in Table II.

The results are given in terms of penetration by CH_3I-131. Tables III to V contain the penetration for fresh carbons (aging time: 0 months) and aged carbons (aging time: 3,6 and 9 months, respectively) at 3 bed depths (12.5, 25.0 and 37.5 cm equivalent to residence times (t) of 0.25, 0.5 and 0.75 s, respectively). Figures 1 to 15 display the same values up to the maximum bed depth (50 cm). Finally, Figures 16 to 18 show the results for the maximum aging time (9 months) for the above-mentioned 3 bed depths.

Figures 1 to 15 show that, using a semilogarithmic plot, with the fresh carbons the usual linear decrease of penetration with increasing bed depth was found. With the aged carbons, a nonlinear course of the penetration curves was obtained: flat at low bed depths and steep at large bed depths, in some cases nearly parallel to the penetration curve of the respective fresh carbon. This form of the penetration curves corresponds to a decrease in aging with an increase in bed depth, with a negligibly small aging in the above-mentioned cases of nearly parallel penetration curves.

Figures 1 to 15 also show that there is a tendency for the same penetration at aging times of 3 and 6 months. (In some cases the penetration at an aging time of 3 months was nominally even higher than at 6 months). Since the aging periods did not overlap, this is possible implying e.g. that the concentration of pollutants in the exhaust air was relatively low during the aging period of 6 months. Some results may also be attributable to experimental errors.

Apart from a few exceptions, the main results obtained for both fresh and aged carbons may be summarized as follows, considering a small bed depth (12.5 cm, t = 0.25 s) and a large bed depth (37.5 cm, t = 0.75 s), respectively:

(a) Fresh carbons:
At a bed depth of 12.5 cm a penetration of between 10^{-4} and 10^{-2} % was found for carbons with KI or KI_3 as the impregnant and of below 10^{-4} % for carbons with the other impregnants. At a bed depth of 37.5 cm the penetration was below 10^{-4} % for all the carbons.

(b) Carbons aged 3 or 6 months:
At a bed depth of 12.5 cm a penetration of about 10^{1} % was observed for carbons with the KI or KI_3 impregnants and of between 10^{-1} to 10^{1} for carbons with the other impregnants. At a bed depth of 37.5 cm the values were $< 10^{-4}$ to 10^{-1} % for the KI impregnant and $< 10^{-4}$ to 10^{-3} % for both the KI_3 and the other impregnants.

(c) Carbons aged 9 months:
At a bed depth of 12.5 cm the penetration was always between 10^1 and 10^2 %. However, at a bed depth of 37.5 cm, a penetration of 10^{-1} to 10^0 % was found for the KI impregnant, of 10^{-4} to 10^{-2} % for the KI_3 impregnant and of $< 10^{-4}$ to 10^{-2} % for the other impregnants.

The main exception was the carbon BPJ which, compared with the other carbons, showed a very high penetration after aging.

The results show a tendency for a comparatively low penetration for the carbons impregnated additionally or exclusively with TEDA or another tertiary amine (or an organic iodide). It is only at the maximum aging time and at low bed depths that this tendency is not evident. The implication of this finding is that the aging of amine-impregnated carbons was stronger than e.g. of KI-impregnated carbons (except for BPJ). The tendency for a comparatively low penetration for the amine-impregnated carbons after aging was consequently due to a better performance prior to aging. This is in agreement with our previous findings. /1/

As for the base materials and grain sizes of the carbons, it is believed that from the investigations performed one should not tempt to draw conclusions on their influence on the aging behavior of the carbons.

As for the additional investigations mentioned above, Figure 19 shows the loading of 207B (KI) with organic compounds of different volatility at an aging time of 9 months. This loading was obtained by gas chromatography. Whereas the compounds of high volatility were found on all the beds, the compounds of low volatility were only observed on beds 1 through 8. The former compounds consisted in particular of toluene, xylene and nonane, the latter compounds of decane and dodecane. Mass spectrometric measurements of the low-volatile compounds revealed methyl and ethyl derivatives of these hydrocarbons and, additionally, of benzene. As the alkalinity of the aged 207B (KI) was found to be unchanged, it is obvious that, in agreement with former measurements, the adsoprtion of organic compounds was a main reason for the aging.

V. Summary and Conclusions

Investigations were performed on the aging of 15 different commercial impregnated activated carbons from several countries in the containment exhaust air of a typical German pressurized water reactor (PWR 3) over periods of up to 9 months. The aging was determined with respect to the penetration by CH_3I-131 under the in-situ conditions (30 °C, 40 % R.H.).

A tendency for a comparatively low penetration was observed for the carbons impregnated additionally or exclusively with a tertiary amine (e.g. TEDA or derivative). This applies in particular to large bed depths (residence times: > 0.5 s) where the good performance of these carbons when fresh is dominant.

Based on the comprehensive data obtained it is concluded that in particular for iodine filters of large bed depths comparatively long stay times should be obtained with amine containing carbons under the conditions investigated. Taking into account the ignition temperature and vapor pressure of amines, derivatives of TEDA are more eligible than TEDA.

Acknowledgment

The following coworkers participated in performing and evaluating the tests:
M. Dehm, S. Engelhardt, A. Hengst, A. Ladanyi and R. Sommerlatt.

VI. References

/1/ Deuber, H.; Gerlach, K.; Kaempffer, R.;
"Investigations on the Aging of Activated Carbons in the Exhaust Air of a Pressurized Water Reactor".
Nucl. Techn. 70 (1985) p. 161.

/2/ Holladay, D.W.;
"A Literature Survey: Methods for the Removal of Iodine Species from Off-Gases and Liquid Waste Streams of Nuclear Power and Nuclear Fuel Reprocessing Plants, with Emphasis on Solid Sorbents".
ORNL/TM - 6350 (1979).

/3/ Kovach, J.L.;
"The Evolution and Current State of Radio-Iodine Control".
16th DOE Nuclear Air Cleaning Conference,
San Diego, 20.-23.10.1980,
CONF-801 038 (1981) p. 417.

/4/ Wilhelm, J.G.;
"Iodine Filters in Nuclear Installations".
Commission of the European Communities,
V/2110/83 (1982).

/5/ Kovach, J.L.; Rankovic, L.;
"Evaluation and Control of Poisoning of Impregnated Carbons Used for Organic Iodide Removal".
15the DOE Nuclear Air Cleaning Conference,
Boston, 7. - 10.8.1978,
CONF-780 819 (1979) p. 368.

/6/ Deitz, V.R.;
"Effects of Weathering on Impregnated Charcoal Performance".
NUREG / CR-2112 (1981).

/7/ Broadbent, D.;
"The Ageing and Poisoning of Charcoal Used in Nuclear Plant Air Cleaning Systems".
CEC European Conference on Gaseous Effluent Treatment in Nuclear Installations,
Luxembourg, 14. - 18.10.1985.

/8/ Collins, R.D.; Hillary, J.J.; Taylor, L.R.; Abbey, F.;
"The Ageing of Charcoals Used to Trap Radioiodine".
IAEA/NEA (OECD) Symposium on Management of Gaseous Wastes from Nuclear Facilities,
Vienna, 18. - 22.2.1980,
STI/PUB/561 (1980) p. 571.

/9/ Hillary, J.J.; Evans, M.G.; Taylor, L.R.;
"The Ageing of European Charcoals Used to Trap Radioiodine".
CEC Seminar on Iodine Removal from Gaseous Effluents in the Nuclear Industry,
Mol, 21. - 24.9.1981,
V/5283/82 (1982) p. 167.

/10/ Furrer, J.; Kaempffer, R.; Wilhelm, J.G.;
"Alterung und Vergiftung von Iod-Sorptionsmaterialien in Kernkraftwerken / Ageing and Poisoning of Iodine Filters in Nuclear Power Plants".
Kerntechnik 18 (1976) p. 313.

/11/ Wilhelm, J.G.; Furrer, J.; Kaempffer, R.;
"Untersuchung zur Ertüchtigung von Iodsorptionsfiltern zur Reinigung der Abluft von Kernkraftwerken".
KfK 3194 B (1981).

/12/ Underhill, D.; Laskie, J.;
"Modified TEDA Impregnants for Methyl Iodide Removal".
16th DOE Nuclear Air Cleaning Conference,
San Diego, 20. - 23.10.1980,
CONF-801 038 (1981) p. 531.

/13/ Kovach, J.L.; Grimm, J.J.; Freeman, W.P.;
"TEDA v s. Quinuclidine: Evaluation and Comparison of Two Teriary Amine Impregnants for Methyl Iodide Removal from Flow Air Stream".
17th DOE Nuclear Air Cleaning Conference,
Denver, 2. - 5.8.1982,
CONF-820 833 (1983) p. 652.

/14/ Deuber, H.; Dillmann, H.-G.; Gerlach, K.;
"Retention of Gaseous Radioiodine with Sorbents Under Accident Conditions".
Fifth International Meeting on Thermal Nuclear Reactor Safety,
Karlsruhe, 9. - 13.9.1984,
KfK 3880 (1984) p. 1505.

/15/ Deuber, H.; Gerlach, K.;
"Parametric Investigations on the Retention of Methyl Iodide by a KI-Impregnated Activated Carbon".
Nuclear Safety 26 (1985) p. 157.

Table I: Data of the activated carbons investigaded

Name	Impregnant	Base material	Size [a]	Supplier
207B	KI	coal	8 - 12 mesh	Sutcliffe Speakman
207B PA	KI	coal	8 - 12 mesh	Sutcliffe Speakman
SA 1565	KI	coal	1.6- 4 mm	Ceca
BPJ	KI	peat	1 - 2 mm	Degussa
727	KI, I_2	coconut shell	8 - 16 mesh	Barnaby and Cheney
463563	KI, I_2	coconut shell	8 - 16 mesh	Mine Safety Appliances
207B	KI, TEDA	coal	8 - 12 mesh	Sutcliffe Speakman
208C	KI, TEDA	coconut shell	8 - 12 mesh	Sutcliffe Speakman
G615	KI, TEDA	coconut shell	8 - 16 mesh	Pica
RI 180	KI, TEDA	coconut shell	1.6- 4 mm	Pica
787	KI, TEDA	coconut shell	8 - 16 mesh	Barnaby and Cheney
Kiteg II	KI, amine [b]	coconut shell	8 - 16 mesh	Nucon
207B	TEDA	coal	8 - 12 mesh	Sutcliffe Speakman
Q	amine [c]	coconut shell	8 - 16 mesh	Nucon
X	org. iodide	coconut shell	8 - 16 mesh	Nucon

[a] 8 - 12 mesh: 2.0 - 1.4 mm (BS 410); 8 - 16 mesh: 2.4 - 1.2 mm (ASTM D2862)

[b] amine: alkyl derivative of TEDA (also additional impregnants)

[c] amine: quinuclidine

Table II: Parameters of the tests with CH_3I-131

Parameter	Unit	Value
Temperature	°C	30 [a]
Relative humidity	%	40 [a]
Throughput	m^3/h	0.88 [a]
Face velocity	cm/s	50 [a]
Preconditioning time	h	1 [b]
Injection time	h	1
Purging time	h	2
Bed depth	cm	50 [a, c]
Residence time	s	1 [a]
I-131 injected	mCi	0.01 - 0.1
I-127 injected	mg	1

[a] Value (largely) identical with that during the aging in the nuclear power plant

[b] $\geq$ 16 h with fresh carbon

[c] 20 beds of a depth of 2.5 cm (diameter: 2.5 cm); sequence identical with that during the aging in the nuclear power plant

Table III: Penetration of various impregnated activated carbons by CH_3I-131 at different bed depths and aging times

(Data of activated carbons: Table I; test data: Table II)

Name	Impregnant	Bed depth (cm)	Penetration (%)[a]			
			0 months	3 months	6 months	9 months
207B	KI	12.5	2.8 (-3)	2.2 (+1)	1.6 (+1)	4.2 (+1)
		25.0	-	1.1 (0)	3.8 (-1)	4.3 (0)
		37.5	-	3.6 (-2)	8.1 (-3)	3.5 (-1)
207B PA	KI	12.5	9.6 (-4)	1.2 (+1)	1.5 (+1)	5.0 (+1)
		25.0	-	1.0 (-1)	3.9 (-1)	5.5 (0)
		37.5	-	2.2 (-4)	1.1 (-3)	3.8 (-1)
SA 1565	KI	12.5	3.3 (-4)	1.1 (+1)	1.8 (+1)	4.8 (+1)
		25.0	-	1.2 (-1)	2.6 (-1)	1.8 (0)
		37.5	-	2.0 (-4)	-	8.9 (-2)
BPJ	KI	12.5	4.6 (-3)	5.5 (+1)	7.4 (+1)	9.3 (+1)
		25.0	-	4.0 (0)	2.0 (+1)	5.7 (+1)
		37.5	-	4.6 (-2)	4.1 (-1)	1.4 (+1)
727	KI, I_2	12.5	1.5 (-3)	1.1 (+1)	1.8 (+1)	5.2 (+1)
		25.0	-	1.0 (-1)	2.8 (-1)	2.4 (0)
		37.5	-	1.2 (-4)	8.7 (-4)	3.3 (-3)

[a] 2.8 (-3) = $2.8 \cdot 10^{-3}$ etc;
-: penetration lower than minimum detectable penetration (10^{-5} - 10^{-4} %)
(also in Tables IV and V)

Table IV: Penetration of various impregnated activated carbons by CH_3I-131 at different bed depths and aging times

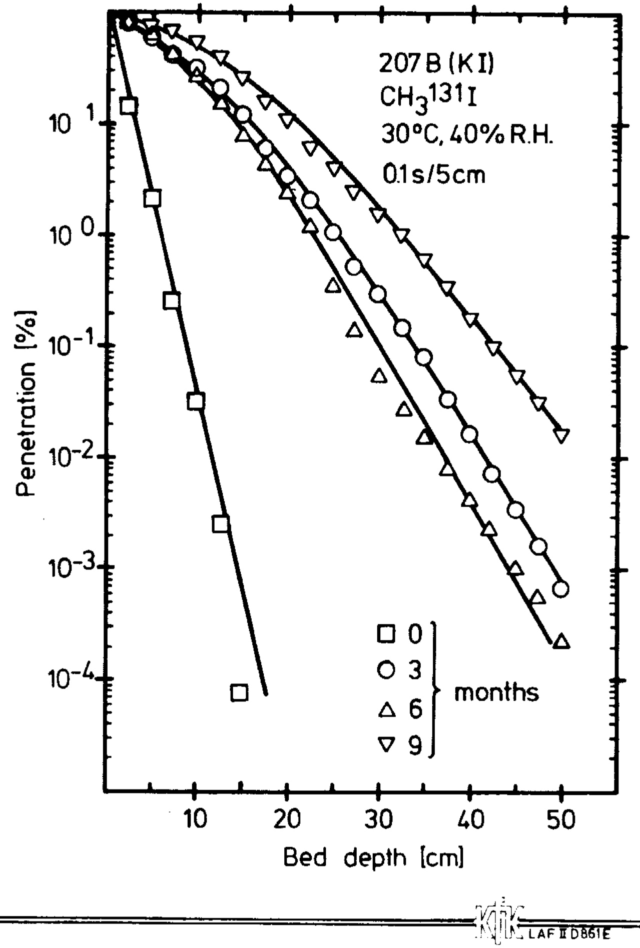
(Data of activated carbons: Table I; test data: Table II)

Name	Impregnant	Bed depth (cm)	Penetration (%)			
			0 months	3 months	6 months	9 months
463563	KI, I_2	12.5	3.8 (-4)	6.9 (0)	6.8 (0)	5.1 (+1)
		25.0	-	2.1 (-2)	3.1 (-3)	1.6 (0)
		37.5	-	-	-	1.2 (-4)
207B	KI, TEDA	12.5	-	2.4 (0)	1.4 (+1)	3.4 (+1)
		25.0	-	1.0 (-3)	1.5 (-1)	1.9 (0)
		37.5	-	-	5.2 (-4)	4.1 (-2)
208C	KI, TEDA	12.5	-	2.2 (-1)	1.1 (0)	1.7 (+1)
		25.0	-	-	-	1.3 (-2)
		37.5	-	-	-	-
G 615	KI, TEDA	12.5	-	4.8 (0)	1.6 (0)	3.2 (+1)
		25.0	-	4.2 (-2)	1.6 (-4)	6.1 (-1)
		37.5	-	5.8 (-4)	-	6.8 (-4)
RI 180	KI, TEDA	12.5	-	4.2 (0)	5.2 (0)	3.3 (+1)
		25.0	-	1.1 (-2)	4.8 (-3)	4.3 (-1)
		37.5	-	-	-	1.1 (-4)

Table V: Penetration of various impregnated activated carbons by CH_3I-131 at different bed depths and aging times

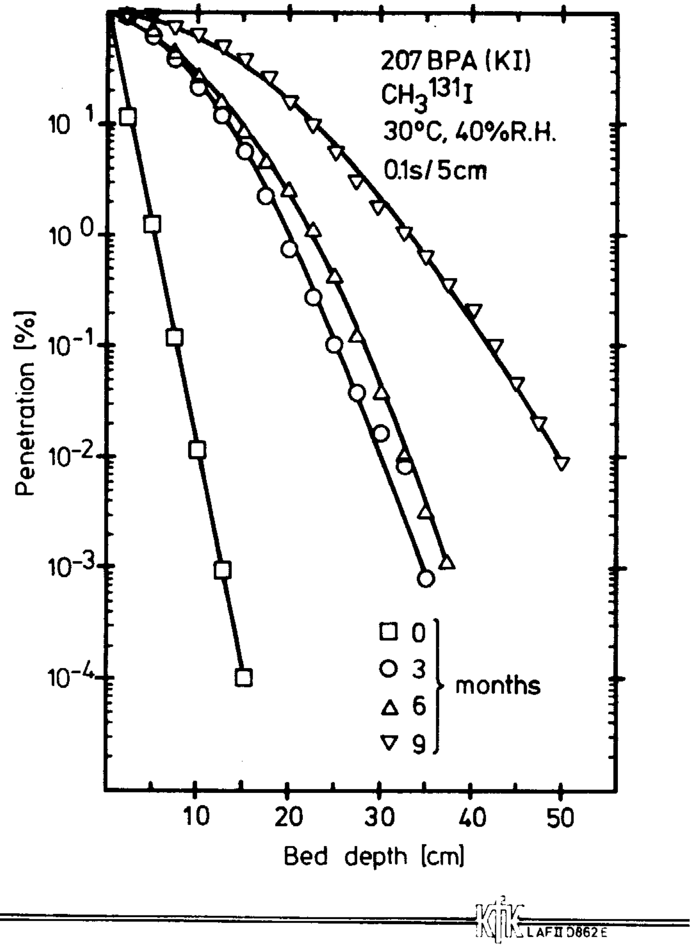
(Data of activated carbons: Table I; test data: Table II)

Name	Impregnant	Bed depth (cm)	Penetration (%)			
			0 months	3 months	6 months	9 months
787	KI, TEDA	12.5	-	2.9 (-1)	2.4 (0)	2.0 (+1)
		25.0	-	-	8.0 (-5)	7.8 (-2)
		37.5	-	-	-	-
Kiteg II	KI, amine	12.5	3.6 (-5)	5.8 (0)	1.8 (1)	5.8 (+1)
		25.0	-	2.7 (-2)	2.3 (-2)	1.8 (0)
		37.5	-	-	-	2.3 (-3)
207B	TEDA	12.5	-	9.9 (-1)	2.8 (0)	2.5 (+1)
		25.0	-	-	9.1 (-4)	1.2 (-1)
		37.5	-	-	-	-
Q	amine	12.5	5.4 (-5)	5.3 (0)	1.5 (+1)	4.9 (+1)
		25.0	-	7.1 (-3)	1.4 (-2)	8.0 (-1)
		37.5	-	-	-	1.7 (-3)
X	org. iodide	12.5	-	4.9 (0)	5.1 (0)	4.4 (+1)
		25.0	-	8.8 (-3)	4.6 (-3)	2.6 (-1)
		37.5	-	-	-	-



Penetration as a function of bed depth at different aging times

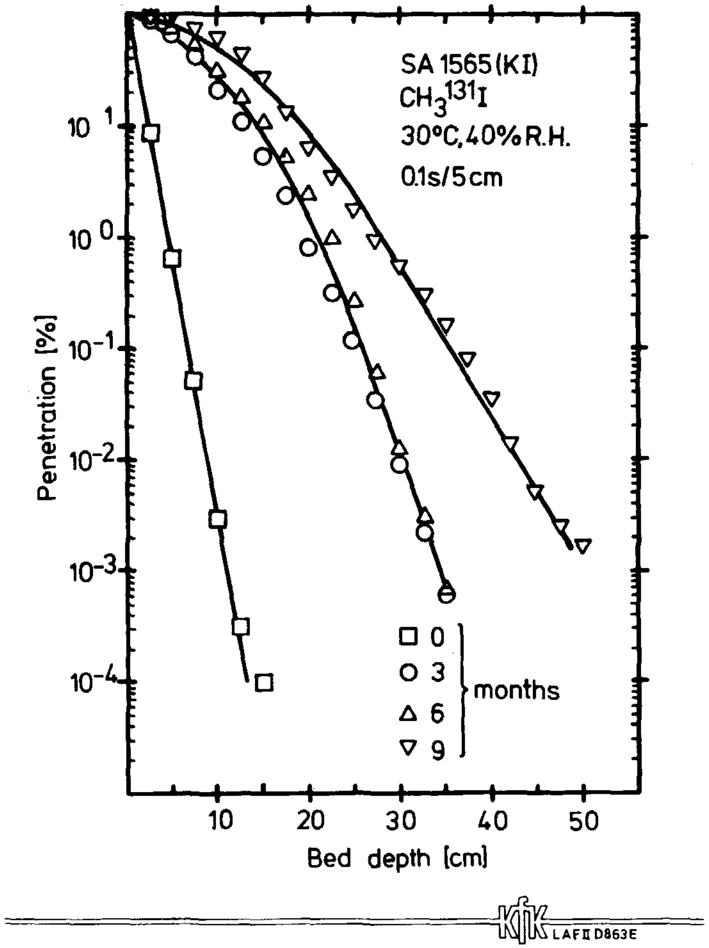
Fig. 1 [a]



Penetration as a function of bed depth at different aging times

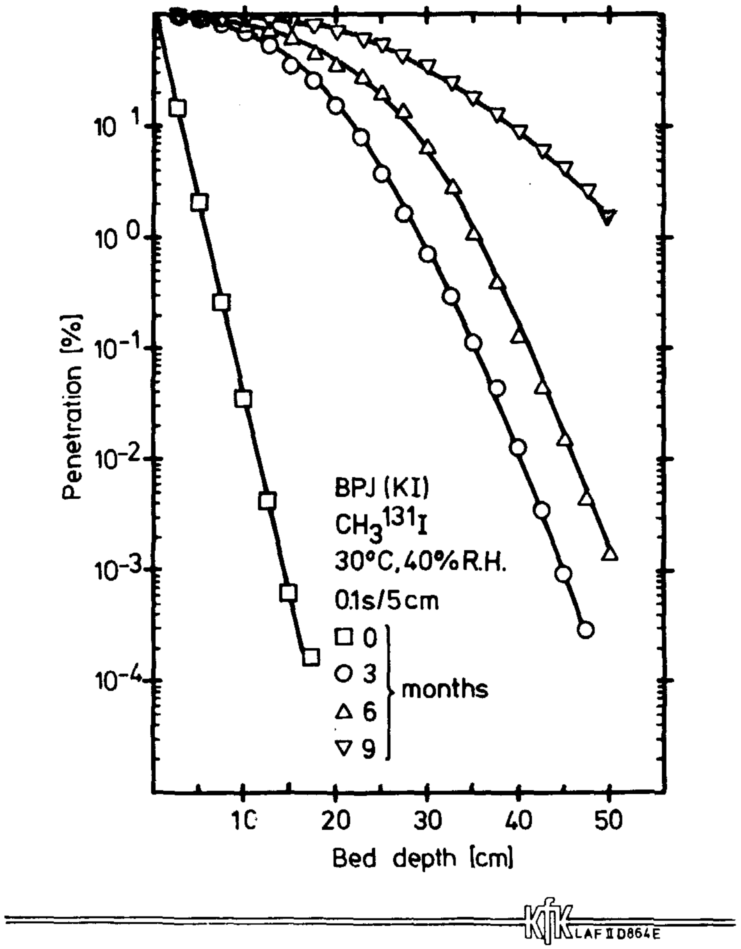
Fig. 2 [a]

[a] No overlapping of the aging periods (also in Figs. 3 - 15)



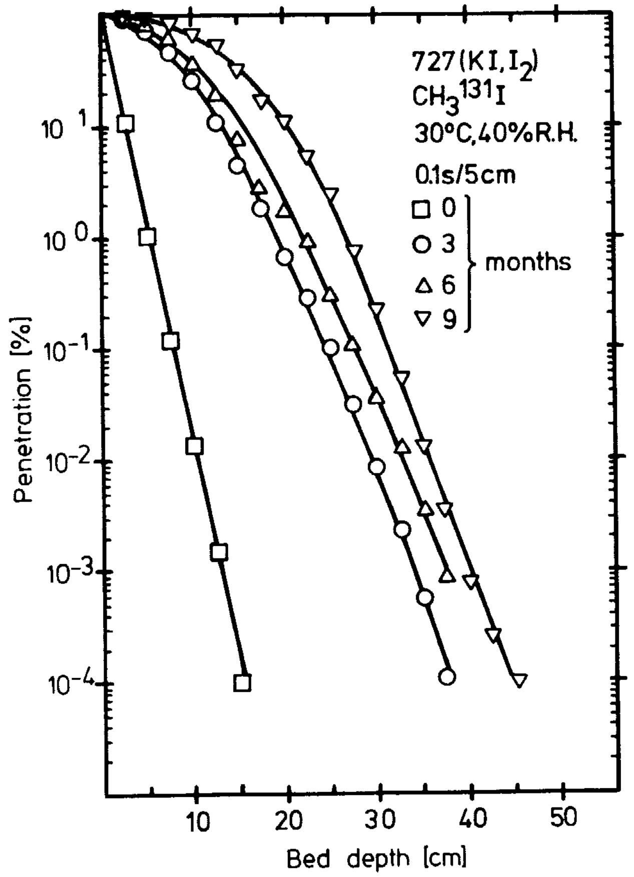
Penetration as a function of bed depth at different aging times

Fig. 3



Penetration as a function of bed depth at different aging times

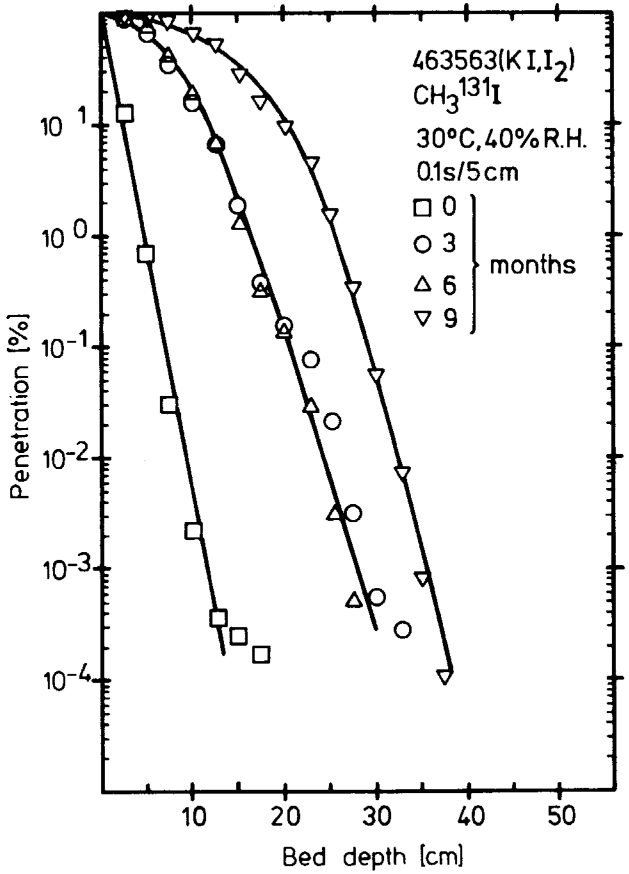
Fig. 4



LAF II D865E

Penetration as a function of bed depth at different aging times

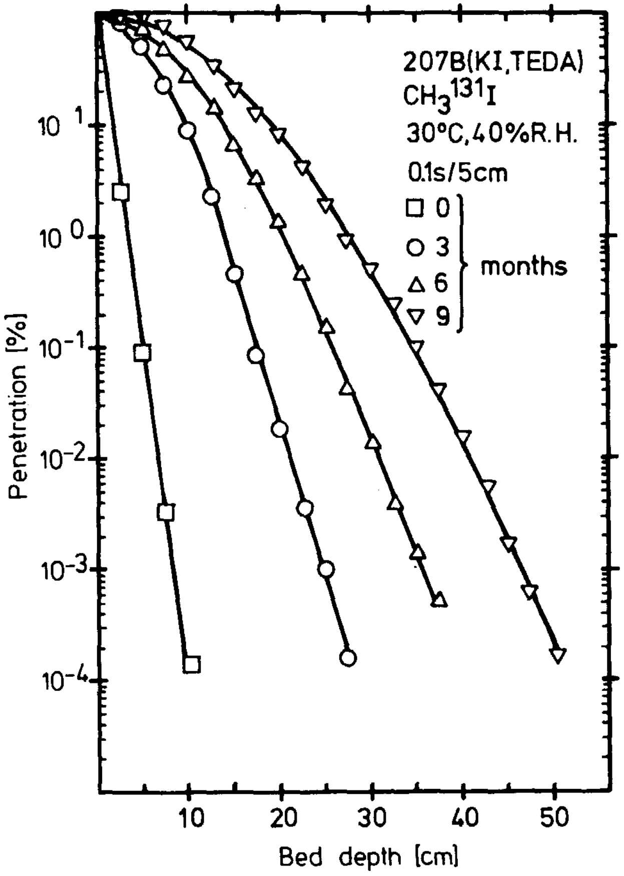
Fig. 5



LAF II D866E

Penetration as a function of bed depth at different aging times

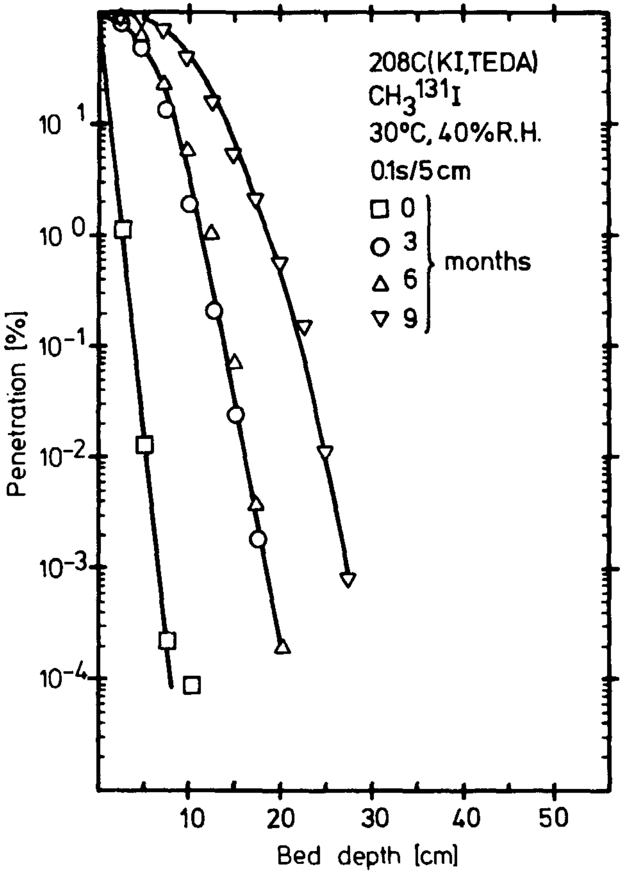
Fig. 6



KfK LAF II D867E

Penetration as a function of bed depth at different aging times

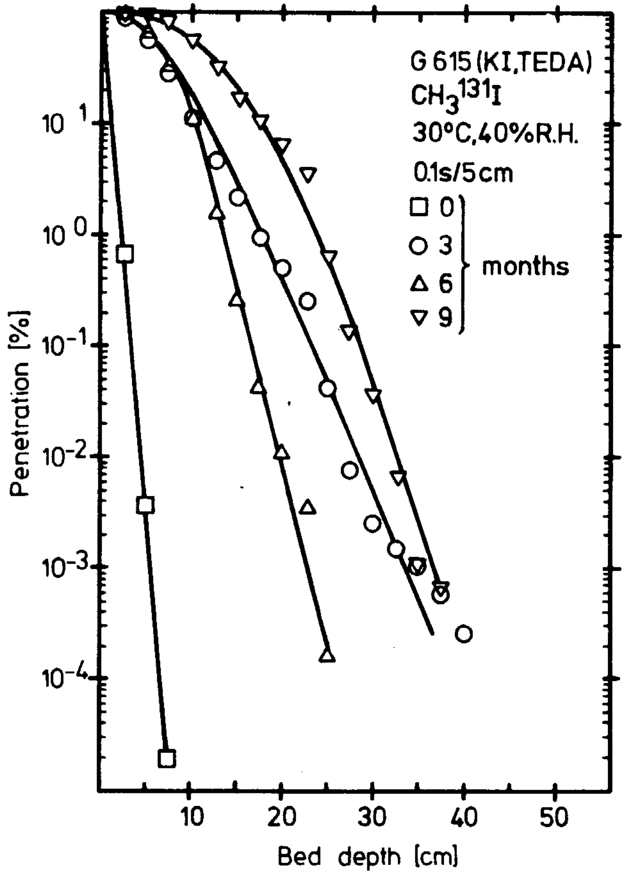
Fig. 7



KfK LAF II D8613E

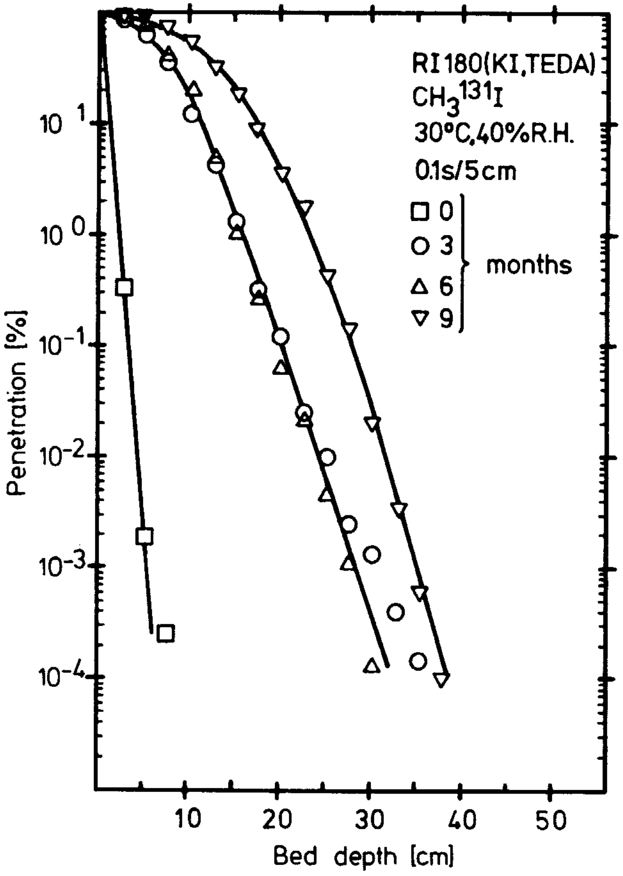
Penetration as a function of bed depth at different aging times

Fig. 8



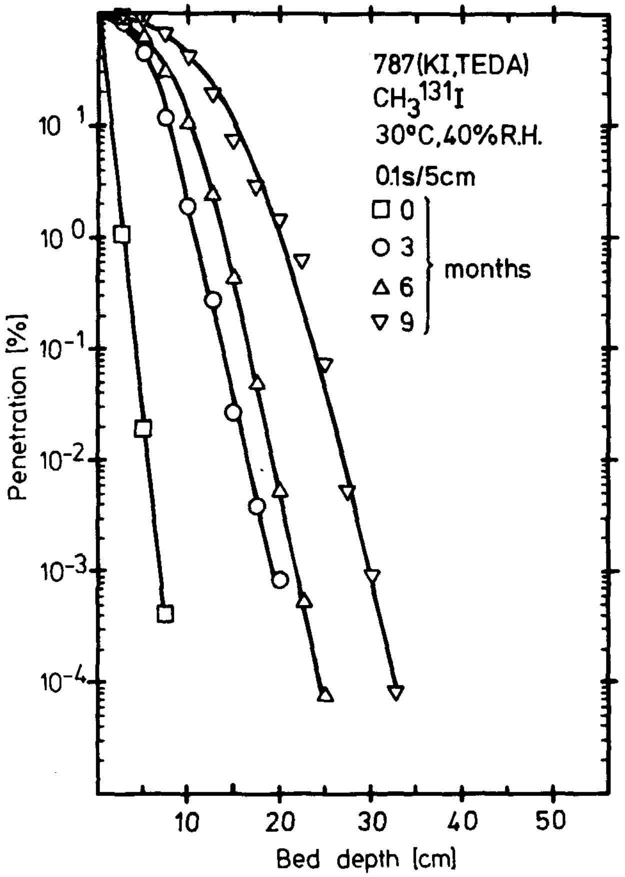
Penetration as a function of bed depth
at different aging times

Fig. 9



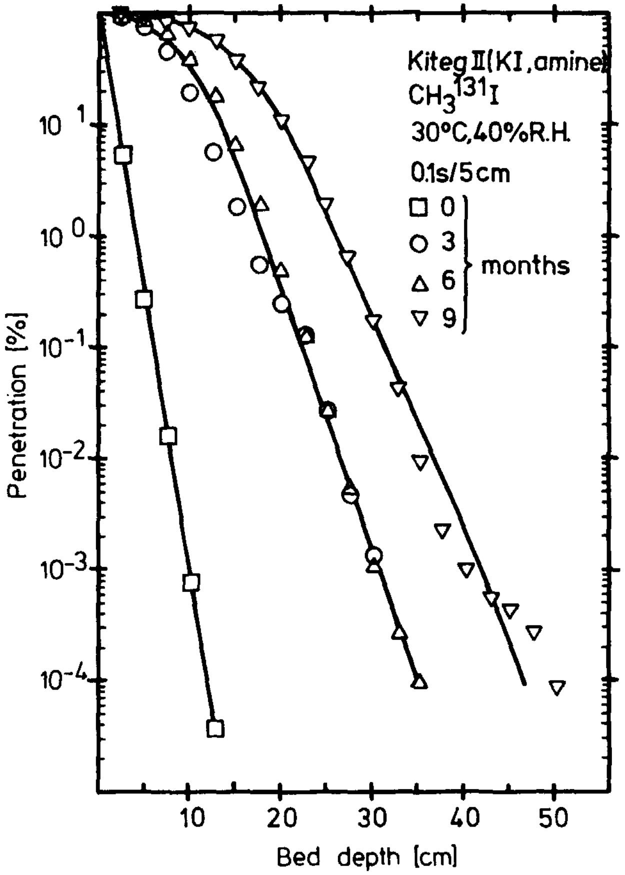
Penetration as a function of bed depth
at different aging times

Fig. 10



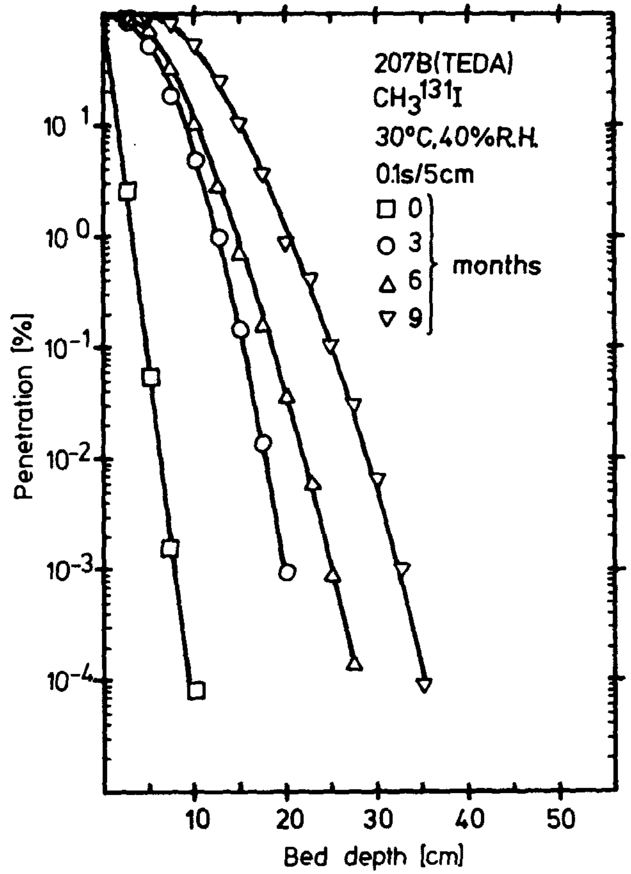
Penetration as a function of bed depth
at different aging times

Fig. 11



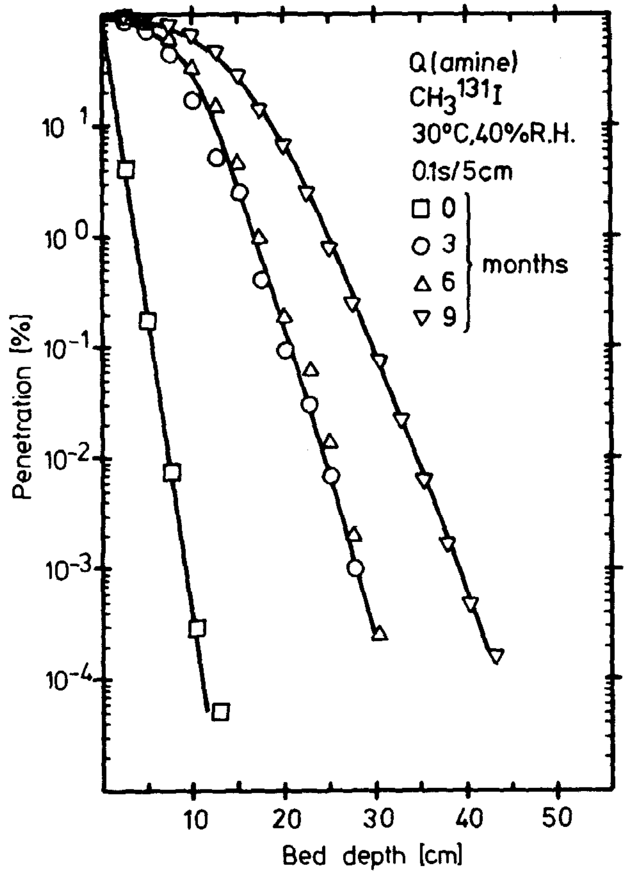
Penetration as a function of bed depth
at different aging times

Fig. 12



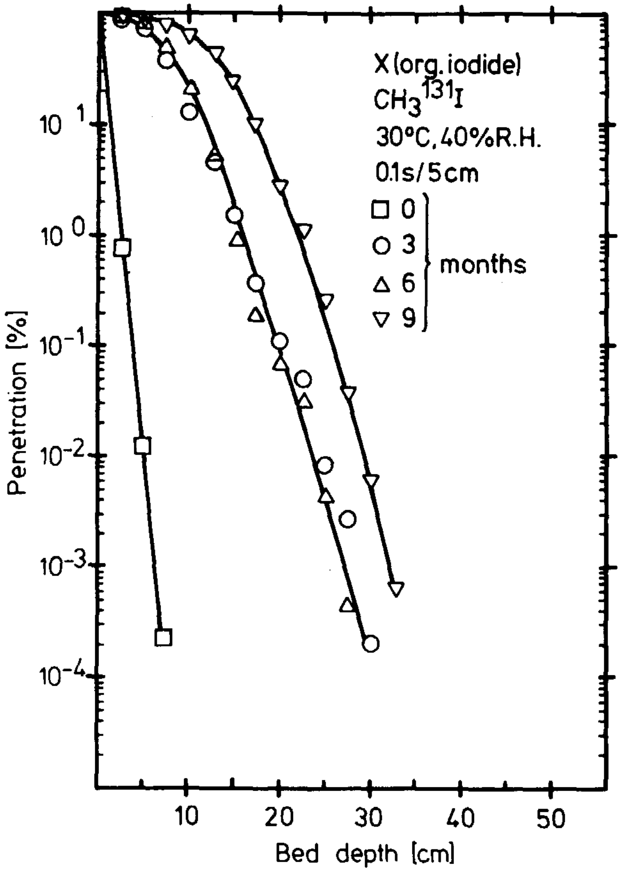
Penetration as a function of bed depth
at different aging times

Fig. 13



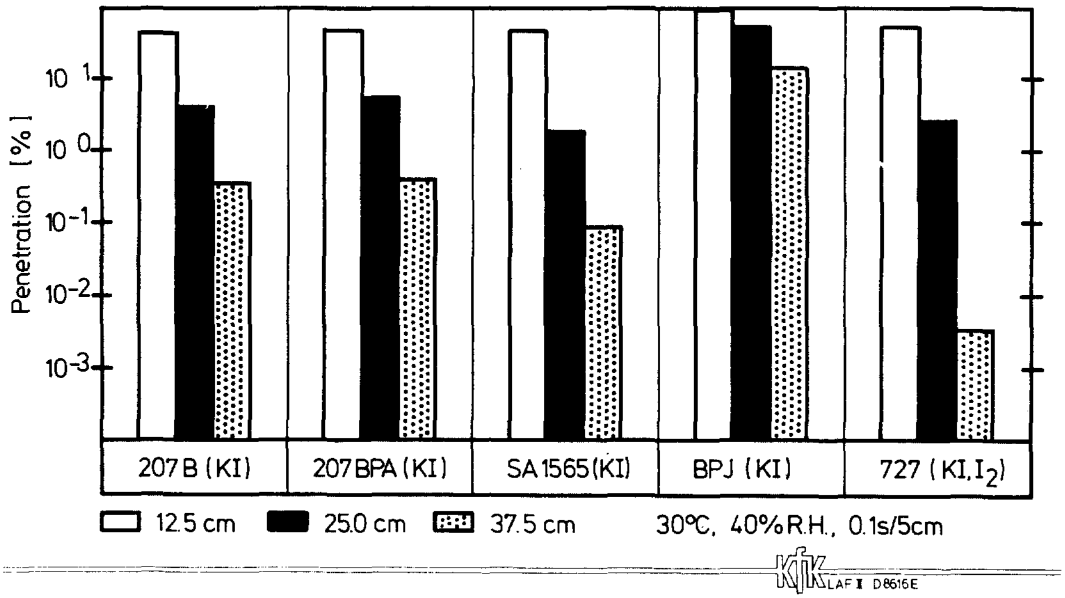
Penetration as a function of bed depth
at different aging times

Fig. 14



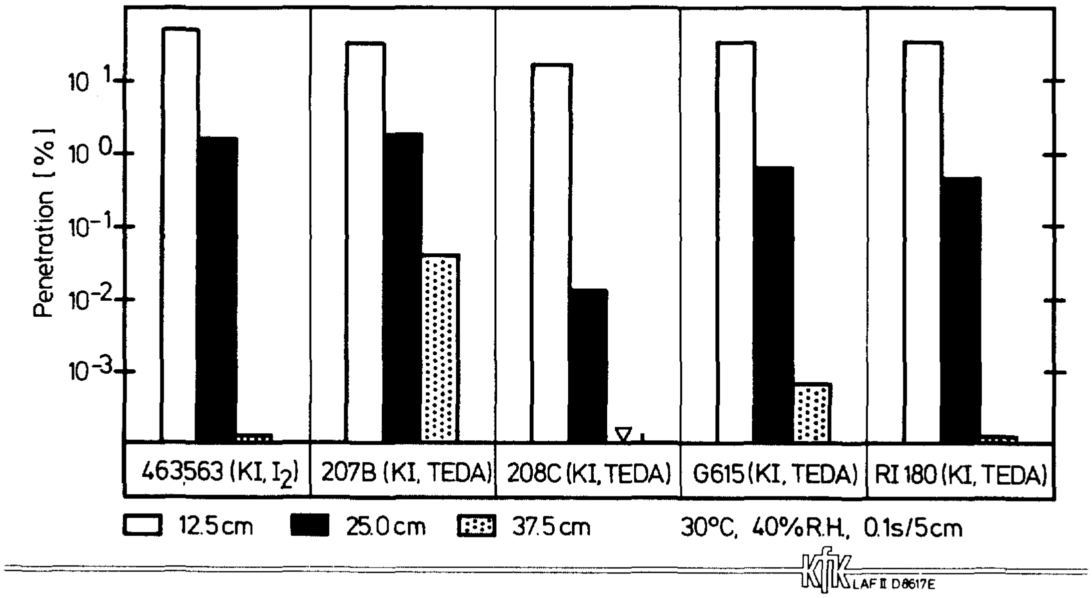
Penetration as a function of bed depth
at different aging times

Fig. 15



Penetration of various impregnated activated carbons by $CH_3{}^{131}I$ at different bed depths (aging time: 9 months)

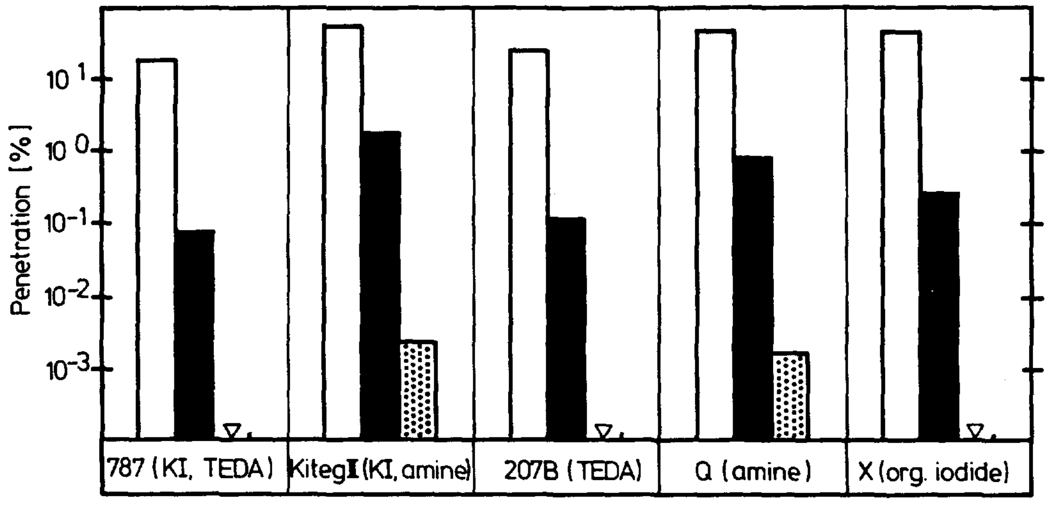
Fig. 16



Penetration of various impregnated activated carbons by $CH_3{}^{131}I$ at different bed depths (aging time: 9 months)

Fig. 17 [a]

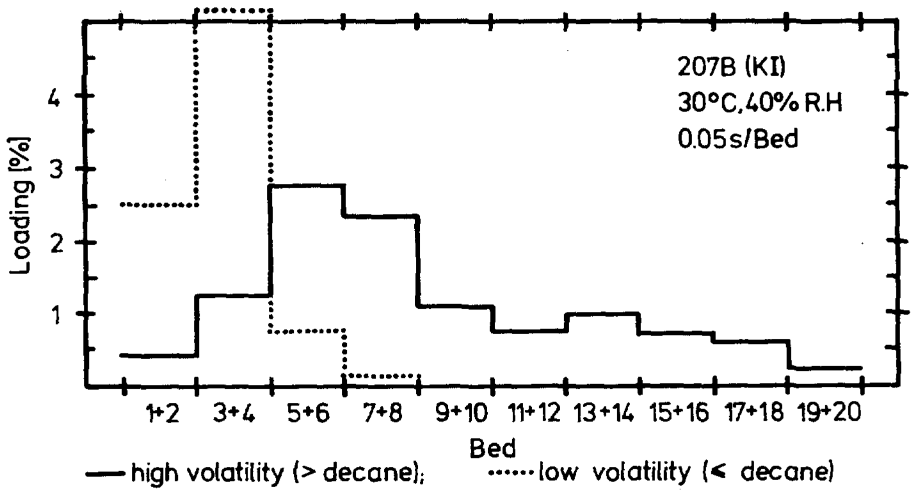
[a] ∇: penetration lower than 10^{-4} % (also in Fig. 18)



LAF I D8618 E

Penetration of various impregnated activated carbons by $CH_3{}^{131}I$ at different bed depths (aging time: 9months)

Fig. 18



LAF II D8548E

Loading of activated carbon beds with organic compounds of different volatility at an aging time of 9 months

Fig. 19

DEPTH PROFILE OF METHYL IODIDE-131 IN THE TESTING OF USED AND REGENERATED ACTIVATED CARBONS

by

Victor R. Deitz
Naval Research Laboratory
Washington, DC 20375

Introduction

The capability of nuclear grade carbons to remove radioiodine is slowly reduced after being placed in service. The methyl iodide-131 penetration measurements to be described in this paper were obtained for such used carbons obtained from different nuclear power installations and after known exposures to flows of outdoor air.

The retention of radioiodine within a carbon bed can be characterized by a depth profile, i.e., by the radioiodine count as a function of depth along the line of flow. In the measurements to be described the sample was thoroughly mixed and the carbon bed (50.8 mm) divided into four equal sections, separated by disks of glass filter paper. After the test each section was removed and counted; the depth profile was calculated from the counts of the four sections. The percent penetration was also determined after mixing the four sections thoroughly and counting the mixture with the same geometry as the backup beds. The calculation was as follows:

$$P = \frac{\text{counts of the backup beds x 100}}{\text{counts of total sample + backup beds}}$$

It will be shown that the depth profile contains information that is additional to the penetration and can be used as an index of the need to replace the carbon in the carbon adsorber.

Experimental

The sources of the used carbons are given in Table 1. All carbons were significantly degraded as judged by the penetration of methyl iodide-131 determined at 30°C and 95% RH. The samples were not prehumidified before the determination. Some of the carbons had been exposed to an unfiltered flow of outdoor air at different locations for known duration (1). The kinetic parameters of the ASTM D-3803 test procedure (2) were used in these measurements.

Table 1. Sources of Used Carbons and Penetration of Methyl Iodide-131 at 30°C and 95% RH.

NRL Number	Exposure	DURATION (months)	% Penetration 30°C
5337	After continued use in Carbon Adsorber	22	49.2
5338	In use in Spent Fuel Vent. System	6	26.7
5339	Removed from Spent Fuel Vent. System	~36	53.5
5340	Removed from Spent Fuel Vent. System	~36	58.6
	Continuous Flow of Unfiltered Outdoor Air		
5226	KI BC 727	15	6.0
5229	KI BC 727	32	22.4
5239	KI BC 727	40	9.5
5214	KI + TEDA BC 787	9	10.0
5235	KI + TEDA BC 787	27	10.1
5151	KI + TEDA KITEG-II	24	38.9
5170	KI + TEDA G-615	20	23.4
5169	5% TEDA on Coal Base	20	76.6
5208	5% TEDA on Coal Base	5	24.4
5208A	5% TEDA on Coal Base	5	30.6

The percentage of total counts of methyl iodide-131 retained in each section are given (Table 2) arranged in the same sequence as the samples in Table 1. The ordering of counts in the inlet to outlet sections decreased in many cases; it remained constant in others, and actually increased for five samples exposed to continuous flows of outdoor air. The behavior shown, Figure 1, for a 5% TEDA impregnated carbon had been exposed for 163 days in Simi Valley, CA (May 14 to October 24). Two determinations were made in order to check this surprising result. The behavior is quite different than that reported for new carbons where the penetration is exponential with depth.(3) Since the carbon test sample was uniformly introduced, the radioiodine-131 must have migrated from the inlet to the outlet

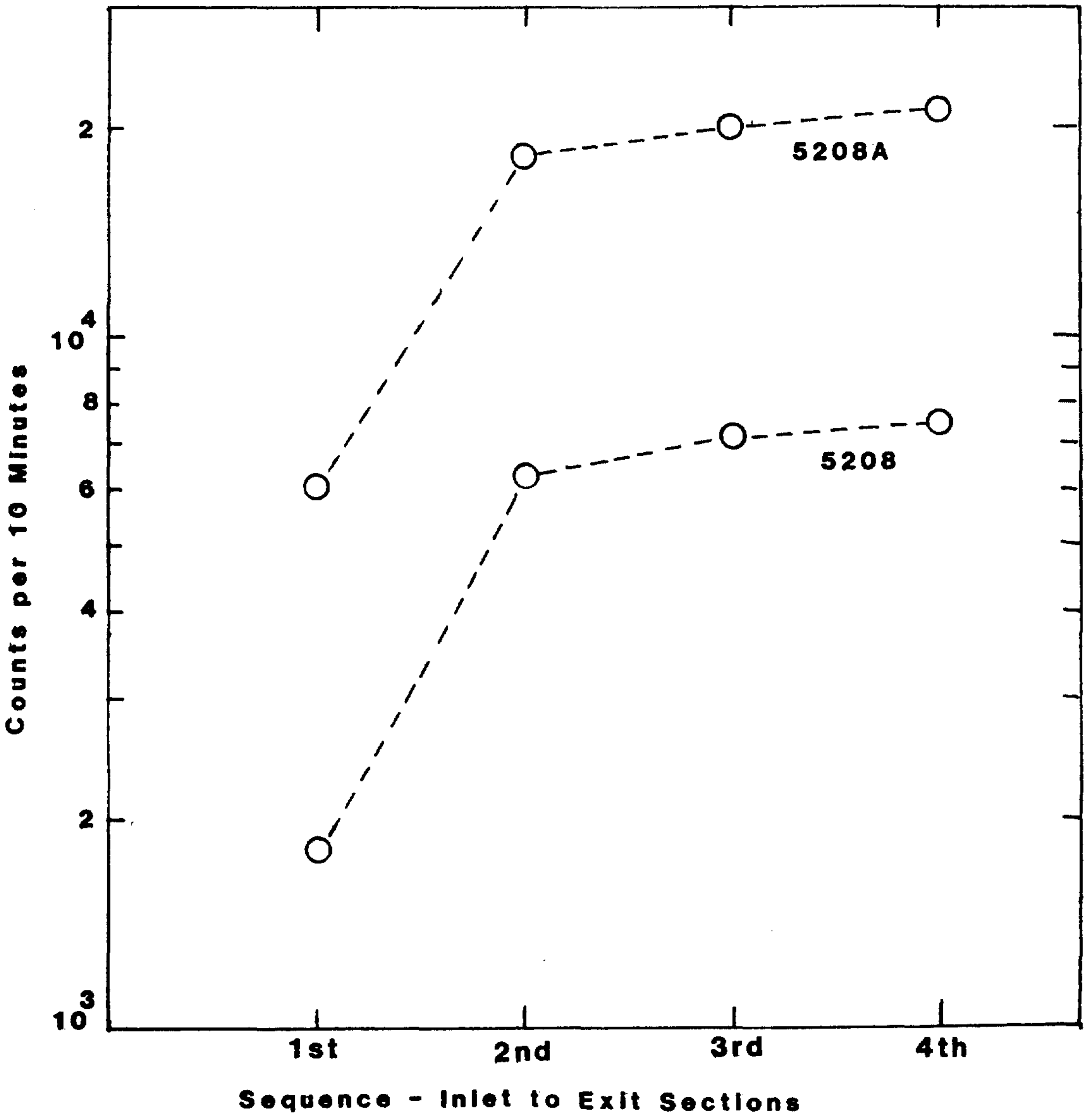


Figure 1. Migration of Methyl Iodide-131 During the Test of a 5% TEDA Charcoal After 163 Days Exposure to an Outdoor Airflow (Simi Valley, CA).

section during the test period. The concentration of methyl iodide-127 in the dose is approximately 10^9 times that of methyl iodide-131 and together with the iodine -127 in a KI impregnated carbon constitutes for new carbons a very large mass-action factor in favor of a successful iodine isotope exchange. However, the radioactive iodine can migrate in used carbons.

Table 2. Percentage of the Total Radioiodine-131 Counts in the Four Layers.

NRL Test	Penetration %	Layer 1	Layer 2	Layer 3	Layer 4
5337	49.2	48.2	24.2	16.1	11.5
5338	26.7	35.9	28.6	19.1	15.7
5339	53.5	25.0	24.1	25.1	25.8
5340	58.6	24.7	25.6	25.7	24.0
Continuous Flow of Unfiltered Outdoor Air					
5226	6.0	44.6	24.2	21.0	10.2
5229	22.4	42.8	23.5	18.0	15.7
5239	9.5	54.3	24.8	12.7	8.3
5214	10.0	16.5	46.5	21.7	15.2
5235	10.1	44.8	22.6	23.6	9.1
5151	38.9	1.6	27.7	37.3	33.4
5170	23.4	2.3	28.8	39.2	29.7
5169	76.6	0.9	4.6	7.1	87.5
5208	24.4	7.9	27.8	31.2	33.0
5208A	30.6	9.3	27.8	30.7	32.2

Discussion

A carbon adsorber is exposed to a variety of environmental contaminants during a long service life without regeneration. The slow degradation of the iodine trapping is due in part to the loss of adsorption, to the loss of chemical reactivity, and above all to the loss of iodine isotope exchange efficiency.

The contaminants that do accumulate can influence a methyl iodide-131 penetration test in several ways. Those that are involved in an iodine isotope exchange mechanism can be divided into two main groups:

Group I: Regenerated by chemical reduction.

Group II: Exposed to high molecular weight compounds and cannot be regenerated.

It has been found that many used carbons of Group I, but not all, can be regenerated by a chemical reaction with hydrazine (N_2H_4)$^{(4)}$. In one mode of degradation the iodide ions (the site of the iodine isotope exchange) are oxidized with the formation of compounds such as IO_x. These sites can be regenerated by a chemical reduction with hydrazine vapor at room temperature. An example of Group A (Figure 2) is a TEDA-impregnated carbon removed after 22 months service of

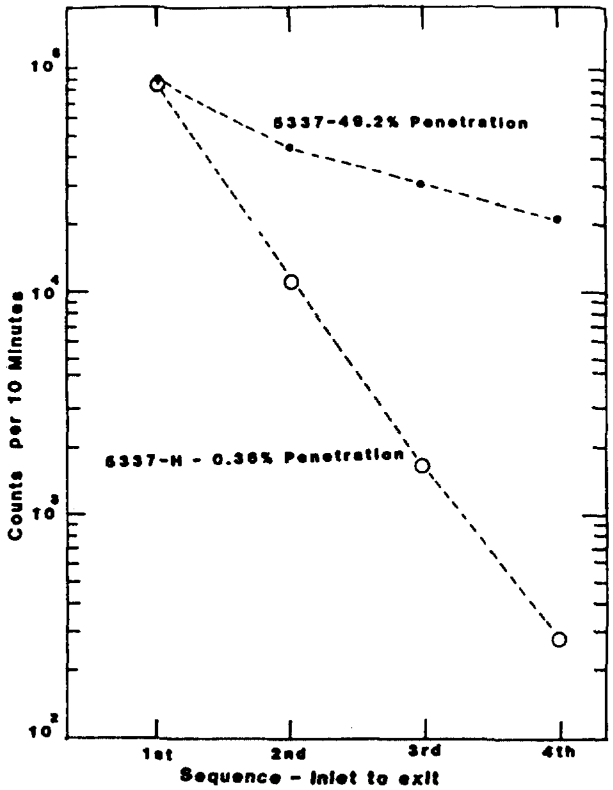


Figure 2. Depth Profile Through a TEDA-Carbon Removed From a Plant-Scale Carbon Adsorber (Before and After Hydrazine Treatment).

continuous use in a plant-scale carbon adsorber. The gradient of counts in the bed after regeneration is quite steep relative to that before regeneration.

Group II of used carbons are those that have adsorbed high molecular weight vapors during service, such as diesel exhaust gases, and these sites cannot be regenerated at room temperature by chemical reduction. The surface areas are very small and presumably the isotope exchange sites have been covered by the adsorbed vapors. One example (Figure 3) is for a carbon removed from the spent fuel handling ventilating system. There appears to be a point of no return for a heavily degraded carbon. The gradient of counts through the bed

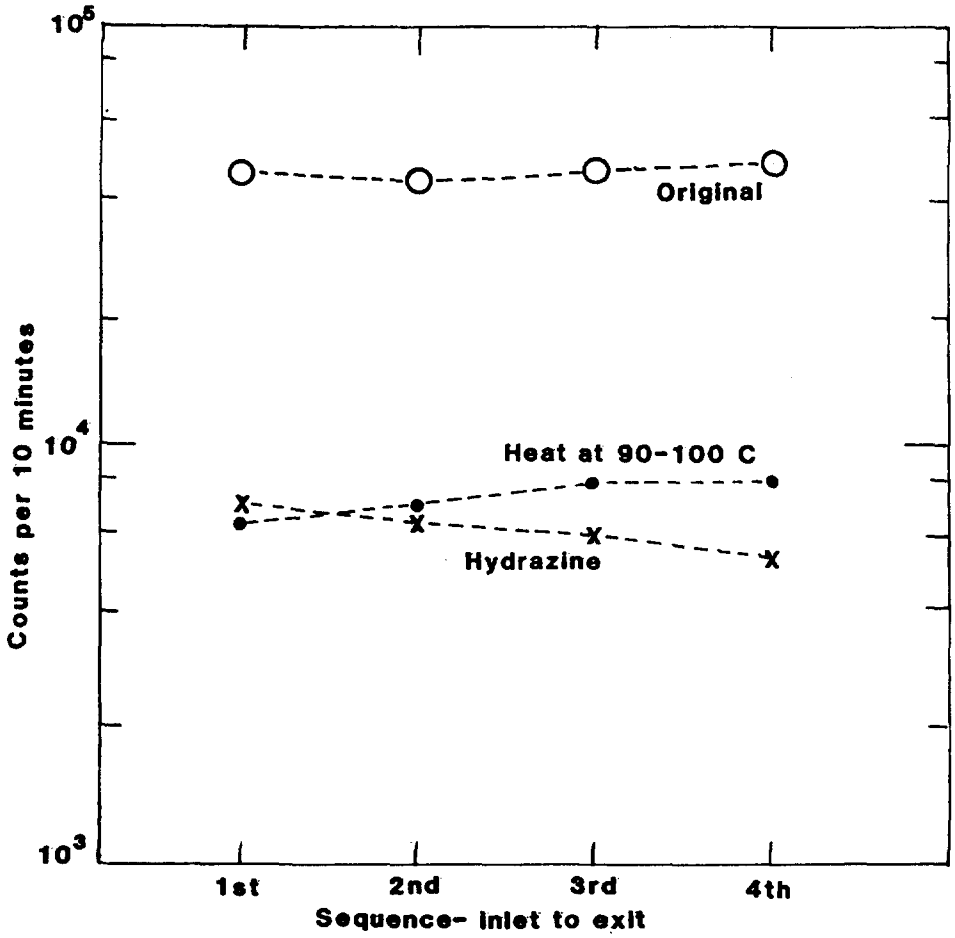


Figure 3. Depth Profile Through a KI-Carbon Removed from a Spent Fuel Ventilating System-Before and After Hydrazine Treatment and After Heating at 100°C for 48 Hours. (NRL 4339).

is obviously quite small. The sample (4339H) after hydrazine treatment was placed in the oven for about 50 hours (90-100°C). This furthered the degradation to almost completion, namely 84.3% penetration (Figure 3). Apparently, the mobility of the contaminants can further lead to the loss of the isotope exchange properties of certain used carbons.

In order to emphasize the uniqueness of iodine isotope exchange in the trapping of methyl iodide-131, three tertiary amine salts were prepared with methyl iodide-127 (Table 3A). Aliquots of the coconut shell charcoal (NACAR G 210-S) were impregnated with 2 wt.% of each salt. The methyl iodide-131 penetration was determined for each preparation and also the percentage of the total counts retained in each section (Table 3B).

Table 3A. Preparation of Methyl Iodide Salts of Three Tertiary Amines

	MP	Iodine	
Impregnant	°C	% calcd.	% obsd.
Quinuclidine · CH_3I	385	50.2	49.9
TEDA · 2 CH_3I	275	64.14	64.07
Hexamethylenetetraamine · CH_3I	205	45.04	45.0

Table 3B. Penetration of Methyl Iodide-131 and the Percentage of Total Counts in the Four Sections (Prehumidification at 95% RH for 16 Hours).

	Percentage of Total Counts				Penetration
	1	2	3	4	%
Quinuclidine · CH_3I	79.0	19.9	1.01	0.03	0.0
5299 TEDA · 2CH_3I	71.4	18.7	7.4	2.4	0.38
5307 " "	80.5	14.6	3.6	1.2	0.23
5307 " "	95.6	3.7	0.26	0.09	0.03
Hexamethylenetetra-amine · CH_3I	73.5	20.9	4.7	0.86	0.23

The measurements show a very high iodine isotope exchange efficiency. Since all of the tertiary amine was tied up in the salt and not available to trap methyl iodide-131, and the physical adsorption of carrier methyl iodide was reversible, the trapping was limited to an iodine exchange with the iodide ion of the impregnated salt. The gradient of counts within the test column was quite steep as previously reported for new carbons.(3) However, the slope of the log count vs depth was not linear for these impregnations (Figure 4) and very little I-131 was found in the outlet sections.

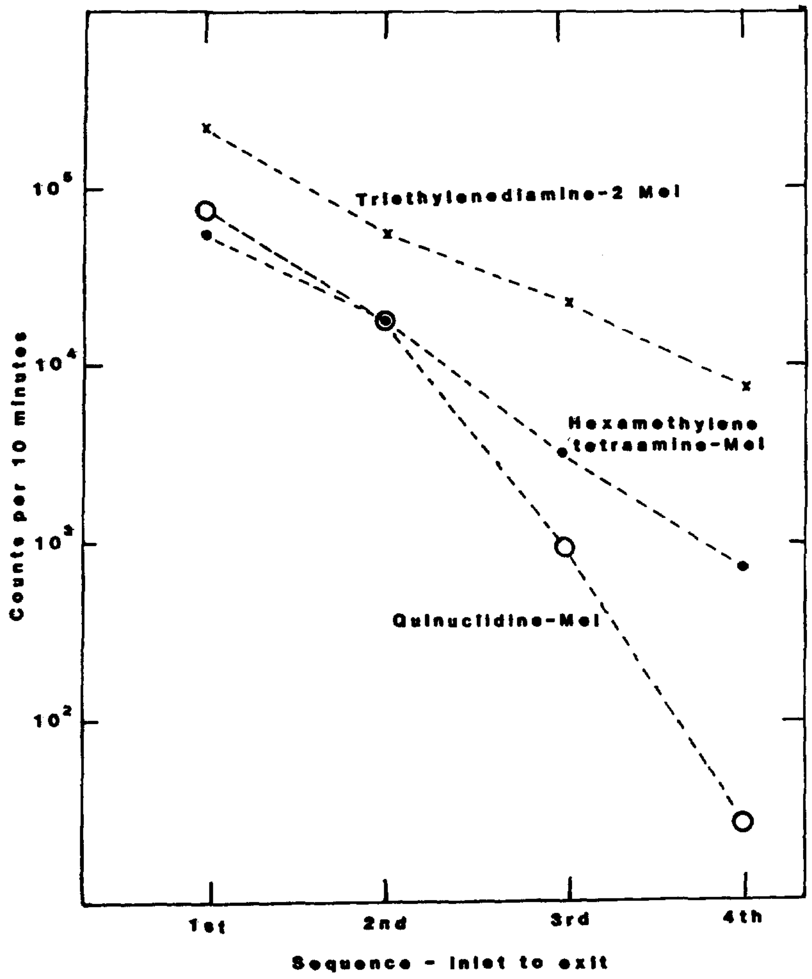


Figure 4. Counts per 10 minutes of Methyl Iodide-131 in the Four Sections of the Test Beds (Salts of MeI and Tertiary Amines on NACAR G-210S).

To determine if the methyl iodide-127 in the dose might react with either a KI or a TEDA impregnation, the adsorption of the vapor was determined. A KI-impregnated coconut shell charcoal (MSA 463563) and a TEDA-impregnated charcoal (S8S 5% TEDA) were used (Figure 5). The adsorption on the KI-carbon was Langmuir-type and that on the TEDA-carbon had a less-sharp knee and followed the Type II isotherm. Of greater importance is the fact that the methyl iodide can be desorbed from new carbons in the observed pressure range with no significant retention by either impregnation. The system was moisture-free in the above measurements. However, the adsorption of methyl iodide-127 on used carbons is not reversible in many cases.

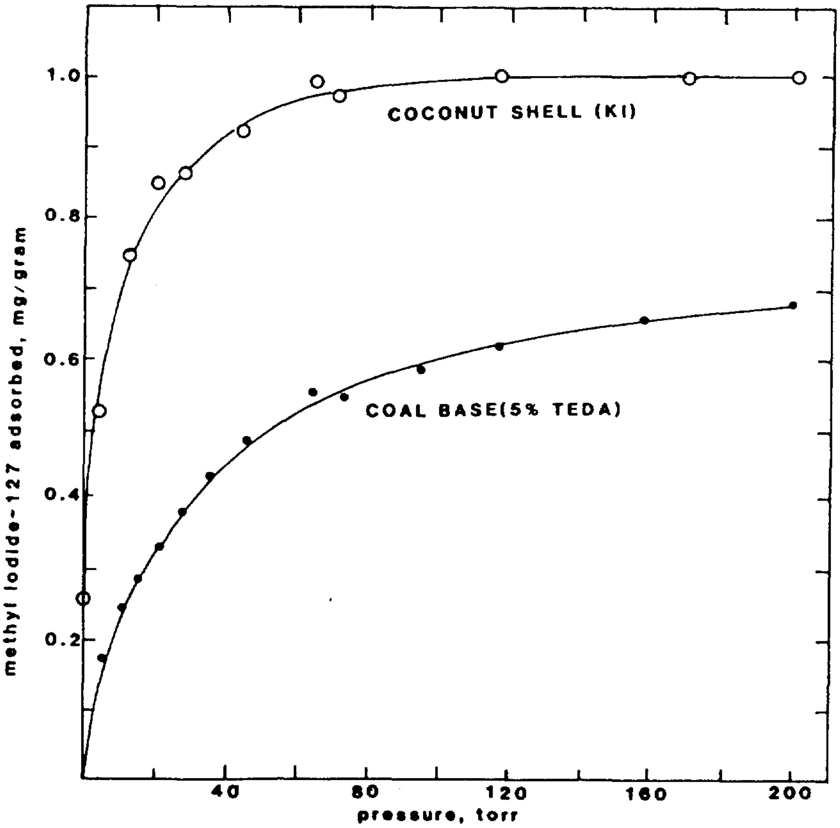


Figure 5. The Adsorption of CH_3I-127 Vapor at 31.7°C on Two New Impregnated Activated Carbons.

A mechanism is required to account for the migration of methyl iodide-131 to the lower sections of a uniformly-packed test column of used carbon. The possibility can be raised that chemical species are present that could react to form elemental iodine and thus move the trapped iodine-131 down the column as $I^{131} - I^{127}$. The changing surface composition of used carbons has developed as follows: First, the KI-impregnated sites have been oxidized during its service forming IO_X groups which do not enter into iodine isotope exchange. Second, in used carbons the acidity of the carbon surface has increased as shown by the low p^H of the water extracts. Third, in the relatively high concentration of methyl iodide-127, addition compounds to contaminants can form yielding iodide species at various places along the length of the column. The stage is thus set for the liberation of molecular iodine:

$$I^- + IO_X \longrightarrow I_2 + IO_{X+1}$$

The exchange reaction forming $^{131}I - ^{127}I$ can then move the activity down the column. Preliminary experiments indicate these reactions are present in testing used carbons.(5)

The BET surface area of a carbon (NRL 5338) in use in a Spent Fuel Ventilation system was $1040 m^2/g$ and this increased after hydrazine treatment to $1540\ m^2/g$. On a carbon removed earlier from the same system (NRL 5339) had a surface area of 670 m^2/g and did not improve with hydrazone treatment. Hence, there are varied and significant structural changes during the life of a used carbon.

Concluding Remarks

1. The depth profiles of CH_3I-131 in a carrier of CH_3I-127 show complex behavior in testing for the penetration of used nuclear-grade carbons. The carbons obtained from nuclear power plants and those from weathering in a continuous flow of unfiltered outdoor air show similar properties.

2. The depth profiles of the CH_3I-131 retained in the test beds of used carbons were either negative, zero, or positive for different samples. The isotope-exchanged iodine located in the entering section can definitely migrate down the bed during the dose period in the presence of methyl iodide-127. This does not happen for new carbons and the explanation must reside in the influence of certain contaminants that accumulate in the carbon.

3. Migration of I-131 takes place along the line of flow due to the formation of adsorbed elemental iodine from the adsorbed impurities. The exchange product ^{131}I -^{127}I can then move the activity down the column.

4. Obviously, one cannot improve the performance of a carbon adsorber until a satisfactory procedure is developed for testing used carbon. A test should reveal the residual life as well as the existing penetration. A used carbon from a carbon adsorber has become so, not only because of contact with water vapor, but after contact with the many local and atmospheric contaminants. The combined influence on the carbon and its impregnation can lead to profound

changes that must be taken into account in a test procedure. The influence of water vapor in itself does not have sufficient generality to be useful in evaluation of used carbons.

References

1. Deitz, V.R., "Effects of Weathering on Impregnated Charcoal Performance," NUREG/CR-2112, NRL Memo Report 4516, 132 pp., April 1981.

2. "Standard Method for Radioiodine Testing of Nuclear-Grade Gas Phase Adsorbents," D-28 Committee Standard D-3803-79, Sept. 1979, ASTM, 1916 Race St., Philadelphia, PA 19103.

3. Deitz, V.R., "Residual Depth Profile in the Evaluation of Granular Adsorbents," Nuclear Tech., 73, 96-101 (1986).

4. Deitz, V.R., "Charcoal Performance Under Accident Conditions in Light Water Reactors," NUREG/CR-3990, NRL Memo Report 5528, 177 pp., January 1985.

5. Deitz, V.R., Romans, J.B. and Bellamy, R.R., "Evaluation of Carbons Exposed to the Three Mile Island Accident," Proc. 16th DOE Nuclear Air Cleaning Conf. 1500-1518 (1981).

DISCUSSION

RIGBY: I would like to ask your opinion. When we talk about the aging of carbon, two different things can happen. One is oxidation due to oxygen and humidity. The second is organic contaminants. The first has been studied in depth. But very little work has been done on what happens with organic vapors. Do you think there is any merit in putting a sacrificial guard bed of unimpregnated activated carbon in front of the impregnated carbon bed? I mention this in particular because the Central Electricity Generating Board, in England, following studies last year, are in fact doing that at one of their power stations.

DEITZ: The contaminants of the air are of constant supply and invariably lead to the degradation of nuclear-grade carbons by weathering. Organic vapors (from solvent spills, paint vehicles, and lubricant sources) are more variable. These vapors, of course, must be sufficiently volatile to reach the carbon filter. A guard bed of activated carbon is not a permanent sink for organic vapors physically adsorbed. For example, benzene vapor may be adsorbed by activated carbon at a low relative humidity (below 50% RH) only to be released by an air flow at a higher relative humidity. Also, a bed of activated carbon acts like a chromatographic column and the incident pulse of organic vapor will eventually be in the effluent during long service in a power station. It is dubious, in my opinion, if a guard bed of activated carbon would be cost effective when placed in front of the nuclear-grade carbon.

MULTIPLE USE FILTER CELL - COMPARATIVE MEASUREMENTS

H.-G- Dillmann, H. Pasler
Laboratorium für Aerosolphysik und Filtertechnik
Kernforschungszentrum Karlsruhe GmbH
Postfach 3640, D-7500 Karlsruhe 1
Federal Republic of Germany

Abstract

A new activated carbon filter cell has been developed that will yield higher decontamination factors for iodine and smaller leakages in the housing in comparison with other filter cell type units. This type of filter cell can be used several times by simply replacing the charcoal and the sealing.

The measured decontamination factors are given and compared with the values of other cells.

I. Introduction

In German nuclear power stations the relevant activated carbon filters are of the deep bed filter type. But filter cells are also used in small iodine filter systems such as for example in laboratory vent air. There are disposable filter cells and multiple use filter cells. The latter offer the advantage that they help to minimize the amounts of waste and hence require less expenditure for eliminating the smaller activity loadings to be expected.

The application of this type of multiple use reuseable filter cells implies a cleaning of the housing and mere replacement of the sealing and the sorption material.

Disposable filter cells and replaceable filter cells offered on the market have been investigated and examined with a view to removal efficiencies. It has appeared that some of the values obtained are far below the values to have been expected from the carbon used and the residence times evaluated. The plots of decontamination factors which, in part, run horizontally or have an extremely flat course while the face velocity undergoes variations, are an indication of mechanical leaks between the upstream and downstream filter sides.

II. The Problem

Considering these results, a new activated charcoal filter cell should be developed which should reduce the number of drawbacks listed above and produce higher decontamination factors than the multiple use filter cell.

As a first step, the greatest carbon volume possible should be enclosed, the carbon should be capable of the highest possible degree of utilization, and the pressure loss should be at a minimum. Carbon compaction in transit and during operation which cannot be ruled out, should be compensated for in order to avoid potential leakages.

III. Design Concept

A major improvement consisted in moving the lid for filling the filter cells to the rear (downstream side), in order to avoid leakages through the lid (Fig. 1). By appropriate design of the boundary zone it has been ensured that the air path through the filter cell is always equivalent to the thickness of the filter layer.

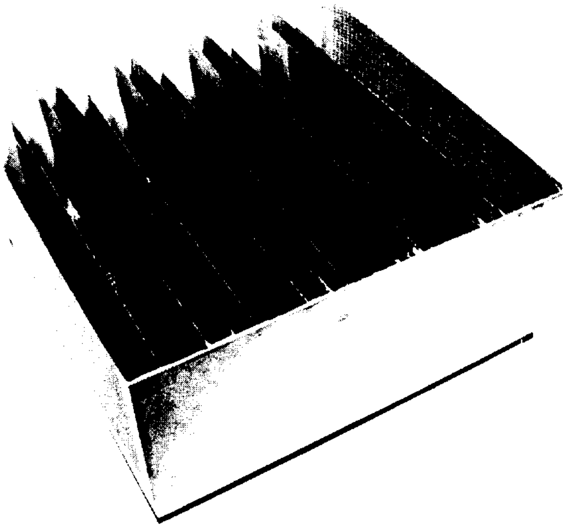


Fig. 1

For this reason, the slits have been arranged up to a distance equivalent to 2.5 cm border periphery for the case of 5 cm filter layer thickness (Fig. 2). The front faces of the V-pleats, except for the outermost entrance faces, are likewise slitted on the upstream and downstream sides in order to avoid that there are carbon zones not passed by the flow.

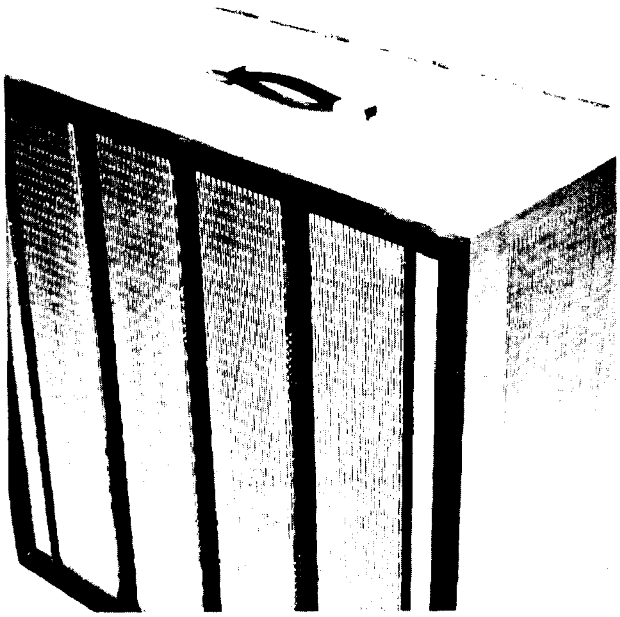


Fig. 2

For this reason, a permanent, elastic, air-transparent fleece has been placed on the lid side which on account of its high elasticity at the same time applies pressure onto the carbon (Fig. 3). Spacers are used to arrange the V-pleats in such a way that the distance between them is always the same. The spacers are equipped with threaded holes on the downstream side for lid mounting and pressing (Fig. 4).

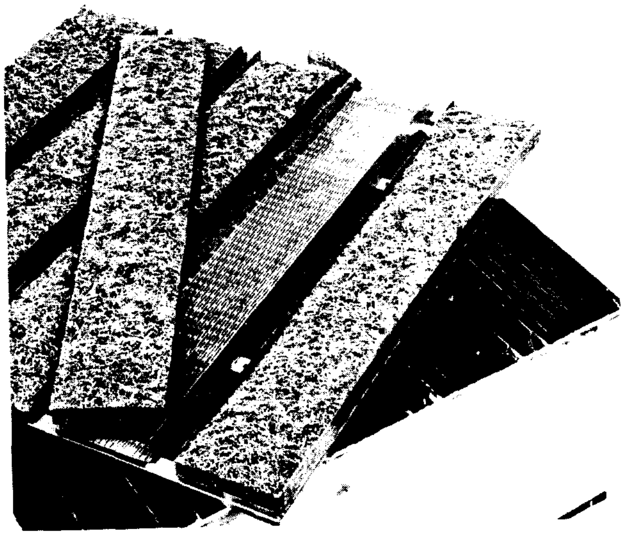


Fig. 3

The elastic material consists of rubberized hog's bristles which are permanently elastic up to 80 °C and cover the rear carbon filling holes.

The cell sealing is glued onto the cell and replaced in each refilling step. The cell is made completely of stainless steel and hence readily amenable to decontamination if necessary (Fig. 5).

The coal fill amounts to about 85 l of activated carbon whose granulation should not be smaller than 1.2 mm because for reasons of fabrication technology the slits are 1 mm in width. During the design stage particular attention was paid to avoiding any sharp edges which ensures that the carbon filters, like the HEPA filters, can be replaced in the plastic bag replacement mode, should the need arise.

The dimensions conform to the current filter cell sizes of 610 x 610 x 292 mm = 24" x 24" x 11.5". By an increase of the face area the pressure drop has been reduced to extremely low values and the volumetric flow rate has been increased, respectively.

Fig. 4

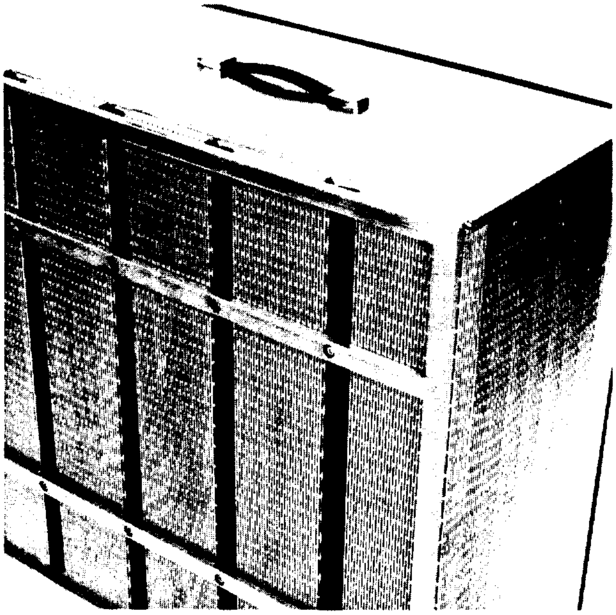


Fig. 5

IV. Test Equipment and Method of Measurement

The tests for determination of the decontamination factor were performed in the TAIFUN test bench under standard conditions at 1 bar, 30 °C, 70% relative humidity, using radioactive methyl iodide CH_3I-131 as the tracer /1/.

The decontamination factor can be calculated from the ratio of upstream air activity to downstream air activity which is determined with a Ge-Li detector.

V. Conducting the Experiment

The filter cells were tested for flows between 800 - 1500 m^3/h in the circulated air mode. The tracer is introduced into the central part of the piping, at several meters distance upstream of the test section (about 2 mCi within 5 minutes).

Upstream and downstream of the filter to be tested samples were collected from a gas stream using a perforated tube frame extending over the whole cross section of the test section, and the samples were transferred to activated carbon filters. The samples were measured and evaluated.

VI. Results

Fig. 6 shows the results of cells tested until now. Fig. 7 shows the improvements made, measured on the first prototypes.

It is to be seen, that the decontamination factors in the Karlsruhe Nuclear Multiple Use Cell are higher by the factor of approximately 10 than the decontamination factors of the welded and the multiple use filter cells, respectively, of different designs.

This is achieved, on the one hand, by the greater carbon volume, and, on the other hand, by the better utilization of the carbon caused by the greater face area in the increased perforated zone.

Comparison of pressure loss plots

Measurements on an industrial scale Karlsruhe Nuclear Multiple Use Cell yielded pressure drops of approximately 1.1 mbar at 800 m^3/h. These values are still below the level of pressure losses of welded disposable cells, despite the higher carbon volumes as can be seen in Fig. 8.

VII. Conclusions

The results show, that the new filter cell will give higher decontamination factors with a lower pressure drop using some simple construction ideas. It is expected, that the number of times such filter units can be reused will be greater than 20. So we use 2 sets of filter cells in our installation. One is under operation, the other set has time for decaying of the iodine and time, thereafter for cleaning and refilling with new charcoal.

VIII. References

/1/ WILHELM, J.G.; DILLMANN, H.-G.; GERLACH, K.;
"Testing of Iodine Filter Systems Under Normal and Post-Accident Conditions".
CONF 720 823 (1972), p. 434.

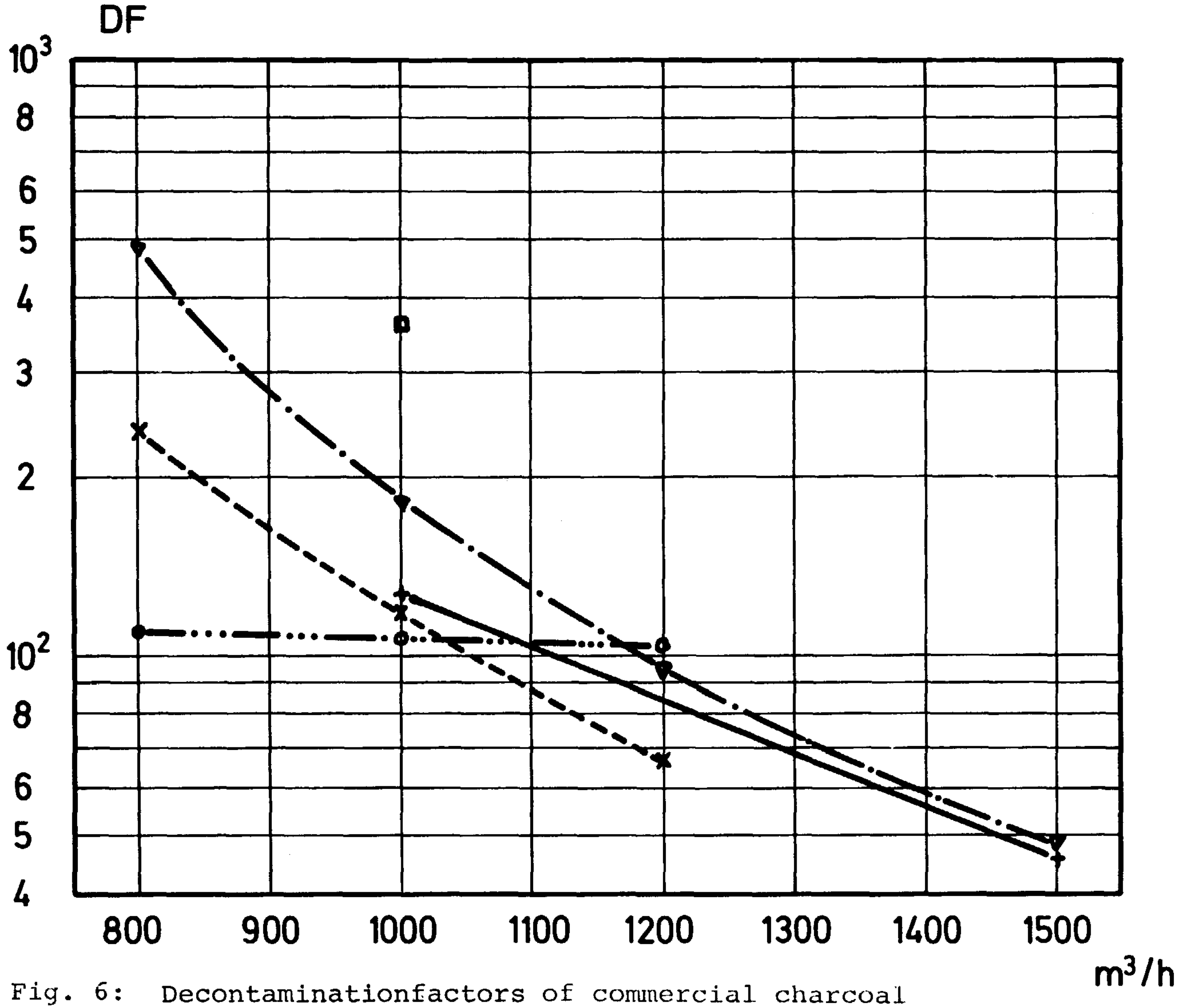
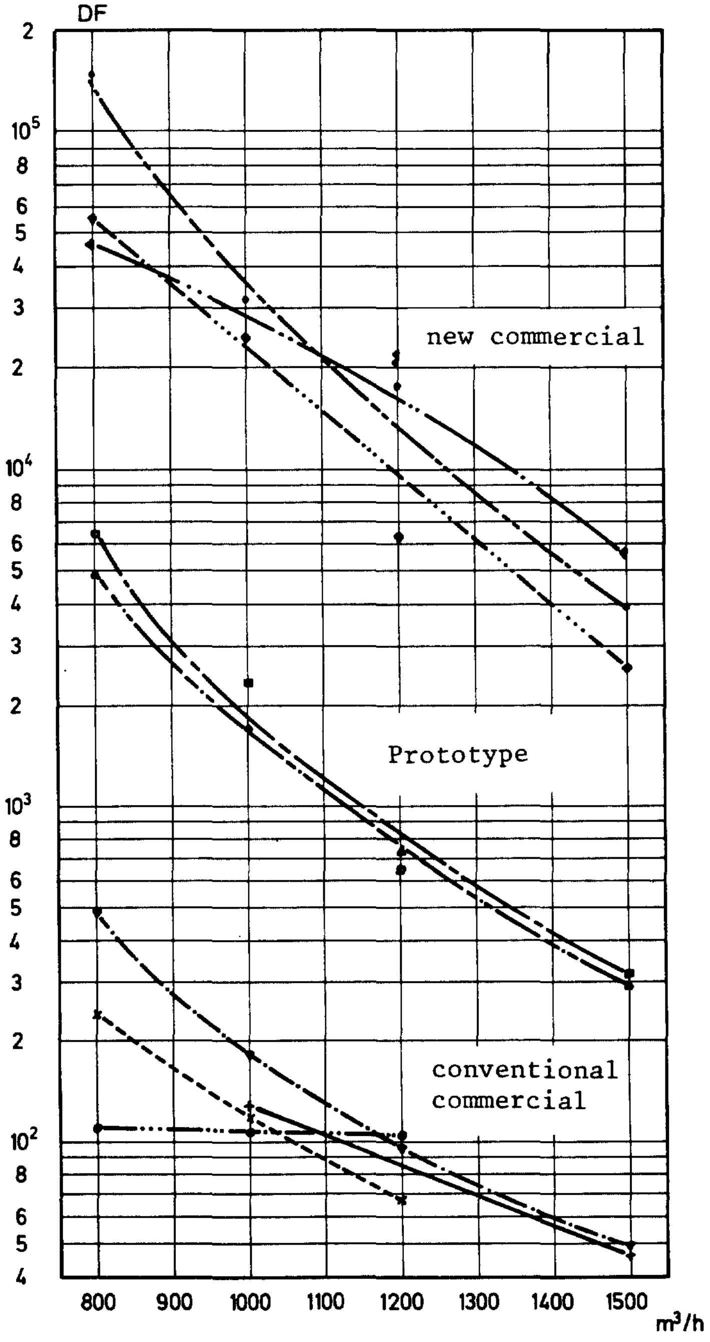


Fig. 6: Decontaminationfactors of commercial charcoal filters as a function of air flow rate

KfK LAF 86



KfK LAF 86

Fig. 7: Decontaminationfactors of conventional commercial and new commercial charcoal filter cells as a function of air flow rate. Each test with the same carbon.

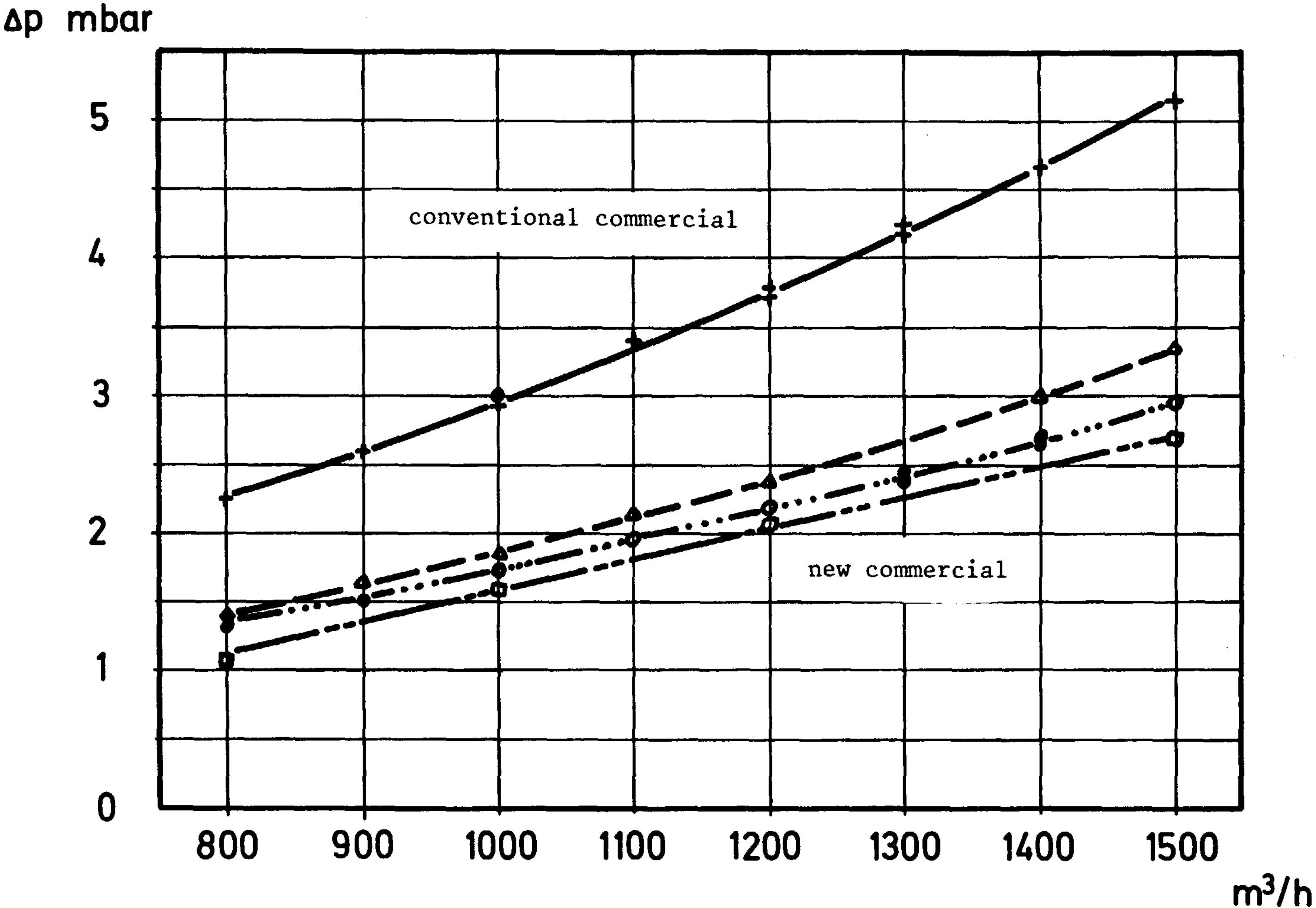


Fig. 8: Pressure drop of industrial and new charcoal filter cells as a function of air flow rate.

DISCUSSION

KOVACH: 1). Was there a difference in DF between the filter cell and an equal bed depth laboratory test?
2) If yes, do you attribute the difference to non-uniformity of flow or non-uniformity of packing?

DILLMAN: 1) Yes, there is a difference in the DF between the laboratory charcoal test and the cell test.
2) The difference may come from:
  a) the sealing of the cell.
  b) variations in the thickness of the charcoal bed, leading to nonuniform flows.
  c) non-uniformity of the charcoal size distribution during filling.
  d) the ratio between the enclosure surface of the housing duct and the charcoal volume, compared with a laboratory test or a deep-bed filter.

PERFORMANCE STUDY ON ACTIVATED CARBON FILTER SYSTEMS AT THE US ARMY CHEMICAL AGENT MUNITIONS DISPOSAL SYSTEM (CAMDS)

by DAVID J. GLADDEN P.E.
& CPT MIKE POST US ARMY

CAMDS Engineering, Tooele Army Depot, Tooele, Utah

Abstract

The US Army has been actively involved in the development and application of activated carbon filter systems for many years. In recent years, this work has expanded to include control of toxic gases generated in industrial activities. Ensuring that plant emissions were controlled and complied with all appropriate state and federal requirements has been a major part of this program.

This paper presents the results of a study of the performance of activated carbon filters used at the US Army Chemical Agent Munitions Disposal System (CAMDS). The filters (constructed with two adsorber banks in series) were installed, tested, and put in continuous service the late 1970's. A wide range of contaminants (e.g., paint solvents and common industrial compounds) as well as the chemical of concern (sarin) passed into the filter systems. Filter performance was monitored on a continuous basis for any detectable sarin breakthrough. It was expected that trace readings would appear and gradually approach a predetermined level at which time the filters would be serviced.

This study was prompted by the appearance of service level readings on a transient basis. An extensive set of data was collected and then charted in various ways to show the performance of the filter banks. The most useful presentation is one which plots the bank performance (C_i/C_o) against inlet concentration (C_i). The performance ratio was calculated when there were readings both upstream and downstream of a bank (a data set).

The plots, including data taken over an extended period, showed that high inlet concentrations tended to yield high filter efficiencies. When inlet concentrations were low however, the performance trended low.

These results are in agreement with general theory for adsorber performance. The operational data cover a wide range of concentrations, thereby extending the total data base over the wider range of high to very low concentrations found in many industrial plants.

However, the results conflict with the commonly accepted value of performance of 10^4 for carbon filter banks. The proper application of adsorbents must consider each compound; its unique characteristics, expected range of concentration. Careful investigation will become increasingly important when low concentrations of high toxicity or valuable compounds are of critical concern.

I. Introduction

The CAMDS facility is serviced by a number of filter systems to provide containment of any toxic vapors released as a result of the plant process. Each filter unit consists of a prefilter bank, a high performance particulate air (HEPA) filter bank, two activated carbon filter banks (in series), and another HEPA bank. A continuous (data averaged over one hour intervals) monitoring system is used to check the levels of toxic contamination within the filter train. The monitoring system is capable of detecting selected vapor contaminates in the parts per trillion(ppt) range. For the purposes of this study, the inlet, midbed and outlet concentrations of the toxic chemical sarin were tracked.

The performance of the filter systems was certified by testing to have a removal performance in excess of 99.99% per carbon bank. This corresponds to a reduction of the inlet concentration by a factor 10,000 (10^4). For the two banks in series a combined reduction of the inlet concentration by 10^8 was expected. The estimates of performance were assumed to apply for any inlet concentration.

Data from several filters at CAMDS was analyzed. The results of the study show that a single bank can not be expected to reduce the agent concentration by the 10^4 factor under all conditions at all times. The data indicate that the filter performance is widely variable and is expected to be a function of many factors. Of particular note was the significant effect of inlet concentration.

II. Discussion

The performance of high performance filters has been studied by many people. These studies have identified the influence of the most common variables including; adsorbent properties, gas flow rates, temperature, contaminant properties, moisture levels and time in service. A number of other minor items may also affect the performance. The point of this study was that the filters did not meet the expectations of the operating personnel and an explanation was desired.

Data Correlation. The monitoring data was examined in an effort to find a correlation. Many of the variables listed above were considered, but did not seem to provide an adequate explanation. The obvious items of time in service, accumulated bed loading and weather conditions were present and seemed to account for some of the events. However, there were enough unexplained events to create an interest in further investigation.

Data Selection. The analysis in this study was complicated by the large volume of data. All data had to be extracted by hand and cross matched with weather, operating and maintenance parameters. It was also complicated by the normal problems of erroneous or missing data.

In an effort to simplify the data extraction, several short time periods were selected for closer study. Within these time periods, only those data sets which met the criteria of having both inlet and outlet readings at the same time were extracted. This resulted in a large reduction in the data. In particular, it eliminated any data point for which there was a high inlet but no measurable outlet. This feature of the data extraction placed an upper bound (UB) on the data that can be expressed in terms of the maximum performance which can be reported. This upper bound is shown on the data plots. Efficiencies above this line exist, but can not be measured. In the case of sarin, this limit is approximately 1x10^{-5} mg/m^3. From this value it can be shown that depending on the inlet concentration (usually less than 10 mg/m^3), the reportable performance will be less than 10^6.

Data Presentation. It was decided to plot the data on log-log paper. Using the inlet concentration as the base and the performance for the ordinate, each data point was plotted. The results of this effort provided graphic evidence of the relationship between filter performance (efficiency) and inlet concentration. Although this data is related to a specific compound, the results should be representative of adsorber performance in general and therefore merits close study when applying adsorber technology.

Operational Data / Performance Ratio. Figures 1&2 are graphs of performance ratio versus inlet concentration for selected Type II filters (with 2 inch beds). The effect of accumulating challenge is seen by the slight shift (downward) of the lines representing different time periods (increasing time) in Figure 1. Figure 2 compares two filters operating in parallel on the same facility (but with different flow rates). Figure 3 is a graph for a Type III filter (with 4 inch beds) and shows the same general trend as seen in Figure 1. Figure 4 compares the characteristics of the TYPE II AND Type III. Figure 5 combines all the data to illustrate the similarity of the several filters regardless of their application or service conditions. The two factors which all units have in common are that they have been in continuous service and have been challenged with widely variable concentrations of sarin.

The performance ratio (PR) is calculated by dividing the inlet concentration by the outlet concentration of that bed. Since the CAMDS filters are constructed with two bed in series, the space between beds is labeled as the mid-bed. Table 1 illustrates this process using typical values.

TABLE 1. Calculation of Performance Ratio PR = C_i/C_o

Sarin Level			Calculated Eff.	
Inlet Reading	Midbed Reading	Outlet Reading	Inlet/Midbed $\frac{C_i}{C_o}$	Midbed/Outlet $\frac{C_i}{C_o}$
1.1x10^{-1}	BDL	BDL	NA	NA
3.6	9.4x10^{-5}	BDL	3.8x10^4	NA
2.1	8.8x10^{-5}	7.9x10^{-5}	2.4x10^4	1.1

All concentrations are in mg/m^3
BDL indicates Below Detectable Limits (approx. 1x10^{-5})
NA indicates that the ratio is Not Available because the outlet reading is BDL

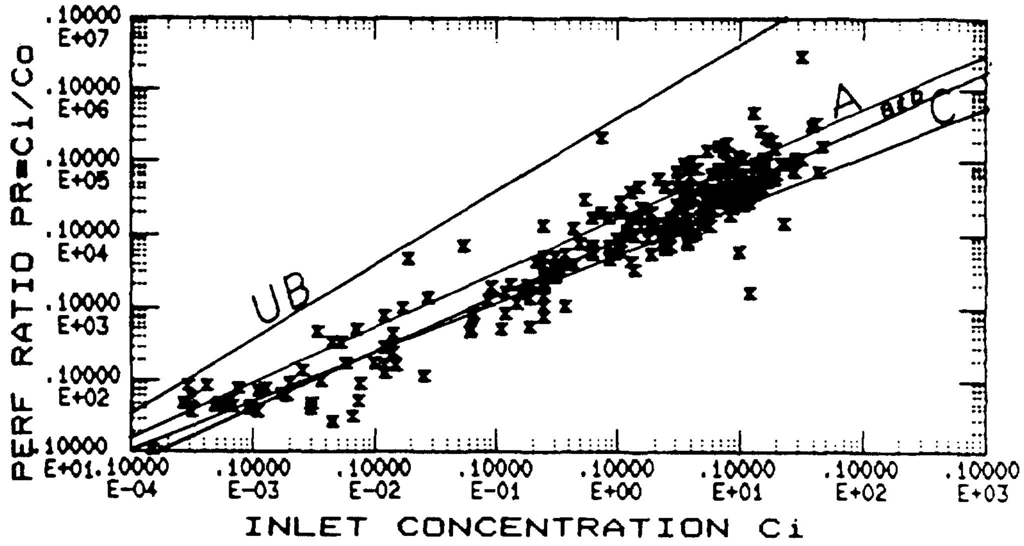


FIGURE 1. TYPE II ADSORBER (PDF)DATA SETS FOR; A=JAN 82 B=APR 82, C=JUL 82, D=COMBINED.

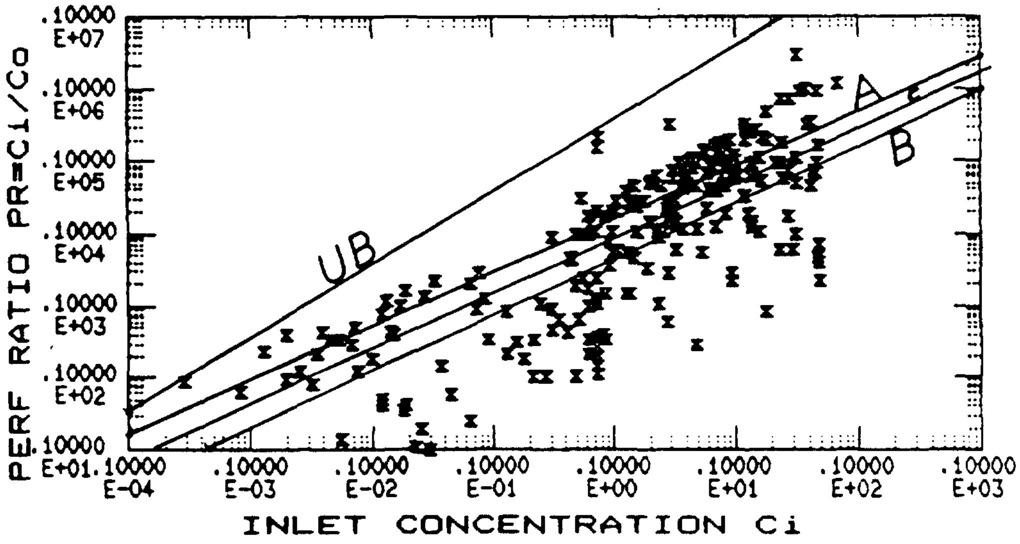


FIGURE 2. COMPARISON OF TWO TYPE II FILTERS IN PARALLEL SERVICE AND EQUAL TIMES (A=PDF, B=PPD, C=COMBINED).

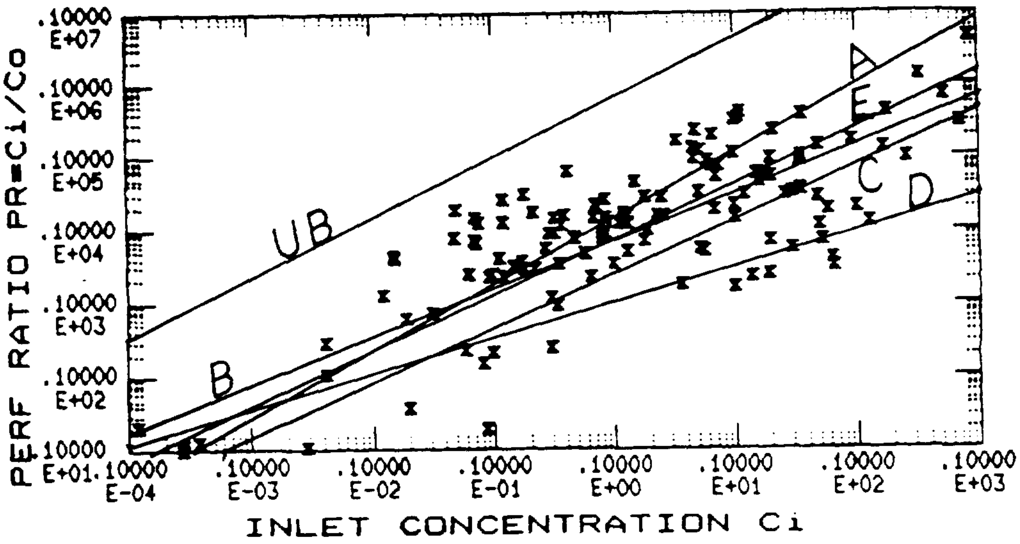


FIGURE 3. TYPE III FILTER (RLS) PERFORMANCE DATA SETS COVERING AN EXTENDED PERIOD; A=FEB 85, B=JUN 85, C=JUL 85, D=SEPT 85, E=COMBINED.

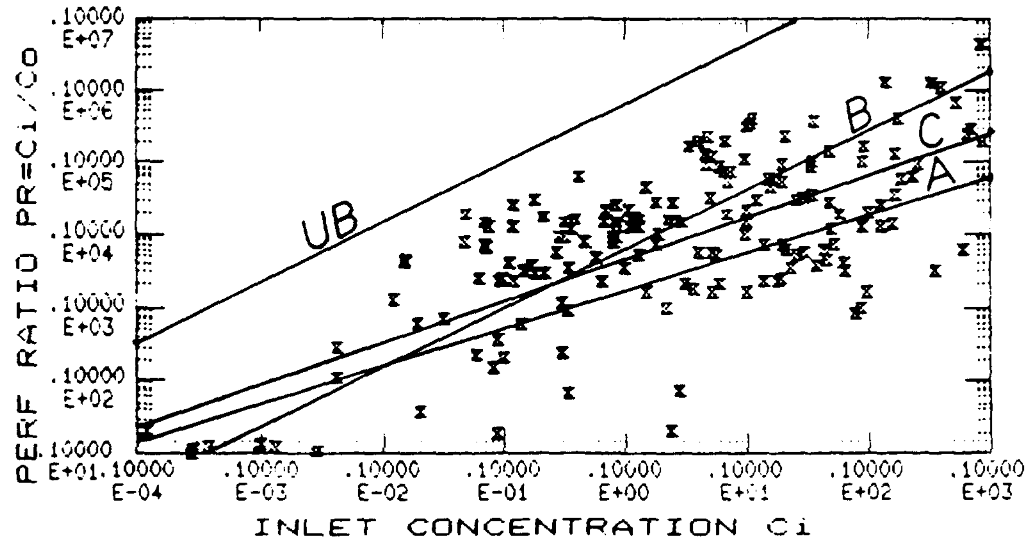


FIGURE 4. COMPARISON OF TYPE II (A=RII) AND TYPE III (B=RLS) FILTERS IN SIMILAR SERVICE (C=COMBINED).

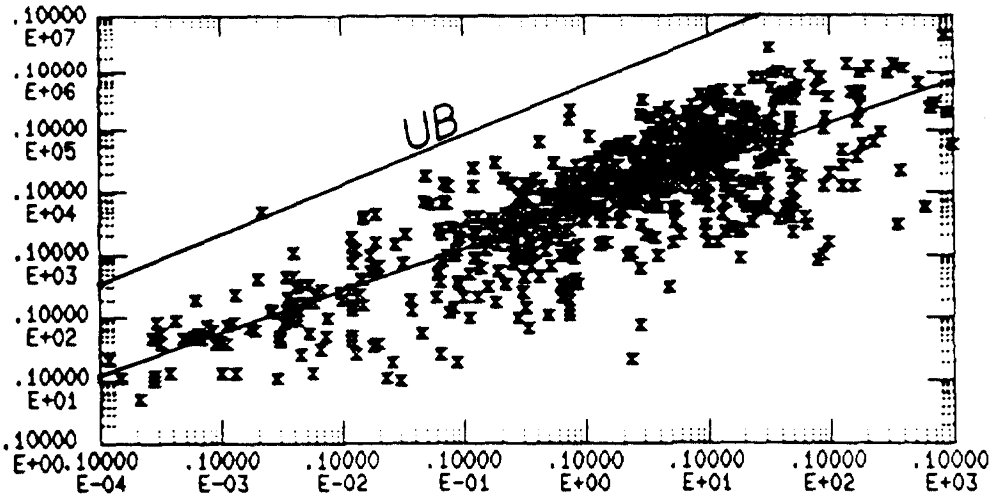


FIGURE 5. COMBINED DATA FOR TYPE II AND TYPE III FILTERS; A=PDF, B=PPD, C=RLS, D=RII, E=LIC, F=COMBINED(737 pts).

Filter Performance Extrapolation. The data points on the graphs show the general trend of decreasing performance ratio at lower inlet concentrations. The lines shown were fitted through the points using the least squares method. The fact that some of the data sets are quite small and contain only those points which met the screening criteria helps explain the wide scatter of the data. However, when there are enough points, the trend becomes apparent.

The equation for this line is expressed quite simply by:

$$PR = C_i/C_o = A(C_i)^B$$

Values for A and B were developed for the selected data sets. Table 2 shows the results of this exercise.

Table 2. Line Fit Coefficients For Combined Data

Figure #	A	B	# of Points
1 (Type II)	5026	0.76	286
2 (Type II)	4019	0.83	231
3 (Type III)	3966	0.72	139
4 (Type II/III)	2029	0.58	188
5 (Type II/III)	3153	0.69	737

At inlet concentrations ranging from 1×10^0 mg/m^3 and higher, the performance ratios were generally 10^3 or greater. This corresponds to the bulk of experience in previous performance estimates. At lower inlet concentrations, the performance ratio is seen to decrease significantly. This raises the question of why there are no data points wherein the inlet was high and there was a low PR. It can be seen that this is never allowed to occur because this represents the condition in which the bed is saturated and has exhibited a massive breakthrough. Usually, the filters are changed before this happens.

Filter Service Life. With the general understanding that the life of a filter is limited, it remains for us to develop a means of predicting the service life of a filter system. Since it has been shown that this life is dependent on the type of service it sees, we should be able to apply the existing theory for adsorbents in this effort. However, it is also apparent that the expected operating conditions must be included as a fundamental part of this analysis.

The performance of carbon filters (expressed as PR) will decrease over time as the carbon matrix becomes loaded with contaminates. The data shown here does not directly show this. However, the data used for Figure 1 & 3 has been broken down into subsets (of time) and then a line fitted to each subset. These lines (A,B,C,etc) illustrate the effects of time on the filter performance (a decreasing PR). Since the filters were in continuous operation, with the inlet challenge varying over a wide range, the data sets represent a combination of adsorption and desorption occurring simultaneously. These processes were being influenced by changes in weather, plant operations and time. They show that given any filter with an indeterminate amount of loading, and letting the process variables drift as they will, the PR of the filter will roughly correspond to the inlet concentration. It should not be unexpected to see PR numbers ranging from greater than 10_4 to less than 1.

III Factors Affecting Filter Performance

It has been shown in many studies that a number of factors affect the overall performance of carbon filters. These include temperature, pressure, challenge concentration, accumulated loading, humidity, the presence of other contaminants, etc. The outlet concentration (of any species of interest) is a complex function of all these factors.

The carbon adsorbers described in this paper are designed in accordance with standard industry practices. The air velocity through the beds is about 40 ft/min. The tray type filter elements have a standard 2 inch bed depth which gives about 0.25 seconds of residence time. The Type III filter with its 4 inch beds would have a 0.5 second residence time (per bed) at rated flow. As a matter of convenience, to allow balancing of the associated ventilation systems, all of the filters were operated below their rated flow. In some cases they were operated at less than 50% of rated flow. This was expected to enhance their performance and extend their service life. The derating does not appear to have had much effect on the overall performance or service life.

The results presented in this paper illustrate the complex nature of the processes occurring within the filter. They extend the experimental data into regions where the adsorption/desorption equilibrium phenomena become a controlling factor.

IV Comparison With Previous Kinetic Studies

Kinetic studies of adsorption of chemical agents using carbon filters have been performed by the US Army at Edgewood Arsenal for many years. For these previous studies, the inlet concentrations have ranged from $5x10^2$ to as high as $6x10^3$ mg/m^3. The operational performance ratios were over 10^4 as previously expected.

As technology has improved and the environmental regulations have become more stringent, the operational limits of processing plants has decreased significantly. In 1985, the inlet challenge from newly developed processes was generally less than 10 mg/m^3. On the other hand, the allowable outlet concentrations had been reduced to $1x10^{-4}$ mg/m^3. This is just above the "best technology detectable limit" of $2.7x10^{-6}$ mg/m^3. It is also several orders of magnitude below the values previously used to predict filter performance in this type of service. It is therefore reasonable to assume that previous estimates of adsorption capacity, the adsorption rate constants, and the equations developed for predicting breakthrough time are suspect at these lower operating ranges.

V Conclusions

First, filter performance has been shown to be a function of inlet concentration. The inlet concentration integrated over time accounts for the total accumulated dosage. As this load increases, the contaminant (through desorption) starts to appear in the outlet stream. Although the scatter of the data points is quite broad, the trend of the data supports this conclusion. By tracking this trend, it should be possible to anticipate the need for service of the filter system.

Secondly, at inlet concentrations greater than 10 mg/m^3, the performance will generally be greater than 10^4. There are times however when due to combinations of factors discussed here that the performance ratio will be reduced to less than 10. It was expected that the type III filter with its 4 inch beds would outperform the type II units. However, the increased bed depth available in the type III units does not appear to represent a significant barrier to low concentrations passing through, even though the deeper bed will contain significantly more contaminant at high concentrations. The design of a filter bed to retain low level concentrations will require greater attention in the selection of adsorbents and flow rates.

The work reported here is part of a development program aimed at the safe disposal of outdated military munitions. In this respect, the Army has insisted on the highest standard of performance. But it has been able to achieve this only by constant monitoring and judicious timing of carbon bed changes.

VI Acknowledgments

This paper is the result of a long term study in the application of activated carbon to the treatment of process air resulting from the US Army CAMDS program at the Tooele Army Depot, Tooele, Utah.

DISCUSSION

GUEST: You didn't tell us what chemicals you were removing. Can we assume that the mechanism is physical adsorption rather than chemical reaction?

GLADDEN: We used unimpregnated carbon of a nuclear grade, the same grade that is specified in N509 and N510. The primary mechanism is physical adsorption. These were organic phosphates. Chemically the name is Sarin, nerve agent GB. It is not classified. It is to our advantage to use standard nuclear grade carbon because it is readily available. The Sarin compound breaks down with time. At low concentrations (C_i), the desorption of some compound is expected and this increases with time and accumulated total dose.

KOVACH: By the way, organic phosphates also react well with methyl iodide, so if you are interested, you can evaluate your used carbon as a feed stock for methyl iodide.

CLOSING COMMENTS OF SESSION CO-CHAIRMAN KOVACH

I would like to summarize by saying that there are still a lot of things we don't know. We have iodine forms that we have not identified, we have erratic behavior of various impregnated carbons that act one way at one time and differently at another and we are not exactly sure why. I think we still have to do a lot more learning before we can really educate others. The one thing I can plead for is an open information exchange between people so we can find out who is doing what, because one lifetime is not long enough for one place to generate all the information that is still needed. This type of cooperation is needed. I think we have to learn from each other as much as we teach each other. This may be a very unconventional summary, but that is my message.

SESSION 4

FILTERS, FILTER TESTING

TUESDAY: August 19, 1986
CHAIRMEN: H.J. Ettinger
R.C. Scripsick
Los Alamos Scientific Lab.

THE EFFECTS OF HIGH RELATIVE HUMIDITIES ON HEPA FILTER MEDIA
B. Normann

HEPA-FILTER BEHAVIOR UNDER HIGH HUMIDITY AIRFLOWS
C.I. Ricketts, V. Ruedinger, J.G. Wilhelm

CALIBRATION TESTS OF A LASER FLUORESCENT PARTICLE SPECTROMETER
Ph. Mulcey, P. Pybot, J. Vendel

A NEW LIGHT SCATTERING SPECTROSCOPY TECHNIQUE FOR RAPID ASSESSMENT OF FILTER MEDIA
Y.W. Kim

HEPA FILTRATION AND MONITORING SYSTEM FOR AN UNDERGROUND NUCLEAR WASTE REPOSITORY
P.S. Parthasarathy, J. Shome

MEASUREMENTS OF REMOVAL EFFICIENCIES PERFORMED ON POWDER METAL AND FIBER METAL CARTRIDGES TO BE USED IN URANIUM ENRICHMENT FACILITIES AND GLOVEBOX EXHAUST DUCTS
H.-G. Dillmann, W. Bier, G. Linder, K. Schubert

EXPERIMENT ON A MULTILAYER TYPE AIR FILTER FOR THE FILTRATION OF SODIUM AEROSOL
N. Otake, O. Nozaki

DEVELOPMENT AT THE KARLSRUHE NUCLEAR RESEARCH CENTER (KfK) OF REMOTELY OPERATED FILTER HOUSINGS AND FILTER ELEMENTS FOR REPROCESSING PLANTS
K. Jannakos, H.J. Becka, G. Potgeter, J. Furrer

OPENING COMMENTS OF SESSION CO-CHAIRMAN SCRIPSICK

This session promises to be a truly global look at the status of filter performance research, and the development of filter test methods. We have papers from Japan and Germany on the performance of high efficiency filters, and papers from Sweden and France on filter testing. As at the 18th Conference in Baltimore, the work being carried out in West Germany on extreme condition filter performance is well represented. We were reminded earlier this year of the importance of such filtration studies with an airborne radioactive material release from the Chernobyl plant. There are predictions that indicate that thousands of extra cancer cases may result from this release. High efficiency air filtration systems are often the last barrier to such releases. The proper design and operation of these filtration systems to a large extent depend on our understanding of filter performance, and the application of meaningful filter performance tests. The presentation of papers from United States and France on filter testing, as well as the activity in this area in Great Britain and Australia, indicates a worldwide interest in filter performance testing. Designing a meaningful performance test requires a broad understanding of filter performance. During this session I will be interested to hear from our speakers how they designed their filter test systems and how they used information on filter performance to design these systems. I will also be interested to hear what information the filter performance authors will have for those performing filter testing. we expect to hear about new and novel methods from the experts in filter testing and in filter performance.

THE EFFECTS OF HIGH RELATIVE HUMIDITIES ON HEPA FILTER MEDIA.

Bård Normann
Studsvik Energiteknik AB
S-611 82 Nykoeping
Sweden

Abstract

The effects of high relative humidities and water on HEPA glass fiber filter media have been investigated.

The experiments also comprised exposure to pure steam for some hours at elevated temperature.

After exposure samples of filter media were dried and tested with respect to particle penetration and tensile strength.

The penetration measurements were carried out with DOP aerosols. The results showed a tendency to increased penetration when exposed to high humidities at elevated temperature.

The tensile strenght of the filter material decreased to about 40 percent of its original value after being exposured to flowing steam for 4 - 5 hours.

As expected the media lost their tensile strength in flowing steam (100 degr C) the strength decreased almost to zero.

Exposure for a few hours to approximately 100 percent humidified airstream at ambient temperatures did not cause any significant changes in the particle retention ability.

The strength was also uneffected by high humidities at ambient temperatures.

No damage to the fibres could be seen on examination in a scanning electronic microscope.

I. Introduction

The introduction of combined carbon- and HEPA filters in the offgas and emergency systems in Swedish nuclear power stations has reduced the folding height considerably.

The depth, or folding height, of the filter, se Figure 1, has thus been reduced from between 200 and 300 mm to about 70 mm, for the HEPA part of a combined filter.

This results in a decrease in the bending strength an thus a reduced tolerance with regard to higher pressure drops. A considerable increase in the pressure drop and reduced structural strength have also been obserwed across particle filters after loading to high humidities and then drying them;

this occurred in the filter material. Steam leakage, either small or large amounts in boiling water reactors, increases the risk for filter failures (3).

The aim with the work reported here (according to (4)) has been to gain a better understanding for the risk of filter failure in triple filters, or other similar installations; and also to determine the causes for filter failure as well as the effects of humidity on

- the structural strength of the type of HEPA filter incorporated in triple filters
- the penetration of DOP aerosol
- the structure of the filter material

II. Test programme

Types of material and filters

Two different filter material which are currently of interest for HEPA filters were studied. The materials are used as the filter medium in the off-gas and accident ventilation systems in Swedish nuclear power stations.

Both materials are glass fibre and are said to be resistant to damp, that is to say they can resist 100 % relative humidity during operation.

The fibres have been treated so as to resist the growth of bacteria, to be flame resistant and to withstand continual operation at 120°C.

One full scale HEPA filter, 610 x 610 mm was tested. It was intended for the triple filter installation, see Figure 1.

The filter comprised 70 pleats with aluminium separators and a pleat depth of 67 mm. The filter packet was housed in a plywood frame and neopren rubber glue was used as the filler material.

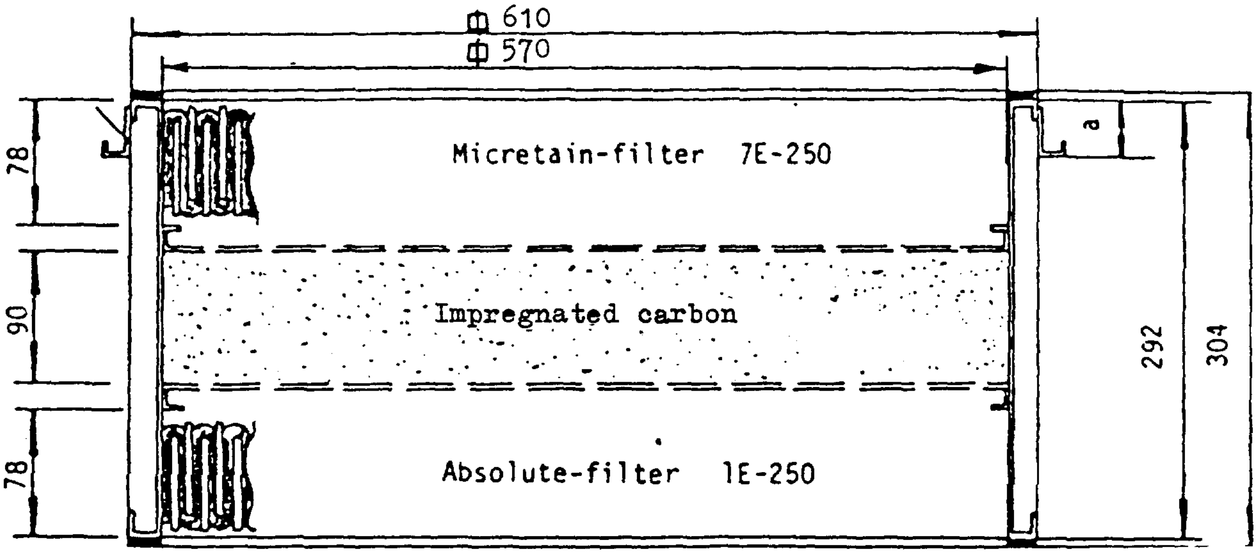


Figure 1. Exempel of a triple filter, manufacturer Camfil. Measurements in mm.

Test of filter material

Test were performed with filter material in damp air at between about 50 and 100 % RH in the temperature region 20 to 100°C, including pure steam.

The following material properties were studied

- the increase in penetration as a function of exposure to steam
- changes in the fracture strength as a function of time for steam exposure
- structural changes after exposure in moist air and steam.

Tests of filter material

Test environment	Material type 1	 type 2
Varying humidity at ambient temperatures	x	
Moisture at elevated temperatures	x	
Moisture att 100°C	x	x
Condensed steam	x	
Water saturated	x	x
DOP	x	x
Mechanical strength	x	x

Full scale filter tests

Tests performed on full scale HEPA filters:

1. Week-long tests at about 90 % RH.
2. 3 day tests at about 100 % RH.
3. Tests with water saturated filters.

III. Test and measurement equipment

Tests of filter material

The equpment shown in Figure 2 was used to perform the material tests in moist air and steam. The temperature, humidity and flow rate can all be controlled.

In order to prevent condensation that part of the equipment containing flowing steam or a mixture of steam and air is insulated.

Temperature and flow rate regulation is manual, based on resistance control and valves. The temperature is measured using a wet and dry bowl mercury thermometer. The flow rate is measured using a Fisher & Porter rotameter.

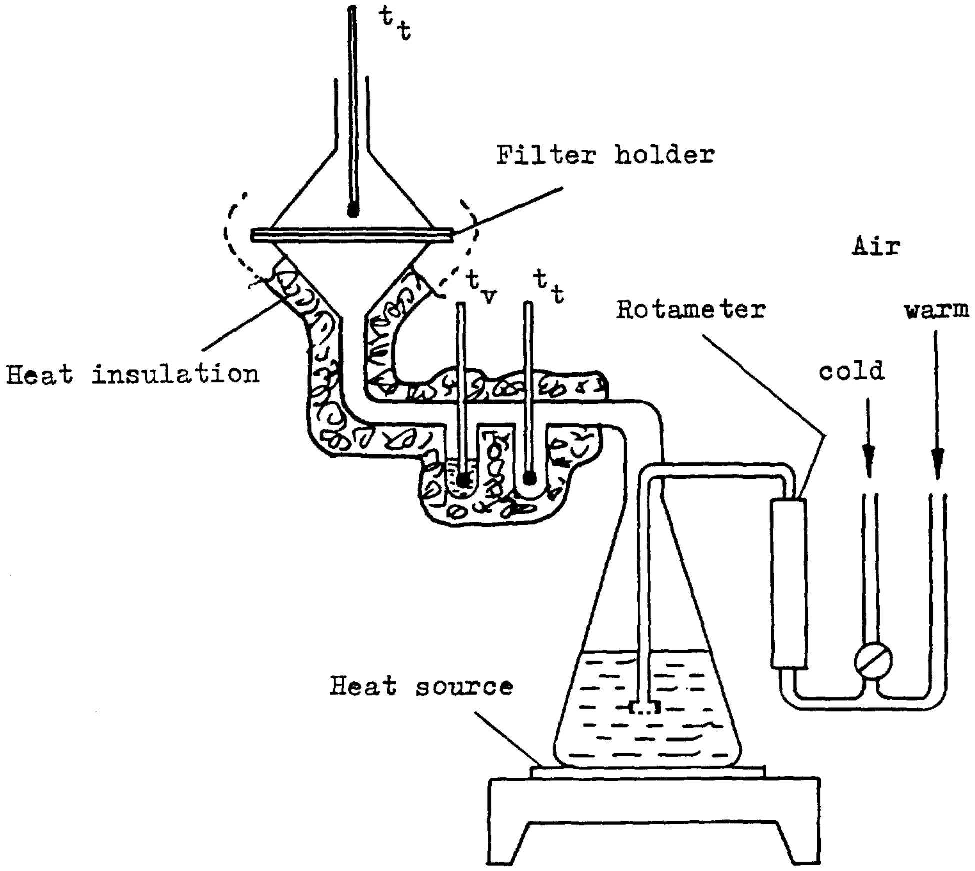


Figure 2. Apparatus for exposing filter material to varying temperatures and pressures. t_t and t_v are the dry and wet temperatures respectively.

The mechanical strength tests, Figure 3, are performed in air in which the flow rate can be increased successively until failure occurs.

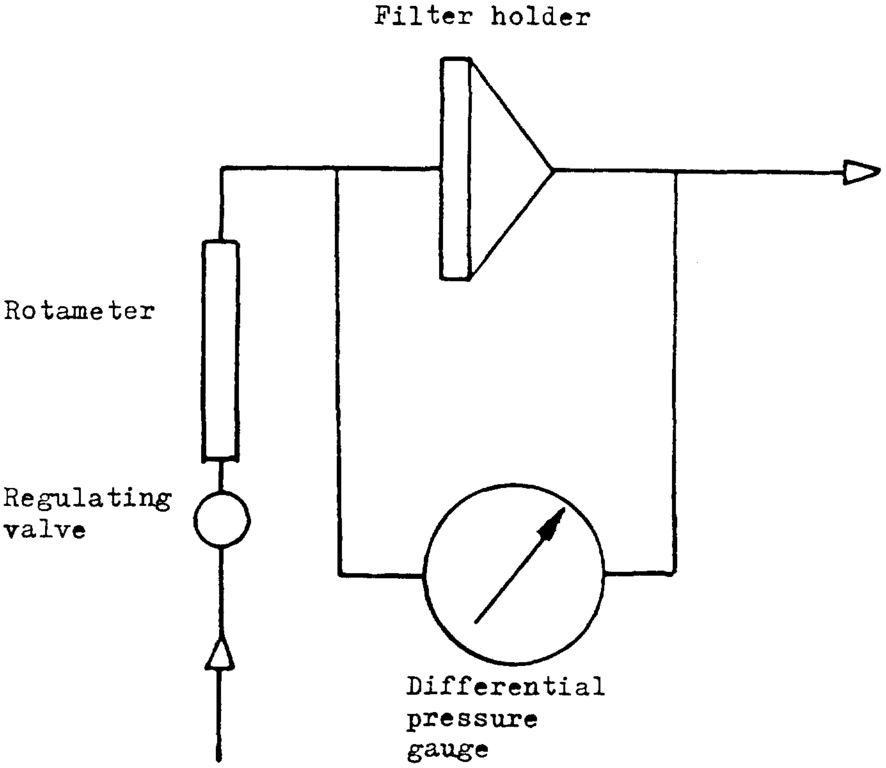


Figure 3. Apparatus for measuring the pressure drop and failure pressure across filter material.

Full scale filter test

The full scale tests of HEPA filters has been carried out in a rig in which the correct humidity and temperature can be attained and maintained automatically.

IV. Results

Filter material

Tests in moist air and water at room temperature

A number of measurements of the pressure drops at high humidities and room temperature are shown in Table 1 and Figure 4. The results show that the pressure drop is almost constant up to about 90 % RH. Above that there is a marked increase, Figure 4.

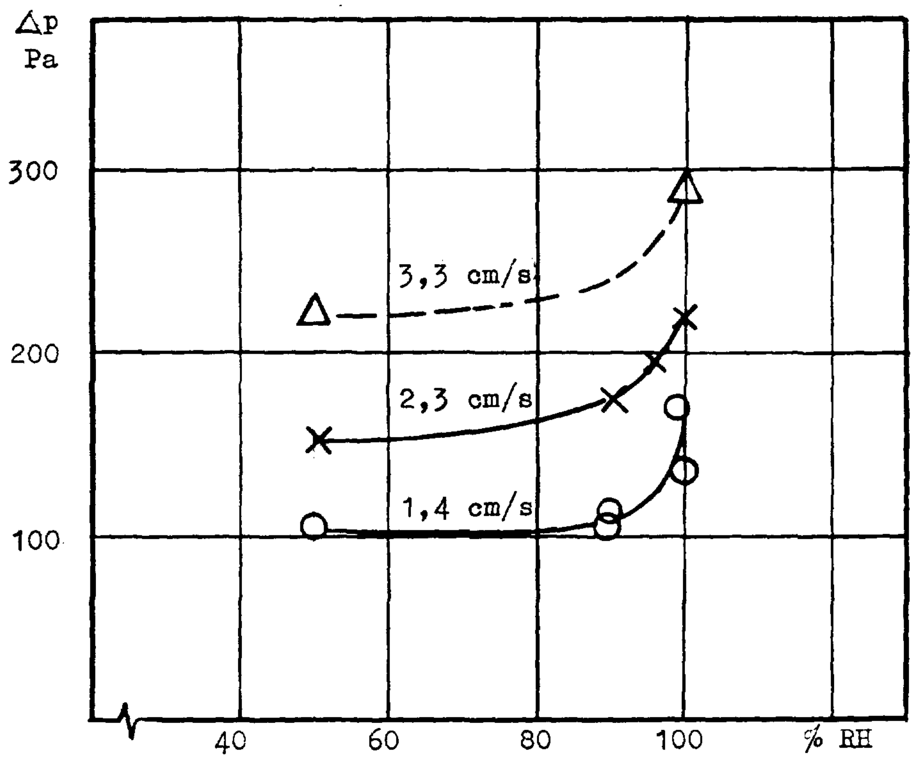


Figure 4. Pressure drop as a function of relative humidity for filter material type 1.

These tests, and tests in which the filter material is wetted in the ionized water for 6 days, show that no signs of change in the material structure could be seen using Scanning Electron Microscopy (SEM).

Material tests in moisture at elevated temperatures

Two tests at approximately 85 and 90 % RH at 85°C were carried out using material type 1. One test lasted 2 hours and the other one hour.

It is difficult to determine from the SEM micrograph, Figure 12, whether there is any damage to the fibres as a result of the relatively high humidity and temperature. On the other hand the mechanical strength has been affected.

Table 1. Pressure drop across filter material, type 1, exposed to flowing air.

Rel hum %	t°C l/min	Flow rate	v cm/s	ΔP mm vp
~90	20	-	1.43	10.8
50	20	14	1.41	10.4
50	20	23	2.32	15.6
50	20	32.5	3.28	22.4
95	20	22.5	2.32	16.8
100	20	13.4	1.35	13.8
100	20	22.4	2.30	22.0
100	20	27.8	2.80	28.4
90	20	13.4	1.35	11.2
90	20	13.4	1.35	13.8
90	20	13.4	1.35	10.8
90	20	14.6	1.47	10.8
97	20	13.4	1.35	16.8

Table 2. Mechanical strength after exposure to pure steam at 100°C. The material samples were dried prior to "pressure testing".

Pressure drop kPa at 28 l/min	Steam flow rate min	Failure pressure kPa	
	Material 1		
4.75	0	100	
4.70	60	67	85°C 85 %
4.70	160	35	
4.60	300	35	
4.60	360	18	
	Material 2		
4.80	0	85	
4.70	130	55	
4.60	300	27	

Tests in pure steam

Tests in pure steam, 6 in all with 2 types of material, were performed with a flow rate of approximately 13 l steam per minute. This gives a flow rate throught the material of 1.3 cm/s, which corresponds to about 50 % of the flow rate through the filter during normal operations.

Measurement of the pressure drop after the material has dried does not show any significant difference between new material and material exposed to steam at 100°C, see Table 2.

Tests of mechanical strength

Tests of the mechnical strength have been carried out using material exposed to pure steam at 100°C. After drying, the specimens were mounted in a circular holder, diameter 42 mm, tilted with the means for measuring pressure and flow rate, see Figure 3.

The results, Figure 5 and Table 2, show that the fracture strength decreases markedly when the material is exposed to flowing steam for some time.

Both types of material had a reduction in failure pressure from about 1 bar to between 0.3 and 0.4 bar after drying.

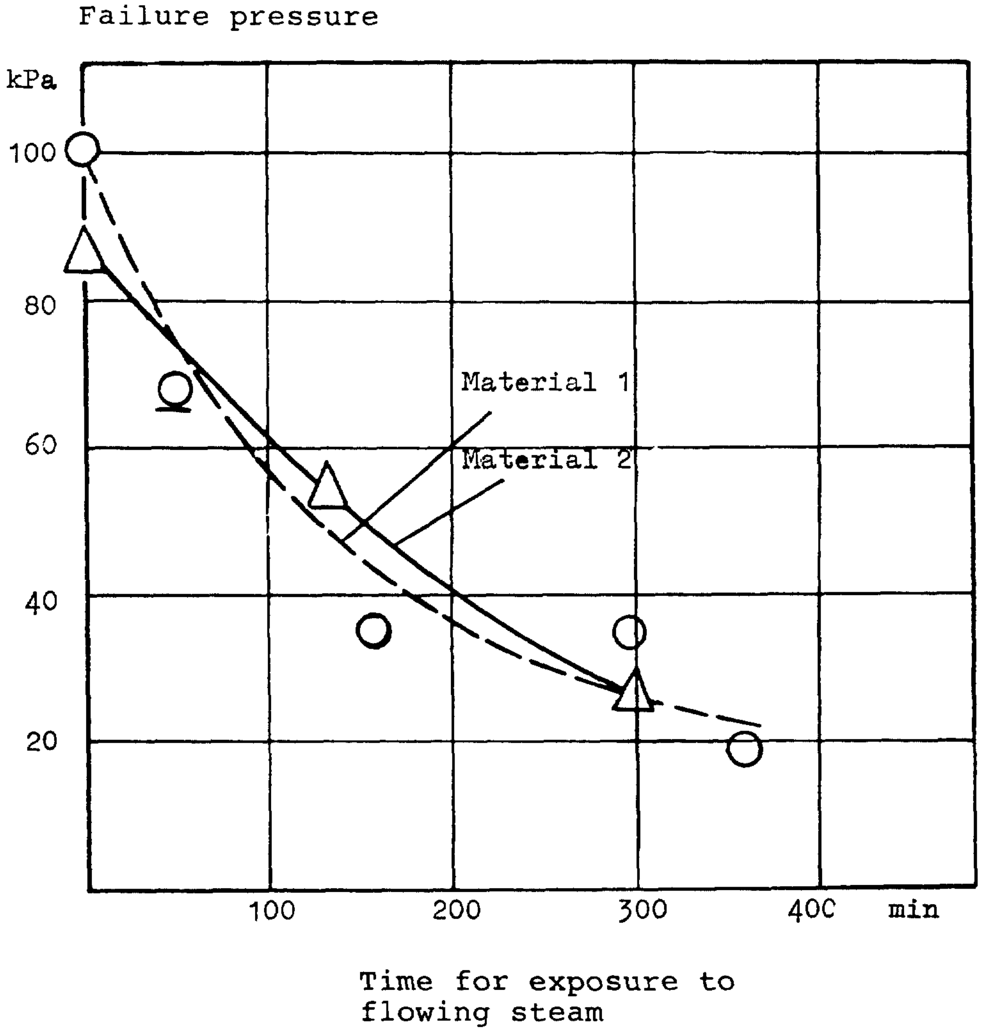


Figure 5. Filter material 1 and 2 mechanical strength sample after exposure to steam at 100°C and a flow rate of approximately 1.3 cm/s. The point 68 kPa/50 min is at about 90°C. The material was dried at about 50°C after exposure to the steam.

Penetration measurements

The tests were performed with DOP on filter material exposed to pure steam at 100°C. A poly dispersive aerosol and particle counter were used for the tests.

The flow rate for the penetration tests was 5.3 cm/s, which is about twice as high as during normal operation.

The filter material was dried in an oven at 50 - 60°C before measurements were made.

The results from the DOP tests indicate that there was an increased penetration, after the filter material had been exposed to pure steam at 100°C for some time, see Table 3 and Figure 6.

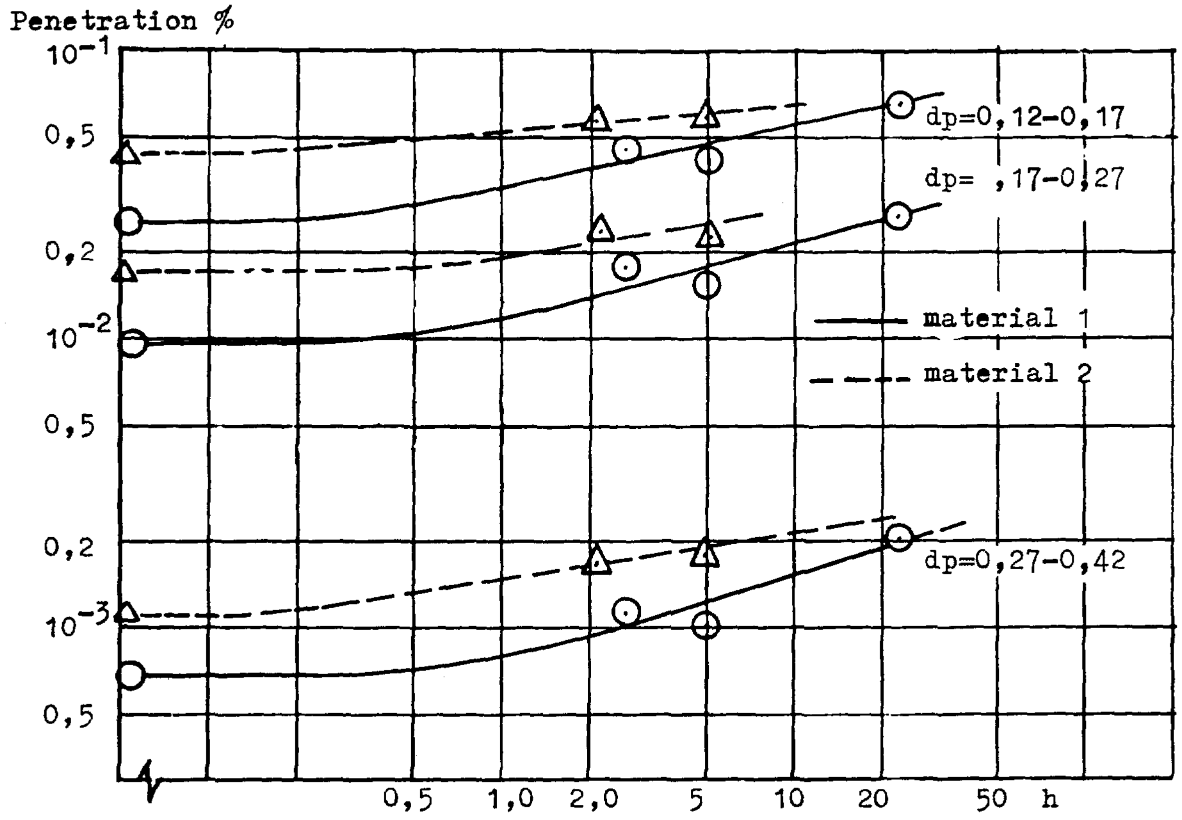


Figure 6. Increase in penetration measured after exposure to steam at 100°C and a flow rate of approximately 2 cm/s. dp in the diagram indicates the particle diameter during the penetration test.

Shape changes

During exposure to the 100°C steam the material specimens had been mounted horizontally; the steam flowed from below. After a while the material deformed into an upturned hemisphere.

The height of the hemisphere increased with time, and was between 5 and 15 mm after 300 minutes exposure to the steam. The diameter of the specimens was 150 mm.

Tests in condensed steam

Two tests were performed in condensed steam. 100°C steam flowing towards the filter ($\sim$0.5 cm/s) down stream from which cold air flowed. Thus the steam condensed in the material. Cracks appeared in the material during this test.

However after drying the remaining parts were mechanically tested and examined using SEM.

The fracture strength of 42 mm circle was about 0.2 - 0.4 bar, compared with 1.0 bar for new material. The results are comparable with these obtained in pure steam.

The SEM micrographs however do not indicate any noteable changes in the structure of the material, Figure 11.

Full scale filters

Tests in moisture at room temperature

The filter was run for a week with 90 % RH at ambient temperatures, and a further 3 days at 100 %. Measurements showed that there was no difference in the pressure drop before or after the prolonged exposure.

Tests in water saturated filters

The pressure drop across the water saturated filter as a function of the flow rate is shown in Figure 7. The filter was mounted horizontally and the air flowed from above it.

It can be seen that the pressure drop at 400 m^3 air/hour is about five times larger with water than without water. The pressure drop across a dry filter is about 200 Pa

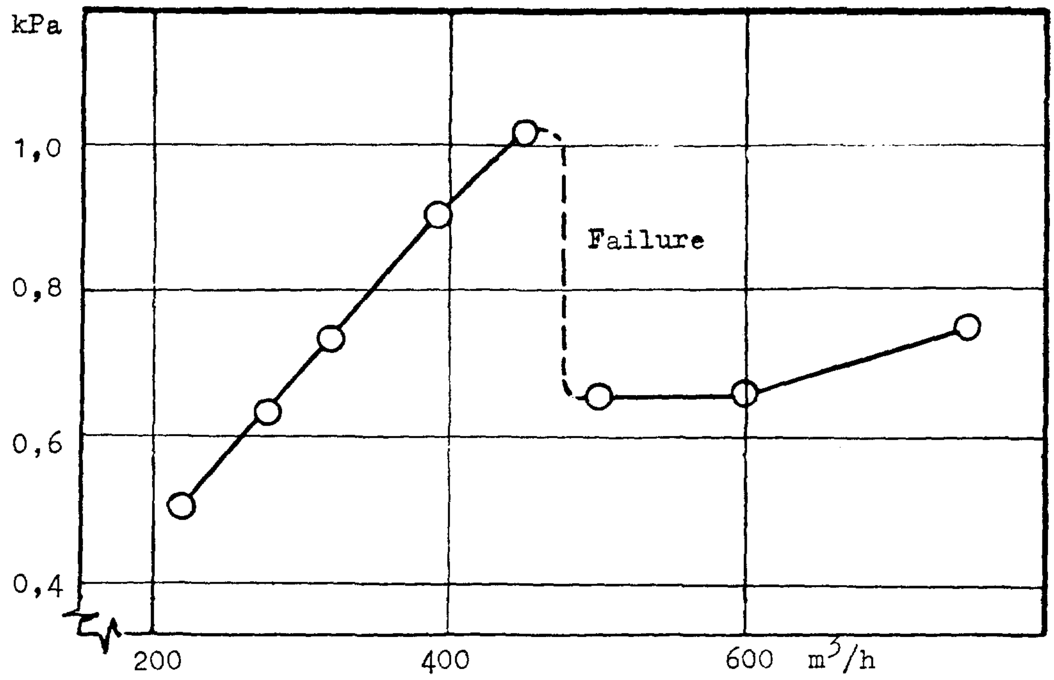


Figure 7. HEPA-filter "triple" design pressure drop after the filter was loaded with water.

During this test the filter contained about 7 l water. The water remained on the horisontal surfaces of the filter during the initial stage of the test.

At about 450 m^3/h and a pressure drop of more than 1 kPa the filter failed, Figure 8. This test shows partly that the material does not allow large amounts of condensate to pass through and partly that this in its turn increases the loading until it exceeds the failure level of the filter at moderate flow rates. As a comparison a standard HEPA filter, bed depth 294 mm , wiel survive 2.5 kPa or about 250 mm water.

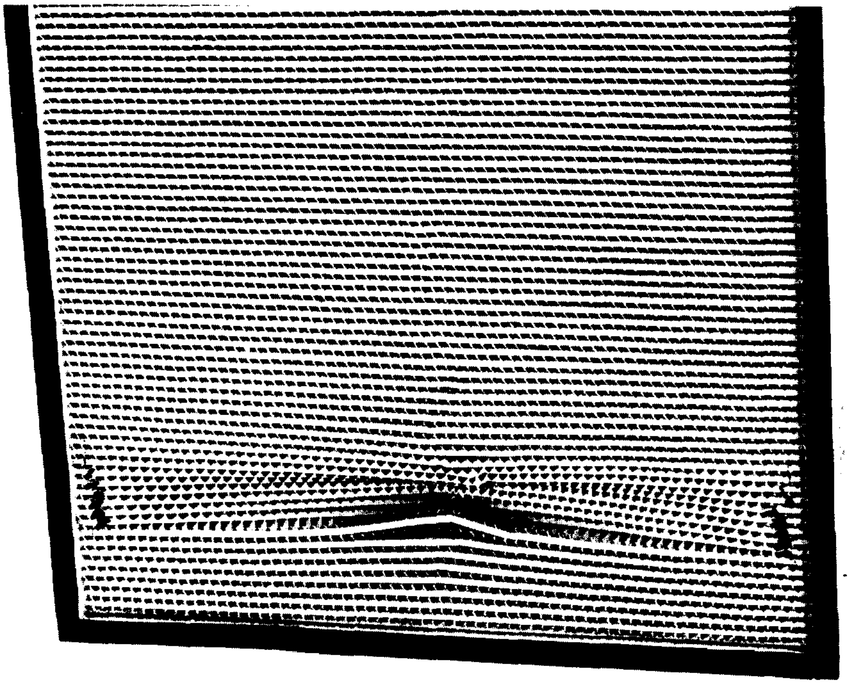


Figure 8a

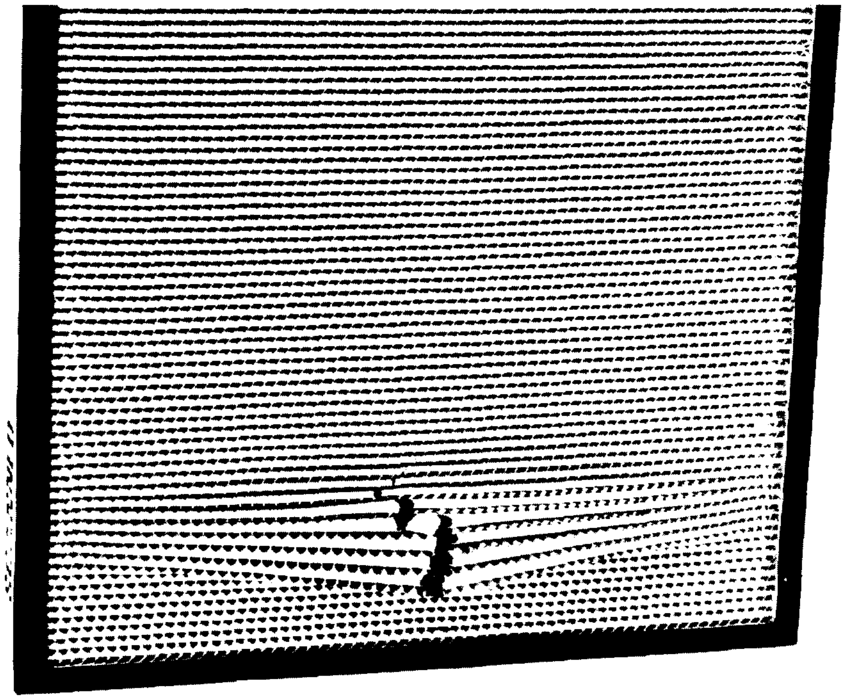


Figure 8b

Figure 8. Failure in HEPA-filter after loading with water and exposure to flowing air. Figure 8a upstream and 8b downstream from the filter.

V. Comments

The following conslucions can be drawn from the results of the filter material tests in moist air and water:

Moisture at room temperature

Moist air, $\leq$ 100 % RH, at about 20°C not had any noticeable effect on the filter material after short exposure times (hours).

Super-saturated air

When super-saturated air flows through the filter material at ambient temperatures and condensation occurs, the pressure drop increases significantly and the material can thus be temporarily overloaded. Overloading and material failure have occurred in boiler water power plants (3).

As noted in (5) an increase has been observed in the penetration during exposure to moisture at ambient temperatures.

Moisture at elevated temperatures

It has been found that HEPA filter material can almost completely loose its strength at high moisture levels and temperatures, 85 - 90°C. Exposure to moisture in the region 20 - 85°C has not been studied. However it is probable that the reduction in strength starts at temperatures lower than 85°C. The mechanical tests were performed after the material had been dried.

Moist and warm material has an almost negligible strength in this temperature region.

Steam and condensation at 100°C

In this environment the filter material is considerably weakend and can fail due to its own weight or in a minimal stream of air.

Filter material which is in practice also partially clogged by dust will probably absurb a larger amount of condensation. It will thus probably "blow" apart in a much less agressive environmen with regard to moisture and temperature even at low flow rates or pressures (6).

Moisture/water, long term exposure

Figure 13 shows deposits on the fibres. The test has not been repeated. However the fact that high moisture levels at moderate temperatures, 20 - 40°C, have an adverse effect on the material cannot be excluded.

Figure 9. Reference Specimen, new filter material, type 1. Magnification x 2 000.

Figure 10. Filter material soaked in water for 6 days. The reason for the deposit on the fibres is uncertain.

Figure 11. Material type 1 after exposure to condensed steam for 2 h.

Figure 12. Material type 1 after exposure to an air steam mixture 85 % RH at 85°C for 2 h.

Full scale filters

As expected the thinner HEPA filter has only moderate resistance to the pressure increase caused by water or condensation.

During prolonged operation with moisture levels of about 100 % an increasing quantity of condensated water can form a layer of water on the surface of the filter.

The filter material is very compact which results in the water remaining in the fibres. This in turn can result in an excessive load and ultimately to filter failure.

It should however be painted out that the carbon filter in triple and combination filters, in which HEPA filters are usually used, are much stronger because of their plate design.

VI. Future work

It is important to note the difference between the affect of moisture/condensation at ambient temperatures and at temperatures around 100°C.

In the current work the filter material has been exposed to moisture or water at various temperatures and pressures.

It has thus been shown that the mechanical strength of the filter material was considerably reduced after it had been exposed to moisture at higher temperatures.

The tests were only conducted on the HEPA part of the triple filter.

Since the triple filter is strengthened by the perforated plates which surround the carbon bed, a number of complementary tests of complete triple filters are planned. these tests will be performed at high pressures and temperatures, and also with air steam mixtures.

The work has been commissioned by the Swedish Radiation Protection Institute and will be performed in collaboration with Kernforshungszentrum, Karlsruhe, where there is a rig for testing at high temperatures and pressures (8)

Table 3. Penetration through the filter material after exposure to flowing steam.

Exposure time h	Particle size, µm 0.12-0.17	0.17-0.27	0.27-0.42	Average
Material type 1				
0	0.0254	0.0093	0.00066	0.0117
2.67	0.0449	0.0177	0.00112	0.0212
5.0	0.0419	0.0153	0.00103	0.0194
23.0	0.0648	0.0262	0.00202	0.0310
Material type 2				
0	0.0433	0.0171	0.00110	0.0205
2.17	0.0583	0.0246	0.00170	0.0282
5.0	0.0585	0.0227	0.00181	0.0277

REFERENCES

1. NEA Group of Experts CSNI,
Report on Air Cleaning in Accident Situations, Febr 1984.

2. RÜDINGER, V RICKETTS, C J and WILHELM, J, G
Limits of HEPA-Filter Application under High-Humidity conditions.
18th DOE Conf, Baltimore, Aug 1984.

3. Normann, B
Iodine and Particle Filter systems in Nuclear Power Stations, Functional requirements and tests.
Studsvik Report NW-83/431 (1983) (In Swedish).

4. National Institute of Radiation Protection, Order Project SSI P271-84, Particulate filters at high humidity, 1984-05-15.

5. DAVIS, R J et al
The effects of Moisture on the Character and Filtration of UO_2-Stainless steel Aerosols.
Symposium, New York, Aug 1968.
Treatment of Airbourne Radioactive Wastes.

6. NORMANN, B, WIKTORSSON, C
Projekcts on filter testing in Sweden.
18th DOE Nuclear Waste Management and Air Cleaning Conference.

7. Nationalinstitute of Radiation Protection Project SSI P313-85, Tests of Carbon/Particulate filters in accident situations.

8. RÜDINGER, V, ARNITZ, T H, RICKETS, C I and WILHELM, J G
Bora - a Facility for Experimental Investigation of Aircleaning during Accident Situations.

DISCUSSION

SCRIPSICK: The penetration measurements that you showed seem rather high, going up to about 0.05% penetration. Is that normal for this type of filter system?

NORMANN: Not for the system, but maybe for the filter media. I can not give you any explanation for this high penetration at the moment. The tests were done with a polydisperse aerosol and the penetration of 0.05 percent refers to very small particles. The penetration decreases with increasing particle size.

SCRIPSICK: My experience with nuclear grade filters suggests that penetration is fairly high, even with the 0.12 to 0.17 m aerosol.

NORMANN: Yes, I noticed that and will look it over. You can see that we have an increase from lower humidity to higher.

WILHELM: Would you indicate the upstream and downstream side of the filter? Is the upstream side the HEPA filter or the prefilter?

NORMANN: In operation, the upstream side is the prefilter, and then you have the carbon filter, and after that you have the HEPA filter.

WILHELM: Our experience has been that small particles pass the prefilter and pass on to the HEPA filter. Now, if the small particles absorb iodine, this iodine will deposit on the HEPA filter but it won't stay there long because it leaves the HEPA filter by oxidation and desorption and will be released. We experienced that in our hot cell work. If you use only a prefilter in front of the carbon filter, I think you won't have good removal efficiency for iodine.

NORMANN: I have not heard about this before. The prefilter is to protect the HEPA filter. When they designed and constructed the filters, I think they divided them into two parts, iodine and particles. No particles stayed in the carbon, all particles stayed in the HEPA, and no iodine reached the HEPA filter. That was the philosophy in Sweden at the beginning.

HEPA-FILTER BEHAVIOR UNDER HIGH HUMIDITY AIRFLOWS*

C.I. Ricketts, V. Ruedinger, J.G. Wilhelm

Kernforschungszentrum Karlsruhe GmbH
Laboratorium für Aerosolphysik und Filtertechnik
Postfach 3640, D-7500 Karlsruhe 1
Federal Republic of Germany

Abstract

A loss-of-coolant accident could threaten the integrity of the HEPA filters in the air cleaning systems of a nuclear power reactor with airflows of high humidity, elevated temperature, and greater than design flow rate. It is important that filter reliability be assured during accident situations since a loss of filter integrity could result in a loss of containment.

A review of the literature concluded that existing countermeasures are either not being fully taken advantage of or are not preventing humidity related filter failure in, for the most part, routine service. The development of filters with greater pack stability and structural strength was judged to be necessary for accident conditions that would involve high humidity airflows.

The average failure differential pressure at 1700 m^3/h for three commercial filter designs under conditions of high air humidity at 50 °C were found to lie between 0.7 and 7.6 kPa. The modes and mechanisms of structural failure were determined for wooden frame deep pleat filters, the design with the most potential for improvement. Initial tests of prototype filter units with a glass fiber medium reinforced by fiber glass cloth proved that structural limits could be increased to a least 10 kPa even with significant decreases in the lateral stability of the filter pack. A similar test of a prototype filter equipped with a special arrangement of the separators and a conventional glass fiber medium showed that pack stability could be maintained during fog conditions that cause failure of conventional glass fiber filters within several hours.

Initial investigations of the water vapor sorption characteristics of glass fiber filter media at 25 °C showed that a dust loading can increase the adsorbed water content in a filter medium by up to a factor of 50 at 97 % r.h.

---

* Work performed under the auspices of the Federal Ministry of the Interior under Contract No. SR 290/1.

I. Introduction

Despite the advanced stage of development and the high levels of performance exhibited by today's commercial High Efficiency Particulate Air (HEPA) filters, reports of moisture related filter failure in nuclear facilities, for the most part during routine service, still appear persistently in the literature /1-7/. The susceptibility of glass-fiber HEPA filters to deteriorations in performance resulting from exposure to humid airflows remains an undesirable and only partially mitigated filter characteristic. As part of the barrier between contaminated zones and the ambient environment, the relatively fragile HEPA filters must be depended upon to perform at design specifications not only during normal operations but also under abnormal operating conditions.

Steam released by a Loss of Coolant Accident (LOCA) /8,9/ or water sprays activated during a fire /9,10/ or LOCA /5,11/ could introduce large quantities of moisture at elevated temperature into the air cleaning systems of a nuclear facility. Filter reliability during such accident conditions is brought into serious question by the reported occurrences of moisture-related filter failure during more routine operations.

A recent study of the literature on the topic of air cleaning in nuclear accident situations /8/ found a lack of information concerning the influence of particulate loading, long term moisture exposure, and filter aging on filter response to high humidity airflows. In a progress report on ongoing filter testing /12/ it was concluded from a literature survey that for dust loaded filters in particular, filter behavior is not documented well enough to assure filter reliability during accident conditions that would involve humid airflows.

Filter reliability depends foremost upon the preservation of filter structural integrity. Assurance of filter reliability requires that filter structural limits not be exceeded by the mechanical loads acting upon filters at their service locations. Values of filter structural limits and mechanical loads are prerequisites to the calculation of realistic safety margins for abnormal or accident conditions.

For practical reasons, the structural limits of individual components such as filters are measured in test facilities. The mechanical loads placed on components in an accident situation can be estimated by the use of computer codes that numerically model the fluid-dynamic and thermodynamic conditions that would prevail in the air cleaning system during the accident/13/. For accurate predictions from such codes, experimentally determined flow-resistance characteristics of individual components are indispensable as input data.

To realistically assess the reliability and the safety margins of in-service HEPA filters under exposure to high humidity airflow, necessitates a knowledge of filter structural limits and flow resistance characteristics under the relevant conditions of operation, and the availability of a suitable computer code for modeling fluid-dynamic and thermodynamic transients in air cleaning systems. Should improvements in filter structural strength be judged to be necessary, the modes and mechanisms of moisture related filter failure would then also become important.

II. Literature

Parameters Governing the Incorporation of Water into the Filter Medium

Moisture induced deteriorations in filter performance and filter structural failure result principally from the presence of liquid water in the fiber structure of the filter medium. The incorporation of water into the filter medium from the airstream can occur by sorption, condensation, or the filtration of water droplets, depending upon the water content of the air /12/.

It is to be assumed that various parameters of the airstream and of the fiber structure influence the moisture content of the filter medium at equilibrium, as well as the rate at which the moisture content attains an equilibrium value. Among these would be the temperature, velocity, and moisture content of the airstream. Relevant characteristics of the fiber structure would include the material, the fiber diameter, the porosity, the water repellency, the particulate loading, and the drainage properties. However, the degree to which and the ranges of values for which these parameters influence the transfer of water into and out of a HEPA-filter medium remain yet largely undocumented in the literature. Summarized below are some of the investigations that have been found to be pertinent.

Sorption: Isotherms and Kinetics. For an air relative humidity, φ , between 0 and 100 %, physical sorption is responsible for the water bound on the surfaces and in the pores of a glass-fiber filter medium. The airborne water vapor, the filter medium, and the surface-bound water correspond in the sorption process to the adsorbate, the adsorbent, and the adsorpt, respectively. One measure of the hygroscopic behavior of an adsorbent is the sorption isotherm. This is defined to be the equilibrium moisture content, at constant temperature, θ, of a sorbent as a function of P_d/P_s, the ratio of the sorbate partial pressure to the sorbate saturation pressure, which is equal to φ /14/. Adsorption measurements are normally made by either volumetric or gravimetric methods /15/. The units commonly used for the moisture content of the sorbent, ψ ,are g H_2O/100 g dry sorbent.

Brunauer et al. /16/ have classified sorption isotherms in the five major groups illustrated in Fig. 1. The characteristic shapes can provide information about the sorption phenomena involved and the ranges of φ for which they dominate.

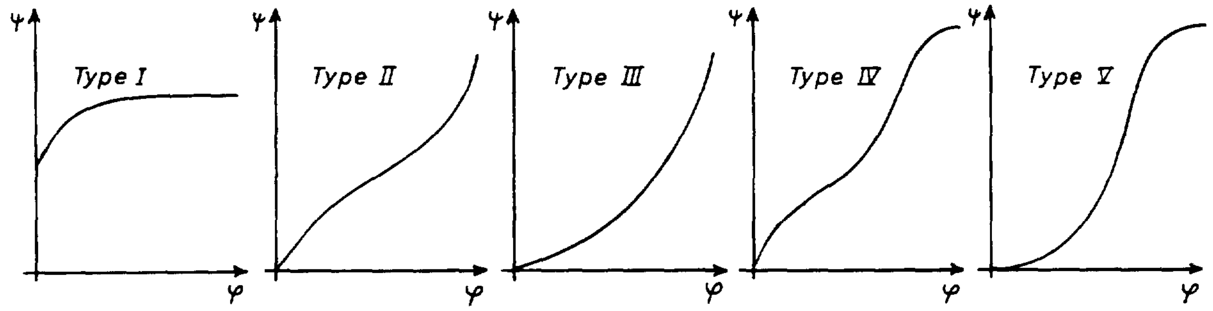


Fig. 1: Classification of Sorption Isotherms from Brunauer, Deming, Deming, and Teller /16/.

In the general case of a type-II isotherm as shown in Fig. 2, monolayer sorption, multilayer sorption, and capillary condensation dominate in respective ranges of φ which are marked by changes in curve slope /17,18/. Due to hysteresis, the desorption curve for a given material generally exhibits a greater moisture content than the adsorption curve.

Studies by Kadlec and Dubinin /19/ of capillary condensation in microporous glass at 20 °C yielded type-IV sorption isotherms for 0-100 % r.h. with large hysteresis loops above 40 % r.h. and maximum ϕ values of about 20 %. The effects of various surface and heat treatments in limiting the adsorption of water vapor on porous glass has been reported by Kawasaki and coworkers /20/. For type-II isotherms at 30 °C and up to 75 % r.h., the maximum values of ϕ for untreated surfaces lay under 3 % and some were reduced by almost one-half after treatment.

Type-II adsorption isotherms for two different presumably clean, unspecified filter papers /17,21/ showed ϕ at 90 % r.h. and 20 ° to be on the order of 10 % for both. For the captured particulates on slightly loaded glass-fiber sampling filters, Tierney /22/ published type-III adsorption curves that at 100 % r.h. and 22 °C show ϕ values between 40 and 90 % depending upon particulate type. Capillary condensation was evident at 50 % r.h. Equivalent type-III isotherms for the loaded filters themselves displayed a maximum of 8 % for ϕ . Clean filters exhibited no apparent adsorption of water vapor. Type-II isotherms from Hofmann /23/ for a number of clean and slightly loaded HEPA filters at 97 % r.h. and 20.5 °C had values of ϕ equal to 2 and 8 %, respectively. The maximum ϕ due to hysteresis for the dust loaded filter units was a factor of 10 greater that that of the clean units. Capillary condensation began to dominate at 70 % r.h. with clean and at 60 % r.h. with loaded units.

These studies illustrate that ϕ at equilibrium can significantly depend on current and prior φ as well as the structure or composition of the glass or particulate adsorbent. Less well documented are effects of other parameters. For a given value of φ , the moisture content of an adsorbent generally decreases with increasing temperature /17,21/. Although adsorption can be measured for an adsorbent in an airstream /14/, it is probable only for large, yet uninvestigated values of φ and superficial velocity that the amount of adsorbed water in capillaries could be influenced aerodynamically.

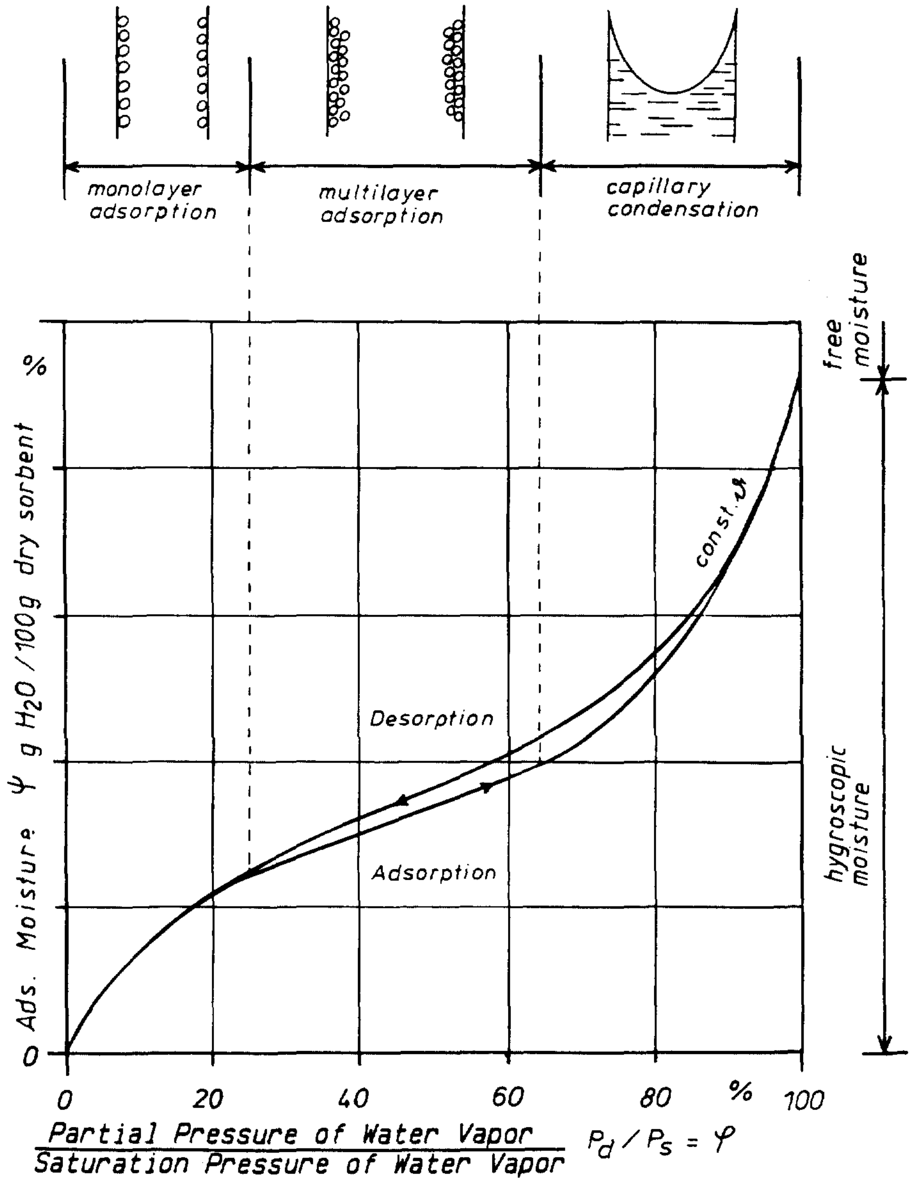


Fig 2: Type-II Sorption Isotherm and Sorption Phenomena /18/.

In addition to the equilibrium values of ϕ , sorption kinetics (see 24), in particular the rates at which equilibrium is attained at constant φ , are also relevant. However, even sorption kinetics in general have been topics of very few published experimental investigations. The rate of physical sorption usually increases with temperature / 25/ and most likely with increasing velocity of an adsorbate through a porous adsorbent. The influence of φ and the surface properties and structure of glass fibers on sorption kinetics do not appear to have been investigated yet.

Condensation. Condensation of airborne water vapor onto filter fibers as free water will occur during any time that the dew-point temperature of the air exceeds the temperature of the fibers or at φ = 100 % r.h., both unstable states in an airstream. Practically, these two special cases can exist in an air cleaning system for short periods of time and only under transient conditions. Nevertheless, saturation of a filter medium by condensation cannot be ruled out. The amount of water incorporated and the rate of incorporation would then depend upon the rate and duration of condensation as well as the water repellency, drainage characteristics, and capillary structure of the filter medium. The airstream velocity would also become important in the rate of water transfer from the downstream side of the filter medium back to the airstream by reentrainment.

Droplet Filtration. For airstreams with a water content above that corresponding to 100 % r.h., filtration of entrained water droplets by the filter will also lead to the incorporation of free water into the filter medium. The rate and extent of water incorporation are dependent upon the duration of exposure to the droplet flow and also the concentration and size distribution of the droplets. Additional factors include the filtration efficiency as well as the other filter medium parameters and the airstream velocity as noted above for the case of condensation.

For filter units in continuous service, Burchsted /9/ proposed for the water content of the filter medium a maximum allowable value of 25 g of H_2O per m^2 of filter medium surface area, said to correspond to a liquid water content in the airstream of 24 g of H_2O/m^3 of air, at design flow rate and equilibrium conditions. Since these values took increased water retention due to dust loads and filter aging into account, values twice as large were said to be permitted for clean new filters at equilibrium. The recommended maximum diameter of airborne water droplets allowed to reach HEPA-filter units in continuous service was given as 1 µm. In comparison with curves from Ruedinger /12/ for ΔP as a function of air humidity, the value of 24 g/m^3 seems to be somewhat high.

The Effects of Moisture on Filter Performance and Construction Materials

Moisture degrades the performance of HEPA-filter units through detrimental changes in the properties of filter construction materials, especially those of the glass-fiber filter medium. Surveys of reports that deal with the moisture effects on filters have appeared elsewhere /8,9,12/ and altogether draw upon such a large number of sources that the brief summary presented here will at times refer to these surveys rather than directly to each original publication.

Filter Medium. The presence of moisture in a clean glass-fiber HEPA-filter medium can effect an increase in the pressure drop /26,27/ and the weight /28/ as well as a decrease in the tensile strength /12,8,29/ and the filtration efficiency /30/ of the medium. The degree to which these changes occur has been shown to increase with decreases in the water repellency of the filter medium /26-28/. Factors which can significantly reduce the water repellency include dust loading /26,27/, aging /31/, creasing /31/, and gamma radiation /27/ of the medium. How water repellency is influenced by moisture exposure time and temperature remains yet unknown. A number of studies have shown that the surfaces of glass fibers can slowly dissolve in water solutions (see 32 and his citations 96, 107, and 103). To what extent the organic binder and water proofing agents inhibit this process also remains yet to be determined.

Increases in pressure drop from about 0.5 to 4 and 4.6 kPa at 100 % r.h. and 30 ^{o}C after 7 h were reported by Jones for specimens from filter units loaded during, respectively, 7 and 48 months of service /27/. In one test, Peters measured a maximum of 10 kPa for a clean water repellent sample after 22 h in a wet steam flow at 100 ^{o}C and design flow rate /26/. Both authors found a tendency toward more rapid and greater increases in ΔP as well as lower values of water repellency for dust-loaded filter papers. Filter medium samples measured by Schwalbe in steam-air mixtures at 65 ^{o}C and 4.5 times rated flow showed in one case a maximum value of 2.5 kPa after 12 min of exposure. A non water repellent sample burst at 3.8 kPa after 5 minutes. After 2 h the weight increase due to water loading of the undamaged samples ranged between 64 and 460 % /28/.

Published decreases in tensile strength of clean wet samples of filter medium compared to dry ones have ranged from 35 to 70 % /12,29/. Tests of wet dust-loaded specimens do not seem to have been reported. Decreases of up to 35 % in the tensile strength of clean dry samples were measured by Jones /27/ after exposure to 5 x 10^7 rad. Similar doses of gamma radiation were also found to reduce the relative water repellency of equivalent samples by up to 100 times. Reductions in tensile strength of up to 60 % /33/ and 70 % /31/ for new dry filter media have been attributed to the creases formed in folding the media during filter manufacture. Values for similar samples from used filters showed decreases of up to 80 % /31/. Water repellencies of both new and used media were determined to be 40-60 % lower for samples that contained creases. Aging has accounted for decreases in tensile strength of up to 5 % for unfolded and up to 26 % for folded dry specimens from filters taken out of normal service /31/. Water repellency also decreased due to aging by some 20 % for those samples without creases and 40 % for those with. Although such deteriorations in tensile strength and water repellency represent more extreme cases and are not necessarily cumulative they provide an outline of the challenges posed to the integrity of filter media in service.

Storage of samples of HEPA-filter media up to 48 h at 100 % r.h. and 80 ^{o}C have led to increases of up to one order of magnitude in the penetration by U_2O-stainless steel aerosols /30/. This was attributed to an increase in effective fiber diameter due to a drawing together of small filters into wetted bundles with filtration characteristics of larger fibers. Here also, water repellent media showed less susceptibility to moisture effects. Similar decreases in the filtration efficiencies for the same aerosols in humid as compared to dry airflows at a 5-cm/s superficial velocity were presumed to be due to moisture-induced shrinkage of agglomerates into more penetrating sizes. A laser spectrometer was used by Deworm /34/ at 20 °C and 100 % r.h. to measure the penetration of DOP, NaCl, and uranine aerosols with diameters of 0.09-0.3 µm through clean HEPA filter units. In spite of the questionableness of using a liquid or a water soluble aerosol at high humidities, results for all three aerosols were of the same order of magnitude and showed an average decrease of some 45 % in penetration in comparison with values at 26 % r.h. Possible explanations for the results were not discussed. Ensinger has reported on the development of a method which will employ a TiO_2 aerosol to measure HEPA filtration at high humidities /35/. This method should fill a large gap in knowledge for actual filtration behavior of full-size filter units during accident situations.

Filter Construction Materials. The effects of moisture on filter construction materials other than the filter medium can include the corrosion of aluminum separators /31,38/ or metal frames /9/. Also reported have been the softening of asbestos separators /9,36/, the swelling of wooden frames , and the separation of pack adhesives from wooden frames /12,37,38/. The availability of suitable corrosion resistant materials such as stainless steel and surface treated aluminum or steel make the prevention of these types of deteriorations primarily matters of cost and of procurement specifications.

HEPA-Filter Units. The principal reported undesirable effects of humid airflow on full-sized filter units are differential pressure increases and structural failure /9,12/, both attributed to water in the filter medium. Ruedinger /12/ published curves for equilibrium differential pressure as a function of air relative humidity and liquid moisture content at design flow rate and 50 °C. Values as large as 9 kPa for clean filters exposed to fog and 7.5 kPa for service loaded filters subjected to saturated airflow were observed. Filter ΔP as a function of accumulated water mass in clean filter units exposed to water aerosols at 20 °C have been reported by Fenton /39/. In one case the differential pressure rose up to 1.5 kPa at reduced flow rate after 27 h and corresponded to a mass of 3.4 kg of water in the medium and the asbestos separators of the filter. At approximately 97 % r.h. and 20 °C Hofmann /40/ found the ΔP of a slightly loaded filter to be 300 % greater than that of a clean unit. Stratmann used a liquid water aerosol to load 11 clean HEPA filters of various types up to 1.2 kPa /38/. The time required varied between 4 and 40 h. Two additional filter units investigated showed little increase in ΔP. Steam-air mixtures at 3 times the design flow rate and 80 °C were used by Jones /41/ to test model filters constructed with filter medium removed from full-size filters following service periods of 4-13 months. The ΔP of one filter increased to 6 kPa after less than 5 min. of exposure. Collins and coworkers /42/ demonstrated the effect of pleat orientation to water entrained airstreams on ΔP by tests of two filters, in tandem with respective prefilters, exposed to water droplets of 1 to 10-µm diameter at initial flow rates of close to 1700 m^3/h. The differential pressure of the filter in the upward vertical flow, despite exposure to 40 % less entrained moisture, had an initial-flow-rate equivalent ΔP twice as large as the filter in the horizontal flow. Other authors have measured filter ΔP increases in humid airflows ranging from some Pa to several kPa /see 12/ often without repeatable or comparable results.

The wide variations in the extent and rate of reported ΔP increase can be attributed to the difficulties encountered in the accurate measurement and control of high humidities in an airstream, as well as the extreme sensitivity of filter ΔP at air humidities close to and above saturation. The ranges and strong influences of dust loading and water repellency on the incorporation of water into the filter medium also contribute to the inconsistency of published results. Tests performed at other than constant flow rate also make comparisons difficult. A reasonable judgment is that values of φ from 95 to 100 % r.h. at $\Theta \leq 50$ °C can lead to differential pressures of up to 5 kPa across dust loaded filters at design flow rate in less than 24 hours of exposure. Likewise, airstreams with liquid moisture contents of about 10 g/m^3 can produce similar ΔP across clean filter units despite the use of a water repellent medium. These values for design flow rates may still be subject to some revisions. Corresponding values of ΔP and humidity for filters in vertical airstreams or with horizontally oriented pleats or at higher than design flow rates require further investigation.

The structural limits for a total of 7 HEPA filters given by various authors and compiled in one literature survey /12/ lay between 0.6 and 2.5 kPa. Ruedinger reported differential pressures at failure which ranged between 1.7 and 9 kPa for 15 deep-pleat filters /12/. Analyses of the modes and respective underlying mechanisms of structural failure were discussed for filters with wooden frames and those with metal frames and pack-to-frame sealants of glass fiber.

Documentation of moisture damage to deep-pleat filters with elastomeric /12,36,38/ and glass fiber /12/ sealants as well as to separatorless /43/ and mini-pleat V-type filters /38,43/ have been published. The reported loosening of the filter pack /12,38,43/ and the transverse tearing of folds close to the top or bottom of the pack /12/ resemble damages sustained by filters during high temperature exposure /44/ or during handling and shipment /45,46/. The ruptures at the downstream ends of the pleats in the medium during exposure to humid airflow /12/ have also been observed for filters under high differential pressure /33,47/, at high temperature /44/, and after shipment /36,46/. The occurrence of the same failure modes under such different conditions serves to point out the locations of structural weakness and instability in deep-pleat filters. A similar argument can be made for the mini-pleat V-type filters which in general are structurally weaker than the wooden frame deep-pleat filters /38,43,47/.

Countermeasures

An important measure taken to counter many of the effects of moisture on HEPA-filter units was the development of water-repellent glass-fiber filter medium. However, dust loading, creasing, radiation, and aging of the medium can compromise the effectiveness of present water repellency treatments, as noted previously.

Metal fiber demisters located upstream of HEPA filters have been shown to be effective in protecting new, clean and slightly loaded filters from damage by water entrained airstreams /see review in 8,12/. Reported structural failures of dust loaded test filters at less than 100 % r.h. /12/ confirm the need for air heaters downstream of the demister to lower the relative humidity of the air upstream of the filter as, for example, in the standard design concept of Standby Gas Treatment Systems (SGTS) /48/. As active components however, heaters require external power and control systems and are therefore subject to failure.

The reliability and safety margins of HEPA filters downstream of demisters could be greatly increased by filters that would, in a dust loaded condition and in the case of a heater malfunction, withstand the temperatures, flow rates, and air humidities to be expected /8/ downstream of the demister during a LOCA. Filters with stainless steel /49/ or polycarbonate /50/ microfibers have been shown to fulfill such a requirement. These filter types however, are still restricted in practice to special applications such as containment venting or acid environments due to size or temperature limitations and costs.

For general nuclear applications, it appears at present that further improvements in HEPA-filter performance during exposure to humid airflow will have to be made with media of glass fiber. The moderate cost, high filtration efficiency, low flow resistance, large surface area per unit volume, reasonable degree of chemical inertness, and fair resistance to radiation, elevated temperature, and fire, form a combination of characteristics for glass fiber which no other material can presently match.

Conclusions

The literature survey has shown that the incorporation of water into the glass fiber medium is an adverse phenomena associated with the exposure of HEPA filters to high humidity airflows. Water in the filter medium leads to an increase in differential pressure and to deteriorations in filter pack stability and in filter medium performance characteristics, especially the tensile strength. The mechanical load on the filter is thus increased at the same time that the structural strength is decreased. The end result is that filter structural failure can occur for unacceptably low values of ΔP, even at design flow rates.

If it is reasonable to assume that existing countermeasures are for the most part being effectively utilized, then the persistent reports of filter failure in normal operations indicate that the effects of humid airflows on filter performance have only been moderated and not yet eliminated by these countermeasures. Filters loaded with dust in service still remain particularly prone to failure and the subject of all too few investigations. The development of filters with greater pack stability and strength is seen to be necessary if containment of airborne radioactive material is to be maintained during accident situations involving high humidity airflows at elevated temperature and greater than design flow rates.

Once filter structural integrity can be assured, removal efficiency tests of filter units under high humidity should be conducted with an appropriate aerosol. To be able to mathematically model or to minimize the increase in differential pressure, additional systematic investigations of water transfer into and out of the filter in terms of airstream and filter medium parameters need to be carried out.

III. Experimental Investigations

Filter Structural Tests

Test Apparatus, Parameters, Filters (Commercial). The filter test facility TAIFUN located at Karlsruhe Nuclear Research Center was used to determine the structural limits of 610x610x 292-mm commercial HEPA filters during exposure to high humidity airflows of 1700 m^3/h in tests which lasted between 2 and 200 h each. The structural limit of a test filter was considered to be the differential pressure at which the first visible structural damage to the filter medium appeared on the downstream side of the filter. Descriptions of the test facility, instrumentation, and general experimental procedures have been reported elsewhere /12/.

The new and dust-loaded commercial filters tested were nuclear grade and contained water repellent glass fiber media. Three principal filter designs have been investigated so far. These include:

- deep pleat filters with an elastomeric sealant and a wooden frame for service at < 120 °C,
- deep pleat filters with a seal of packed glass fiber and a metal frame for service at < 250 °C, and
- mini pleat V-type filters with an elastomeric sealant and a wooden frame for service at < 120 °C.

Approximately 35 filters have been tested at 50 °C. An additional 4 filters each were tested at 20 and at 80 °C to evaluate possible influences of temperature on filter behavior.

Most loaded filters in a test group of 21 had pretest pressure drops of 1 to 2 kPa at 1700 m^3/h after 12-18 months of continuous service in a laboratory building in which radioactive iodine is the primary airborne contaminant. Four mini - pleat filters tested had average differential pressures of 0.6 kPa at design flow rate following removal from up to 12 months of active service in a decommissioned reactor building currently used as a containment safety test facility /51/. All loaded filters had served with prefilters in exhaust air ventilation systems and had been mounted in individual caissons or banks through which the vertical airflow was upward into the horizontal upstream face of the filter /52/. None of the filters showed measurable residual levels of radioactivity at the time of the humidity tests.

During a test the air humidity was increased in steps from about 50 % r.h. until either structural failure occurred or an equilibrium ΔP at 100 % r.h. was attained. The test filters that remained undamaged at saturation were then exposed to an airstream with a liquid water content of some 10 g/m^3 and consequently failed within a few hours. The removal efficiency of most test filters was measured for a 0.7-μm DOP aerosol before and after each test at high humidity.

Test Results. The results of the structural tests of commercial filters are summarized in Table 1. For new clean filters, the filter design can be seen to greatly affect the average ΔP at failure. If an average value for the AP and CN filters, 4 kPa, is compared to one for the RNF and DPF types, 0.9 kPa, it is noted that the structural limits of the mini pleat design are only 1/4 of those for the deep pleat design. The use of a packed glass-fiber seal instead of an elastomeric one in deep-pleat filters can account for a decrease of almost 50 % in the average structural limit. This is seen by comparison of the average for clean AP and CN filters, 4 kPa, to the VM value of 2.2. The 50-mm pack depth of the special RNFS filters accounted for no more than a 17 % increase in average ΔP at failure compared to the loaded RNF filters with the standard 20-mm depth.

A greater influence on average structural limit is exerted by the humidity itself. Comparison of the values of Table 1 with those obtained for equivalent filters procured from the same manufacturers and then tested with dry air at high flow rates /47/, show that prolonged exposure to moisture can reduce the differential pressure at structural failure by 60 to 90 %. For instance, the value of 3.4 kPa for clean CN filters is only 15 % of the average value of 23 kPa reported for tests with dry air.

The effect of dust loading on the average ΔP at failure for the deep pleat AP & AN+ and CN filters appears here to be insignificant. This is not the case for the mini-pleat RNF filters where the average failure ΔP of the loaded filters is twice that of the clean ones. This effect can be partially explained if a loading of dust can increase the wet tensile strength of an uncreased filter medium, as discussed later.

+ The difference between the AP and AN filters (from the same manufacturer) was in materials; the AP type had a particle board and the AN a plywood frame. Test results for these 2 types are considered to be independent of this difference and therefore comparable.

Table 1: Structural limits of commercial 610x610x292-mm glass fiber HEPA filters tested under high air humidities at 1700 m^3/h and 50 °C.

Filter		Structural Test Results				
Design Pack / Flow Rate (m^3/h) Frame / Serv. Temp. (°C)	Manf. Code	Test Cond.	No.of Tests	Range of ΔP at Failure (kPa)	Std. Dev. (kPa)	Average ΔP at Failure (kPa)
Deep Pleat (270 mm) / 1700 Al.Separators Wood / < 120	AP	new	4	2.5 - 5.9	1.4	4.6
	AN	loaded	7	4.1 - 6.3	0.8	5.0
	CN	new	5	1.8 - 5.9	1.6	3.4
	CN	loaded	11	0.9 - 5.7	1.4	3.3
	DN*	new	2	6.3 - 9.0	1.9	< 7.6
Deep Pleat (270 mm) / 1700 Al. Separators Metal / < 250	VM	new	3	1.2 - 3.3	1.1	2.2
Mini Pleat (20 or 50 mm) / 1700 V Panels Wood / < 120	RNF	new	2	0.8 - 1.4	0.4	1.1
	RNF	loaded	4	1.4 - 3.6	0.9	2.3
	RNFS	loaded	3	2.5 - 3.4	0.3	2.7
Mini Pleat (25 mm) / 3000 V Panels Wood / < 120	DPF	new	2	0.4 - 0.9	0.4	0.7

* ΔP is that by first visible damage on downstream face of filter. Filters had probably failed sooner within pack at lower ΔP.

The most important effect of a dust loading on the structural failure of any filter type is to lead to structural failure at lower air humidities than those for clean filters. The dust loading and its characteristics determine the degree to which this happens. For the dust and loadings involved here, 15 of 23 loaded filters failed at $\varphi \leq 100$ % r.h. The average value of φ at failure was 97 % r.h. after an average exposure time of 21 h. Two deep pleat filters that failed at 80 % r.h. in less than 3 h represented the lowest values of φ and exposure time. Only 1 of 18 clean filters failed at $\varphi \leq 100$ % r.h. and it was later determined to have had an only partially water repellent medium. Clean filters in general failed after an average of < 10 h in airflows with moisture contents of some 10 g/m^3.

Documentation, Modes, and Mechanisms of Structural Failure

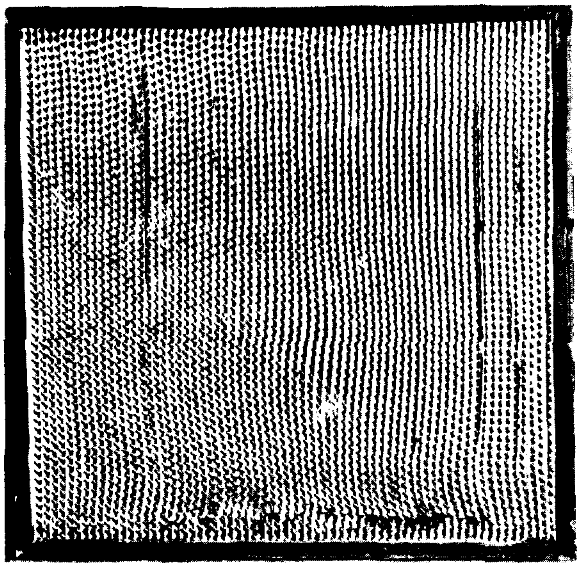
Loss of Tightness in the Filter Pack. Typical structural damage caused by humid airflow through wooden frame deep pleat filters is shown in Figs. 3-5. Two different modes of failure can be seen. One results in ruptures along the creases in the downstream ends of the pleats of the filter medium. Consequences of the second are horizontal tears in the sides of pleats, which normally extend through the depth of the filter so that the damage is also visible on the upstream ends of the pleats.

Damage to the filter medium of 610x610x292-mm deep pleat filters caused by humid airflows.

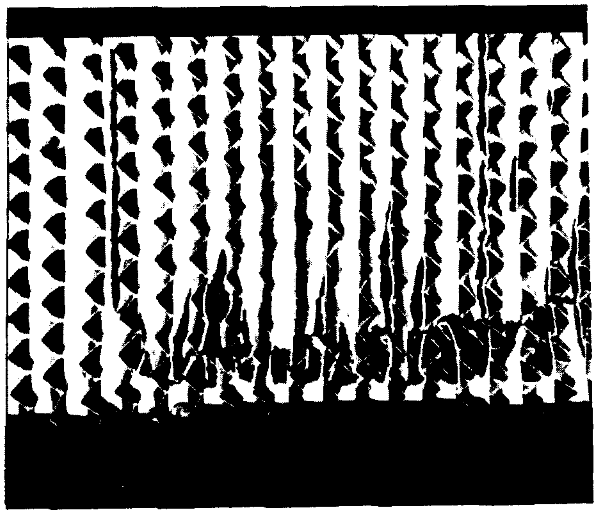
Fig. 3: Ruptured and torn downstream ends of pleats.

Fig. 4: Close-up of downstream ruptured ends of pleats.

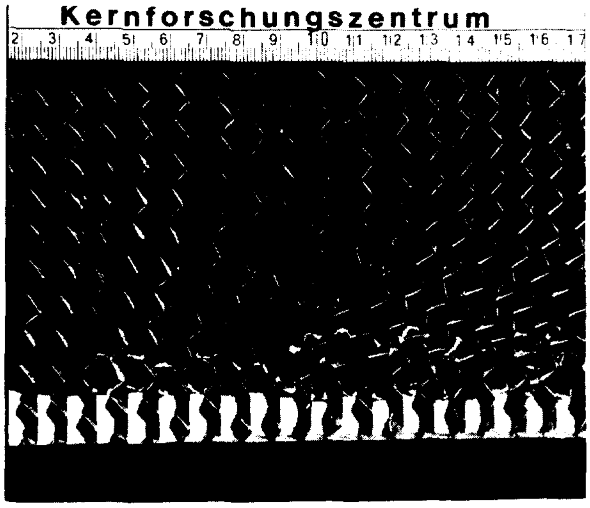
Fig. 5: Close-up of play and torn upstream ends of pleats.



3.



4.



5.

Structural limits varied among filter manufacturers by about 50 % for both the deep pleat and mini pleat designs. This is observed in comparisons of results for AP & AN with those of CN filters and of results for RNF with those of DPF filters. The value of 7.6 kPa listed for the DN filters is probably somewhat higher than the actual value. These filters, outfitted with a webbing of glass fiber between the ends of the separators and the filter medium, likely failed earlier within the pack than on the downstream filter face monitored during tests.

The influence of temperature between 20 and 80 °C on the ΔP at failure remains inconclusive due to the limited number of tests performed at other than 50 °C, the relatively large scatter in results as well as the weak effect anticipated. There is some circumstantial evidence to suggest that airflows with large and numerous fluctuations in humidity and differential pressure can play a role in structural failure. A tendency toward failure with less service time and at lower ΔP may be expected for such cases. Both moisture cycling and temperature remain topics for further study.

One mechanism which contributes to both failure modes is the loosening of the filter pack. An example of the resulting play between adjacent pleats of the filter medium is evident in Fig. 5. Every filter tested sustained a loss in pack tightness which at zero flow rate resulted in an average of 20 mm of total play on the downstream filter face. The amount of play in the filters tested ranged from 10-40 mm. Service history or dust loading had no effect on the average value but considerably accelerated the relaxation process. Several loaded filters showed 10 mm of play within 1 h of exposure to airflow with $\varphi \leq 80$ % r.h.

The geometrical configuration of adjacent separators which makes the relaxation possible is illustrated in Fig. 6. In the lower detail is the most adverse arrangement of the peaks on adjacent separators. Under the combined effects of differential pressure, moisture, and exposure time the filter medium takes on the corrugated form of the separators. This results in a loss of tightness in the pack and the creation of play which allows those pleats that are internally pressurized to grow in width.

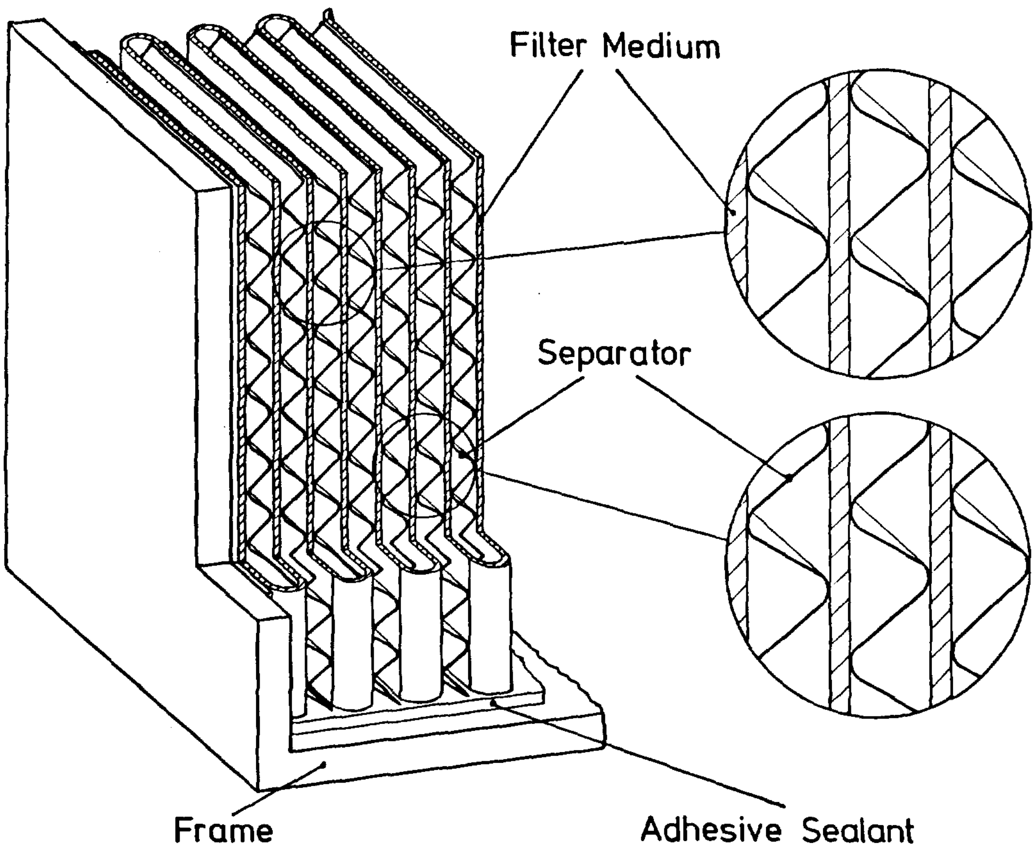


Fig. 6: Two extreme cases for the arrangement of adjacent separators in a deep pleat HEPA-filter pack.

This increase in pleat width is a second mechanism for both failure modes. For the downstream ends of the pleats, any increase in width can have several adverse consequences, one of which is greater circumferential tensile stresses in the filter medium /12/. Additionally, contact and interaction between the edge of the separator and the end of the pleat become possible by a movement of the pleat end into the edge of the separator during swelling /54/ or a movement of the then loosely bound separator into the pleat end after swelling.

Ruptures in the Downstream Ends of the Pleats. Quantitative knowledge of the effects of wetness, creases, prior moisture exposure, and dust loading on filter medium strength are needed to evaluate the extent to which decreases in strength contribute to ruptures and tears in the filter medium. Consequently, samples of filter medium were removed from a total of 40 new, loaded and humidity tested 610x610x292-mm filters for testing according to DIN 53 857 Part 2. These tensile strength tests were performed with wet and dry specimens 50 x 250 mm in size using a ZWICK Model 1425 testing machine. To thoroughly saturate the wet specimens, they were submerged in water and conditioned for 1-2 h in a vacuum chamber at an absolute pressure of 5 kPa and at the temperature of the prior filter humidity test, usually 50 °C. Dry specimens were tested at 20 °C. The average values of tensile strength and of elongation at rupture are listed in Table 2 for groups of samples classified by filter manufacturer and by filter service and test histories.

Results are presented for specimens oriented in the cross direction of the filter medium as well as in the machine direction, with and without the creases caused by pleating. Analysis of the averages in the machine direction for all filter types, excluding those of the prototype MP filter, show that acting alone wetness caused a 60 % decrease in tensile strength, creases 45 %, humidity-test moisture exposure with subsequent drying 40 %, and dust loading (for creased samples) 5 %. If the decreases are assumed to be cumulative the residual tensile strength can be calculated to be 13 % of the value for a new uncreased filter paper. The residual value for AN and CN filters after pleating, loading, high humidity exposure (& drying), and wetting is a comparable, yet even lower, 7 %. That for the single, only partially water repellent CN filter was 3.5 %. This indicates that the effects of these factors acting together can be greater than the sum of their individual effects acting alone. The cumulative effect of the above 4 factors on elongation was to decrease the values for AP (& AN) and CN filters by 40 and 80 %, respectively.

It is noted for CN filters that dust loading brought about an avg. 35 & 70 % increase, resp., in tensile strength of uncreased dry and wet specimens without prior high humidity exposure. This effect was reduced for wet and dry samples after exposure to high humidities during filter testing. Creased, loaded, CN dry samples without humidity exposure exhibited no increase in tensile strength; probably because folded creases do not load during service (see Figs. 8,9). Similar wet loaded samples showed, as yet inexplicably, a 45 % larger residual strength than wet clean ones. However, creases, humidity exposure (& drying),and wetting produced equal and the lowest values,in both clean and loaded samples.

Table 2: Results of tensile strength tests of filter medium samples removed from new and humidity tested HEPA filters.

Samples of Filter Media			Tensile Strength (kN/m)						Elongation (^{m}/m %)	
			Dry			Wet			Dry	Wet
Manf. Code	Pretest Cond.	No. of Filters Sampled	XD$^+$	MD^{++} (w/o fold)	MD (w/fold)	XD	MD (w/o fold)	MD (w/fold)	MD (w/fold)	MD (w/fold)
AP	cl./new	1	0.92	1.08	0.33	0.63	0.86	0.24	0.29	0.30
AP	cl./h-t.	3	0.47	1.03	0.20	0.37	0.72	0.13	0.22	0.20
AN	cl./new	1	0.52	0.83	0.42	0.35	0.63	0.25	0.31	0.26
AN	ld./h-t.	6	0.56	0.85	0.19	0.28	0.40	0.06	0.29	0.30
CN	cl./new	3	0.57	0.87	0.38	0.14	0.20	0.11	0.51	0.47
CN	cl./h-t.	5	0.34	0.50	0.17	0.14	0.20	0.07	0.29	0.32
CN*	cl./h-t.	1	0.26	0.49	0.13	0.02	0.03	0.03	0.36	0.32
CN	ld.	1	0.82	1.08	0.36	0.25	0.32	0.16	0.34	0.40
CN	ld./h-t.	10	0.56	0.81	0.21	0.17	0.21	0.06	0.36	0.37
DN	cl./new	2	0.37	0.81	0.31	0.06	0.14	0.05	0.35	0.42
DN	cl./h-t.	3	0.64	0.83	0.29	0.13	0.17	0.09	0.43	0.45
VM	cl./new	1	0.69	1.05	0.58	0.13	0.20	0.16	0.58	0.48
VM	cl./h-t.	1	0.41	0.74	0.31	0.21	0.44	0.17	0.47	0.50
MP**	cl./new	1	10.3	14.7	14.7	6.9	11.8	11.6	1.52	1.52
MP**	cl./h-t.	1	9.8	16.0	15.5	7.4	12.9	11.4	1.91	1.50

$^+$cross direction $^{++}$machine direction *partially water repellent medium **Lydair 255LW1 medium
cl. = clean filter h-t. = humidity tested filter ld. = loaded filter

In addition to exhibiting the lowest values of tensile strength and of elongation to rupture, the ends of the pleats are also more vulnerable to wetting by liquid water. This was observed both during the conditioning of wet samples for tensile tests and during water repellency tests on samples from the same 40 filters. An apparatus equivalent to the Q101 Water Repellency Indicator of MIL-F-0051079B was used to test 10 creased and 10 uncreased samples from each filter. With the exception of the one partially water repellent filter medium (value of 2 kP),and filters from one manufacturer who makes no claim to meet the military standard, the averages for uncreased samples from the other 34 filters all exceeded the 5 kPa minimum value. The results for creased samples were significantly different; only for new untested filters were water repellency values above 5 kPa evident. Average values for loaded and humidity tested filters in the groupings of Table 1 ranged between 1 and 4 kPa. A number of filters, particularly loaded humidity tested ones, showed consistent values of zero for creased samples and hydrophilicly absorbed water along the creases within 1 sec after initial contact with a water surface.

The loosening of the filter pack and swelling of the pleats together with the results of the tensile strength and water repellency tests provide strong evidence as to why the downstream ends of the pleats are so prone to rupture in high humidity airflows.

Given values for the total amount of play in the pack, x_p, the tensile strength across the wet ends of the pleats, and the failure differential pressure, the validity of Equation (1),

$$\sigma_c = \frac{r \cdot \Delta P}{d} \quad , \qquad (1)$$

which was proposed /53/ and qualitatively confirmed /47/ by Ruedinger for dry swollen pleats, can be checked for wet swollen pleats of lesser width. In Equation (1), σ_c is the circumferential tensile stress in the pleat end, r the radius of a downstream pleat end, ΔP the differential pressure across the filter, and d the thickness of the filter medium. If it is assumed that up to 1/4 of the total play is at any one time available to any one pleat for room to swell, then the maximum radius of a swollen pleat end is approximated by

$$r \approx r_0 + \frac{x_p}{8} \quad , \qquad (2)$$

where r_o is the average initial radius of the downstream pleat ends before the humidity test. For the filters tested , the calculated values of r ranged from 3-7 mm and the recorded values from 2-10 mm. These calculated values of r, based on the amount of play in the filter pack at failure, were used because the video-recorded values were available for only some few cases. How well Equation (1) predicts the circumferential stresses in the pleat end is seen in Fig. 7 where the actual structural limits of 26 filters tested are plotted against respective calculated structural limits. This diagram for new and loaded deep pleat 610x610x292-mm wooden frame filters from 3 different manufacturers indicates with a correlation coefficient of 0.56 a qualitative confirmation of Eq. (1). The slope of the linear regression curve fit to the points is seen to be a factor of 3. 5 smaller than the value needed for a quantitative confirmation. One explanation for this is that Eq. (1) does not take into account the additional stresses in the pleat end caused by any contact and interaction with the enclosed separator.

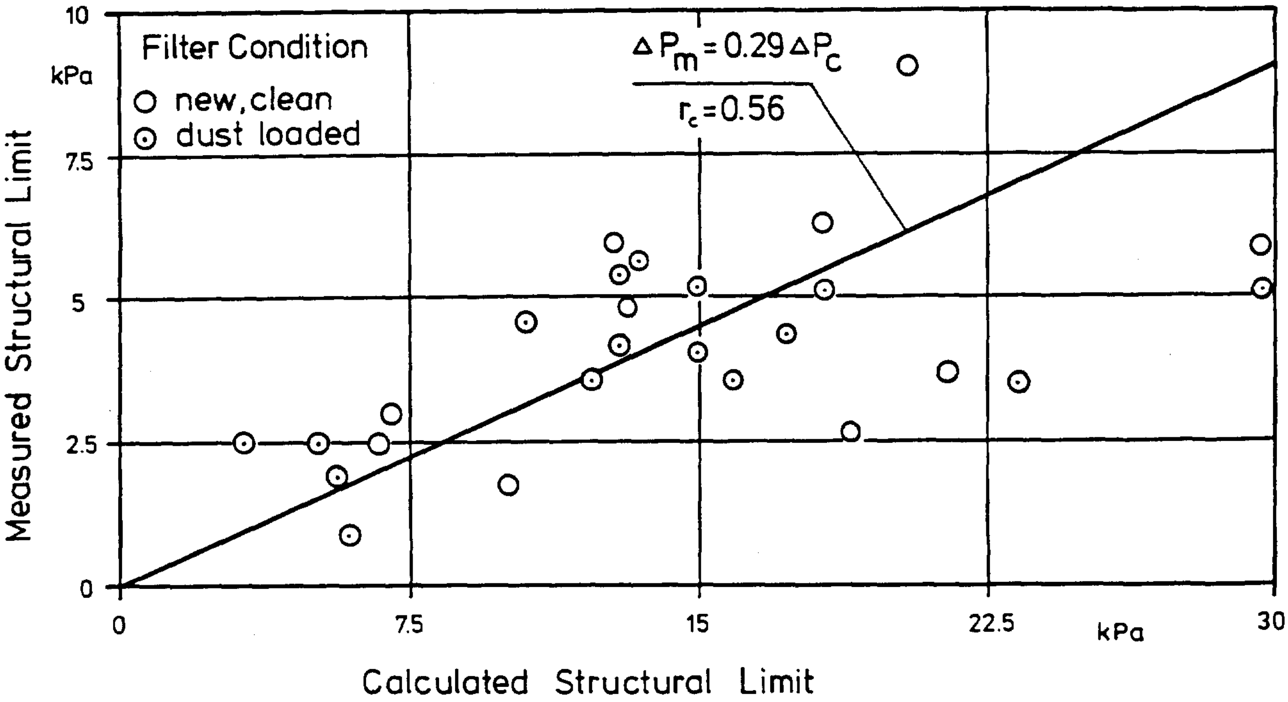


Fig. 7: Comparison of the measured to the calculated differential pressure at structural failure for the case of a wet swollen pleat end.

Damage caused by such contact is shown in Fig. 8. Here, the upstream side of pleat ends removed from the downstream face of clean and dust loaded filters after humidity tests are shown. The small gap between separators and filter medium, typically 0-3 mm in commercial filters, is illustrated in Fig. 9 by similar photos taken of dust loaded pleat ends after humidity tests. The horizontal white lines are the creases in the end of the pleat whereas the vertical ones indicate the areas of contact between the sides of the pleats and the separators during service.

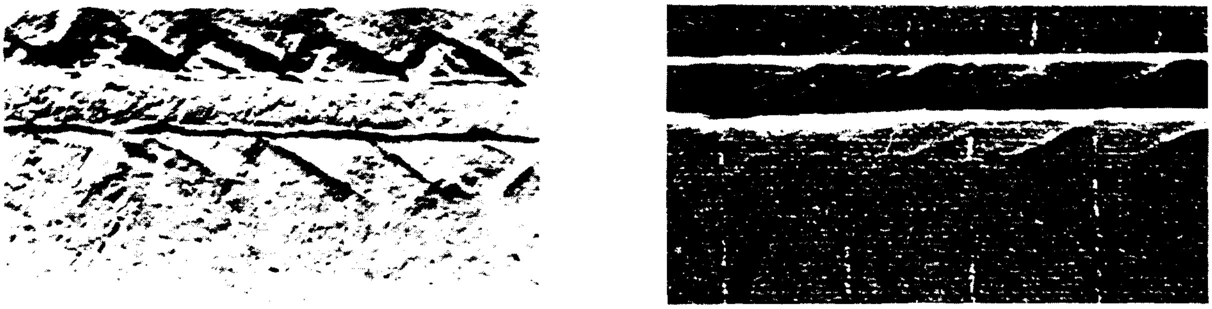


Fig. 8. Damage caused to downstream pleat end of clean (l.) and dust loaded (r.) filter medium by contact with the edge of an aluminum separator during humidity test.

The black intervals by which each vertical line fails to fully intersect with a horizontal line is a record of how large the gap between separator edge and pleat end was during service. The photo on the right in Fig. 9 shows the edge of an aluminum separator at the position it was in with respect to the pleat end during service and the subsequent humidity test. These four photos illustrate how separators can threaten filter medium integrity and how little relative movement is required to bring the edges of the separators into contact with the filter medium. They also explain why Equation (1) does not fully account for all the stresses in the pleat ends during filter exposure to humid airflows.

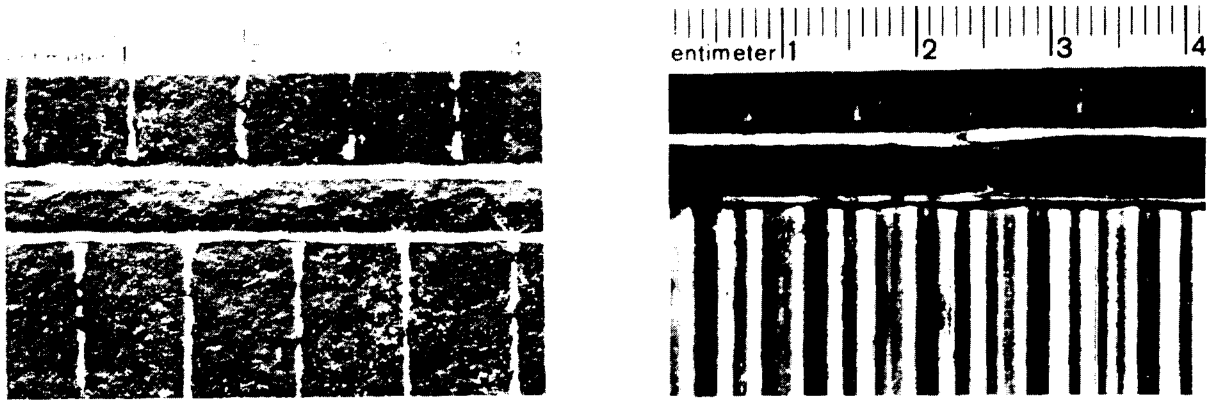


Fig. 9. Record of the gap between the edge of separators and the ends of pleats during service for two dust loaded HEPA filters.

Tears in the Filter Medium Along the Sides of the Pleats. The loosening of the filter pack also leads to the tears observed in the filter medium along the sides of the pleats, frequently for the full depth of the filter. These tears were found to run parallel to the edge of the adhesive on the medium, within 30 mm of the frame on both the top or the bottom of the filter pack. The number of pleats damaged per filter ranged from 3 to 50 for the 7 new and the 9 loaded filters that exhibited this failure mode. A filter pack that is no longer tight leaves the pleat room, not only for swelling but also for movement and thus subjected to the influence of the aerodynamic forces in the airstream.

The pleats in a pack loosened by humidity and differential pressure can have two degrees of freedom in movement. One is a lateral motion made possible by the play between the pleats. The second is a relative lateral motion of the upstream end of a pleat with respect to the downstream end. The potential for this relative motion to occur is increased by the 8-mm, or 40 %, greater amount of average total play measured on the upstream side of the filters tested. Tears in the upstream ends of pleats were observed to appear twice as frequently and sooner than on the downstream side, and in some cases sooner than the rupture of the swollen downstream ends of pleats.

A plausible explanation for the locations of these tears is evident if the side of a swollen pleat is modeled as a beam with fixed ends and a distributed load, and without the support provided by the adjacent downstream separator. The maximum stresses are found to occur at the top and bottom of the pleat and for a conventional filter medium, ruptures can be expected for a $\Delta P < 1$ kPa. This simple model does not take into account any residual support from the separators or any fatigue factor due to pleat movements. For several filters under test, lateral oscillations with an amplitude of up to 5 mm and a frequency of about 1/60 Hz have been observed among the downstream ends of the pleats /55/.

These tears then can be attributed to the longitudinal stresses in the sides of the pleats, stresses normally present due to filter ΔP, but increased by the swelling and movement of the pleats and by decreases in the rigid support of adjacent separators. Moisture and the fatigue due to pleat movement reduce the ability of the filter medium to withstand these increased stresses. This failure mode requires more study before a suitable model can be developed and verified.

First Improvements in Filter Structural Strength Under Humid Airflows

Several options are available to improve filter construction and thus prevent structural damage to glass-fiber HEPA filter units in humid airflows. An increase in the tensile strength of the filter medium or in the stability of the filter pack are two principal ones. Measures to protect the ends of the pleats from contact with the separators constitute a third option.

Tests of Prototype Filters with Glass Fiber Medium. Initial tests of 3 deep pleat 610x610x292-mm prototype filters which incorporated some of these countermeasures have been carried out. A glass fiber filter medium+ reinforced by a fiber glass cloth was used to fabricate++ 1 filter with a wooden frame and an elasto-

[+] Grade 255LW1 from Lydall Inc. (See Table 2 for tensile strengths.)

[++] Manufacturer: Mitchell Cotts Air Filtration Ltd.

meric adhesive and 1 filter with a metal frame and a ceramic adhesive. Each was tested with high humidity airflow in the manner described for the structural tests of the commercial HEPA filters.

Under fog conditions and despite a resultant 80 mm of play in the pack, the wooden frame filter sustained without damage a differential pressure of 10 kPa, the maximum ΔP of the test facility at 1700 m^3/h. Filter DOP filtration efficiency after the humidity test and drying of the filter was measured to be 99,98 % compared with a value of 99,99% before the test. The metal frame filter, with a maximum 60 mm of resultant play, withstood a 5-kPa differential pressure for 2 h before the ceramic adhesive separated abruptly from the frame at 7 kPa. These results represent, by factors of 2-3, increases in failure differential pressure above the values for respective commercial filters given in Table 1. This improvement was achieved with only an increase in the tensile strength of the filter medium, without any attempt to minimize the loosening of the pack.

To evaluate one possible method to minimize the loosening of the filter pack, 1 prototype filter with a wooden frame , conventional glass fiber medium, and special aluminum separators was also tested in an airflow which had a liquid moisture content of some 10 g/m^3. These separators were formed such that the corrugations were slightly inclined to the direction of the airflow and were installed so that the corrugations on adjacent separators were not parallel but instead skew with respect to each other. This filter failed at a differential pressure of 9 kPa during exposure to the fog conditions, but more importantly, the play in the filter pack was limited to < 5 mm. This represented less than 6% of the final play exhibited by the wooden frame filter with the reinforced medium.

That the increase in filter structural strength and the minimization of the resultant play in the pack were achieved by two different means, indicates the potential for even greater improvement in filter strength in a combination of the two. Tests of prototype filters with both the reinforced filter medium and the special separators are planned for the near future.

Initial Investigations of Adsorption Phenomena

To investigate the effects of dust loading, humidity, exposure, and fiber material on the water vapor adsorption characteristics of HEPA filters, samples were removed from a total of 14 new and humidity tested filters and used to determine the adsorption isotherms for the filter media. Preparation of the 100-mg samples consisted of drying them at 65 °C and an absolute pressure of 100 Pa for 48 h. With a discontinuous gravimetric method /56/, the adsorbed moisture contents at 25 °C were established after an equilibration time of 14 d. The 7 different values of relative humidity between 32 and 97 % r.h. were established with an uncertainty of $\leq$ 1.5 % r.h. by solutions of salt-saturated water.

The range of adsorption isotherms for samples from 9 clean glass fiber filters is shown in Fig. 10. This range is bounded

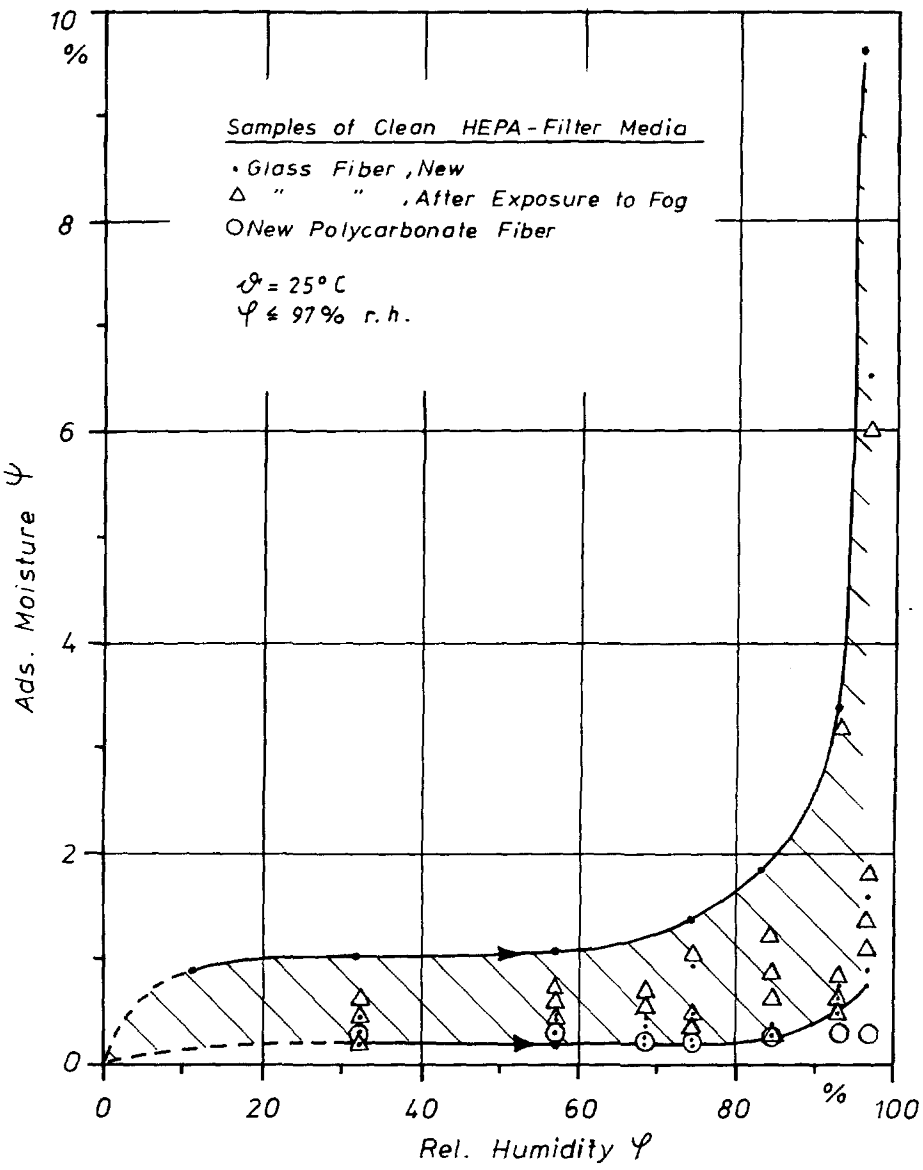


Fig. 10: Range of adsorption isotherms for media removed from 10 clean, new and humidity tested HEPA filters.

by points on isotherms for 3 new filter media, because points on the top curve represent the higher respective values for two intersecting isotherms. The curves of 2 other new and 4 humidity tested filters all lie within this range. No significant influence from fog exposure on adsorption is evident for the clean glass fiber filters sampled. The values of ϕ at 97 % r.h. are noted to vary by 1 order of magnitude for the media tested so far. Only the points for the new polycarbonate microfiber medium+ /50/ show no increase in moisture content up to 97 % r.h. It is also of interest to note that the sorption isotherm for the partially water repellent medium lies at the bottom of this range with nothing in

---

+ Manufacturer: Carl Freudenberg, Weinheim, FRG.

particular to distinguish it from those of the water repellent media.

When compared to that of Fig. 10 the range of isotherms for 4 dust loaded glass fiber filter media in Fig. 11 shows clearly the effects of dust loading on ψ. At 97 % r.h. the amount of adsorbed moisture can be a factor of 5-50 greater

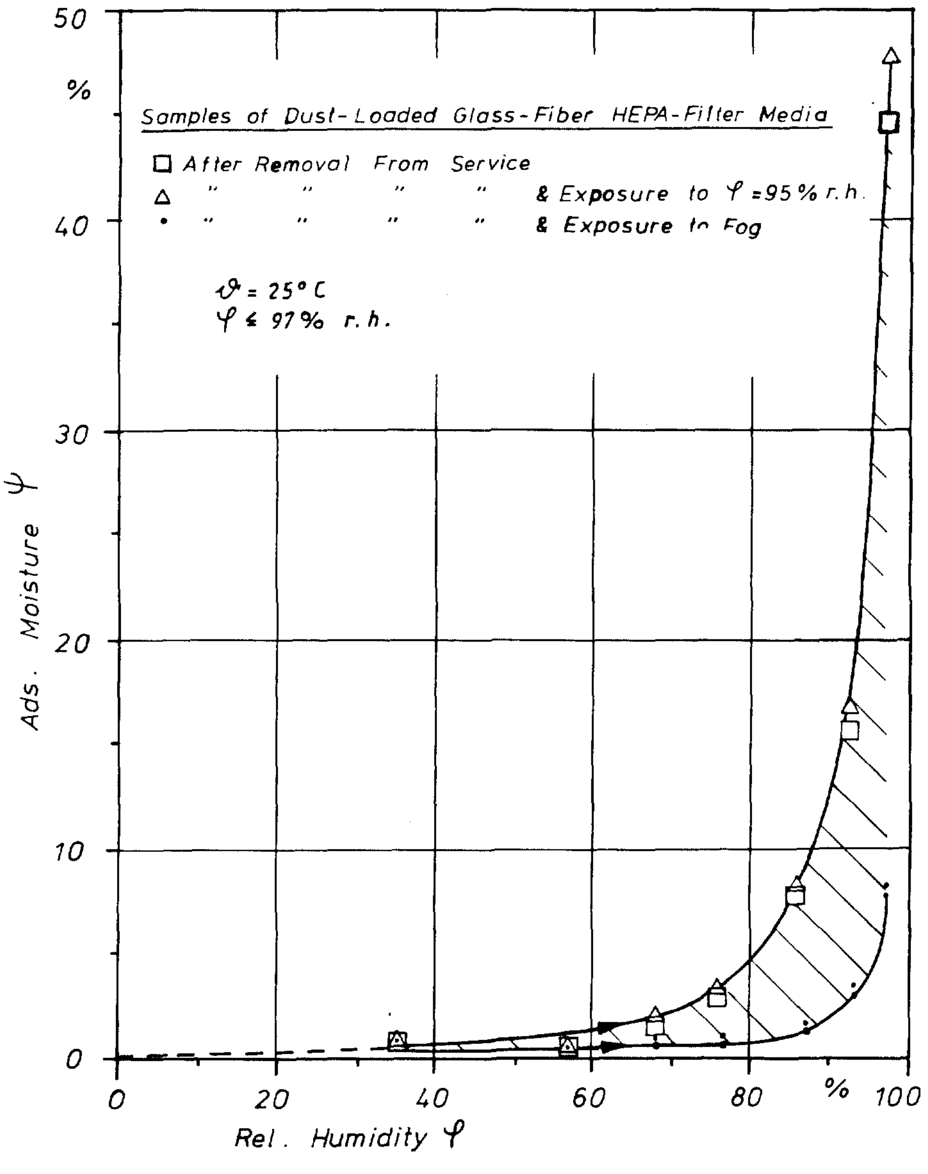


Fig. 11: Range of adsorption isotherms for media removed from 4 dust loaded HEPA filters.

in a dust loaded as compared to a clean filter medium. The two media, with ψ values presumably reduced through exposure to fog conditions, show maximum values of ψ corresponding approx. to the equivalent ϕ of the most hygroscopic clean media. Comparison of the points for the dust loaded filter without prior high humidity exposure, to those of a filter humidity tested with up to 95 % r.h. show no significant difference. This suggests that for the type of dust involved here, φ > 95 % r.h. is required to change the structure of the adsorbing surfaces enough to alter the hygroscopic properties of the dust loaded filter medium. This is somewhat unexpected given that values of φ as low as 80 % r.h. normally alter the dust loaded fiber structure enough to reduce the ΔP of a loaded filter almost to that of a new filter /12/.

These results are still preliminary and require further study and investigation. Sorption kinetics and the effects of temperature and fiber diameter on adsorption isotherms are also topics pending the analysis of already recorded experimental data.

IV. Conclusions

Despite the many threats posed by high humidity airflows to the structural integrity of commercial glass fiber HEPA filters, studies of the modes and mechanisms of structural failure can lead to significant and cost effective improvements in filter structural strength. As a result, the reliability and safety margins of filters exposed to moisture, particularly in accident situations, can be increased considerably. The preliminary results obtained for the 3 new prototype filters need to be extended and verified for filters with combined improvements and also for filters with dust loads and radiation exposure.

Once structural integrity is assured, tests to evaluate filtration efficiencies at air humidities up to 100 % r.h. can be carried out. Studies of the incorporation of water into the filter medium should be continued in order to minimize humidity related increases in filter differential pressure and to model filter ΔP as a function of airstream and filter medium parameters in support of computer code accident analyses. The need for an international, stringent test standard for the qualification of HEPA-filter units under high humidity airflows also provides an important item on the agenda for future work.

Acknowledgements

The authors wish to express their appreciation to Messrs. H. Leibold, E. Demmer, and U. Geckle for their valuable contributions to the filter test program and to Mr. H.-G. Dillmann who kindly made possible the use of the TAIFUN test facility. Thanks are also due to Mr. W. Wolf of the Federal Institute for Nutrition in Karlsruhe for the adsorption isotherm data and related consultations.

V. References

/ 1/ Carbaugh, E.H.
A Survey of HEPA-Filter Experience,
CONF-820 833 (1983) p. 790 ff.

/ 2/ Glissmeyer, J.A.; Mishima, J., and Bamberger, J.A.
Prototype Firing Range Air Cleaning System,
CONF-840 806 (1985) p.846 ff.

/ 3/ Moeller, D.W. and Sun, L-S.C.
Failures in Air-Cleaning, Air-Monitoring, and Ventilation Systems in Commercial Nuclear Power Plants, 1978 - 1981,
Nuclear Safety, 24 (3) (1983) p. 352 ff.

/ 4/ Moeller, D.W.
Failures in Air-Monitoring, Air-Cleaning, and Ventilation Systems in Commercial Nuclear Power Plants (Jan. 1, 1975 - June 30, 1978),
Nuclear Safety, 20 (2) (1979) p. 176 ff.

/ 5/ Moeller, D.W.
Current Challenges in Air-Cleaning at Nuclear Facilities,
Nuclear Safety, 18 (5) (1977) p. 633 ff.

/ 6/ Moeller, D.W.
Problems in Nuclear Air Cleaning Systems,
Nuclear Safety, 16 (4) (1975) p. 469 ff.

/ 7/ Bellamy, R.R.
Experience with High Efficiency Particulate Air Filters at United States Nuclear Installations,
Proc. of Seminar on High Efficiency Aerosol Filtration in the Nuclear Industry, Comm. of the European Communities, Luxembourg (1977).

/ 8/ Air Cleaning in Accident Situations,
Report to CSNI by an NEA Group of Experts
ISBN 92-64-12633-3
OECD, Paris, 1984.

/ 9/ Burchsted, C.A.; Fuller, A.B., and Kahn, J.E.
Nuclear Air Cleaning Handbook,
ORNL, Oak Ridge, TN, ERDA 76-21, 1976.

/10/ Alvares, N.J. et al.
Fire Protection Countermeasures for Containment Ventilation Systems,
CONF-801 038 (1981) p. 1213 ff.

/11/ Glibert, R.G. et al.
Design of a PWR Gaseous Radwaste Treatment System Ensuring Safe Control of Gaseous Radionuclides Released Under Normal and Severe Conditions,
in: Management of Gaseous Wastes From Nuclear Facilities, IAEA, STI/PUB/561, Vienna, 1980, p. 545 ff.

/12/ Ruedinger, V.; Ricketts, C.I., and Wilhelm, J.G.
Limits of HEPA-Filter Application Under
High-Humidity Conditions,
CONF-840 806 (1985) p. 1058 ff.

/13/ Arnitz, Th. and Ruedinger, V.
Untersuchungen zur Genauigkeit zweier Rechencodes zur
Vorhersage der Störfallauswirkungen innerhalb kerntechnischer Lüftungsanlagen,
in: Gaseous Effluent Treatment in Nuclear Installations,
ed. G. Fraser & L. Luykx, CEC, EUR 10580, Graham & Trotman,
London (1986) p. 416 ff.

/14/ Thurner, F. and Stietz, M.
Bestimmung der Sorptionsisothermen lösungsmittelfeuchter Sorbienten nach der Durchströmungsmethode,
Chem. Eng. Proc., 18 (1984) p. 333 ff.

/15/ Kantro, D.L.; Brunauer, S., and Copeland, L.E.
BET Surface Areas - Methods and Interpretations,
in: The Solid-Gas Interface, ed. E.A. Flood,
Edward Arnold Publishers Ltd., London, 1967, p. 413 ff.

/16/ Brunauer, S.; Deming, L.S.; Deming, W.E., and Teller, E.
J. Am. Chem. Soc. 62 (1940) p. 1723 ff.

/17/ Lück, W.
Feuchtigkeit: Grundlagen - Messen - Regeln,
R. Oldenbourg, München - Wien, 1964.

/18/ Poersch, W.
Sorptionsisothermen: Ihre Ermittlung und Auswertung,
Die Stärke 11 5 (1963) p. 405 ff.

/19/ Kadlec, O. and Dubinin, M.M.
Comments on the Limits of Applicability of the
Mechanism of Capillary Condensation,
J. Colloid & Inter. Sci. 31 4 (1969) p. 479.

/20/ Kawasaki, K.; Senzaki, K., and Tsuchiya, I.
Adsorption Studies of Water Vapor on Porous Glass,
J. Colloid Sci. 19 (1964) p. 144 ff.

/21/ Krischer, O. and Kast, W.
Die wissenschaftlichen Grundlagen der Trocknungstechnik,
3. Auflage, 1. Bd., Springer-Verlag,
Berlin - Heidelberg - New York, 1978.

/22/ Tierney, G.P. and Conner, W.D.
Hydroscopic Effects on Weight Determination of
Particulates Collected on Glass-Fiber Filters,
American Industrial Hygiene Association Journal,
(August 1967) p. 363 ff.

/23/ Hofmann, W.M.
Feuchtigkeitsaufnahme von Schwebstoffiltern,
Zeitschrift für Heizung, Lüftung, Klimatechnik
und Haustechnik (HLH), 25 3 (1974) p. 77 f.

/24/ Rachinskii, V.V.
The General Theory of Sorption Dynamics and
Chromotography, (translation from the Russian),
Consultants Bureau, New York, 1965.

/25/ Mantell, C.L.
Adsorption,
1st Edition, McGraw-Hill Book Co. Inc.,
New York and London, 1945.

/26/ Peters, A.H.
Applications of Moisture Separators and Particulate
Filters in Reactor Containment,
USAEC DP-812, Savannah River Laboratory, 1962.

/27/ Jones, L.R.
Effects of Radiation on Reactor Confinement
Systems Materials,
CONF-720 823 (1972) p. 655 ff.

/28/ Schwalbe, H.C.
Redevelopment of the Savannah River Laboratory
Moisture Resistance Test for Filter Paper,
CONF-680 821 (1968) p. 86 ff.

/29/ Belvin, W.L. et al.
Development of New Flouride Resistant HEPA Filter
Medium: Final Report,
USERDA, TID-26649, Savannah River Operations Office, 1975.

/30/ Davis, R.J. et al.
The Effects of Moisture on the Character and
Filtration of UO_2-Stainless Steel Aerosols,
in: Treatment of Airborne Radioactive Wastes,
IAEA STT/PUB 195, Vienna (1968) p. 345 ff.

/31/ Robinson, K.S. et al.
In-Service Aging Effects on HEPA Filters,
in: Gaseous Effluent Treatment in Nuclear Installations,
ed. G. Fraser & L. Luykx, CEC, EUR 10580,
Graham & Trotman, London (1986) p. 60 ff.

/32/ Kvetoslav, R. et al.
Methodik und Untersuchungen zur Beständigkeit von
Mineralfasern in vitro und in vivo,
Staub-Reinhalt. Luft 44 4 (1984) p. 169 ff.

/33/ Horak, H.L. et al.
Structural Performance of HEPA Filters Under
Simulated Tornado Conditions,
LA-9197-MS, Los Alamos National Laboratory, 1982.

/34/ Deworm, J.P.
Laser Particle Spectrometry for Testing HEPA -Filtration Systems in the Nuclear Industry, Filtration & Separation July/Aug. (1984) p. 254 ff.

/35/ Ensinger, U.; Ruedinger, V., and Wilhelm, J.G.
A Procedure to Test HEPA-Filter Efficiency Under Simulated Accident Conditions of High Temperature and High Humidity,
CONF-840 806 (1985) p. 1036 ff.

/36/ Gilbert, H. and Palmer, J.H.
High Efficiency Particulate Air Filter Units,
TID-7023, USAEC, 1961.

/37/ Burchsted, C.A.
Environmental Properties and Installation Requirements of HEPA Filters,
in: Treatment of Airborne Radioactive Wastes,
IEAE, STI/PUB/195, Vienna, 1968, p. 175 ff.

/38/ Stratmann, J.
Bericht eines Großverbrauchers von Schwebstoffiltern der Klasse S,
in: Proc. Seminar on High Efficiency Aerosol Filtration, Comm. European Community, Luxembourg, (1977), p. 411 ff.

/39/ Fenton, D.L. and Dallman, J.J.
HEPA-Filter Loading by Simulated Combustion Products, Report submitted by New Mexico State University Mechanical Engineering Department to Los Alamos National Laboratory, Sept. 1982.

/40/ Hofmann, W.M.
Druckverlust feuchter Schwebstoffilter,
Zeitschrift für Heizung, Lüftung, Kimatechnik und Haustechnik (HLH), 25 11 (1974) p. 370.

/41/ Jones, L.R.
High Efficiency Particulate Air (HEPA) Filter Performance Following Service and Radiation Exposure,
CONF-740 807(1974) p. 565 ff.

/42/ Collins, R.D.; Hillary, J.J., and Taylor, J.C.
Air Cleaning for Reactors with Vented Containment,
CONF-660 904 (1966) p. 419 ff.

/43/ Gunn, C.A. and Eaton, D.M.
HEPA-Filter Performance Comparative Study,
CONF-760 822 (1977) p. 630 ff.

/44/ Pratt. R.P.
The Performance of Filters Under Hot Dynamic Conditions,
in: Gaseous Effluent Treatment in Nuclear Installations,
ed. G. Fraser & L. Luykx, CEC, EUR 10580,
Graham & Trotman, London (1986) p. 824 ff.

/45/ Richardson, W.J. and Palmer, J.H.
The Installation, Handling, and Storage of High
Efficiency Filters,
TID-7593, USAEC, 1960, p. 182 ff.

/46/ Thaxter, M.D.
Condition of Commercial High-Efficiency Filters
Upon Receipt or Installation
TID-7593, USAEC, 1960, p. 157 ff.

/47/ Ruedinger, V.; Ricketts, C.I., and Wilhelm, J.G.
Versagensgrenzen und Schadensmechanismen von Schwebstoffiltern unter Beanspruchung durch trockene Luft
hoher Strömungsgeschwindigkeit,
in: Gaseous Effluent Treatment in Nuclear Installations,
ed. G. Fraser & L. Luykx, CEC, EUR 10580,
Graham & Trotman, London (1986) p. 792 ff.

/48/ NRC
Regulatory Guide 1.52, Revision 2,
USNRC, Washington, D.C., 1978.

/49/ Dillmann, H.-G.
Ergebnisbericht über Forschungs- und Entwicklungsarbeiten
1981 des Laboratorium für Aerosolphysik und Filtertechnik,
KfK-Bericht 3294, (1982) p. 12.

/50/ Alken, W. et al.
Development of a HEPA Filter with High Structural Strength
and High Resistance to the Effects of Humidity and Acid,
CONF-840 806 (1985) p. 1085 ff.

/51/ HDR Sicherheitsprogramm - Gesamtprogrammphase II, PHDR
Arbeitsbericht 05.19/84, Kernforschungszentrum Karlsruhe
GmbH, Karlsruhe, FRG, Jan. 1984.

/52/ Ohlmeyer, M.
Vor-Ort-Prüfung von Schwebstoffiltern und Entnahme von
Sorptionsmaterialproben bei Iod-Sorptionsfiltern,
Atomenergie- Kerntechnik 40 (1982) p. 259 ff.

/53/ Ruedinger, V. and Wilhelm, J.G.
HEPA-Filter Response to High Airflow Velocities,
CONF-820833 (1983) p. 1069 ff.

/54/ Ruedinger, V.; Ricketts, C.I., and Wilhelm, J.G.
Development of Glass-Fiber HEPA Filters of High Structural Strength Based on the Establishment of Failure Mechanisms,
in: Proceedings of the 19th DOE/NRC Nuclear Air Cleaning Conference, Seattle, WA, Aug. 1986, to be published.

/55/ Leibold, H.
Private Communication, KfK, July 1986.

/56/ Wolf, W.; Spieß, W.E.L. and Jung, G.
Standardization of Isotherm Measurements, (COST - Project 90)
in: Properties of Water in Foods, eds. D. Simatos, J.L. Multon, NATO ASI Series, Series E,
in: Applied Sciences No. 90, Martinus Nijhoff Publishers, Dordrecht - Boston - Lancaster, 1985, p. 661 ff.

DISCUSSION

SCRIPSICK: Please comment on the potential effects these extreme conditions might have on penetration and please refer to Normann's paper.

RICKETTS: We have not yet performed filtration efficiency measurements on HEPA filter units under high humidity conditions. However, a suitable method which employs a submicron TiO_2 condensation aerosol and AAS analysis has been developed at Karlsruhe Nuclear Research Center and will soon be used to test full scale filters at air relative humidities up to 100%. Published measurements of filtration efficiency under high humidity conditions are somewhat contradictory. The work of Adams, Davis, et al. at Oak Ridge National Laboratory some 20 years ago seem to be the most credible. Tests performed on samples of filter medium with a UO_2-stainless steel aerosol showed increases of up to 1 order of magnitude in penetration after exposure to 100% RH. Reference No. 30 in our paper provides a good summary of the work at Oak Ridge. The measurements presented here by Mr. Normann show the same tendency toward increased penetration after exposure of the filter medium to high humidity.

ETTINGER: How does the type of dust affect the sorption of water and the resulting change in pressure drop characteristics? Would this effect be as much as an order of magnitude?

RICKETTS: The material constituents, the particle size, and the amount of dust on a filter medium greatly influence the hygroscopic, and hence the sorption properties of a dust-loaded filter medium. A salt, such as NaCl, can be very hygroscopic whereas some materials, such as the polycarbonate microfiber filter medium investigated here, show very little if any sorption of water vapor. At relative air humidities above 95%, and temperatures less than 30°C, the absorbed moisture content for different types of dust could vary by up to 2 orders of magnitude. For the dust in our laboratory exhaust airstream, in comparison to clean filters we have measured, the differential pressures of loaded filters were greater by 1 order of magnitude at 1700 m^3/hr and >97% RH. We are currently developing a mathematical model to describe filter ΔP as a function of the amount of adsorbed water on a clean filter.

SANDOVAL: Please explain in further detail the mechanisms involved in increasing the dust loading capabilities by high humidity airflows for the filters studied.

RICKETTS: For the particular room air dust captured in the HEPA filter units that we have tested at 1700 m^3/hr and 50°C, air relative humidities between 70 and 90% RH have reduced the ΔP of dust loaded filters from 2 kPa to values as low as 0.5 kPa within several hours of exposure. This effect has also been observed by other authors for other types of filter media and captured particulates. This phenomenon has been attributed to capillary condensation in the particulate-loaded filter medium and subsequent surface tension effects which draw particles together into agglomerates. It is hypothesized that the formation of the agglomerates partially reopens the pore structure of the filter medium and thus decreases the flow resistance of the filter. For further information and other literature citations, we refer you to reference No. 12 of our paper.

ETTINGER: You talked about the fact that dust loading increased the amount of moisture which can be picked up by a filter and produce a change in pressure drop. I assume that would vary, depending upon the type of dust. What kind of material did you use, and what range would you expect, depending upon the type of dust?

RICKETTS: The amount of water adsorbed by the dust is very much dependent upon the quantity of dust on the filter medium as well as the sorption characteristics of the dust itself. The type of dust that was on the filters that we tested was essentially room air dust. These filters had been removed from the exhaust air system of a laboratory building at Karlsruhe.

ETTINGER: Do you think there would be orders of magnitude difference depending upon the type of dust and whether it is hydroscopic or hydrophilic?

RICKETTS: Yes, definitely; perhaps up to two orders of magnitude difference.

SCRIPSICK: I have a couple of questions, both relating to penetration. Dr. Normann's paper, previous to yours, indicated that there was an increase in penetration associated with humidity-conditioned filters. I am wondering if you would comment on his results and tell me if you have investigated the effect on penetration. You mentioned that the two major problems were Δ P and tensile strength changes. Have you looked at penetration?

RICKETTS: We haven't made penetration measurements under high humidity conditions. That is planned for the future. A colleague in our group is presently working on the development of a test method that uses a titanium dioxide aerosol that will be used to test filter efficiency under both high humidity and high temperature.

SCRIPSICK: Are you familiar with the penetration increases associated with filters exposed to high humidities in Dr. Normann's paper?

RICKETTS: There are contradictory results published in the literature. Work done at Oak Ridge National Laboratory by Mr. Davis and Mr. Adams showed an increase in penetration under high humidity conditions. There was a recent report published by Mr. DeWorm who found decreases in penetration for DOP, NaCl, and uranine aerosols. I am rather skeptical of results for aerosols that are water soluble or that are liquids, when used under high humidity conditions.

CALIBRATION TESTS OF A LASER FLUORESCENT PARTICLE SPECTROMETER

Ph. Mulcey, P. Pybot, J. Vendel
Commissariat à l'Energie Atomique
Institut de Protection et de Sûreté Nucléaire, Département de Protection
Technique, Service de Protection des Installations Nucléaires, Section d'Etudes
Industrielles de Protection - CEN/Saclay, 91191 GIF SUR YVETTE Cédex, France

Abstract

The recent development of laser particle spectrometers enables measurements of higher decontamination factors of filters than those provided by standard test methods. Specific aerosols, as fluorescent ones, increase sensibility and eliminate interference due to background aerosol leaking into the downstream sampling location. The best compromise seems to be reached with a laser fluorescent particle spectrometer.

Before using this kind of method for testing HEPA filters, we have made a calibration of such a device (Laser Fluorescent Particle Spectrometer; PMS model ASAS.XF). Fluorescent (FL) and all particle (ALL) counting modes have been investigated.

For the fluorescent particle mode the DOP solutions are tagged with fluorescent dye (Potomac Yellow) in various concentrations in order to estimate the threshold detection.

For the all particle mode, the calibration is achieved using monodispersed polystyrene latex (PSL) aerosols reselected, for the smaller dimensions (RANGE 1), by an electrostatic classifier (DMPS; TSI model 3071).

The response of the ASAS-XF spectrophotometer to DOP particles in the same size range was finally investigated, using the same electrostatic selection device, and compared to the one obtained from PSL aerosols.

I. Introduction

The conventional standard test methods used to perform efficiency measurements of particulate air filters suffer limitations which makes difficult, or even impossible, the control of tandem HEPA or ULPA filters.

Recent developments in the field of optical single particle counters lead to dispose of instruments performing real time measurements of aerosol size distributions over a wide size range and, by using an appropriate dilution system, over a wide concentration range (1), (2).

The use of a laser beam as a light source can allow, if coupled with a selected dye tracer, a specific detection of the test aerosol avoiding high backgrounds due to particles entering the circuit downstream of the filter.

The calibration tests performed on a laser fluorescent particle spectrometer (PMS Model ASAS-XF) using polystyrene latex (PSL) and DOP aerosols, are presented below.

II. Aerosol Spectrometer Description

The ASAS-XF laser aerosol spectrometer is a single particle counter which allows size classification from the amplitude of the scattered light emitted by the particle when passing through the laser beam. The laser can operate either in the "ALL" mode, for which every particle is taken into account whatever fluorescent or not, or in the "FLUORESCENT" mode for which the classification is achieved for the fluorescent particles only these being detected owing to their fluorescent emission induced by excitation in the laser wavelength if the aerosol particles are tagged with an appropriate fluorescent dye.

The spectrometer is equiped with a 10 mW He-Cd TEM_{00} mode laser tube emitting a 442 nm wavelength. The corresponding dye, which can be used with DOP aerosols, is the Potomak Yellow (excitation wavelength : 440 nm, emission wavelength : 490 nm).

The optical diagram of the instrument is shown in figure 1.

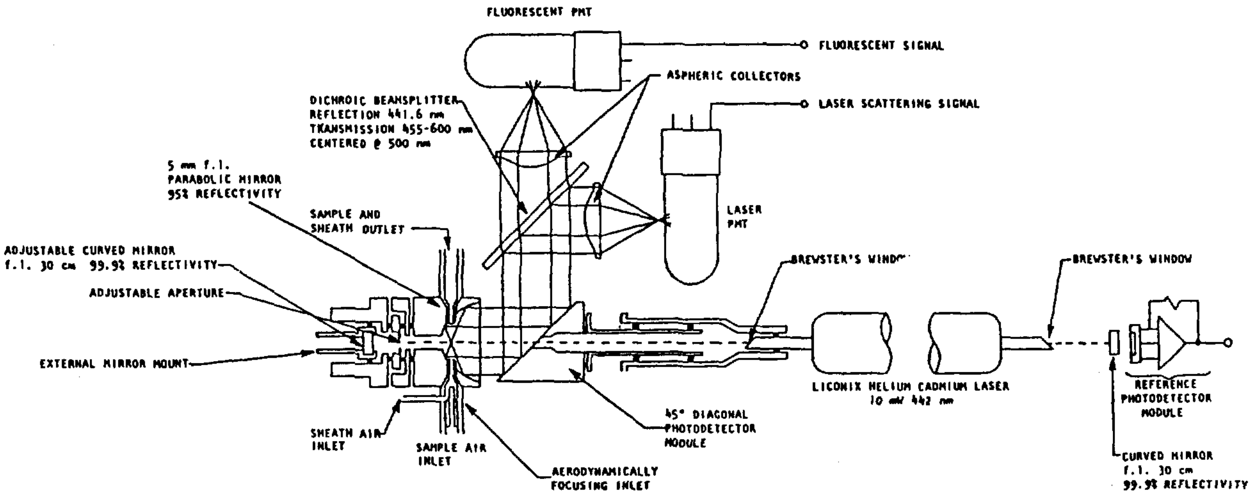


Figure 1 - ASAS-XF Optical system diagram (from Operating Manual)

The ASAS-XF airflow diagram is shown in figure 2.

III. Calibration Tests of the Spectrophotometer

Two different series of tests were conducted :

- tests with polystyrene latex (PSL) monodispersed aerosols in order to check the initial supplier's calibration,
- tests with DOP aerosols. For some of these tests the DOP solution was tagged with Potomak Yellow.

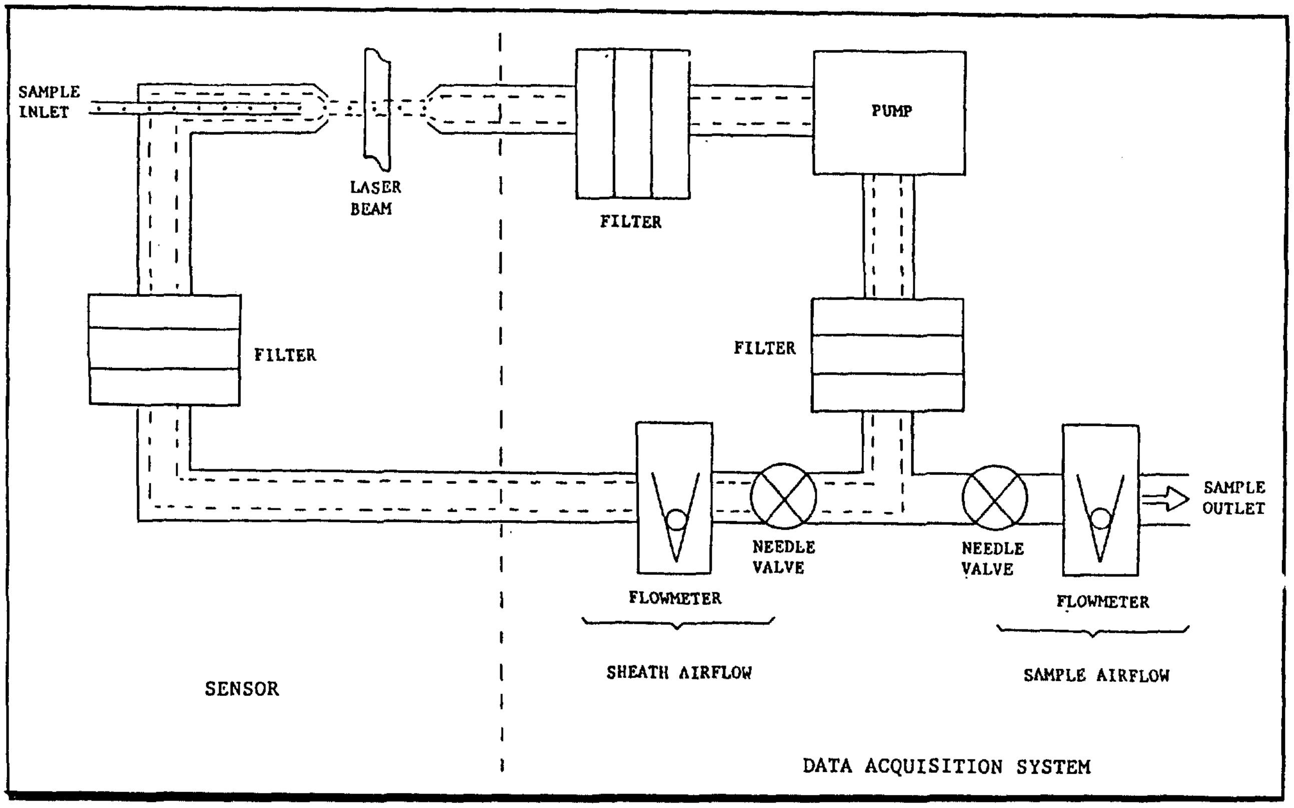


Figure 2 - ASAS-XF airflow diagram (from Operating Manual)

Calibration tests with polystyrene latex

Supplier's initial calibration

The ASAS-XF has two size ranges : RANGE 1 from 0.125 to 0.85 μm; RANGE 0 from 0.85 to 3.10 μm each of them divided into 15 channels. The size partition is given in table I.

Table I. ASAS-XF size partition

CALIBRATION DATA

ASAS-XF (Liconix 4110 HeCd Laser [10 mW])

Size Range #1 (0.125 - 0.850 μm)

Channel	Size (microns)	Interval (microns)
1	0.125 - 0.150	0.025
2	0.150 - 0.200	0.05
3	0.200 - 0.250	
4	0.250 - 0.300	
5	0.300 - 0.350	
6	0.350 - 0.400	
7	0.400 - 0.450	
8	0.450 - 0.500	
9	0.500 - 0.550	
10	0.550 - 0.600	
11	0.600 - 0.650	
12	0.650 - 0.700	
13	0.700 - 0.750	
14	0.750 - 0.800	
15	0.800 - 0.850	0.05

CALIBRATION DATA

ASAS-XF (Liconix 4110 HeCd Laser [10 mW])

Size Range #0 (0.85 - 3.10 μm)

Channel	Size (microns)	Interval (microns)
1	0.85 - 1.00	0.15
2	1.00 - 1.15	
3	1.15 - 1.30	
4	1.30 - 1.45	
5	1.45 - 1.60	
6	1.60 - 1.75	
7	1.75 - 1.90	
8	1.90 - 2.05	
9	2.05 - 2.20	
10	2.20 - 2.35	
11	2.35 - 2.50	
12	2.50 - 2.65	
13	2.65 - 2.80	
14	2.80 - 2.95	
15	2.95 - 3.10	0.15

The initial calibration delivered by the supplier from PLS aerosols is given in figure 3.

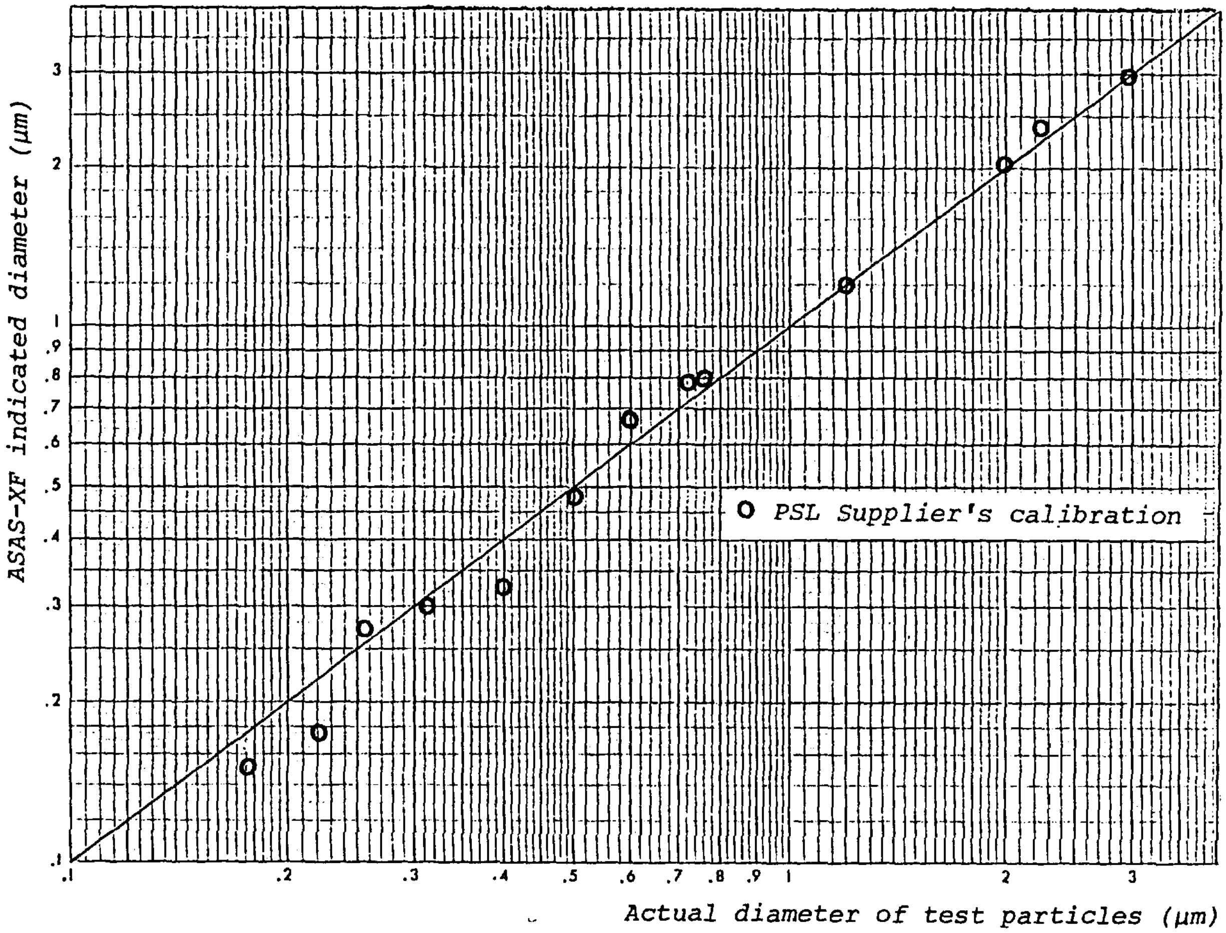


Figure 3

Laboratory calibration

A laboratory calibration was carried out to establish the spectral response of the instrument for monodispersed aerosols. For that purpose, the PSL aerosol is reselected by means of a Differential Mobility Particle Sizer (DMPS) in order to eliminate, especially for the lower dimensions, the interfering aerosol produced by spraying and the multiplets due to coagulation of PSL particles.

The results of this calibration expressed in the form of histograms are shown in figure 4 for 0.12 µm, 0.481 µm, 0.721 µm and 3.1 µm.

The PSL aerosol sizes used for these calibration tests are the following : 0.12 µm, 0.312 µm, 0.481 µm, 0.721 µm, 0.94 µm, 1.65 µm and 3.1 µm. For the first four sizes the PSL aerosol has been reselected by means of the DMPS. The response curve of this apparatus is given in figure 5.

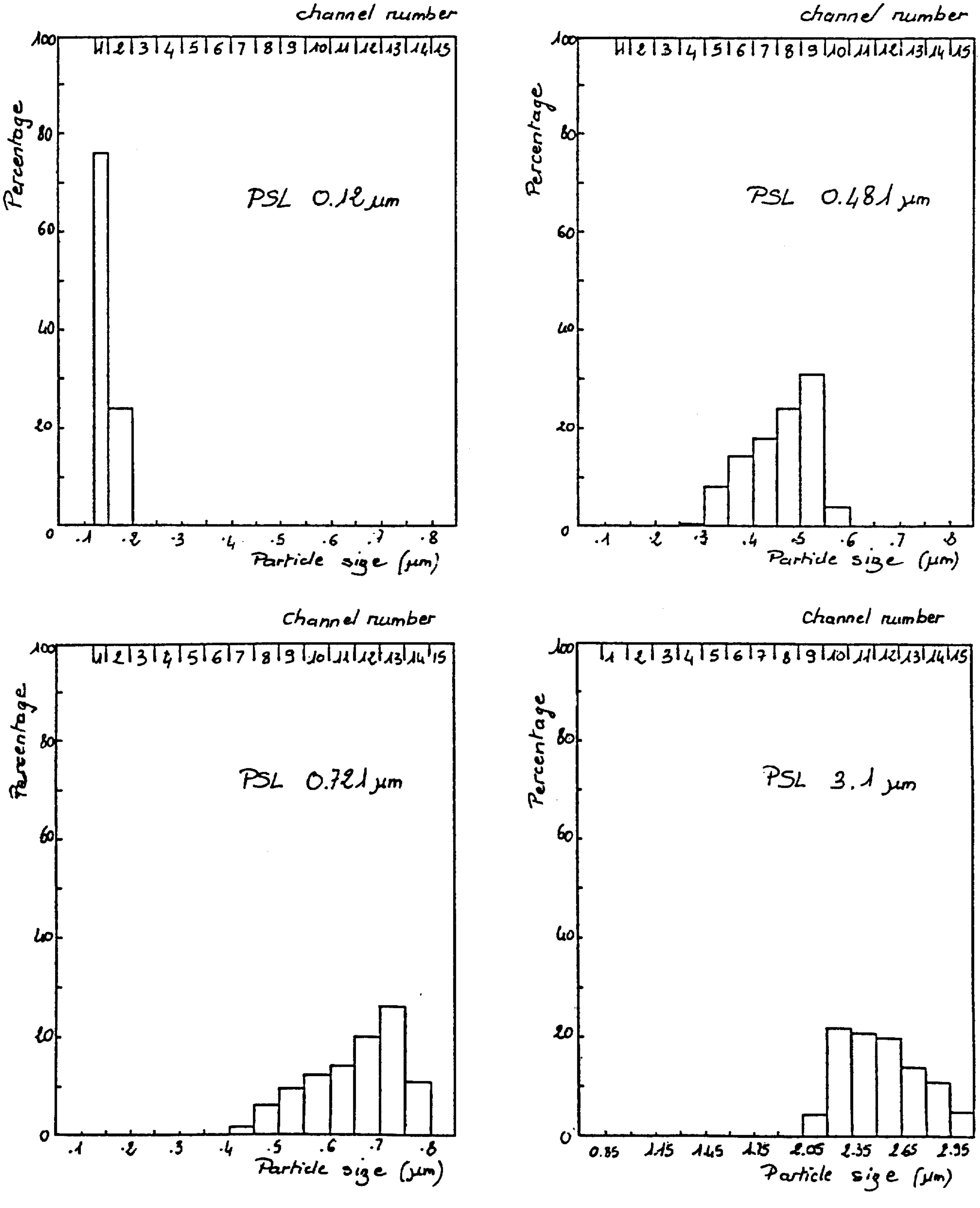

channel number
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
100
80
60
40
20
0
Percentage
PSL 0.12 µm
.1 .2 .3 .4 .5 .6 .7 .8
Particle size (µm)
channel number
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
100
80
60
40
20
0
Percentage
PSL 0.481 µm
.1 .2 .3 .4 .5 .6 .7 .8
Particle size (µm)
Channel number
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
100
80
60
40
20
0
Percentage
PSL 0.721 µm
.1 .2 .3 .4 .5 .6 .7 .8
Particle size (µm)
Channel number
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
100
80
60
40
20
0
Percentage
PSL 3.1 µm
0.85 1.15 1.45 1.75 2.05 2.35 2.65 2.95
Particle size (µm)

Figure 4

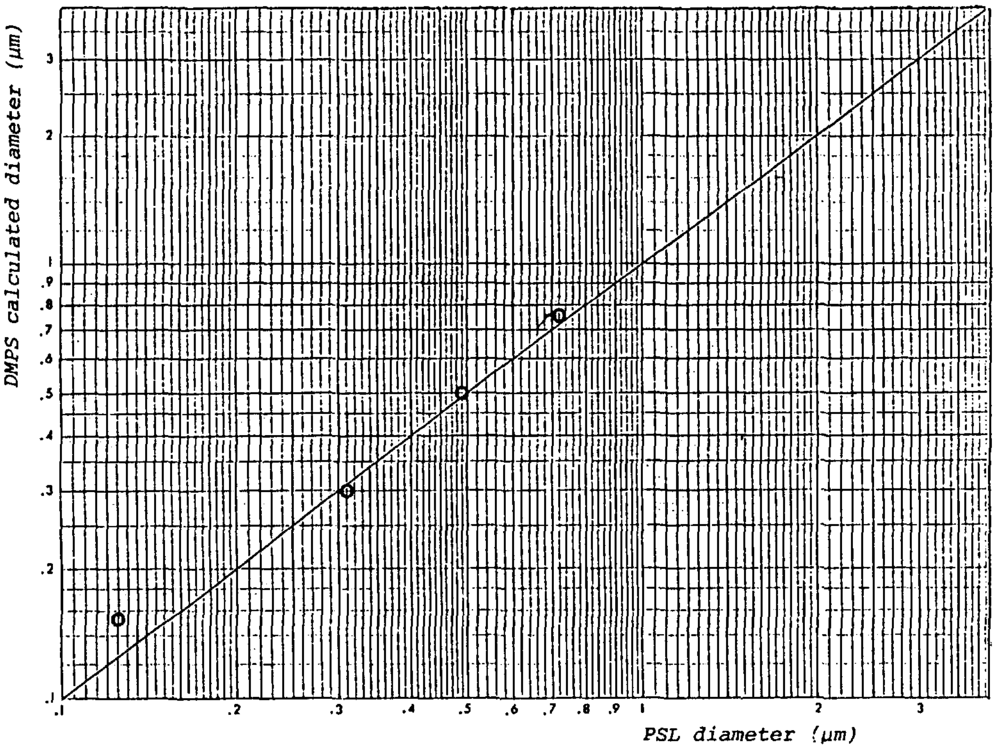


Figure 5

From these data, cumulative distributions were derived from which the mean diameter and standard deviation are calculated. The results are plotted in figure 6.

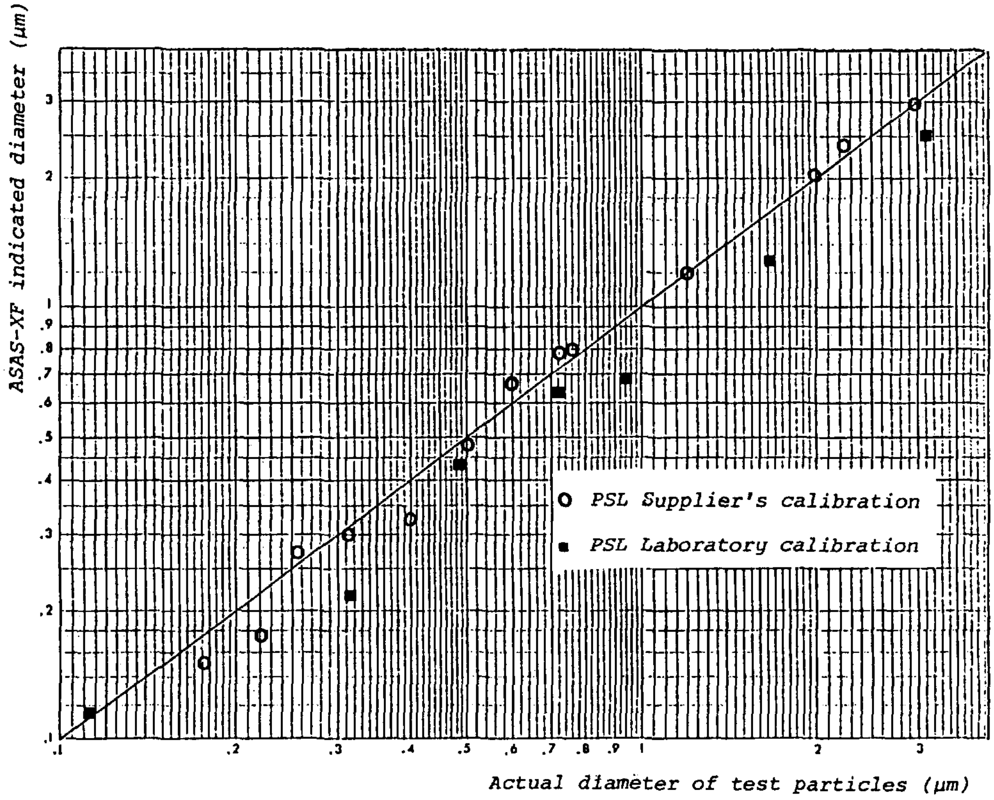


Figure 6

Calibration tests with DOP

Fluorescent detection check

A test was conducted with a polydispersed DOP aerosol tagged with Potomak Yellow. From this test it results that the fluorescent detection has a 100% efficiency except for the very first channel. This can be explained by the low amplitude of the fluorescent emission due to the small amount of Potomak Yellow (maximum solubility in DOP : 2 g/l) contained in DOP aerosol particles of about 0.1 µm.

DOP calibration tests

As for the PSL calibration tests, a polydispersed DOP aerosol was generated and then reselected by means of the DMPS. The size distributions given by the ASAS-XF for different particle sizes are illustrated in figure 7.

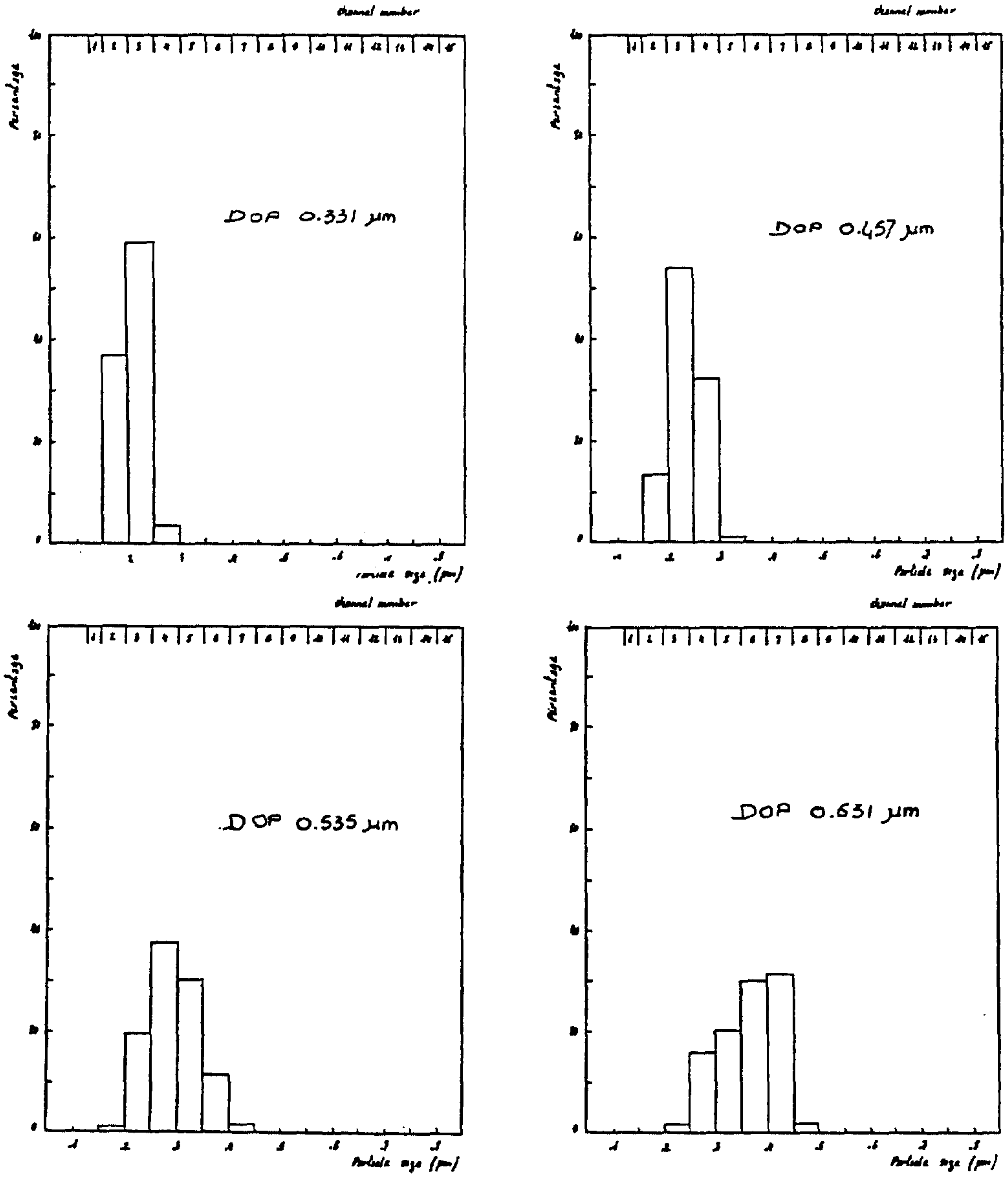


Figure 7

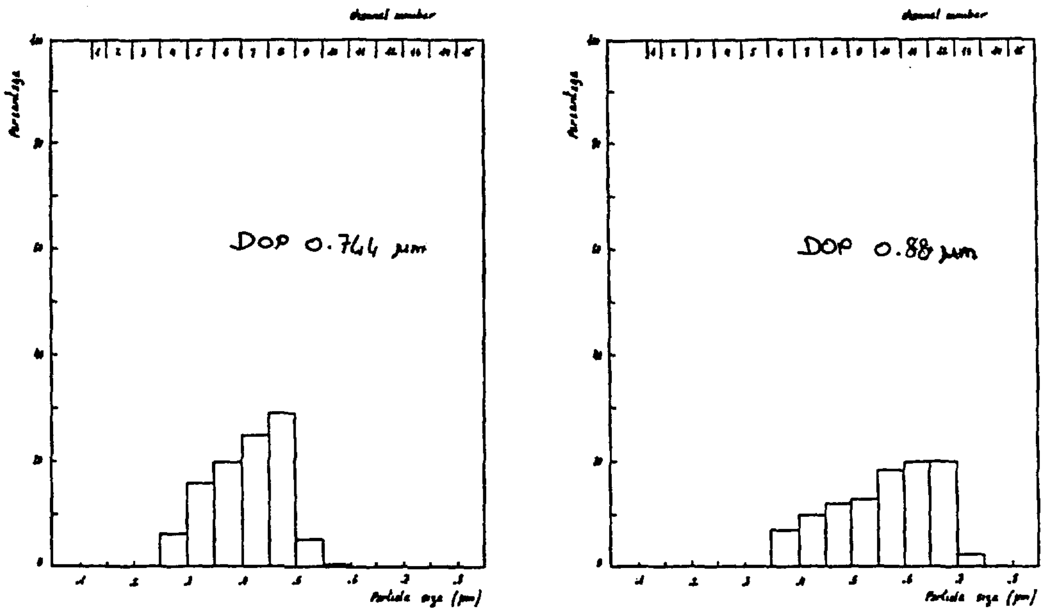


Figure 7 (followed)

Mean diameters and standard deviations were also calculated from cumulative' distributions. These data are plotted in figure 8.

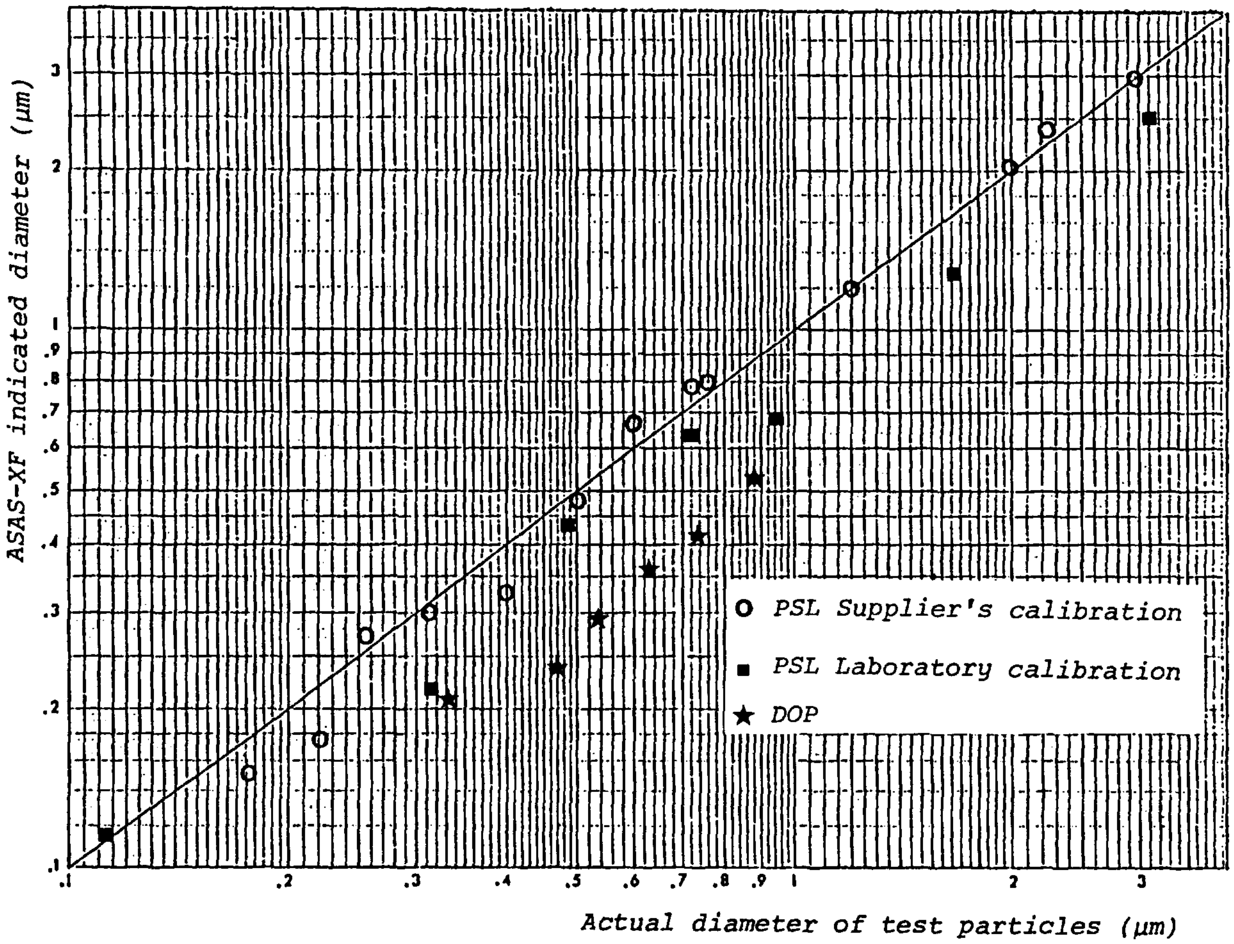


Figure 8

IV. Discussion

The calibration of the ASAS-XF laser aerosol spectrometer with PSL and DOP monodispersed aerosols (all reselected by the DMPS if ranging below 0.84 µm) are summarized in table II.

Table II

Actual particle diameter (µm)	Mean particle diameter calculated from instrument indication (µm)	Standard deviation
0.12 (PSL)	0.13	1.09*
0.312 (PSL)	0.22	1.14*
0.481 (PSL)	0.43	1.19*
0.721 (PSL)	0.63	1.15*
0.94 (PSL)	0.68	1.26
1.65 (PSL)	1.27	1.23
3.10 (PSL)	2.50	1.11
0.331 (DOP)	0.21	1.135*
0.457 (DOP)	0.23	1.15*
0.535 (DOP)	0.29	1.18*
0.631 (DOP)	0.36	1.18*
0.744 (DOP)	0.41	1.20*
0.88 (DOP)	0.52	1.18*

* Aerosol reselected by means of the DMPS

The results for the PSL calibration are in a good agreement with the initial calibration carried out by the supplier.

The standard deviation σg ($\sigma g = \frac{\text{diameter at } 50\%}{\text{diameter at } 16\%}$) either for PSL or DOP monodispersed aerosols is less than 1.2. This corresponds to a good spectral resolution.

On the other hand, these tests confirm previous works (3) with regard to the role played by the refractive index of the particle on the diameter measured by the instrument. As a matter of fact, the diameter indicated for DOP particles in the measurement RANGE 1 is about 50% less than the actual diameter. This is of particular importance when size distribution and spectral efficiency measurements are carried out.

References

(1) J.C. Elder, T.G. Kyle, M.I. Tillery and H.J. Ettinger. In place testing of tandem HEPA filter stages using fluorescent aerosols. 16th DOE Nuclear Air Cleaning Conference (1980)

(2) J.P. Deworm. Laser particle spectrometry for testing HEPA filtration systems in the nuclear industry. Filtration and Separation. (July/August 1984)

(3) W.C. Hinds and G. Kraske. Performance of PMS model LAS-X optical particle counter. J. Aerosol Sci. 17 n° 1 (1986)

DISCUSSION

HOLUB: Who makes the laser fluorescent particle spectrometer?

MULCEY: The manufacturer is Particle Measuring Systems, Boulder, Colorado.

ETTINGER: On your last viewgraph, where you showed the difference between polystyrene latex and DOP, did you correct DOP for index of refraction?

MULCEY: No, we did not. The data plotted on this viewgraph were directly obtained from the apparatus.

ETTINGER: Would a correction for index of refraction have brought them into closer agreement?

MULCEY: Certainly.

SCRIPSICK: A specific analysis of the difference between the responses for DOP and PSL particles of the same diameter has not been performed. This would take into account that the wavelength of our laser (He - Cd : 442 nm) is different from other's (He - Ne : 633 mn).

A NEW LIGHT SCATTERING SPECTROSCOPY TECHNIQUE FOR RAPID ASSESSMENT OF FILTER MEDIA

Yong W. Kim
Professor of Physics
Lehigh University, Bethlehem, Pa. 18015

ABSTRACT

Rapid execution of particulate filter evaluation for performance as a function of particle size is of great importance both from an applications stand point and for studies of particle removal mechanisms because any filter medium ages in the course of such testing activities. New instrumentation for essentially instantaneous determination of the particle size distribution function both upstream and downstream of a filter medium has been developed in this connection. The technique is based on real-time measurement of the light scattering loss spectrum over a wide wavelength range covering the ultraviolet to near infrared extremes. The spectrum is deconvoluted for determination of the full size distribution function in under a minute. Mineral oil drops produced by a modified Laskin generator and other atomizers are used, with concentrations varying over five orders of magnitude and particle size reaching 0.1 μm or less on the small size end. Critical analysis of the overall methodology is presented.

I. Introduction

Small particles in suspension are encountered in a variety of situations, ranging from natural phenomena, such as volcanic events, forest fires and photochemical smog to manufacturing and basic investigations of fluid phenomena. The particle size distribution function of the suspension invariably are required in any quantitative analysis.

Of particular importance to the general program of aerosol filter evaluation is a rapid method for measuring the particle size distribution function. This is because the performance of any given filter changes as a function of its total exposure to the particle-laden gas flow. The method must also be applicable to a wide range of particle concentration because the filter efficiency as a function of particle size requires measurement of the size distribution function both on the upstream and downstream side of the filter.

In this paper, a new technique for size distribution measurement will be described, which is based on the light scattering properties of small dielectric spheres. The technique is new in that the light scattering from a suspension is observed and analyzed as a spectroscopic phenomenon covering a continuous spectral range from 2000Å to 9000Å. In contrast, the conventional light scattering methods deal with the angular distribution of scattered intensities at one or a few selected wavelengths. As a spectroscopic process, the total scattered intensity spectrum exhibits a broad resonance structure as a result of a convolution of the light scattering cross-section and the particle size distribution function. The particle size distribution function is then obtained through deconvolution of the measured spectrum aided by the fact that the light scattering cross-section is known.

In the present implementation of the above concept of light scattering spectroscopy, measurement is made of the light scattering loss spectrum. A collimated beam of continuum light suffers a loss of intensity due to scattering as it goes through a suspension of small particles. It can be shown that the light scattering loss spectrum, when reduced to the form of the absorption coefficient, exhibits the same resonance structure as referred to earlier. The advantages of this approach are: i) the range of particle number density over which the technique can be applied can be as large as five orders of magnitude and ii) the technique is truly non-invasive, permitting in-situ applications.

In the following sections, we will describe first the basic framework of the technique in the light scattering mode. A description of the experimental implementation will then be given. The results of some measurements will be also presented, together with a critical analysis of the overall methodology.

II. The Light Scattering Loss Spectroscopy

In the present implementation of the light scattering loss spectroscopy technique, we envision a particle suspension in a gas flow leading to, and departing from, a filter medium. A narrow, highly collimated, beam of light is directed into the suspension. The intensity of the beam is attenuated by scattering of the light from the particles and by absorption of the light by the bulk of the particles and the host gas molecules. The scattering loss alone can be determined if the incident intensity is defined to be that transmitted through the particle free gas of the same path length and gas density. The bulk absorption by the particles can also be excluded by considering the measured absorption coefficient of mineral oil, the liquid used for dispersal of the aerosol in the present experiment, but this turns out to be negligible here.

It can then be readily shown that the transmitted intensity $I(x,\lambda)$ as affected by the scattering loss alone, is given by

$$I(x,\lambda) = I_o(\lambda)\exp[-K_f(\lambda)x] \qquad (1)$$

where

$$K_f(\lambda) = \pi n \int_0^\infty R^2 Q_s(R,\lambda) f(R)\, dR . \qquad (2)$$

x denotes the length of the suspension along the beam path, λ the wavelength of light, R the particle radius and n the particle number density in the suspension. $I_o(\lambda)$ is the incident intensity of light at wavelength λ, as defined by the intensity transmitted through the gas without particles suspended in it. $Q_s(R, \lambda)$ is the total scattering efficiency, which is the total light scattering cross-section for a particle of radius R at wavelength λ expressed in units of the geometrical cross-section of the particle, πR^2. Derivation of eq. (1) is described in detail elsewhere.(1)

Of particular interest here is the attenuation coefficient $K_f(\lambda)$. It is given as a convolution of $Q_s(R,\lambda)$ and the particle size distribution function $f(R)$. Here $f(R)$ is normalized according to

$$\int_0^\infty f(R)\, dR = 1 . \qquad (3)$$

Implicit in the expression for $K_f(\lambda)$ is that full knowledge of the index of refraction, m, for the bulk material of the particle is required because $Q_s(R, \lambda)$ depends on it. The resonance structure mentioned earlier in the introduction pertains to the functional form of $K_f(\lambda)$. It comes about as a result of going in, and out of alignment, of the maxima of $Q_s(R, \lambda)$ with the maximum of $f(R)$ when the wavelength is varied. This is because the undulating structure of $Q_s(R, \lambda)$ are expanded or contracted due to changes in n, whereas $f(R)$ remains unchanged.

The particle size distribution function is extracted from the measured $K_f(\lambda)$ by a deconvolution process because the functional form of $Q_s(R, \lambda)$ is known. The uniqueness in determination of $f(R)$ is a critical issue in such a deconvolution process; it simply hinges on the breath of the wavelength range in measuring the resonance structure of $K_f(\lambda)$. $K_f(\lambda)$, of course, is obtained from measurement of the intensity ratio $I(x, \lambda)/I_o(\lambda)$. In view of the wide dynamic range required of the technique as a means for aerosol filter evaluation, the ratio must be measurable over a wide range of the particle number density n. The main strength of the light scattering loss mode lies in the fact that the quality of the measured ratio $I(x, \lambda)/I_o(\lambda)$ can indeed be maintained as the particle number density undergoes steep changes over several orders of magnitude because the ratio depends on the product of x and n and the product of xn can be kept in a narrow range of value by appropriately adjusting x in response to the changes in n. Consequently, $K_f(\lambda)$ can be measured accurately even though the magnitude of $K_f(\lambda)$ may decrease sharply with decreasing n.

The procedure for determination of the particle size distribution is to search for a distribution function of definite amplitude, width and maximal radius parameters in such a way that the calculated

$K_f(\lambda)$ best fits the measured $K_f(\lambda)$. The search is carried out on a dedicated computer by means of an efficient numerical algorithm which we have devised for this application.

Several two parameter size distribution functions are employed:

Zeroth order log-normal distribution

$$f(R) = (2\pi z^2 R_m)^{-\frac{1}{2}} \exp\left[-z^2/2 + (\ln R_m - \ln R)^2/2z^2\right] \qquad (4)$$

Schultz distribution (unnormalized)

$$f(R) = (R/R_m)^z \exp\left[-(z+1)R/R_m\right] \qquad (5)$$

Maxwell distribution (unnormalized)

$$f(R) = (R/z)^{M-3} \exp(-R^2/z^2), \text{where } R_m = z[(M-3)/2]^{\frac{1}{2}} \qquad (6)$$

Here R_m represents the mean or maximal radius and z the width parameter. It is noted that the actual values of R_m and z have different meanings for different distribution functions.

III. The Experimental Arrangement

Fig. 1 shows schematically the overall arrangement of the experiment. It consists of a source of mineral oil aerosols, two light scattering stations, two collimated continuum light sources, a filter section, a flat field spectrograph with photodetectors, two digital signal processors and a 16-bit microcomputer. The particle source has two generators (a modified Laskin generator and an atomizer), a settling chamber and a flow manifold for arbitrary dilution of the particle suspension with particle-free gas.

The two light scattering stations are basically vacuum-tight chambers of cylindrical shape, equipped with two fused silica windows on the ends. The one on the upstream side of the filter section is short in length, variable from 0.1 to 10 cm, in keeping with a severe loss of the light intensity by scattering at very high particle concentration. The second station is much longer, 134 cm in length, and is located between two wide area mirrors whose orientation can be controlled independently. By changing the relative orientation of the mirrors, the incoming collimated light beam can be steered to make multiple traversals through the chamber. The net effect is to essentially arbitrarily increase the path length x, as appearing in eq. (1), in response to a sharp decrease in the particle number density in the gas stream after passing through the filter section.

As can be seen in the figure, the light sources employ all-mirror

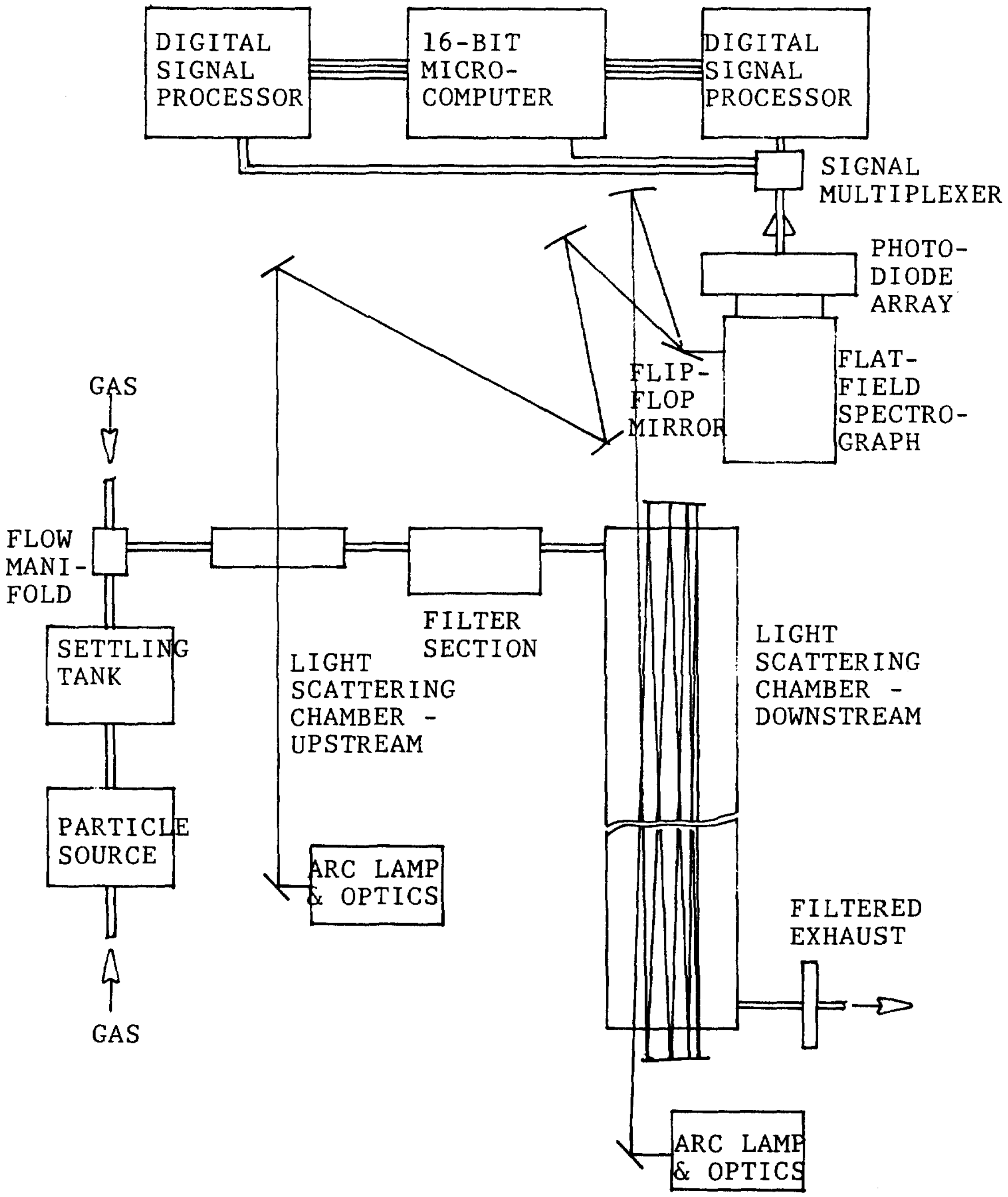


Fig.1. Schematic diagram of the experimental arrangement, consisting of a particle source, two light scattering chambers, a filter section, two continuum light sources, a spectrograph and data processor electronics.

optics in order to produce a highly collimated beam whose beam quality is independent of wavelength over the entire spectral range from 2000 to 9000Å. The xenon arc lamp provides a strong continuum of light with only minor line spectra superposed on it.

Both of the collimated beams terminate at the entrance slit of one spectrograph on a time-shared basis by means of a flip-flop mirror, which can be set at one of the two detented orientations under computer control. The full intensity spectrum is measured with a 1024-element photodiode array detector, which is mounted on the image plane of the spectrograph. The spectrograph provides a flat image plane with a linear dispersion of 360Å/mm.

The intensity spectrum detected by the photodiode array detector is read out in the form of sequential analog voltages corresponding to individual photodiode pixel elements and stored in one of two digital signal processors. Each read-out takes less than 100 msec. After the two light beams are so interrogated, the data stored in the signal processors are moved to the computer under the RS-232C protocol. A full measurement consists of taking $I_o(\lambda)$, which is the intensity spectrum seen by the exact same optical system except that the gas glow is free of particles, for each of the two beams and storing the two spectra in the computer as references, followed by measurements of $I(x,\lambda)$. Experience shows that the entire experiment can be conducted for hours without measureable changes in $I_o(\lambda)$ and therefore only occasional updating of $I_o(\lambda)$ is needed.

The two sets of spectra $I(x,\lambda)$ and $I_o(\lambda)$, corresponding to the two light scattering chambers, are then processed to extract the two attenuation coefficient spectra $K_f(\lambda)$. Subsequent numerical searches bring out the particle size distribution functions. The entire process of measurement and analysis takes two minutes at present. It is reasonable to expect that this time can be further reduced at least by a factor of two.

IV. Results and Critical Analysis

We will now focus on the measurement of the particle size distribution in the light scattering chamber on the upstream side of the filter section. The particles are droplets of heavy mineral oil. The index of refraction for the oil is needed in order to carry out computation of the total light scattering efficiency $Q_s(R,\lambda)$. The measurement of the index over the full wavelength range of interest turned out to be somewhat taxing due to a strong absorption at wavelengths below 3000Å. As a result, we have developed a special instrument for measurement of the index of refraction of liquids. The instrument will be described in another publication but the result for the mineral oil is shown in Fig. 2.

Fig. 3 shows a measured $K_f(\lambda)$ spectrum for a suspension with particle number density of 2 x 10^7 particles/cm^3, as produced by the atomizer. The plot is prepared in such a way that the magnitudes of the $K_f(\lambda)$ spectrum are shown for 100 different wavelengths and are normalized to the maximum value, $(K_f)_{max}$, of the $K_f(\lambda)$ spectrum.

The particle number density is determined by two methods: a) counting of particles, as suspended, by means of an ultramicroscope(1) and b) counting of the particles which are collected on an electron microscope grid by sweeping out all entrained particles in a volume of gas onto the walls of a container. In the second method, the grid is mounted flush with the inner wall surface of a cylindrical electrostatic precipitator. After a steady flow of the suspension is established through the precipitator section, the flow is stopped and all particles precipitated electrostatically. The collected particles are first exposed to the vapor of osmium tetroxide before examination by electron microscopy. From the known gas volume, surface area and the total number of particles per unit area of the grid, one can deduce the particle number density. This latter method also provides an independent measurement of the particle size distribution function.

Deconvolution of the measured $K_f(\lambda)$ spectrum of Fig. 3 is carried out by comparing it to a bank of calculated $K_f(\lambda)$ spectra resident in the computer files. The calculated spectra have been generated by varying the maximal radius and width parameters of a given size

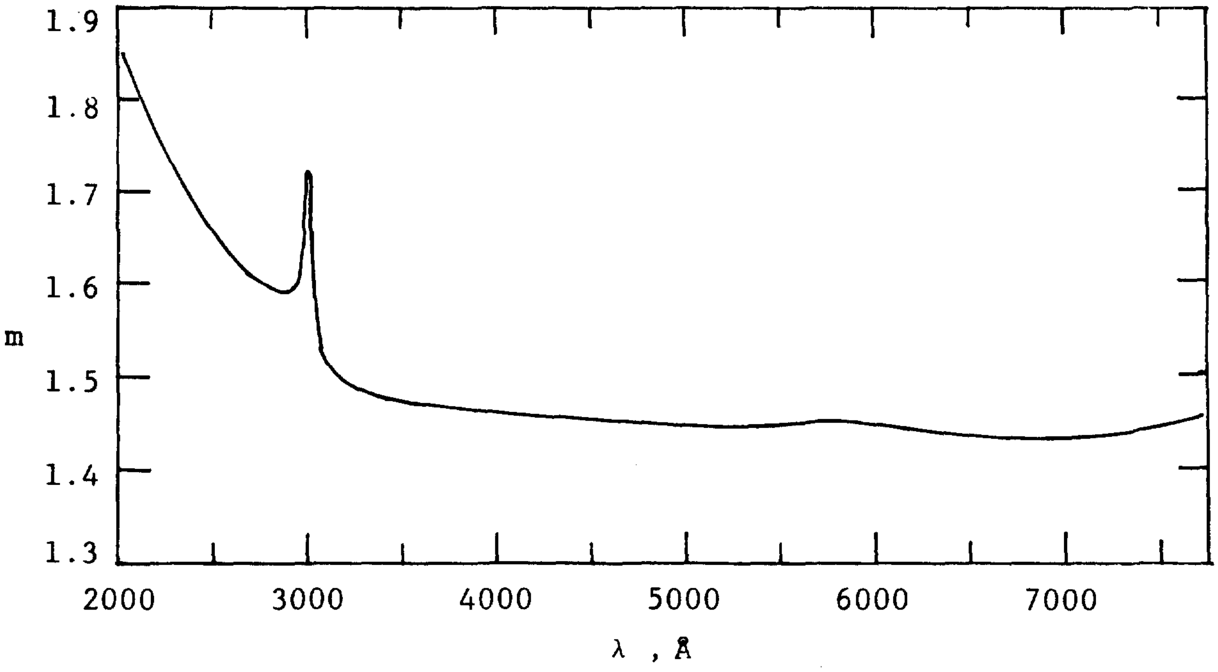


Fig.2. Measured index of refraction of heavy mineral oil as a function of wavelength.

distribution function in the neighborhood of the actual size distribution of the particles used in the experiment. Altogether, five different functions have been considered to represent the particle size distribution and three of them were shown earlier in eqs. (4)-(6). 105 to 132 $K_f(\lambda)$ spectra have been calculated and stored in our computer files for each form of the size distribution function.

The search is conducted by minimizing the absolute difference between the measured and calculated $K_f(\lambda)$ spectra. The search is broken up into two or three levels of coarseness in the increments of parameters R_m and Z. In this way, not only the time for search is significantly reduced, as compared to a sequential search algorithm, but also the precision of the fit can be arbitrarily improved on in

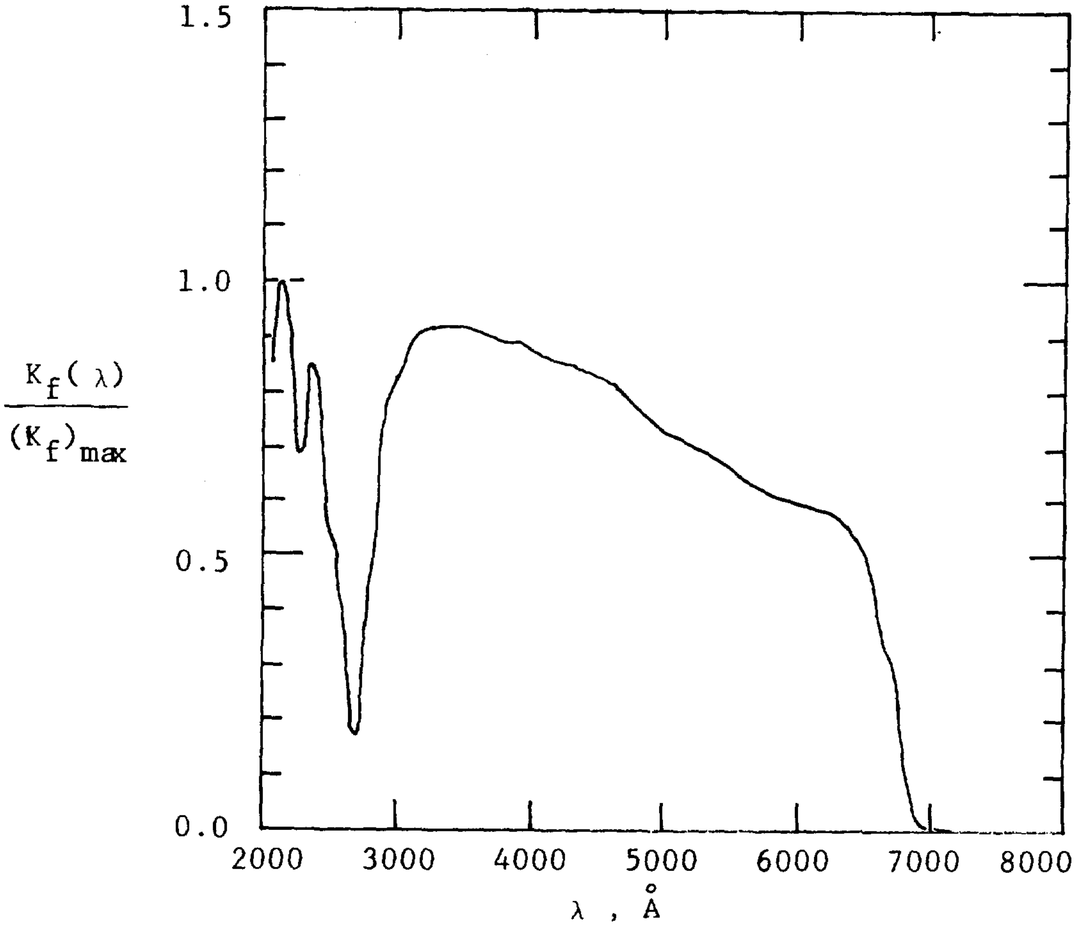


Fig.3. Normalized attenuation spectrum as measured for a suspension of atomized mineral oil drops.

succeeding levels of search, given that the $K_f(\lambda)$ spectrum has been sufficiently accurately determined.

Fig. 4 shows the resulting particle size distribution function. Also shown in the figure is the result of particle size measurements by electron microscopy based on the sampling procedure employing the technique of the electrostatic precipitation outlined earlier. Agreement is remarkably good between the two results.

In an attempt to evaluate the overall sensitivity of the final determination of the size distribution function to experimental uncertainties, the measured $K_f(\lambda)$ spectrum has been subjected to a variety of random errors, simulating the shot to shot variations in

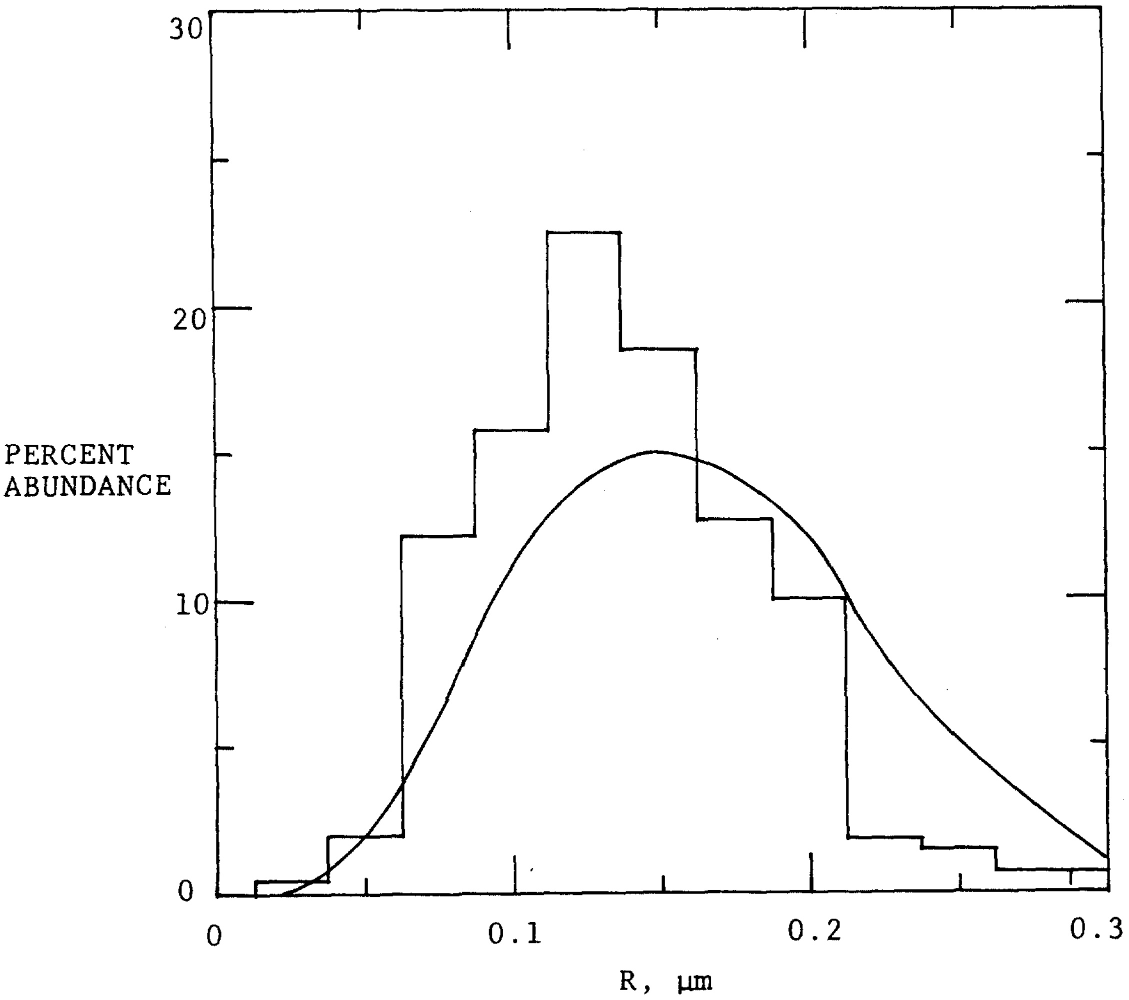


Fig.4. The particle size distribution function as reduced from the measured $K_f(\lambda)$ of Fig.3, using the Maxwell distribution function. The histogram gives the electron microscopy data.

its measurement. The magnitude of the $K_f(\lambda)$ spectrum at each given wavelength is subjected to random errors not exceeding a fixed percentage error δ. Two white-noise random number generators are used for this purpose, one to choose the size of the number to be added to the measured $K_f(\lambda)$ spectrum and the other to select its sign. The resulting $K_f(\lambda)$ spectrum is then processed through the search routine to arrive at a new best-fit particle size distribution function. This is repeated several times for each given δ-value in order to determine the range of spread in the best-fit parameters R_m and Z. The results of this sensitivity analysis are that for the zeroth order log-normal distribution function, the maximum range of variation in Rm is ± 2.64 and ±7.5% when $\delta = \pm 2\%$ and ±5%, respectively. The corresponding spread for Z shows much the same response.

V. Conclusions

Our research program on light scattering as a branch of spectroscopy has been described with particular emphasis on implementation of the light scattering loss spectroscopy technique for measurement of the size distribution function of aerosol particles. The technique has been adapted to filter evaluation applications and all aspects of carrying out measurements with the technique have been examined, including its application under real-time conditions. As the evidence presented in the preceding sections demonstrate, the technique is indeed rapid, versatile and information rich in the sense of broad size range coverage and wide dynamic range in particle concentration. In addition, with proper calibrations, one can determine the particle number density as well as the size distribution function. Furthermore, there is ample room for working with more complex forms of the size distribution function, which may have more than two parameters, in order to attain finer details of the particle size distribution.

The author acknowledges valuable assistance of M. Reuter, R. Marlowe, S. H. Kim, R. Millard, J. D. Kim, E. Gretzinger, M. Lee, J. Gregoris and M. Masters. This work has been supported in part by the Chemical Research and Development Center of the U. S. Army.

References

1. Y. W. Kim, "Development of a New Technique and Instumentation for Rapid Assessment of Filter Media", Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Baltimore, 12-16 August 1984, ed. M. W. First (1985). p. 1193

DISCUSSION

BERGMAN: A general comment on ensemble scattering, which I think may be applicable to this method, is the non-uniqueness of effectively deconvoluting turbidity spectra or extinction spectra. Namely, for the same spectra you can fit many different particle size distributions. Previous investigators had been well aware of the non-uniqueness of a deconvolution technique for many years. Would you care to comment on how your particular technique of ensemble scattering addresses the problem of non-unique solutions of size distributions?

KIM: The non-uniqueness comes from the meager nature of the experimental data. If you make a measurement at two wave lengths, the non-uniqueness, or ambiguity, in your result becomes enormously multiplied. Our approach exactly addresses that because we are deliberately looking over a very wide spectral range, making 1,024 separate wave length measurements simultaneously. That is to say, we are adding 1,024 boundary conditions which a size distribution function can be fitted to. The function has essentially three characters, two parameters and the nature of the function, so you are trying to solve three unknowns out of 1,024 boundary conditions and therefore a stringent fit can be arrived at. We find non-uniqueness not to be an issue. On the other hand, when you go to experimental methods, where measurements are made at 5 or 6 different laser wave lengths, non-uniqueness becomes a big issue and then you throw in experimental uncertainties right on the top. So, the wealth of information is critical.

BERGMAN: Just to elaborate on that point. Have you tried to fit many different size distributions to the same light scattering spectrum? For example, if you maintain a constant average volume for the particle size distribution while you vary the degree of heterodispersion from monodisperse to heterodisperse, I expect you will see the same light scattering spectra. However, you are absolutely right, the previous work has been restricted to very narrow wave lengths between 400 to 600 nanometers. Increasing the spectrum will resolve some of the ambiguities, but aren't some of the 1,024 boundary conditions redundant?

KIM: I think your points are well made, but I must disagree with you because the measurement is a unique value as you change the wave length of light. It is not measuring it in a more clever way because the physical measuring stick is being changed. When you go through the many different size distribution functions one can decide the best fit for each function. On the other hand, when you look at the chi-square values absolutely, it shows that certain functions fit better than others. That is how we arrived at the Maxwell distribution as being the best fit. What I am really saying, as I mentioned before, is that you are dealing with more than two parameters. By choosing different types of functions, you are introducing a third and fourth additional variable. If you look at the experimental data, the $K\lambda$ spectrum, they show rather significant details. All the local maxima are real results that come from the fact that the light scattering function has an undulating structure and the size distribution function is scanned through it as you change the wave length. You run into resonances, i.e., numerous bumps, and the depth of the bump, or how sharp it is will depend on how narrow the size distribution function is. These are the kinds of data that potentially can help accommodate many more independent parameters to deal with the size distribution.

ANDERSON: Your theoretical approach seems to be oriented toward Mie scattering regions. Since filtration interest is restricted to only the portion .01 to .3 μm, how does the Rayleigh and/or Gonz transition affect the interpretation of your Fourier Analyses of the total spectrum? We are looking in the Rayleigh area pretty much where it is a 6th power of the function rather than square. We do not see resonance bumps in the 0.1 - 0.3 μm light scattering patterns because there is a uniform envelope in that area. When you look at your overall theory, I agree with everything you say, but when you look at applications to filter testing, I have reservations.

KIM: Your point is well taken. It is one of the reasons why we are looking at light scattering over a very wide range of wavelengths, with particular emphasis on the very short wavelength end (near 2000 Å). In this way, we can force the Mie parameters into a large value regime and enhance the role of the periodic structure of the total scattering efficiency function. I am curious about your lack of observation of the bumps. How was your experiment done?

ANDERSON: When you are down around 0.1 - 0.2 μm the scattering function is uniform, with the majority of it in the forward direction.

KIM: This is at what wave length?

ANDERSON: This is at the visible wave length which is the same as you are working, 400 to 600 nanometers.

KIM: We are operating with a shorter wave length light source.

ANDERSON: You do not get the pronounced bumps at the other wave length, also.

KIM: I think we are talking about different bumps. I am sure you are referring to the angular dependence.

ANDERSON: That is right.

KIM: The quantity we are dealing with is the total amount of light that is lost in all directions. It involves a spatial integration and that produces the bumps that are indicated here. If you look at the scattering efficiency as a function of the Mie parameter, you see that is behaves like a sort of a damped oscillation. These features are the ones which contain the Rayleigh regime behavior. We are not just limiting our studies to one wave length, or to a narrow range. We are looking at a much longer wave length range where the measuring stick is in fact being changed.

ANDERSON: Would you put up the equation you had on earlier? Maybe I am misinterpreting what I saw in your equation. In the very last one, K_f, is R is the radius of the particle?

KIM: Right.

ANDERSON: You have a square function in there.

KIM: That's right.

ANDERSON: The Rayleigh theory says it is a 6-power function.

KIM: The scattering efficiency function is conventionally expressed in units of the geometrical cross-section of the particles, so πR^2 comes from that. What is left over is still contained in Q the scattering efficiency function that has the additional size dependence. That is the steep part you are referring to, the very short end of it.

ANDERSON: That is where we are working, down at the steep part.

KIM: I gather that accounts for your concern.

HOLUB: Do you think that your method of deconvolution is better than the traditional iterative methods of Twomey and Maher used in diffusion battery measurements?

KIM: It is better in terms of the time of execution. The attenuation coefficient spectrum takes very large amounts of time to compute and so does any deconvolution scheme. It also is less vulnerable to numerical errors, such as round-offs. Inspection of many calculated spectra helps in developing intuition about the most important features of the relationship between the light scattering techniques and the particle size distribution. We make a quick search and then go through another level of search, and a third. This process is the fastest method we could come up with. We had to provide a distribution function in a minute, or less if we could. It looks as though we can probably push for about 10 seconds in a reasonable fashion.

HOLUB: Do you think that this method could actually replace the other deconvolution approaches?

KIM: I don't have any comments on that. All I can say is that this is our best solution given the boundary conditions that we set up for ourselves.

SCRIPSICK: I am wondering in the vein of Vern Bergmann's and Andy Anderson's questions if it would be possible to do some mathematical experiments looking at different size distributions and how your method would deconvolute them. Would it be able to determine a difference in the region we are interested in for filter testing? If it could, it might go a long way to putting to rest some of the concerns in this area.

KIM: I think that is a fair question and we have done our share of it. You are really pointing out, I think, the need to look at some of these results. They are forthcoming but at this point we have looked at only these five very well exercised distribution functions and have gone through a fit with all of them, i.e., essentially through a field of about 100 to 150 files for each distribution function. Each distribution function gives you a best fit, as expected, but there is a better fit with some than with others. I think this is something that will take its legitimate course through discussions in the community. On the other hand, as a filter evaluation program, there are some issues still remaining. The dynamic range of the technique is certainly one. I might mention that on the upstream side you may have a distribution function of the type that I showed you, but after going through a filter what will be left in the stream will be very, very small particles at low concentration. Using wavelength of light as a measuring stick, pressure is on the light source to be even shorter in wavelength. When these light sources get to 2,000 - 2,500 Å, the scattered light becomes very hard to find. Therefore, there is a need to find a new light source, which we are also exploring. These are the issues for which we have made significant progress but quite a bit more work needs to be done, and we have to be educated about filters in general.

SCRIPSICK: I have a few more questions of a practical nature. First of all, the time for the rapid method of detection is indicated to be 50-60 seconds for one size distribution, and then you would have to double it in order to do a filter test. Would that take two minutes?

KIM: At present, the measurements, both upstream and downstream of the filter, can be made well under a second. The best fit search takes just under a minute for both when the searches are carried out in parallel. The time needed for the filter to be in the flow stream is well below one minute. We have, in fact, done the best fit searching for five different types of the size distribution functions. Each yields two best-fit parameters but the fit is better with some functions than others. The results from such analysis will be made available in the near future. Now that personal computers are becoming powerful and inexpensive, I don't see the need to burden one computer with both tasks. With two running simultaneously, I expect that 10-20 seconds would be sufficient for both measurements and analysis.

SCRIPSICK: On the order of one minute for a filter test in the worse case. Is that correct?

KIM: Yes.

SCRIPSICK: Also, your system depends on the shape of the particle being circular. Have you looked into the sensitivity of your calculational techniques on the shape of the particles?

KIM: We have been told about this problem by just about everybody we talk to and I have one simple answer. If the particles are agglomerates, you may see their rotational motion, which is occurring at an extremely high rate. What you are looking at is a time-average contribution. A second point is that electron microscopy shows that agglomerates are not as prevalent with oil drops as among latex aerosols. At high concentration, latex forms very large aggregates. Given the experimental conditions, I am reasonably comfortable with the spherical assumption. It would only add a small adjustment to the analysis scheme.

SCRIPSICK: I was thinking more in terms of a quality assurance test. You use an oil aerosol but in the field you can count on solid particles being irregular.

KIM: I think that is an entirely different problem because the index of refraction has to be dealt with for those particles anyway. I would rather limit my discussion to a situation where an air filter is evaluated by the same type of liquid droplets day in and day out.

HEPA FILTRATION AND MONITORING SYSTEM FOR AN UNDERGROUND NUCLEAR WASTE REPOSITORY

P.S. Parthasarthy and J. Shome
Bechtel National Incorporated
San Francisco, California

Abstract

The design of a HEPA filtration system for the underground areas of a nuclear waste disposal facility involves some unique requirements, normally not encountered in other nuclear facilities. Many of these requirements have been addressed in the design of the Waste Isolation Pilot Plan (WIPP). WIPP is a defense activity of the U.S. Department of Energy and is intended to provide a research and development facility to demonstrate the safe disposal of radioactive wastes resulting from the defense activities and programs of the United States, exempted from regulation by the Nuclear Regulatory Commission.

The primary function of the HEPA filtration system is to minimize the release of radioactive particulate contaminants from the underground facility to the outside environment during and after a postulated nuclear accident. It was established during the design, that the consequences of unfiltered releases from the WIPP facility, during normal operations and accidental occurrences, will be well below the appropriate maximum permissible limits. HEPA filtration for the underground exhaust system is provided solely to mitigate the consequences of any activity release to a level as low as reasonably achievable.

The underground exhaust air stream normally bypasses the filter units. Upon detection of any abnormal radioactivity underground, air is diverted through the HEPA filter units. Filtration mode can be initiated manually or automatically. Manual control is provided locally as well as in a remote central monitoring room.

The main components of the system are:

A) HEPA filter units consisting of two banks of prefilters and two banks of HEPA filters in series;

B) Two 10 feet diameter bypass isolation valves;

C) Underground radiation monitoring system;

D) Effluent monitoring system;

E) Exhaust fans.

Some of the concerns addressed during the design were:

A) Effect of dust generated by underground activities on the HEPA filters;

B) Effects of an underground fire on the HEPA filters;

C) Effect of the time lag between detection of underground radioactivity and initiation of filtration mode, on offsite releases.

An economic analysis was also performed comparing the present system design to a continuous filtration system with no bypass. Results of the analysis are summarized later in this paper.

Functions of the System

The primary functions of the underground ventilation system at WIPP are:

A. To provide adequate quantities of fresh air to all underground personnel and equipment.

B. To maintain an airflow pattern which will preclude movement of potentially contaminated air from areas where waste emplacement operations are performed to areas where construction is in progress.

C. To minimize the spread of smoke and fire within the underground areas during a postulated fire accident.

D. To minimize and control the release of airborne radioactive particulate contaminants from the underground facility to the outside environment during and after a postulated nuclear accident.

The focus of this paper will be on the last item listed above.

System Description

General

Storage level in the WIPP facility is approximately 670 meters (2200 feet) below the surface and is served by three shafts.

A. Construction and salt handling (C&SH) shaft, 3 meters (10 feet) diameter.

B. Waste shaft, 5.8 meters (19 feet) diameter.

C. Exhaust shaft, 4.3 meters (14 feet) diameter.

The C&SH shaft is used as the primarily air intake to the underground, the waste shaft as the auxiliary air intake and the exhaust shaft is used for the ventilation exhaust. Near the bottom of the C&SH shaft the supply air is split into three streams, one serving the waste storage areas, the second serving the construction areas and the third serving the experimental areas (Fig. 1 and Fig. 2). Air coming down the waste shaft is used to

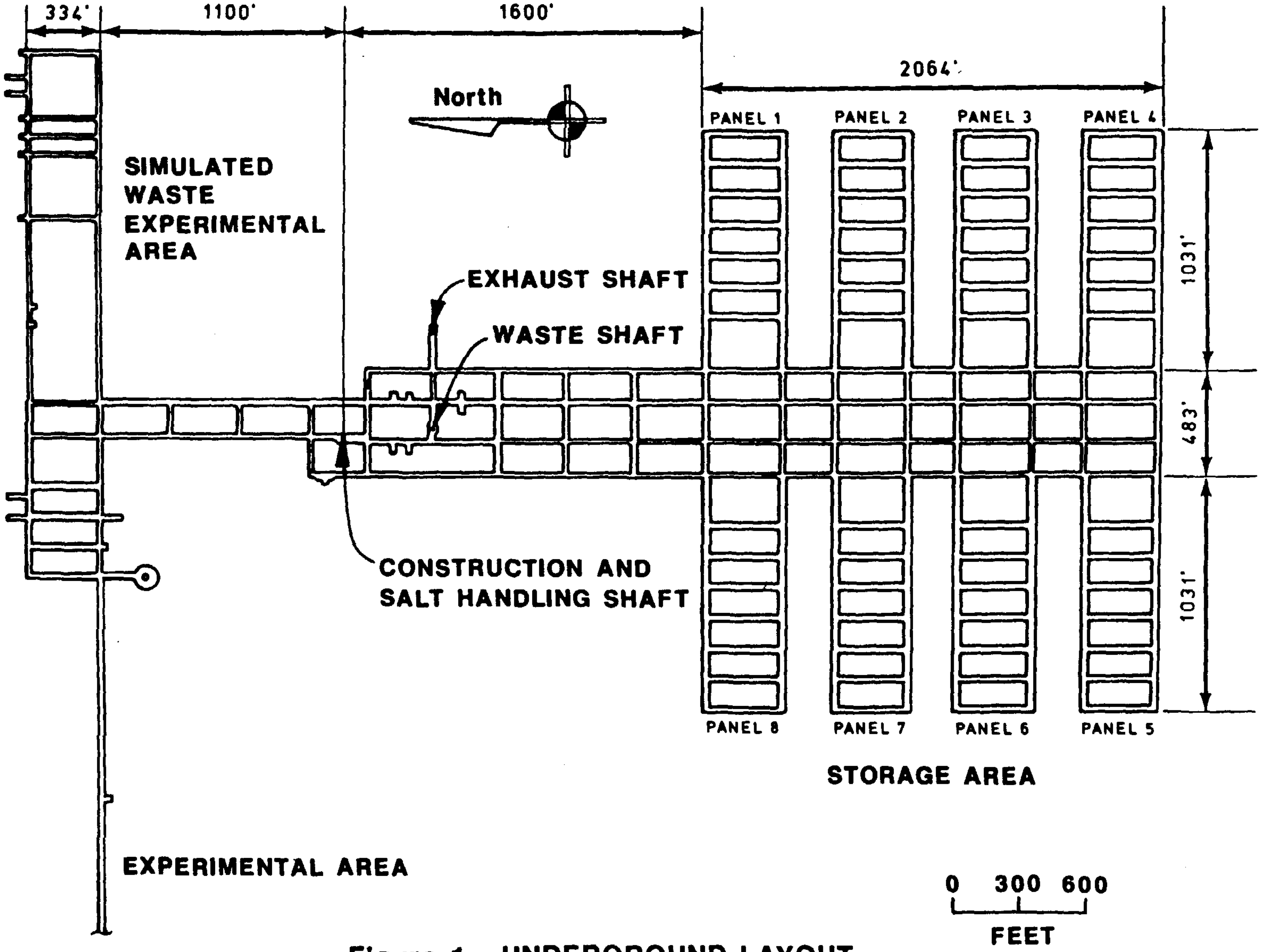
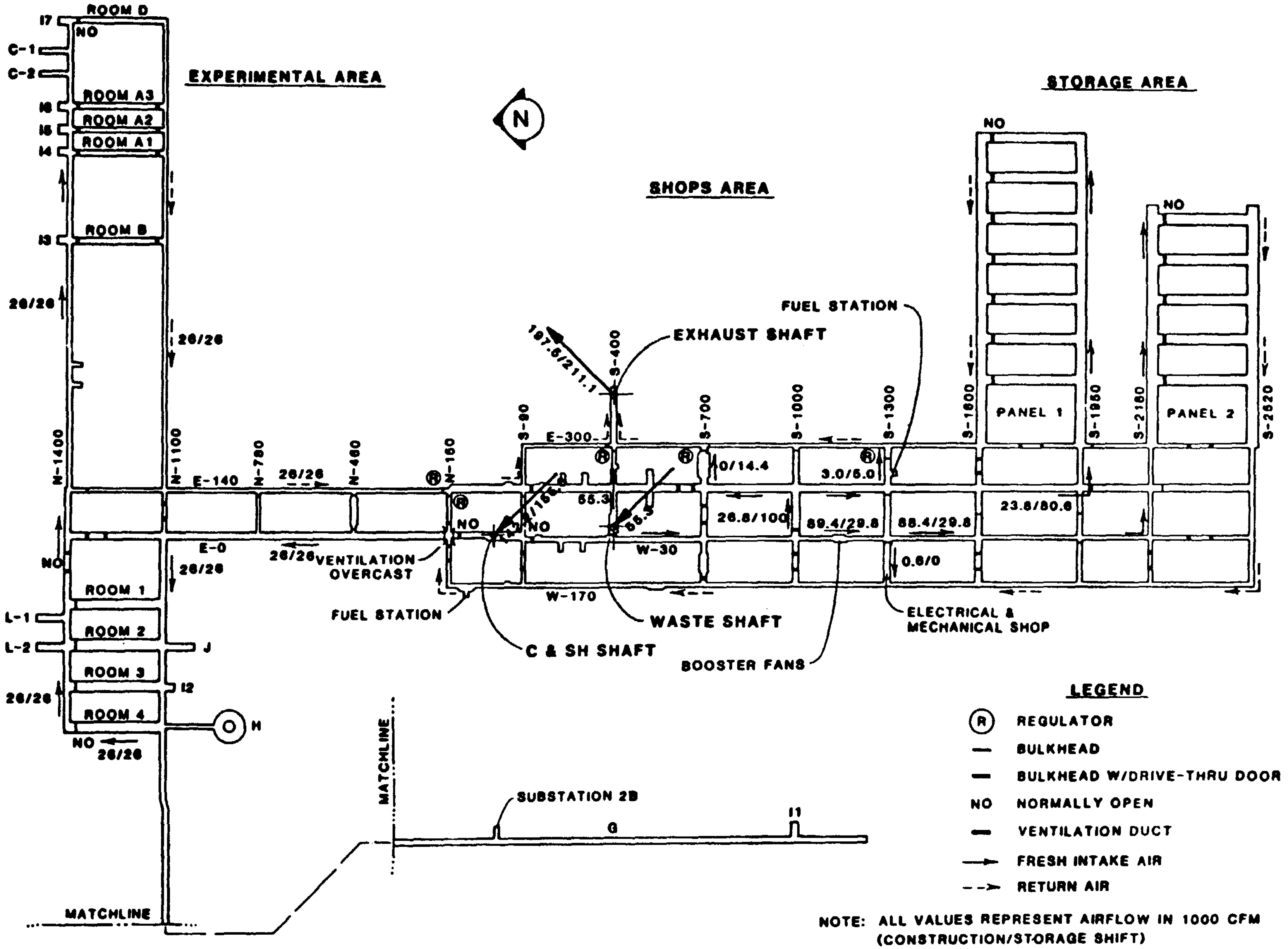


Figure 1 UNDERGROUND LAYOUT



NOTE: ALL VALUES REPRESENT AIRFLOW IN 1000 CFM (CONSTRUCTION/STORAGE SHIFT)

Figure 2 UNDERGROUND VENTILATION SYSTEM

ventilate only the waste shaft station and the immediate work areas and is then exhausted directly through the exhaust shaft. A common exhaust shaft serves all underground areas.

Construction and waste storage operations will be performed in separate shifts and the bulk of the fresh air will be switched from one area to the other at the end of each shift.

Within the underground facility, ventilation separation is maintained between the waste storage and construction areas. A pressure gradient is established which ensures that air leakage is always from construction areas towards waste storage areas. To facilitate separation of the two sides, a unique sequence of construction, waste emplacement, and backfilling operations will be used. With such sequencing, personnel will not be required to work in the exhaust air stream from the waste storage side.

Three main exhaust fans are located on the surface. In addition, booster fans are provided underground, on the construction side only, to ensure a pressure differential between the waste storage and construction sides.

Each exhaust fan is sized for 33 M^3/sec (70,000 cfm) at a static pressure of 3237 pascals (13.00 inches water gage), to give a total ventilation capacity of 99M^3/sec (210,000 cfm).

HEPA Filtration System

It was established by a study(1) that the consequences of unfiltered releases from the WIPP underground areas, during normal operations and accidental occurrences, will be well below the appropriate maximum permissible limits. So a HEPA filter system was designed which permits the underground exhaust air stream to bypass the filters during normal operation. HEPA filtration is provided solely to mitigate the consequences of any activity release to a level as low as reasonably achievable. Upon detection of any abnormal radioactivity underground, exahust air is diverted through the HEPA filters.

The filtration system is housed in the Exhaust Filter Building (Fig. 3). Air is ducted from the exhaust shaft to the building as shown. During normal operation, air flows straight through a 3 meter (10 feet) diameter duct to the outlet plenum and exhausted by the fans to the atmosphere. In the filtration mode, the two butterfly valves in the 3 meter duct are closed and air is diverted through the 1.8 meters (6 feet) duct to the filter inlet plenum. Two filter housings, each of 14.15M^3/sec (30,000cfm) capacity, are provided between the inlet and outlet plenums. During the filtration mode the total ventilation exhaust is reduced from the normal flow of 99M^3/sec (210,000 cfm) to 28.3M^3/sec (60,000 cfm). Only one out of the three main exhaust fans is required to operate.

All underground activities will cease immediately after switching to the filtration mode. The reduced level of ventilation is adequate to prevent the spread of contamination underground and to support recovery operations.

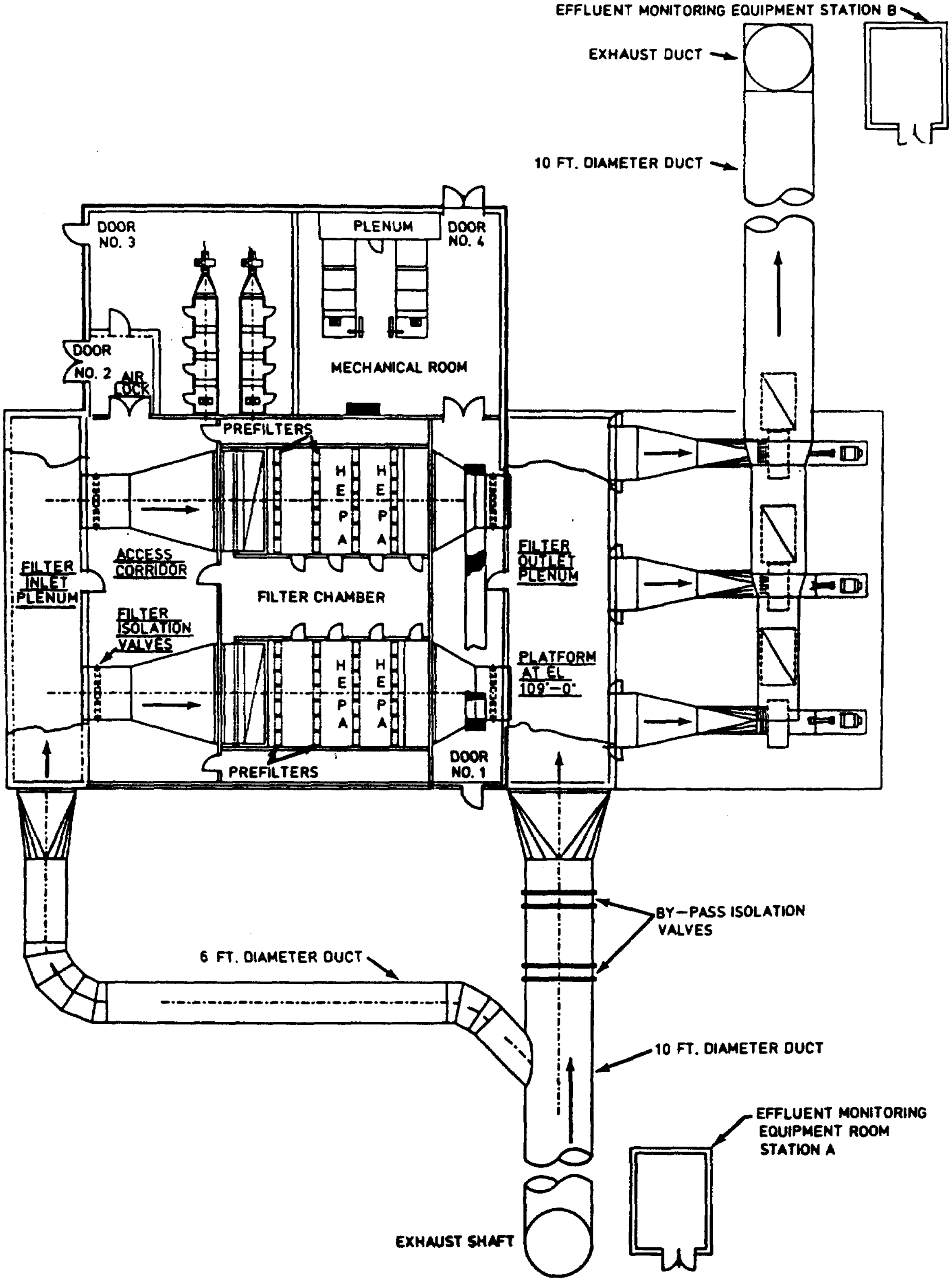


Figure 3 EXHAUST FILTER BUILDING

Two banks of HEPA filters are provided in series to obtain a decontamination factor of 10^{-6}. Because of high levels of salt dust in the air, the HEPA filters are preceded by two banks of prefilters, a moderate efficiency bank and a high efficiency bank.

Monitoring System

The monitoring system consists of the following:

A. Effluent monitoring system consisting of isokinetic probes in the main exhaust duct and monitoring equipment on a nearby skid. The probes are installed at the top of the exhaust shaft and downstream of the HEPA filter units (Fig. 2). The extracted air sample is split into two parallel streams of equal volume. Each stream has a particulate filter. One is viewed by an alpha detector and the other by a beta/gamma detector.

B. Continuous air monitors (CAM) with alpha and beta-gamma detectors underground.

C. Non instrumented fixed air samplers (FAS) underground.

D. Gamma area radiation monitors (ARM) underground.

E. Differential pressure indicators for each filter bank.

F. Flow indicators for each exhaust fan.

G. Status indicators for bypass isolation valves and fan motors.

Controls

The ventilation system will be automatically switched from the bypass mode to the filtration mode if any of the following events occur:

A. Detection of airborne radioactive contaminants above a predetermined level by either the alpha or beta CAM associated with the isokinetic probes in the main exhaust duct on the surface.

B. Receipt of a signal from at least two alpha CAM's, two beta CAM's, or one alpha and one beta CAM in the same undergroud airstream.

C. A sustained 15 second loss of signal between the Central Monitoring Room and the local processing unit controlling the valves and fans.

D. Loss of offsite power.

Items C and D provide a fail safe condition.

Upon receiving an activating signal, the controls automatically divert airflow through the HEPA filter units by closing the bypass isolation valves, opening the filter isolation valves and stopping two of the three exhaust fans. Construction side booster fans underground are also stopped at this time.

Effects of Fire and Dust on HEPA Filters

Fire

Smoke particles generated from a fire can rapidly plug HEPA filters and eventually damage them. Lawrence Livermore Laboratary (2)(3) has conducted a number of studies and experiments on this subject. These results could not be directly applied to WIPP because of the considerable distance between the HEPA filters and the potential source of fire underground at WIPP. Also, sufficient data is not available on the concentration and size distribution of smoke particles that would be in the airstream at the time of reaching the HEPA filters. A considerable amount of the smoke particles are expected to settle down in the underground drifts(4).

The Lawrence Livermore studies also indicate that prefiltration is the most promising of the various countermeasures being considered to prolong HEPA filter life. While WIPP does not have a rolling prefilter system referred in these studies, it has two stages of extended media prefitlers.

The design basis fire underground consists of the following sequence of events(4):

A. A 60 gallon diesel fuel tank on an underground vehicle ruptures.

B. An ignition source sets the diesel fuel and the vehicle on fire.

C. One stack of drums containing contact handled nuclear waste is involved in the fire. It is assumed that the other rows of drums behind the first row are protected by backfill material.

A computer program(5) that was used for ventilation network analysis on the WIPP project was also used to study the temperature distribution at various points of the network based on the design basis fire. The results indicated that the temperature in the vicinity of the HEPA filters would be less than 37.7°C (100°F). Based on this analysis, fire suppression features were deemed unnecessary for the HEPA filters.

Dust

In the WIPP underground facility dust is a major problem, when compared to other nuclear facilities. However, there are two alleviating factors:

A. The HEPA filter units are part of a standby system and hence will not be exposed to dust laden air at all times.

B. The filter units are located on the surface and are a long distance away from the source of dust generation. A major portion of the dust generated will settle down in the underground drifts (tunnels) before reaching the surface.

The key to prolonging the life of the HEPA filters during the filtration mode is to provide an efficient pre-filtration system which will remove most of the dust particles. A study of the available data on dust concentration and particle size distribution in the exhaust air stream was made. Two sources of data available are:

A. A survey made by the Inhalation Toxicology Research Institute (ITRI) at WIPP(6).

B. A survey made at the Potash Company of America (PCA) near Carlsbad,(7), Table 1.

The surveys indicate that a decrease of nearly two orders of magnitude in concentration occurs as the sampling point is moved away from the mining equipment to the bottom of the exhaust shaft. The PCA survey provides a size distribution and concentration for each size range at the bottom of the exhaust shaft (Table 1).

Table 1 PCA Survey: Dust concentration and particle size distribution at the bottom of the exhaust shaft.

Particle Size Micron	Concentration μg/cft
7 to 11	1.68
3.3 to 4.7	2.39
1.1 to 2.1	3.44
0.43 to 0.65	2.23
0.4 to 0.05	1.19

These data were used to study prefilter and HEPA filter life during the filtration mode. This is a very conservative analysis because,

A. During filtration mode, no mining activities are scheduled to take place, minimizing dust generation.

B. During filtration mode the exhaust volume, and hence the velocity in the shaft, are reduced to a third of the normal operating value. This will cause more dust to settle down before reaching the filter units.

C. PCA data was taken underground at the bottom of the shaft. No credit is taken for any plate out in the shaft, ducts, and the plenum upstream of the filters.

Based on vendor information(8) on dust holding capacity and filter efficiency, it was determined that during the filtration mode the prefilters will last approximately 21 days of continuous use before requiring replacement. HEPA filters appear to last for more than 2 years of continuous use (See Tables 2 and 3). They may need replacement for other reasons.

Table 2 Dust removed by various filter banks.

	Mod. Effy. Prefilter		High Effy. Prefilter		First Bank HEPA Filter	
Particle Size Micron	Removal[a] Efficiency	Weight of Dust Removed μg/cft	Removal[a] Efficiency	Weight of Dust Removed μg/cft	Removal[a] Efficiency	Weight of Dust Removed μg/cft
7-11	98%	1.65	100%	0.03	100%	0.00
3.3-4.7	80%	1.91	95%	0.46	100%	0.02
1.1-2.1	60%	2.06	80%	1.10	100%	0.28
0.43-0.65	40%	0.89	70%	0.94	100%	0.40
0.4-0.05	10%	0.12	40%	0.43	99.99%[b]	0.64
TOTAL		6.63		2.96		1.34

[a] The removal efficiencies indicated above are approximate and are based on values indicated in ERDA Manual 76-21(9), Page 51.

[b] Conservative assumption for this analysis.

Table 3 Filter life.

	Moderate Effy. Prefilters	High Effy. Prefilters	First Bank HEPA Filters
Design air flow, each cell	1430 cfm	1430 cfm	1430 cfm
Dust loading from Table 2	6.63 μg/cft	2.96 μg/cft	1.34 μg/cft
Dust loading, gm/hour	0.569	0.254	0.115
Dust holding capacity, gms (at final resistance)	270[a]	130[a]	2700[b]
Filter life, hours	475	512	23,478
Filter life, days of continuous use	20	21	978

1. [a] Information, courtesy of American Air Filter. Values at 2000 cfm flow rate. Values at 1430 cfm would be higher.

2. [b] Information, courtesy of American Air Filter. Result of test using ASHRAE dust without lint.

3. All filters are 24"x24"x11 1/2".

Filter Bypass Feasibility Study

The study(1) evalauted the feasibility of discharging the ventilation exhaust air from the underground storage areas directly to the atmosphere, without continuous filtration.

The two main components of the study were,

A. Radiological protection analysis and,

B. Reliability analysis.

The radiological protection analysis evaluated the maximum potential radioactivity releases to the atmosphere. These releases depend on the time lag between the postulated releases of radioactivity to the underground air stream, the detection of the radioactivity by the CAM's or by air sample analysis, and the initiation of the filtration mode of operation. The analysis considered, among other things, delay time between underground detection and release to the atmosphere, and response time of the CAM's. Results of the study indicate that the maximum radioactivity that could be released is within the maximum permissible concentration limits established by 10 CFR 20 and DOE Order 5480.1A.

The reliability analysis considered levels of redundancy in system design, equipment failure rates, test intervals, and average time for repair. The results indicate that for each year of plant operation, the probability that the system will fail to detect and filter abnormal concentrations of radioactivity is less than 1.24×10^{-3}.

Economic Analysis

During the design of the WIPP facility, a study (10) was performed to evaluate if a continuous filtration system would be more economical than a bypass system. A continuous filtration system will be simpler since it will not require any bypass hardware and control needed for switchover from one mode of operation to the other.

The study considered:

A. Capital costs for each alternative,

B. Operating and maintenance costs, including filter replacement and disposal costs for the life of the facility and,

C. Annual energy consumption.

A lifecycle cost analysis was performed using Department of Energy guidelines including discount rates and energy escalation rates. The results indicate that a filtration system with bypass is clearly more cost effective than a continuous filtration system. The major difference is in the filter replacement costs over the life of the facility. Continuous dust loading on the filters from the underground air stream makes filter replacement a very significant factor. Fan energy consumption increases considerably when the filters are in the air stream continuously.

While significant design changes have occurred since the study was performed in terms of airflow quantities, number of filter housings, etc., the results of the study are still valid in principle.

Acknowledgements

The support and cooperation of Samuel Chu, Nuclear Engineering Group Supervisor, Bechtel National, Inc. in reviewing this paper is sincerely appreciated. His input on nuclear related matters has been invaluable.

References

1. Bechtel National, Inc.; "Storage exhaust filter bypass feasibility study"; Document No. 41K510AA-019, Rev 3.

2. Gaskill, J.R., Alvares, N.J., Beason, D.G., Ford H.W., Jr.; "Preliminary results of HEPA filter smoke plugging tests using the LLL full scale fire test facility"; Procedings of the 14th ERDA Air Cleaning Conference.

3. Alvares, N.J., Beason, D.G., Ford, H.W.; "In-duct countermeasures for reducing fire generated smoke aerosol exposure to HEPA filters"; Proceedings of the 15th DOE Nuclear Air Cleaning Conference.

4. Bechtel National, Inc.; "Design bases fire study, repository level; Document No. 76-F-510-02.01, Rev 3.

5. United States Bureau of Mines; "Mine ventilation simulation system (MNVNTLN); February 1980.

6. Newton, G.J., Cheng, Y., Wong, B.A., Boecker, B.B., "Aerosol measurements in the partially completed underground waste isolation pilot plant; final report", July 1983, Inhalation Toxicology Research Institute.

7. Stinebaugh R.E.; "Results of air samples taken in Potash Company of America"; Sandia Laboratories memorandum Metcalf J.H.; February 10, 1978.

8. Data Received by WIPP Project From American Air Filter, March 1985.

9. Burchsted C.A., Kahn J.E., Fuller A.B.; Nuclear Air Cleaning Handbook; ERDA 76-21.

10. Bechtel National, Inc.; "Storage exhaust filtration system cost study"; Document No. 41-F-510-07, Rev 1.

DISCUSSION

HOLUB: Have you measured radon and radon daughter levels? How did you measure the activity levels?

PARTHASARATHY: As far as my information goes, we do not have a radon problem at this time and radon is not being measured at WIPP. An air monitor continuously samples the air stream and any buildup of radioactive particles on the filters will actuate the detectors.

HOLUB: What kind of instrument is used?

PARTHASARATHY: We have detectors for all three forms, alpha, beta, and gamma.

HOLUB: Do you measure continuously on a filter paper?

PARTHASARATHY: Yes, in addition to the continuous air monitors, we have fixed air samplers that collect particles on a filter paper but do not have detectors. The filter papers are taken to the lab and checked on a periodic basis. In addition, we have continuous air monitors with detectors built-in that can automatically turn on the filtration system. Radon detected underground is higher than atmospheric levels above ground.

HOLUB: Yes, I know that Carlsbad Caverns have a pretty high radon concentration.

MURTHY: Your first slide showed that one purpose of the HEPA filters is to "confine radioactivity, if any." Since WIPP is not yet operating, what are the sources of radioactivity now?

PARTHASARATHY: The design is based on having a clean facility during operations, which means all waste is contained and we don't have releases into the airstream. You would only get releases when there is a mechanical breach of a container.

MURTHY: When will WIPP become operational?

PARTHASARATHY: The end of 1988.

MURTHY: You will receive waste at that time?

PARTHASARATHY: Yes, we will receive the first waste in October, 1988.

MEASUREMENTS OF REMOVAL EFFICIENCIES PERFORMED ON POWDER METAL AND FIBER METAL CARTRIDGES TO BE USED IN URANIUM ENRICHMENT FACILITIES AND GLOVEBOX EXHAUST DUCTS

*H.-G. Dillmann

**W. Bier, G. Linder, K. Schubert

*Laboratorium für Aerosolphysik und Filtertechnik
**Institut für Kernverfahrenstechnik

Kernforschungszentrum Karlsruhe GmbH
Postfach 3640, D-7500 Karlsruhe 1
Federal Republic of Germany

Abstract

Development work is described which has been performed on fiber metal filter cartridges used to remove extremely small particles in separation nozzle plants for uranium enrichment. The separation nozzle method is explained. The removal efficiencies measured in filters previously used as well as in new prototypes are indicated.

I. Introduction and Summary

Under the separation nozzle method developed by the Karlsruhe Nuclear Research Center, uranium-235 is enriched by centrifugal forces in a semi-circular flow of a gas mixture consisting of uranium hexafluoride (UF_6) and a light gas, i.e. helium or hydrogen /1/. Figure 1 shows a separation nozzle system; its semi-circular wall along which the gas flows, at the present state of the art has a radius of only 50 µm. Due to the chemical properties of the UF_6, very fine (sub-micron) particles of UF_6 decomposition products are formed continuously in the gas flow /2/. Therefore, special filters must be used to protect the separation nozzles which, in terms of their performance data, are highly sensitive to dust.

A powder metal filter unit, the filter material of which had originally been fabricated for liquid filtration was built and tested successfully in a technical prototype separation nozzle stage similar to the stages needed in numbers of several hundreds in an enrichment plant.

The developmental work on the separation nozzle method aims at the realization of advanced nozzle systems which are further reduced in size by a factor of two or three (radius of curvature 15-25 µm). This will increase considerably the performance of the separation stages but, on the other hand, will make the nozzle systems more sensitive to dust particles.

Therefore, measurements of removal efficiencies were performed on the existing powder metal filter cartridges using the uranin method /3/ and new upgraded metal filters were developed on the basis of sintered stainless steel fiber mats. First prototype filter cartridges of 2-8 µm fiber thickness were tested with the uranin method and removal efficiencies up to 99 % with low pressure drops of 1-2 % were measured.

With the fiber filters even higher removal efficiencies can be achieved by further optimization. This makes this type of filter interesting for other applications, e.g. in glovebox exhaust ducts.

II. Test of Powder Metal Filter Cartridges in a Separation Nozzle Stage and by the Uranin Method

The separation nozzle stage in which the powder metal filter unit was tested is shown in Figure 2. The feed gas is compressed by a two-stage radial compressor with intermediate and final cooling. The gas enters the separation tubes on which the nozzle systems are mounted from below via the set of filter cartridges. The gas flow through the filter is about 13 000 m^3/h; the operating pressure which is inversely proportional to the dimensions of the nozzle system, is 0.5 bar for nozzles with a radius of 50 μm (Fig. 1). The total nozzle length of one separation stage is about 500 meters.

The powder metal filter unit which, on account of the aggressiveness of UF_6 is made of stainless steel, is shown in Figure 3. It consists of 21 cartridges each having 19 filter tubes of 20 mm diameter and 600 mm length. The filter unit has been optimized for reasons of economy to attain minimum pressure drops of about 1-2 % in the filter and a relatively small filter surface of about 15 m^2. This implies relatively high face velocities of about 30 cm/s. The filter material has been taken from a fabrication program for liquid filtration; its mean grain size is about 70 μm, its porosity about 50 % , and the wall thickness is 1 mm. The specific inner surface of the filter is 1500 cm^2/cm^3.

The results of endurance tests of the stage with the filter and, for reasons of comparison also without the filter, are shown in Figure 4. By operation without the filter it was demonstrated that due to the permanent deposition of very small amounts of powder particles in the nozzle system the mass flow through the nozzle decreases slightly but steadily. When operating the stage with the filter no change in the mass flow was measured within the limits of accuracy over a test period of 2000 hours. Hence sufficiently long service lives of at least several years can be expected for separation nozzle stages.

In order to examine the filter more closely with regard to the removal mechanism and the development of improved filters which will be described in the next chapter, single filter cartridges similar to those shown in Figure 3 were measured with the uranin method which has already been described repeatedly at this conference /3/.

Compared with the normally applied aerosol filter testing methods, this method yields the most conservative results because the spectrum of particles is very narrow and lies around 0.2 µm. The measured results have been presented in Table 1 and Figure 5.

Table 1: Decontamination factors of sintered metal powder cartridges from various suppliers. (See also Fig. 5)
Test aerosol: uranin

	Volume Flow m^3/h	Decontamination Factor	Removal Efficiency %
P1	100	1.44	30
	200	1.37	27
	300	1.29	23
	400	1.22	16
	500	1.1	10
P2	100	1.3	24
	200	1.2	17
	300	1.29	22
	400	1.29	22
	500	1.22	18
P3	200	1.35	26
	400	1.28	22
	600	1.17	15

It became apparent that under comparable conditions only removal efficiencies of up to approximately 30 % had been attained. This did not come as a surprise because, as mentioned before, the filter material had been taken from liquid filtration where removal efficiencies greater than 99 % have been attained only for particles greater than 10 to 15 µm. This means that the filter unit with only comparatively low removal efficiencies for the particle sizes of approximately 0.2 µm used in the uranin method is sufficient in practical application of the separation nozzle process where the dust particles are much smaller.

The particles deposited in the nozzles during the test run of the stage without filter (Fig. 4) result from the continuous decomposition of very small amounts of the UF_6; at first the decomposition products have the size of one or two molecules. Then, by agglomeration with other decomposition molecules in the gas flow, they grow and, without a filter provided, can reach only a certain size before they are deposited in the curved nozzle by means of the centrifugal forces. For the nozzles of a 50 µm radius used in the test run, this maximum particle size can be calculated to be about 0.01 µm; for particles of this size the nozzle would act as a total filter barrier.

The good results of the powder metal filter unit (Fig. 4) which filters out all particles smaller than 0.01 um can only be explained by diffusion effects taking place at relatively high face velocities and in multiple filtration.

During growth the decomposition particles which touch the inner surface of the filter (approx. 1000 m^2) are deposited by diffusion and they stay at the wall due to van der Waals forces. Thus the filter prevents the particles from growing and obviously they do not reach sizes of about 0.01 µm at which they would be deposited in the nozzles downstream of the filter. The probability of contact between growing particles and the inner filter surface is greatly enhanced by the fact that the total gas flow is pumped through the filter every three seconds (multiple filtration). This holds for short circuit operation of one stage and for plant operation with the stages, each equipped with a filter, connected in series and the gas flowing from stage to stage.

Table 2 contains the removal efficiencies calculated for multiple filtration comprising 1, 10, 100, and 1000 runs.

Table 2: Removal efficiencies calculated for multiple filtration. 1, 10, 100, and 1000 filtrations.

η/1	η/10	η/100	η/1000
10	65	99.99	100
1	9.5	63	99.99
0.1	0.99	9.5	63
0.01	0.099	0.99	9.5

III. Development and Testing of Improved Filters on the Basis of Sintered Stainless Steel Fiber Mats

As mentioned earlier, developmental work on the separation nozzle method aims at reducing the nozzle size by a factor of two or three (radius of curvature 15-25 μm). This will increase considerably the performance of the separation stage. On the other hand, the reduction in size will make the nozzle more susceptible to dust particles.

Therefore, new upgraded metal filters were developed on the basis of stainless steel fiber mats. The thickness of the fibers was varied from 8 μm down to 2 μm. Figure 6 shows one of the first prototype cartridges. As cut fiber particles have to be avoided, sintered fiber material was used. This also simplifies the fabrication of filter cartridges. The wall thickness of the sintered fiber mats is 0.3 mm, the mean porosity about 65 %.

A major improvement has been achieved by pleating the sintered filter cartridges and thus increasing the filter surface. Actually, a fiber filter material has a greater specific surface compared to a grain material of the same weight fabricated from powder metal, and it enhances the number of particles removed by diffusion. In the material tested here, the specific surface of 7000 cm^2/cm^3 is about 5 times larger than in the powder metal filter.

The results obtained from measurements performed with the uranin method on these fiber filter cartridges are shown in Table 3 and in Figure 5.

Figure 7 shows the respective pressure losses of the filter cartridges at 1 bar and with air as the testing gas.

The measured results show that fiber filter cartridges under comparable conditions yield much higher (up to 99 %) removal efficiencies than powder metal cartridges. The removal efficiencies could be further improved by implementing certain developments, e.g. an increase in the fiber areal weight or an increase in the area subjected to the flow which would result in lower face velocities.

Considering that a removal efficiency of only 30 % of the powder filter was sufficient to protect the 50 μm radius nozzle from all dust particles below 0.01 μm in size, it is supposed that a considerably higher removal efficiency, i.e. between 90 and 99 %, as measured with the filters of 2 μm fiber thickness, should suffice to protect the smaller advanced nozzles with a radius of 15-25 μm.

Table 3: Decontamination factors of fiber metal filter cartridges, measured with uranin as the test aerosol. (See also Fig. 5)

Fiber Diameter Type	Volume Flow m^3/h	Decontamination Factor	Removal Efficiency %
F1: 8 μm	100	5	80
	200	4.5	78
	300	3.3	70
	400	2.99	66
	500	4.2	76
F2: 4 μm	100	10.6	91
	200	8.4	88
	300	7.0	86
	400	6.4	84
	500	6.1	83
F3: 2 μm	100	26.2	96
	200	21	95
	300	14	93
	400	12	91
	500	11	90
F4: 2 μm	100	630	99.8
	200	41	98
	300	73	99
	400	112	99.1

Consequently, a complete filter unit identical to the prototype filters made of 2 μm fiber mats was ordered and is currently being fabricated by industry. Filter testing in the separation nozzle stage with a process gas of UF_6 and helium will be performed in early 1987 when the advanced separation nozzles will be available.

IV. Prospects

For the fiber filters even higher removal efficiencies than the measured 99 % can be achieved by further optimization of the depth of pleats of the filter mats, by improvement of the geometric configuration, and by variation of the layer thickness of the fibers. This makes this type of filter interesting in other applications.

Use of these corrosion and temperature resistant filter cartridges with high removal efficiencies for particles with diameters in the sub-micron range is optimum, e.g. in fuel element fabrication plants, (trains of boxes) if fire aerosols have to be removed. Another advantage offered by fiber metal filters is that they act as automatic barriers to flames. This means that the propagation of flames is avoided by passive measures.

Moreover, these all-metal filter elements can be cleaned and hence - unlike the previously used fiber glass filters - are suitable for repeated use.

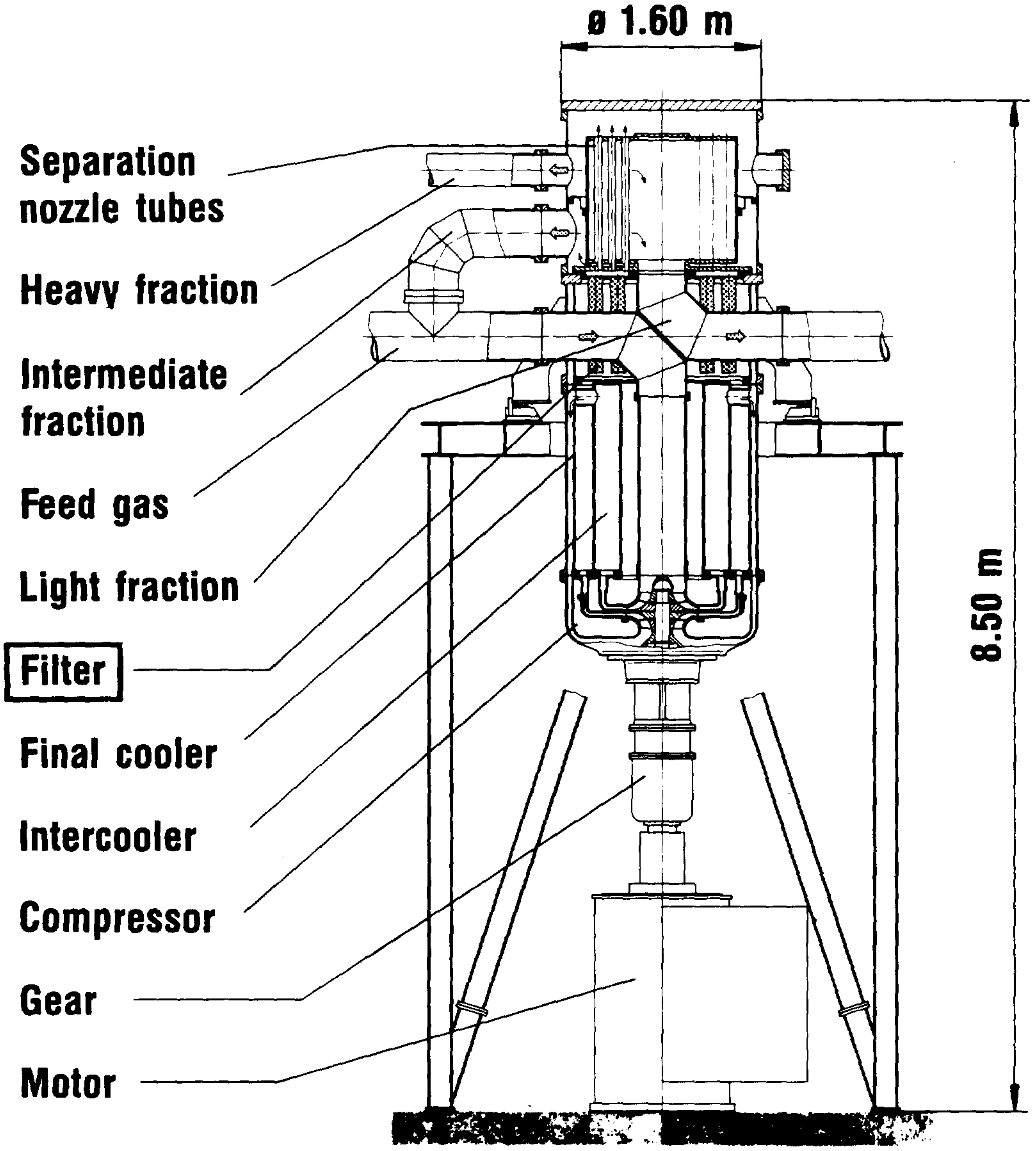
V. References

/1/ BECKER, E.W.; BIER, W.; BLEY, P.; EHRFELD, W.; SCHUBERT, K.; SEIDEL, D.:
"Development and Technical Implementation of the Separation Nozzle Process for Enrichment of Uranium-235".
AIChe 1982 Winter Meeting, Orlando, Florida, USA, March 2, 1982.

/2/ BECKER, E.W.; BIER, W.; HAGMANN, P.; MIKOSCH, F.:
"Bedeutung der radiolytischen Selbstzersetzung von Uranhexafluorid bei der Anreicherung von Uran-235 nach dem Trenndüsenverfahren."
KfK-Bericht 3332, Kernforschungszentrum Karlsruhe GmbH, (April 1982).

/3/ DILLMANN, H.-G.; PASLER, H.;
"Experimental Investigations of Aerosol Filtration with Deep Bed Fiber Filters".
CONF 820 833, p. 1160 (1983).

Fig. 1: Separation nozzle system for the enrichment of uranium-235. A gas mixture of uranium hexafluoride and a light gas enters the nozzle from the left side and is expanded along the curved wall. At the present state of development, the radius of the curved wall is 50 µm.



S1.55.883 IKVT

Fig. 2:

Separation nozzle stage SR33 for the enrichment of Uranium235

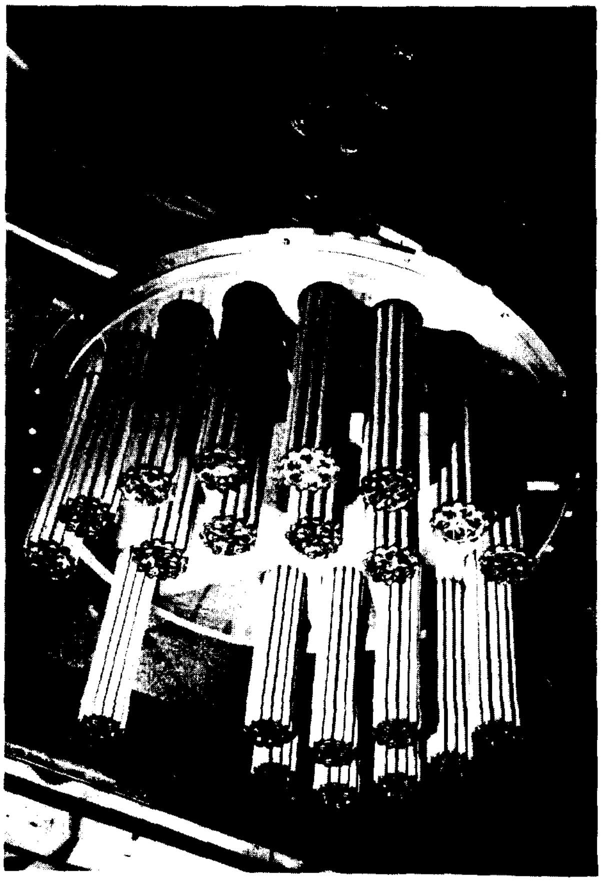
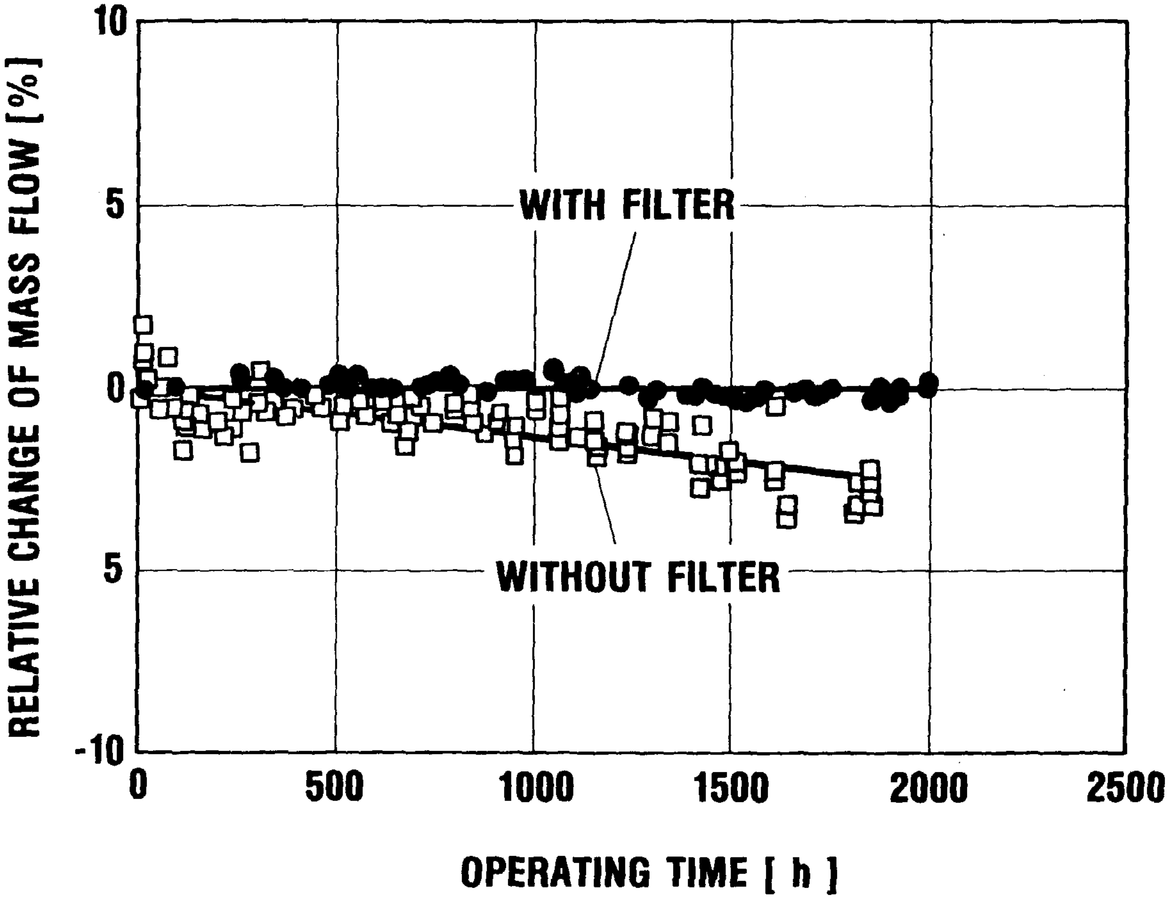


Fig. 3: Filter unit with sintered powder metal cartridges as used in the prototype separation stage (shown in Fig. 2) to protect the separation nozzle systems from the very fine particles (submicron range) formed in small amounts in the process gas.



K4.632.686 IKVT

Fig. 4: **ENDURANCE TEST OF A SEPARATION NOZZLE STAGE**

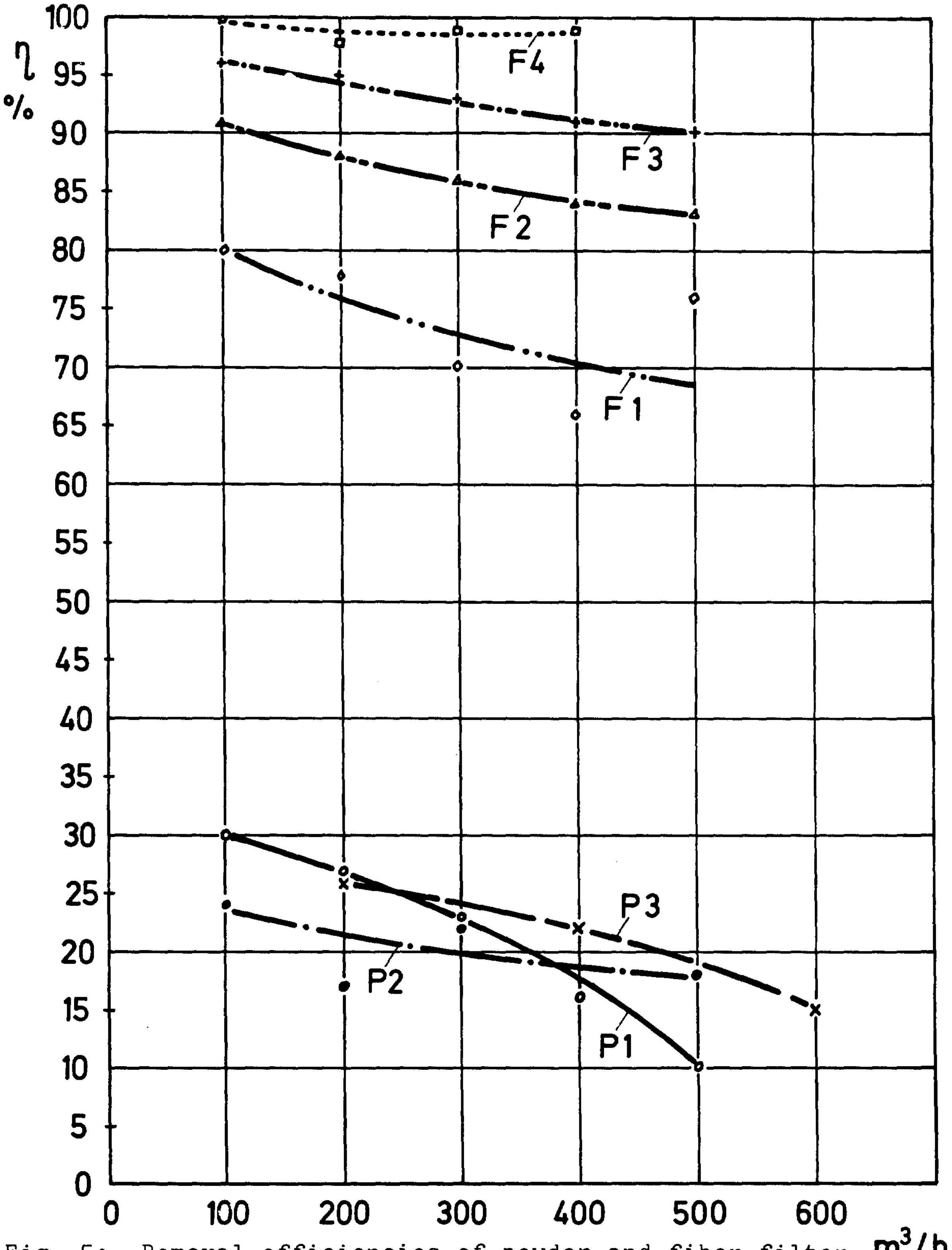


Fig. 5: Removal efficiencies of powder and fiber filter cartridges as a function of air flow, measured with uranin as the testing aerosol.

KfK LAF 86

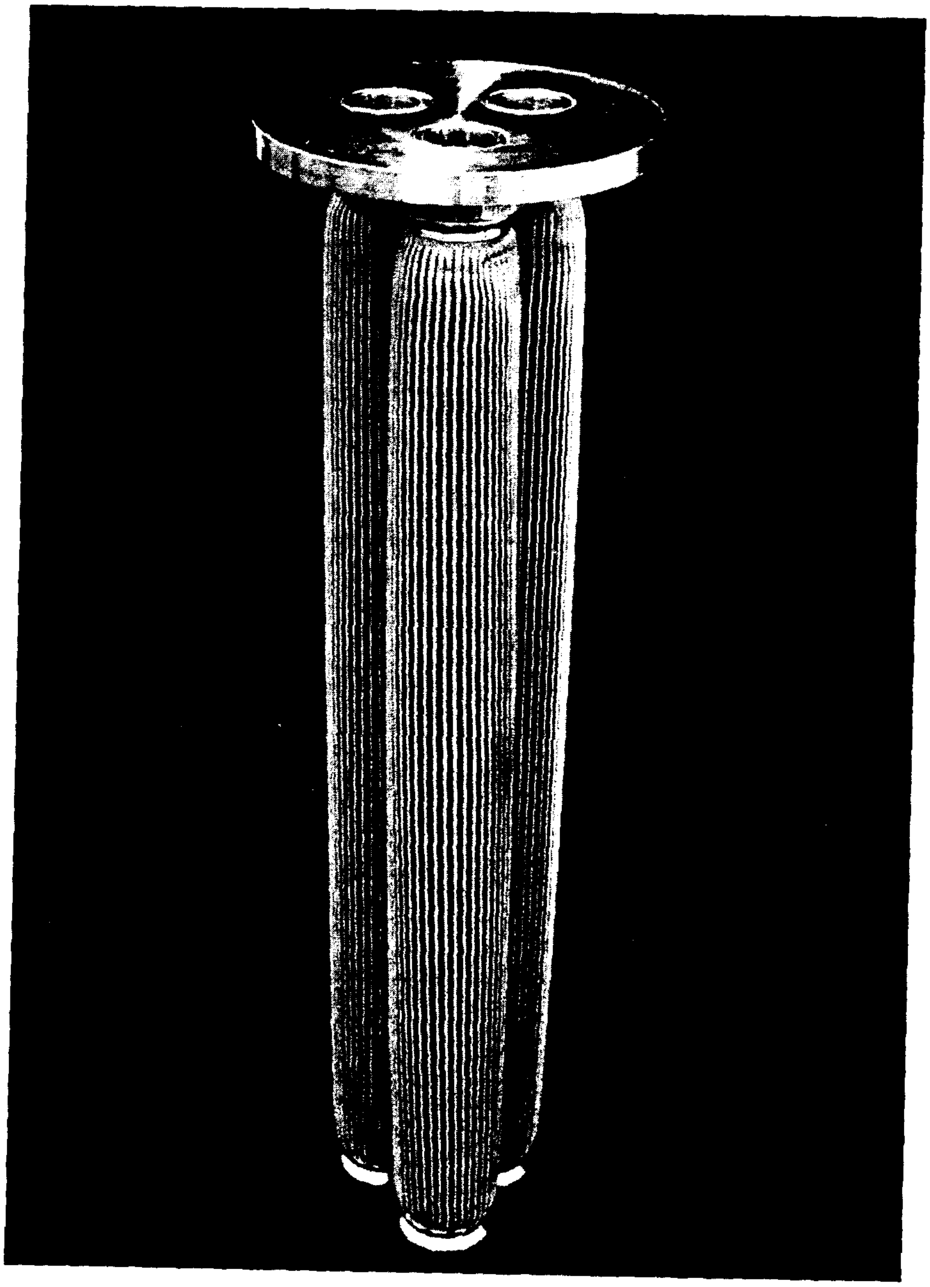


Fig. 6: Prototype of a 2 μm stainless steel fiber filter cartridge.

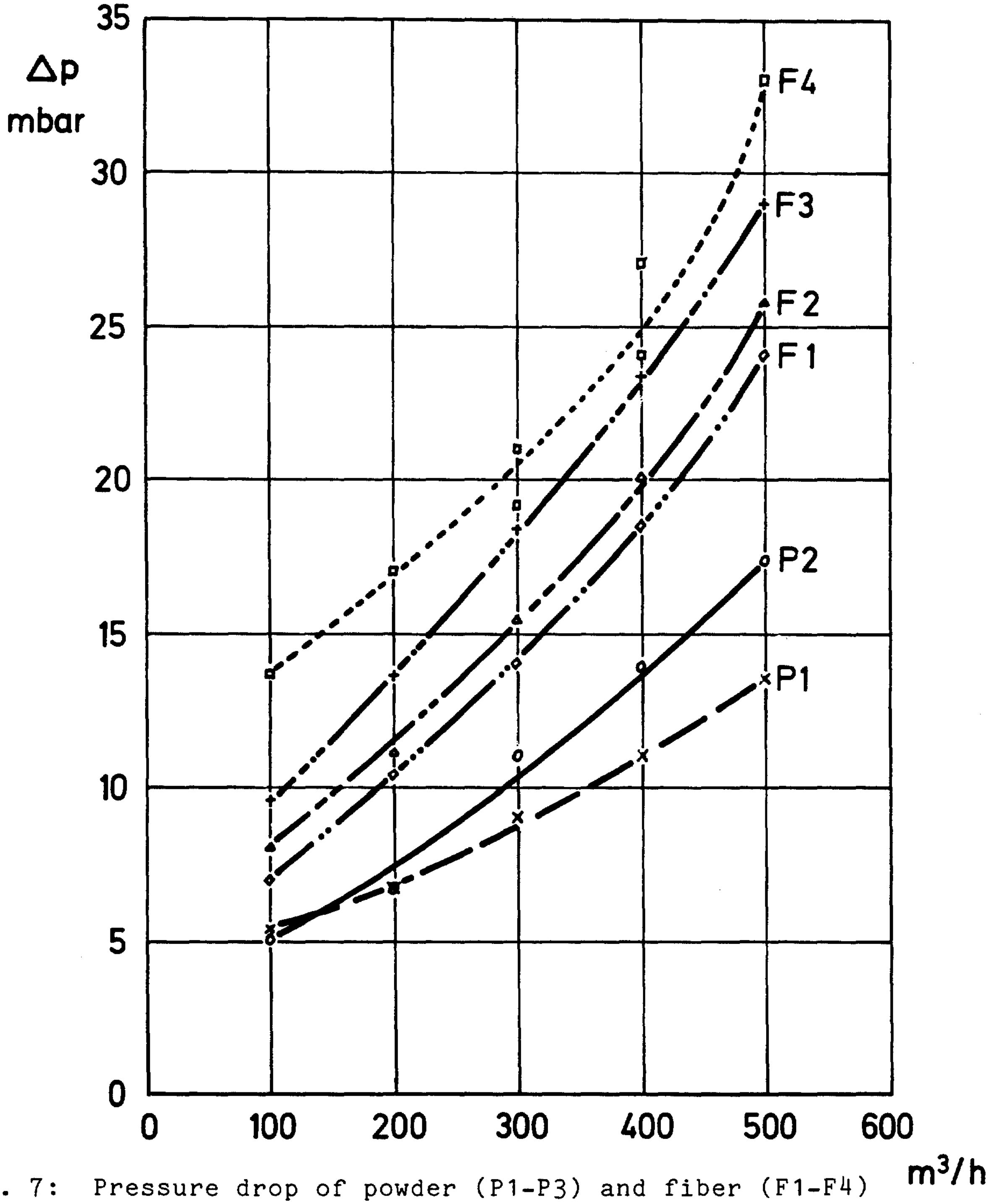


Fig. 7: Pressure drop of powder (P1-P3) and fiber (F1-F4) filter cardridges as a function of air flow.

KfK LAF 86

EXPERIMENT ON A MULTILAYER TYPE AIR FILTER FOR THE FILTERATION OF SODIUM AEROSOL *

N.Otake and O.Nozaki
Nippon Muki Co.,Ltd.
Yuki,Ibaraki-Ken, JAPAN

Abstract

An emergency air filter system of FBR was developed by using a multilayer type filter to protect the function of HEPA filter from clogging due to loading of sodium aerosol. To examine the effect of loading of sodium aerosol on the filter system,sodium aerosol consisting of sodium oxides and the related compound was supplied to the filter system. Several parameters to determine the effectiveness of the multilayer type filter were surveyed. It was confirmed that the emergency air filter system of FBR consisting of the multilayer type filter - a medium filter - HEPA filter with standard size(610mm × 610mm) in series could hold 800 g-Na at 1.5 kPa without clogging.

I. Introduction

The present work was carried out to develop a pre-filter for a high efficiency particulate air (HEPA) filter for an emergency air filter system of a Fast Breeder Reactor (FBR). The emergency filter system is required to remove sodium oxide aerosol released in an event of the posulated accident of FBR.

Sodium aerosol,when generated in the air,may consist of Na_2O,Na_2O_2, $NaHCO_3$,Na_2CO_3,or their mixture according to the condition of atomosphere. The sodium aerosol, if collected by HEPA filter, may cause clogging of the air filter so that the function of air cleaning system will be lost. It was recommended by Kitani et al. that the loading capacity of HEPA filter should be limited to 1 mg-Na/cm^2.(1) Jordan reported 16 mg/cm^2 at 5 kPa for 2.6 m^2 glass fiber filters.(2) McCormack et al. reported 2.7 ~ 16 mg/cm^2 at 5 kPa for HEPA filters.(3) Thus, the problem of severe clogging of air filter by sodium aerosol should be solved.

For example, a sand bed filter was developed by a group of KfK in West Germany.(4) On the other hand, McCormack et al. reported that none of the pre-filter tested were effective in increasing the sodium oxide/hydroxide holding capacity by the combination of pre-filters and HEPA filters.(3)

This work is concerned with the filtration of sodium aerosol by applying a multilayer type filter to the filter system without a special blower with the strong sucking force or to install many filters for a large surface area in ventilation systems. The multilayer type filter consists of a few layers of perforated filters by which sodium aerosol may be holded without clogging and can protect the function of HEPA filter.

The emergency air filter system for a FBR, considering one unit of system, is possible to design as shown in Fig. 1, in which the system consists of a multilayer type filter, a medium filter, a HEPA filter and

---

* This work was performed under contracts between the Power Reactor and Nuclear Fuel Development Corporation and Nippon Muki Co.,Ltd.

a charcoal filter, respectively. Thus, the HEPA filter is protected by the multilayer type filter and medium filter.

The effectiveness of the multilayer type filter was investigated with respect to kind of filter media,opening diameter,opening area ratio and intervals among layers. Another important subject in the test was to evaluate the effect of humidity.

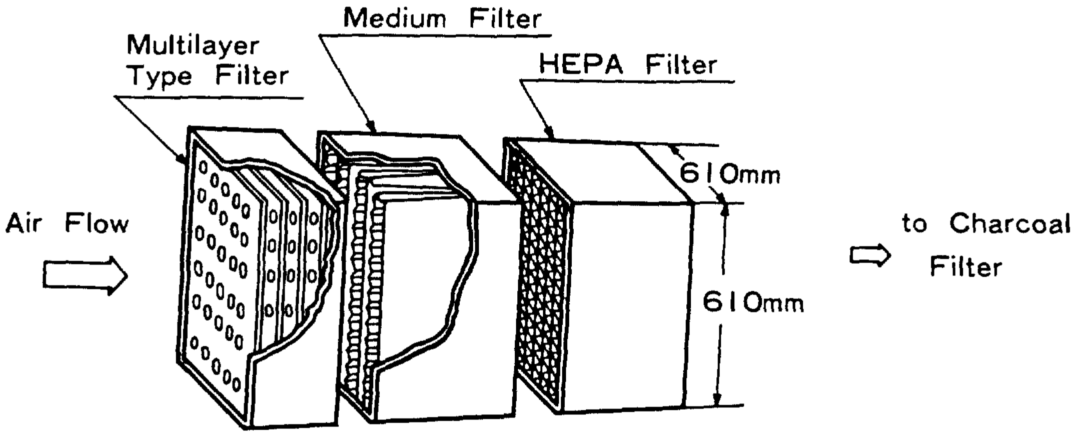


Fig. 1 A filter assembly developed for an emergency filter system of FBR.

II. Experimental

Test apparatus

The test apparatus is shown in Fig. 2. Filter units with standard size (610 mm×610 mm) were installed singly or in series. The apparatus consists of filter system,a buring pan for sodium aerosol generation, a blower,ducts and measurement systems. The blower can suck 77 m^3/min at 5 kPa of static pressure. The concentration of aerosol was monitered by Sibata digital dust counter which could measure the scattered light intensity of white light at 50° from the direction of light beam propagation.

Particle size analysis

The particle size distribution of sodium aerosol was analyzed by sampling the aerosol on meshes of an electron microscope by a thermal depositor and the particles on the meshes were analyzed by Curl Zeiss-TRZ 3 particle size analyzer.

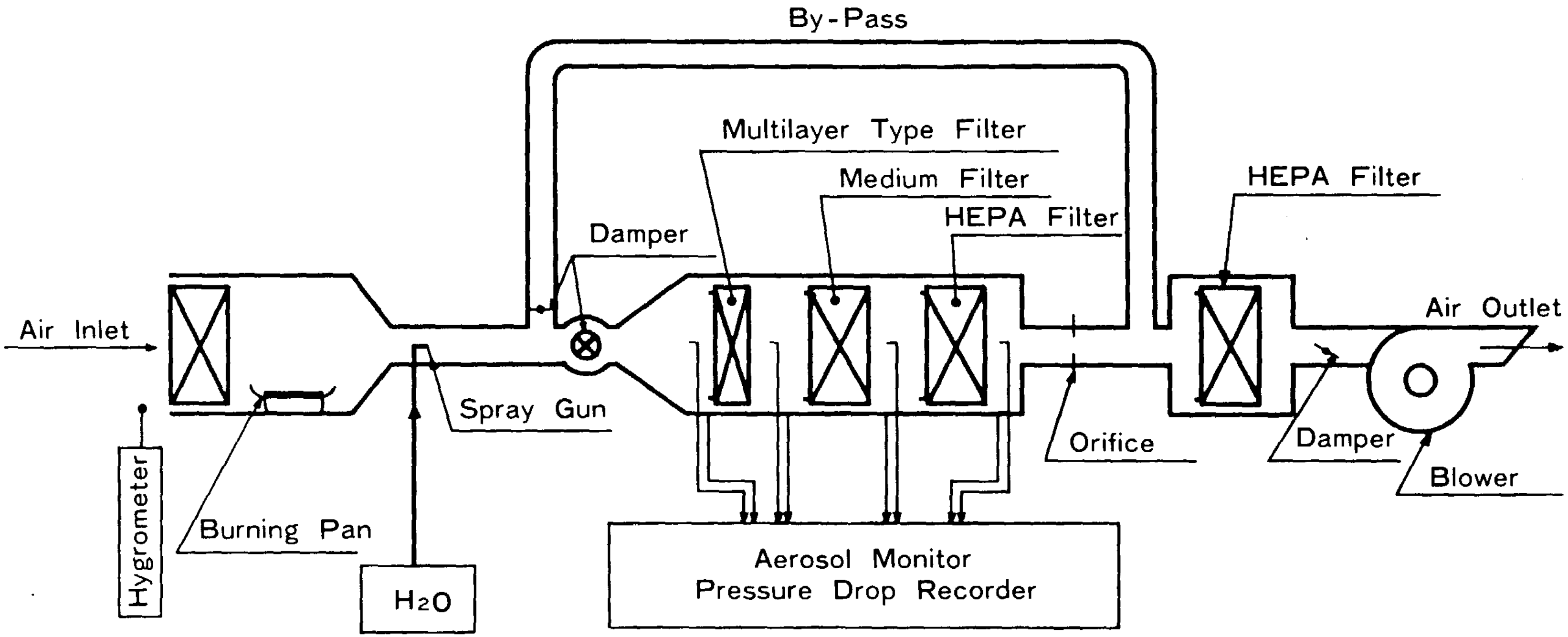


Fig. 2 Sodium aerosol test apparatus.

Sodium analysis

The amount of sodium trapped on filters was analyzed by atomic absorption spectroscopy. The values of Na on each filter were used to obtain the average removal efficiency of aerosol.

Medium filter

The specification of medium filter is given in Table 1.

Table I The specification of medium filter.

Filter Media : High Silica Content Fiber
Filter Size : $610^H \times 610^W \times 290^D$ (mm)
Air Flow Capacity : 28 m^3/min
Initial Pressure Drop: 17 kPa maximum
Removal Efficiency : 45 % minimum(0.3μmDOP)

Multilayer type air filter

Fiber media such as stainless steel, Zirconia, alumina, alumina-silica and high silica content glass were investigated for the multilayer type filter to remove sodium aerosol. As a whole, high silica content glass mat was selected from view points of high loading of sodium oxide aerosol and disintegration due to corrosion by sodium hydroxide. Fig. 3 shows a configuration of multilayer type filter with the diagonal stagger perforation of round hole (stagger angle 45°). The opening were arranged so as to meet flat part of the neighbor layer alternately. Diameter of openings and opening area ratio are connected with an equation regarding piches of openings as follows,

$$R = 157\ D^2/P^2 \qquad (1)$$

where

D : diameter of openings
P : center to center pich
R : opening area ratio.

The condition for openings on a layer to meet the filter media of the neighbor layer is given as $P > 2D$ and the maximum value is about 39 %. In this work, the opening area ratio was taken less than 30 %.

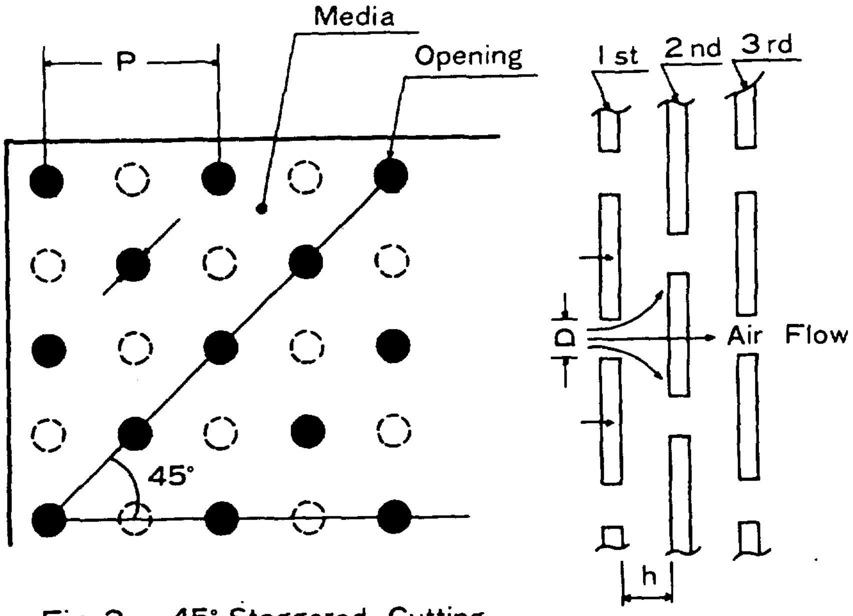


Fig. 3 45° Staggered Cutting

As to the diameter of openings, a range of 10 ~ 35 mm was taken for the practical usage. Table 2 shows the experimental parameter for diameter of openings, opening area ratios and number of openings.

Table 2 Experimental parameter of media.

Opening Area Ratios & No. of Openings / Diameter of Opening		10mmφ	15mmφ	20mmφ	25mmφ	30mmφ	35mmφ
※ 10%	Number of Opening	450	200	128	72	50	32
	Opening Ratio(%)	9.5	9.5	10.8	9.5	9.5	8.3
15	Number of Opening	722	338	162	128	72	50
	Opening Ratio(%)	15.2	16.1	13.7	16.9	13.7	12.9
20	Number of Opening	968	450	242	162	98	72
	Opening Ratio(%)	20.4	21.4	20.4	21.4	18.6	18.6
25	Number of Opening	1152	512	288	200	128	98
	Opening Ratio(%)	24.3	24.3	24.3	26.4	24.3	25.3
30	Number of Opening	—	648	338	242	162	128
	Opening Ratio(%)	—	30.8	28.5	31.9	30.8	33.1

※ Approximate opening area ratio.

On the other hand, the interval among layers was another factor. The effectiveness of intervals among layers were investigated according to Table 3.

Table 3 Setting of various intervals among layers.

Type of Multilayer	Interval Among Layers			
	1 st	2 nd	3 rd	4 th
A	20	20	20	20
B	30	20	20	10
C	45	20	20	10
D	70	20	20	10
E	25	20	15	10
F	40	30	20	10

(mm)

Filter system design

At first, a prior condition was given by a relation between the maximum holding capacity of sodium aerosol as Na and the maximum pressure drop regarding a HEPA filter at flow rate of 28 m^3/min. The holding capacity was taken as 200 g from the condition of 1 mg-Na/cm^2 and the pressure drop was 0.5 kPa,i.e., a standard value for exchange of HEPA filter in industries.

Table 4 A strategy of a filter system for an emergency filter system for FBR.

Filter Unit / Item		Pre-Filter	Middle Filter	Final Fiter	Total
		Multilayer Type Filter	Medium Filter	HEPA Filter	
Pressure Drop	Initial (kPa)	X_1	0.15	0.25	X_1 +0.4
	Final (kPa)	Y	0.50	0.50	1.50
Sodium Aerosol Holding Capacity (g) ※		X_2	130	200	X_2+330

※ Na base

The data on the medium filter were obtained experimentally by using sodium aerosol under a condition of 16.5 ℃, 41 % R.H. and flow rate of 28 m^3/min. The concentration of aerosol was 0.6 ~ 0.8 g/m^3 as Na. The data is shown in Fig. 4, where η_D is the value by Sibata digital dust counter. The amount of Na on the filter was determined by sodium analysis mentioned above. Thus, the maximum pressure drop was taken as 0.5 kPa and the corresponding aerosol holding capacity of Na as 130 g-Na for the medium filter.

The overall relation among filter units are shown in Table 4, in which each role of medium and HEPA filters are given respectively. The overall maximum pressure drop was taken as 1.5 kPa. From the table, it is required that the multilayer type filter should be designed properly with respect to pressure drop.

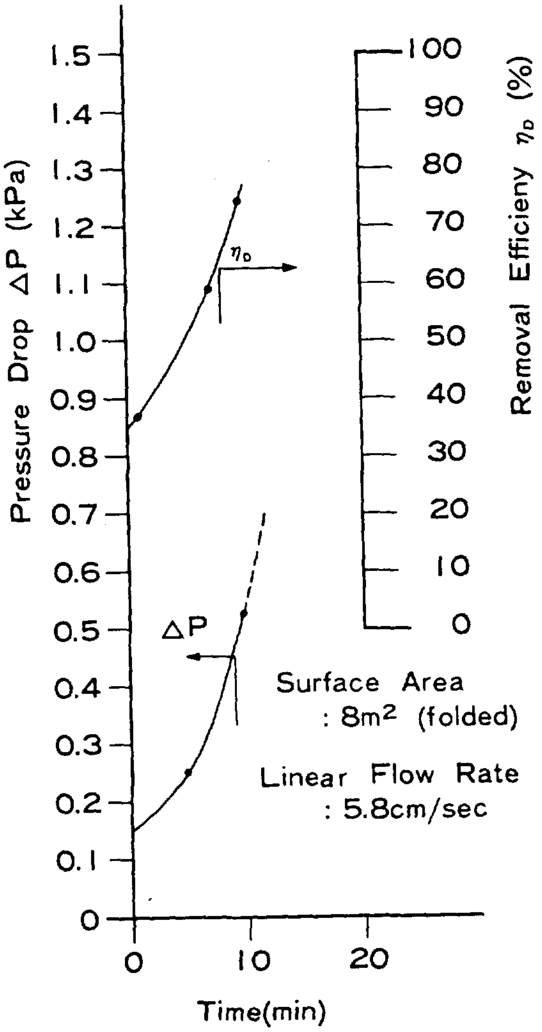


Fig. 4 The change of pressure drop and removal efficiency by high silica content fiber mat as a function of time (Medium filter).

Ⅲ. Result

Opening area ratio

The initial pressure drop for the multilayer type filter of five layers was investigated as a function of opening diameter and opening area ratio at the flow rate of 28 m^3/min. The result is shown in Fig. 5. From the figure,it is seen that the pressure drop is mainly determined by opening area ratio. The diameter of opening did not give much influence on the pressure drop. From Table 4, it was required that the highest pressure drop at the initial condition should be less than 0.4 kPa. From Fig. 5 the value corresponds to 20 % for opening area ratio.

Media : High Silica Content Fiber
Layers : 5 Layers
Interval : All 20 mm
Flow Rate : 28 m^3/min

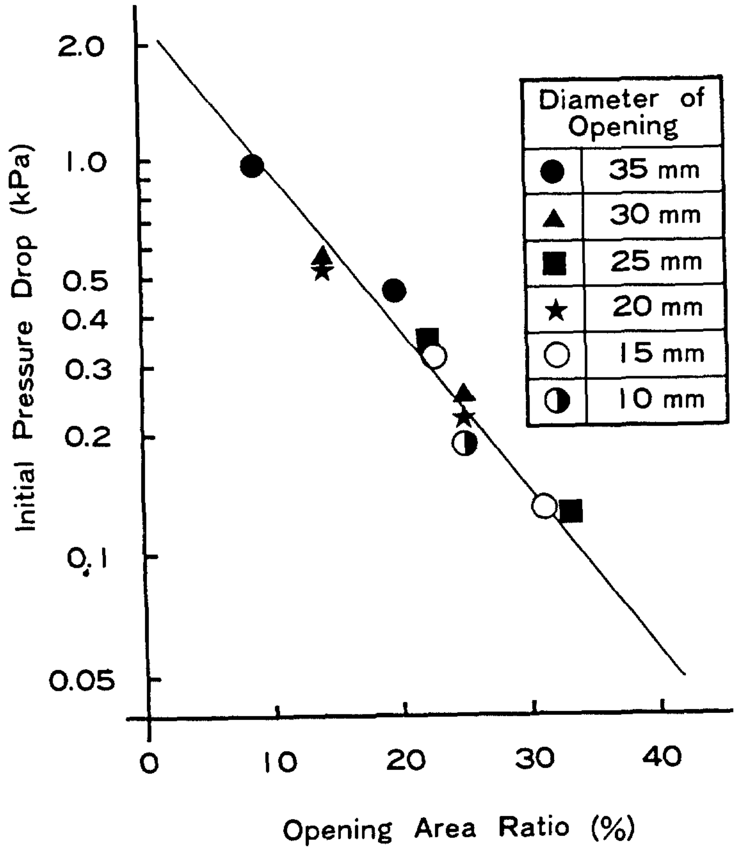


Fig. 5 The pressure drop as a function of opening area ratio.

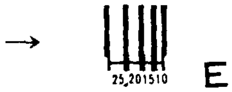
Diameter of opening

The effect of diameter of opening on the removal of sidium aerosol was examined. If the diameter of opening was smaller than 15 mm, the opening was clogged due to loading of sodium aerosol. So the diameter of opening was decided to fix 20 mm.

Intervals among layers

With a constant value of 20 % as the opening area ratio, the intervals among layers were set as shown in Table 3. The pressure drop for the types of A ~ F in Table 3 is given in Table 5.

Table 5 The initial pressure drop on several combination of multilayer at a room condition.

Run No.	Intervals of Layer(mm)	Flow Rate	Initial Pressure Drop
53-15		$28\ m^3/min$	0.338 kPa
53-16			0.345 kPa
53-17			0.348 kPa
53-18			0.357 kPa
53-20			0.39 kPa
53-21			0.375 kPa

Sodium aerosol

It was tried to generate sodium aerosol uniformly as much as possible by feeding sodium metal on buring pan manually in reference to the dust counter. Fig. 6 shows the data in which the concentration of sodium aerosol was relatively constant at the inlet of multilayer type filter. The average concentration of sodium aerosol was 0.6 ~ 0.8 g/m^3 as Na. The difference of sodium concentration before and behind filter gave the removal efficiency.

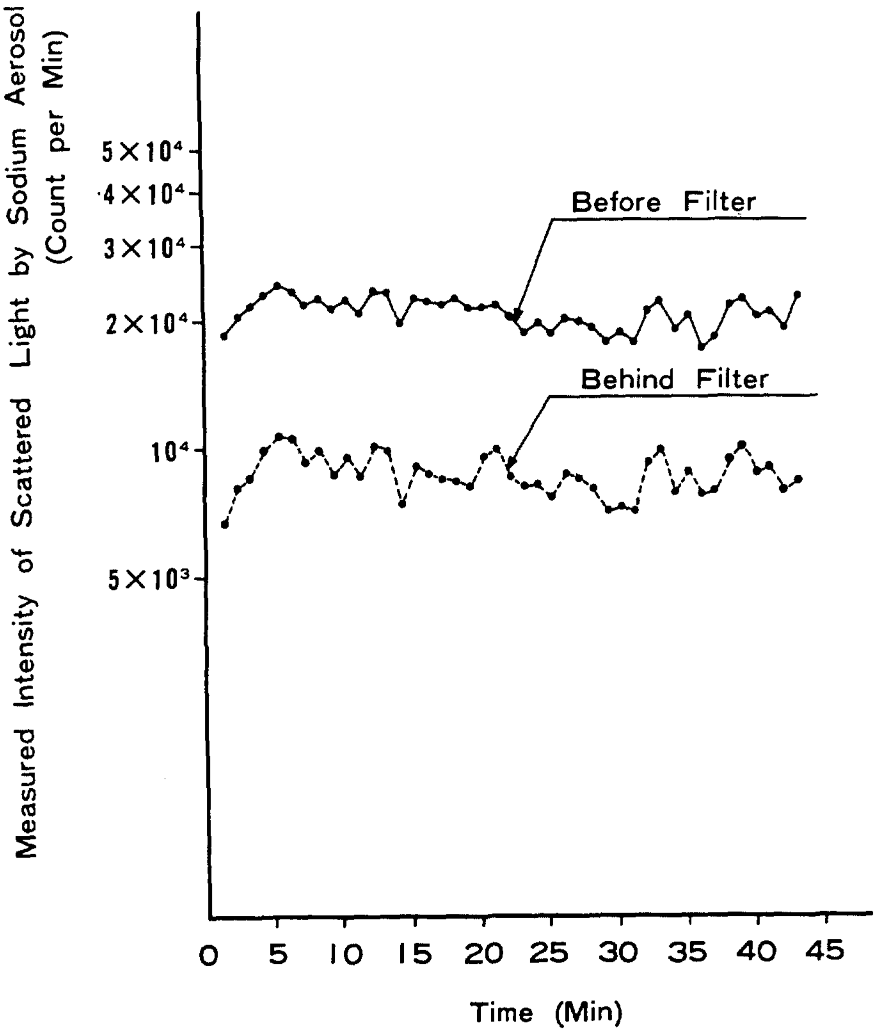


Fig. 6 Intensity of scattered light of sodium aerosol before and behind the multilayer type filter during run (Run 53-14).

Fig. 7 shows the photograph taken by an electron microscope. The particle size distribution was characterized with CMD (count median diameter) as 0.29 μm and geometric deviation σg =1.6.

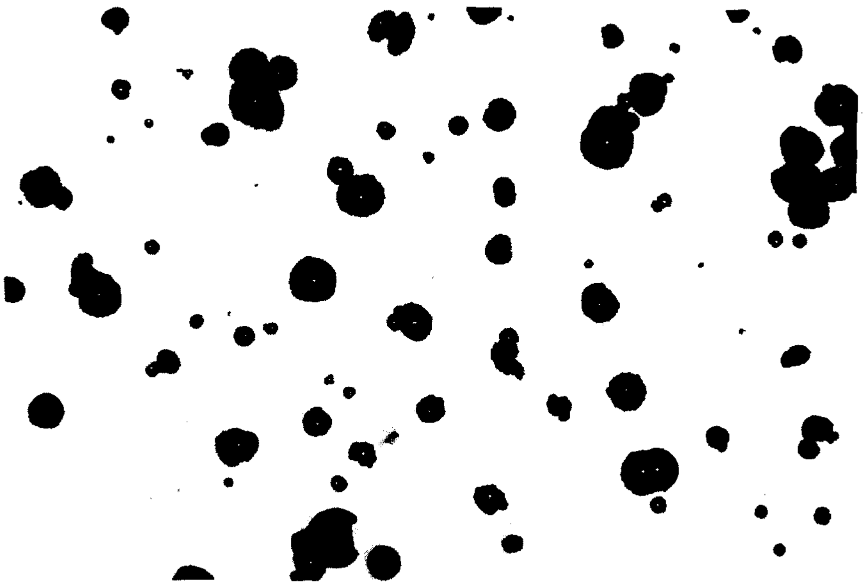


Fig. 7
Sodium Oxide Aerosol
Electron Microscope
(× 21656)
CMD = 0.29 μm
σg = 1.6

The appearances of trapped sodium aerosol on the first and second layers of the multilayer type filter is shown in Fig. 8. The sodium compound was distributed uniformly over the first layer,but it scattered on the second layer as lumps corresponding to openings of the first layer. On the downward layers after the second one, it was slight and uniform. It was noticed that the color of it was yellowish in dry condition, but white in a wet condition.

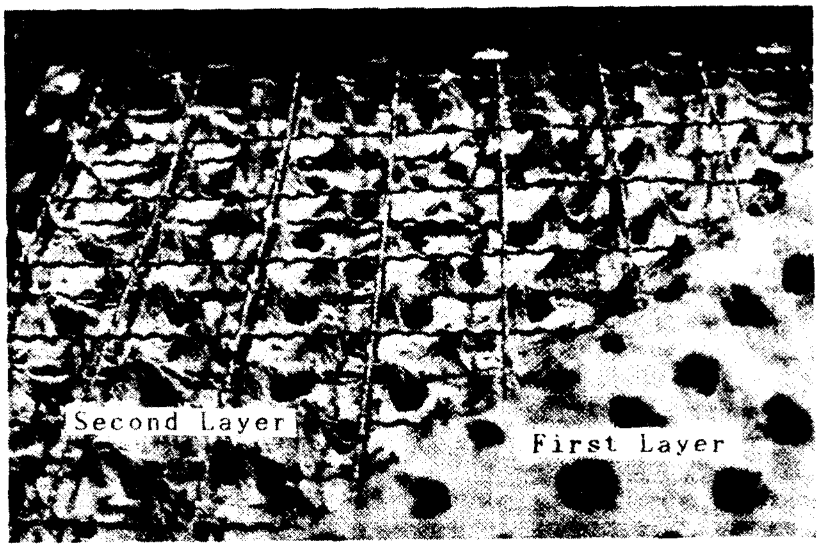
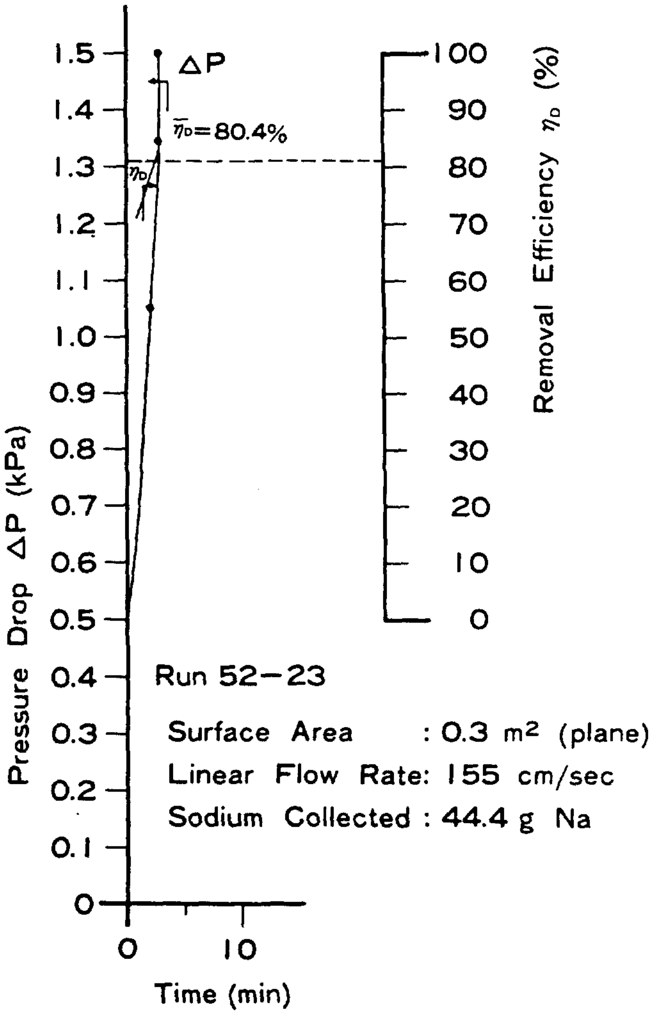


Fig. 8
Distribution of sodium aerosol on filter.

Sodium aerosol trapping on layer

a) Monolayer

Using monolayer of the same filter media,two runs were carried out to investigate the removal efficiency of sodium aerosol as a function of flow rate. One run was done at linear flow rate of 155 cm/sec which was corresponding to the first layer of multilayer type filter with 0.3 m^2 in area if it had no opening. The other was done at 5.8 cm/sec by setting the medium filter with 8 m^2 in area. The test results are shown in Fig. 9 for the former test and Fig. 4 for the latter one. It is seen from the figures that the removal efficiency increases with flow rate. At 0.7 kPa, the plane filter could trap sodium aerosol as much as about 16 g of Na per one filter unit or 5 mg-Na/cm^2. While the folded filter could trap about 150 g of Na per one unit or 1.9 mg-Na/cm^2.



Run 52-23

Surface Area : 0.3 m^2 (plane)
Linear Flow Rate: 155 cm/sec
Sodium Collected : 44.4 g Na

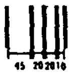
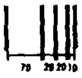
Fig. 9 The change of pressure drop and removal efficiency by high silica content fiber mat as a function of time (One layer of the multilayer type filter with no opening).

b) Multilayer

The runs for removal of sodium aerosol using several multilayers were carried out under 28 m^3/min up to 0.7 kPa of pressure drop. The results are shown in Table 6 and Fig.10. From Fig.10 it is seen that the first layer could trap 68 ~ 158 g-Na per layer or 18 ~ 42 $mg\text{-}Na/cm^2$ under 155 cm/sec of linear flow rate. These values are quite different from the case of monolayer in Fig. 9 in spite of the same linear flow rate, i.e. 5$mg\text{-}Na/cm^2$. It is considered that although most of aerosol would pass through openings in the first layer, a portion of aerosol might pass the filter media and the aerosol would be filtered efficiently as much as 100 % due to very low flow rate.

On the other hand, the second layer could trap sodium aerosol efficiently because of high air flow rate during passing through openings of the first layer: One mechanism is by filtration through filter media and the other is by inertia collision on the surface of filter media on which sodium aerosol was deposited. When sodium aerosol is loaded gradually on the second filter media, the air jets would be enlarged around the clogged portions.

Table 6 Result of Na loading on each layer.

Run No.	Intervals of Layers(mm)	Temp. (°C)	R.H. (%)	1 st (g)	2 nd (g)	3 rd (g)	4 th (g)	5 th (g)	Total (g)	Total Feed (g)	η ※ (%)
53-15	Air Flow →  A	11	50	156.5	196.0	78.6	41.1	47.3	519.4	694.4	74.8
53-16	→  B	6.5	54	112.4	233.2	82.7	54.1	47.2	529.6	1052.9	50.3
53-17	→  C	9	59	108.0	108.0	74.0	87.5	128.0	505.5	604.7	83.6
53-18	→  D	14	68	68.0	52.8	41.3	33.0	28.8	223.9	313.6	71.4
53-20	→  E	6.5	62	104.0	145.5	75.0	58.3	46.7	429.5	672.1	63.9
53-21	→  F	6.5	62	82.0	102.0	40.5	44.2	40.3	309.5	918.4	33.7

$$※\ \eta = \frac{\text{Total Loading (g)}}{0.8g/m^3 \times 28m^3/min \times \text{Time (Min)}} \times 100$$

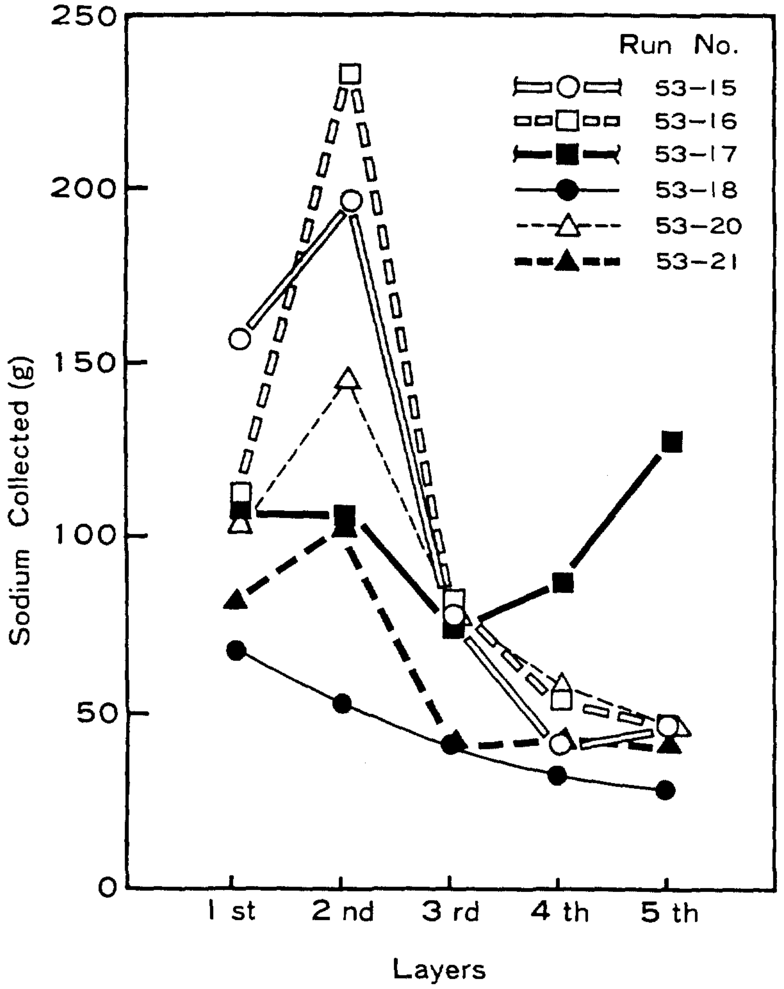


Fig. 10 Results of Na loading on each layer.

The controlling factor of the pressure drop was examined by appling metal plates with openings by arranging them according to Table 3. The type A showed 0.3 kPa and the type F 0.32 kPa. This means that the pressure drop mainly arises by air flow through openings, but not by collision of air jet onto the plate. It is estimated that the increase of pressure drop may be arisen by clogging of openings by sodium aerosol. So that, it is necessary to evaluate not only the removal efficiency of aerosol, but also the pressure drop. It is found that the second layer contributes effectively towards the solution of high holding of sodium aerosol with low increase of pressure drop. In other words, if tried to distribute sodium aerosol equally over every layer, the aerosol deposited worked to clog the openings of layers. From the results, the interval among the layers was taken 20 mm equally, because it gave better effectiveness and also simplicity for manufacturing.

Effect of humidity

The effect of humidity on the emergency air filter system of FBR was concerned. The test was carried out for the overall filter system. The results are shown in Table 7. The increase of pressure drop was given as a function of time in Fig.11. The overall pressure drops with loading of sodium aerosol on the filter system are summarized in Fig.12 with respect to the absolute humidity. It is found that the higher the absolute humidity, the lower the increase of pressure drop. From the result, it is concluded that the minimum loading of sodium aerosol is 800 g-Na per one filter system.
The effect of water droplets on the filter system was also examined. Water droplets could reduce the increase of pressure drop definitely as shown in Fig.13.

Table 7 Na aerosol loading test.

	Run No.	Temp. (°C)	R.H. (%)	D.P. (°C)		Multilayer Type Filter	Medium Filter	HEPA Filter	Total
In Normal Air	53−1	22.5	74	17.5	Na Loading (g)	527.5	179.6	274.9	981.0
					η_w (%)	53.7	39.5	≒ 100	
	53−3	26.5	34	9.1	Na Loading (g)	616.3	199.1	120.1	935.5
					η_w (%)	65.9	62.4	≒ 100	
	53−14	11.5	90	9.6	Na Loading (g)	623.9	148.8	231.3	1,004.0
					η_w (%)	62.1	39.1	≒ 100	
	53−22	6.5	62	0	Na Loading (g)	392.7	277.1	152.2	822.0
					η_w (%)	47.8	64.5	≒ 100	

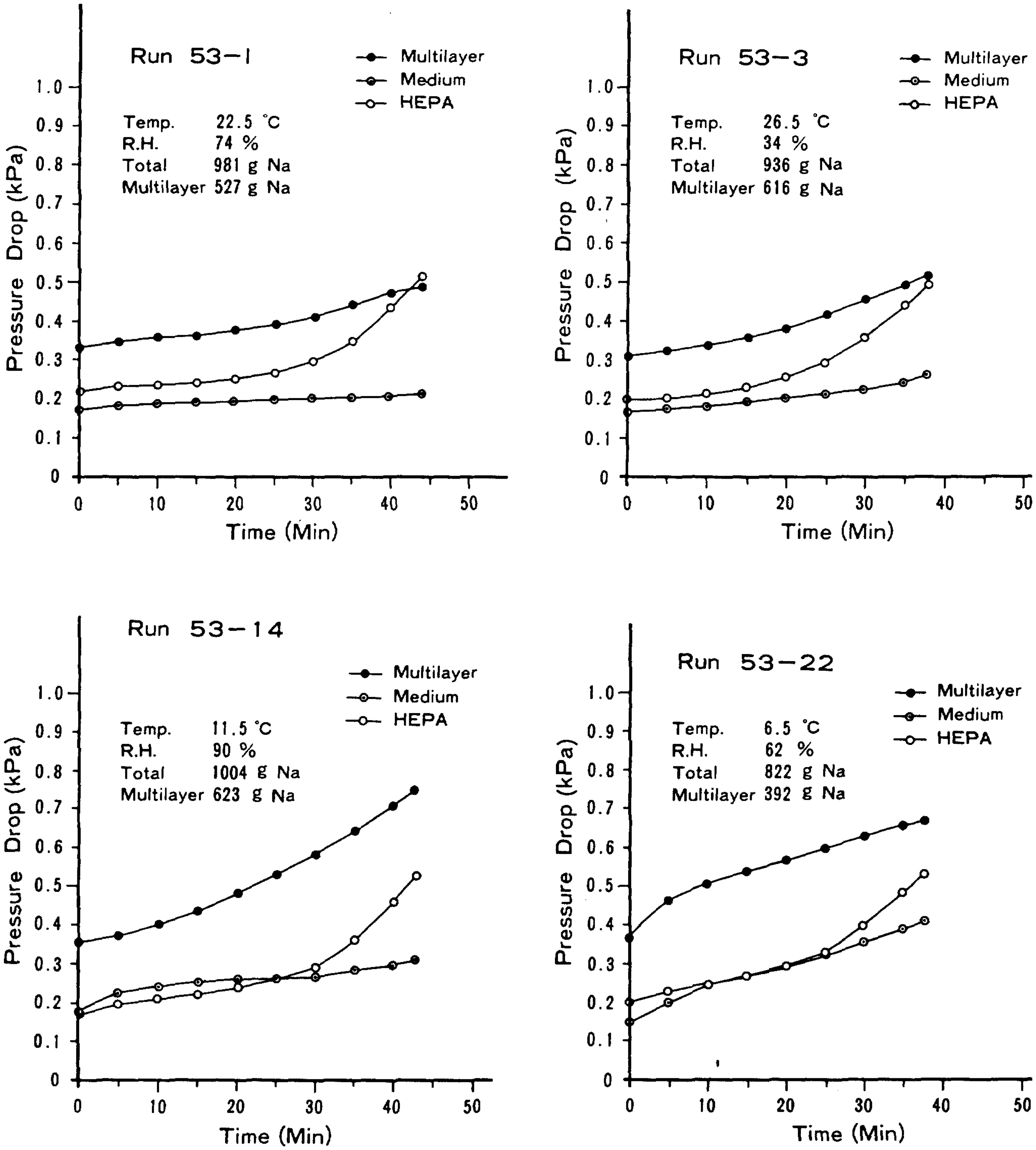


Fig. 11 The change of pressure drop as a function of time.

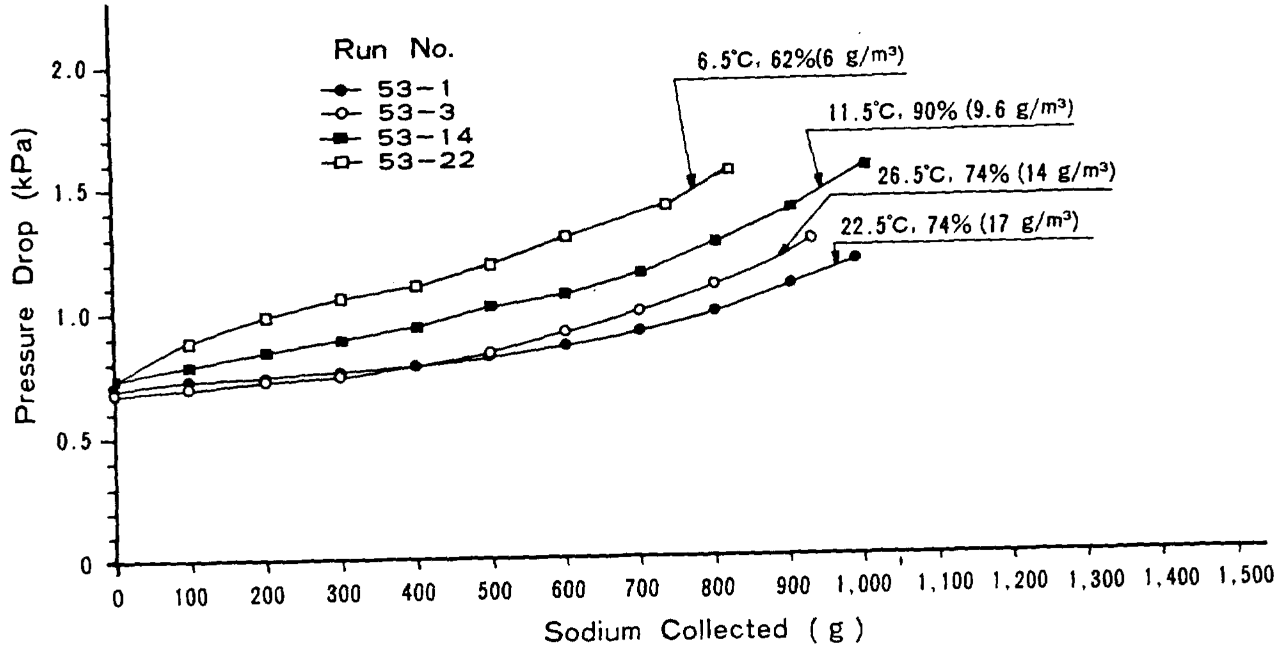


Fig. 12 The change of pressure drop during loading of sodium aerosol.

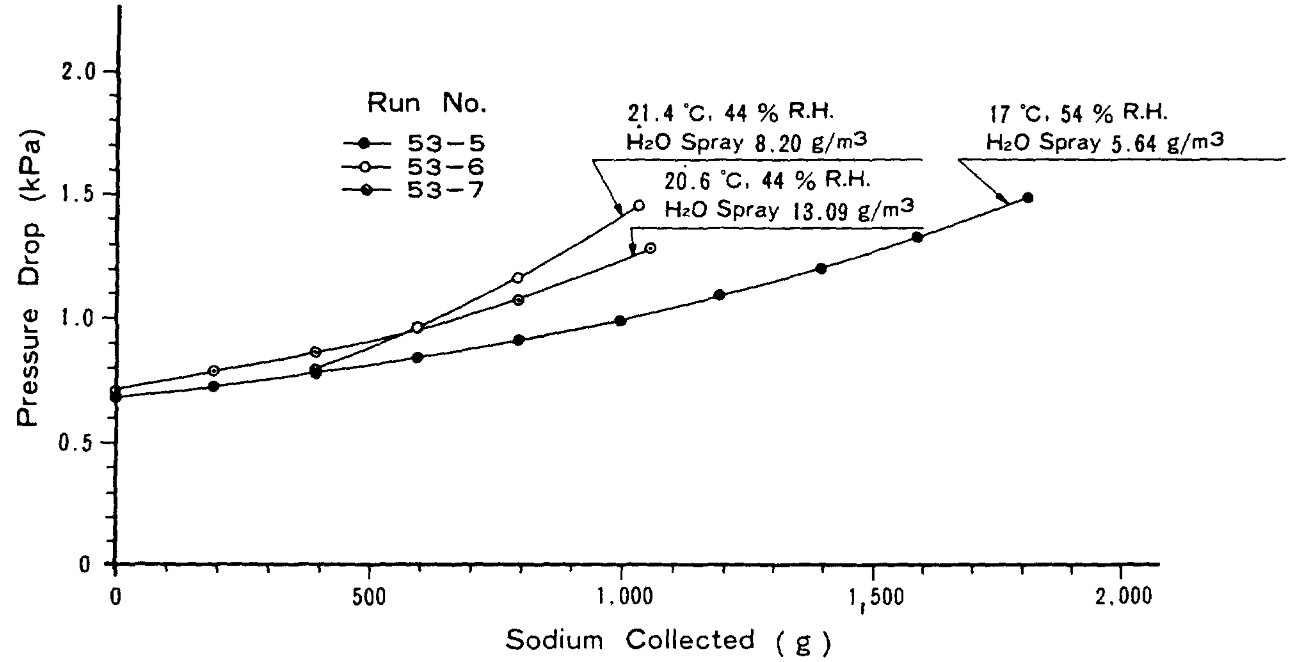


Fig. 13 The change of pressure drop during loading of sodium aerosol.

IV. Summary

(1) A filter system consisting of a multilayer type filter - a medium filter -a HEPA filter with standard size(610mm × 610mm)were examined to develop an emergency air filter system of FBR, where the multilayer type and medium filters are for protection of HEPA filter.
(2) High silica content glass fiber was selected for the filter media for the multilayer type filter as well as the medium filter.
(3) A series of run were carried out for the design of multilayer type filter to remove aerosol effectively. The filter consisted of 5 layers of fiber mat with openings of 20 mm in diameter and opening area of 20 %.
(4) The high humidity reduced the increase of pressure drop of the filter system.
(5) It was confirmed that an emergency air filter sustem of FBR consisting of a multilayer type filter - a medium filter - a HEPA filter with standard size as 610mm × 610mm in series could hold 800 g-Na at 1.5 kPa of the overall pressure drop.

Reference

(1) Private comunication with Dr.Kitani.S, Messrs.Uno.S, Takada.J, and Takahashi.K of Japan Atomic Energy Research Institute.(1972)
(2) Jordan.s, Alexas.A, Lindner.W, Filtration of Sodium Fire Aerosol, Trans.Am. Nucl. Soc.,24,526(1977)
(3) McCormack.J.D, Hilliard.R.K, and Barreca.J.R, Loading Capacity of Various Filters for Sodium Oxide/Hydroxide Aerosols, Proceeding of 15th DOE Nucl. Air Cleaning Conf.,
(4) Bohn.L, Jordan.S and Schikarski.W: Experiments on Filtration of Sodium - Aerosol by Sand-Bed-Filters, CONF-740401, 461 (1974)

DEVELOPMENT AT THE KARLSRUHE NUCLEAR RESEARCH CENTER (KfK) OF REMOTELY OPERATED FILTER HOUSINGS AND FILTER ELEMENTS FOR REPROCESSING PLANTS

K. Jannakos, H.J. Becka, G. Potgeter, - IT
J. Furrer - LAF II
Kernforschungszentrum Karlsruhe
Federal Republic of Germany

Abstract

Under the Reprocessing and Waste Treatment Project (PWA) development work is carried out at the KfK on fabricating and testing remotely operated filter housings.and filter elements for wet and dry aerosol retention. The remotely operated filters are used mainly to clean gaseous effluents containing a high portion of radioactive aerosols, e.g. for cleaning dissolver offgases from reprocessing plants.

Development work on the remotely operated filter housings has produced a third generation of filter housings which are modular in design so that in an accident parts of the housing and of the remote handling mechanism, respectively, can be replaced by remote operation using a crane and a manipulator.

The development of filter elements for aerosol retention started from the rectangular design via a quasi-circular design to a polygonal filter element for dry aerosol retention. This filter element is suited for gas flows up to 3000 m^3/h and temperatures up to 200 °C. It can be replaced by remote handling and compressed to about one third of its volume so that the waste volume can be minimized.

Remotely operated filter elements with circular packed fiber glass (single ring layer filter) are used for wet aerosol retention. The packed surface available with a packing thickness of about 50 mm governs the maximum gas flow achievable. It has been the goal of development work to optimize the packed surface for a specified size of the filter element in order to keep low the number of filter units needed to purge a gas flow. Two variants of filter elements have been developed, the double ring layer filter and the star-shaped layer filter.

The technical development of the filter elements has now been completed but testing is still going on, above all as regards remote handling, tightness, decontamination factor for various offgas parameters (humidity, temperature, pressure, NO_x), corrosion, and under accident conditions.

I. Introduction

For cleaning offgases carrying a high activity inventory in nuclear facilities - e.g. the dissolver offgas in reprocessing - special filters and remote handling devices are needed which guarantee a high removal efficiency largely verifiable, are safe in operation and amenable to remotely operated replacement, packaging and removal of the loaded filter elements from the site.

Considering the development work underway as well as planning work on a reprocessing plant in the Federal Republic of Germany, development of suitable filters and remote handling devices for cleaning the dissolver offgas of large reprocessing plants was started in 1975 under the Reprocessing and Waste Treatment Project (PWA). The objectives included:

- Development and testing of a remotely operated filter housing for accommodation of aerosol and iodine filter elements inclusive of the necessary handling devices.
- Development and testing of filter elements capable of remote replacement for retaining total volume flows of aerosols and iodine of up to 3000 m^3/h.

Some of the development work, mainly that part related to filter housings and remote handling, was already reported in 1980 at the 16th DOE Nuclear Air Cleaning Conference (1).

The technical development has meanwhile been completed. Testing of the improved filter housing and filter elements is still going on, above all as regards remote handling, tightness, removal efficiency for various offgas parameters (e.g. humidity, temperature, pressure, NO_x), accident conditions, and corrosion phenomena.

In this report the remotely operated filter housing and the filter elements are described and operating experience accumulated until now on remote handling, readiness for operation, corrosion and sealing behavior is presented.

II. Remotely Operated Filter Housing

In the course of development work concentrating on special applications three generations of filter housings have been developed. They differ mainly in their remote handling and maintenance concepts which had been worked out for the filter cell depending on the status of development work.

Figure 1 shows the filter housing of the third generation. It consists of the vessel with the hinged lid and clamping mechanism. The vessel is of vertical design. The connections for gas inlet and outlet and for cleaning the inner housing wall and monitoring the tightness of the filter element seat are provided on the vessel. The filter element is installed in the vessel in such a manner that it ensures a gastight isolation between the upstream and downstream air faces. The filter element can be passed by the flow either axially or radially, depending on the type of the filter element (iodine or aerosol filter) used.

The filter housing is suited for accommodating filter elements capable of retaining iodine, wet and dry aerosols. The lid and clamping mechanism are the same in all filter elements; only at the filter housing minor modifications are necessary. Figure 1 shows an

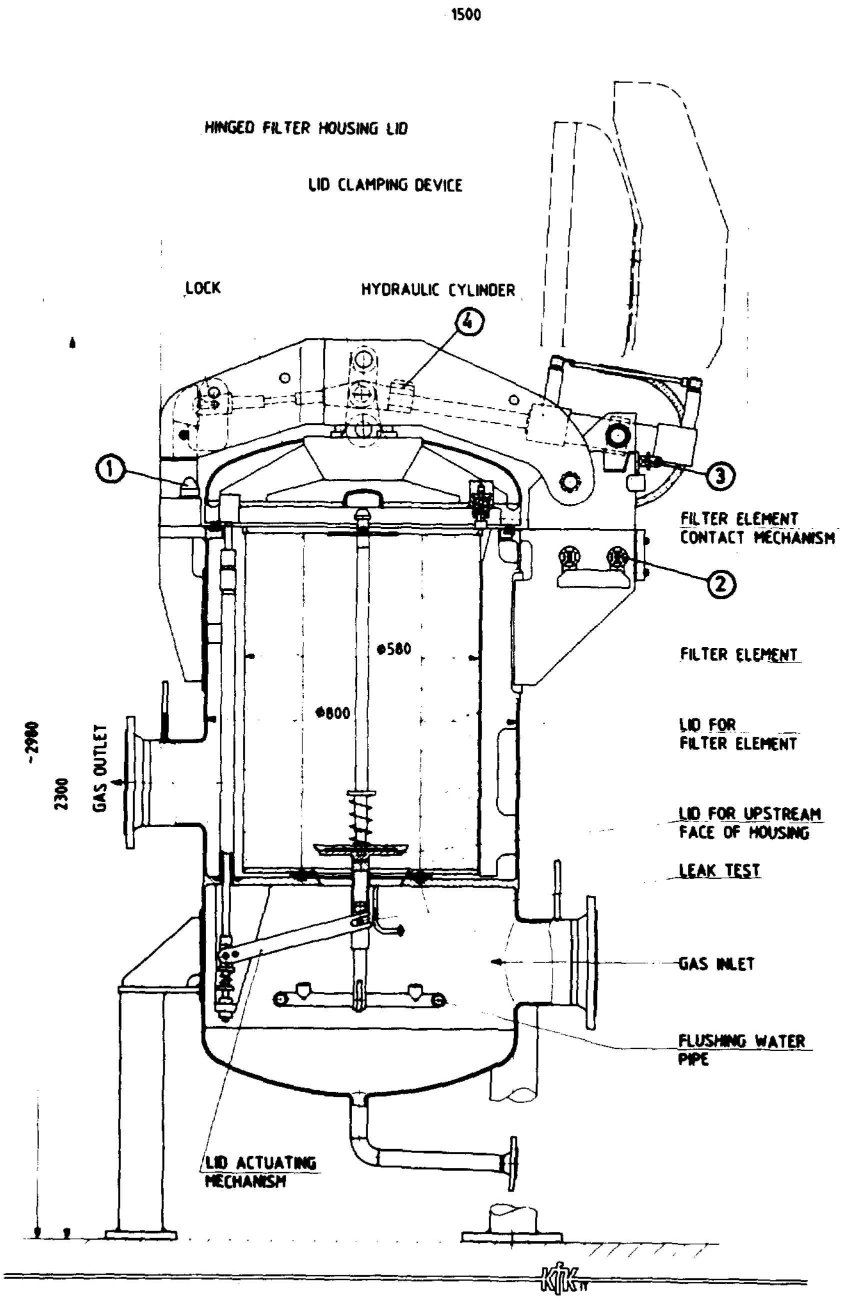


FIGURE 1
REMOTELY OPERATED FILTER HOUSING WITH HEPA FILTER

aerosol filter element. The filter housing can be optionally provided with a lock mechanism in order to avoid that the downstream face of the filter housing becomes contaminated during filter changeout. The locking plate of the filter housing at the inlet of the filter element is automatically controlled via an actuating bar from the lid of the housing. The aperture towards the filter element is open when the lid of the housing is closed and closed when the lid of the housing is opened.

The clamping mechanism (carrying the lid of the housing) is installed at the housing in such a manner that it can be hinged and is actuated hydraulically. When the housing lid is closed the clamping mechanism is the first part that contacts the locking hook. The lid does not yet contact the sealing of the housing. As the movement of the hydraulic piston continues, the contact mechanism gets locked, the lid is moved vertically downwards via the elbow lever joint and the guide bolts and pressed onto the sealing. When the hydraulic piston has reached its end position the elbow level joint is above the central axis and self-supporting. This means that the lid of the housing remains closed in the absence of hydraulic pressure. The filter element is pressed by the lid via plate spring gaskets. These also serve to compensate fabrication tolerances or elongations of the filter element during operation. The maximum possible clamping force for the lid and filter element is about 100 kN. A maximum of 40 kN is required. About 20 kN each are needed for the lid of the housing and the filter element. The total clamping force or the individual clamping forces for the lid and filter element, respectively, can be optionally indicated at the operator's panel. This indication supplies valuable information about the condition of the sealing of the housing and the pressure applied to the filter element.

The hydraulic system for opening and closing of the housing lid is outside the filter cell. Only two lines of about 10 mm outer diameter lead to the filter housing.

The inner diameter and the height of the filter housing can be adapted to the requirements. To this day filter housings of up to 790 mm inner diameter and 840 mm inner height have been tested. The filter housings have been designed for the pressure range of 0 to 2.5 bar absolute pressure. Figure 2 shows the filter housing installed in the test bench.

The description above applies to all three generations of filter housings. For the third generation the lid and the hydraulic cylinder have been designed in conformity with the status of the handling technique developed for the German reprocessing plant in such a manner that they can be replaced remotely in case of failure. It can be noted in Fig. 3 that a crane and a heavy load manipulator are required to equip the filter cell. This equipment is adequate for filter cells accommodating a large number of filter housings.

Should the hydraulic cylinder fail, the hydraulic lines equipped with rapid action couplings are decoupled from the housing with the heavy load manipulator, the fastening bolts for the hydraulic cylinder are withdrawn and laid down in the magazine and the hydraulic cylinder is lifted with the crane, removed and posted out. A new hydraulic cylinder is mounted in the reverse order.

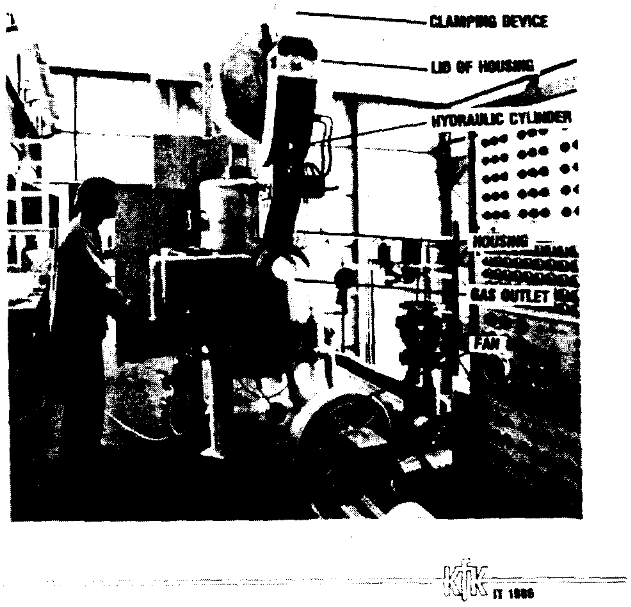


FIGURE 2
TEST FACILITY
FILTER HOUSING WITH OPEN LID

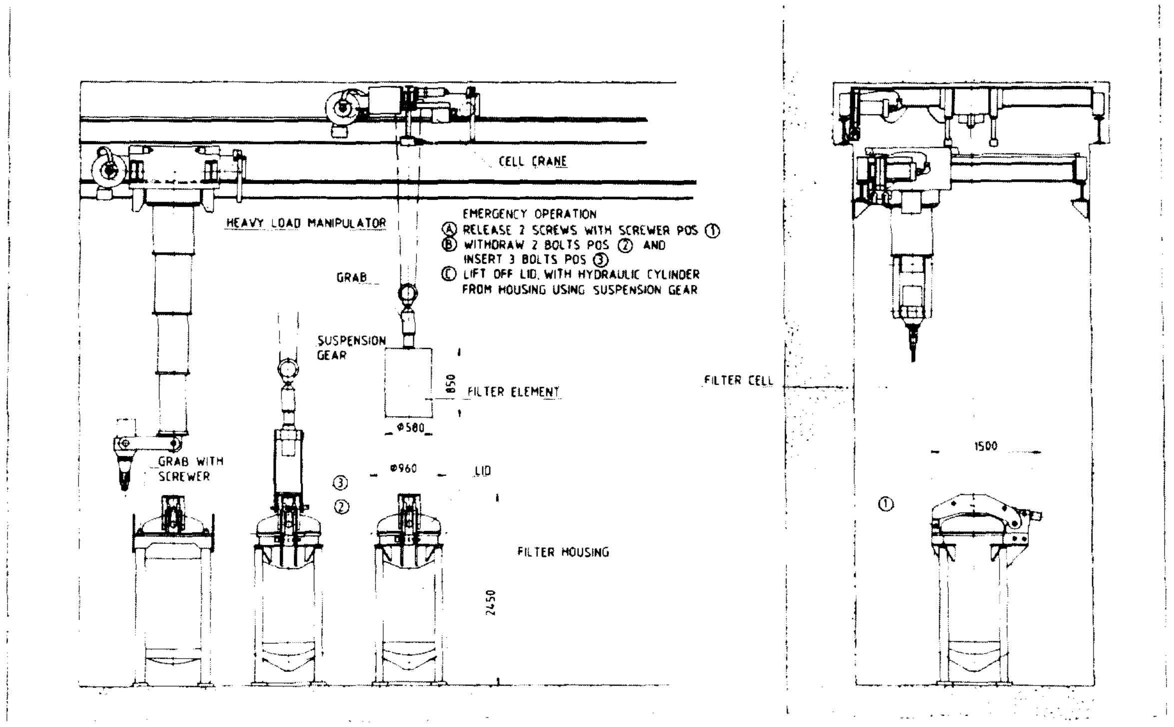
Figure 4 shows the withdrawal of the fastening bolt for the hydraulic cylinder and Fig. 5 shows the removal of the disassembled cylinder during the phase of testing.

If the mechanism at the housing lid fails, it is replaced together with the clamping device. The lid in the contacted state (Fig. 1) is dismounted as follows: The two fastening screws (¹) are screwed out with a screwer provided at the manipulator (thus relieving the clamping device), the two fastening bolts (²) are withdrawn and the hydraulic lines decoupled. A transport device is coupled to the lid using the crane and the manipulator so that the lid can be lifted and removed (Fig. 6). The repaired or the new lid is mounted on the housing in the reverse order.

The sealing between the lid and the housing which is attached to the housing is replaced remotely using a special device.

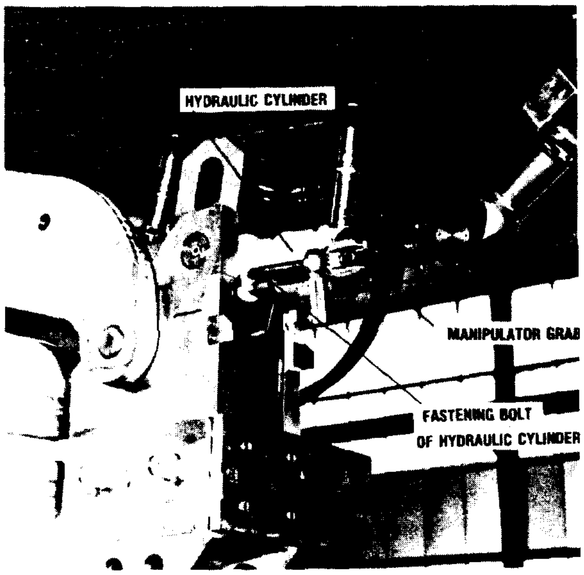
The third generation of filter housings has been tested since early 1986. Testing has related mainly to remote handling because the clamping mechanism and the filter housing had been tested already sufficiently for the first and second generations.

The first generation of filter housings (Fig. 7) has been designed in such a way that in case of failure of the mechanism of the lid the latter can be opened and closed with an emergency lever operated by the crane (without locking). After the loaded filter element has been withdrawn and the inner housing surfaces cleaned the mechanism of the lid can be repaired either in situ by staff wearing protective clothes or the lid can be dismounted from the

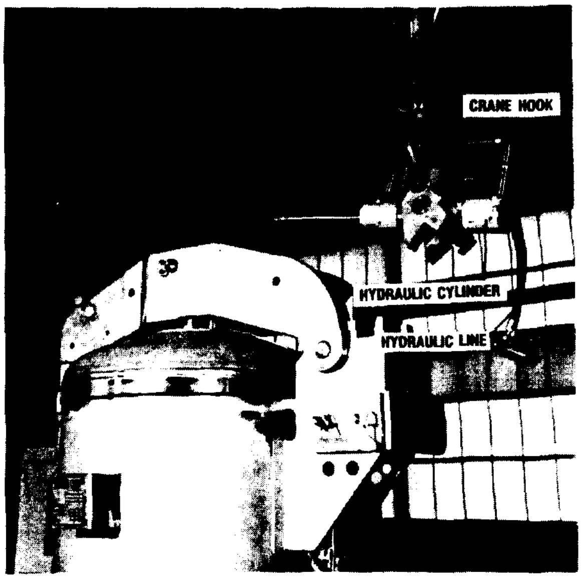


KfK IT

FIGURE 3
REMOTELY OPERATED FILTER HOUSING WITH NECESSARY CELL EQUIPMENT



IT 1986



IT 1986

FIGURE 4
FILTER HOUSING
REMOTELY OPERATED REMOVAL OF
HYDRAULIC CYLINDER

FIGURE 5
FILTER HOUSING
REMOTELY OPERATED REMOVAL OF
HYDRAULIC CYLINDER

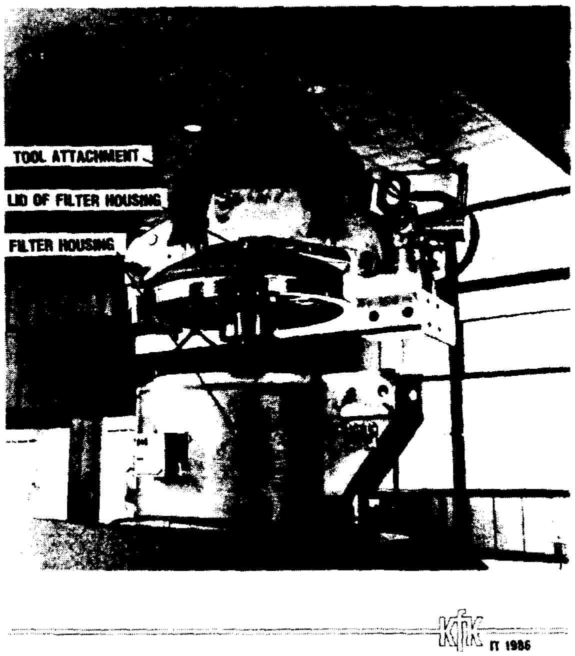


FIGURE 6
FILTER HOUSING
REMOTELY OPERATED LID REMOVAL

housing and repaired outside the filter cell. This design of the housing is particularly suited for small filter systems which can be cleansed and taken out of service before the filter cell is entered.

The filter housings of the first generation have undergone a sufficient amount of testing since 1978 with iodine and aerosol filter elements (three filters). The offgas values (temperature, pressure, iodine and NO_x fractions, aerosols, steam) selected for the filter testing phase resembled largely those which are expected to occur in the dissolver offgas of a reprocessing plant.

Since 1984 housings of this type have been used together with HEPA and fiber pack filters in the PAMELA (vitrification pilot plant for HLLW) facility which started active operation in 1985. According to information received from the operator, DWK, they have so far performed well with a high decontamination factor (fiber pack filter = 3×10^3, two HEPA filters in series $\geq 10^5$).

As the filter housing can be readily handled, there have been no errors in handling so far. No wear and corrosion phenomena have been visible although the filter housings have been made from the usual stainless steel, material No. 1.4541. The sealing of the housing consists of a silicon mixture resistant to NO_x, steam and HNO_3. Following the initial permanent deformation of the sealing (about 5%) no further changes have been detected.

In the filter housings of the second generation (Fig. 8), developed for a specific purpose, the vessel of the filter housing has been screwed to the bottom of the filter cell. The bottom of the filter cell has been made as shielding so that the radiation emanating from the gas lines and the loaded filters is adequately attenuated and work can be performed in the filter cell. The lid of the filter housing which is likewise provided with a shielding of the same thickness as the bottom of the cell, is installed on the cell bottom. Emergency handling devices as used for the first and third generations of housings can be dispensed with because the filter cell can be entered in protective clothes for doing repair work. This filter design has been tested since 1980 using the same offgas values as in the first filter generation (two filters). No differences have been observed in filter handling, corrosion, wear, removal efficiency and reliability, compared with the first generation of filter housings.

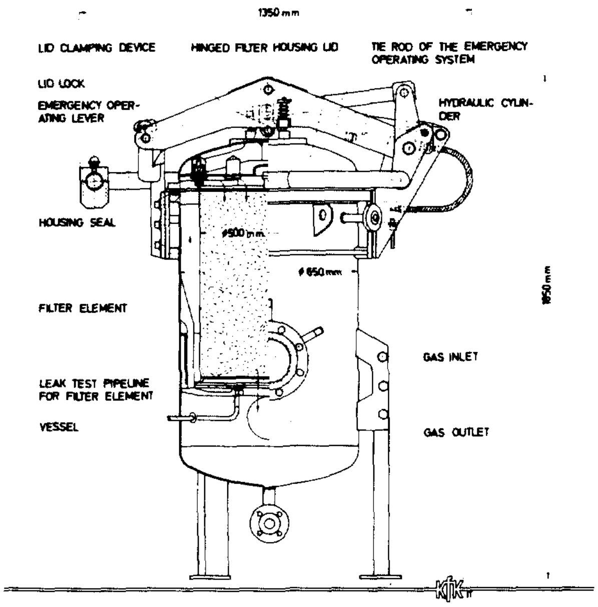


FIGURE 7
REMOTELY OPERATED FILTER HOUSING
WITH IODINE FILTER ELEMENT
(VERTICAL DESIGN)

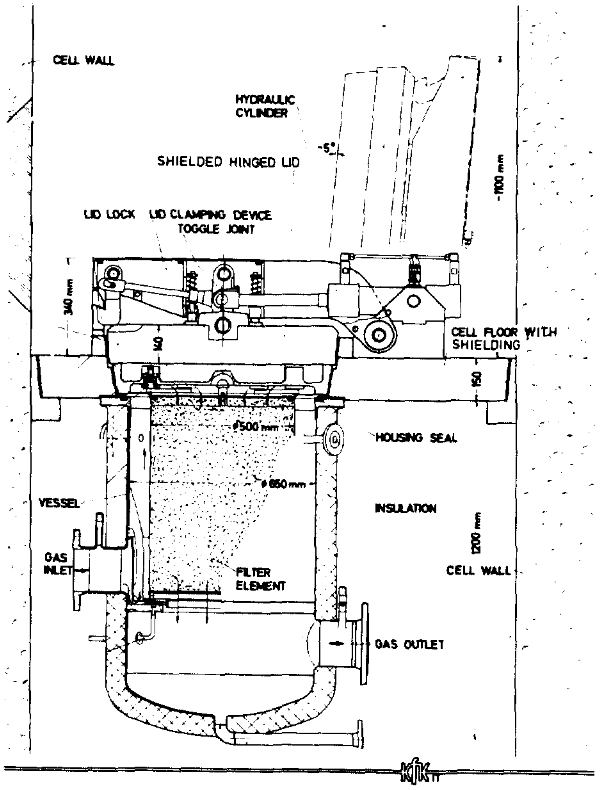


FIGURE 8
REMOTELY OPERATED "SUSPENDED"
FILTER HOUSING WITH SHIELDING
AND THERMAL INSULATION

III. Filter Element for Aerosols (HEPA)

To retain aerosols in offgases carrying a high portion of radioactive aerosols, e.g. dissolver or vessel offgas in reprocessing, filter elements are needed which largely meet the following conditions:

a) Operating conditions: The filter elements shall be resistant to
   - high permanent temperatures of about 150 °C,
   - pressure variations,
   - acid,
   - NO_x and iodine.

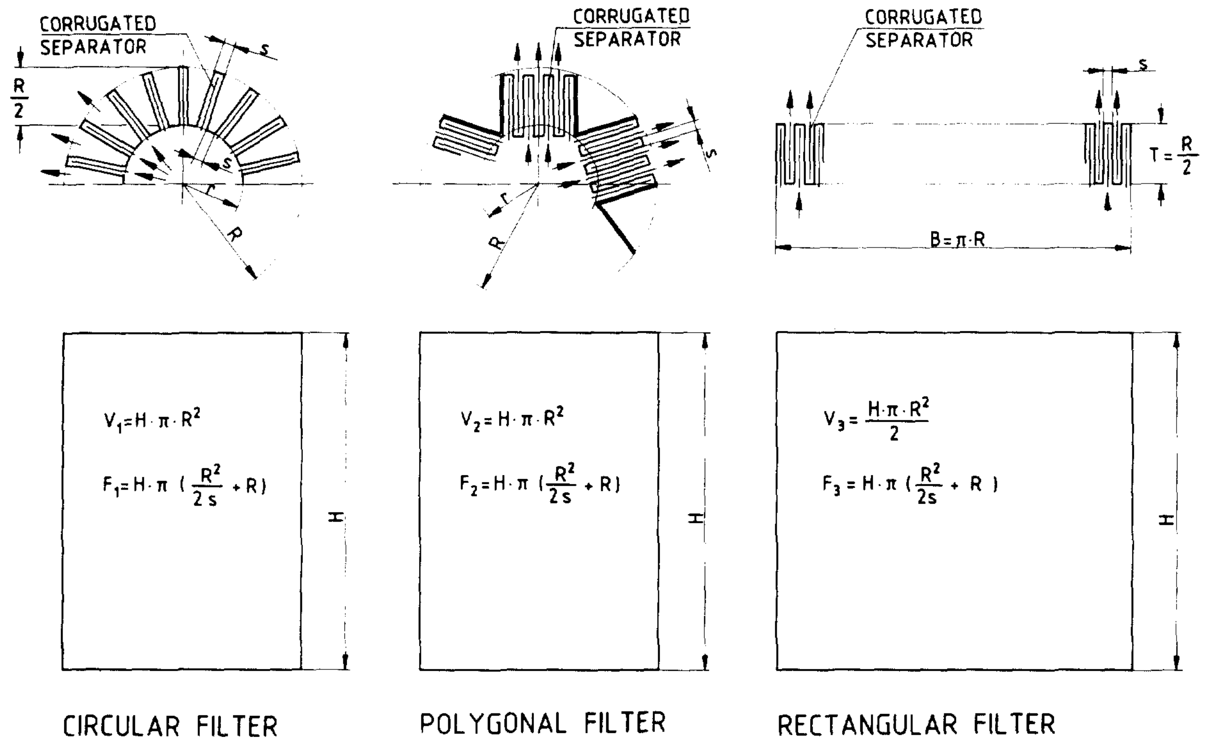
   Moreover, they shall ensure
   - a high permanent removal efficiency,
   - a good and verifiable seat of the sealing.

b) Handling conditions: The filter elements shall have the following characteristics:
   - mechanical protection from damage from outside (transport, installation) and from inside (gas entrained particles),
   - adequate strength,
   - capability of remote handling.

c) Conditions regarding ultimate disposal and rentability: The filter elements shall have the following characteristics:
   - small volume of waste (compressible),
   - low weight (minimum amount of stainless steel material),
   - large paper filter surface and small filter volume (low number of filters),
   - convenient fabrication (little machining of individual components).

III.1 Generalities

Until this date rectangular filter elements (about 140 mm width, about 270 mm height) have been used in the Federal Republic of Germany to filter offgases carrying a high fraction of radioactive aerosols, e.g. the dissolver offgas in pilot and testing facilities. These rectangular filter elements are screwed to a pentagonal stainless steel frame and sealed so that they make up a quasi-circular filter element. In terms of the paper surface available, the sealing, the required weight of the stainless steel mass and the compression, this design is highly unfavorable. Therefore, a new filter element has been developed which meets largely all requirements indicated under III. As the circular shape is the most favorable with a view to remote handling and ultimate disposal, it has been adopted. In order to be able to transfer to the new filter element the good results in operation and removal efficiency which had been obtained during operation and in many years of testing quasi-circular filter elements, the paper quality, the paper format, the spacing of pleats, the spacers and the packing density have been maintained so that only a new polygonal filter paper support has been developed. The polygonal filter element combines the advantages of the circular and rectangular filter elements in an optimum manner. The surface of the filter paper (Fig. 9) is of the same size with the radius (R), height (H) and spacing of pleats (s) specified in advance. However, no spacers can be provided in the circular filter element in the pleats, which are open towards the outside, because the distances between the paper pleats undergo radial changes. Also preliminary pressing of the filter paper, as performed in the rectangular filter element in order to make the block more stable, cannot be practiced. In the polygonal filter element both disadvantages can be avoided.



$r = 145$
$R = 290$
$H = 820$
$F_1 = F_2 = F_3$

FILTER PAPER SURFACE FOR CIRCULAR FILTER $F = H\,[\frac{2 \cdot \pi \cdot r}{s}(R-r) + 2 \cdot \pi \cdot r\,]$

CONDITION FOR MAX. FILTER PAPER SURFACE $\frac{dF}{dr} = 0 \longrightarrow r = \frac{R+s}{2}$

FIGURE 9
COMPARISON OF SIZES AND ARRANGEMENT OF FILTER PAPERS OF VARIOUS FILTER ELEMENTS

The conditions to be met for a maximum filter paper surface of circular and polygonal filter elements, respectively, with the external radius specified, is dF/dr = 0. The paper surface is

$$F = H[\frac{2\pi r}{s}(R - r) + 2\pi r]$$

where:

F = filter paper surface,
r = inner radius of the filter,
R = outer radius of the filter,
H = filter height.

In order to satisfy the condition above, r must be approximately R/2 [precisely r = (R+s)/2]. For this value the paper surface is

$$F = H\pi\left(\frac{R^2}{2s} + R\right)$$

Consequently, with the values given for the filter radius and filter height, it depends solely on the spacings between the pleats. Considering the filters used and tested until now, it has been chosen to lie between 2.5 and 4 mm.

Figure 9 shows for the same filter paper surface the true scale size of a circular, a polygonal and a normally used rectangular filter element.

It can be expected (the relevant tests have not yet been completed) that the polygonal filter element behaves more favorably as regards tearing and blowing out of the filter paper exposed to fluctuations in pressure and mass, compared to the currently used rectangular filter element, because it is made up of smaller individually supported paper surfaces.

III.2 Description of the Polygonal Filter Element

The polygonal filter element, Fig. 10, consists of the filter paper and the frame. The frame is made up of a bottom and a lid connected with each other by a supporting tubular structure. The seat gasket is glued to the bottom and fastened in addition by a holding ring. Both faces of the gasket (glued and contacting faces) are monitored for tightness during operation. The force from the lid to the bottom gasket is transmitted via the supporting structure. The filter paper provided is endless and glued at both ends: It is divided into five filter chambers by the V-spacers; the inner and outer surfaces of these chambers are of the same width. To provide better protection under conditions of pressure and gas mass fluctuations, the filter paper is contacted with the help of the V-spacers and glued to the latter. The lower front faces of the filter paper and the V-spacers are embedded into a special sealant on the sheet metal bottom and sheet metal lid, respectively. Except for the installed condition, the bottom opening for gas inlet is always closed so as to be largely tight with the help of the locking plate which is closed automatically by gravity.

As a protection against damage from outside a screen has been provided in front of the filter surfaces. Figures 11 and 12 show a polygonal filter element for a maximum gas flow of about 3000 m^3/h. Dimensions of this filter element: diameter 580 mm, height 900 mm. The surface of the filter paper is 35.4 m^2 for s = 2.8 mm; this gives a maximum face velocity of the gas of 2.4 cm/s. The filter element weighs about 30 kg. The maximum pressure loss of the not loaded filter element, without locking plate, is about 25 mm WC for a gas flow of 3000 m^3/h.

In order to find out whether the filter element is capable of withstanding higher temperatures, leading e.g. to tearing of the paper as a result of deformation of the frame in the axial direction, the frame was heated to 200 °C in a furnace. No damage of the filter element was observed up to this temperature level.

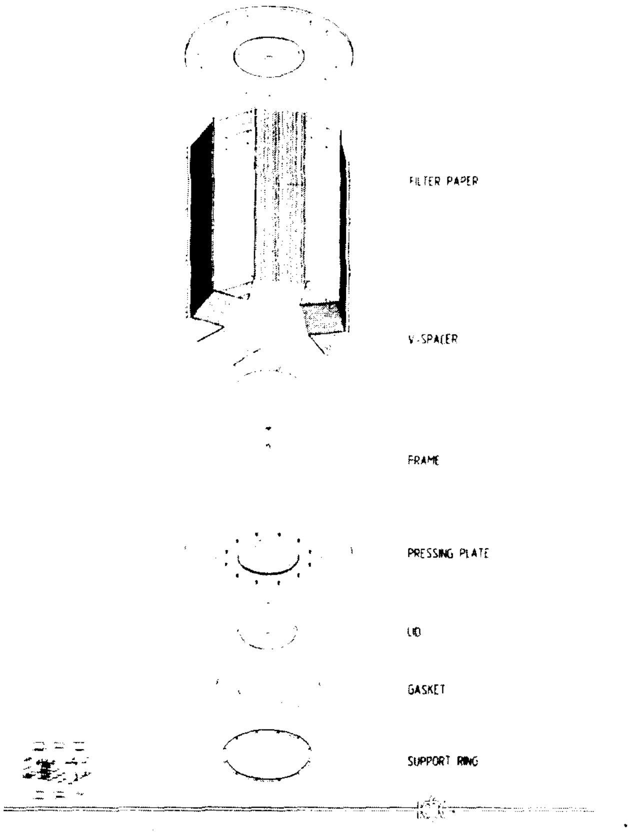


FIGURE 10
POLYGONAL FILTER ELEMENT (HEPA)

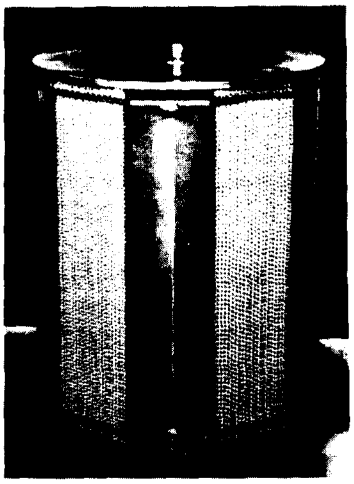


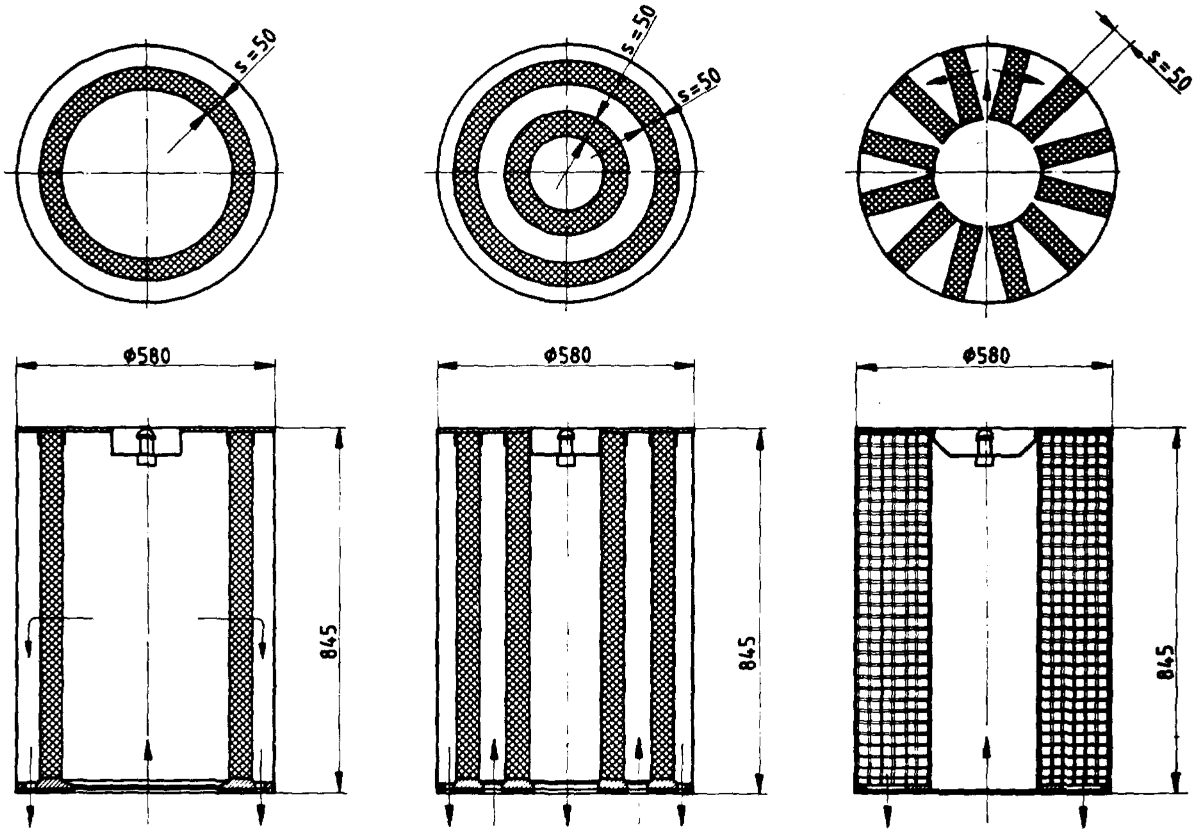
FIGURE 11
POLYGONAL FILTER ELEMENT (HEPA)
TOP PLAN VIEW

FIGURE 12
POLYGONAL FILTER ELEMENT (HEPA)
INVERTED PLAN VIEW

IV. Filter Elements for Wet Aerosol Retention

IV.1 Generalities

To filter wet aerosols from offgases having high contents of radioactive aerosols, remotely operated filter elements with fiber glass packings are used. In the Federal Republic of Germany single ring layer filters with fiber glass packing are used to filter radioactive offgases from pilot and test facilities upstream of the HEPA filters (Fig. 13). The gas face velocity is limited (about 6 -8 cm/s) in order to achieve optimum operating conditions (low pressure loss, high removal efficiency).



SINGLE ANNULAR FILTER F = 0,9 m^2

DOUBLE ANNULAR FILTER F = 1,57 m^2

STAR-SHAPED FILTER F = 1,6 m^3

F = FILTER SURFACE   s = THICKNESS OF FILTER MATERIAL

FIGURE 13
PACKED FIBER MIST ELIMINATOR
COMPARISON OF THE FILTER SURFACES

Consequently, for a given gas flow the number of the filter units required is dependent on the filter surface of the fiber glass packing available. The number of filter units should be kept as low as possible. In the single ring layer filters the filter surface depends only on the filter height provided that the diameter is defined in advance. However, if standard waste drums are used for ultimate disposal, the filter height is limited to 850 mm. In order to optimize the filter surface for a given volume of the filter element, two further solutions have been elaborated, namely a double ring layer filter and a star-shaped layer filter.

Figure 13 shows for comparison of the three alternatives the filter surfaces available for identical external sizes of the filter elements, as well as the schematic layout of the three solutions. The single ring layer filter has been tested under cold conditions in the PASSAT facility since about 1978 and under active conditions in the PAMELA facility since 1985. The decontamination factor is 3×10^3.

IV.2 Double Ring Layer Filter

The double ring layer filter, Fig. 14, is composed of the filter frame and the filter material. The filter material consists of glass fibers and is identical with the filter material used in the proven single-ring layer filter. The layer thickness is about 50 mm. The filter frame is made up of the bottom to which the gaskets of the seats have been mounted similar to the mounting in the HEPA filters, the jacket, the lid and the tubular structure connecting the lid with the bottom. The contact forces for the gaskets of the seats are transmitted to the bottom via the tubular structure. To support the fiber glass layers concentric stainless steel screens, cylindrical in shape, have been fastened between the bottom and the lid. The non-filtered air flows from the bottom into the annular space between the two fiber glass ring layers, passes them in the radial direction and leaves the filter element towards the bottom through the central opening and the external annular gap. Water nozzles have been provided below the gas inlet annular gap so that the fiber glass layers in the filter housing can be flushed.

The main disadvantage of this filter element consists in three times sealing required with respect to the filter housing: two gaskets between the inflowing and outflowing gas and one between the filter element jacket and the filter housing in order to avoid a high contamination of the inner surfaces of the filter housing. The main advantage consists in the easy mounting of the filter layers. The filter element has not yet been tested.

IV.3 Star-shaped Layer Filter

The star-shaped layer filter, Fig. 15, consists of the filter frame and the filter material. The filter material is composed of glass fibers and is identical with the filter material of the proven single-ring layer filter. The layer thickness is about 50 mm. The filter frame is made up of the bottom to which the gaskets are mounted as in the HEPA filter, the jacket, the lid and the inner tube which transmits to the bottom the forces needed to press the sealing. To support the fiber glass layers, star-shaped stainless steel screens have been installed between the jacket and the inner tube. To flush the fiber glass layers, a nozzle system has been installed at the filter element and connected to the water pipe of the filter housing when the filter element is inserted into the filter housing.

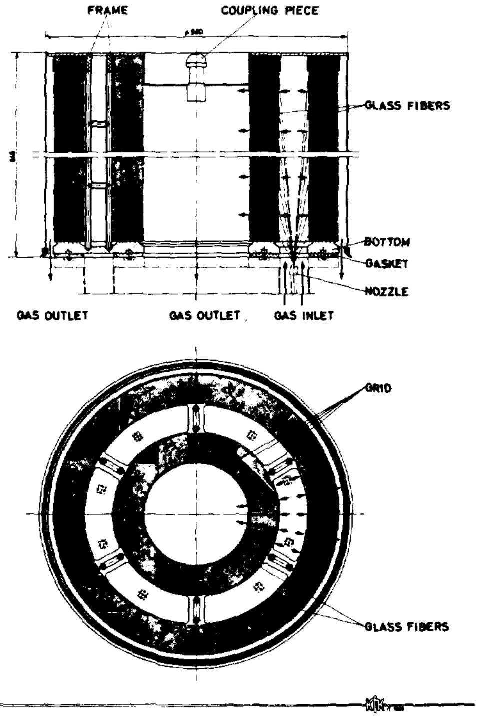


FIGURE 14
PACKED FIBER MIST ELIMINATOR

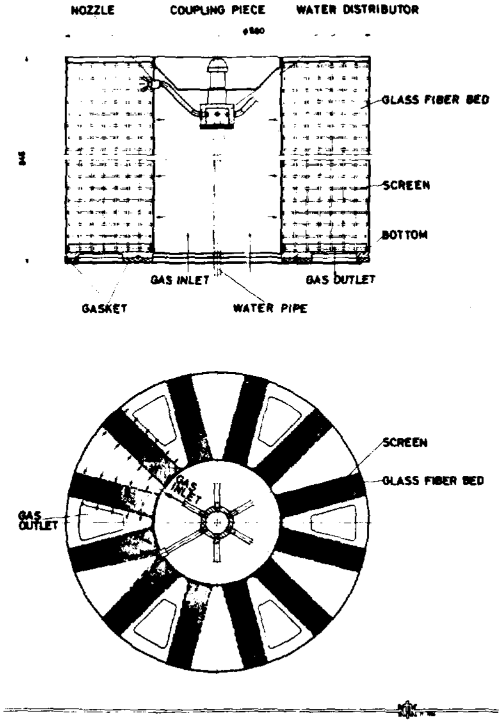


FIGURE 15
PACKED FIBER MIST ELIMINATOR

The unfiltered air flows from bottom (Fig. 15) through the centric bottom opening into the filter element and through the longitudinal gaps of the filter element to the filter layers, passes them and leaves the filter element flowing downwards through the openings in the bottom provided for this purpose. The filter element is equipped with a gasket between the inflowing and outflowing gas and a gasket between the filter element jacket and the filter housing in order to avoid excessive contamination of the inner surfaces of the filter housing.

The main disadvantages of this filter element is the costly provision of the stainless steel screen as well as the requirement of stuffing the filter material. The main advantage consists in simple sealing and force transmission to the sealings. The filter element has not yet been tested.

References

(1) Jannakos, K., Lange, W., Potgeter, G., Furrer, J., Wilhelm, "Selected solutions and design features from the design of remotely handled filters and the technology of remote filter handling. Previously operating experience with these components in the PASSAT facility," Proceedings of the 16th DOE Nuclear Air Cleaning Conference, Vol. 1, 317 (1980)

(2) Furrer, J., Wilhelm, J.G., Jannakos, K., "Aerosol and iodine removal system for the dissolver offgas in a large fuel reprocessing plant," Proceedings of the 15th DOE Nuclear Air Cleaning Conference, Vol. 1, 494 (1978)

(3) Jannakos, K. et al., Furrer J., Wilhelm, J.G., "PASSAT - eine prototypische Abgasfilteranlage. Beschreibung und bisherige Betriebserfahrungen," KfK-Nachrichten 3/79

CLOSING COMMENTS OF SESSION CO-CHAIRMAN ETTINGER

We have had a look at unique applications of filtration and new approaches to filter testing. It has been a varied discussion ranging from waste disposal to sodium aerosols to uranium enrichment facilities. There have been unique concerns identified, ranging from remote operations, to a standby automated system, to a filter system where you can reuse the filters and meet corrosion and resistance requirements. The speakers have expressed a lot of concern about humidity and temperature effects. I think the message is that people are looking at new approaches to filtration. They range from a multi-layer approach, to the use of powders or fibers for filtration, and the drawing on other technologies such as liquid filtration. Also, some of the papers are concerned with very basic physics principles as related to the application of test methods using light scattering and fluorescence spectroscopy. An important consideration is how to adopt some of the new methods to produce new filter test methods and new filtration techniques. The two go together because we need new test methods to go with new filtration procedures. We also have to learn how to apply some of the new methods to reduce the harmful effects of humidity and temperature. That will permit us to design better filters, the aim of two of our speakers. It has been a very interesting and informative session.

SESSION 5

STANDARDS AND REGULATIONS

TUESDAY: August 19, 1986
CHAIRMAN: W.H. Miller, Jr.
Sargent & Lundy Co.

REPORT ON THE 1985 CONAGT FIELD TESTING WORKSHOPS
D.M. Hubbard

HIGHLIGHTS OF PROPOSED CHANGES TO ANSI/ASME N509-80
S.C. Ornberg

HIGHLIGHTS OF PROPOSED CHANGES TO ANSI/ASME N510-1980 TESTING OF NUCLEAR AIR-CLEANING SYSTEMS
D.D. Whitney

AN ASME CODE FOR FIELD TESTING OF NUCLEAR SAFETY RELATED GAS PROCESSING SYSTEMS
S.A. Hobart, F.J. Cannito, L.B. Nesbitt, D.P. Werkeiser, J.M. Pleva, R. Reda

REVIEW OF CURRENT HALIDE AND AEROSOL LEAK TESTING METHODS OF NUCLEAR AIR TREATMENT SYSTEMS, COMPONENTS AND BANKS
J.W. Jacox

CONFLICTS AND MISAPPLICATIONS OF ANSI N509, N510 USNRC REGULATORY GUIDE 1.52 AND 1.140
J.L. Kovach

CONTROVERSIAL ISSUES WITH AIR CLEANING AT NUCLEAR POWER STATIONS
R.R. Bellamy

REACTIVE INSPECTION RESPONSE OF NRC REGION III TO POTENTIAL TECHNICAL DEFICIENCIES IDENTIFIED IN RECENT NUCLEAR AIR CLEANING CONFERENCE PAPERS
C.F. Gill

OPENING COMMENTS OF SESSION CHAIRMAN MILLER

Since the last Air Cleaning Conference in Baltimore two years ago, we in the nuclear air cleaning business have been experiencing an upsurge of activity, energy, and interest in regulations, codes, and standards. This has come at a time when the nuclear power industry is generally not vigorous, as far as new design goes. The prediction that we would have trouble keeping people involved in standards and regulations has not come to pass. Construction of our third generation of commercial nuclear power plants has progressed through the testing phase of nuclear air cleaning systems, and we now know that we only need to keep the communication lines open to tap the wealth of knowledge that exists in the hands of the users. This session fully opens the communication lines between the standards and regulation writers and the users.

In this session we will hear eight papers, seven by active ASME CONAGT Committee Members. I have to make a statement that will keep four of our presenters from having to make a disclaimer. ASME has a disclaimer that they would like me to read. I will paraphrase it. What it basically says is that this morning you are going to hear papers that concern proposed changes in standards and codes currently under consideration. Because we are involved in an ANSI consensus code process, we will be going through a number of different stages of review. What will be said here is our current intention as a committee, but the final code and standard may be a little different because of the approval processes.

Our first paper concerns workshops that CONAGT held in 1985 to obtain feedback from standards users. Three papers concern CONAGT's efforts to incorporate this effort into our ANSI/ASME N-509,N-510, and Gas Processing standards and codes. Two papers presented by professional nuclear air cleaning field testers will give us a testers' perspective on various issues related to field test methods and misapplications of some of these standards. Lastly, two papers will give us an NRC regional staff perspective on the issues and the potential deficiencies in the literature and various system applications. This is a power packed lineup of practical papers.

REPORT ON THE 1985 **CONAGT** FIELD TESTING WORKSHOPS

Dean M. Hubbard
Duke Power Company
Charlotte, North Carolina

Abstract

In 1985 the ASME Committee on Nuclear Air and Gas Treatment Equipment (CONAGT) held two workshops on the field testing of nuclear air cleaning systems. The primary goal of the workshops was to provide the users of ANSI/ASME N510 "Testing of Nuclear Air-Cleaning Systems" and ANSI/ASME N509 "Nuclear Power Plant Air Cleaning Units and Components" an opportunity to provide input to the standards writers on changes needed to reduce implementation problems. The result was a forum to discuss the design and testing of nuclear air-cleaning systems that drew a combined attendance of over 100 professionals. CONAGT received valuable input for the revision of ANSI/ASME N510 and N509 as well as input that has been used for the development of other CONAGT codes and standards.

An important by-product of the workshops was the opportunity for the attendees to meet with a cross section of their industry peers to discuss problems and solutions. Previously few opportunities existed for this type of needed communication in the area of nuclear filtration. CONAGT took a bold step in initiating this practical type of interchange in an effort to better understand the needs of the industry it was organized to serve. CONAGT plans to continue this type of direct industry contact and is recommending that the Board on Nuclear Codes and Standards encourage other nuclear code and standard writing committees to follow suit.

The workshops were organized into multiple round table discussions on the following topics: ASMI/ANSE N510 & 509 Revisions, Source Term Implications, Regulatory Concerns, Field Testing of Nuclear Air Cleaning Systems, Operation and Maintenance of Nuclear Air Cleaning Systems and Field Testing Personnel Training. Significant information from each of the round table summaries is discussed with an emphasis on the practical aspects of designing and testing nuclear air-cleaning systems.

I. Introduction

In 1985 ASME's Committee on Nuclear Air and Gas Treatment (CONAGT) established a first in code and standard development. Sensing the changing needs of the utility industry it serves, CONAGT organized two open workshops on the field testing of Nuclear Air Cleaning (NAC) systems. These workshops provided participants an opportunity for face to face dialog with the authors of the codes and standards. The workshops were held on the east and west coast to encourage maximum industry participation at a reasonable cost. Combined attendance totaled 112 people representing design engineers, manufacturers, regulatory agencies, consultants, construction firms, and end users of NAC systems.

II. Workshop History

CONAGT was chartered by ASME in 1975 to "develop, review, maintain, and coordinate codes and standards for design, fabrication, installation, testing and inspection of equipment for nuclear power plant air and gas treatment systems." Before the chartering of CONAGT, the American National Standards Committee N45 had largely completed two standards on the design and testing of air and gas cleaning systems for nuclear facilities. These two standards became the responsibility of CONAGT and were designated N509-1976 "Nuclear Power Plant Air Cleaning Units and Components"(1) and N510-1975 "Testing of Nuclear Air Cleaning Systems"(2). N509 and N510 have since undergone one maintenance revision in 1980 and were due to be reaffirmed or revised in 1985 according to ANSI procedures. CONAGT's original plans were to replace N509 and N510 with the CONAGT code AG-1. The delaying of the complete issue of AG-1 necessitated consideration of the maintenance revisions.

When the possibility for a second revision became known, users in the industry showed considerable interest in contributing. The revision was an opportunity to use the previously unavailable operating experience with N509 systems to refine the two standards. In response to the interest expressed by the industry, the Main Committee of CONAGT proposed that a workshop be used as the mechanism for input. The idea was so well received that two workshops were held yielding benefits well beyond the original intent.

The first workshop was held on the west coast in Los Angeles, California on February 16, 1985. The second was held on the east coast in Charlotte, North Carolina on April 22 & 23, 1985. Both workshops centered around six main topics pertaining to the design, operation, testing, and maintenance of NAC systems. Each topic had one or two moderators that would give an introductory statement on the topic and then host a round table discussion. Round tables were attended by a cross section of the industry where participants had an opportunity to ask questions and provide input. At the end of the round table sessions summaries were given by the table moderators. The conclusion of the workshop was followed by a comprehensive summary that was mailed to all the attendees. As planned this summary became a significant source of input for the maintenance revisions of N510 and N509 which are expected to be issued in 1986 or early 1987.

III. Round Table Results

Some of the more significant information from each of the round table topics were condensed and are listed by round table title in the subheadings below.

ANSI/ASME N509 & N510 Maintenance Revisions

Both of these standards are currently being revised to incorporate new information, provide clarification, and resolve conflicts between N509 & N510. A significant number of comments were received ranging from changing definitions to modifying the basis of the standards. Six of the comments are described below.

Requirement for Downstream HEPA Bank. The need for a second bank of HEPA filters downstream of the carbon filters was questioned. A second HEPA bank was originally intended to catch carbon fines from the carbon adsorber bank and would provide the additional benefit of backup for the primary HEPA bank. The argument was presented that HEPA filters are unnecessary overkill for filtering carbon fines. Carbon fines are typically in the 5 to 10 micrometer range and could be easily be captured by high efficiency dust filters with significantly less pressure drop.

It was also noted that carbon fines have not become the problem they were predicted to be. Activated carbon meeting the specifications outlined in N509 does not generate significant fines. Small quantities of fines result from loading the carbon into the filter housings but are quickly removed by the air flow in the first few minutes of operation.

Fire, Painting, and Chemical Release. Incorporate guidance on the effect of fire (smoke), painting, and chemical release on activated impregnated carbon. Regulatory Guide 1.52 Rev. 2(3) requires in-place testing if any of the above releases occur in any ventilation zone communicating with the filter system. The question remains as to how much fire, painting, or chemical release is required to affect the carbon (e.g. Does smoking a cigarette constitute a fire?).

Through discussion it became clear that most of the participants were convinced that more research needs to be done to quantify the effects of contaminants on carbon. The results of that research and the present knowledge should be incorporated into N510.

Ideal Versus Actual N509 Design. A great deal of concern was expressed over whether the standard for testing (N510) should address only "ideal" N509 design NAC systems; or should guidance be provided for testing systems that don't meet the performance capabilities of "ideal" N509 design systems. It was generally agreed that a majority of NAC systems now in use are not "ideal" N509 systems. Users of N510 desire practical guidance to enable them to perform the necessary testing. CONAGT will attempt to provide this kind of practical guidance in the appendix of N510.

Expand Scope of N509/N510. Attendees not affiliated with nuclear power plants expressed the desire to expand the scope of N509/N510 to include other nuclear facilities such as fuel processing. CONAGT members responded to this suggestion by stating that expanding the scope would require CONAGT charter revisions. There is considerable resistance to expanding the charter to avoid further complicating these standards. It was suggested that other facilities could use N509/N510 to develop their own guidelines.

Clamping/Mounting Requirements. Clamping/mounting devices are not addressed in the current revisions of N509/N510. Clamping/mounting devices need to be added due to the potential of component test failure due to poor design. Inadequate clamping has been a source of failure for in-place leak testing when the clamping devices are unable to provide the proper gasket compression on the filter to frame seal.

Laboratory Testing of Carbon. Used carbon laboratory testing criteria are specified in Regulatory Guide 1.52 Rev. 2 but are not covered in N509. It was recommended that N509 should either directly refer to the Regulatory Guide or state that the requirements are plant specific and are established between the utility and the regulator.

New Source Term Implications

Source Term was included in the workshop because of the potential effects that new source terms could have on the assumptions for radioactive releases and therefore the type and efficiency of filtration required.

Source Term is defined as the amount and type of radioactive materials that would be available for escape to the environment from a reactor which has undergone a severe reactor accident. Current assumptions on source terms used for light water reactor siting evaluations were established during the early sixties. These assumptions were developed using calculations and data from graphite moderated reactors. The effects of reactor design, fission product chemistry, and aerosol behavior were not considered. The key factor that was not considered was the role of water or steam in limiting the release of fission products, especially iodine. Accident records and destructive tests in light water reactors to date show that no more than 0.5% of the available iodine has ever been released to the atmosphere versus the 50% release assumptions used for siting evaluations.

The impact of these finding on nuclear air filtration could mean the elimination of carbon adsorbers on some air filter systems. However, source term reevaluation work is far from complete. Work remains to complete a thorough review of all accident mechanisms. Post accident release of organic iodine was cited as one example where additional work is required.

In some areas the current source term studies have been sufficient to justify regulatory relief. In one case of control room habitability retrofit, relief given for iodine thyroid dose resulted in not having to install two new engineered safety filter trains at an approximate cost savings of $900,000.

It was suggested by one attendee that if carbon adsorbers are removed from a system, the space could be used for an additional HEPA bank if needed.

Regulatory Concerns

Temporary Sealants. The regulatory topic raised by the greatest number of attendees concerned the use of temporary sealants in ductwork on ESF (engineered safety feature) and non-ESF systems. Due to the widespread use of temporary sealants, it was suggested that some of the sealants should be qualified as permanent. No general guidance has been developed on the use of such materials. The NRC is reviewing the use of temporary sealants on a case-by-case basis, taking into account the safety significance of increased leakage resulting from the deterioration of the sealant. It was suggested that periodic leak testing of the ductwork, replacing sealant with welded joints, or partial replacement of the ductwork, are possible options where safety problems are identified.

Isolation/By-pass Damper. Leakage testing of isolation dampers and by-pass dampers seems to be resulting in higher leakage rates than had been assumed. This has been particularly true for dampers associated with high volume recirculating systems where the fan can induce a high differential pressure across the damper. One solution suggested to reduce the high leakage rate is to replace parallel blade type dampers with bubble tight dampers.

Fire Damper Testing. Fire dampers are also presenting problems with testing. The problem occurs when trying to test the damper(s) with the system in operation. This is especially true where sequential testing results in increased velocity or pressure drop across the damper. It was noted that EPRI has been asked to develop guidance on the subject.

Field Testing of Nuclear Air Cleaning Systems

QA Review of Testing. The major item of concern centered around Quality Assurance (QA) personnel reviewing the in-place testing based on the requirements of N510. A majority of NAC systems being tested cannot meet the guidelines of N510 to the letter and are therefore being questioned by QA. The problem has resulted in part because a significant number of systems were designed before N509/N510 were issued. A NAC system not designed to N509 cannot be expected to meet the testing requirements of N510. This is stated in N510 in Section 1.2, "Limitations of the Standard", but still presents problems.

The matter is further complicated by NAC systems that were specified to N509 that still cannot be rigorously tested to N510. Part of this problem is that N510 is not being used as the basis for developing test programs but is being applied in the absolute sense. An example is the visual inspection check list that has items for all types of NAC systems and cannot be applied in its entirety to any one system. It was noted that QA check list or procedures are often too detailed and rigid to allow proper technical judgement to be exercised.

Carbon Aging and Storage. Early purchases of carbon made during plant construction have not been used until five to eight years later. There is no formal guidance on how long carbon can be stored and still meet testing criteria. When should carbon be questioned and therefore retested before be used? In general, the method being used is to retest if there is any doubt. Five years has been suggested as the maximum storage time before retest if the carbon is stored under ideal conditions. Storage conditions will greatly affect carbon life. Most agreed that guidelines are needed and should be based on additional testing of aging under "normal" storage conditions.

Instrument Sensitivity to R-11. Limiting R-11 concentrations to the sensitivity of the detecting instrument is necessary to achieve accurate testing results. It was suggested the upper and lower R-11 challenge concentration limits be established for the specific instrument in use. Instrument linearity or correction for nonlinearity should be taken into account in the evaluation. The limits should also reflect consideration for unstable field environments.

Carbon Adsorber Fire Protection. Fire protection for carbon beds continues to trouble the owners of NAC systems. There have been over 180 reported cases of accidental initiation of fire protection systems. Testing has shown that water spray is not effective in extinguishing a burning carbon bed unless the bed is completely flooded for a period of time. A credible source of ignition has yet to be identified. The only case of a bed fire in a power plant NAC system is where the heaters used to reduce relative humidity were placed directly in contact with the carbon. Defeat of the heater temperature controls led to the ignition of the carbon. Heaters of this type are no longer used.

Wetting the carbon presents numerous problems. Due to the impregnates on the carbon, acids form with the addition of water that are corrosive to stainless steel. Damage to the filter screen will occur unless the carbon is removed quickly and the filters washed. Wet carbon is extremely difficult to remove from any type filter. Disposal of the wet carbon is difficult and expensive. The entire process of putting the NAC system back into service is time consuming and could force the unit off-line due to plant technical specification limitations.

The resistance to removal of water spray fire protection of carbon beds comes primarily from the insurance industry. Insurance concerns are based on experiences in the solvent recovery industry where certain keytones undergo exothermic decomposition which can lead to ignition. This is not a consideration for power plant NAC systems.

Maintenance of Nuclear Air Cleaning Systems

Carbon Sampling. Carbon sampling from Type III carbon adsorber filters can be accomplished using a grain thief (slotted tube) if carbon test canisters are inadequate or unavailable. It was suggested that the sample be taken as close as possible to the air entering side to get the most weathered sample for conservatism. One concern with this method is the possibility of leaving voids in the carbon bed. Most felt that this was highly improbable due to the nature of carbon as evidenced by the settling that occurs when air flow is established through the carbon. To avoid any opportunity of bypass at the top of the filter it was suggested that carbon be added at the top of the bed (above the screen) after removing the sample. The air flow distribution test should be used to locate a representative carbon sample location.

Carbon Loading. Loading carbon in Type III carbon adsorbers has presented several problems. Loading devices provided by manufacturers are often ineffective or too slow. An alternative being used is to pour the carbon in as rapidly as possible and use a pneumatic vibrator to settle the carbon to its maximum density. The carbon should be monitored for excessive settling that could lead to filter bypass. Other carbon loading suggestions include temporarily taping the slots between the bed sections to contain the fines during carbon loading (remove tape after loading), and use a 2 inch roll type filter downstream of the carbon beds during initial start-up to catch carbon fines.

Field Testing Personnel Training

Training opportunities in nuclear air filtration are limited to formal training and apprentice type training. Harvard University and Duke Power Company currently offer the only formal training courses. Some testing consultants are starting to offer on-site training for in-place testing.

A major concern expressed by some individuals is the lack of management priority in the area of NAC systems. Adequate staffing and training of personnel in the testing and maintenance of NAC systems are required to assure that the systems will perform as designed when needed. Even if testing or maintenance services are contracted, supervision needs to be knowledgeable of the testing and maintenance required. Ultimately the licensee is responsible for their NAC system in the event of failure. Ideally all personnel that have contact with NAC systems should have training relevant to their responsibility for the systems.

High turnover rates of testing personnel is a major concern. Trained and motivated personnel tend to move up in the organization making it difficult to keep enough trained and experienced personnel available. No solutions were proposed for this problem which is common in most other technical areas.

Training is one aspect of employee qualification. Experience is also necessary to assure qualification of field testing personnel. It was noted that neither training or experience guarantees that an individual is qualified. A case was discussed where a person was trained and had in excess of seven years experience but was still doing unsatisfactory work. The bottom line is that owner supervision is responsible for the quality of testing and maintenance done on NAC systems to assure their operability.

IV. By-products of the Workshops

Even though the attendees were enthusiastic about the opportunity to provide input into the revisions of N510 and N509, it was obvious that the participants would use the time together for other reasons as well. Surveys were taken after both workshops. In the survey category of "expectations fulfilled", a majority of the participants cited the opportunity for information exchange on their particular concerns. Their concerns included such areas as: testing difficulties, personnel qualification, management support, and regulatory expectations. The workshops were an excellent opportunity to ask questions and share solutions where few opportunities for such exchange exist. Most participants felt they had gained a better perspective of the NAC industry and understanding of the other NAC industry members.

V. Conclusion

Workshops are not a new mechanism for information exchange. What is new is that the CONAGT workshops were used to encourage input into code and standard development. The results of these two workshops should ensure that they will continue in some form for CONAGT activities. CONAGT is also discussing the feasibility of holding NAC short courses under ASME auspices. The people of CONAGT recognize the need to know and address the practical needs of the industry they were chartered to serve. CONAGT is recommending to the Board on Nuclear Codes and Standards that other nuclear code and standard writing committees explore the workshop method for communicating with the industries they serve.

References

1. American National Standard, "Nuclear power plant air cleaning units and components," ANSI/ASME N509-1976, The American Society of Mechanical Engineers (1976).

2. American National Standard, "Testing of nuclear air-cleaning systems," ANSI/ASME N510-1975, The American Society of Mechanical Engineers (1975).

3. USNRC Regulatory Guide 1.52 (Rev. 2), "Design, testing and maintenance criteria for atmoshpere cleanup system air filtration and adsorption untis of light-water-cooled nuclear power plants," (March 1978).

DISCUSSION

HYDER: Mr. Hubbard stated that he felt more work was required in N510 on the effects of organic materials and other insults to carbon beds, and his paper calls for more research on the effect of fumes, solvents, etc. on carbon. A study of the effect of organic materials on carbon performance was made at Savannah River to establish criteria for carbon replacement there. These results should be published within the next year. We did establish requirements for our own carbon beds based on historical studies and analysis of our own carbons.

HIGHLIGHTS OF PROPOSED CHANGES
TO
ANSI/ASME N509-80

S. C. Ornberg, Vice Chairman
Subcommittee on General Support Services, ASME CONAGT
Sargent & Lundy
Chicago, Illinois

Abstract

ANSI/ASME N509 and N510 are undergoing revision based on the results of a required 5-year review and comments received from users of the standards at workshops and through inquiries.(1)(2) This paper discusses the highlights of the significant revisions to ANSI/ASME N509 and explains the reasons for the changes.

I. Introduction

Standards which are approved by the American National Standards Institute (ANSI) are required to undergo a review every 5 years to determine whether they should be reaffirmed as published, withdrawn, or revised.

In 1984, the ASME Committee on Nuclear Air and Gas Treatment (CONAGT) considered whether to perform maintenance revisions of ANSI N509 and N510. CONAGT's original intent was to supersede N509 with the Code on Nuclear Air and Gas Treatment (ANSI/ASME AG-1).(3) However, due to delay in preparation and approval of some code sections, it was apparent that N509 could not be superseded in the near future. In addition, CONAGT committee members noted that many utility Technical Specifications referenced N509-80 and it was apparent that N509 needed clarification based on inquiries received from users.

In order to obtain more industry user input for a possible N509 revision (as well as an N510 revision), CONAGT decided to hold two open workshops in 1985. (For more information on these workshops, refer to D. Hubbard's paper, "Report on the 1985 CONAGT Field Testing Workshops," from these proceedings.) The conclusion reached by CONAGT after these workshops was that N509 needed revision to incorporate comments received from CONAGT members, workshops participants, and technical inquiries; and that N509 needed interface with N510 revisions also presently underway. I was selected by CONAGT to coordinate the resolution of comments and present the revisions at the next DOE/NRC Air Cleaning Conference. These comments have now been incorporated into an N509 draft revision, and an initial CONAGT Main Committee ballot on these changes, as of the writing of this paper, is currently in progress.

At this point, it is necessary to emphasize that these revisions are not yet approved by ASME CONAGT, the Board of Nuclear Codes and Standards, or ANSI. The review of this material is intended as a constructive public service in the hope of spurring interest and comments to improve the final version, and to demonstrate that the consensus standards process is responsive to industry needs. The results of the ASME review process will be made available to the public for comment prior to issuance. It is anticipated that the resolution of Main Committee comments and the ASME Board of Nuclear Codes and Standards (BNCS) approval process will not be completed until the end of 1986. It is expected that N510 revisions will be available for public comment in 1987. (For additional information on the N510 revisions presently being contemplated, refer to D. Whitney's paper, "Highlights of Proposed Changes to ANSI/ASME N510," from these proceedings.)

It is hoped that interested parties will review these revisions and provide constructive comments. The explanation of the revisions contained in this paper will help reviewers to better understand the basis for the changes. Those interested in receiving the revised standard when available for public comment should contact the Secretary for the Committee on Nuclear Air and Gas Treatment through the American Society of Mechanical Engineers in New York.

II. Discussion of Proposed Changes

The following discusses the significant proposed changes and the basis for the changes.

Definitions (Section 3*)

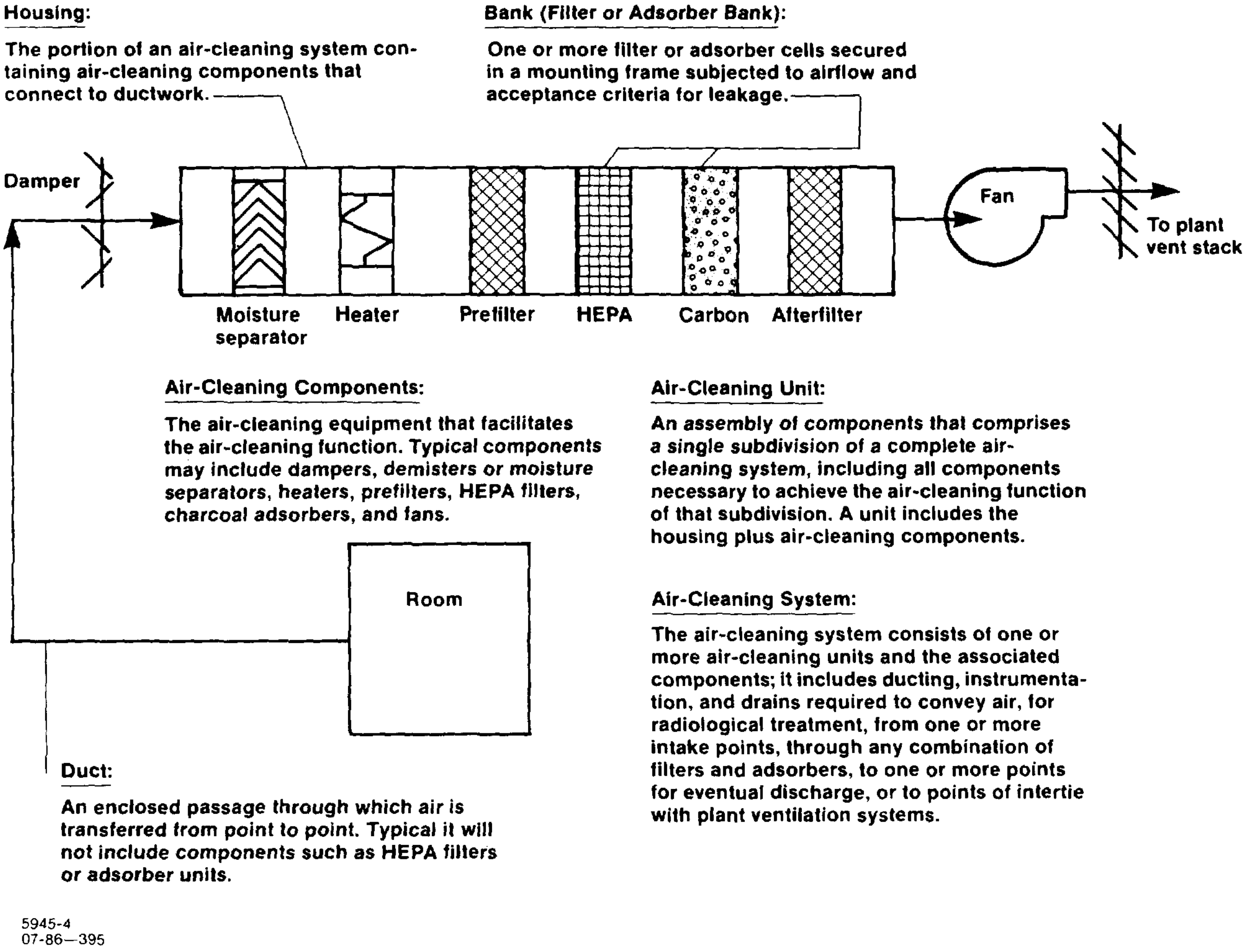
Definitions are often initially overlooked by readers of a standard (and sometimes the preparers as well) until a question of interpretation occurs. Then, definitions are reviewed very carefully, because often they hold the key to the preparer's intent.

System, Unit, Bank. Over the last 5 years, it became apparent that several clarifications were required and several new definitions were needed to better explain the intent of the standard. For example, there has been much discussion on what is an air-cleaning system, what is an air-cleaning unit, and what is a bank of filters. In some cases, where "system" was used, "unit" or "bank" was meant. Definitions are being added or revised to clarify this intent. Figure 1, "Air-cleaning system, unit, component, and filter banks," depicts these terms.

Bypass. Bypass can have many meanings, depending on the situation. For N509 and N510, when bypass is used, it refers to "a path by which contaminated air can escape treatment by the installed HEPA and/or adsorber banks. Examples are leaks in filters, filter frames, defective or inefficient bypass dampers, unintended passage through adjacent plenums or penetrations such as electrical conduits, pipes, or floor drains." This definition is extremely important to proper interpretation of testing

---

*of ANSI/ASME N509-1980



Housing:

The portion of an air-cleaning system containing air-cleaning components that connect to ductwork.

Bank (Filter or Adsorber Bank):

One or more filter or adsorber cells secured in a mounting frame subjected to airflow and acceptance criteria for leakage.

Air-Cleaning Components:

The air-cleaning equipment that facilitates the air-cleaning function. Typical components may include dampers, demisters or moisture separators, heaters, prefilters, HEPA filters, charcoal adsorbers, and fans.

Air-Cleaning Unit:

An assembly of components that comprises a single subdivision of a complete air-cleaning system, including all components necessary to achieve the air-cleaning function of that subdivision. A unit includes the housing plus air-cleaning components.

Air-Cleaning System:

The air-cleaning system consists of one or more air-cleaning units and the associated components; it includes ducting, instrumentation, and drains required to convey air, for radiological treatment, from one or more intake points, through any combination of filters and adsorbers, to one or more points for eventual discharge, or to points of intertie with plant ventilation systems.

Duct:

An enclosed passage through which air is transferred from point to point. Typical it will not include components such as HEPA filters or adsorber units.

Figure 1. Air-cleaning system, unit, component, and filter banks.

requirements, and therefore, the design must adequately prevent this type of bypassing.

Afterfilter (Paragraph 4.1)

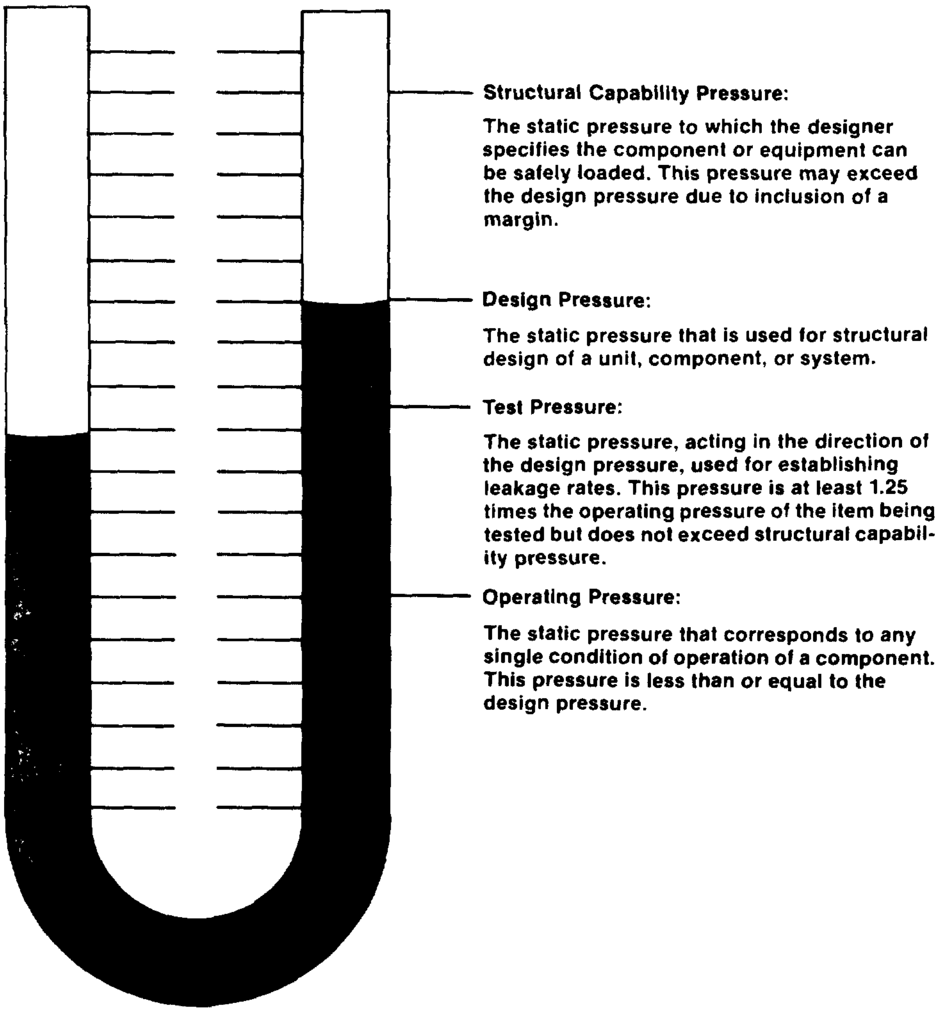
The requirement for an afterfilter was added to Section 4 on Functional Design. An afterfilter is required when adsorbers are used to retain carbon fines. Previously, the standard was silent about this requirement. However, Regulatory Guide 1.52 requires downstream HEPA filters and Regulatory Guide 1.140 suggests they be considered.(4,5) Comments received from the industry indicate that downstream HEPA filters are overly conservative, and since the filters are for retaining carbon fines, a filter with a lower efficiency (85% per ARI 680) is more than acceptable.(6)

The use of an extended media-type filter rather than a HEPA filter simplifies testing and resolves many problems of leak testing series HEPA banks. This is considered a significant, but rather subtle change.

Design Pressures (Paragraph 4.6)

The term design pressure had been used extensively throughout N509 as well as N510, to identify different conditions. Four definitions are being incorporated to properly define the intended pressure and to better interface with N510. These terms are operating pressure, design pressure, structural capability pressure, and test pressure. Figure 2, "Pressure definitions and relationship," identifies the relationship and definitions of these terms. The section on design pressures is being revised to incorporate these terms. For instance:

Units and Components Subject to Only Rated Flow. These requirements are being revised to state that components not required to withstand peak pressure may be designed for maximum operating (static) pressure rather than design pressure. This is especially important for the components at the end of a duct system that may actually experience less than 1 inch Wg, instead of a design pressure of 10 to 15 inches Wg. This also impacts duct leakage analysis and testing as discussed later.



Structural Capability Pressure:

The static pressure to which the designer specifies the component or equipment can be safely loaded. This pressure may exceed the design pressure due to inclusion of a margin.

Design Pressure:

The static pressure that is used for structural design of a unit, component, or system.

Test Pressure:

The static pressure, acting in the direction of the design pressure, used for establishing leakage rates. This pressure is at least 1.25 times the operating pressure of the item being tested but does not exceed structural capability pressure.

Operating Pressure:

The static pressure that corresponds to any single condition of operation of a component. This pressure is less than or equal to the design pressure.

5945-3
07-86—395

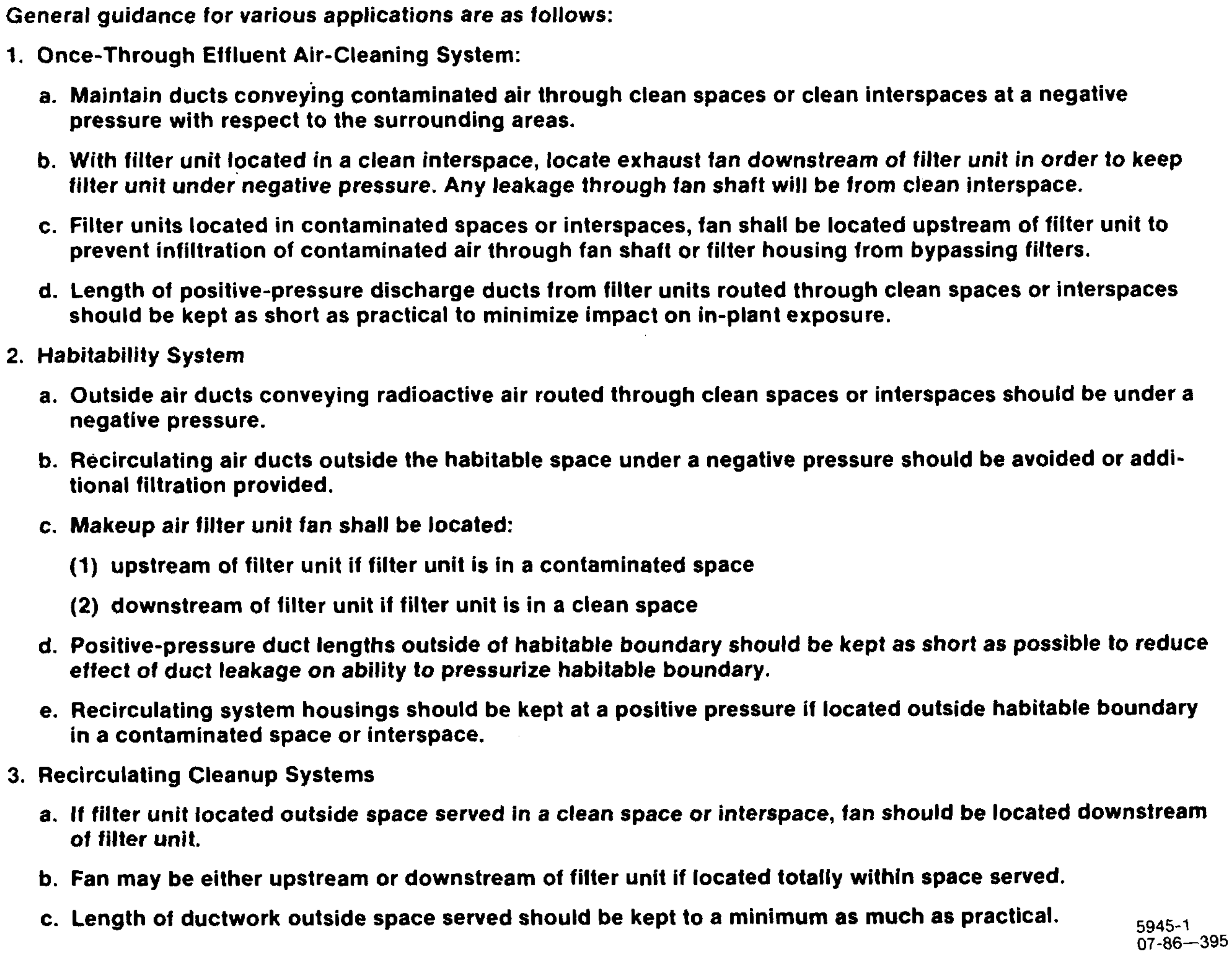
Figure 2. Pressure definitions and relationship.

Components Exposed to Fan Peak Pressure. The design pressure for components exposed to fan shutoff conditions is still the fan peak pressure. However, if the system design incorporates a provision to trip the associated fan on a static pressure increase, then the components can be designed for this trip pressure, plus margin to allow for rate of pressure rise during the instrument response time.

Location of Fans Relative to Filters (Subparagraph 4.6.4)

This section is being revised to provide general guidance for specific types of air-cleaning systems: once-through, habitability, and recirculating cleanup systems. Previously, habitability systems were not explicitly discussed.

By following these guidelines, as noted in Figure 3, "Guidelines for air-cleaning system configuration and location," the radiological impact of duct or housing leakage may be minimized and the effectiveness of air-cleaning systems improved.



General guidance for various applications are as follows:

1. **Once-Through Effluent Air-Cleaning System:**
    a. **Maintain ducts conveying contaminated air through clean spaces or clean interspaces at a negative pressure with respect to the surrounding areas.**
    b. **With filter unit located in a clean interspace, locate exhaust fan downstream of filter unit in order to keep filter unit under negative pressure. Any leakage through fan shaft will be from clean interspace.**
    c. **Filter units located in contaminated spaces or interspaces, fan shall be located upstream of filter unit to prevent infiltration of contaminated air through fan shaft or filter housing from bypassing filters.**
    d. **Length of positive-pressure discharge ducts from filter units routed through clean spaces or interspaces should be kept as short as practical to minimize impact on in-plant exposure.**
2. **Habitability System**
    a. **Outside air ducts conveying radioactive air routed through clean spaces or interspaces should be under a negative pressure.**
    b. **Recirculating air ducts outside the habitable space under a negative pressure should be avoided or additional filtration provided.**
    c. **Makeup air filter unit fan shall be located:**
        (1) **upstream of filter unit if filter unit is in a contaminated space**
        (2) **downstream of filter unit if filter unit is in a clean space**
    d. **Positive-pressure duct lengths outside of habitable boundary should be kept as short as possible to reduce effect of duct leakage on ability to pressurize habitable boundary.**
    e. **Recirculating system housings should be kept at a positive pressure if located outside habitable boundary in a contaminated space or interspace.**
3. **Recirculating Cleanup Systems**
    a. **If filter unit located outside space served in a clean space or interspace, fan should be located downstream of filter unit.**
    b. **Fan may be either upstream or downstream of filter unit if located totally within space served.**
    c. **Length of ductwork outside space served should be kept to a minimum as much as practical.**

5945-1
07-86—395

Figure 3. Guidelines for air-cleaning system configuration and location.

Fire Protection (Paragraph 4.10)

The present N509-80 does not include any requirement for fire protection, except that water deluge systems are not acceptable for adsorbent cooling, "although they may be used for fire extinguishment."

Based on recent review of data on frequency and probability of impregnated activated carbon adsorber fires versus the number of spurious activation of automatic or manual fire protection systems, there is a greater need to protect the carbon from the fire protection system rather than from a fire.(7) (A panel discussion on this topic is scheduled for this conference.)

While it is recognized that more data is needed in this area, N509 is being revised to provide for:

- Deluge nozzles inside the housing but not hard piped to a fire protection system. Instead, two isolation valves external to the housing should be provided to allow the plant fire brigade to connect manually to the plant fire protection system if they confirm a carbon adsorber fire exists. Automatic deluge systems are not recommended.
- Automatic shutdown of the fan and isolation of the filter unit based on a first-stage alarm from a two-stage detection system.
- Detection systems chosen based on ability to respond quickly to fire conditions (i.e., product-of-combustion detectors may be faster responding than temperature detectors). In addition, detection system response should be demonstrated.

Monitoring of Operational Variables (Paragraph 4.8)

Several changes are being proposed to the requirements for instrumentation on engineered safety feature (ESF) and non-ESF air-cleaning units.

Recording Function. The requirement for recording inlet flow rates for ESF and non-ESF units is being deleted as is the recording of the ESF upstream HEPA filter pressure drop on the remote manual control panel. These changes are being made based on feedback reporting that the rate of change is so gradual (1 inch Wg over a few years) that it is imperceptible. Recording of filter pressure drops requires a great deal of space in the main control room with little benefit, especially for those plants with many ESF filter systems. Alternately, plants usually keep logs of filter pressure drops for trending and predicting changeout time based on weekly, biweekly, or monthly surveillance. Furthermore, rapid changes in flow or pressure are monitored and alarmed if acceptance values are exceeded.

Unit Pressure Drop. The present N509 standard calls for a high pressure drop alarm in the main control room for each ESF unit. This is now being noted as an optional alarm if each individual air-cleaning unit component, whose pressure drop is subject to change over time, is alarmed in the main control room.

Deluge Valves. The requirement that handswitch and position indicators are only required for power-actuated valves is being added. Manual valves with local position indication at the valve are also acceptable (in fact, preferred).

In-Containment Units. A note is being added to clarify that local controls for non-ESF air-cleaning units located inside the containment should be located outside the containment.

Testability - Test Canisters

The present standard only requires a minimum of six test canisters. The proposed revision changes this to one test canister per 2000 cfm or a minimum of six test canisters dispersed througout the adsorber bank.

Pressure Boundary Leakage (Paragraph 4.12 and Appendix B)

Philosophy. Determination of allowable pressure boundary leakage is undergoing perhaps the most philosophical change from the previous revision. The revised philosophy is a direct outcome of comments received by the N509-80 users and experience with how N509-80 was being interpreted.

N509-80 contained the requirement that pressure boundary leakage be determined based on the minimum allowable leakage of health physics, air-cleaning effectiveness, and duct/housing quality. Consider a specific system design that can withstand 20-cfm leakage to meet health physics requirements, 10-cfm leakage for air-cleaning effectiveness, and 1-cfm leakage for duct/housing quality. By N509-80 criteria, the system would have to be designed to 1-cfm leakage. This could be overly conservative and would require more labor and different construction (or even reconstruction).

The present criteria do not recognize the difference between types of systems, nor can they handle systems with multiple functions (ventilation, air cleaning). Furthermore, they do not allow the designer any decision making authority based on the specific system design requirements.

Because of this, the pressure boundary allowable leakage limits are being revised to be based on health physics requirements and the specific system design. The maximum allowable leakage rates (based on health physics criteria) may be modified by the owner/designer to meet plant specific ALARA (as low as reasonably achievable) programs or, if the owner/designer so chooses, be based on air-cleaning effectiveness and duct/housing quality guidelines, which are contained in nonmandatory Appendix B.

The designer can now choose the appropriate criteria based on the health physics design requirements, the specific system design/function, and plant requirements. The allowable leakage will not exceed the value to meet the health physics requirement, which is the base criterion, just as temperature is one of the base criteria for equipment qualification. This is the true measure of the air-cleaning system's flow effectiveness. An identified margin may then be applied, if so desired.

System Pressure. Another change is that allowable leakage is to be determined at the maximum system operating pressure, rather than design pressure. The designer must also specify the test pressure to be used to perform N510 leak testing.

Examples. Appendix B is being expanded to include suggested procedures and more detailed examples of how to determine allowable leakage to meet health physics criteria. Procedures to determine allowable leakage are being included, based on maintaining MPC (maximum permissible concentration) space levels for:

- ducts under a positive pressure located in a clean interspace (as defined in N509), and
- ducts under a negative pressure located outside the contaminated space they serve.

Also being included in Appendix B are procedures and detailed examples for evaluating iodine protection factor reductions for habitable areas, such as the control room, due to air-cleaning and air-conditioning duct leakage. This material is adapted from previous Air Cleaning Conference papers on this subject.(8,9)

HEPA Filters (Paragraph 5.1)

The qualification requirements previously included in Subparagraph 4.3.1 of N509 are being incorporated in Paragraph 5.1. In addition, it was clarified that listing of a HEPA filter in the U.S. Army Qualified Products List is not required, but that the HEPA filters must be requalified to MIL-F-51068 every 5 years. Qualification documents must be retained by the manufacturer.

Adsorbers (Paragraph 5.2)

Design. The requirements for design of tray-type and deep-bed adsorbers are being revised to reference the imminent issue of ANSI/ASME AG-1, Code Sections FD and FE.(10,11) This supersedes the presently referenced standard AACC-CS 8T.(12)

Adsorbent. New and unused adsorbent must meet the requirements in the newly issued ANSI/ASME AG-1, Code Section FF, "Adsorbent Media."(13)

Testing. The testing requirements presently contained in Table 5-1 are being split into three tables (5-1Q for Qualification Tests, 5-1B for Batch Tests, and 5-1S for Benchmark Surveillance Tests). Tables 5-1Q and 5-1B are in agreement with the previous Table 5-1, as well as with the requirements of Section FF of AG-1. The benchmark surveillance test requirements conform to Regulatory Guide 1.140 requirements, but recognize that Plant Technical Specifications override these requirements.

Unused Carbon. The testing of adsorbent is based on unused carbon (i.e., carbon that has not been placed in service). Carbon that has been originally tested then stored at site for a long period of time has commonly been referred to as "spinster" carbon. Carbon is considered unused if it has not been exposed to process air (or gas) streams. Regulatory agencies require that this carbon be retested to N509.

Reactivation/Reimpregnation. ANSI/ASME AG-1, Code Section FF further prohibits reactivation of activated carbon, but "reimpregnation of carbon that has been in service or outdated impregnated carbon is permitted, but must be qualified in accordance with this Section (FF)."

Housing Design (Paragraph 5.6)

This area is being considerably expanded due to feedback from testing personnel based on their recent experience with air-cleaning units designed and constructed to N509-76 and N509-80.

Many of the design details for smaller components such as housing doors, door lugs and latches, door seals, drains, flexible connections, and manifolds were either only superficially addressed or not addressed at all. Recent experience has shown that these components can be the biggest obstacles to achieving an installation that can be successfully tested to N510. Lack of attention to these details has resulted in systems that are either more difficult to test or unable to meet the test acceptance criteria without extensive, expensive field modifications.

The guidance given in the proposed revision to N509, while intended for new systems, should be reviewed for existing systems that have experienced difficulty in meeting N510 test requireents or acceptance criteria.

Modifications should be thoughtfully considered where economically justified. Reducing test time by 8 hours on a unit may not appear to be a great deal of time, but it adds up when applied to many units over a 40-year life or during a specific outage.

For new equipment designs, or review of existing designs, it is highly recommended that knowledgeable personnel, with N510 test experience, review the manufacturer's drawings and provide recommendations or comments on the testability of a unit. For new designs this will resolve many problems before they get to the field where the best resolutions may not be possible. Some of the specific housing design revisions are:

Housing Doors. Additional requirements are being added to improve the door seals. Door design should protect gaskets and a knife-edge seal should be provided to ensure compression. A means for adjusting compression forces and gasket compression should be included in the door design. The number of gasket joints should be minimized to limit leakage.

More attention needs to be given to door lug design. Human factors need to be considered. Many door designs in use today are not very "user friendly." It is difficult to hold all the lugs in the proper position to close the doors. Therefore, the one or two sentences on lug requirements are being expanded to include the requirements stated in Figure 4, "Housing door latching requirements."

1. **Latches must be located on all four sides of any door.**
2. **There must be six or eight latches minimum, depending on door size (one top, one bottom, four or six on sides). This assumes a standard configuration door, with height two to four times width. Doors with a square shape may need more than one latch on top and bottom.**
3. **Latches shall seal in less than 270-degree motion.**
4. **Latches shall not have more than one handle per location. That is, there shall not be a handle to position the inside clamp and a separate handle to tighten the clamping down.**
5. **Latches shall be configured such that when open, gravity will hold them in the open position.**
6. **Latches shall indicate (or have permanent indication on the door) which direction to turn to open or close. This shall be for each latch, or if all work the same, then indicated once on each door.**
7. **Ideally, latches should open and seal with only the torque that can reasonably be applied by an average person while suited up. If additional torque is required, a specific tool to provide the torque shall be supplied for each door, and so attached as to reasonably assure it will be available during the life of the plant.**
8. **Latch assemblies shall have a minimum number of components and be designed so no loose components can fall apart.**
9. **Latches shall be designed to operate with no lubricant required within the pressure boundary.**

Figure 4. Housing door latching requirements.

Additionally, requirements have been added to submit door design drawings to the purchaser, prior to fabrication, that show:

- location of hinges,
- latch details,
- viewports, and
- gasket installation.

Drains. The requirements for drains are also being expanded to resolve reported problems with recent field installations. Drain systems must be designed to meet the allowable leakage criteria. This requires that drains from individual compartments be valved, sealed, or trapped to prevent bypassing air around filters, inducing air from surrounding contaminated interspaces into the air-cleaning units, or blowing contaminated air from the filter out to a clean interspace.

The number of normally functioning drains should be kept to a minimum; that is, those that are not expected to be required for use during normal operation should be valved or capped off.

Where loop seals are provided in the functioning drains, the seals should be sized based on the highest operating static pressure the unit may experience during system startup, normal operation, or system shutdown. Automatic filling of loop seals should be considered or, if manual filling of the seals is utilized, periodic inspection and fill procedures should be written.

The drain lines should be initially tested to ensure operability. If shutoff valves or check valves are utilized, they should be initially tested for operability and leakage.

In the recent past, two reported problems have been identified with drain lines systems.(14,15) By following the above criteria, the number of reported field problems with drain systems will be reduced.

Housing Connections. No requirements were previously included in N509 for housing connections. The proposed revision includes the following requirements: Housing connections shall be designed with consideration of the air distribution uniformity requirements of N510 to preclude field testing problems. Flexible connections shall be provided between the housing and any prime movers. Flexible connections shall be rated for fan shutoff pressure and a qualified life established based on environmental and operating conditions. Allowable leakage shall be identified and flexible connections shall be tested for fabric and joint leakage following environmental qualification.

As with drains, flexible connections can be a weak link in the air-cleaning system. Degradation or failure of the flexible connection integrity can severely reduce the effectiveness of the air-cleaning system and, depending on fan location, can increase personnel radiation exposure. More attention needs to be given to this component since it, too, is usually a consumable product that requires periodic replacement.

Housing Drawings. Requirements are being added to clarify that manufacturer's housing drawings be submitted to the purchaser depicting the location and size of each door, drain, housing connection/penetration, as well as lights, switches, instrumentation, and other appurtenances. Flexible connection details, door details, and drain valve details shall also be submitted prior to fabrication.

The purchaser should have these drawings reviewed by personnel who are knowledgeable about N509 requirements and by capable filter testing personnel to ensure that housing design can be tested to N510 requirements.

Testing. Another significant addition to the housing design section is the requirements for manifold design. These requirements are being included so that housing designs incorporate provisions for manifolds early in the design phase, prior to shipment to the site. Permanent mounting of manifolds is recommended, although not mandatory, to obtain repeatability of test results. If permanent manifolds are not included, then manifolds shall be provided that can be reassembled in the field with each piece numbered, tagged, and marked for reinstallation. Additional guidance on locating and sizing manifolds is being provided in a new Appendix D. Furthermore, for manifolds located within the housing, manifold design, and location shall be qualified, in the shop prior to shipment, to N510 requirements for airflow distribution and air-aerosol mixing uniformity. Results shall be documented and submitted to the purchaser.

It is expected that early attention to housing testing, and specifically to the design and location of manifolds, will reduce the number of units delivered to the field in the future that are difficult to test adequately.

Fan Selection (Paragraph 5.7)

A clarification is being included to advise the designer that the fan selection should make provision for N510 test conditions. The system designer should identify the maximum allowable differential pressure for each filter bank plus a margin to allow for degradation between surveillance (typically 25%).

Feedback has shown that this requirement (which was in N510-80) has not been incorporated into the design and selection of the fan, and therefore, has caused problems. This clarification is being added to N509 for further emphasis and to assign the responsibility of determining these criteria to the system designer during selection of the fan.

Duct Construction (Paragraph 5.10)

The proposed revision recognizes the use of mechanical lock seams and silicone sealant for air-cleaning ducts and requires that these be qualified for the intended applications.

Testing (Subparagraph 5.10.8)

Leak Testing. Several comments were received concerning inequalities in the present N509 leak testing exemptions and requests for clarifications. Based on these comments, the following changes are proposed to clarify the intent, taking into account the effect of leakage in health physics.

Ducts exempted from quantified leak tests are:

- All ESF and non-ESF ducts serving the protected space, located within the protected space, regardless of length.
- All negative pressure ESF and non-ESF ducts passing through a clean interspace.
- All positive pressure ESF and non-ESF ducts passing through contaminated interspace having an MPC concentration within the duct (Cd), which is no greater than

1.1 times the MPC concentration in the space (Cr), where:

$$Cd \leq 1.1\ Cr. \qquad (1)$$

- Non-ESF and ESF positive-pressure duct passes through a clean interspace, and when the effective concentration within the duct is less than 5 MPC.
- Non-ESF and ESF duct under negative pressure passing through a contaminated interspace having an MPC concentration (Cr), which is no greater than 1.1 times the MPC concentration within the duct (Cd), where:

$$Cr \leq 1.1\ Cd. \qquad (2)$$

- Plant vent stacks or ducts outside plant buildings when high-level or mixed-mode release credit is not required to meet offsite dose requirements.

Balancing. The requirements for balancing of air-cleaning systems are being modified to allow the owner/designer to set the upper and lower acceptance criteria for system flow rather than the present ±10%. These limits shall be selected such that the design function of the system is maintained and equipment capabilities not exceeded. Furthermore, a statement is being proposed such that the air-cleaning system must be balanced prior to initially declaring it operable. The system need not be balanced prior to air-cleaning unit field testing if the airflow rate through the air-cleaning unit during testing is within the upper and lower acceptance values and the final balanced system flow is within the upper and lower acceptance criteria.

Fan Peak Test. This test requirement is being clarified such that it need only be performed for those housings, ducts, and components that would be subjected to fan peak pressure due to closure of dampers on suction or discharge of fan. Therefore, duct sections far from the fan that will never be subjected to fan peak pressure need not be tested to the fan peak pressure.

III. Summary

The above proposed revisions are currently being balloted by the ASME CONAGT Main Committee. After resolution of ballot comments and Main Committee approval, the revisions are balloted by the ASME Board of Nuclear Codes and Standards. Around the beginning of 1987, the final version of these revisions should be available for public comment. Interested parties should contact the ASME Secretary for information. Constructive input from users of the standard is eagerly sought.

The guidance given in these proposed revisions, while intended for new systems should be reviewed for existing systems that have experienced difficulty in meeting ANSI/ASME N510 testing requirements. In addition, because N509 and N510 are standards which are meant to be used in conjunction with each other, the proposed changes to N510, which are also following the same timetable as N509, should also be reviewed. Potential modifications should be thoughtfully considered where economically justified.

New equipment designed, constructed, and installed to these revised requirements should be better able to meet the intended testing requirements by reducing the number of field-resolved problems.

Acknowledgments

I wish to thank the participants at the 1985 CONAGT Air Cleaning Workshops for their comments and inspiration to initiate this revision. Also, I would like to thank all the many CONAGT members who contributed comments and resolutions.

References

1. ANSI, "Nuclear power plant air cleaning units and components," ANSI/ASME N509-1980, The American Society of Mechanical Engineers, (1980).
2. ANSI, "Testing of nuclear air cleaning systems," ANSI/ASME N510-1980, The American Society of Mechanical Engineers (1980).
3. ANSI, "Code on nuclear air and gas treatment," ANSI/ASME AG-1-1985, The American Society of Mechanical Engineers, (1985).
4. USNRC Regulatory Guide 1.52 (Rev. 2), "Design, testing and maintenance criteria for atmosphere cleanup system air filtration and adsorption units of light-water-cooled nuclear power plants," (March 1978).
5. USNRC Regulatory Guide 1.140 (Rev. 1) "Design, testing, maintenance criteria for normal ventilation exhaust system air filtration and adsorption units of light-water-cooled nuclear power plants," (October 1979).
6. Air Conditioning and Refrigeration Institute, "Standard for air filter equipment," ARI 680, (1980).
7. Kovach, J. L., "Differences between solvent recovery & air cleaning applications," ASME Committee on Nuclear Air & Gas Treatment Main Committee Meeting, (April 24, 1986).
8. Murphy, K. G., and Campe, K. M., "Nuclear power plant control room ventilation system design for meeting general criterion 19," Proceedings of the 13th AEC Air Cleaning Conference, (August 1974).
9. Miller, W. H., Ornberg, S. C., and Rooney, K. L., " A consistent approach to air cleaning duct design," Proceedings of the 16th DOE Nuclear Air Cleaning Conference, (October 1980).

10. ANSI, "Code on nuclear air and gas treatment," ANSI/ASME AG-1-1985, Section FD, "Type II Sorbers," The American Society of Mechanical Engineers, (1985).

11. ANSI, "Code on nuclear air and gas treatment," ANSI/ASME AG-1-1985, Section FE, "Type III Sorbers," The American Society of Mechanical Engineers, (1985).

12. Institute of Environmental Science (IES), "Standard for high-efficiency gas-phase adsorber cells," AACC-CS-8T, (1973).

13. ANSI, "Code on nuclear air and gas treatment," ANSI/ASME AG-1-1985, Section FF, "Adsorbent Media," The American Society of Mechanical Engineers, (1985).

14. IE Information Notice 85-85, "Systems interaction event resulting in reactor system safety relief valve opening following a fire protection system malfunction," United States Nuclear Regulatory Commission Office of Inspection and Enforcement, (October 31, 1985).

15. Notice of Violation and Proposed Imposition of Civil Penalty, Portland General Electric Company, Trojan Nuclear Plant, Docket No. 50-344, License No. NPF-1 EA-86-54, (May 14, 1986).

HIGHLIGHTS OF PROPOSED CHANGES
TO
ANSI/ASME N510-1980
TESTING OF NUCLEAR AIR-CLEANING SYSTEMS

D. D. Whitney
Chairman,
Subgroup on Field Testing of
Nuclear Air Treatment Systems (NATS)

Abstract

This paper will review the significant proposed changes to ANSI/ASME N510-1980, "Testing Nuclear Air-Cleaning Systems", which are currently under review by the ASME Committee on Nuclear Air and Gas Treatment (CONAGT). These changes are a result of many sources of input to ASME from users, CONAGT members, and two workshops that were held in 1985 to discuss the proposed revisions to N510-1980 and N509-1980, "Nuclear Power Plant Air Cleaning Units and Components."

The paper discusses the proposed changes and the reasons for the changes.

I. Introduction

All standards which are approved by the American National Standards Institute (ANSI) are required to undergo a review every five years to determine if the standard should be reaffirmed as presently written, withdrawn or revised.

In 1984, the ASME Committee on Nuclear Air and Gas Treatment (CONAGT) considered what course of action to take regarding ANSI/ASME N510-1980(1) as well as N509-1980(2). CONAGT's original intent was to supersede both standards with the Code On Nuclear Air and Gas Treatment (ANSI/ASME AG-1)(3). However, due to the schedule for preparation and approval of the code, and the fact that many utility Technical Specifications refer to N509 and N510, it was apparent that it was appropriate to update and revise these standards to resolve several items needing clarification.

In order to obtain industry wide input to be used in the revisions, it was decided to hold two open workshops. This was done in 1985. (For more information on these workshops, refer to D. Hubbard's paper "Report on the 1985 CONAGT Field Testing Workshops" from these proceedings.)

These workshops provided many helpful comments and questions in addition to those received from CONAGT members and formal technical inquiries relating to both N509 and N510. These comments have been incorporated into N510 and will soon be submitted to the CONAGT Main Committee for balloting.

It is necessary to emphasize that these revisions are not yet approved by CONAGT, the Board of Nuclear Codes and Standards, or ANSI. The presentation of this material is intended as a constructive public service in the hope of spurring interest and comments to improve the final version. The results of these revisions will be available for public comment prior to final issue of the revised standard. It is anticipated that resolution of Main Committee Comments and the ASME Board of Nuclear Codes and Standards (BNCS) approval process will take until the end of 1986. The public comment period will extend into 1987, so actual publication is expected in early 1987.

It is hoped that all interested parties will review these revisions and provide constructive comments. In addition, it is hoped that the explanation of the revisions contained in this paper will help those preparing comments to better understand the basis for the changes. Written comments or suggestions may be submitted to the author. It is doubtful that indivdual responses will be practical, but all received comments will be reviewed by the Subgroup.

II. Discussion of Proposed Changes

The following discussion presents a description of the major changes and their associated background.

Definitions

Field use of N510 has at times been made tedious due to inconsistencies between N509, the NATS design standard, and N510, the NATS inplace testing standard. Furthermore, some fundamental definitions were either missing, contradictory, or simply not useful. A major effort to resolve these concerns has resulted in a consistent set of definitions for the two standards. They are described in the Air Cleaning Conference paper highlighting the changes to N509 by Steve Ornberg(4). One of the most important aspects in these revisions has been to clarify the relationships between Operating Pressure, Test Pressure, Design Pressure, and Structural Capability Pressure.

Application of N510

Although N509 and N510 have been in use since 1976/1975 respectively, field experience has shown that many systems were either built before these standards existed, or the actual installations have not accomplished all that the design standards intended. Simply, there are many systems in-place which do not provide all of the N509 design features, usually with respect to testability. For these reasons, this revision of N510 is specifically structured for the comprehensive testing of N509 designed systems. At the same time, additional test techniques are provided which will assist owners/operators/test personnel in the testing and qualification of those filter systems which do not fully meet N509-1980 requirements. These additional methods and techniques are contained in non-mandatory Appendices of the N510-1986 standard and typically deal with inspection requirements and sampling methods. It is the stated purpose of N510-1986 that the mandatory body shall be rigorously applicable only to N509-1986 systems. It may be used for technical guidance for systems that do not meet the requirements of N509-1986 or N509-1976.

General Changes

A number of general changes are proposed. The proposed revision to the standard is less wordy and does not attempt to provide as much detail in the body as did the 1980 edition. The visual inspection checklist has been expanded and compared line by line with the draft of N509-1986 so all "shall" statements in N509 will be covered in the N510-1986 inspections.

There is consideration being given to require the housing leak test to be repeated at least once each operating cycle. In the 1980 (and 1975) edition this is a one-time test. Experience has shown many leak paths do develop (doors, instruments, conduits, drain lines, etc.) following startup, hense the need for the test.

Where possible, we simply refer to an existing standard that provides a needed method. We feel strongly that there is no need to duplicate an existing standard that provides the required data. (Examples are SMACNA and ASME.)

Reporting requirements are no longer specifically listed, as there were usually too many exceptions General report content guidance is given, but not an item-by-item listing.

It is the committee's intent to provide only minimum functional requirements for test instrumentation and to make the requirements as hardware independent as possible. This will allow new instrument developments to be used without inquiry or special justification.

The Airflow Capacity/Distribution Section has been revised to eliminate confusion as to which part is for Acceptance, and which for Surveillance. An exemption for Airflow Distribution and Air/Aerosol Mixing Uniformity tests for banks having only a single HEPA filter, has been added.

Effect of Inquiries

Since N510-1980 was issued there have been several formal inquiries from field users which have questioned certain of its requirements, methods, and criteria. The number and scope of these inquiries is indicative of the amount of usage N510 receives. The revision to N510 now being proposed incorporates the resolutions of the received inquiries, and has attempted to resolve the underlying cause or concern, which led to the inquiry. The following major issues began as inquiries and are discussed to provide a basis for their treatment in the proposed N510 revision.

a. Requirement for 1 cfm Penetrometer Sample Flowrate

Every effort is being made in the proposed N510 to produce a Standard and not an "operating manual". Requirements such as this are often times historically traceable to a specific piece of once available test equipment, and its requirements, or to some antiquated government test laboratory specification. In this case the "requirement" is that the sample be representative of the air being measured. As such, N510-1986 notes that the upstream and downstream sample line lengths should be similar and that sample delay time needs to be considered when taking and analyzing data. It is left to the specific test procedure to establish flow rates and other related parameters to insure this objective is achieved.

b. Audible Leak Testing

N509 allows for this technique when seeking to identify leaks prior to system qualification testing. As N510-1980 did not provide the associated procedure or method, one has been developed and included. Still, some confusion may exist on the part of quality or regulator inspectors relative to its application. Everyone concerned must recognize that the audible leak test is not quantifiable. Should it be necessary to quantify a leak, then one of the quantitative leak rate test methods in N510-1986 must be used.

c. Testing at "1.25 Times Dirty" Filter Pressure Drop

This issue has been a major problem with field use of N510-1980. With the confusion over definitions, between N510 and N509, and the fact that designers were not required to specify the appropriate field test or surveillance criteria, there are nearly as many interpretations of this requirement as there have been systems to test. Furthermore, given the relatively "flat" fan head/flow characteristics typical of most fans used in air filtration systems, 125% of design pressure drop is often simply not available. The original intent of this requirement was to demonstrate that the system would be able to supply the specified flowrate, even under "dirty filter" conditions. As proposed, N510-1986 will test each system, or filter bank, at both the "clean" and "dirty" pressure drops and demonstrate that the design flow requirements are met. The designer must specify the appropriate "dirty" value. This is important, for it is seldom appropriate to use the sum of the filter manufacturers "maximum dirty pressure drops." This is due to installations where adsorber banks are preceded by HEPA filters, or similarily, downstream or "guard" HEPA's following carbon filters. They usually do not see the particulate material necessary to develop high pressure drops, while the effects of other components in a housing (moisture separators, finned tube heaters, etc.) may significantly effect the system pressure drop and are not always taken into account.

The previous requirement to obtain a flow condition midway between "clean" and "dirty" pressure drop has likewise been eliminated as unnecessary and not meaningful.

d. Use of Flowrate Meter in lieu of Totalizing Gas Meter.

When determining leak rate from an enclosed volume, N510-1980 had specified a method which utilized a totalizing gas meter. As a result of an inquiry, it is being proposed that an acceptable alternate method is to use an appropriate flowrate meter. This is acceptable providing that the necessary changes in the technique and data collection/analysis are made. The SMACNA 15.d(5) test standard is referenced for the direct flow leak test method. Again, this is in an effort to make the standard less of a "handbook" or "procedure" specification.

Multiple Sampling Technique

Air filtration systems and equipment are often large and difficult to test, one of the major reasons for N509 and N510 to exist in the first place. Central to effective testing is the need to obtain representative challenge samples in a timely and repeatable manner. For these reasons, N509 suggested that injection/sample manifolds be installed in filter systems. To date, adoption of this recommended approach has been poorly implemented.

The multiple sampling technique is useful for qualifying air-aerosol mixing uniformity (N510-1980 Section 9, "Air-Aerosol Mixing Uniformity, Upstream of a Filter Bank"), and by comparison, allows qualification of an injection or sample manifold installed ahead of a filter bank.

The proposed revision to N509-1980 aids in this effort by providing design guidelines for these manifolds. Still, field tests are required to "qualify" any installed manifold, and for this reason the Multiple Sampling Technique is presented in the proposed N510-1986, along with a non-mandatory appendix which elaborates on the method and its limitations. Acceptance criteria are unique to each installation, and are to be developed by the system owner.

It must be remembered that Multiple Sampling is intended to show the actual distribution of challenge across a filter bank. If the challenge distribution is non-uniform for any reason, it will be observed during this test. N510 continues to utilize air velocity distribution measurements to qualify the air distribution, while the air-aerosol mixing test is done solely for the purpose of qualifying the location of injection and sample ports. Multiple Sampling is not necessarily meaningful downstream of a filter since there is no assurance that a leak will be aligned with a sample point. A well mixed downstream sample is always needed. The method is useful in situations where non-uniform upstream tracer-air mixing is a concern, or where it is needed to qualify an injection or sample manifold.

Duct-Damper Bypass and System Leakage Tests

Many filtration systems are equipped with ducts and dampers which bypass the filter banks during normal operations. To demonstrate that the filter system is performing at the desired level, it is important to insure that these dampers are such that the filter banks will perform their intended function at the required high efficiency without being bypassed due to damper leakage.

The proposed N510-1986 includes a test of the bypass dampers which is similar to the tests performed on HEPA filter banks. DOP Aerosol provides a quick method for quantifying leak tightness and can usually be used to locate leaks should the leakage exceed acceptance criteria.

Following the testing of the various HEPA filter banks, adsorber banks, and bypass dampers, a NATS "system" test is now specified. For this test, the challenge aerosol is injected far enough upstream, and sampled far enough downstream, to determine the overall effectiveness of the filter system, including ALL bypass and leakage pathways. Acceptance criteria must be carefully developed to consider the systems purpose (as established by the designer in the relevant Design Study) and the effects of any filter banks installed in series. It is this test which demonstrates that the system is performing its intended function. This has resulted in two new sections in the standard to insure there is no ambiguity as to the specific test requirements.

Surveillance Testing

Operating Nuclear Plant Technical Specifications require surveillance testing of those air filtration systems important to safety of plant, its personnel, and the public. N510 provides the test methods to accomplish this testing, although it does not provide acceptance criteria unique to each nuclear facility. That criteria is contained within the facility Technical Specifications. The methods and techniques provided in the proposed revisions to N509-1986 and N510-1986 make available to the nuclear industry the basis for upgrading systems, criteria, and operations to provide a higher standard of performance of these important and expensive systems. Facility owners should review their system designs and operations in light of these revised standards for specific enhancements and improvements within their facility. Experience has shown that these changes will not only improve the filter systems technically, but can provide cost savings too.

Summary

ANSI/ASME N510-1986 "Testing of Nuclear Air-Cleaning Systems" is in the final stages of revision. This is the second revision since original issue in 1975, and should prove to be much more useful to the nuclear industry as a result of the incorporation of the numerous "lessons-learned" during its first eleven years of field use. We equally understand it will not be perfect, and look forward to continued user input.

The coordinated updating, with ANSI/ASME N509 "Nuclear Power Plant Air Cleaning Units and Components," has resolved numerous difficulties faced during the field testing of these filter systems. At the same time, industry participation, through workshops and the inquiry process, has provided impetus and direction to the CONAGT Subcommittees working to develop the most useful and pertinent standards for assuring the quality and performance of nuclear air-cleaning systems.

References

(1) ANSI/ASME N510-1980, "Testing of Nuclear Air-Cleaning Systems," published by the American Society of Mechanical Engineers, 345 East 47th Street, New York, N.Y. 10017.

(2) ANSI/ASME N509-1980, "Nuclear Power Plant Air Cleaning Units and Components," published by the American Society of Mechanical Engineers, 345 East 47th Street, New York, N.Y. 10017.

(3) ANSI/ASME AG-1-1985, "Code On Nuclear Air and Gas Treatment," published by the American Society of Mechanical Engineers, 345 East 47th Street, New York, N.Y. 10017.

(4) "Highlights of Proposed Changes to ANSI/ASME N509-1980" by S.C. Ornberg of Sargent & Lundy, Chicago, Illinois. Paper presented at 19th DOE Air Cleaning Conference, August 1986, Seattle, Washington.

(5) Sheet Metal and Air-Conditioning Contractor's National Association, Inc. (SMACNA)

AN ASME CODE FOR FIELD TESTING OF NUCLEAR SAFETY-RELATED GAS PROCESSING SYSTEMS

S. A. Hobart, F. J. Cannito, L. B. Nesbitt
D. P. Werkheiser, J. M. Pleva, R. Reda*
ASME CONAGT Subgroup on Field Testing of Gas Processing Systems

Abstract

"Field Testing of Gas Processing Equipment [Nuclear Safety-Related]" is a new test code applicable to equipment designed for processing containment atmospheres in a postaccident condition. This paper provides a discussion of the code section and a perspective of its relationship to the other Nuclear Air and Gas Treatment Code (AG-1) sections. The need for a unified field test code is presented, along with a comparison of existing codes, standards, and regulations. Particular attention is given to providing the potential user with an understanding of the structure of this code section and requirements unique to particular equipment types.

I. Introduction

The ASME Board on Nuclear Codes and Standards (BNCS) originally chartered the Committee on Nuclear Air and Gas Treatment (CONAGT) in 1975 to "develop, review, maintain and coordinate codes and standards for design, fabrication, installation, testing, and inspection of equipment for nuclear power plant air and gas treatment systems."

Under this charter, a draft code section for field testing of nuclear safety-related gas processing systems has been developed and is undergoing the required peer review process.

For the purpose of this code, gas processing equipment either produces a change of state or composition, or alters the concentration of gases. Specifically, equipment designed to stabilize the containment atmosphere or process gasses containing radioactive material during normal or emergency conditions is addressed by this code.

The equipment addressed by the code can generally be distinguished from nuclear air treatment system (NATS) equipment by the following characteristics:

---

*Affiliations of authors:
S. A. Hobart, Electric Power Research Institute, Palo Alto, California.
F. J. Cannito and D. P. Werkheiser, American Nuclear Insurers, Farmington, Connecticut.
J. M. Pleva, Tennessee Valley Authority, Athens, Alabama.
R. Reda, Quadrex Corporation, Campbell, California.
L. B. Nesbitt, General Electric Company, San Jose, California.

- Relatively high pressure,
- Relatively low flow, and
- Relatively high radioactivity content.

In the case of "mixed" safety-related air treatment and gas processing systems, this code section applies to gas processing-type equipment (e.g., compressors, piping, and valves) and references other code sections for testing air treatment equipment (e.g., blowers, ductwork, dampers). This code section therefore applies to gas processing equipment such as igniters, hydrogen recombiners, and sampling and analysis equipment. It may also apply to mixing, diluting, inerting and purge equipment, if that equipment is classified as safety-related and contains gas processing-type equipment.

Excluded from the code are gas supply systems and systems not designed to modify the containment atmosphere.

Field acceptance and surveillance tests for identified equipment are included in this code, while manufacturers's acceptance tests are not included. The equipment performance that ultimately matters is functionality under actual intended operating conditions. Therefore, this code section requires equipment to be tested under actual or simulated operating conditions.

In the code section, the term "equipment train" is used synonymously for the term "system." The equipment train is defined as all components between the process gas influx and release point, including associated instrumentation and controls.

II. Need for Testing Code Sections

It is an established practice for codes to address functional testing of equipment important for the protection of plant personnel and the general public. Usually, codes are sets of minimum specific requirements, written in explicit terms, and are often invoked by law.

The concept of including test sections in codes is not new. There are three sections in the ASME Boiler and Pressure Vessel Code (B&PVC) that address testing: Sections VI, VII, and XI. Sections VI and VII of the B&PVC are guidelines for the care and operation of heating and power boilers, respectively. Section XI covers mandatory in-service inspections and tests for nuclear power plant components. All three sections of the code call for functional testing, in addition to pressure boundary tests.

Other ASME test requirements were issued in 1906 as Power Test Codes. These codes are now known as Performance Test Codes (PTCs) and are invoked to verify conformance with design criteria or federal, state, or local laws.

In addition to codes, many standards such as ASME, ANSI, and IEEE standards also address functional equipment testing. These standards are not legally enforceable, except as invoked by a code. One example of this type of cross-reference is the intent of B&PVC

Section VI to reference the ASME Operations and Maintenance (O&M) Standard for in-service testing of valves.

Nuclear Regulatory Guides, Plant Technical Specifications, and Standard Review Plans also require functional testing of equipment. Since these requirements are subject to unannounced change and have no process for industry review, they do not necessarily form the basis for industry consensus standards.

The need for a specific test code for nuclear gas treatment systems was identified by the ASME BNCS for the following reasons:

- Gas processing equipment functionality is important in order to protect plant personnel and the general public;
- General requirements of the B&PVC must be tailored to the specific equipment designed to process gases;
- Minimum test requirements for this type of equipment are currently lacking; and,
- Test recommendations for gas process equipment are scattered among many different documents.

Therefore, this code section was written to augment and consolidate requirements of existing codes and standards, and to provide a viable single-source document.

III. Relationship to Other CONAGT Code Sections

The Code on Nuclear Air and Gas Treatment is organized into four divisions, as shown in figure 1. The code section described in this paper is part of Division IV, Testing Procedures. Definitions and references included in Section AA, Common Articles, are adopted by inference, unless specifically altered or supplemented. Other sections within Division IV are referenced, as applicable.

IV. Generic Examination and Test Requirements

One article of this code section contains generic examinations and test techniques. Also included are general requirements common to all equipment addressed by this code, as well as requirements pertaining to test personnel and test instruments.

Personnel

Because test personnel must have the necessary training to understand the equipment as well as the purpose of the required test, Section TC, Qualifications of Test Personnel (figure 1), was developed and is referenced here.

Test Instruments

General requirements for test instruments are modeled after similar requirements in Section XI of the ASME B&PVC.

Examination Techniques

There are three major types of examination and test techniques:

- Visual examination,
- Pressure integrity examination, and
- Functional examinations.

The generic test requirements of this article and the specific test requirements of subsequent articles are categorized according to these techniques.

The first technique, visual examination, is used in order to assess the general condition of the equipment that may affect its operability and functionality. By conducting visual examinations prior to other examinations and tests, it is intended to discover gross anomalies that may impact the result of a subsequent examination or test.

The second technique verifies the integrity of the pressure boundary. For acceptance testing, if the entire pressure boundary of a new or existing equipment train has been previously verified as acceptable to the rules of Section III or Section XI of the ASME B&PVC, this test need not be repeated. Periodic surveillance tests are required to reverify the pressure boundary integrity of gas processing equipment. As stated before, this code has provisions for utilizing the test results of the B&PVC (Section XI), provided that the entire pressure boundary is subjected to test.

The last technique, functional tests, is used to verify the operability of the entire equipment train. Due to the unique nature of the various types of equipment covered by this code, functional test requirements are specific in nature and are only addressed in subsequent articles.

IV. Specific Examination and Test Requirements

Specific test requirements are addressed by system or component type in two separate articles. The first article addresses field acceptance tests, while the other article addresses periodic surveillance tests. The components/systems addressed are:

- Igniters,
- Recombiners,
- Mixing, Diluting and Purge Equipment,
- Sampling and Analysis Equipment,

- Blowers and Compressors,
- Piping Components, and
- Electrical, Instrumentation and Control Equipment.

Some of the requirements unique to particular types of equipment are discussed in the following paragraphs.

Igniter Equipment

Igniter equipment is an assembly of electrical equipment utilized to burn hydrogen in primary reactor containment prior to the gas reaching critical concentrations. The equipment train usually consists of igniter elements (e.g., glow plugs), power supplies, supports, and instrumentation, controls, cabling, and other interconnections.

The surface temperature of each igniter element must be measured and recorded during operation for the field acceptance test and the periodic surveillance test. The results of periodic surveillance tests must be compared to the field acceptance test results and analyzed for degradation trends.

Recombiners

Recombiners for containment hydrogen control present a particular difficulty for development of a test code. The problem arises because there exist three fundamentally different types of recombiners, two of which involve assemblies of several processing equipment components. The three types of recombiners currently used for this service are:

- Electric
- Catalytic
- Flame

Electric recombiners are of two types. One type of electric recombiner consists of an enclosure containing electrical resistance heaters. The enclosure serves as a duct for funneling the flow of gases past the heating elements. These units are placed in containment and require only electrical connections in order to function. The other type of electric recombiner consists of a furnace external to the containment. Containment-grade piping and a prime mover (e.g., blower) conduct containment atmosphere to and from the furnace.

Catalytic recombiners typically consist of piping, valves, pressure vessels, heat exchangers, a blower, a catalyst bed, and the associated instrumentation and controls required for monitoring and controlling operation of these miniprocess systems.

Flame recombiners typically consist of piping, valves, a blower, fuel injectors, gas supply and lines, an igniter, and the associated instrumentation and controls.

Because of the physical intricacies of all types of recombiners, both internal and external visual examinations are required. The internal examination is necessary to verify the equipment is configured in accordance with design requirements for internal features, and to ensure that no construction debris remains or construction-related damage occurred. Also included are requirements to verify the integrity of support plates after catalyst installation, and to verify the presence of water in loop seals.

Because catalysts are generally adversely affected by contact with water, hydrostatic tests are required to be conducted prior to catalyst installation. If a hydrostatic test of a catalyst vessel is performed, thorough drying is required before catalyst installation. A leak test of affected joint is required after catalyst installation.

Various components, such as liquid level, flow, and spray controls are to be functionally tested with fluids over the entire range of process input conditions. For example, liquid level controls are to be tested by raising and lowering an actual liquid interface past controller setpoints. This testing may be performed during integrated equipment functional tests.

For catalytic recombiners, the integrated flow test is to be followed with a test using a combustible test gas (hydrogen), to ensure recombination functionality at design concentrations and flow rates. After this testing is complete, the equipment is to be internally inspected for migration of catalyst fines.

Because some plant designs allow shared use of a recombiner at more than one operating unit, the situation may occur where the recombiners are kept in a storage location, rather than being permanently installed. To ensure that the recombiner and permanent connections properly join and the soundness of the equipment was not affected by transportation, the code section requires that each shared recombiner be installed at each of its potential use sites and all connections then be leak-tested and/or functionally verified. Integrated flow testing is required at only one of the potential use sites.

Stored units are to be inspected periodically to assure that they have been maintained in accordance with manufacturers' recommendations (or ANSI NQA-2, if no manufacturer's recommendations exist).

A mandatory appendix detailing safety precautions to be taken when handling hydrogen and oxygen has been provided. The appendix references precautions specified in NFPA-50 and NFPR-50A, and includes material found in the Compressed Gas Association pamphlets G-4 and G-5, with only minor modifications.

Mixing, Diluting, Inerting and Purge Equipment

Test requirements for the majority of components in these systems are covered in other articles. Code Section TA, Field Testing of Air Treatment Systems, is referenced for ductwork and damper test requirements.

Sampling and Analysis Equipment

Containment atmosphere sampling and monitoring equipment used to measure hydrogen and oxygen concentration are of two types:

- Out-of-containment analyzers (external system), and
- In-containment sensors (internal system).

The external system consists of a sensor-analyzer unit outside the containment. Tubing and a prime mover (e.g., vacuum pump) transport a continuous sample of containment atmosphere to and from the analyzer. The internal system consists of a sensor(s) mounted inside containment and electrically connected to out-of-containment instrumentation and controls. The sensor must be qualified for postaccident environments.

Prior to pressure and leakage tests, sample lines must be checked for obstructions by blowing dry air or nitrogen through them. This precaution is necessary because the sample lines are usually small (1/4 in.) and the potential for plugging by construction debris is fairly high.

The component test for gas analyzers consists of an operational test using zero and span gases. The span gas mixture is required to be within the upper range of interest for the analyzer. A nonmandatory appendix provides guidance for calibrating analyzers and verifying results. This appendix includes information from ANSI/ASTM D3249-79, with minor modifications.

Certain in-line hydrogen and oxygen analyzers have experienced operational failures due to ingress of water. Because there is a significant potential for condensation of containment atmospheric steam within sample lines with subsequent flow blockage, an integrated flow test under design basis accident conditions (pressure, temperature, relative humidity and time) is required for field acceptance. During this exposure test, the flow rate must be continuously monitored and performance of the analyzer must be verified at least every ten hours. This test is intended to verify that sample line design and installation have precluded the formation of loop seals by condensate and will free-drain back to the containment penetrations.

Condensation and resulting flow blockage could be prevented by maintaining the containment air sample above its dew point temperature along the entire length of sample line. Provision has therefore been made to reduce the required exposure test time to four hours if the sample line is heat-traced. This test will ensure that the heat tracing is capable of maintaining sample temperature during post-accident conditions.

Testing of Individual Components

Test requirements for types of components that are contained in more than one equipment train are detailed in the subarticles following those for equipment train testing. Those components include blowers, compressors, piping components, electrical and instrumentation and control equipment.

Blowers and Compressors. Section TA, Field Testing of Nuclear Air Treatment Systems (figure 1), is referenced for component tests of blowers.

Component tests for compressors consist of vibration amplitude measurements. The compressor test requirements found in ASME B&PVC Section XI, Part IWG, were used as guide for this testing. Acceptance criteria of IWG were modified so that allowable ranges of vibration are not expressed as a function of a reference temperature and pressure, but only of the reference vibration measurements. The tolerances for allowable vibration remain the same as those in IWG.

Piping Components. Valves are the only piping components requiring tests not specified by pressure boundary, electrical or instrumentation and control tests.

ASME B&PVC Section XI, Part IWV, is referenced for component functional testing. The equipment is already under the purview of Section XI; this provides merely a cross-reference, not a new requirement. As previously noted, it is intended that the O&M Standard on valves will eventually be referenced by Section XI. Section TA, by referencing Section XI, will thus also be linked to the O&M Standard.

Electrical and Instrumentation and Control Equipment. Component functional testing required includes:

- Manual and automatic (when applicable) operation of electrical equipment and observation of response,
- Introduction of a test signal to verify equipment trip setpoints, and
- Testing the functionality of any equipment bypass controls and the associated status indicators.

References for continuity, resistance to ground, calibration, and functional performance tests include IEEE-43-1974, NEMA Electrical Standard ICS-1-1978, Part 109, and IEEE-498-1985. In addition, the following component functional tests are required:

- Insulation resistance tests on all electrical drivers,
- Current measurement on each phase of power supplies,
- Polarity of 120 vac, 50-Hz, 3-wire systems,
- Resistance between the neutral side of power supplies and ground, and
- Resistance between the ground connections of all enclosures and earth ground (per IEEE-27-1974).

Acceptable methods for continuity testing are also described.

All instrumentation and controls necessary to operate the equipment train shall be subject to the same test and calibration requirements as for permanently mounted test and calibration equipment.

Annunciator circuits must be tested for functionality by introduction of a test signal, as well as the physical manipulation of process conditions.

VI. Testing Following an Abnormal Incident

Abnormal incidents have the potential for affecting the equipment covered by this code section. An evaluation must be performed and documented to determine the scope of required retesting.

VII. Repair and Retest

Unacceptable equipment conditions must be corrected by replacement, repair, or analysis. The analysis must demonstrate that the condition does not impair equipment performance to design specifications. A simple successful repetition of a test cannot be used to negate a failed test; corrective action must precede the retest.

If the cause of the deviation cannot be determined by inspection or analysis, corrective action may consist of recalibration of test instruments and subsequent retesting. If it is determined that the unacceptable situation is attributable to equipment condition or malfunction, rather than problems with the test equipment or procedure, the equipment must be taken out of service or declared inoperative until corrective action has been taken.

VIII. Documentation

Documentation for the examination, tests, replacements, repairs, corrective actions, analyses, and evaluations must be prepared and maintained for the service lifetime of the nuclear power plant, in accordance with the utility's quality assurance program.

IX. Summary

The code section has received preliminary review from test, operations, and engineering personnel from five utilities, as well as personnel from an equipment manufacturer/supplier, insurers, and an engineering firm. Comments from these sources have been invaluable and all were addressed and resolved by the subgroup. Additional comments received during the ASME review and public comment process will ensure this document is usable, while setting forth minimum test and performance requirements.

Figure 1

Organization of
ASME Code on Nuclear Air and Gas Treatment

Division I - General Requirements

Section AA - Common Articles

Division II - Ventilation Air Cleaning and Ventilation Air Conditioning

---

Section BA - Fans and blowers
Section CA - Conditioning equipment
Section DA - Dampers and louvers
Section FA - Moisture separators
Section FB - Prefilters and frames
Section FC - HEPA filters and frames
Section FD - Sorbers and frames
Section FE - Sorbent media
Section IA - Instrumentation and control
Section RA - Refrigeration equipment
Section SA - Ductwork

Division III - Process Gas Treatment

Section GA - Pressure vessels, piping, heat exchangers, and valves
Section GB - Noble gas hold-up equipment
Section GC - Compressors
Section GD - Other radionuclide equipment
Section GE - Hydrogen recombiners
Section GF - Gas sampling

Division IV - Testing Procedures

Section TA - Field testing of air treatment systems
Section TB - Field testing of gas processing systems
Section TC - Personnel qualification
Section TD - Laboratory qualification

Review Of Current Halide and Aerosol Leak Testing Methods of Nuclear Air Treatment Systems, Components and Banks

BY J.W. JACOX
JACOX ASSOCIATES

ABSTRACT

All nuclear power plants, and nearly all other types of nuclear facilities, have one or more Nuclear Air Treatment Systems (NATS) to control influent, effluent, or process flow. Many NATS are required to be periodically tested by the facility licensee, and all should be tested per good engineering practice. The basis for this testing in the United States is ANSI/ASME N510-1980 (or -1975), "Testing of Nuclear Air Cleaning Systems."

There are a number of different testing methods, and this paper will discuss the advantages and disadvantages of each method along with ALARA considerations and cost considerations.

With the information and analysis presented in this paper the engineer will have a reasonable basis in both theory and practical experience upon which to help propose the best possible method for testing existing systems and for design of new NATS.

INTRODUCTION AND BACKGROUND

Over the approximately 30 years of the commercial nuclear power industry's history, there has been significant evolution of the Nuclear Air Treatment Systems (NATS) used in these plants. Both the number of systems per plant and system complexity have increased. Concurrently, the requirements (both regulatory and technical standards) for testing the NATS have also increased. Unfortunately the physical provisions in the systems to allow performance of the up-graded testing has not kept pace with the testing requirements. This has led to the development of a number of test methods designed to cope with the problems of attempting to meet testing requirements without the needed physical provisions built in the NATS. This paper will review the historical, technical and standards aspects of this situation.

When the first commercial nuclear electrical power production plants in the United States came on-line in the early 1960's (actually starting with Shippingport in 1957), there were no industry standards on nuclear air filtration systems, no specific regulations on these systems, no specific textbooks or hand books on them, and no formal courses or seminars for training. The only source of specific technical papers was the series of Nuclear Air Cleaning Conferences (1) originally sponsored by the AEC (and now by DOE). The first was held in 1951 and was classified "Secret" (it was never published in the open literature). Later ones in 1952, 1953, 1955, 1957, 1959 and every other year thereafter, were not classified, or if so, were later declassified and proceedings were published. Unfortunately the timing of these early conferences was at, best parallel, for the design of the first plants. Even with the much shorter lead time enjoyed in the early days of the nuclear industry at least 5 years was required so the early plants were contracted for in the mid to late 1950's. This meant that the only basis for NATS design, testing or regulation was the expertise of the technical personnel involved. Fortunately many of these engineers had experience from the Naval Reactors Program, or the earlier Manhattan District Project. This experience, along with good engineering judgement, produced NATS that, perhaps primitive by today's standards, did the required job well enough that there have been no radioactive releases through these systems that have had an adverse effect on the public or the environment.

Two areas that did, and in some cases still do, suffer from a lack of experienced technical personnel, were maintenance and testing. Maintenance is outside the scope of this paper and testing is its substance.

The very first plants had only one or two air filter systems and would hardly be recognized as NATS today. HEPA filters were well known from the Manhattan District Project and follow-on weapons work. Carbon filters for adsorption of gaseous radioactive fission products were even more primitive. The existance of organic forms of Iodine was not considered in those early days. Initially the only actual testing, if any, beyond visual inspection, was an aerosol leak test. The provision for this test was usually the ability to run a sample line into a housing by removing some component and using its pressure boundary penetration. From this we have evolved to NRC Regulatory Guides 1.52 Rev 2 (2) and 1.140 Rev 1 (3), ANSI/ASME N509-1980 and N510-1980, (4)(5)(6)(7) (with 1986 revisions in-process) and detailed NRC Inspection and Enforcement Procedures for HVAC in the "Inspection and Enforcement Manual" section 50100 (8).

The earliest generally available guidance for Halide leak testing of adsorbers was from the Savannah River Production facility of the AEC in publication DP 1082 (9,11). This was published in 1967. ANSI N101.1-1972 (10) was the first industry standard for DOP aerosol leak testing of HEPA filters. It was published in 1972. These early standards and technical reports were of great assistance to the industry, and were the basis for later documents. They were, however, limited in scope and not written for broad nuclear power plant use. The need for broader standards was recognized and an ad-hoc group was formed in July of 1971 under the American National Standards Institute (ANSI) N45 Committee on Reactor Plants. The American Society of Mechanical Engineers (ASME) was the direct sponsor. This group authored the original issue of N509 "Nuclear Power Plant Air Cleaning Units and Components" in 1976, and N510 "Testing of Nuclear Air Cleaning Systems" in 1975. Need for continuing development and ongoing revision was immediately recognized, and the permanent ASME Code "Committee on Nuclear Air and Gas Treatment" was accredited in January, 1976. CONAGT has produced the 1980 editions of N509 and N510, and is working on the 1986 editions. CONAGT is also writing an expanded Code including both NATS and Gas Processing systems for future use.

In addition to the overall and unavoidable problem of a learning curve there is the very real problem created by the long lead time between design of a NATS and its Acceptance Testing. Today this is unfortunately on the order of 10 years. This means the testing requirements may be more demanding than the equipment design anticipated even when the letter and intent of the requirements at the time of design were fully implemented. Unfortunately this full implementation of the intent of the standards is less common than it should be so the overall problem is further complicated.

The most common NATS consists of a series of component banks in a filter housing. The simplest would be a prefilter and HEPA bank with the most complex a Standby Gas Treatment or Control Room Emergency Makeup with a Moisture Separator, Heater, Prefilter, HEPA filter, Adsorber and Final HEPA filter (Figures 1, 2 and 3). Many variations and combinations exist including multiple adsorber banks in series. It is the series aspect of component banks that is the most common source of testing problems. By regulation and good engineering practice, each bank must be leak tested individually for credit to be taken for that bank. This requires each bank to be individually challenged and sampled. How to introduce an aerosol challenge between two HEPA banks, and how to sample between them in a manner that evenly challenges and samples the entire bank equally is a significant problem. The same problem exists for series adsorbers further complicated by the time dependency of a halide leak test. Fortunately series adsorbers are very unusual, and therefore will not be treated as a separate item in this paper. Most comments on series HEPA banks will apply to series adsorbers.

Over the years a number of common, de facto, procedures were developed to allow a bank leak test of some type to be performed. Some of these procedures provide reasonably accurate indications of bank leak tightness but at a great cost in time and effort. Others have significant technical flaws. That is not to say they are worthless. Simply, that as the sophistication of the industry has evolved, better methods have been developed, so older methods can no longer be justified. One method stands out as the most technically correct, and also the most cost efficient in the long run. It adds slightly to the upfront capital cost (which is the most visible), and therefore is often resisted. The analysis in this paper will call attention to the significant costs, as well as the purely technical aspects of each method.

The most common methods in use today to perform ANSI/ASME N510 Leak Testing can be summarized as:

1. Shroud Test Method (STM)
2. Component Removal Method (CRM)
3. Selective Insertion Method (SIM)
4. Multiple Sampling Method (MSM)
5. Test Manifold Method (TMM)
6. Single Point Sampling (SPS)

DEFINITIONS

For consistancy and to reduce possible confusion the following definitions are used in this paper:

1. Aerosol - A suspension of small particles, solid or liquid, in air. The diameter or major dimension of the particles may vary from 100 microns down to 0.1 micron or less (a micron is 1/1,000,000 meter or about 1/25,000 inch.

2. Challenge Agent - Either DOP or Halide used to measure leakage through a component bank.

3. Component Removal Method (CRM) - The removal of one or more components from a component bank to allow challenge agent to flow through said component bank without filtration, delay, or reduction of the challenge agent concentration.

4. DOP - Dioctyl Phthalate (di-2-ethyl hexyl phthalate) an oily, clear, noncorrosive liquid that forms an aerosol of repeatable dimensions under given parameters of temperature, pressure, flow, etc.

5. DOP Aerosol - By N510-1980 definition, the polydispersed aerosol shall have:

"an approximate light-scattering mean droplet size distribution as follows:
99+% less than 3.0 microns
50+% less than 0.7 micron
10+% less than 0.4 micron"

6 Halide (per ANSI B79.1) (11)
2.6.1 R-11; Tricholoro (mono) fluormethane
2.6.2 R-112; 1,1,2,2-Tetrachlorodifloroethane

7. Multiple Sampling Method (MSM) - The taking of many individual samples in a defined cross section or housing area over a period of time to be averaged by calculation per Section 11 of N510-1980. (Note: "Section" references are to N510-1980 unless otherwise specified.)

8. Selective Insertion Method (SIM) - The selective insertion or installation of components during bank loading to allow passage of challenge agent through said bank before all components are installed.

9. Shroud Test Method (STM) - The use of a device (shroud) to limit flow from a generator, or to a detector, through a subset of the entire bank under test. See Section 10.6 of N510-1975.

10. Test Manifold Method (TMM) - The use of devices (manifolds) to inject or sample challenge agents over a defined area or housing cross section with the concentrations averaged by physical mixing to provide the equivalent of a single point sample to a detector or even distribution from an injection position.

11. Single Point Sampling (SPS) - The taking of a single sample as representative of the Challenge Agent concentration over a defined area or housing cross section. For a SPS to be used, test data must have been obtained to verify the sample is representative (Section 9).

Each of the listed methods will be described and discussed as to their strengths and weaknesses.

DISCUSSION

SHROUD TEST METHOD (STM)

The STM is one of the oldest methods, and while very useful in the past, it has been superseded by better methods. It was deleted as an acceptable integrated leak test method by N510-1980. The only current application of this method would be to assist in scanning for locating leaks (Section 10.1).

A shroud is a device to enclose a portion of a filter or adsorber bank to limit the challenge injection and sampling to this restricted section (Figures 4,5). The original basis of need was very real. It was the limited output of DOP Aerosol Generators. Laskin nozzle air operated generators were usually limited to an output sufficient for about 15,000 SCFM. Even at this output, they were being pushed to their limits. High output DOP Aerosol Generators for 80,000 SCFM systems have been commercial units since 1975.

For the bank, other than the perimeter, shroud design was not too difficult. It would enclose one or more filters in a box form on both upstream and downstream faces of a bank. There is the immediate and obvious trade off of size/weight with attendant handling and problems vs. the number of setups and tests made. At best handling any object in a housing with installed components is difficult and time consuming. Such handling often causes damage to the installed components. Once the shrouds were installed, a local leak test was performed. The total leakage was then obtained by calculation when all locations had been tested.

The perimeter filters are very difficult to test with a shroud. Since the frame to housing seal must be included as a part of any leak bank test, the shroud must seal to the wall, floor, or ceiling depending on the filters being tested. This usually means three shroud sets, or at least three setups or configurations of a single, more complex shroud set. Obviously complexity, weight, expense, and delay are increased by these different configurations.

An additional problem with shrouds is the inherent probability of both Type I and/or Type II errors. That is, failing an acceptable bank or passing an unacceptable one. The basis for this problem is the need to ensure total coverage of all possible leak paths. This is usually done by overlap of the area covered by different shroud locations/setups. Double counting the same leak will occur when the shroud overlaps to include the entire bank. Certainly this is the conservative and safe approach.

If the shroud does not cover all possible leak paths, then leaks could obviously be missed. The housing to frame seal welds were the most commonly missed potential leaks as many shrouds were never designed to seal to the wall.

As mentioned, damage to HEPA filters and other NATS components was a serious problem when using a shroud even with faceguards on the HEPA filters.

Uniform air flow distribution is a question universally (in this author's experience) neglected when using shrouds. To meet the requirements of N510-1975, an Air Flow Distribution Test must be performed for each Shroud Setup.

ADVANTAGES

1. In the past it allowed testing of large systems when there were no alternate options due to DOP Aerosol generator limitations.

2. If leaks must be found on a bank that failed some other form of integrated leak test, it could assist in scanning by DOP, loading only a portion of the bank at a time.

DISADVANTAGES

1. The requirement of fabricating, handling, mounting, and moving the shrouds. Inside a housing handling is time consuming and a potentially serious problem. If the system has any contamination, it also becomes an ALARA problem. The frame requires

provision to hold the shroud in place.

2. Missing frame to housing seals and potential leaks.

3. Type I and Type II errors.

4. Damage to HEPA filters (and other components).

5. Much longer time required for test.

6. Requirement to perform an Air Flow Distribution Test for each Shroud Setup.

7. It has been deleted as an acceptable test in N510-1980 (and draft-1986).

8. The need for shrouds no longer exists as high output DOP Aerosol generators have long been commercially available.

COMPONENT REMOVAL METHOD (CRM)

The CRM method is also a long used method. The method is exactly as the title suggests. One or more HEPA filters in the first (upstream) bank are removed to allow DOP challenge to pass through to test the second (downstream) bank. (Figures 7,8). The DOP flow is also through the adsorbent when such a bank exists between the HEPA banks.

This can provide an acceptable test of the second HEPA bank but the problem remains of how to test the first bank. After the removed filters are reinstalled in the first bank it must be leak tested. Obviously HEPA filters cannot be removed from the second bank, or it would need to be retested, and you would be in an endless loop of testing. The usual method of downstream sampling for the first HEPA bank is the MSM.

The CRM has been used on high efficiency prefilters to provide sufficient challenge to the first HEPA bank. This is more acceptable than removing HEPA's since prefilters do not need testing, but it is still a less than ideal process. Airflow Distribution will change when some prefilters are removed. Again, the question of Air Flow Distribution arises. To meet the requirements of N510, an Air Flow Distribution Test is required prior to a Bank Leak Test if any modifications are made. Upstream filter removal is certainly a modification. Even if the same filters are always removed, the dirt loading, and, therefore, the pressure drops will be different, so the test is always required. By the same reasoning, the Air/Aerosol Test is also always required.

At times, Type II carbon trays are removed along with the selected HEPA filters in the first bank. Such removal is a good practice since it keeps the DOP from passing through the actual adsorbent. It is, however, a labor intensive process and adds additional possibilities for damage and resealing problems. This practice also further compounds the Air Flow Distribution and Air/Aerosol Mixing situation.

In any CRM approach, the units removed must be carefully chosen to provide uniform air flow and challenge/air mixing for the bank to be tested.

ADVANTAGES

1. It allows testing of series systems that would not be otherwise (except by some method discussed here) testable.

2. It does not require any fixtures or extra test apparatus.

3. Depending on the system, it is usually reasonably rapid compared to a shroud method.

4. Properly performed it challenges the entire bank so the Type I/Type II error problem is eliminated.

DISADVANTAGES

1. It requires considerable time inside a housing which can be an ALARA problem.

2. It requires removal and reinstallation of a number of HEPA filters which will usually result in some damage. It also presents the problem of reseating a filter with a deformed gasket. Leaks often result from re-using a HEPA filter that has been clamped in place since the gaskets take on a permanent deformation. The same problem exists for removal and replacement of Type II Trays.

3. For the method to provide uniform challenge, the position and number of filters removed requires careful analysis. Both an Air Flow Distribution test and an Air/Aerosol Mixing Uniformity test per N510 is required to confirm that proper air flow and challenge is obtained at the bankface.

4. Unless the entire bank of Type II Trays is removed, DOP will go through the adsorbent. On a two inch Type II tray, this is not a serious problem, but equally, not good practice.

5. If a Type III deep bed adsorber is in the flow path, a problem exists with DOP flow through the adsorbent. At 4 inches or greater, a significant quantity of DOP will be removed by the carbon. As the bed depth increases so does the problem. Additionally, DOP appears to be a potential poison to the impregnated carbon. Unfortunately there is <u>very</u> little data on DOP as a

poison in the quantities used for leak tests, but we know most hydrocarbons have a negative effect on the impregnated carbon.

6. Much longer time required for test.

7. The test is not specifically recognized by N510

SELECTIVE INSERTION METHOD (SIM)

The SIM approach is very similar to the CRM except that by definition it applies only at startup (or in some cases filter changeout). Filters and adsorbent are installed in an order such that the various tests may be performed the same as they are in the CRM. One advantage is that the DOP testing can (if a deviation is taken on the Airflow Distribution test, Section 8.2) be completed before the carbon is installed. (Figures 9,10)

The problem of Airflow Distribution testing is critical to the use of this method. N510-1980 requires the Airflow Distribution Test as a prerequisite to the DOP leak test. An Airflow Distribution Test requires all the actual filters and carbon be installed. If all components are installed, the situation is that of using the CRM not the SIM. In either case if the Airflow Distribution test is not performed and passed, then N510-1980 has been violated.

Most significant in an analysis of the SIM is the impact on plant operation. It may allow a valid leak test if the Airflow Distribution problem is somehow addressed during Startup but except in exceptional circumstances, it CANNOT BE REPEATED AS A SURVEILLANCE TEST during plant operation. This point is often "neglected" to be mentioned when the SIM is used for Acceptance Tests.

ADVANTAGES

1. It allows a test to be performed that would not otherwise (except by some other method discussed here) be possible.

2. It does not require any fixtures or extra test apparatus.

3. Depending on the system, it is usually reasonably rapid compared to a shroud method.

4. Properly performed, it challenges the entire bank so the Type I/Type II errors are eliminated.

DISADVANTAGES

1. It is essentially a Startup/Acceptance test only, and leaves the problem of life of plant Surveillance Tests to the plant operating staff. This is seldom acknowledged at the time of the test.

2. The problem of Airflow Distribution testing is critical and often simply overlooked. This will make the Test invalid on procedural grounds. Further, when the test is performed, if it does not meet the N510 requirement of all components installed, an Air/Aerosol Mixing Test is performed under conditions contrary to the requirements of N510.

3. The problem of downstream sampling of the first HEPA bank is left open as in the CRM. Multiple sampling is the usual approach for this requirement.

MULTIPLE SAMPLING METHOD (MSM)

The MSM is performed as the name implies. A number of samples (usually large) are taken over the subject cross section, and the average concentration is calculated along with the standard error, and the 95% confidence level of the concentration per Section 11 of N510-1980. (Figures 11, 12)

While the first formal step in the procedure (11.4) is to define a set of sample points, the <u>prerequisite preliminary step</u> is to scan the entire bank including the frame to housing interface, locate, and have all leaks repaired. The immediate problem is that leak is not defined. How small or large an indication of increased DOP concentration constitutes a leak? At best, this is a very time consuming step, and loads considerable DOP on a large bank. It will also be a significant ALARA problem in a contaminated system. The question of what level of DOP concentration is acceptable without being defined as a leak is not addressed in N510-1980. It is not an easy question. If the level is set too high, then too much leakage may be accepted. If set too low, it will take an unreasonable length of time to eliminate all these indications. This prerequisite is a major undertaking, and again is usualy ignored when a case is being made for the MSM.

After the scanning repair and rescanning, the formal sample areas must be chosen. The guidance given is theoretically correct, but could lead to missing frame to housing leaks unless executed with great care. If the prescanning was well executed, then the formal data taking is essentially instrument noise or random particles that always penetrate or come off the actual media. Further the scanning in step 11.4.2 does not take into quantitative account the area of the maximum reading. It could be a point or an area equal to (or even larger than) the traverse area selected. This reduces the quantitative confidence of the method regardless of mathematical calculations based on statistics. It is also very time consuming.

Note that the MSM does not in any way address the problem of injection of a challenge for a down stream component bank. In a HEPA-carbon-HEPA system, DOP must be injected after the carbon to challenge the down stream HEPA. While the MSM can provide concentration data, it simply ignores this injection problem.

N510-1980 in 11.1 discusses penetration, but in 11.4 defines "Cu" as the upstream concentration. For upstream it would be an Air/Aerosol Mixing Uniformity Test per Section 9. When used for penetration, it could make sense as providing necessary data per discussion in other methods.

When the MSM is used, the samples should be taken where there is the best mixing provided by the system itself. For example in a Type III adsorber, the inlet slots provide good mixing for the downstream sample of a first HEPA bank.

ADVANTAGES

1. For upstream readings of a system where the ±20% challenge uniformity cannot be met (and modification is not made to bring the NATS up to standard), this is a reasonable method.

2. In general, it can always be used to take readings, IF there is any physical access, either for man entry or for remote traverses.

3. It does not require any component removal, or matter in what order components are installed. There is usually no problem of Airflow Distribution. If a person is inside a small system, some air flow disruption obviously takes place, but does not seem to constitute a problem.

DISADVANTAGES

1. Long exposure of HEPA filters to DOP loading.

2. NO possible application to Halide leak testing due to strict time dependency of a Halide leak test.

3. Serious potential ALARA problems due to long duration of work required inside the housing.

4. Limited confidence in the value of downstream readings when there are no leaks. The value should be low and provides good indication that the bank integrity is leak free, but the actual calculated penetration values are very shaky.

5. Based on the prerequisite scanning, it is more of a qualitative exercise that there are no leaks over some arbitrary

value. This may be all that is needed, but the MSM must be recognized for what it is in relation to a real integrated leak test penetration (or efficiency) value.

6. The problem of injection for challenge of a down stream component bank remains totally unaddressed by the MSM. This is a critical flaw that is usually not discussed in relation to the MSM.

7. Access to the entire bank face is required.

TEST MANIFOLD METHOD (TMM)

A Test Manifold (TM) is a device that takes a number of representative samples over a defined cross section of a housing, all at the same time, and averages them by physical mixing for a single sample representative of the entire cross section (Figures 13,14 and 15). The TM may be a permanent part of the housing (greatly preferred), or temporary and installed only for the test. Usually temporary TM's are only used if they are added after the housing is installed, and were not designed and built in as a part of the original system.

Since a TM is operating "real time" (less a very short delay due to sample transmit time), it can be used for Halide tests as well as DOP tests. This "real time" sample also allows a very efficient and rapid test. With permanently installed TMs (or after temporary TMs have been installed), the actual leak test is performed in a few minutes; the same as for the ideal Single Point Sampling Method.

The design of the TM should use the inherent mixing of the housing. The previously mentioned slots of a Type III adsorber are excellent locations for manifolds.

Based on the N509 and N510 (all editions) requirements for testability, TMs should be designed and built in systems to allow leak testing as necessary. As discussed in this paper, many NATS did not have them included so they have been added as backfits. This presents the problems of seismic analysis, possible interference for maintenance, and component changeout, as well as "after the fact" design in the field. These backfit TMs are usually designed for temporary use to avoid the seismic and interference problems. While far better than no manifolds, temporary TMs are not as desirable as permanent TMs. The increased initial cost of a permanent TM is quickly paid back in plant operational use. With a permanent TM, a leak test can be conducted without otherwise disturbing an operating (or operational) system. That is, it is a leak test that is possible without any intrusion into the system housing.

ADVANTAGES

1. It can be used for both Halide and DOP Leak Tests.

2. It provides the same convenience and accuracy as Single Point Sampling.

3. It will allow a test to be performed without entry into the housing (for permanently installed TMs).

4. Usually, a shorter time is required in a housing to install a TM (designed for temporary use) than for the MSM, STM, CRM, or SIM.

5. It provides greater repeatability since a fixed sample configuration is used for each test based on the fixed physical construction of the TM. This applies to both permanent or temporary TMs.

6. It can be used with generally less requirements for training and skill of the test engineers than any of the other methods (except SPS, of course).

DISADVANTAGES

1. Some increase in initial cost.

2. If temporary, controlled storage is critical so the TM will be available when needed.

3. The TM takes up some space in the housing where space is usually in short supply.

CONCLUSION

Given the noted and obvious disadvantages of the CRM, MSM, SIM, and STM, the conclusion is firm that the TMM is the method of choice when SPS is not possible for series components. Further, it is specifically required (or at least such was the intent) in N509, N510, R.G 1.52, and R.G. 1.140. From a "life of plant" cost standpoint, the TMM is clearly the least costly based only on simple test time. Unfortunately the "life of plant" cost does not seem to be used in most design analyses, or bid evaluations for NATS. When ALARA considerations are added, the TTM advantage is greatly reinforced. Adding the significant technical deficiencies discussed makes this conclusion even more obvious.

Certainly in some NATS configurations such as a HEPA-HEPA system, a manifold may be difficult to design, and end up with rather complex baffles added to provide mixing. (An excellent reference on basic mixing baffles and related testing topics is

"A Comparison of the Methods Used in the Nuclear Industry to Test High Efficiency Filters") (12). However, the same type of problems will exist for any alternate method, and be repeated each time the system is tested.

The problems of Airflow Distribution and Air/Aerosol Mixing as required prerequisite tests each time an alternate Leak Test method is performed have historically been ignored. This brings to question many leak tests performed in the past.

TMs are often argued to be unnecessary added cost, but when there is any real analysis of the technical merits and life of plant costs; rather simply the cost of a day's time for a test team on a "one shot" basis vs. the one time cost of TMs, the answer always is the same on paper. It is not clear why all plants have not upgraded existing systems to the required standards, and to good engineering practice.

Whatever the specifications, regulations, or industry standards require, or are interpreted to require, the real design objective should be for a NATS that will operate, and be testable and maintainable; at a maximum technical level and at minimum cost for the life of the plant. TMs clearly are required to meet these criteria if Single Point Sampling and Injection is not possible.

REFERENCES

[1] AEC/ERDA/DOE-Harvard Nuclear Air Cleaning Conferences, (1951 to present)

[2] US NRC Regulatory Guide 1.52, Rev 2, "Design, Testing, And Maintenance Criteria For Post Accident Engineered-Safety-Feature Atmosphere Cleanup System Air Filtration And Adsorption Units Of Light-Water-Cooled Nuclear Power Plants"

[3] US NRC Regulatory Guide 1.140, Rev 1, "Design, Testing, And Maintenance Criteria For Normal Ventilation Exhaust System Air Filtration And Adsorption Units Of Light-Water-Cooled Nuclear Power Plants"

[4] ANSI/ASME N509-1976, "Nuclear Power Plant Air Cleaning Units and Components"

[5] ANSI/ASME N509-1980, "Nuclear Power Plant Air Cleaning Units and Components"

[6] ANSI/ASME N510-1975, "Testing of Nuclear Air Cleaning Systems"

[7] ANSI/ASME N510-1980, "Testing of Nuclear Air Cleaning Systems"

[8] US NRC "Inspection and Enforcement Manual," 17 October 1975

[9] USAEC DP-1082, July 1967, "Standardized Nondestructive Test of Carbon Beds for Reactor Confinement Applications" *

[10] ANSI N101.1-72, "Efficiency Testing of Air Cleaning Systems Containing Devices for Removal of Particulates"

[11] ANSI B79.1, "Number Designation of Refrigerants"

[12] "A Comparison Of The Methods Used In The Nuclear Industry To Test High Efficiency Filters," R.G. Dorman, Commission Of European Communities, June 1981

*NOTE: A series of DP reports in the mid-sixties were written on the technical area of leak testing of adsorbent beds. DP-1082 is the one most commonly referenced.

PUBLICATIONS SOURCES

[1] U.S. Nuclear Regulatory Commission Documents

A) U.S. Government Printion Office
P.O. Box 27982
Washington, DC 20013-7082
(202) 275-2060 (202) 275-2171

B) Publication Service
U.S. NRC
Washington, DC 20555
(301) 492-7914

C) NRC Public Document Room
(202) 634-3273

D) National Technical Information Service (NTIS)
U.S. Department Of Commerce
5285 Port Royal Road
Springfield, VA 22161
(703) 487-4650

[2] ANSI/ASME Standards

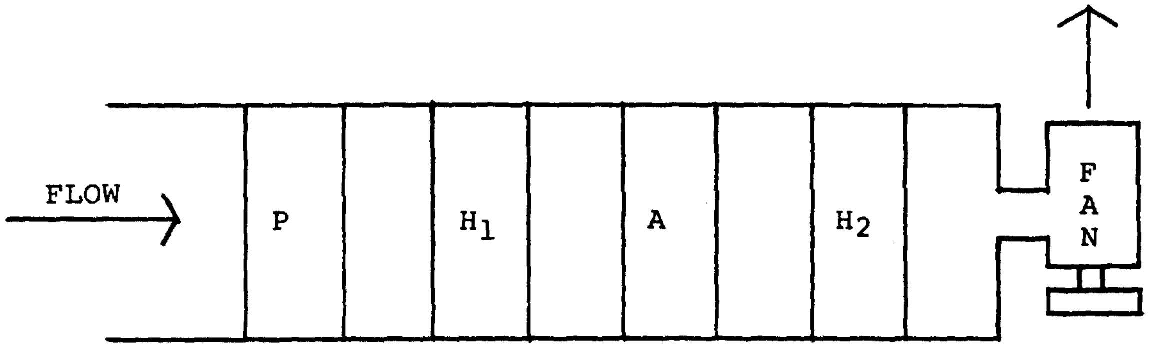
A) The AMerican Society Of Mechanical Engineers (ASME)
345 East 47th Street
New York, NY 10017
(212) 705-7801, Nuclear Department

B) American National Standards Institute (ANSI)
Sales Department
1430 Broadway
New York, NY 10018
(212) 642-4900

[3] Organization for Economic Co-operation and Development (OECD)

A) Nuclear Energy Agency
Organization for Economic Co-operation and Development
Director of Information, OECD
2, rue Andre-Pascal
75775 PARIS CEDEX 16
FRANCE

B) OECD (USA Address)
OECD Publications and Information Center
1750 Pennsylvania Avenue N.W.
Washington, DC 20006-4582
(202) 724-1857



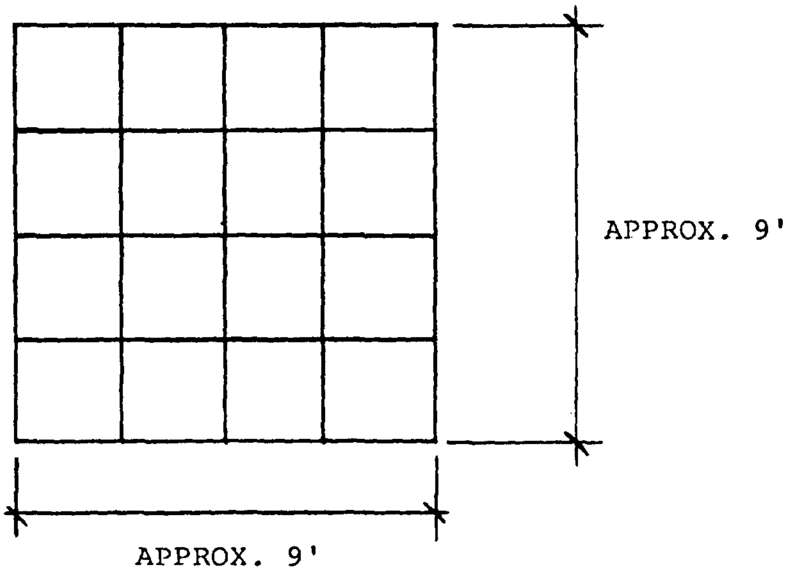
Typical NATS configuration with Prefilter, HEPA filter, Adsorber and HEPA filter banks in series. This will be used for all examples

CONFIGURATION:

P = Prefilter Bank
A = Adsorber Bank

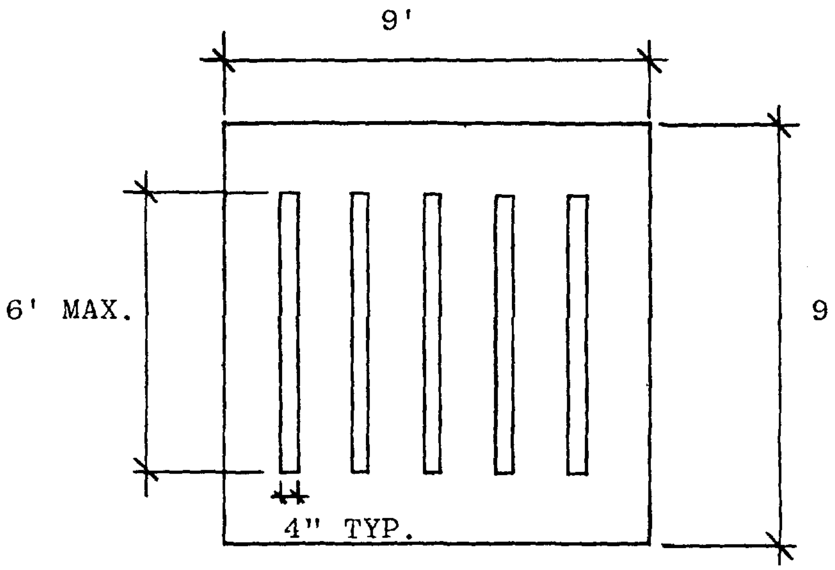
H_1= First HEPA Bank
H_2= Second HEPA Bank

FIGURE 1



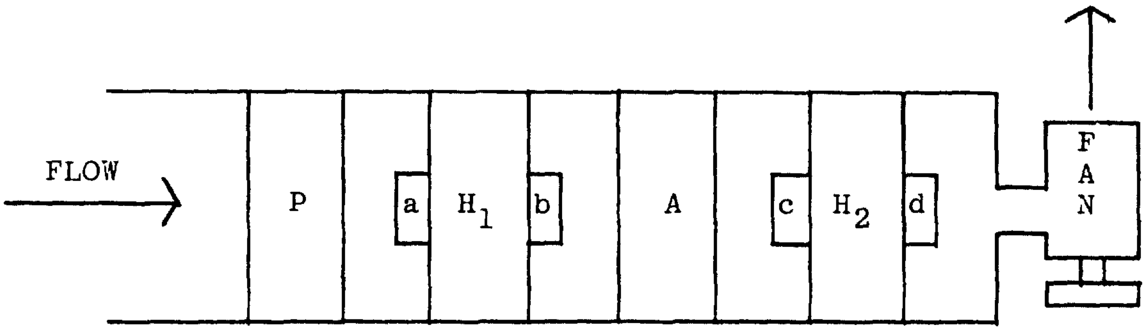
Typical 4 x 4 HEPA Filter Bank. This configuration will be used for all examples.

FIGURE 2



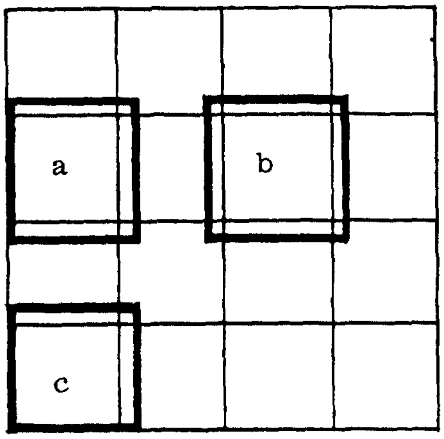
Typical Type III Deep Bed Adsorber showing flow slot configuration. Note small face area of slots vs. that of HEPA Banks. Slot area is typically 10% to 15% of HEPA face area.

FIGURE 3



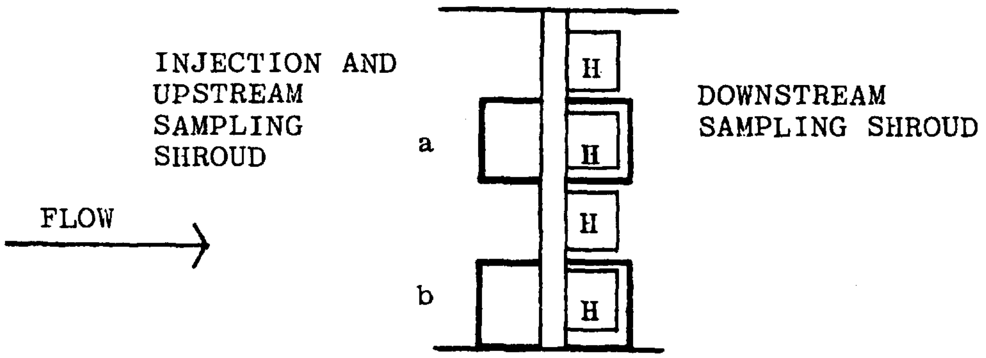
Typical placement of Shrouds to test the HEPA Banks. Shrouds a/b and c/d must be moved in pairs to perform the test. There is usually only one pair and the tests done sequentially.

FIGURE 4



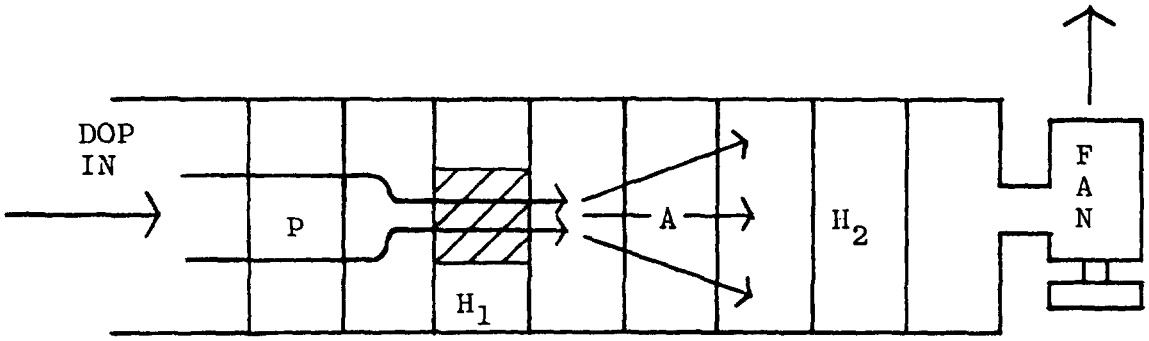
Example of the three configurations that shroud design must address--a is filter plus filter bank seal to side (or top or bottom) wall, b is filter(s) only and c is the corner. All three exist on all NATS.

FIGURE 5



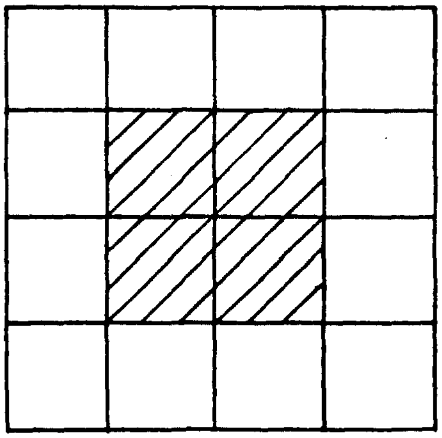
Side view of typical shroud used on a 4 high HEPA bank. Two shroud sets are shown in place but usually only one set at a time is used. Note the different design required to test a center filter vs. a perimeter filter where the housing wall must be included.

FIGURE 6



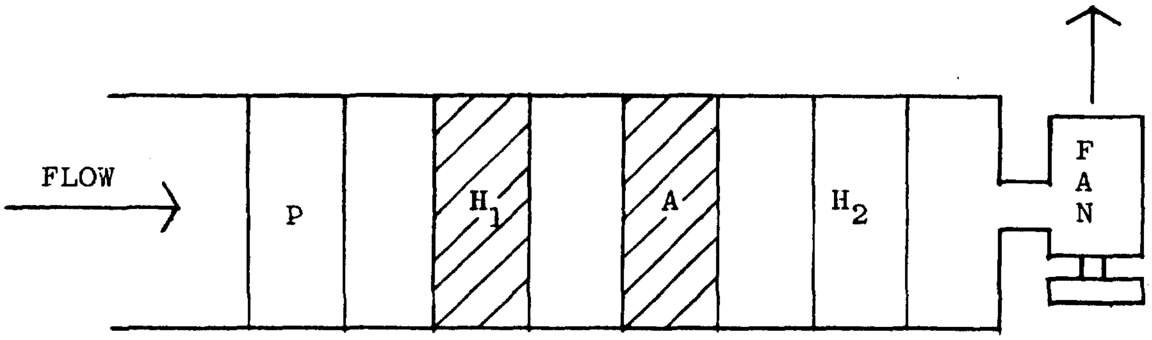
Example of CRM configuration to test HEPA Bank H_2 by removing filters from Bank H_1. Injection is in the up stream duct. Shaded area in H_1 shows removed filters. DOP is injected through the Adsorbent.

FIGURE 7



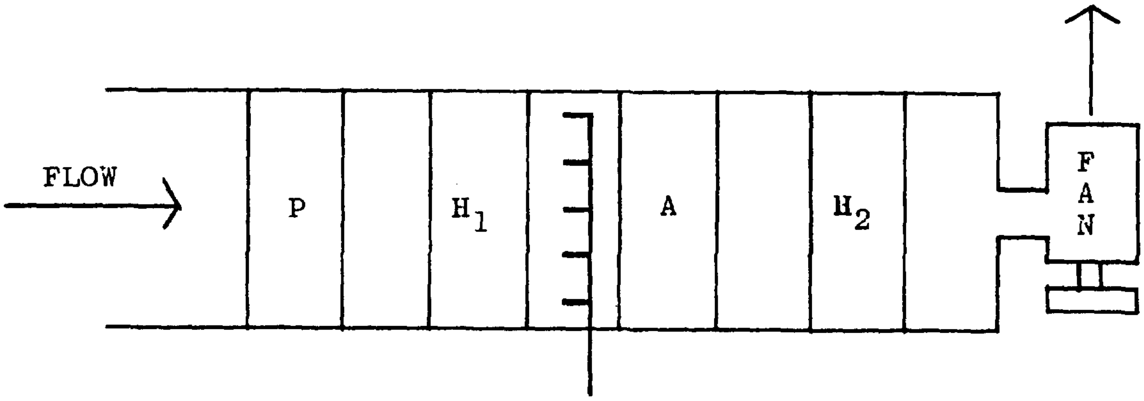
Example of CRM with shaded areas showing a possible set of filters removed.

FIGURE 8



Example of SIM with H_2 under test and H_1 and A not yet installed.

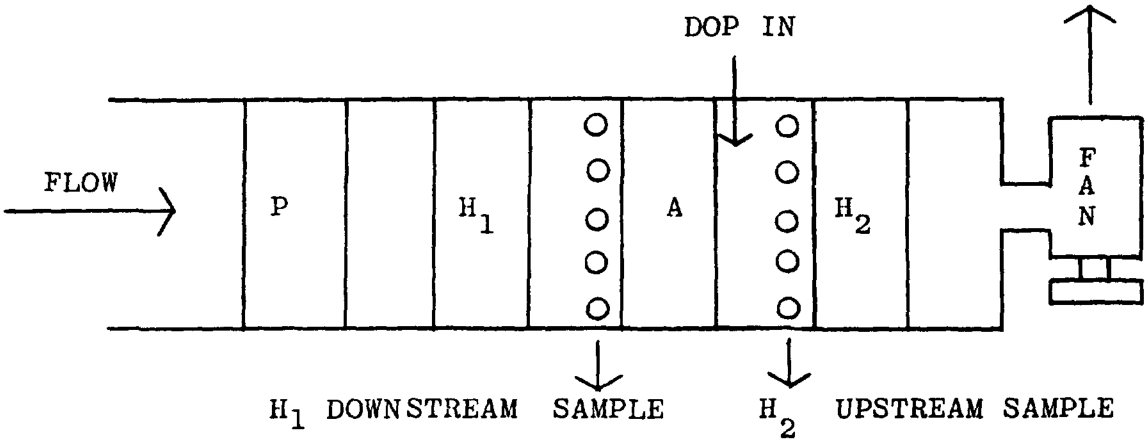
FIGURE 9



H_1 Down Stream Sample

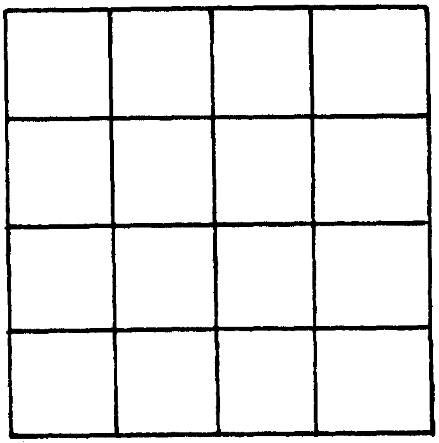
Example of SIM with H_1 under test after H_2 has been tested and accepted. Note sampling for downstream sample must be taken between H_1 and H_2.

FIGURE 10



Example of MSM required downstream of H_1. Note need for DOP injection for H_2 test. A second MSM may be required upstream of H_2 depending on Air/Aerosol Mixing.

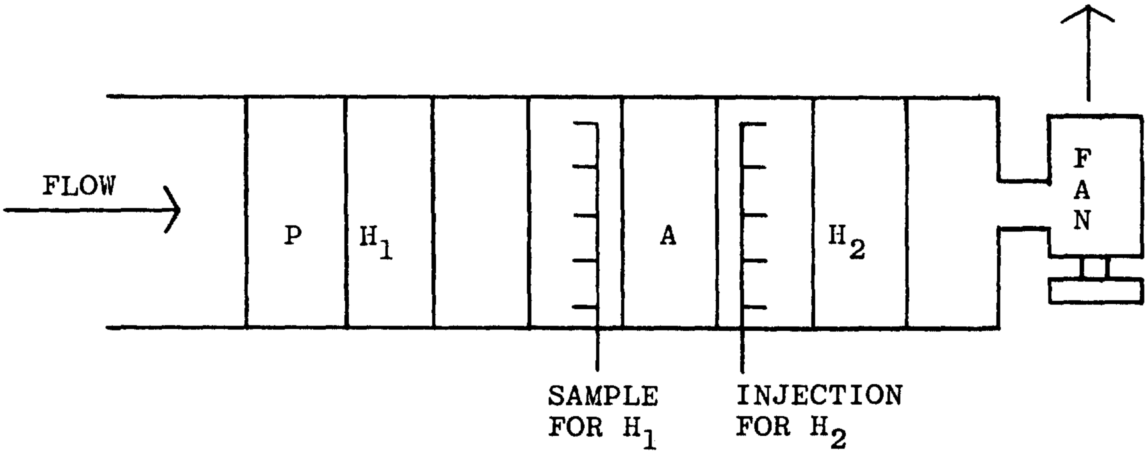
FIGURE 11



AREA APPROXIMATELY
80+ ft^2, HOUSING
PERIMETER 36 ft AND
HEPA GASKET
PERIMETER 128 ft.

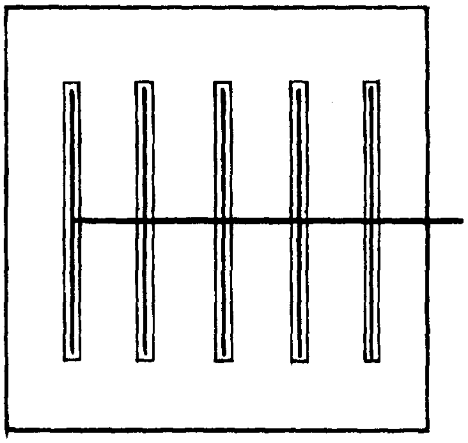
Face of HEPA Bank for MSM. Both the face area and listed perimeters are critical and must be included in MSM test point selection. Laminar flow leaks are easily missed.

FIGURE 12



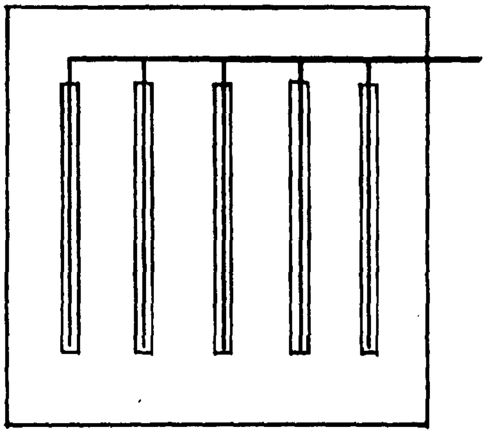
Typical placements of Manifolds. Sample Manifold for downstream sample of H_1 and Injection Manifold for H_2.

FIGURE 13



Typical example of a Manifold used in relation to a Type III Adsorber. Basic configuration is the same for sampling and injection to take advantage of reduced flow area and turbulence.

FIGURE 14



Common alternate Manifold configuration. Usually found on Manifolds added in the field. They may be permanent or removable.

FIGURE 15

DISCUSSION

ORNBERG: I think your examples of selective removal of upstream filters show how the downstream HEPA complicates testing. I strongly urge people to discuss this with their regulatory people to find out if, indeed, they can eliminate testing requirements for the downstream HEPA. It would make it a lot easier to test the upstream HEPA and result in a better system all around.

JACOX: An additional problem with the downstream HEPA is that even when you do have carbon dusting, a HEPA is the worst possible filter to use because it blinds very quickly on the particle sizes of carbon you usually get during dusting. When you have severe carbon dusting, rather than helping, the filter will simply blind, your pressure drop will go out of sight, and you will end up with little or no flow. When there is a carbon dusting problem, a HEPA is the worst filter to use. You should use an 95% NBS filter, instead.

BURWINKEL: Has anybody gone to the black box approach by challenging the entire package as a unit regardless of the number of HEPAs or carbon filters installed?

JACOX: What I interpret you to be referring to is an overall system test. A plant I have been working with is doing that. I am aware by NRC participation in two workshops that it is the intent of the NRC that you shall test the entire system as a blackbox. And that is a good approach.

MILLER: I think he is saying that you can take one sample of the upstream and one sample on the downstream at the very end. Is that what you are agreeing to, Jack?

JACOX: When you have carbon and HEPA filters, you have to use both an aerosol and an halide. From a strictly procedural standpoint, it is definitely the intent and the requirement of N-510 and NRC Regulatory Guides that you have bank tests first to prove integrity on an individual bank basis and then work your way up to the bypass ducts and the system as a whole. The analogy I use is that when you have a primary loop, you don't turn it on to see if it works. You check your valves, check your pumps, check your electricals, check your cutouts. You don't just turn a system on and say "Does it work or not."

ANON: If we eliminate the individual HEPA challenge, what we are left with is a system test.

JACOX: How are you going to show that both a HEPA and an absorber bank are leak tight if you use a single test?

EDWARDS: I think what is being said is that in lieu of testing each bank separately, one might test an entire system with DOP and if it passes, no matter how many filters are in it, it is an acceptable system. Then, one would test the overall system with Freon and if the carbon adsorbers remove the Freon, no matter how many there are in series, the system is acceptable.

JACOX: The Nuclear Regulatory Commission and the CONAGT standard say very clearly that you get no credit for a bank unless it is challenged individually.

EDWARDS: In one of your drawings, you showed that if you remove some filters to challenge the carbon beds you have to do an aerosol uniformity test. You also say that you have to do an air distribution test if the system does not have carbon filters in it. Is an air distribution test a technically reasonable requirement for HEPA filters only?

JACOX: I feel it is technically reasonable and the standards, as well as the NRC, state that it is procedurally required because the justification of residence time on carbon is easier to measure. Some systems that I have seen, at least when they were first tested and before there were modifications made, would have an order of magnitude difference in the flow distribution. I think you would agree that even for HEPA filters, if you have such a flow distribution, you have a pretty poor system, one that is not going to function as intended. You could disagree with the tolerance figure, perhaps, but I don't think you could argue that it is an unnecessary test.

EDWARDS: I disagree technically that a uniform air distribution system is a requirement on a HEPA bank for several reasons. One involves your order of magnitude. If you have 200 fpm in one place and 2,000 fpm in another, very soon the 2,000 fpm filter is going to load up and you will tend to have a fairly even flow distribution. In other words, the HEPA filters are going to correct themselves. When you don't have carbon in the system, it really doesn't make any difference.

CONFLICTS AND MISAPPLICATIONS OF ANSI N509, N510
USNRC REGULATORY GUIDE 1.52 AND 1.140

J. Louis Kovach
Nuclear Consulting Services, Inc.
Columbus, Ohio

I. Abstract

The nuclear industry in the early 1970's attempted to standardize air cleaning system design and testing by the development of ANSI N509, "Nuclear Power Air Cleaning Units and Components" and ANSI N510, "Testing of Nuclear Air Cleaning Systems". Parallel to these and leaning somewhat on the same information, the USNRC has prepared first, Regulatory Guide 1.52, "Design, Testing, and Maintenance Criteria for Atmosphere Clean Up of System Air Filtration and Adsorption Units of Light Water Cooled Nuclear Reactor Plants", and Regulatory Guide 1.140, "Design, Testing, and Maintenance Criteria for Normal Ventilation Exhaust System Air Filtration and Adsorption Units of Light Water Cooled Nuclear Power Plants".

These documents were a good starting point and should have been the basis for the evolution of sound engineering practices. Instead of that path, the subsequent revisions were narrow in scope, uncoordinated, rarely based on experience and became nearly unworkable.

Starting with the scope statement (or equivalent for the regulatory guides) the problems began to occur.

ANSI N509

"This standard covers requirements for the design construction, and testing of the units and components which make up Engineered Safety Feature (ESF) and other high efficiency air and gas cleaning systems used in nuclear power plants".

Thus, there is no system design specification, only air cleaning component and unit design requirements. Requirements are not standards, and as further review of N509 demonstrate may items are "recommendations" and some critical system related specifications are missing.

ANSI N510

"This standard covers field testing of ESF (Engineered Safety Feature) and other high efficiency air cleaning systems for nuclear power Plants and other high efficiency air cleaning systems for nuclear power plants and other nuclear applications." The standard provides a basis for the development of test programs and detailed acceptance and surveillance test procedures, and specifies minimum requirements for the reporting of test results. The standard does not include acceptance criteria except where the results of one test influences the performance of other tests.

Here we are talking about testing systems which are not designed to any standard. N509 requirements are for units and components, but in fact, further review of N510 does not indicate a single "system" test. At best, sections of units are tested instead of systems. Additionally, N510 is taken by both regulatory personnel and utilities as a test specification. Section are copies verbatim, instead of realizing that the scope states that "it is a basis for developing detailed procedures for the testing". As a result, test procedures are applied incorrectly to system testing and the actual application suitability of the particular system is not determined.

Regulatory Guide 1.52

"This Guide presents methods acceptable to the NRC staff...with regard to design, testing and maintenance criteria for air filtration and adsorption units of engineered safety feature (ESF) atmosphere cleanup systems in light-water-cooled nuclear power plants.... It addresses the ESF atmosphere cleanup system, including the various components and ductwork, in the postulated DBA environment".

This is accomplished mainly by referring to ANSI N509, N510 and ORNL-NSIC-65 (ERDA 76-21) the latter being not a standard but a collection of good practices, not a standard.

An example of this type of reference is the following: "Duct and Housing leak tests should be performed in accordance with the provisions of Section 6 of ANSI N510-1975". Unfortunately, there is no duct testing procedure in ANSI N510. Only a housing test is described. A detailed analysis of the Reg Guide shows numerous other conflicts.

Currently the omissions, mistakes, and inconsistencies of these basic documents make any procedure and work subject to various individual interpretations and did not result in good standardized design and/or design verification. As many of these problems as possible will be pointed out to assist those laboring on future revisions of these documents, or preparing test procedures for start-up and surveillance of air cleaning units and systems.

II. Environmental Design Parameters

Regulatory Guide 1.52, Section B, Paragraph A, requires design to DBA conditions as specified in Table 1 where pressure surge is specified for in containment systems as a "result of initial blowdown" and for outside containment as "generally less than primary". This pressure rating resulted in systems being built ranging from ASME Section III to flimsy sheet metal construction as the two extremes. Even the two classifications are inadequate. The pressure surge on a Standby Gas Treatment System can be significantly higher than the pressure surge on a Control Room Recirculation Clean Up System.

In the same section Reg Guide 1.52 specifies maximum pressure for outside of the containment systems as "near-atmospheric" and the inlet relative humidity for all systems of 100%.

ANSI N509 simply states that "design parameters shall be specified when invoking this standard..." The net result is that the environmental design conditions are highly subjective and generally not consistent from plant to plant.

The same Table 1 of Reg Guide 1.52 specifies (for outside of containment systems) elemental iodine inlet concentrations as 10 mg/m^3 and both methyl iodide and particulate iodine as 1 mg/m^3. Aside from the lack of reality in the source term on which these values are based (particularly as ratios of the various forms) the values do not correspond to ANSI N509, RDT M16 or ASTM D3803 loading requirements. Furthermore, Reg Guide 1.52 Section 3.C.i specifies a minimum loading of 2.5 mg total iodine (radioactive & stable) per gram of activated carbon. This value again does not correspond to either ANSI N509, RDT M16 or ASTM D3803 loading conditions. The same paragraph of Reg Guide 1.52 requires that "the radiation stability of the type carbon specified should be demonstrated and certified". There is no standardized test method for this requirement. Many carbons currently sold have never been tested and certified for this requirement and, in fact, the requirement is being ignored by the NRC, the Architect Engineers, the Utilities and many of the carbon suppliers.

Regulatory Guide 1.52, Section B, paragraph 6 states "Average temperature and relative humidity also vary from site to site, and the potential buildup of moisture in the adsorber should also be given design consideration. The effects of these environmental factors on the ESF atmosphere cleanup systems can be determined by scheduled testing during operation". Unfortunately, there are no standard test procedures for the "effects" of "these environmental factors" in ANSI N510 or any site specific "scheduled testing".

Summarizing the Environmental Design Conditions for which the engineered safety feature(ESF) atmosphere clean-up units are supposed to be designed, it can be said that they are inadequately specified in Regulatory Guide 1.52, left to individual design selection in ANSI N509 and not covered adequately (or at all) in ANSI N510. Where certain requirements are specified, the appropriate test methods do not match the requirements.

III. Inter-relation of Test Conditions

Regulatory Guide 1.52 requires "periodic testing during operation to verify the efficiency of the components..." and "the adsorber system should be designed for an average residence time of 0.25 seconds per two inches of bed depth".

The ANSI N510 airflow capacity test acceptance criteria is +/-10%. This is normally performed by pitot tube measurements in ducts which measurement has an error limit of +/-5% to +/-10%. The ANSI N510 Airflow distribution test acceptance criterion within a single bank is +/-20%.

Thus there can be a significant velocity variation within a single air cleaning unit from the design assumption. Airflow will be always highest through the thinnest part of the adsorber and the efficiency of the unit for a contaminent will not follow flow averages. In the Three Mile Island 2 accident, the iodine activity within a single adsorber bank varied by approximately one order of magnitude from top to bottom.

At the same time, the performance of the radioiodine test of samples removed from adsorbers being in use, is performed within +/-4% velocity tolerance and currently the test condition is being restricted to even lower tolerances.

Where is the correlation between the various standards? Or is it assumed that if the laboratory test for the adsorbent is performed under the best controllable conditions, that the obtained test result will, in fact, resemble the performance of an air cleaning unit where the aggregate contaminant flow is variable by +/-30% at any point?

IV. Representative Samples

Regulatory Guide 1.52 states "The efficiency of the activated carbon adsorber section (presumably bank) should be determined by laboratory testing of representative samples of the activated carbon exposed simultaneously to the same service conditions as the adsorber section. Each representative sample should not be less than two inches in both length and diameter and each sample should have the same qualification and batch test characteristics as the system adsorbent... . The design of the samplers should be in accordance with the provisions of Appendix A of ANSI N509-1976..."

ANSI N509-76 states "The superficial velocity of any test canisters shall be shown by calculation or direct measurement to be within +/-20% of the superficial velocity of the adsorber bed." (Same value is permitted in the 1980 version)

This +/-20% is apparently on top of the possible +/-30% variation discussed above, and review of the illustrations attached to ANSI N509-76 shows methods which almost guarantee that the flow through the canisters will be, if anything, -20% compared to bank flow.

The Type 1 test canisters show elbows, reductions and extensive velocity loss in lines. No aerodynamic analysis is required to realize that the velocity will be less than the velocity across the main adsorbent bed. The Type 2 test canister shown in the illustration, was tested by the author and had more than double the pressure drop at the same flow rate of an equivalent size standard adsorber tray.

This type of sampling is being performed on approximately 80% of the currently operating nuclear air cleaning units.

However, ANSI N509-1980 in Table 9-1 Acceptance Test, requires that Adsorber Residence Time is a Field Determination and the "Avg = minimum, design value specified". Typically the residence time is not evaluated on the basis of all of the tolerances permitted.

While Reg Guide 1.52 requires correctly, that the same Batch of adsorbent be used to fill the test canisters, both 1976 and 1980 versions of ANSI N509 requires only that the same Lot be used. Thus the loading of the canisters is not specified, particularly for cases where more than a single batch (approximately 12,000 lbs) is required to load the adsorbent bank.

In this case, Regulatory Guide 1.52, by specifying the illustrated ANSI N509-1976 methodology promotes the use of incorrect engineering.

It is estimated that even if interlaboratory carbon sample testing agreement is reached, the laboratory test results will rarely predict the actual performance of the adsorber unit if designed to ANSI N509 and Regulatory Guide 1.52 conditions.

V. Unenforced and Unenforcable Sections of Reg Guide 1.52

Section C.2.d

"Each component should be protected with such devices as pressure relief valves so that the overall system will perform its intended function during and after the passage of a pressure surge..."

If there is a relief valve, how can the system perform its function while it is venting? If designs exist meeting this criterion verbatim, what detail requirements are they designed to, there is no discussion of this subject in ANSI N509 and no test method in ANSI N510.

Section C.2.j

"Each train should be designed and installed in a manner that permits replacement of the train as an intact unit or as a minimum number of segmented sections without removal of individual components".

The author is not aware of a single unit at any US power reactor which could satisfy this criterion.

Section C.2.1

"ESF atmosphere cleanup system housings and ductwork should be designed to exhibit on test a maximum total leakage rate as defined in Section 4.12 of ANSI N509-1976".

Why should anyone design to exhibit a "maximum" leakage? Furthermore, ANSI N509-1980 states in Table 9-1 that housing test is performed per paragraph 4.12, which states that "The test pressure shall be equal to the design pressure as defined in Par. 4.6.". Par. 4.6.3 states "Design pressure shall be determined by summing the losses in total pressure of all air path components between the open atmosphere and the point in the system under consideration. Losses shall be based on the most severe condition of resistance to rated air flow.". Which pressure is obviously that with dirty filter pressure drop. However, ANSI N510 does not agree that the test pressure is the design pressure because in Section 6.4.2.3 it states that "Start blower and run until pressure in the enclosed space is equal to 1.25 times the system design pressure.".

Section C.3.j

"Adsorber cells should be designed, constructed and tested in accordance with the requirements of Section 5.2 of ANSI N509-1976".

ANSI N509-76 in turn states in Paragraph 5.2.1 "Pleated bed and tray type adsorber cells shall meet the requirements of Type I or Type II cells respectively of AACC CS-8...and in paragraph 5.2.5.1 "A report giving the information specified in Par. C.4 of AACC CS-8...shall be furnished to the purchaser".

In spite of these requirements, tray units are supplied which do not meet the size requirements of the AACC CS-8 and are not interchangeable with standard adsorbers. Filling methods and construction methods are often not qualified according to the AACC CS-8 procedures. As a matter of fact, several types do not even meet the structural test requirements of the CS-8 specification (or its current replacement standard IES-RP-CC-008-83-T).

Section C.3.k

"The design of the adsorber section should consider possible iodine desorption and adsorbent autoignition that may result from radioactively-induced heat in the adsorbent and concomitant temperature rise. Acceptable designs include a low airflow air bleed system, cooling coils, water sprays for the adsorber section, or other cooling mechanism..."

The low flow air bleed presents a significantly higher hazard (that of ignition) than no flow at all.

Cooling coils would increase relative humidity and lower the organic iodide removal efficiency.

Water deluge systems remove the adsorbed iodine, and can cause extensive corrosion if free iodine containing carbon is used.

Therefore, none of the three listed methods would prevent iodine desorption. Fortunately, it is unlikely that the massive amounts of radioiodine would be present to cause overheating of the adsorbent bed.

Section C.3.o

"Straightening vanes should be installed where required to ensure representative air flow measurement and uniform flow distribution through the cleanup component."

Very few systems are in existence where this method is used to ensure uniform flow distribution either with clean or with dirty filters.

Section C.4.c

"The system design should provide for permanent test probes with external connections in accordance with the provisions of Section 4.11 of ANSI N509-1976."

ANSI N509 4.11 states "Where required for proper challenge agent mixing and/or sampling, multiple inlet or outlet distribution manifolds shall be provided to allow injection and sampling per ANSI N510".

Neither of these sections allow shroud testing, removing filters to perform testing or using internal multiple sampling techniques. In spite of that there are many systems installed without required manifolds and, therefore, violating Regulatory Guide 1.52, without any enforcement action.

The performance of test with pre or HEPA filters removed obviously voids any air flow distribution or air aerosol mixing uniformity test, which was performed prior to the removal of those components. Therefore, it generates only numbers instead of meaningful results.

Section C.5.c

"The use of silicone sealants or any other temporary patching material on filters, housing, mounting frames, or ducts should not be allowed."

This is spottily enforced, without complete review and results in great inconsistency in system quality.

VI. Iodine Removal Efficiency Credits

Table 2 of Regulatory Guide 1.52 assumes that all systems are either inside the containment or have humidity control installed in the unit which is not always the case.

A credit of 95% removal is given if adsorbent from a test cartridge (which needs to be equalized only to within +/-20% of the main system velocity) results in less than 1.0% penetration. The velocity effect on the penetration is well known and such latitude is not realistic, particularly when most sampling cartridges have lower velocities than the main adsorber banks (more likely -20% rather than the +20% side).

The same problem exists with Table 2 of Regulatory Guide 1.14 also.

Why tests should be performed at 70% RH for any system sample if there is no humidity control component is not explained.

The exact test conditions for used samples are currently completely non uniform. Earlier issues of the Reg. Guide 1.52 required running tests under DBA temperature, velocity, etc., conditions and early ORNL work stated that worst test conditions were to be 130°C, 95% RH. It is well known now that low temperature at high humidity is a more stringent test conditions, but industry practice has not followed this change in knowledge.

Many plants also miss the somewhat hidden requirement of Reg Guide 1.52 which states that "Similar (to Table 2) laboratory tests should be performed on an adsorbent sample before loading into the adsorbers to establish an initial point for comparison of future test results". This requirement should be clearly listed on requirements to be performed at the time of loading which can be significantly later than when the carbon is purchased.

The wide variety of test conditions - often under unrealistic test parameters - does not permit a comprehensive evaluation of adsorbent performance in the US.

VII. Recommendations

1) Review the overall system requirements and expectations before codifying obsolete or incorrect designs.
2) Evaluate tolerances of design conditions and operation to assure that tolerances are realistic for each step and commensurate with the tolerances of the safety analysis. Require justification of all tolerances.
3) Require proof testing of representativeness of any test canister.
4) Establish correlation between requirements and test methods used to meet the requirements.
5) Write design and test specifications sequentially rather than simultaneously.
6) Both Regulatory Guides 1.52 and 1.14 should be revised to eliminate unnecessary requirements and to contain only critical uniformly enforceable and enforced items.
7) Establish adsorbent test parameters commensurate with adsorbent use conditions.
8) Quantify chemical release effects, requiring retesting.
9) Develop actual system design requirements and overall system tests for air cleaning systems.
10) Eliminate personal interpretation possibilities by clear wording in all standards and Regulatory Guides.

DISCUSSION

GUEST: Given the discussion you just gave us on the impossibility of getting representative samples in these canisters, and the discussions we have heard this week and previously on no laboratories being able to agree on what the chemical analysis of the efficiency of the charcoal is, is it not, perhaps, time to rethink the entire process. Perhaps we shouldn't be testing samples. Perhaps we should be performing in situ tests within the station i.e, the blackbox approach suggested earlier, testing the entire system with radioiodine.

KOVACH: I agree that the entire approach has to be reviewed to make sure that what we are doing matches the safety requirements for the system. Just because we can test in the laboratory to the second decimal, if the sample is 50% non-representative, we are not going to have an adequate evaluation of our system.

Controversial Issues with Air Cleaning at Nuclear Power Stations

Dr. Ronald R. Bellamy
U.S. Nuclear Regulatory Commission
Region I
631 Park Avenue
King of Prussia, PA 19406

Abstract

The guidance documents available for design, installation, and testing of air cleaning systems at commercial nuclear power stations are quite numerous. They have been written and subsequently revised to provide sufficient specificity, yet at the same time allow flexibility to the plant operator. These documents have received continual review and updating by recognized experts in nuclear air cleaning. Nonetheless, issues continually arise that are subject to interpretation, and discussion on these issues sometimes becomes quite controversial. The 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference discussed a number of these concerns that were long-standing. Even with the discussion at that time taking place in an open, public forum, questions still remain on those issues. In addition, new questions continually surface with respect to the applicable guidance.

This paper will provide a forum to attempt to resolve long-standing issues, such as the definition of "significant", with new input and ideas discussed since the 18th Air Cleaning Conference. It will also discuss new issues, to include time between sampling and testing activated carbon, definition and timing of an air cleaning system being out-of-service, best available guidance for laboratory testing of used carbon and resultant technical specification conflicts, and installation of sub-standard HEPA filters.

I. The Definition of Significant

The interpretation of the phrase "significant painting, fire, or chemical release in any ventilation zone communicating with the filter system" is an issue that is still subject to considerable controversy. Although discussed in great detail at the 18th Air Cleaning Conference (1), it is clear that confusion continues to exist. It is important to recognize that the plant technical specifications, and their interpretation by the regulatory authorities, are the controlling documents. There is no question that the performance of activated carbon will be severely degraded for iodine removal by the adsorption of the products of paint or fire, or by chemicals. Therefore, if there are any concerns about how well the carbon might perform, the best guidance would be to run a laboratory radioiodine test. Any decisions made by nuclear power plant staff should be documented with approval by station management.

A better approach, and one many licensees have asked about in the last two years, is to protect the carbon so that the question of whether a laboratory radioiodine test should be run does not arise. Ways to accomplish this include:

1. Turn off the filter system during any painting, and leaving it turned off for days after the termination of painting.
2. Use a scavenger carbon system during the painting that will be discarded after use.
3. Schedule painting just before a test or changeout of the carbon is required for other reasons.

Significant should be interpreted as implying any release that could affect the performance of the filter system. Thus, filter system operation should be planned so that painting does not affect its performance, and if this is not possible, any decision pertaining to whether to run a laboratory radioiodine test should be documented, including the applicable reasoning.

II. Laboratory Testing Criteria

Whenever activated carbon is tested in the laboratory for radioiodine removal, the resultant penetration depends very heavily on the test conditions used: temperature, relative humidity, equilibration, loadings. Of equal if not greater importance is the procedure followed in performing the test. Recent round robin testing is reported in the Panel Session "Nuclear Carbon Test Protocols" as part of this Conference.

Until the results of these recent studies and round robins are complete and additional consensus guidance issued to the industry, licensees should follow their plant technical specifications. This can, however, cause difficulties when it is obvious that the results for the laboratory radioiodine testing are inconclusive, not representative, or misleading. In these cases it is important to realize that the purpose of this test is to verify that the carbon has not degraded to a level where it would not perform satisfactorily in the event of an accident. Therefore, it is incumbent on the licensee to identify such inadequacies, and pursue resolution with the appropriate regulatory authorities.

III. Reactivation and Re-impregnation

Spinster carbon has previously been defined as qualified carbon that has not seen service but has been stored at a site for a number of years. Prior to being put in service, this carbon should be tested to verify that it has not degraded. Inquiries have been made concerning whether this carbon can be re-treated in any fashion to improve its performance, even temporarily. All guidance published to date (specifically ASME Code Sections on Sorbent Media) do not allow reactivation of carbon, irrespective of what filter system will use the carbon. However, these same code sections very clearly allow re-impregnation of carbon that has been in service or of outdated impregnated carbon, so long as the carbon is qualified in accordance with the applicable requirements.

IV. Heating of Service Carbon

Lengthy discussions were held with a licensee in June 1986 concerning the heating of air for purging carbon prior to an in-place freon test. The system in question does not have installed heaters, but the licensee was using a portable heater to heat the air, thus removing atmospheric contaminants and greatly improving the chances of passing the in-place freon test. At first glance this approach sounds like adding a bias to the system and testing the system under conditions that would not approximate those expected during an accident. However, it must be remembered that the in-place test is only for leaks; it is not designed to check the capacity, efficiency, or retentivity of the carbon. The licensee was removing representative carbon samples at the specified frequency to verify the condition of the carbon. In addition, freon testing was being performed at all other specified intervals - maintenance, replacement of trays. Therefore, it was concluded that although the idea of "cleaning" carbon prior to a freon test may not sound logical, it is within the guidelines of plant technical specifications, and is improving the performance characteristics of the carbon.

V. Sub-standard HEPA Filters

Industry consensus over the last 10 years has shown that, although not required by regulation, the use of a DOE operated Quality Assurance Filter Testing Station to visually inspect and dioctyl-phthalate (DOP) test every HEPA filter prior to installation is an excellent industrial practice (2,3). An incident discovered in late 1984 by an NRC licensee and subsequently indicted by a federal grand jury shows the wisdom of using Quality Assurance Filter Testing Stations.

During a routine audit of a vendor by the NRC licensee, it became apparent that non-nuclear (commercial grade) HEPA filters were reported to be nuclear-grade (safety-related). A total of 250 filters were supplied to the licensee and certified as nuclear grade by a local distributor and supplier of HEPA filters. Contrary to this certification, the audit at the vendor's manufacturing plant revealed that these 250 HEPA filters supplied to the distributor by the vendor were not nuclear grade, but had been purchased by the distributor as commercial grade. However, the filters were received by the NRC licensee bearing labels indicating that they were nuclear grade. Documentation supporting the nuclear grade labeling was furnished by the distributor. Ninety-seven (97) of these filters were installed in operating air cleanup systems in the nuclear power plant where nuclear grade filters are required.

Once reported to the NRC by the licensee, in accordance with 10 CFR 21.3(e) as a potential for non-conformance by a supplier of nuclear grade material, the NRC referred the matter to the Department of Justice for possible prosecution against the supplier/distributor. It was clear that the supplier had simply re-labeled the commercial grade HEPAs as nuclear safety-grade. After testimony supplied to a federal grand jury, the supplier/distributor pleaded guilty and was fined.

It is important to emphasize three facts. First, the vendor/manufacturer of the HEPA filters was not involved in any way, and his performance was never questioned. He was asked by the supplier/distributor to provide commercial grade filters, and he did just that. During the investigation the manufacturer/vendor very easily traced the serial numbers for all 250 HEPAs, and readily acknowledged they were commercial grade. Second, no substantial safety concern was created by this deviation. All releases from the plant were controlled, filtered, and monitored at all times, and were always within applicable release limits. Third, the concern was identified and corrected by the "self-police" results of the licensee's audit program at the vendor/-manufacturer.

VI. Out-of-Service Air Cleaning Systems

Plant technical specifications are very clear and specific concerning what constitutes operability of an air cleaning system. The surveillance requirements list specific tests to be performed, the acceptance criteria, and surveillance frequency. If any parameter is out-of-specification, the filter system is defined as inoperable, and a time limit is given within which corrections to the system must be made.

The importance of having operable engineered-safety-feature air cleaning systems was emphasized after an inspection of a Portland General Electric facility (4). Serious concerns arose over the failure to maintain the control room emergency ventilation systems in an operable status over an extended period of time. Inleakage pathways through a condensate drain and housing opening existed, and excessive makeup flow was measured, that would have resulted, under design bases accident conditions, in doses significantly exceeding those specified in General Design Criterion 19 of Appendix A to 10 CFR 50 to the control room operator. The limits specified in these regulations are 5 Rem whole body, 30 Rem thyroid, and 30 Rem skin dose for the duration of the accident. In addition, the two control room emergency ventilation systems were not independent in that a cross connection (common drain line) existed which allowed air flow between each system. To emphasize the importance of maintaining these ventilation systems in an operable status, a Severity Level II violation (Level I being the most severe, Level V being the least severe) was issued, along with a Civil Penalty.

VII. Recent Operating Event

Perry, Unit No. 1, a BWR in Ohio, had a recent malfunction leading to a charcoal fire (5). Since the malfunction occurred during pre-operational testing, no radioactive materials were involved, and the charcoal did not contain any radioactive materials.

The plant's main condensor offgas system consists of two trains connected for series flow, with four tanks in each train. Each tank is approximately 20 feet tall by 4 feet in diameter, containing 6 tons of charcoal. The licensee was heating the rooms containing the charcoal tanks with space heaters on June 20, 1986, for a startup test of the HVAC system for the rooms. This test required an initial air temperature of 150°F. Although the offgas charcoal was not in service during the HVAC system test, thermocouples in the center of the charcoal in two tanks (one in each train) measured between 1100°F and 1400°F. The tank surface temperature remained below 150°F.

Room air returned to normal temperature as the HVAC test was stopped. Nitrogen (25 cfm) was used to purge the tanks of free oxygen, however, residual oxygen kept the charcoal burning (smoldering) for days, and although the temperatures decreased, the maximum measured charcoal temperature on June 23, 1986 was 324°F. The tanks have been opened and the charcoal is being analyzed.

The source of heat leading to the high charcoal temperatures appears to be the placement of the space heaters too close to the charcoal tanks. Once the charcoal began smoldering, it was difficult to extinguish the charcoal. An attempt at a retest on July 6, 1986, resulted in further elevated temperatures (700°F) being observed in the charcoal tanks, simply by blowing 80°F instrument air over the charcoal.

VIII. Summary

This presentation has discussed those air cleaning issues that have led to some confusion, discussion, and even disagreements since a similar presentation at the 18th Air Cleaning Conference. Its purpose was not to give definitive guidance, but to allow the opportunity for discussion and input from all interested parties. Consensus standards are the best source of guidance, but even these are subject to interpretation. It is clear that nuclear air cleaning is still an evolving art, and concerns that arise need to be discussed to achieve the best possible guidance and clarification.

REFERENCES

1. Bellamy, Ronald R., "Regulatory Experience With Nuclear Air Cleaning," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF - 840806, pp. 897-903.

2. Burchsted, C. A., "A Review of DOE Filter Test Facility Operations," Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF - 820833, pp. 775-787.

3. Allan, Thomas T., and Cramer, Robert V., "Evaluation of HEPA Filters Meeting MIL-F-51068 Purchased on the Open Market. Are They Nuclear Grade?", Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF - 820833, pp. 1115-1120.

4. Martin, John B., "Notice of Violation and Proposed Imposition of Civil Penalty (NRC Inspection Report No. 50-344/86-06)," to Portland General Electric Company, May 14, 1986.

5. Stello, Victor, USNRC Executive Director for Operations, Daily Staff Notes, June 23, 1986.

DISCUSSION

ORNBERG: What are your comments regarding requirements for a downstream HEPA, since it is mentioned in the Regulatory Guide 1.52? 2) Comment also on the substandard filters. I think the biggest potential problem exists with the non-ESF units because those filters are required to be just like the ESF filters, yet many purchasing agents, when they see they are non-ESF systems, figure that they don't need any quality assurance at all. I think there is more potential there for getting the commercial-grade filters. The last question is, whether you or any other NRC member has any comments to make about when, or if, Regulatory Guides 1.52 and 1.140 are going to be revised?

BELLAMY: I will start with your second comment and say, I agree with it. Addressing your question on the downstream HEPAs, I would say that the basis for the Regulatory Guide was a perceived lack of confidence in some of the installers in the field and the fact that I, personally, viewed maintenance individuals having lunch inside the housings. After testing was done, they would walk around inside with wrenches in their back pockets and put holes in the HEPA filters. They didn't know what a HEAP was. Because the filters have fragile components, we figured it made sense to have a redundant downstream bank of HEPA filters. Is it still necessary today? You can make a good case that it is not. The recommendation that it be taken out of N509 and all future codes is one I can agree with. I think that is the statement you were looking for. Getting back to the previous blackbox discussion on testing an entire system, it is important to realize that, to the NRC, it doesn't matter how many banks of HEPA filters you have, one or many, the credit given for particulate removal during an accident will be the same.

ORNBERG: For people with downstream HEPAs installed now, what do you suggest they do regarding discussions with the NRC about continuing to test them.

BELLAMY: I suggest that the filters be left in, but that a case be made to treat the system as a blackbox and test the two banks simultaneously.

Regarding your third question, I have been pushing to get the Regulatory Guides revised. I am not going to stand up here and tell you the 1978 issues are perfect. Obviously they are not perfect and they are not even adequate in a lot of cases. We have figured how much it would cost us to revise one of the Regulatory Guides. The point I am going to make is that the money to do this is not in the NRC's budget. There are no plans that I know of to initiate a revision to the Regulatory Guides. What I am looking for is someone or some group willing to volunteer their time to revise the Regulatory Guides for us.

JACOX: The comment you made about a blackbox test seems to be a fairly radical change from the previous NRC/DOE position. When you have a system where you have one HEPA bank only, I can see some technical basis for it, but I know there are a number of systems, one in a plant that is still to be started up, where they have charcoal banks in series. Would you also apply the blackbox test to charcoal in series. You could have one that is leak tight when the other leaks grossly.

BELLAMY: The answer is, yes, I would.

JACOX: Well then, how can you justify the need or the existence of the second bank? If one leaks and one doesn't, the one that leaks doesn't exist for practical purposes, so how do you justify having it in?

BELLAMY: I would think that the visual test would point out a lot of the potential gross leakages. Certainly, a molecule that has passed through the first bank of carbon doesn't know that there is a space between the first bank and the second bank. So, for this purposes two two-inch banks would be the same as a four-inch bank. There could be some inconsistences with gross bypass, but that is the purpose of the visual test, to catch such a defect.

JACOX: I agree that a good visual inspection catches most of the defects. I disagree with the blackbox approach. Will you cite specific technical data that constitute a basis for allowing reimpregnation, but not allowing reactivation, I find this a highly difficult differentiation to understand.

BELLAMY: We have had a lot of discussions on reactivation versus reimpregnation in the code work that we have done. For ESF systems, in particular, I have taken the position that it is a nickel/dime item whether you reactivate the carbon or not. It doesn't make any sense to me to reactivate the carbon. I think, in the overall operating and maintenance cost of the filter system, the cost of new carbon versus the cost of possibly reactivating and reimpregnating it just doesn't make enough sense to go ahead with the reactivation process. I know there are some of my learned colleagues here today who do not agree with that point. In the interest of publishing an adsorber code section, we negotiated and compromised. We agreed that the reactivation would not be allowed in the present code section but that we would very actively and quickly try to come to some happy medium on exactly what might be allowable for reactivation. Reimpregnation, I think, has a basis in the fact that it is permitted for spinster carbon (a term that you really should take credit for, Jack.) This means that if a carbon has never been used, but its impregnation has deteriorated, you can come to the conclusion that it is financially beneficial to go ahead and put a second impregnant on it and then run the whole gamut of tests. If it then passes all the tests, as a regulator, I cannot tell you it is not allowable to use that carbon in the filter system. You might say I am talking out of both sides of my mouth because you could use a similar argument to permit reactivation. I think it is just a philosophical point, reimpregnation is one step, reactivation is a second step. This is where we draw the line for the present. I know that is not going to give you a warm feeling but that is the best I can do.

RIGBY: My question relates to the last question. I think that, quite frankly, reimpregnation is full of dangers. You are into the stage of what is a spinster carbon and how that can be developed and changed. I am sure that one would think that, in the industry that we are talking about, the utilities, we are not really in the secondhand car market.

BELLAMY: My answer to the original question was more from a regulatory NRC concern. Now, I have to agree with you 100%. As an engineer, I would say that reimpregnation is not a recommended thing to do. If I worked for a utility, I would very strongly push to prevent reimpregnated carbon being used on my site. I personally would not recommend it.

REACTIVE INSPECTION RESPONSE OF NRC REGION III TO POTENTIAL TECHNICAL DEFICIENCIES IDENTIFIED IN RECENT NUCLEAR AIR CLEANING CONFERENCE PAPERS

C. F. Gill
U.S. Nuclear Regulatory Commission/Region III
Glen Ellyn, Illinois

ABSTRACT

In order to effectively meet its responsibility to protect the public health and safety, the Nuclear Regulatory Commission (NRC) nuclear power plant licensing and inspection programs respond to potential technical deficiencies identified by conference and professional society meeting papers when deemed appropriate. The NRC staff's response mechanisms for such technical deficiencies include: generic letters, Bulletins, Information Notices, Standard Review Plan (NUREG-0800) revisions, docketed Final Safety Analysis Report (FSAR) questions, special studies, special (reactive) inspection, and inspection program revisions. This paper describes reactive inspection efforts by Region III in response to potential technical deficiencies identified in recent air cleaning conference papers, including: post-accident effluent sample line deposition losses; failure to implement good engineering practices in the design, construction, and testing of Nuclear Air Treatment Systems (NATS); filter bypass via filter housing drain lines; spinster carbon degradation; use of silicone sealants and other temporary patching material in NATS; filter housing fire protection deluge system problems; lack of charcoal batch traceability; Quality Assurance records problems involving equipment, vendor, filter, and personnel qualifications; inadequate ANSI/ASME N510 acceptance criteria and tests; and failure to adequately demonstrate control room habitability per 10 CFR 50, Appendix A, General Design Criterion-19. Region III inspections indicate that many of these deficiencies appear to be prevalent. Inspection findings and utility responses to the findings are discussed. NRC Region III and Headquarters programmatic reactions to the identified generic problem areas are also discussed.

I. INTRODUCTION

Each year potentially generic technical deficiencies of significance to the nuclear industry are identified in technical journal articles and conference and professional society meeting papers. Once the NRC becomes aware of these reported deficiencies, the staff reviews each item and responds as deemed appropriate by management. The NRC staff's response mechanisms include: generic letters, IE Bulletins and Information Notices, Standard Review Plan (NUREG-0800) revisions, docketed Final Safety Analysis Report (FSAR) questions, special studies, special (reactive) inspections, and inspection program revisions. This paper deals specifically with the reactive inspection response of the NRC Region III to potential technical deficiencies identified in recent Nuclear Air Cleaning Conference papers.

The 17th (1982) and 18th (1984) Nuclear Air Cleaning Conference proceedings published a number of papers which the NRC Region III and the Office of Nuclear Reactor Regulations (NRR) felt might represent potentially generic technical deficiencies. The areas of concern included post-accident effluent sample line deposition losses; failure to implement good engineering practices in the design, construction, and testing of Nuclear Air Treatment Systems (NATS); filter bypass via filter housing drain lines; spinster carbon degradation; use of silicone sealants and other temporary patching material in NATS; filter housing fire protection deluge system problems; lack of charcoal batch traceability; Quality Assurance records problems involving equipment, vendor, filter, and personnel qualifications; inadequate ANSI/ASME N510 acceptance criteria and tests; and failure to adequately demonstrate control room habitability per 10 CFR 50, Appendix A, General Design Criterion-19. In order to determine the extent of these problems, the Chief, Meteorology and Effluent Treatment Branch, NRR, and the Chief, Emergency Preparedness and Radiological Protection Branch, Region III agreed that Region III should conduct regional directed (reactive) inspections at two Near Term Operating License (NTOL) commercial nuclear power plants. Based on the inspection findings at the first two NTOLs, this special inspection program was extended to include the other three Region III NTOLs and several operating plants. These inspection findings were shared with other NRC regional offices, the Office of Inspection and Enforcement (IE), and NRR.

NTOL HVAC acceptance test programs receive review by several groups of regional inspectors, including: Test, Quality Assurance (QA), Resident, Fire Protection, and Facilities Radiation Protection Section (FRPS) inspectors. Although each of these inspection groups reviews a portion of each NTOL applicant's HVAC preoperational program for compliance with regulations and commitments in accordance with existing standard inspection procedures (modules), none of these procedures specifically addresses the potential deficiencies discussed in recent Nuclear Air Cleaning Conference papers. Therefore, in addition to the completion of the scheduled inspection modules, the FRPS was assigned to conduct special (reactive) inspections of Region III NTOL's to determine the extent to which the reported deficiencies existed at these facilities.

Region III generally encouraged the applicants to conduct a compliance review to identify and correct any potential regulatory violations before the NRC inspectors formally began the inspections. The FRPS inspectors initially met with applicant representatives to: (1) determine the status of the ANSI/ASME N510 acceptance test program; (2) inform them of the types of documents which should be available onsite for NRC inspector review; (3) discuss programmatic deficiencies discovered at other NTOL plants; and (4) request that spinster carbon be laboratory retested, an ANSI/ASME N510 acceptance test compliance analysis be prepared, and the use of silicone sealant on HVAC ductwork and filter housings be evaluated.

The types of documents which were reviewed during these special inspections included: (1) acceptance test inspector qualification records; (2) applicant quality assurance vendor audits; (3) HEPA filter and charcoal adsorber qualification documents; (4) applicant ANSI/ASME N510 test acceptance criteria; (5) acceptance test procedures and reports; (6) HVAC and ANSI/ASME N510 acceptance test program commitment and compliance documentation; (7) deficiency tracking system documentation; and (8) applicable HVAC surveillance and operational procedures. The inspectors also interviewed applicant and contractor personnel to assure that proper use was being made of the applicant's deficiency tracking systems and that all identified design, test, and procedural discrepancies were satisfactorily resolved. The review of the HVAC systems also included physical inspections of ductwork, filter housings, housing drainage systems, and fire protection water deluge systems to assure compliance with design and construction commitments to regulatory guidance and industry standards.

Although an applicant's ANSI/ASME N510 acceptance test program was not usually developed sufficiently to warrant detailed NRC inspector review during the first site visit, the inspectors met with applicant representatives to discuss programmatic deficiencies discovered at other NTOLs. Potential deficiencies discussed included: (1) performance of tests by uncertified personnel; (2) unresolved vendor audit findings and observations; (3) inadequate HEPA filter and carbon adsorber qualification records; (4) lack of carbon adsorber batch traceability; (5) lack of a deficiency formal reporting and resolution tracking system; (6) misuse or lack of adequate test acceptance criteria; (7) inadequate timing of visual inspections; (8) significantly degraded "spinster" carbon; (9) lack of detailed compliance with N510 test procedure and report specifications; and (10) improper use of silicone sealants. Subsequent NRC inspections were conducted to review applicant documentation, interview personnel, and inspect installed HVAC systems to assure compliance with regulations and commitments.

In response to Region III requests, applicants for operating licenses generally prepared two documents to track compliance of their ANSI/ASME N510 acceptance test programs. These documents were internal reports made available for NRC review. The first report was a commitment and compliance analysis which provides a detailed (line-by-line) identification of each commitment associated with ANSI/ASME N510, addressed reviews of QA qualification documentation and specific potential HVAC system or acceptance test deficiencies, ascertained compliance with regulations and commitments, identified corrective measures needed or variance requests required, and identified actions needed to document compliance. The second report was a detailed action plan providing a tracking system for actions needed to comply with commitments and regulations, for design deficiencies and other discrepancies and their resolution, and to document compliance. This report was usually detailed enough to include specific tasks, individuals assigned to each task, a schedule for completion, and a periodically updated status for each action item.

All of the Region III NTOL applicants identified a number of apparent violations of regulations and deviations from commitments. Under the provisions of 10 CFR 2, Appendix C, the NRC does not usually issue notices of violation when the violation is licensee or applicant identified and the corrective actions are deemed adequate, if the other criteria of Appendix C are also met. The following example is typical of the results of an applicant's ANSI/ASME N510 commitment and compliance analysis. The applicant's ANSI/ASME N510-1980[1] line-by-line commitment and compliance analysis was divided into 55 Engineered Safety Feature (ESF) commitments (Regulatory Guide 1.52[2]) and 55 non-ESF commitments (Regulatory Guide 1.140[3]). The applicant also reviewed QA qualification documentation and specific potential deficiencies based on discussions with NRC Region III personnel. Approximately 180 action plan tasks were assigned, 35 variances from ANSI/ASME N510 specifications were noted, acceptance test criteria and procedures were modified, FSAR amendments were issued, operational and surveillance procedures were revised, HVAC and fire protection deluge systems were modified, and some technical specifications were redrafted. Technical deficiencies discovered included lack of QA documentation on charcoal adsorber batches and HEPA filters, improper vendor charcoal adsorber qualification tests, lack of carbon adsorber batch traceability, lack of detailed compliance with ANSI/ASME N510 acceptance test specifics, improper use of silicone sealants, and filter bypass via filter housing drainage systems. The applicant initiated adequate corrective actions for the delineated deficiencies by use of existing deficiency reporting and resolution tracking systems.

II. FINDINGS

Based on the reactive inspection of four of the five Region III NTOLs and a partial review of several operating nuclear power plants, it appears that at least some of the potential technical deficiencies identified in recent Nuclear Air Cleaning Conference papers are prevalent. The findings for each identified potential deficiency will be presented separately.

Post Accident Effluent Sample Line Deposition Losses

In 1982 Kabat[4] reported deposition velocities for various species of iodine for a variety of sample line materials. These deposition velocities were used by the NRC to obtain the concentration reduction factor results in Table 1 for a certain station's auxiliary building vent stack post-accident effluent monitoring system. Because the high range sample line losses were found to be excessive by NRR, the licensee has agreed to modify the system (before startup after the first refueling outage) to greatly reduce the iodine deposition potential. NUREG-0737[5], Item II.F.1, Attachment 2 specifies that representative samples are to be obtained from post-accident effluent sampling systems. As stated in Footnote 14 of Table 3 of Regulatory Guide 1.97. Revision 3[6], "collection of representative samples" means obtaining the best samples practicable given the exigencies that attend the accident environment; line losses or line deposition should be empirically predetermined and appropriate loss correction factors should be applied. This statement from the regulatory guide is utilized as further clarification of the NUREG-0737 requirement by the NRC. As a result of this interpretation, Region III NTOL applicants had their operating licenses conditioned by the requirement to empirically predetermine post-accident effluent sample line losses when supplied by NRR with the criteria for an acceptable methodology. NRR has contracted Battelle Pacific Northwest Laboratories and the Idaho National Engineering Laboratory (INEL) to aid in the development of guidance for iodine sample line loss determination. It is noted that INEL and NRC representatives will be presenting a paper at this conference regarding this matter[7].

Table 1. Calculated iodine deposition factors for a certain station's auxiliary building vent stack low and high range sampling lines.

EXPOSURE CONDITIONS			EXPERIMENTAL PARAMETER	LOW RANGE SAMPLING LINE CALCULATED IODINE LOSS			HIGH RANGE SAMPLING LINE CALCULATED IODINE LOSS		
Iodine Form	Relative Humidity % (±3%)	Surface	Deposition Velocity Vg (m/s)	Deposition per length Du (m^{-1})	Total Conc. Reduction Factor	D_L, Total Deposition Fraction	Deposition per length Du (m^{-1})	Total Conc. Reduction Factor	D_L, Total Deposition Fraction
I_2	5	N.Cl.	1.8(-4)	1.367(-2)	1.72	.418	1.268(-1)	1.52(+2)	.993
		Ch.Cl.	8.7(-4)	6.606(-2)	1.37(+1)	.927	6.129(-1)	3.53(+10)	1.00
	97	N.Cl.	1.6(-3)	1.215(-1)	1.23(+2)	.992	1.127	2.50(+19)	1.00
		Ch.Cl.	2.0(-3)	1.519(-1)	4.11(+2)	.998	1.409	1.76(+24)	1.00
HOI	5	N.Cl.	4.0(-6)	3.037(-4)	1.01	.0120	2.818(-3)	1.12	.106
		Ch.Cl.	3.3(-5)	2.506(-3)	1.10	.0945	2.325(-2)	2.51	.602
	97	N.Cl.	1.8(-5)	1.367(-3)	1.06	.0527	1.268(-2)	1.65	.395
		Ch.Cl.	4.4(-5)	3.341(-3)	1.14	.124	3.100(-2)	3.42	.707
CH_3I	5	N.Cl.	1(-7)	7.593(-6)	1.0003	.0003	7.045(-5)	1.003	.003
		Ch.Cl.	7(-8)	5.315(-6)	1.0002	.0002	4.931(-5)	1.002	.002
	97	N.Cl.	8(-8)	6.075(-6)	1.0002	.0002	5.636(-5)	1.002	.002
		Ch.Cl.	8(-8)	6.075(-6)	1.0002	.0002	5.636(-5)	1.002	.002

Where:

N.Cl. = non-cleaned surface

Ch.Cl. = chemically cleaned surface

Vg = deposition Velocity of Stainless Steel (exp)

Du = deposition of iodine per unit length (exp) = $\frac{(\pi d)\ Vg}{F_s}$

e^{Du*L} = total iodine concentration reduction factor

d = inside diameter of sample tubing = 1/4" and 3/4" for High and Low range, respectively

Fs = sample line volumetric flow rate = .06 and 1.67 cfm for High and Low range, respectively

L = length of sample line = 130 feet

D_L = $1 - e^{-Du*L}$ = total iodine deposition fraction

In 1984, the methodology for experimental determination of the transmission of radioiodine through sample lines was apparently advanced as reported by Unrein, Pettetier, Cline, and Voilleque[8]. Although the deposition velocities determined earlier by Kabat[4] are reaffirmed by this paper, the more recent experimental program also investigated the effects of resuspension. It was found that over a period of time the radioiodine transmission factors improve significantly over that predicted by deposition velocity alone. Although this more recent data indicates that equilibrium iodine transmission may be much higher than originally predicted, it also indicates that the transmission factor changes over a period of time (until equilibrium is reached) and the transmitted iodine may have changed chemical species and undergone significant radiodecay. This variation of transmission characteristics and chemical species with time will make the development of regulatory guidance for post-accident empirical predetermination methodology even more difficult.

Failure to Implement Good Engineering Practices

At the 17th DOE Nuclear Air Cleaning Conference, Moeller and Casper Sun[10] presented a paper which analyzed failures in air-cleaning, air-monitoring, and ventilation systems in commercial nuclear power plants based on data gleaned from Licensee Event Reports (LERs) submitted from 1978 through 1981. It was concluded by the authors that although information needed to prevent and/or correct such failures is available, it is not being effectively utilized. The authors also concluded that the primary reason for the failures is a shortage of personnel who are knowledgeable about HVAC systems, in general, and about nuclear air cleaning technology, in particular. To correct this situation, the authors recommended an increase in the training of both nuclear power plant personnel and members of the NRC staff in the subject areas. The NRC Region III special inspection effort regarding potential HVAC deficiencies is, in part, in response to this paper. The NRC Region I training efforts in response to nuclear air cleaning concerns were well expressed by Dr. R. R. Bellamy in the discussion portion of Panel 13, "Nuclear Air Cleaning Field Experiences," of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference (see Pages 923-934 of the proceedings).

The lack of adequately trained personnel seems, however, to account for only part of the nuclear air cleaning system failures. As pointed out by Moeller and Casper Sun[9], LERs are written to address mostly problems which represent violations of Technical Specifications. In order for the NRC staff to properly assess problems associated with air cleaning systems, data supplemental to information contained in LERs must be obtained. The papers presented at the Nuclear Air-Cleaning Conferences is a good source of that supplemental data. Additional information of this type was suppled by Jacox[10,11] who indicated that although lack of sufficient training and knowledge concerning Nuclear Air Treatment Systems (NATS) is a generic industry problem, failure to implement good engineering practices in the design, construction, and testing of NATS is also prevalent. The effect of poor engineering practices on HVAC system and equipment reliability is well documented by conference papers including those by Kovach[12] and Graves, Hunt, Jacox, and Kovach[13] from the proceedings of the 13th and 15th Nuclear Air Cleaning Conferences, respectively.

During the NRC inspector review of the Region III NTOL ANSI/ASME N510 acceptance test programs, many of the examples of poor engineering practices described by Moeller and Casper Sun[9], Jacox[10,11], Kovach[12], and Graves[13] were noted. These examples included: damper failure or excessive leakage; other HVAC system ductwork under positive pressure which passes through the Control Room gas control envelope; use of silicone sealant and other temporary patching material on ductwork and filter housings; hydrogen explosions and fires in BWR offgas systems; poor cooperation between design engineering, system engineering, startup test engineering, and operations; significant difficulty of low quality ductwork and filter housings to pass ANSI/ASME N510 acceptance test criteria; improper interpretation of regulations and standards; improper QA documentation and qualification tests of charcoal adsorbers and HEPA filters; lack of adequate provision of testing manifolds for series filter banks; floor drain check valves installed so that the filter housing doors cannot be opened far enough to change out filters; and other examples of poor engineering practice which appear elsewhere in this paper.

Regional NRC inspectors are, in general, restricted to assuring that an applicant for an operating license complies with regulations and commitments. The regional inspector, however, is aided by technical and regulatory guidance from NRR and inspection program guidance from I&E. If an applicant meets the requirements of the relevant regulations and commitments, it is difficult for the NRC inspector to additionally require that generically recognized good engineering practices be followed. Thus, although nuclear standards, codes, and regulations are meant to be additional requirements over and above the use of good engineering practices, all too often it appears that NATS are designed, constructed, tested, and operated by applicants and licensees who take full advantage of their minimal commitments to standards, codes, and regulatory guidance. Although not readily able to directly require the application of good engineering practices, NRC inspectors do assure that the applicants' deficiency reporting systems are properly utilized, management controls are adequate, and all allegations are thoroughly reviewed and resolved.

Filter Bypass Via Housing Drains

Regulatory Guide 1.52, Revision 2, March 1978[2], (Regulatory Position 3.h) and Regulatory Guide 1.140, Revision 0, March 1978[3], and Revision 1, October 1979[14], (Regulatory Position 3.e) state that the filter housing water drains should be designed and constructed in accordance with the recommendations of Section 4.5.8 of ERDA 76-21[15] and Section 5.6 of ANSI N509-1976[16], respectively. These recommendations include individually valving, sealing, or otherwise protecting drain lines from individual chambers of the housing to prevent bypassing of contaminated air around filters or adsorbers through the drain system. At the 17th Nuclear Air Cleaning Conference, Bellamy[17] described how filter and system bypass via housing drains was discovered at the TMI Station in early 1982. He considered this incident a problem of potential generic applicability.

During the special Region III reactive inspection program regarding potential generic HVAC deficiencies, filter bypass pathways via housing drain systems were found at all five NTOLs and many of the operating plants which were reviewed for this deficiency. As an example, a recent inspection at an operating plant revealed that out of 19 filter housings inspected, only two had drainage systems which precluded filter bypass. The most common deficiencies at this plant were drain lines connected to a common header without isolation valves in each drain line and uncapped drain lines left open to room atmosphere. Even for those housings which had valves or loop seals in each uncapped drain line, in general, applicants and licensees usually did not have adequate administrative controls on use of isolation valves and loop seals, air leak-tightness of isolation valves had never been verified, either water check valves had not been a design consideration or installed water check valves were of inadequate design, loop seals were also often of inadequate design, and significant potential existed for either allowing loop seals to dry-out or to inadvertently flood the housing by overfilling or by drawing water out of the loop seal during system startup. The Region III licensees and applicants for operating licenses all initiated adequate corrective actions when informed by NRC inspectors that filter housing drain systems or administrative controls are not adequate.

Spinster Carbon

Partly because of significant delays in the startup dates for many reactors, qualified carbon has been in storage at some sites for five years or more. This unused carbon is commonly referred to as "spinster" carbon. Due to the lengthy storage times, spinster carbon may be significantly degraded by the time it is used and therefore may have to be retested to verify adequate retention of performance characteristics. The amount of degradation depends on many factors, including: storage period; damage due to handling, moving, and storage techniques; packaging methods; and exposure to contaminants. Hubbard[18] states that unused carbon is generally expected to have a shelf life (when properly sealed in storage) of three to five years when it can meet the specifications of new carbon. Jacox[11] points out that although carbon may be several years old before it is used in a system, it should meet all the requirements of new carbon, but this is nowhere stated in standards or regulatory guidance. Bellamy[19] recommends, as a rule of thumb, if the carbon was stored properly, it probably need not be retested if the storage time is one or two years or less. He also states that if storage approaches five years, retesting should be performed.

All the Region III NTOLs were asked to voluntarily test spinster carbon using the following guidelines.

- If the carbon has been properly stored, it probably need not be retested if the storage time is 18 months or less. Retesting should be considered for longer storage times and if storage approaches five years, retesting should definitely be performed.
- Batch samples should be tested with methyl iodide to Regulatory Guide 1.52, Table 2 or Regulatory Guide 1.140, Table 2 (as appropriate) acceptance criteria. The carbon should be replaced if it fails the prescribed test.

All Region III NTOLs agreed to test spinster carbon which had been stored for longer than 18 months. For the four NTOLs which have completed this test, about one-half of the carbon stored longer than five years and less than 10% of the carbon stored less than five years failed the laboratory tests. Although given the poor quality of the recent roundrobin carbon adsorber test results, as reported by Miller[20], First[21], Kovach[22], and Bellamy[23], the significance of these spinster carbon tests is uncertain. It should also be noted that the Region III spinster carbon which had been stored greater than five years was all at one NTOL and was purchased approximately ten years before the laboratory retest. Region III NTOL applicants who failed spinster carbon tests have discarded the failed charcoal adsorber batches except for one batch for which special permission for use was granted by NRR.

Use of Silicone Sealants

Regulatory Guide 1.52, Revision 2 (March 1978[2]), Regulatory Position 3.n states that ESF ductwork should be designed, constructed, and tested in accordance with the Section 5.10 of ANSI N509-1976[16]. ANSI N509-1976, Subsection 5.10.4 states that longitudinal seams shall be either all welded, seal welded mechanical, or in accordance with SMACNA - High Velocity Duct Construction Standards (Pittsburgh Lock or Acme Lock Seam) as required to meet structural and leak-tightness requirements of Pars. 5.10.3 and 4.12, respectively. ANSI N509-1976, Subsection 4.12 states that the allowable leakage will, by reference to Par. 4.12.3, indicate the required type of duct construction; i.e., welded or nonwelded; however, ducts for ESF systems and all housings shall be welded. It should, however, be noted that Subsection 4.12 of ANSI/ASME N509-1980[29] allows welded or flanged transverse joints and mechanical lock type longitudinal seams for ESF ductwork.

Regulatory Guide 1.52, Revision 2 (March 1978), Regulatory Position 5.c states that the use of silicone sealants or any other temporary patching material on ESF filters, housings, mounting frames, or ducts should not be allowed. Regulatory Guide 1.140, Revision 0 (March 1978[3]), and Revision 1 (October 1979[14]) Regulatory Positions 3.f and 5.c have the same wording as Regulatory Guide 1.52, Revision 2 (March 1978), Regulatory Positions 3.n and 5.c, respectively (which are discussed above).

Contrary to the above regulatory positions and to the applicants' commitments to the regulatory guides, Region III inspectors have identified the use of silicone sealant at all five NTOL's and one operating plant. Common uses of silicone sealant to pass ANSI/ASME N510 leakage tests includes application on ESF and non-ESF mechanical lock longitudinal seams inside sheetmetal ductwork, external longitudinal and transverse ductwork seams, between stitch welds on companion angle flanges, companion angle flange gaskets, installation of instruments into ductwork, and on non-ESF housings. After the use of silicone sealant was identified at each plant, the matter was referred to NRR for resolution.

The responses of NRR to the use of silicone sealants and other temporary patching material at Zion[24] and Clinton[25] are a matter of public record. In both cases, NRR issued general guidance on the use of temporary sealants and patching materials in air cleaning systems. This guidance states that while the quality of some of these sealants has improved in recent years, the NRR staff has not yet accepted such material as being good for the life of the plant; thus it is the staff's position that these materials can be expected to degrade over a period of years and may result in unacceptably high leakage in ductwork or filter housings. The guidance also states negative pressure ductwork outside the Control Room gas control envelope is of particular concern. Several alternatives are available if an applicant for an operating license has used temporary sealant on this ductwork including replacement of the sealant with welded joints or a leakage testing program which would establish the long-term integrity of the sealant or patching material used. Other ESF and non-ESF air cleaning systems which incorporate these sealants in construction or leakage repair are evaluated on a case-by-case basis. The NRR resolution of each plant's use of sealants on air cleaning systems has resulted in license conditions or technical specification modification.

Fire Protection Deluge System Problems

During their review of LERs submitted from 1981 through 1983, Moeller and Kotra[26] noted that unintentional actuation of fire suppression systems was reported at several nuclear power plants during the three-year study period. This rendered the filters and adsorbers inoperable and they had to be replaced. IE Information Notice No. 83-41[27] also addresses this topic. Perhaps the most serious incident of this type occurred on May 15, 1985 at Hatch, Unit 1 (LER 85-018-00, INPO SER 34-85, and IE Information Notice No. 85-85[28]) where inadvertently flooded ductwork leaked water onto an Analog Transmitter Trip System (ATTS) panel. This introduced moisture into the ATTS panel which, in turn, resulted in the malfunction of a safety relief valve and the High Pressure Coolant Injection System (HPCI). For approximately 15 minutes a safety relief valve could not be closed and the HPCI system could not initiate.

As part of the special (reactive) inspection of potentially generic HVAC deficiencies, FRPS inspectors discussed the Hatch incident with applicant and licensee representatives, reviewed applicant and licensee internal responses to INPO SER 34-85 and IE Information Notices No. 83-41 and 85-85, and inspected the physical configuration of filter housing drains and the fire protection water deluge systems at Region III commercial nuclear power plants. Although response to this concern was initially inadequate at some facilities, licensee meetings with NRC Region III personnel led to acceptable resolutions. Some of the modifications Region III NTOLs and operating plants made to fire protection deluge systems included: changing from automatic to manual activation (with NRC concurrence); modifying the valving arrangement from one valve under pressure (which when opened initiates deluge flow) to a system with two closed valves in series with an open low-point drain line isolation valve between them (which requires the operation of the three valves to initiate deluge flow), and an actuation sequence which required two permanently installed fire hoses to be connected and two valves to be opened.

Although the final design of the installed deluge systems and the applicants' and licensees' internal responses to INPO SER 34-85 and IE Information Notices No. 83-41 and 85-85 indicated that there was little likelihood of the fire protection systems inadvertently actuating, it often appeared that the applicants and licensees had not instituted administrative controls beyond those needed for the initiation of the deluge systems. The administrative controls which were initially lacking, included: (1) assurance that the filter housing will not overfill to the extent that water backs up into the ductwork, housing integrity is jeopardized, or seismic and static loading become concerns; (2) training or procedural cautions to warn the fire brigade that the water in the housing is contaminated and radwaste (or radiation protection) personnel should be notified; (3) assurance that the housing will be drained in a timely manner without overloading the radwaste system, with procedural steps to ensure that the proper isolation valves are correctly manipulated; (4) addition of the filter housing drain line isolation valves to the valve check list of the ventilation system startup procedure; and (5) assurance that the filter housing drain line isolation valves are verified closed as part of the (monthly) fire protection surveillance program. Region III FRPS inspectors assured that proper administrative controls were eventually established.

In addition to assuring that Region III licensees and applicants for operating licenses had properly designed systems and administrative controls to significantly reduce the probability of inadvertently actuating filter housing fire protection water deluge systems and to ensure proper recovery from deluge system operation, the FRPS inspectors also assured that potential for wetting of charcoal adsorbers during HVAC system operations due to leaky fire protection water deluge systems was minimized. Several conference papers discuss the effects of wetted carbon beds in detail including those by Kovach[12] and Graves, Hunt, Jacox, and Kovach[13] from the proceedings of the 13th and 15th Nuclear Air Cleaning Conferences, respectively. Fortunately, system modifications and administrative controls taken in response to inspector concerns and IE Information Notices 83-41 and 85-85 also usually reduced the probability of charcoal adsorber wetting during normal plant operations. However for several plants additional corrective actions were required, as the following example illustrates.

In March 1985 a Region III licensee noticed during surveillance that the an ESF filter housing was leaking onto the charcoal adsorbers in several trays. The problem was traced to a plugged check valve in the deluge valve drain line which was quickly corrected; however, a condition report for this incident was not prepared. In part because a condition report was not written for this incident, deluge valve drains apparently were not checked for the other HVAC filter housing deluge systems. During a plant tour by NRC inspectors in September 1985 it was noted that many of the deluge valve drain line sight glasses were partly filled with residue, indicating a significant potential for plugged drain lines or check valves. Apparently a small leakrate into the deluge line can produce significant wetting of the charcoal adsorbers because the high negative pressure in the filter housing atomizes the water and sprays it directly onto the charcoal. With the fan turned off, the deluge water would instead drip to the floor of the housing. The five filter housings that were opened for visual inspection during the

September 1985 plant tour indicated water had pooled in each, which may indicate a chronic problem with the deluge systems. The pooled water had evaporated, leaving residue (dirt, scale, and/or rust) on the floor of each filter housing. The licensee's subsequent investigation found corrosion and evidence of water damage on some charcoal trays. Licensee corrective actions included: (1) replacing damaged charcoal and trays (some had split seams); (2) cleaning filter housings of all dirt, scale, rust, or other material which might damage filters, (3) repainting corroded filter housing areas; (4) replacing all deluge valve drain line check valves by more reliable stainless steel check valves, and (5) providing adequate maintenance of the deluge systems and sight glass surveillances.

Lack of Charcoal Batch Traceability

Each original or replacement batch of impregnated activated carbon used in the adsorber section of air-cleaning systems is required to meet the qualification and batch test results specified by Regulatory Position 3.i of Regulatory Guide 1.52[2] or Regulatory Position 3.g of Regulatory Guide 1.140[3;14], as appropriate. NRC inspectors reviewed the Region III NTOLs regarding charcoal batch traceability; all five facilities had significant difficulty demonstrating batch traceability. Examples include: (1) 32 out of 35 drums had lost their batch identification labels; (2) several barrels in each of four batches in a given lot did not have the stenciled batch number designation; (3) a lot of a size great enough to constitute several batches was procured by a purchase order which did not require the charcoal vendor to designate batches; (4) at two facilities the charcoal vendor filled all the batch test canisters with charcoal from the same batch to represent all batches in the lot; and (5) during the NRC inspection at one NTOL, it was noted that most of the charcoal drum batch designation labels were poorly affixed due to insufficient adhesive properties. In all cases the applicants either restored batch traceability or discarded the charcoal.

Quality Assurance Records Problems

As part of the NRC Region III special HVAC inspection program, Quality Assurance (QA) records of equipment, vendor, filter, and personnel qualifications were reviewed at NTOLs. Since these inspections revealed only minor problems regarding the applicants' QA audit records of HVAC equipment and vendor qualifications, only filter (HEPA and charcoal adsorber) and personnel qualifications are discussed in this paper. It should be noted that the FRPS inspectors did not conduct full QA reviews of the above delineated areas. The purpose of the special inspection program was the potential verification of specific HVAC deficiencies. The comprehensive reviews of NTOL QA programs are conducted by Region III Quality Assurance Program Section (QAPS) inspectors with assistance from the resident inspectors.

At the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Jacox[11] gave an example of applicant QA personnel, "who admit that they have no idea of the technologies involved (and no need to learn it), down-grading experienced test personnel from Level III to a Level II based on a utility decision to use the N45.2.6 <u>recommended</u> experience periods as an absolute minimum." He also points out the delays some utilities inflict on the N510 acceptance test program while personnel are qualified to the utilities interpretations of ANSI N45.2.6[30,31]. Regional inspectors review the applicants' QA and training records to ascertain whether commitments made to the NRC regarding personnel qualifications are met. Region III FRPS inspectors identified no significant problems with the ANSI/ASME N510 acceptance test personnel qualification records at NTOLs. CONAGT's efforts concerning the development of a specific qualification standard for field-test personnel was discussed by Miller[21] at the last air-cleaning conference.

Major QA problems at two Region III NTOLs resulted in all the charcoal adsorbers being disqualified for use in commercial nuclear power plants. At the first facility, half of the charcoal failed the spinster carbon laboratory test and the other half was without new charcoal qualification test records. The utility discarded their entire charcoal inventory. The second facility also discovered that all of its charcoal was unqualified for various reasons, including: (1) lack of batch traceability; (2) purchase orders did not require qualification documentation; and (3) improper new charcoal qualification tests. With the concurrence of NRR, the utility successfully requalified the ESF charcoal (four batches, 40,000 pounds) according to the new charcoal acceptance criteria of ASNI N509-1980. The non-ESF charcoal (three batches, 7,000 pounds) was discarded. The applicant also found a number of HEPA filters, designated for use in non-ESF, N510 filter systems, which did not have adequate qualification documentation.

Inadequate N510 Acceptance Criteria and Tests

A primary goal of the NRC Region III reactive inspection response to concerns raised by certain Nuclear Air Cleaning Conference papers is to assure that NTOL ANSI/ASME N510 acceptance test programs comply with regulations and applicant commitments. During the preparation of the commitment and analysis reports, the applicants found numerous failures to meet regulations and commitments. The inspectors reviewed the corrective actions for adequacy. Usually the corrective actions were adequate; however, controversy often developed between the NRC and the applicants in several key areas. The disputed areas were mostly detailed specifics involving adequacy of N510 test acceptance criteria and apparent lack of detailed compliance with N510 test procedure and report specifications. Although these disagreements reached satisfactory conclusions for all but one NTOL, it is important that utilities pay close attention to these sorts of details. One Region III NTOL received notices of violation when similar failures were discovered by the NRC, rather than the licensee. It is imperative that applicants for licenses state in their FSAR commitments to Regulatory Guides 1.52 and 1.140 any exceptions they wish to the specific requirements/recommendations given in ANSI/ASME N509 and N510.

Control Room Habitability

At the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, several papers were presented which discussed control room habitability potential deficiencies, including papers by Moeller and Kotra[26] and Hayes, Muller, and Gammill[32]. The first paper discussed deficiencies reported by licensees in LERs and the second paper discussed the NRC working group study of control room habitability. It is noted that interim findings of the NRC study concerning operating plant reviews will be presented at this conference by Driscoll[33]. Because the generic NRC study group was conducting a thorough review of control room habitability, the Region III special HVAC inspection program concentrated inspector efforts toward assuring that the calculated dose guidelines of 10 CFR 50, Appendix A, General Design Criteria (GDC)-19[34] were met at Region III NTOLs.

Section 4.12 of ANSI/ASME N509[16,29] states that the criteria for leakage across the pressure boundary of any portion of an air cleaning system are: (a) air cleaning effectiveness requirements; (b) health physics requirements; and (c) duct and housing quality requirements. The standards also states that the lowest value as determined by items (a), (b), or (c) shall be used as the allowable leakage for design and testing. Based on the review of Region III NTOLs, it appears common for applicants to base the value of allowable unfiltered inleakage into the control room gas envelope on the health physics requirements without doing the required evaluation to choose the lowest value derived from the three methods. It should be noted that the applicants also have the option of requesting NRR concurrence such that they need consider only the allowable inleakage value obtained from the health physics requirements evaluation.

Section 6.4, "Control Room Habitability System," of the Standard Review Plan (NUREG-0800[35]) provides NRC personnel with guidance on the review of applicants' control room design, including the requirements of 10 CFR 50, Appendix A, GDC-19. The Standard Review Plan references a Nuclear Air Cleaning Conference paper by Murphy and Campe[36] for further guidance concerning GDC-19 compliance. Further guidance on the mathematical modeling of control room ventilation systems, which might aid in the calculation of post-accident doses to control room operators, is contained in several other Nuclear Air Cleaning Conference papers, including those by Miller, Ornberg, and Rooney[37] and Almerico, Michaels, Ornberg, and Lahti[38]. The Region III FRPS inspectors reviewed the calculational basis for each NTOL's control room allowable unfiltered inleakage and assured that this N510 acceptance test parameter agreed with the applicant's regulatory commitments. All Region III NTOL reviews of the documentation of this parameter and the acceptance test programs for the control room ventilation systems have led to several rounds of negotiations between Region III, NRR, and the applicant before final resolution.

Although Standard Review Plan Section 6.4 states that applicants for licenses should determine the control room unfiltered inleakage flowrate (infiltration) conservatively and include such infiltration sources as leaking dampers, none of the approximately 20 FSAR control room dose evaluations reviewed by the inspectors assumed infiltration except for a few cases where lack of acceptable two-door vestibules resulted in the standard assumption of 10 cfm inleakage for post-accident control room ingress and egress. Inspector reviews of N510 acceptance test criteria and test results revealed that control room infiltration rates of several hundred cfm are apparently common. Much of this infiltration is due to high leakage ductwork which has mechanical lock longitudinal seams and gasketed flanged transverse joints. Addition leakage at some plants is also due to the use of opposing blade isolation dampers rather than Class I bubble-tight dampers required by ANSI/ASME N509[16,29]. Despite the poor quality of HVAC construction at some facilities, all plants are required to comply with the same regulations and to their FSAR commitments. Three of five Region III NTOLs have passed these criteria to date, although some system modifications were necessary.

By not including the actual control room infiltration values in the FSAR evaluation of control room operator post-accident doses, the nuclear industry is left with the impression that these HVAC systems are of considerably higher quality than field experience indicates. This concept is illustrated by the following example. Acceptance Criterion 2.a of Standard Review Plan Section 6.4 states that dampers used to isolate the control zone from adjacent zones or the outside should be leak-tight and the degree of leak-tightness should be documented in the FSAR. The example utility purchased an opposing blade isolation damper which was specified to leak at no more than 2 cfm at 10 in. wg. When tested in-situ, the damper leaked at 670 cfm at 4 in. wg. It should be recognized that vendor idealized laboratory tests may not reflect the conditions imposed on an installed and cycled nuclear power plant HVAC damper. The total allowable infiltration for the example control room was 650 cfm. This plant's mechanically connected thin-gauge sheet metal control room ductwork also had significant leakage problems.

The inspectors noted that all Region III NTOLs reviewed to date had significant difficulty passing the acceptance criteria imposed by GDC-19 and indeed usually failed the initial control room habitability acceptance tests. It should be noted that although ANSI/ASME N510[1,39] does not specifically require in-situ damper leakage tests, these tests are required for some control room HVAC isolation dampers to satisfy the conditions of ANSI/ASME N509[16,29] and GDC-19[34]. In fact, the measurements of all control room infiltration pathways, including other systems' ductwork under positive pressure which passes through the control room gas control envelope, are required when the health physics requirements option of N509 is used.

III. SUMMARY

During the last two years, the NRC Region III has conducted a reactive inspection program partially in response to potential technical deficiencies identified in recent Nuclear Air Cleaning Conference papers. The technical concerns reviewed at Region III NTOLs were in several areas, including: post-accident effluent sample line deposition losses; failure to implement good NATS engineering practices; filter bypass via filter housing drain lines; silicone sealant; fire protection water deluge systems; charcoal batch traceability; QA records; ANSI/ASME N510 acceptance criteria and tests; and control room habitability per GDC-19 of Appendix A to 10 CFR 50. Without exception, these concerns were identified as apparently prevalent technical deficiencies in the Region III NTOLs and operating plants reviewed. Based on Region III inspector discussions with numerous industry experts in the past two years, deficiencies in the above delineated technical areas may be common in the nuclear industry. Other NRC regions, NRR, and IE headquarters have been kept informed of the findings of the special Region III HVAC inspection program.

IV. REFERENCES

1. American National Standard, "Testing of Nuclear Air-Cleaning Systems," ANSI/ASME N510-1980, The American Society of Mechanical Engineers (1980).
2. U.S. Nuclear Regulatory Commission, Regulatory Guide 1.52, "Design, Testing, and Maintenance Criteria for Post-Accident Engineered-Safety-Feature Atmospheric Cleanup System Air filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants," Revision 2, March 1978.
3. U.S. Nuclear Regulatory Commission, Regulatory Guide 1.140, "Design, Testing, and Maintenance Criteria for Normal Ventilation Exhaust System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants," Revision 0, March 1978.
4. Kabat, M. J., "Deposition of Airborne Radioiodine Species on Surfaces of Metals and Plastics," Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF-820833, Volume 1, Page 285 (August 1982).
5. U.S. Nuclear Regulatory Commission, NUREG-0737, "Clarification of TMI Action Plan Requirements," November 1980.
6. U.S. Nuclear Regulatory Commission, Regulatory Guide 1.97, "Instrumentation for Light-Water-Cooled Nuclear Power Plants to Assess Plant and Environs Conditions During and Following an Accident," Revision 3, May 1983.
7. Mandler, J. W., S. W. Duce, J. W. Tkachyk, J. Y. Lee, and C. A. Willis, "Monitoring Gaseous Radioiodine at Power Reactors," paper presented at the 19th DOE/NRC Nuclear Air Cleaning Conference, August 1986.
8. Unrein, P. J., C. A. Pelletier, J. E. Cline, and P. G. Voilleque, "Transmission of Radioiodine Through Sampling Lines," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 1, Page 116 (August 1984).
9. Moeller, D. W., and L. S. Casper Sun, "FAilures in Air-Cleaning, Air-Monitoring, and Ventilation Systems in Commercial Nuclear Power Plants (January 1, 1978 Through December 31, 1981), "Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF-820833, Volume 2, Page 178 (August 1982).

10. Jacox, J. W., "NATS Field Testing Observations and Recommendations," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 918 (August 1984).

11. Jacox, J. W., "Experience in Startup, Preoperational and Acceptance Testing of Nuclear Air Treatment Systems in Nuclear Power Plants," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 1209 (August 1984).

12. Kovach, J. L., "The Users View of the Reliability of Air Cleaning Systems in Nuclear Facilities," Proceedings of the 13th AEC Air Cleaning Conference, CONF-740807, Volume 1, Page 50 (August 1974).

13. Graves, C. E., J. R. Hunt, J. W. Jacox, and J. L. Kovach," Operational Maintenance Problems with Iodine Adsorbers in Nuclear Power Plant Service," Proceedings of the 15th DOE Nuclear Air Cleaning Conference, CONF-780819, Volume 1, Page 428 (August 1978).

14. U.S. Nuclear Regulatory Commission, Regulatory Guide 1.140, "Design, Testing, and Maintenance Criteria for Normal Ventilation Exhaust System Air Filtration and Adsorption Units for Light-Water-Cooled Nuclear Power Plants," Revision 1, October 1979.

15. Burchsted, C. A., A. B. Fuller, and J. E. Kahn, ERDA 76-21, Nuclear Air Cleaning Handbook, U.S. Energy Research and Development Administration, Second Edition, March 31, 1976.

16. American National Standard, "Nuclear Power Plant Air Cleaning Units and Components," ANSI/ASME N509-1976, The American Society of Mechanical Engineers (1976).

17. Bellamy, R. R., "HEPA Filter Experience During Three Mile Island Reactor Building Purges," Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF-820833, Volume 2, Page 1003 (August 1982).

18. Hubbard, D. M., "Field Testing of Nuclear Air Cleaning Systems at Duke Power Company," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 904, (August 1984).

19. Bellamy, R. R., "Regulatory Experience with Nuclear Air Cleaning," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 897 (August 1984).

20. Miller, Jr., W. H., "CONAGT's Place in ASME's Centennial Year," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 1480 (August 1984).

21. First, M. W., "Results of CONAGT-Sponsored Nuclear-Grade Carbon Test Roundrobin," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 1186 (August 1984).

22. Kovach, J. L., "Air Cleaning in Accident Situations," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 1495 (August 1984).

23. Bellamy, R. R., "CONAGT's Nuclear Carbon Roundrobin Test Program," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 2, Page 1190 (August 1984).

24. U.S. Nuclear Regulatory Commission Memorandum, Hugh L. Thompson, Jr., Director, Division of Licensing, NRR to John A. Hind, Director, Division of Radiation Safety and Safeguards, Region III, Subject: ADEQUACY OF ZION CONTROL ROOM EMERGENCY AIR CLEANING SYSTEM MODIFICATION AND REPAIR (AITS F03018285), August 26, 1985. (available in the NRC Public Document Room)

25. U.S. Nuclear Regulatory Commission Memorandum, Robert M. Bernero, Director, Division of BWR Licensing, NRR to John A. Hind, Director, Division of Radiation Safety and Safeguards, Region III, Subject: RESPONSE TO REQUEST FOR TECHNICAL ASSISTANCE RELATED TO AIR CLEANING SYSTEMS AT CLINTON (TIA 85-70), March 10, 1986. (available in the NRC Public Document Room)

26. Moeller, D. W. and J. P. Kotra, "Commentary on Nuclear Power Plant Habitability - Including a Review of Related LERs (1981-1983)," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 1, Page 145 (August 1984).

27. U.S. Nuclear Regulatory Commission, IE Information Notice No. 83-41, "Activation of Fire Suppression System Causing Inoperability of Safety-Related Equipment," June 22, 1983.

28. U.S. Nuclear Regulatory Commission, IE Information Notice No. 85-85, "Systems Interaction Event Resulting in Reactor System Safety Relief Valve Opening Following a Fire Protection Deluge System Malfunction," October 31, 1985.

29. American National Standard, "Nuclear Power Plant Air Cleaning Units and Components," ANSI/ASME N509-1980, The American Society of Mechanical Engineers (1980).

30. American National Standard, "Qualifications of Inspection, Examination, and Testing Personnel for the Construction Phase of Nuclear Power Plants," ANSI N45.2.6-1973, The American Society of Mechanical Engineers (1973).

31. American National Standard, "Qualifications of Inspection, Examination, and Testing Personnel for Nuclear Power Plants," ANSI N45.2.6-1978, The American Society of Mechanical Engineers (1978).

32. Hayes, J. J., D. R. Muller, and W. P. Gammill, "NRC Study of Control Room Habitability, "Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 1, Page 162 (August 1984).

33. Driscoll, J. W., "Control Room Habitability Study - Findings and Recommendations," paper presented at the 19th DOE/NRC Nuclear Air Cleaning Conference, August 1986.

34. Appendix A to Title 10 of the Code of Federal Regulations, Part 50, "General Design Criteria for Nuclear Power Plants," Criterion 19 - Control Room.

35. U.S. Nuclear Regulatory Commission, NUREG-0800, "Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Power Plants," Revision 5, December 1984.

36. Murphy, K. G. and K. M. Campe, "Nuclear Power Plant Control Room Ventilation System Design for Meeting General Criterion-19," Proceedings of the 13th AEC Air Cleaning Conference, CONF-740807, Volume 1, Page 401 (August 1974).

37. Miller, Jr., W. H., S. C. Ornberg, and K. L. Rooney, "A Consistent Approach to Air-Cleaning System Duct Design," Proceedings of the 16th DOE Nuclear Air Cleaning Conference (August 1980).

38. Almerio, F., A. J. Michaels, S. C. Ornberg, and G. P. Lahti," Performance Evaluation of Control Room HVAC and Air-Cleaning Systems Under Accident Conditions," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF-840806, Volume 1, Page 188 (August 1984).

39. American National Standard, "Testing of Nuclear Air-Cleaning Systems," ANSI/ASME N510-1975, the American Society of Mechanical Engineers (1975).

DISCUSSION

JACOX: This is more of a comment and commendation than a question. Initially, you made a point about good engineering practice. This is something that many of the people here know I have harped on for years and will again at a later panel session. I find it very difficult to understand why people will take a minimum regulatory approach rather than apply good engineering practice, when in the long run, just fighting about it often costs more than the perceived savings that you anticipate. I particularly commend your citing specific examples in previous papers presented by many of us at these Conferences. In the long run, good engineering practice will, in my experience, generally meet all the NRC requirements. That should be the place to start.

MILLER: A comment I would like to add is that I saw my own name and your name (Jack) in the specific example table too many times. I hope that in the next few Conferences, we will see some fresh authors because we need to hear from everyone having relevant experiences.

CLOSING COMMENTS OF SESSION CHAIRMAN MILLER

We have come a long way in terms of finding out what is bothering users of the Nuclear Codes and Standards that apply to air and gas cleaning systems, and changes for the better are well along. We still have a lot of inconsistences between regulations and corresponding codes standards. We still have some practices and some systems in the field that need additional work to bring them into compliance. Let us conduct a little examinations of conscience when we get back home to see if there is anything we can do to improve the situation.

SESSION 6

KRYPTON, RADON, TRITIUM, CARBON-14 TREATMENT

TUESDAY: August 19, 1986
CO-CHAIRMEN: W. Motes
Westinghouse Idaho Nuclear
A.G. Croff
Oak Ridge National Lab.

FINAL RESULTS AND CONSEQUENCES OF THE DEVELOPMENT OF A CRYOGENIC KRYPTON SEPARATION SYSTEM
E. Hutter, J. Amend, R. von Ammon, W. Bumiller, G. Neffe

KRYPTON RECOVERY BY CRYOGENIC DISTILLATION WITHOUT PRECEDING OXYGEN ELIMINATION
L. Geens, W.R.A. Goossens, J. Marien

PILOT PLANT DEVELOPMENT FOR ADSORPTIVE KRYPTON SEPARATION FROM DISSOLVER OFF-GAS
H. Ringel, R. Printz

CONTAINMENT OF KRYPTON-85 BY AN ION-IMPLANTATION/SPUTTERING PROCESS USING RARE EARTH ALLOYS
E.D. McClanahan, R.W. Moss, E.N. Greenwell, E.R. Bradley, E.L. McDonald

CONDITIONING OF RADIOKRYPTON USING ION SPUTTER PUMPS
J. Romer, E. Henrich, T. Fritsch

OPENING COMMENTS OF SESSION CO-CHAIRMAN CROFF

This session is, in principle, concerned with the recovery, treatment, and disposal of relatively long-lived volatile radionuclides such as tritium, Kr-85, C-14, and I-129. The primary source of these radionuclides would be from nuclear fuel reprocessing plant effluents. They are important because their uncontrolled release has the potential to significantly increase the radiological impact of nuclear electricity generation.

It is apparently accepted that krypton, carbon, and iodine will have to be (or are being) recovered, treated to yield an acceptable waste form, and isolated using a technology appropriate to each radionuclide. The need to reduce releases of other radionuclides is still open to question, although recovery processes are currently being investigated. The subject of this session overlaps consideration of dissolver off-gas cleaning (Session 8), and it is there that some papers on iodine and tritium recovery will be given.

As I've noted, the need to investigate management of long-lived volatile radionuclides is closely related to the degree to which nuclear fuel will be reprocessed. The many worldwide delays in undertaking reprocessing have clearly resulted in greatly reduced work in the subject area of this session. Consideration of the likely near-term directions of reprocessing indicates that this situation is likely to persist for some time unless repository-related activities require the use of some of this technology.

FINAL RESULTS AND CONSEQUENCES OF THE DEVELOPMENT OF A CRYOGENIC KRYPTON SEPARATION SYSTEM

E.Hutter, J.Amend, R.v.Ammon, W.Bumiller, G.Neffe,

Kernforschungszentrum Karlsruhe GmbH,
Karlsruhe, Federal Republic of Germany.

Abstract

After some ten years of development the test program carried out with the ADAMO and KRETA facilities for cryogenic separation of Krypton from the dissolver offgas of a reprocessing plant was brought to an end in a continuous campaign of three months of industry-like operation. The feasibility of the chosen concept has been demonstrated in some 20,000 hours of test operation.

The most important activity was the investigation, in cryogenic columns of the behavior of impurities within the offgas stream (O_2, CO_2, CH_4,NO_x) and of compounds which may form in the columns (O_3, N_2O_4) due to radiolysis.

Progress was made

- in effective removal of NO_x in the preparatory catalytic reduction step; NO_x can be removed from the process gas down to less than 1 ppm_v, which prevents plugging of the cryogenic column;
- in effective removal of ozone together with the xenon bottom product from the krypton-xenon separation column. Operating the column with continuous head and bottom product withdrawal, and suppressing pressure oscillations by careful control, allows the Xe-O_3 product to be kept free from krypton down to a few ppm_v.

Another topic of development was the reduction of the krypton inventory of the columns to reduce the activity inventory:

- While the principle of sieve plates was maintained, the column diameter was reduced and the design features of the plates were changed.
- Bottom product withdrawal of the first column was switched from the liquid to the gaseous from, which reduced the Kr concentration in the bottom liquid almost by a factor of 10.
- The most drastic activity reduction would result from replacing the sieve plate column by a packed columm.

In three months of continuous operation no major malfunctions occurred. No long-term effects, such as dangerous accumulations or gradual plugging of parts of the installations, were observed.

Introduction

Ten years ago, in 1976, we built and commissioned the KRETA test facility, which has since been operated in 19 campaigns, each of them comprising several weeks of continuous operation, including a final long-term campaign of three months, to develop and test the low-temperature rectification technique for krypton removal from the dissolver offgas of a reprocessing plant (Fig. 1). This trial period corresponds to a cumulated period of operation of approximately 20,000 hours. In this way, we have proved the usefulness and reliability of the method, although we did not work with radioactive krypton in that phase.

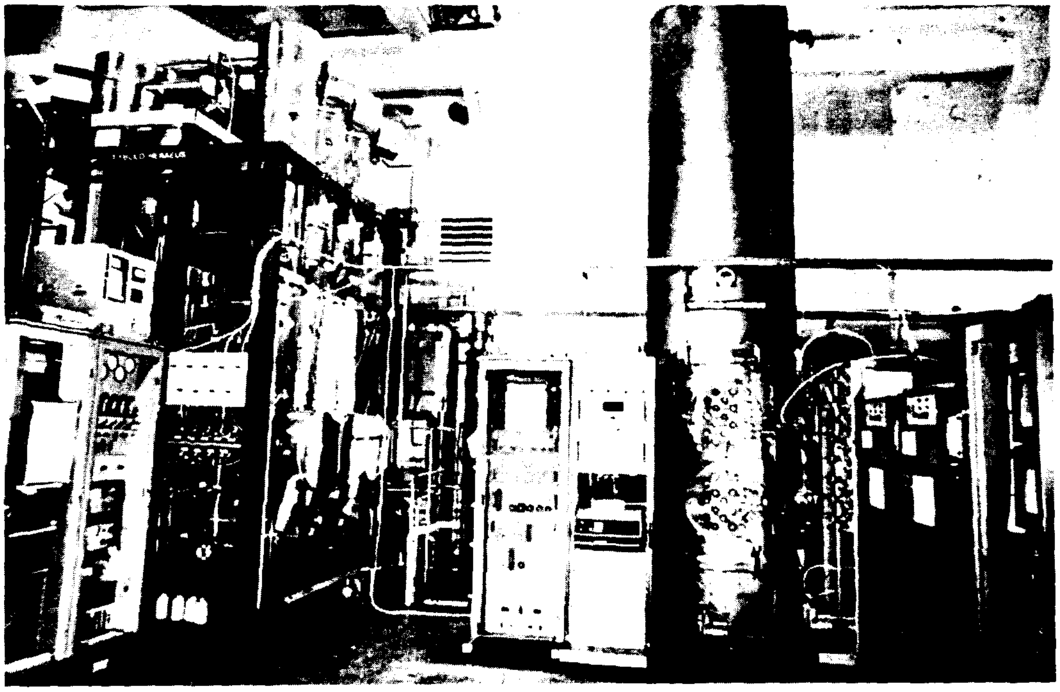


Fig.1: KRYPTON RETENTION FROM DISSOLVER OFFGAS TESTFACILITIES ADAMO-KRETA

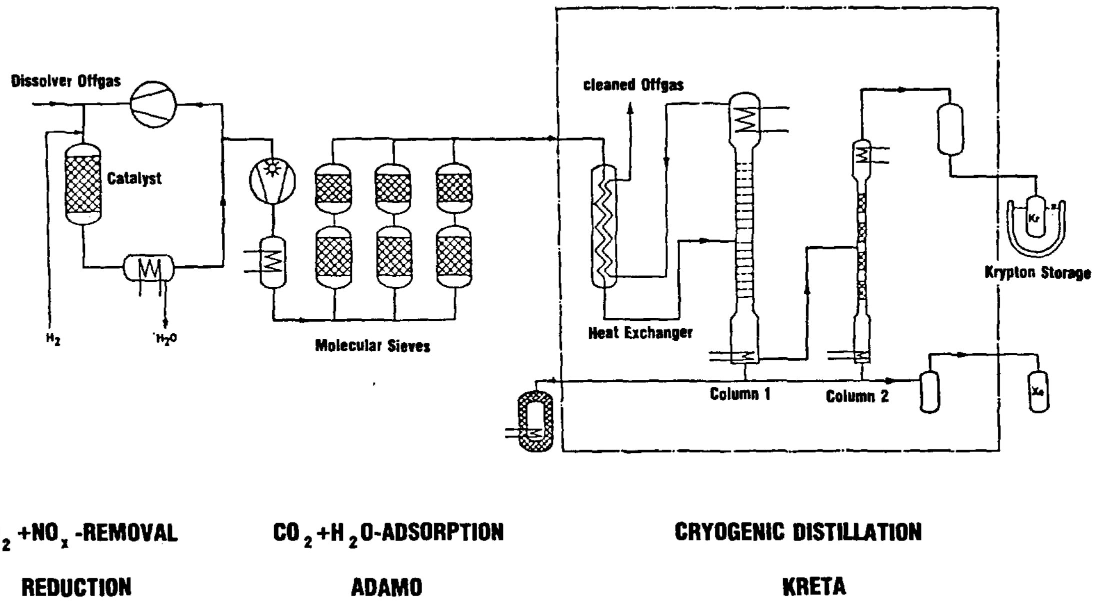


Fig.2: KRYPTON RETENTION FROM DISSOLVER OFFGAS
KfK TESTFACILITIES

The progress of our work has been reported at all Nuclear Air Cleaning Conferences since 1978 (1) (2) (3) (4).

The basic flowsheet of the process steps in cryogenic krypton removal (Fig. 2) as built up in Karlsruhe for a gas throughput of 50 m^3/h STP shows the preparatory removal of those impurities, which would freeze out at lower temperatures, in the following facilities:

- REDUKTION, for the catalytic reduction of oxygen and nitrogen oxides by means of hydrogen and removal of the water generated,

and, after a pressure increase to 5 bar by a water ring compressor,

- ADAMO, for the adsorptive retention of CO_2 and water in series connected silicagel and molecular sieve beds. Three such lines are arranged in parallel so that adsorption, flushing back of co-adsorbed krypton and xenon and regeneration can be carried out simultaneously in one operating cycle.

This is followed by the cryogenic section installed in a cold box (KRETA facility), consisting of the inlet heat exchanger, in which the gas is cooled to approx. 125 K, the first separation column (sieve plate column), in which the Kr and Xe noble gases are separated from the nitrogen carrier gas, and the second separation column (packed column), which separates krypton from xenon.

In addition, there is a dump tank to accept the liquid contained in the plant in cases of maloperation, and a filling device for noble gas.

Besides investigating the reliability of this process, our activities were concerned with the following main topics:

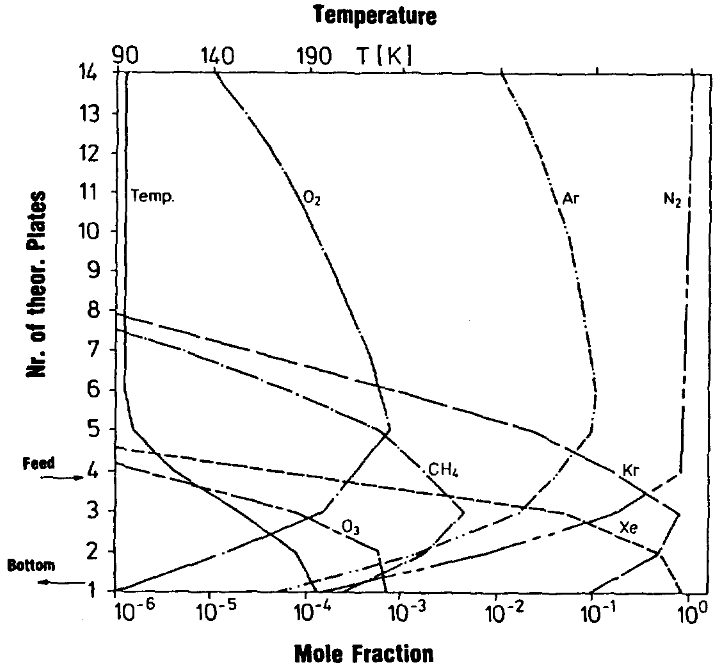
- Behavior of the highly volatile impurities (mainly O_2, CO_2, NO, and CH_4), which can still enter the cryogenic facility in ppm_v concentrations even after preparatory cleaning (Fig.3 and 4).
- Behavior of the ozone generated radiolytically out of oxygen, which we simulated by feeding ozone to the first separation column (Fig..3 and 4), and the movement of radiolytically generated NO_2/N_2O_4.
- The influence operating conditions exert on product composition.
- Determination of the decontamination factor of the whole facility.
- Accident studies (failures of column feed, heating and cooling) as well as equipment behavior in the presence of fluctuations in throughput and concentration.
- Development of cryogenic separation equipment so as to reduce the krypton inventory, i.e., the activity inventory.

Final Results

Let us now discuss the most recent findings and describe the latest process concept and equipment design.

Behavior of Nitrogen Oxides in the First Cryogenic Column

A distinction must be made between the NO_2 arising only in the first column as a result of radiolysis of N_2 or NO with O_2 (3) or of residual NO impurities carried into the column with O_2, and the NO_2 not retained in the preparatory cleaning step.



Feed: 50m^3 STP/h

Reflux Ratio: 1

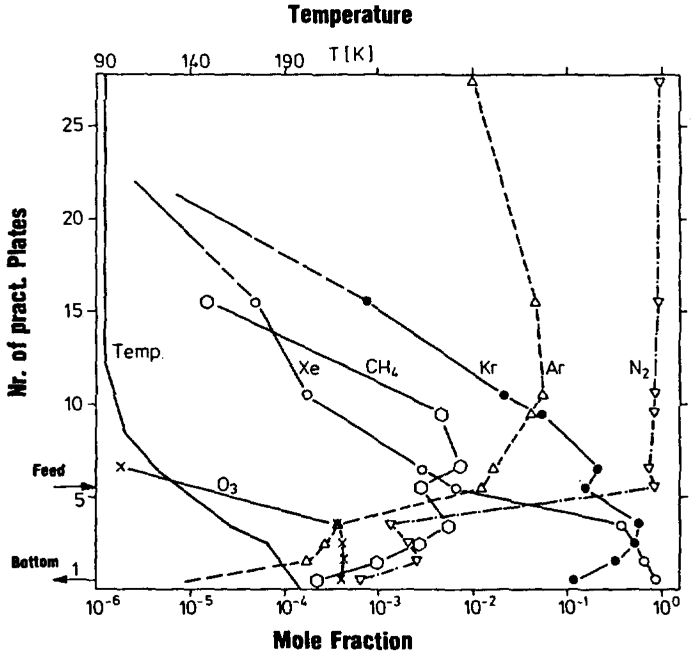
Pressure: 5 bar

Feedgas Concentration

Ar	1 vol%
Kr	400 vpm
Xe	4000 vpm
Ch_4	1 vpm
O_2	10 vpm
O_3	3 vpm
N_2	98.5586 vol%

calculated Data, Gas

Fig. 3: CONCENTRATION PROFILES IN THE FIRST KRETA COLUMN



Feed : 50 m^3 STP/h

Reflux Ratio: ~ 1

Pressure: 5 bar

Feedgas Concentration

Ar	1 vol%
Kr	400 vpm
Xe	4000 vpm
CH_4	1.2 vpm
O_3	3 vpm
N_2	98.55958 vol %

experimental Data, Gas

Fig.4: CONCENTRATION PROFILES IN THE FIRST KRETA COLUMN

In earlier experiments on the behavior of nitrogen oxides we were able to see that NO fed to the column indeed reacted with the oxygen present in the column at this low temperature to form NO_2 which, however, seems to dissolve in the liquid immediately and go to the column bottom. Only when the solubility limit in xenon of about 1 Mol% is exceeded it is precipitated there as a solid and thus does not upset column operation (aside from plugging up measurement lines, e.g., for measurement of the bottom level by means of the differential pressure, a situation which could be remedied by adopting a different measurement principle, such as capacitance level measurement) (3). The same behavior is expected also of NO_2 generated by radiolysis.

However, when adding NO_2 as a residual impurity to the column feed we found that, at NO_2 concentrations above 1 ppm_v, the differential pressure across the first sieve plate gradually rises above the feed point and, over prolonged periods of operation, gradual plugging of the column must be expected. This results in the need to reduce in the preparatory cleaning step (catalytic reduction and adsorption) NO_x to levels of $<$ 1 ppm_v. After we had begun to use a computer-supported hydrogen control system in the REDUCTION test facility we were able to meet this criterion even in process gas streams of varying compositions (Fig. 5). As NO is oxidized to NO_2 in the presence of O_2 on the molecular sieve even at room temperature, and the molecular sieve adsorbs NO_2, we also studied this retention possibility in a separate test series by means of ADAMO. We found that, at an O_2 concentration of at least 120 ppm_v, a residual NO concentration of 5 ppm_v was retained completely by our technical molecular sieve arrangement. In the presence of a lower O_2 excess, all NO was converted into NO_2, but NO_2 still broke through with >1 ppm_v.

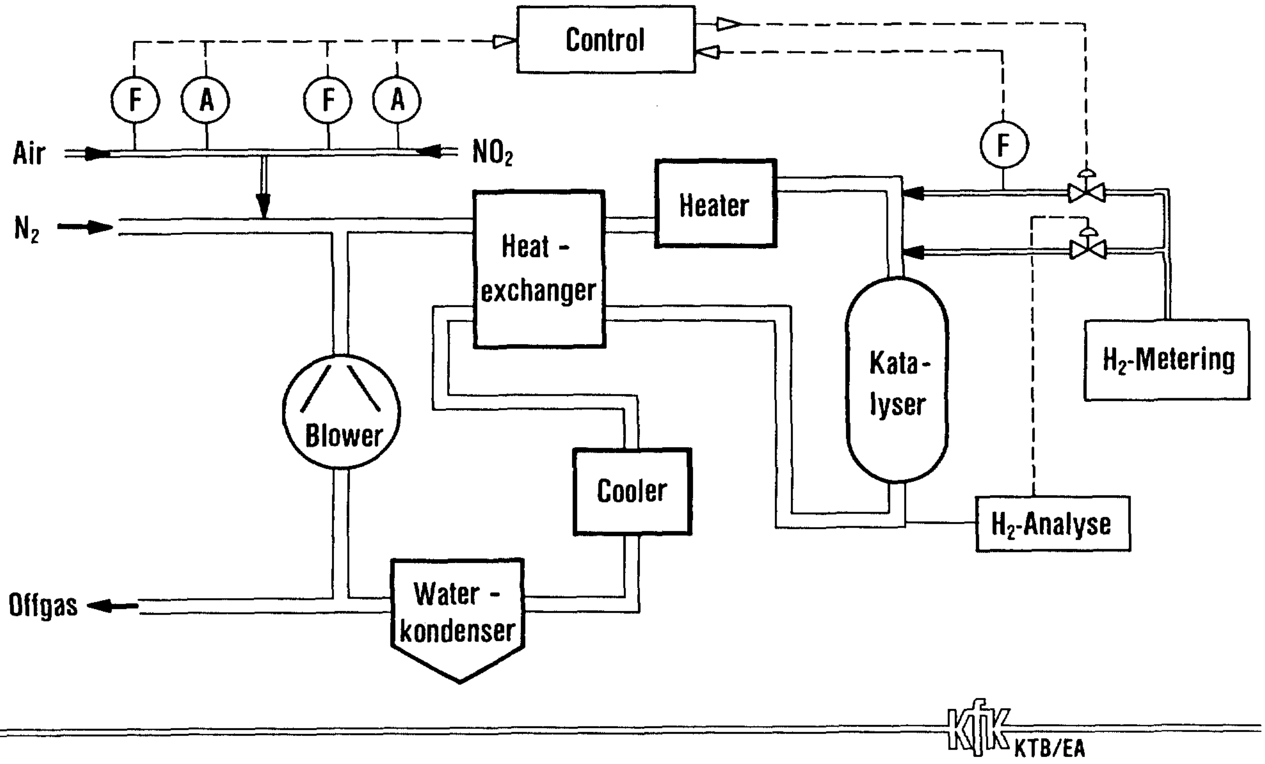


Fig.5: H_2-METERING AT THE REDUCTION

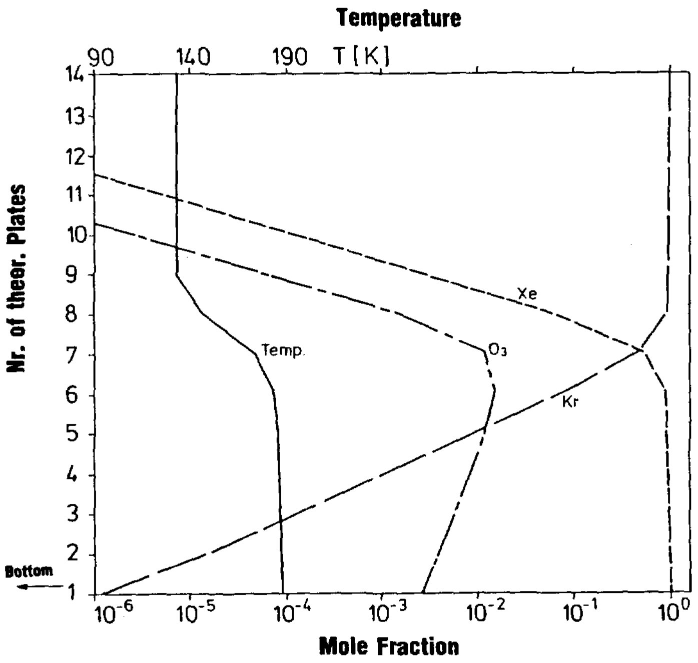
This aspect of making use of oxygen would be attractive if preparatory cleaning with selective NO_x reduction were applied, i.e., feeding O_2 to the separation facility. This would allow the criteria to be slackened which this NO_x retention would have to meet, for the adsorption unit would accommodate the residual nitrogen oxides.

In any case, however, in order to be on the safe side, we would arrange for two inlet heat exchangers and design them as regenerable cold traps, in case increased NO_x concentrations or also CO_2 and H_2O were to enter the cryo-facility as a result of maloperation or irregularities.

Purity of the Xenon Product of the Second Cryogenic Column

We were able to show that separating krypton from xenon in the second column is no problem if only these two substances are present or if highly volatile gases, such as N_2, Ar, O_2 or CH_4, get into this column in the mode of operation selected here, assuming, of course, continuous extraction of these gases at the top of the column. In this way, a xenon product can be achieved whose residual krypton fraction is in the ppb rang.

If one now traces the path of the ozone to be removed from the second column with the xenon bottom product, even a simulation calculation (Fig. 6) shows that the three component system of Kr, O_3,Xe is separated to the desired product purities in only a small number of theoretical stages. We put this separation part into the lower region of our column, the number of stages which is vastly overdimensioned anyway. This mode of operation minimizes the O_3 inventory and helps to keep the O_3 concentration far from the explosion limit of 8% ozone in xenon.



Feed: 176 l STP/h

Reflux Ratio: 150

Pressure: 3 bar

Feedgas concentration

Kr 9.083 vol%

O_3 0.068 vol%

Xe 90.849 vol%

HETP: 5 cm

calculated Data, Gas

Fig. 6: **CONCENTRATION PROFILES IN THE SECOND KRETA COLUMN**

As far as technical control is concerned, this mode of column operation is easily achieved, but now the Kr front is so close to the xenon extraction that the residual krypton content in the xenon product may become worse as a result of minor fluctuations in the operating parameters. For example, our control concept included temperature control acting on the bottom heating system. Because of the steep slope of the temperature profile at this point, the transition from xenon to krypton, the set temperature was easily exceeded or underrun, the consequence being that bottom heating was controlled as in on-off control. This plus the batch extraction of the top product also resulted in minor pressure fluctuations which, in turn, affected the temperature. The result was a residual content of krypton in xenon of 40 to 400 ppm_v in the continuous mode of operation. This means that this product can be released through the stack only after dilution which, at the same time, detracts from the decontamination factor of the whole plant.

However, the product would be quite suitable for commercial utilization of the xenon after a final cleaning procedure, e.g., by adsorption.

If the objective were to keep the residual Kr content in xenon very low in the continuous mode of operation, column control would have to be improved. The column pressure and the position of the separation interface between krypton and xenon must be maintained within very narrow limits. Control concepts serving this purpose are proposed in the literature (5). We decided to do without such backfitting, but do not consider it to be a major problem.

Reduction of the Activity Inventory

One important objective of our work was the minimization of the krypton inventory, i.e., the activity inventory, in the plant. On the one hand, this was based on safety considerations and, on the other hand, less activity reduces the radiolytic formation of ozone and NO_2 (3).

We did two things in the first column:

(a) Maintaining the sieve plate principle, which we still consider to be most suitable for reasons of operational reliability, because of its low sensitivity to fluctuations in operation (throughput, concentrations and maloperation), we designed a new first column.

The modifications affecting the inventory were these:
- Reduction of the column diameter from 160 mm to 120 mm in the top part and 100 mm in the bottom part (with the reflux being increased from 0.58 to approx. 1.0).
- Reduction of the liquid content on the plates by changing the weir levels from 55 mm to 30 mm.

Design possibilities are limited by the area of the sieve holes needed for the gas flow. However, it was found that the area of the sieve holes as determined by the usual methods of computation can be underrun by a clear margin without entailing any drawbacks.

What is more important is a pronounced surge zone around the inlet and the outlet ducts to prevent flooding of the column. This was achieved by segmented weirs (Fig. 7).

(b) The activity inventory was further reduced by changing from the original extraction of the liquid of the noble gas-bottom product at the base of the bottom tank to gas extraction above the liquid level. In normal operation, the Kr/Xe ratio at the point of extraction is always established as a function of the ratio of the feed concentration, i.e. 10/90%. In the case of liquid extraction, this is the composition of the bulk liquid in the bottom tank while, in the case of gas extraction, the liquid contains only some 1% of krypton.

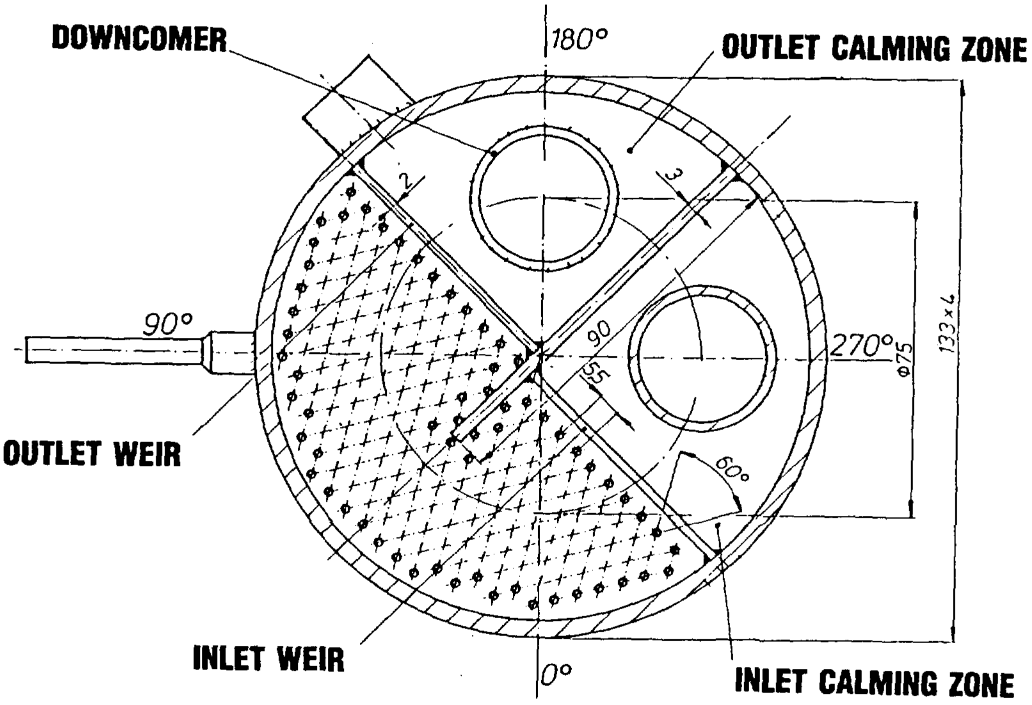


Fig. 7: IMPROVED SIEVE TRAY

Table 8 shows the activity levels of the old and the new KRETA columns designed for 50 m³/h STP. For extrapolation to 100 m³/h STP of offgas, wich would roughly be applicable to a reprocessing plant of 350 tons/a, this value would have to be multiplied by a factor of $\sqrt{2}$.

The most drastic activity reduction is brought about by the change from sieve plate columns to packed columns (Table 8). We decided not to take this step because our Belgian colleagues in Mol, in a similar, smaller facility, operated both separation units as packed columns and also achieved good results. In our second column we accumulated experience with packings.

In our opinion, packed columns are less stable to fluctuations in throughput and process gas composition; their reaction to disturbances is also less damped. Yet, if a low activity inventory in the plant is regarded as the more important factor (for safety reasons or because of the radiolysis products), we think the use of packings is feasible also in the first cryogenic separation column.

Table 8. Activity Inventory

FEED: 50m^3 STP/h

FEED COMPOSITION [vpm]:

N_2	Ar	O_2	CH_4	NO	Kr	O_3	Xe
98.56%	1%	10	1	1	400	—	4000

	KRETA first Design	KRETA new Design	KRETA package	
ACT.INV.	284300	120123	6060	Ci
COOLING	3293	3293	3293	Watt
HEATING	2378	2652	2743	Watt
	built	built	calcul.	

Total Inventory

Proposal of a Cryogenic Krypton Removal Facility

In conclusion of our work, our proposal for a cryogenic krypton removal plant for dissolver offgas is as follows (Fig. 9):

- Catalytic reduction of oxygen and nitrogen oxides with the assistance of a computer-supported H_2 control system.
- Pressure increase by means of a water ring compressor; a proven combination of materials in the slide bearings is special cast CrMo and Sb-impregnated hard carbon.
- Adsorptive retention of CO_2 and water by the molecular sieve. Flushing and regeneration of the adsorber bed can be carried out simultaneously with adsorption in the parallel tank. This saves one line, or allows one line to be kept for standby purposes.
- Zeolon 900H as an adsorber material proved to work just as satisfactorily as did the 10A molecular sieve, while the silicagel material tested is subject to accelerated aging in the presence of nitrogen oxide bearing offgas. For this case, a combination of Zeolon 900 H and a 10A molecular sieve is proposed.
- The cryogenic part of the facility is installed in a vacuum insulation tank, which makes for good accessibility.

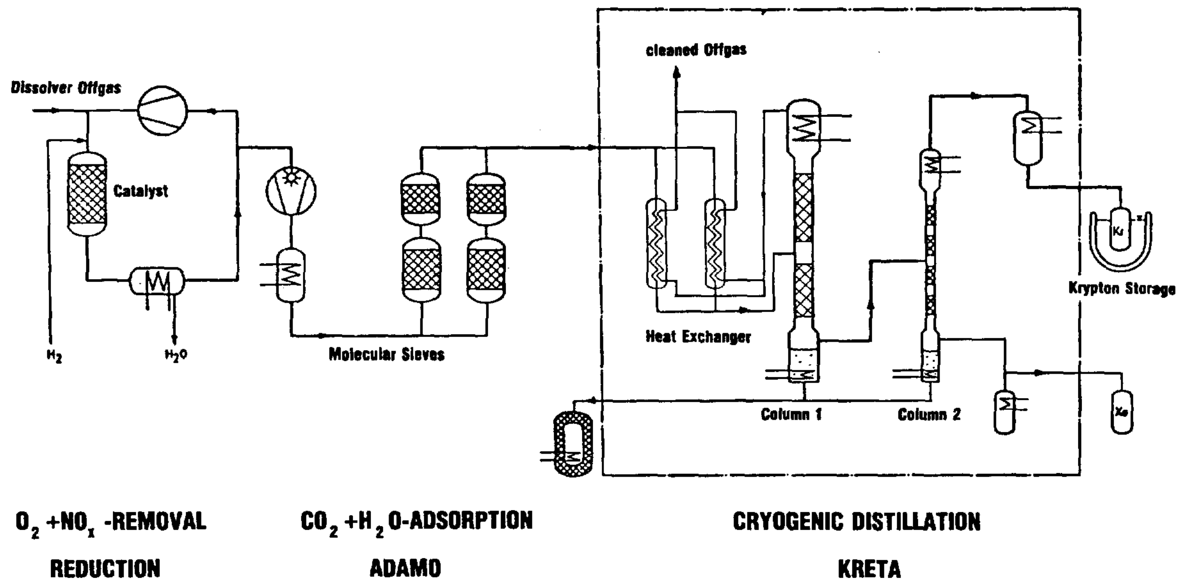


Fig. 9: KRYPTON RETENTION FROM DISSOLVER OFFGAS PROPOSED FLOWSHEET FOR A PILOTFACILITY

- For the inlet heat exchangers two parallel units are used, which are designed as a cold trap, can be switched and regenerated.
- For reasons of activity minimization, the first separation column is a packed column.
- The second column, also a packed column, is equipped with a sensitive control system for product extraction and pressure.

The next step now will have to be the construction of a pilot plant for active operation in conjunction with a reprocessing plant to allow radiological influences to be studied. This is being planned for the Wackersdorf facility; however, it is not yet certain which method of Kr removal will be employed.

References

(1) von Ammon R., Burkhardt H.-G., Hutter E., Neffe G., "Development of a cryogenic krypton separation system for the offgas of reprocessing plants" Prodeed. 15th DOE Nuclear Air Cleaning Conf. (First M.W.Ed.), CONF-780819, p. 640, 1979

(2) von Ammon R., Bumiller W., Hutter E., Neffe G., "Steady state operation of the first cryogenic column in a krypton separation system" Proceed. 16th DOE Nuclear Air Cleaning Conf. (First M.W.Ed.), CONF-801038, p.202, 1981

(3) von Ammon R., Bumiller W., Hauss E., Hutter E., Neffe G., Hammer R.R., "Formation and behaviour of nitric oxides in a cryogenic krypton separation system and consequences of using air as process gas" Proceed. 17th DOE Nuclear Air Cleaning Conf. (First M.W.Ed.), CONF-820833, p. 683, 1983

(4) von Ammon R. Bumiller W., Hutter E., Knittel G., Mas C., Neffe G., "Behaviour of impurities in a cryogenic krypton removal system" Proceed. 18th DOE Nuclear Airborn Waste Management and Air Cleaning Conf. (First M.W.Ed.), CONF-840806, p.951, 1985

(5) Ullmanns Enzyklopädie der Technischen Chemie, Vol. 2, 1972, p. 535.

KRYPTON RECOVERY BY CRYOGENIC DISTILLATION WITHOUT PRECEDING OXYGEN ELIMINATION

L. Geens, W.R.A. Goossens, J. Marien
S.C.K./C.E.N., Mol, Belgium

Abstract

By applying the proper operation mode for cyrogenic distillation, ozone accumulation can be reduced or even be prevented to the extent that oxygen might be allowed in the feed gas of a cryogenic krypton recovery unit in future installations.

The behaviour of oxygen and ozone in the S.C.K./C.E.N. cryogenic distillation unit was investigated. The presence of oxygen resulted in new temperature and concentration profiles in the first rectification column, without affecting the operation of this column. No oxygen was detected in the liquid krypton-xenon bottom product, so that no oxygen was transferred to the second batch distillation column. Ozone accumulated in the krypton-xenon mixture. Its decay was measured in absence and in presence of a thulium-170 radiation source. The higher decay rate, observed with the radiation source, indicates that ozone accumulation might be prevented by providing the proper radiation intensity in the kettle of the rectification column.

The feasibility of cryogenic distillation for krypton removal from an air carrier stream was demonstrated with an inventory of 4.44 TBq krypton-85, during a 900 hours campaign.

I. Introduction

Two operation modes are possible for the cryogenic recovery of krypton, depending on the absence or presence of oxygen in the feed gas of the cryodistillation unit. In the past, absence of oxygen was preferred in order to avoid the accumulation of ozone, which represents a serious explosion risk. Ozone was indeed observed to accumulate in the rectification column of the ICPP krypton recovery plant, which was fed with air ([1], [2], [3]). However, the removal of oxygen creates a similar safety problem by the use of a catalytic oxygen reduction process with hydrogen. Moreover, this process substantially increases the costs of an integrated off-gas cleaning system.

Presently, the second operation mode, allowing oxygen in the feed gas of the cyrogenic distillation unit, is more favoured. Laboratory experiments were carried out to investigate the formation and the destruction of ozone in a β-γ radiation field. These experiments revealed that the G-values for ozone destruction were much higher than for ozone formation, in both gaseous and liquid krypton-xenon mixtures ([5]). This information, combined with the operational control for a cryodistillation unit as developed at the S.C.K./C.E.N., indicates that the accumulation of ozone in the rectification column can be reduced to a great extent ([4], [6]). In this framework, the behaviour of both oxygen and ozone in the S.C.K./C.E.N. cryodistillation unit was studied. Particularly the ozone decay rate in the krypton-xenon bottom product of the rectification column has been measured. The influence of a radiation source on the decay rate was investigated. Finally, the feasibility of cryogenic krypton recovery from an air stream was demonstrated with radioactive krypton-85, during a campaign of limited duration.

II. The S.C.K./C.E.N. cryodistillation unit

Figure 1 shows a flow sheet of the cryogenic distillation unit, as it was developed at the S.C.K./C.E.N. in Mol. The unit essentially consists of two parallel heat exchangers, a first packed rectification column and a second packed batch distillation column, mounted in a insulated cold box.

The feed gas is cooled in the heat exchangers by the exhaust stream and by the vapours of the liquid nitrogen fed to the condensors of the distillation columns. Krypton and xenon are separated continuously from the feed gas in the first rectification column. The liquid krypton-xenon mixture is transferred from the first to the second distillation column, where both noble gases are separated from each other by batch distillation.

A more elaborate description of the unit and its operation mode has already been given earlier ([4]). Table 1 summarizes the main operational characteristics of the principal part, the rectification column.

Table 1 : Characteristics of the rectification column

Gas flow rate	:	25 $Nm^3.h^{-1}$
Operating pressure	:	0.70 MPa
Inlet temperature	:	124 - 134 K
Reflux ratio	:	0.3 - 1.0
Packing material	:	3 x 3 x 0.4 mm S.S. springs
Upper packing	:	h = 0.80 m, d = 0.10 m
Lower packing	:	h = 0.72 m, d = 0.07 m

III. Oxygen behaviour in the cryodistillation unit

Three different oxygen concentrations were fed to the cryodistillation unit : 6, 12 and 21 vol. %. This resulted in new temperature and concentration profiles in the rectification column, without affecting the operation of the column itself. Those profiles were already reported and commented in earlier publications ([7]).

In Table 2 the oxygen concentrations, measured in the gas phase, at the different sampling points of the first column are summarized. The oxygen concentration measured in the krypton-xenon bottom product was less thant 0.1 vpm, the detection limit of the oxygen monitor used. This means that no oxygen will be transferred to the second batch distillation colun, if also ozone accumulation can be avoided in this bottom product. The differences observed between the feed gas and the outlet gas concentrations were within the accuracy of the analytical method used.

Table 2 : Oxygen concentrations in the first column

Experiment number	Feed vol. %	Out vol. %	AP 2 vol. %	AP 7 vpm	Bottom vpm
1	6.0	6.2	7.0	< 0.1	< 0.1
2	12.0	12.1	13.0	< 0.1	< 0.1
3	20.9	20.9	22.8	< 0.1	< 0.1

Note : AP 2 = in the feed zone of the column
AP 7 = in the pure krypton zone, in the lower packing

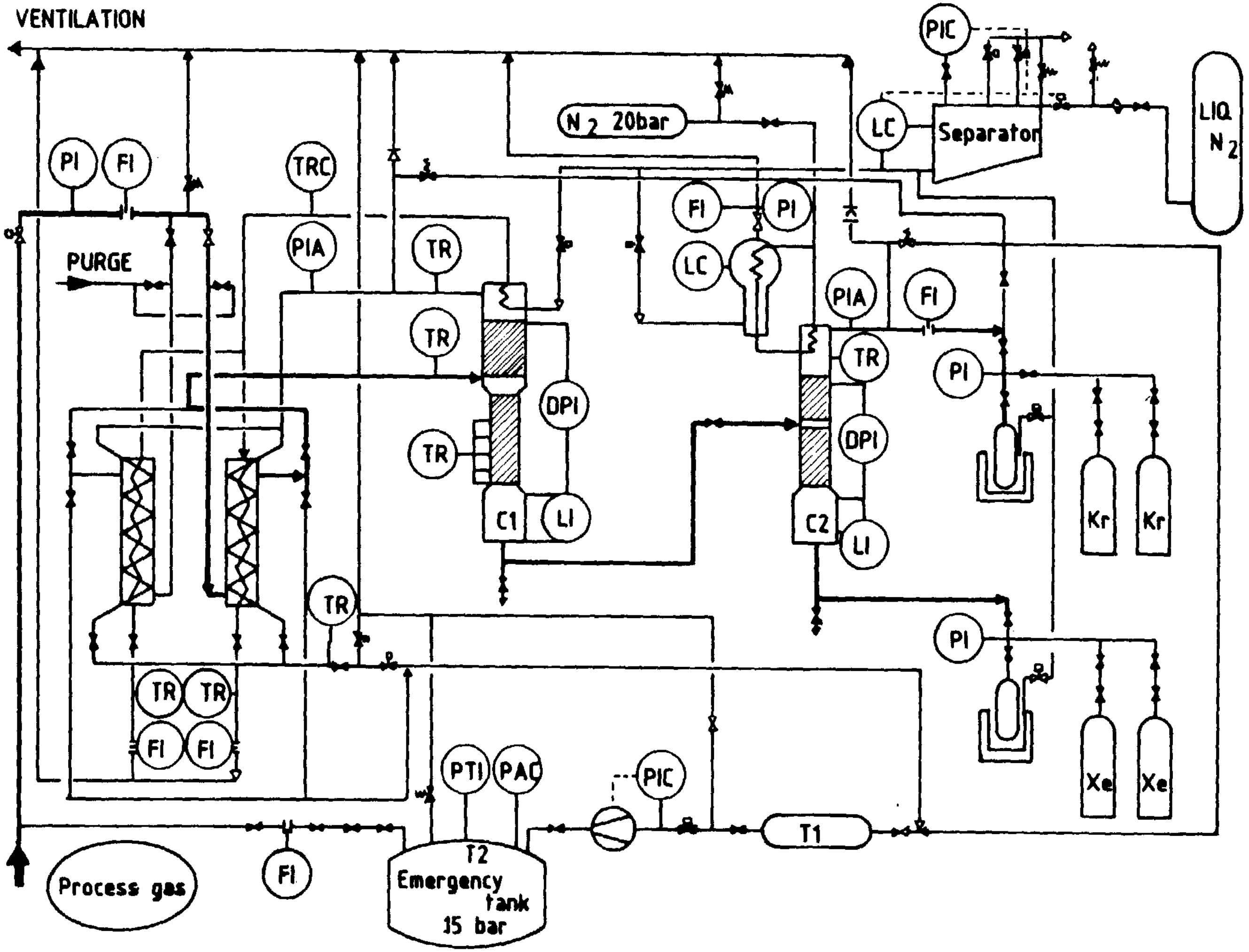


Fig. 1 : Cryogenic distillation unit

IV. Ozone behaviour in the rectification column

Ozone was fed to the rectification column at concentrations from 30 to 140 vpm, in an air carrier stream. No ozone was detected in the upper packing, nor in the outlet gas stream. All the ozone was found to accumulate in the liquid bottom product. For safety reasons, the accumulation was limited to about 1 vol. %. Table 3 summarizes the results of the ozone analyses at the different sampling points.

Table 3 : Ozone concentrations in the first column

Experiment number	Feed vpm	AP 4 vpm	Out vpm	Bottom
1	35	< 0.4	< 0.4	acc.
2	90	< 0.4	< 0.4	acc.
3	140	< 0.4	< 0.4	acc.

Note : AP 4 = in the upper packing
acc. = accumulation

Since ozone was accumulated in the krypton-xenon bottom liquid of the first column, its chemical behaviour there was investigated in greater detail. Particularly, to study the influence of the radiation field, the ozone decay rate was measured in absence and in presence of a thulium-170 source, which was installed in the kettle of the rectification column. Additionally, a short campaign was run to gain some information on the formation of ozone.

The use of the ^{170}Tm source for studying the influence of radiation on the ozone decay rate resulted from a survey of potential radiation sources. Taking into account the local implantation and shielding of the cryodistillation unit, a γ-emitter was excluded. For radioprotection reasons a β-emitter with limited γ- and bremsstrahlung was recommended, and on the basis of decay energy, half-life and availability considerations a thulium-170 source was chosen. The ^{170}Tm was obtained by irradation of the monoisotopic natural thulium metal (^{169}Tm, 100 %) in the BR2 reactor. By decay, the β-emitter ^{170}Tm is transformed to the stable ytterbium-170 isotope. The β-decay energies of ^{170}Tm are somewhat higher than those of ^{85}Kr : 23 % of 0.89 MeV and 77 % of 0.97 MeV, rather than 100 % of 0.67 MeV. On the other hand, for practical reasons the 129 days half-life of ^{170}Tm, is very well suited for the relatively short experiment which was planned to last 1000 hours. Besides its radiation properties, thulium metal has the advantage that it is chemically stable in the medium where it was to be used.

In Figure 2 the decay curves for ozone in the liquid bottom product are given, with and without the radiation source. Without the thulium-170 source, it took almost 30 days to observe a decay from 0.90 to 0.45 vol. %. The decay rate slightly decreased with decreasing ozone concentration. In the presence of a 18.5 TBq thulium-170 source an almost constant decay rate was observed, which resulted in a complete decay from about 1.05 vol. % to zero in 4 days. After two half life periods of the thulium-170, i.e. with a 4.63 TBq source, also a constant decay rate of about 0.20 vol. % per day was observed during the first days. Later on, the decay rate decreased with decreasing ozone concentration, which resulted in a complete decay from 1.05 to 0 vol. % in about 10 days.

The qualitative conclusion which can be drawn from these experiments, is that the possibility exists to prevent ozone accumulation in the bottom liquid of a cryodistillation column by providing the proper radiation intensity. However,

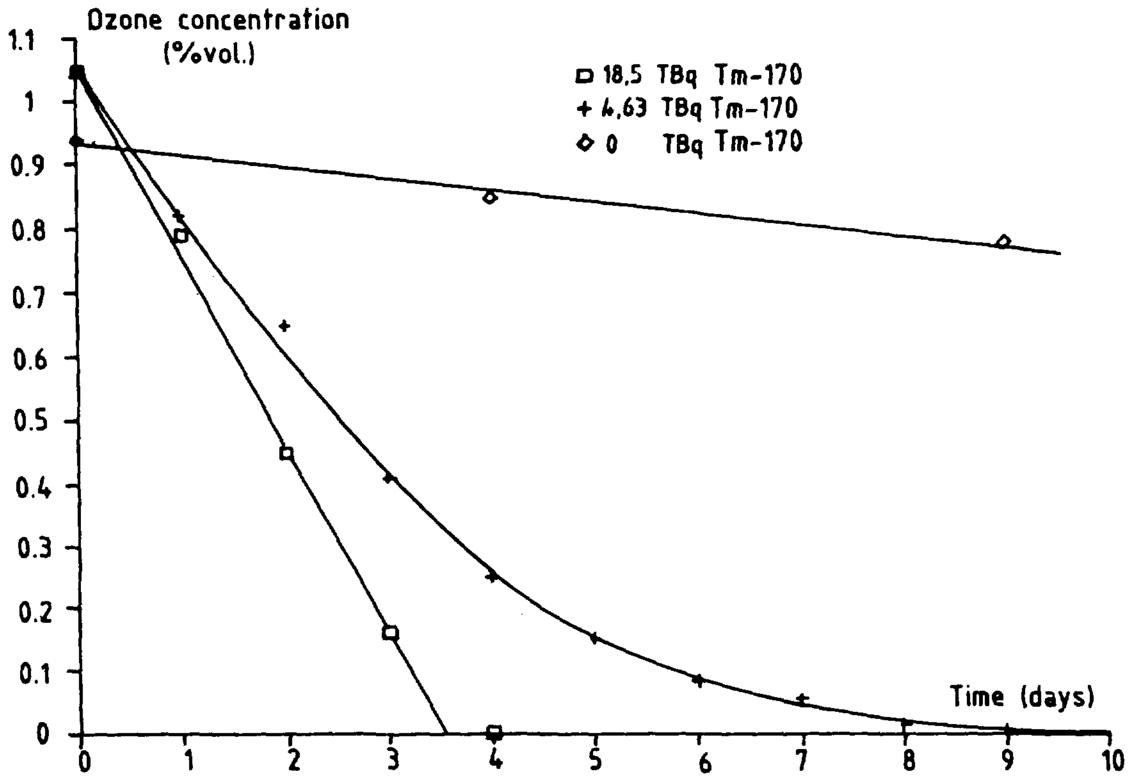


Fig. 2 : Ozone decay:influence of radiation

quantitative interpretation of these data is difficult. Indeed, there are too many unknown parameters, which influence the decay rate :

- the presence of radioactive by-products,
- the degree of dispersion of the ^{170}Tm source.

In addition to the ozone decay characteristics, also the ozone formation was investigated. In particular non standard operating conditions of the unit, oxygen was accumulated in the noble gas mixture in the kettle of the rectification column, up to 12 vol. % in presence of the 18.5 TBq thulium source. Even at this relatively high oxygen concentration, no ozone was detected after a period of 18 hours. This experiment was limited in time, since under the prevailing operating conditions, proper control of the column is very difficult.

V. Cryogenic distillation in presence of krypton-85

As the final step of the experimental programme the feasibility of cryogenic distillation in presence of oxygen was demonstrated under moderate radioactive condition, using 4.44 TBq krypton-85. For this purpose, both columns and all krypton and xenon containing bottles of the installation were shielded with 2 cm of lead. A 6 cm lead shielding as provided, for the kettle of the rectification column, which also contained the 18.5 TBq ^{170}Tm source.

In the experiment, radioactive krypton-85 was fed to the cryodistillation unit in an air carrier stream, at about 1.11 GBq.h^{-1}. The maximum inventory of

^{85}Kr in the rectification column amounted to 1.5-1.8 TBq. The campaign lasted for almost 900 hours, and no special problems due to the presence of the radioisotope were encountered. The only problems that occurred with the ^{85}Kr, concerned the transfer of the rather concentrated ^{85}Kr from the delivery containers into the storage bottles of the cryodistillation unit.

VI. Conclusions

The experiments with oxygen in the feed stream of the cryogenic distillation unit revealed that the operation of the unit was not influenced by the presence of oxygen, not even at concentrations corresponding to an air feed stream. Furthermore, in the liquid bottom product of the recification column no oxygen has been detected.

On the other hand, ozone was found to accumulate in the bottom liquid, Nevertheless, when its concentration was limited below 1 % volume, no operational problems occurred. After accumulation up to 1 % volume, its decay was measured in the absence and in the presence of a ^{170}Tm source. The conclusion of these experiments was that ozone accumulation can be prevented by providing the proper radiation intensity in the kettle of the rectification column. The observation of the oxygen and the ozone behaviour allows to conclude that no oxygen, neither as molecular oxygen, nor as ozone, will be transferred to the batch distillation column in this case.

Finally, the feasibility of cryodistillation for krypton recovery is presence of oxygen was demonstrated with 4.44 TBq krypton-85, during a 900 hours campaign.

References

[1] BENDIXSEN, C.L., GERMAN, F.O., "Operation of the ICPP rare gas recovery facility during fiscal year 1970", ICP-1001, 1971.

[2] BENDIXSEN, C.L., et al., "1972 Operation of the ICPP rare gas recovery facility", ICP-1023, 1973.

[3] BENDIXSEN, C.L., GERMAN, F.O., "1974 Operation of the ICPP rare gas recovery facility", ICP-1057, 1975.

[4] GEENS, L., et al., "Experience gained in the cryodistillation unit for krypton removal", BLG 547, 1981.

[5] HUYSKENS, P., et al., Personal communications.

[6] COLLARD, G.E.R., et al., "Recent developments in krypton removal by cryogenic distillation", Proc. CEC Specialists' Meeting on "Methods of Krypton-85 Management", Brussels, June 1982.

[7] GEENS, L.P., et al., "Alternative modes for cryogenic krypton removal", Proc. 18th DOE Nuclear Air Cleaning Conference, Baltimore, August 1984.

Acknowledgement

The authors would like to thank the C.E.C for their financial support, with which the experimental work, reported here, could be carried out.

PILOT PLANT DEVELOPMENT FOR ADSORPTIVE KRYPTON SEPARATION FROM DISSOLVER OFF-GAS

H. Ringel
Kernforschungsanlage Jülich,
Institut für Chemische Technologie

R. Printz
Wissenschaftlich-Technische Ingenieurberatung,
Titz-Rödingen

Federal Republic of Germany

Abstract

In view of hot cell application a separation process was investigated for the retention of Kr-85 from gaseous effluents. In the flow sheet only adsorption beds are applied. The most efficient process scheme is adsorption of the noble gas on activated charcoal and thereafter separation of the coadsorbed gas species like N_2, O_2, Xe and CO_2 from the krypton by gas chromatography. Adsorption is at normal pressure and low temperatures of up to -160°C, whereas desorption is at elevated temperatures and under helium purge. Influences on the process operation like off-gas composition, adsorption temperatures and adsorbent are experimentally investigated, as well as the behaviour of trace impurities in the adsorption columns. On the basis of pilot plant operation the main components for a full scale facility are being designed.

I. Introduction

In recent years at KFA Jülich a new process has been developed for the separation of radioactive noble gases from the gaseous effluents of nuclear installations. A specific goal is the retention of Kr-85 from the dissolver off-gas of a reprocessing facility, such as the planned German reprocessing plant at Wackersdorf. The entire flow sheet of this Kr-85 retention process is based on adsorption/desorption principles only and should therefore result in a simple and reliable process for hot cell application.

II. Process Principles

In a number of different laboratory experiments for adsorptive Kr separation from air, including: pressure swing, temperature swing and gas chromatography, a specific process scheme was selected (1,2). The main principles of the selected process are adsorption of krypton and xenon on activated charcoal at very low temperatures and regeneration of the adsorption beds by temperature rise and helium purge. Figure 1 shows the process principles. One process line consists of two adsorption columns of indentical design filled with chromatography-grade activated charcoal. During adsorption the first column is cooled to -130°C and the second one is cooled as low as -160°C.

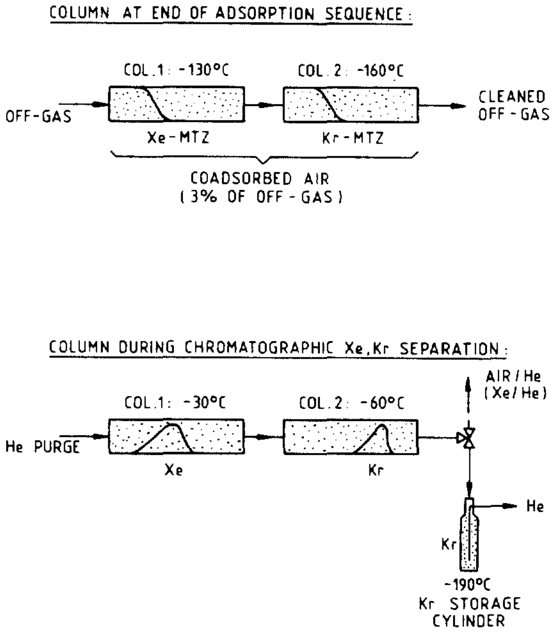


Figure 1: Process principle for adsorptive/chromatographic krypton separation process

Under these conditions Xe will adsorb completely in column 1 and the Kr will adsorb partly in column 1 and partly in column 2. At the end phase of the adsorption sequence the following gas distribution will be approximately present in the two columns:

The xenon is adsorbed in the first third of column 1 whereas about one half of the krypton is adsorbed across in column 1 and the other half part is adsorbed in the first third of column 2. Together with the noble gases, air is co-adsorbed across both columns and namely about 3 % of the full amount of the off-gas air will be adsorbed, i.e. 97 % of the off-gas was cleaned during the adsorption sequence and left the columns.

For column regeneration and final separation of the krypton from the co-adsorbed air, both columns are heated up in a specific manner, while being purged with a small amount of helium. These regeneration parameters are selected in such a way that a pure Kr/He mixture can be collected at the column exit.

Final Kr/He separation is done directly in the krypton storage cylinder. Therefore this storage cylinder is filled with activated charcoal as well and has to be cooled in the lower part with LN_2 for the filling operation. While the Kr/He gas mixture is fed into the top of the storage cylinder, the krypton is completely adsorbed on the charcoal and pure helium leaves the storage cylinder from the cylinder bottom by means of a lower exit pipe.

III. Laboratory Experiments

The results of laboratory experiments are reported in detail in ref. (2) and therefore they will only be summarized here. For the experiments adsorption columns as shown in Fig. 2 were used. The columns had an adsorption bed of 35 mm diameter and 500 mm length, filled with activated charcoal of .50 to .85 mm grain size. Two such columns form one separation unit. The pressure drops for both columns (including two heat exchangers) at adsorption temperature of -130°C and -160°C respectively and a typical gas flow of 1 m^3/h STP was .2 bar.

The parameters for a typical experiment were as follows:

- gas batch for one separation cycle: 2 m^3 STP
- gas composition: dry air with .01 vol% Kr and .10 vol% Xe
- adsorption time: 2 h
- gas flow during adsorption: 1 m^3/h STP
- temperature of column 1: -130°C
- temperature of column 2: -160°C
- pressure at column entrance: 1.2 bar
- pressure at column exit: 1.0 bar
- time for column regeneration (including subsequent time for column cooling): 2 h
- helium comsumption for regeneration: 4 % of gas batch
- decontamination factor for Kr: $\geq$ 1000
- concentration of separated Kr: $\leq$ 99 %

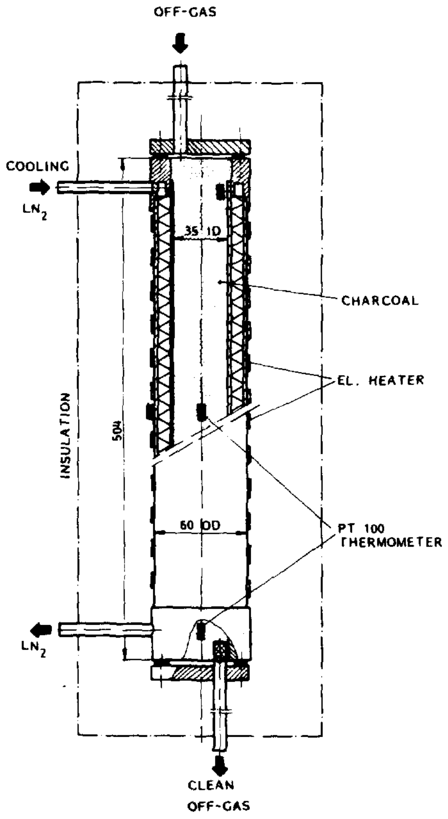


Figure 2: Kr separation column for laboratory experiments

The influences of the following operation parameters were investigated:

Off-Gas Composition

An increase of the typical noble gas concentration by a factor of 2 or 3 hardly influenced the separation results. Whereas a considerable noble gas increase to e.g. .2 vol% Kr and 2 vol% Xe does reduce the off-gas batch which can be cleaned in one process cycle.
Temporary high fluctuations of the noble gas concentration in the off-gas did not influence process operation; all these fluctuations will be levelled out in the adsorption columns during the adsorption sequence.

Adsorption Temperatures

In principle, the separation capacity increases e-functionally with decreasing adsorption temperatures; so the above mentioned column temperatures are the lowest temperatures possible for the following reasons:

For 1st column the lower temperature limit is about -130°C, because below that temperature xenon will freeze out. Even if plugging is avoided, the separation of Kr and N_2 from Xe is considerably impaired if the adsorption temperature is below -130°C because the xenon is too greatly polluted by Kr and N_2.

For the 2nd column the limit is about -160°C if 20 vol% O_2 is in the off-gas. Below -160°C this O_2 will condense in the charcoal micropores, so that the adsorption capacity of the charcoal for Kr is reduced. For off-gas without O_2 the temperature could be decreased as low as -180°C.

Behaviour of Trace Impurities

In the precleaned off-gas are - besides krypton and xenon - further gas species with small concentrations. The behaviour of the following gases in an adsorptive separation process was investigated:

- N_2O from dissolution process
- CO_2 mainly (300 ppm) from the dissolver purge air (containing some C-14)
- argon (about 1 vol% in air)
- CH_4 (ppm traces in air)

The operation diagram Figure 3 shows the behaviour of these gases in the experimental facility for one operation cycle. In order to achieve a distinct separation of all gas species only 1 m^3 STP off-gas was cleaned and a relatively small temperature increase, i.e. a long-lasting column regeneration programme, was chosen.

Argon behaves like N_2 and O_2, and is therefore not separated from the basic air species; N_2 and O_2 are also hardly separated from each other on charcoal. N_2O and CO_2 behave very similarly and therefore could only be separated from each other with an even longer temperature rise programme.

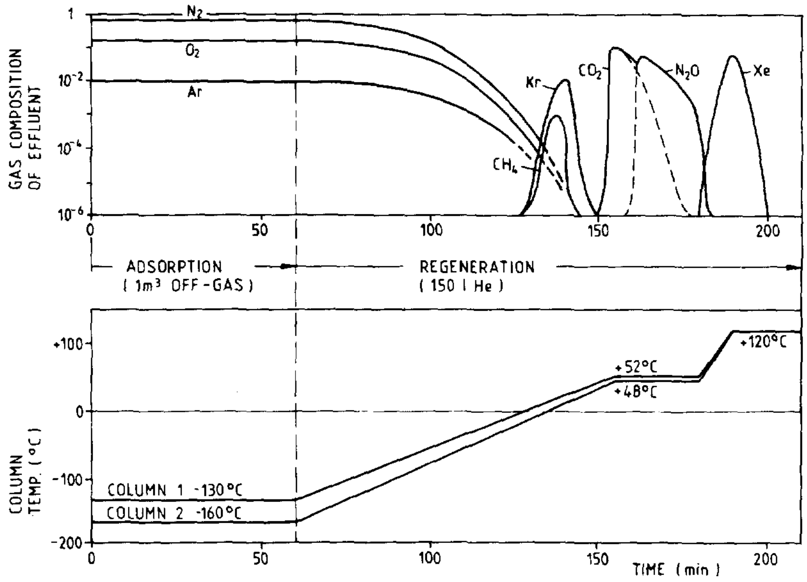


Figure 3: Behaviour of trace impurities in the activated charcoal columns during Kr separation process (dry air with 1000 ppm Xe, 1000 ppm N_2O, 100 ppm Kr, 10 ppm CH_4)

At normal process operation Xe, N_2 and CO_2 will not be separated from each other, i.e. these gases will be collected in one batch. They could be separated from each other by a different gas separation process if desirable. The methane trace is not separated from krypton, i.e. it will be flushed from the column at each operation cycle and therefore does not represent a safety risk.

Selection of Adsorbent

The typical adsorbent throughout the experiments was a gas chromatography grade activated charcoal with the following specifications:
- grain diameter: .5 to .85 mm
- poured density: 440 g/l
- specific surface: 1050 m^3/g

This charcoal was most efficient for chromatographic gas separation. But due to its relatively small grain size the pressure drop during adsorption is high, so that for technical application a charcoal with larger particles might be more economical.

Apart from this, for safety considerations in hot cell application one would consider applying a ceramic adsorbent instead of activated charcoal. The experimental investigation showed that for large-scale Kr/N_2 separation charcoal can hardly be replaced by molecular sieve. In contrast to that, for preseparation of Xe 5A molecular sieve could be used. This would result in a process flow sheet with one enlarged molecular sieve and one charcoal column.

Ag exchanged molecular sieve showed a very poor adsorption capacity for Kr and is therefore not practical.

IV. Pilot Plant Experiments

For full-scale plant developments adsorptive off-gas cleaning will be experimentally tested for an off-gas flow of up to 10 m^3/h STP, and both for NO_x, H_2O separation as well as for Kr, Xe separation.

NO_x, H_2O Separation

After precleaning, (NO_x wash column, aerosol and iodine filtration) the dissolver off-gas will still contain, besides the noble gases, NO_x and H_2O residues in the order of .5 vol% NO_x and 2 vol% H_2O. This water will also be contaminated with some tritium, as HTO.

For the separation of these species the adsorptive retention on molecular sieve was investigated on a laboratory scale (3).

Applying the selected acid resistance molecular sieve an appropriate process was tested on a 1 m^3/h STP off-gas scale. The flowsheet consisted mainly of two adsorption columns in parallel arrangement so that the simulated off-gas could be cleaned continuously. The main test parameters were as follows:

- column dimensions: 64 mm ID., 430 mm length
- molecular sieve: "900 H Norten", SiO_2/Al_2O_3 = 10/1, 8-9 Å, 1.8 mm DIA. extrudates
- gas flow: 970 l/h STP
- gas composition: wet air with .51 vol% NO_x and .5 vol% H_2O
- time for 1 ppm NO_2 breakthrough: 10.7 h
- time for breakthrough of 1/2 NO_2 inlet concentration: 14.2 h

From these data, one can calculate the length of the MTZ as 210 mm and the dynamic loading as 47 l STP NO_2/l molecular sieve.

The experiments showed that NO_2 would always break through before NO breakthrough occurred, which is a strong indication that in the column all NO of the gas will be oxidized to NO_2 and afterwards adsorbed.

Regeneration of the molecular sieve was accomplished by heating to 250°C and flushing the column with a small amount of air. For up to 50 adsorption/desorption cycles the adsorption characteristics of the molecular sieve did not change.

Kr and Xe Separation from Off-Gas

The final krypton/air separation is accomplished by gas chromatography. For this process sequence the separation column is purged with helium and heated up in a certain way. Since only a very small helium flow is needed, the column has to be heated by heat conductivity from the column outside. In order to have an undisturbed gas separation process during this heat-up it is most important to achieve a relatively small temperature gradient perpendicular to the gas flow, i.e. in the cross section of the column. The poor heat conductivity of activated charcoal cannot fulfil this process demand.

Therefore, for the process scale-up a different column design was chosen with respect to the laboratory-scale column. A cross section of this pilot plant column is given in Figure 4. In principle the charcoal bed is shaped like an annulus, with an annulus thickness of 17.5 mm. The inner and outer wall of the annulus can be cooled by a cooling medium and heated by an electric resistance heater, which is integrated into both walls. With the dimension of the charcoal bed being OD = 60 mm, ID = 25 mm, and length = 615 mm one separation column would contain 1.45 l charcoal.

With two of these columns in series an off-gas flow of 3 m^3/h STP containing air with .1 vol% Xe and .01 vol% Kr could be processed. The result of a gas separation experiment is shown by the operation diagram in Figure 5.

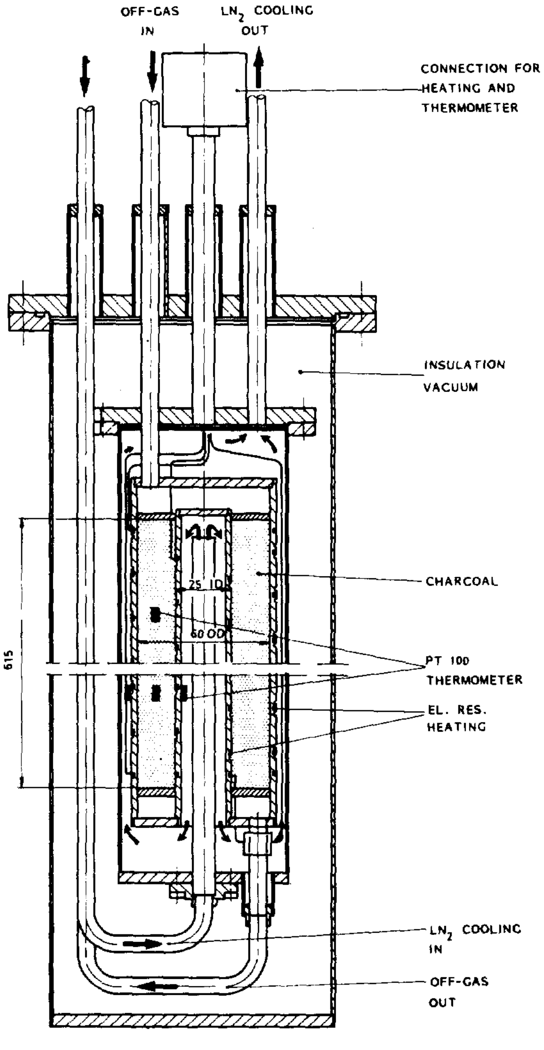


Figure 4: Kr separation column for pilot plant

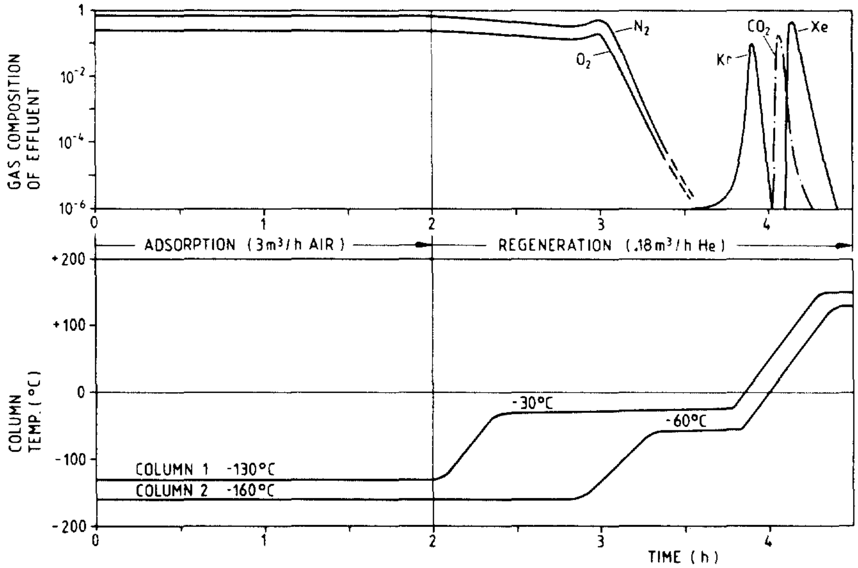


Figure 5: Operation diagram for cleaning 6 m^3 STP off-gas in pilot plant (2 columns with 1.45 l activated charcoal each)

During the adsorption sequence of two hours 6 m^3 STP of off-gas flowed through the columns. Pressure at the column entrance was 1.2 bar, the off-gas temperatures were -130°C and -160°C, respectively. During column regeneration and gas chromatography sequence the helium flow was 3 l/min STP, so that for 2.5 h regeneration time 450 l He was used. The separated Kr batch contained about 1 vol% N_2, the Kr decontamination factor was determined as about 1000. The accuracy of these two values is determined by the accuracy of the quadrupole mass spectrometer which was applied for gas analysis. For Kr the detection limit was about 1 ppm and for air (N_2, O_2) some 100 ppm.

Subpressure Operation

For hot cell operation, it is typically favourable if a process operates at a pressure below atmospheric. Therefore adsorptive Kr separation was also tested under subpressure conditions.

In this case, the off-gas was sucked through the two-column arrangement by a vacuum pump at the exit of the second column. For 3 m^3/h STP off-gas flow and process temperatures in the columns, pressure at the column exit was .6 bar if pressure at the column entrance was 1.0 bar, i.e. the pressure difference was doubled with respect to operation at normal pressure at the column exit.

The results for the gas separation was in principle similar for both cases, i.e. identical gas batches could be processed. A main difference was noted for the second part of the chromatography sequence. About 10 min before full Kr breakthrough some Kr (up to 100 ppm) would elude from the column together with the N_2/He effluent. In order to reduce or to avoid this Kr leakage the heating rate during column regeneration has to be reduced, which results in a longer regeneration time or a few litres of Kr-contaminated effluent have to be recycled.

Apart from the column for 3 m^3/h STP gas flow, columns for 10 m^3/h STP with an identical design principle to that shown in Figure 4 have been constructed. Testing of these columns has yet to be done.

V. Engineering Design for Full-Scale Plant

Adsorptive Kr-85 separation from dissolver off-gas is a batchwise process as well as fuel dissolving itself. Therefore it might be favourable to adapt the sequence of this Kr-85 separation process to the sequence of the dissolution process. The main point is that only fuel element chopping and primer dessolution are the principle process steps when Kr-85 is liberated into the dissolver off-gas. During the other dissolution process steps, like: - secondary dissolution, iodine stripping, dissolver cooling and emptying - no Kr-85 is set free and therefore for this sequence no krypton cleaning of the off-gas would be necessary.

For design purposes the following criteria can be assumed:

- heavy metal batch for one dissolution: 1000 kg
- krypton content for typical burn-up (40,000 MWd/t): 120 l Kr STP
- Kr-85 content: 8800 Ci Kr-85 (5 % Kr-85 after 7 a interim storage)
- time for Kr-85 liberation: 6 h
- time for full dissolving cycle: 12 h
- average off-gas flow: 100 m^3/h STP
- average Kr content in off-gas: .02 vol%
- cylinder size for interim of final Kr-85 storage: 50 l volume for 200,000 Ci Kr-85 (4)

A flow sheet for a full-scale krypton retention facility for the off-gas line of one dissolver is shown in Figure 6. It is assumed that NO_x and H_2O (and HTO) are separated continuously by batchwise operation of two molecular sieve units whereas only one krypton separation unit is necessary.

H_2O/NO_x Column Design

After off-gas precleaning the moisture amounts to 2.5 vol% H_2O and the NO_x concentration may vary between 0.04 vol% and o.3 vol%. Before separating the krypton from the dissolver off-gas, these NO_x and H_2O concentrations must be reduced to values below 1 ppm.

In order to minimize the adsorber volume required for H_2O/NO_x adsorption, the off-gas moisture is reduced to approx. 1 vol% by a condenser before entering the NO_x/H_2O adsorption columns. In this case the off-gas must be cooled to approx. 6°C.

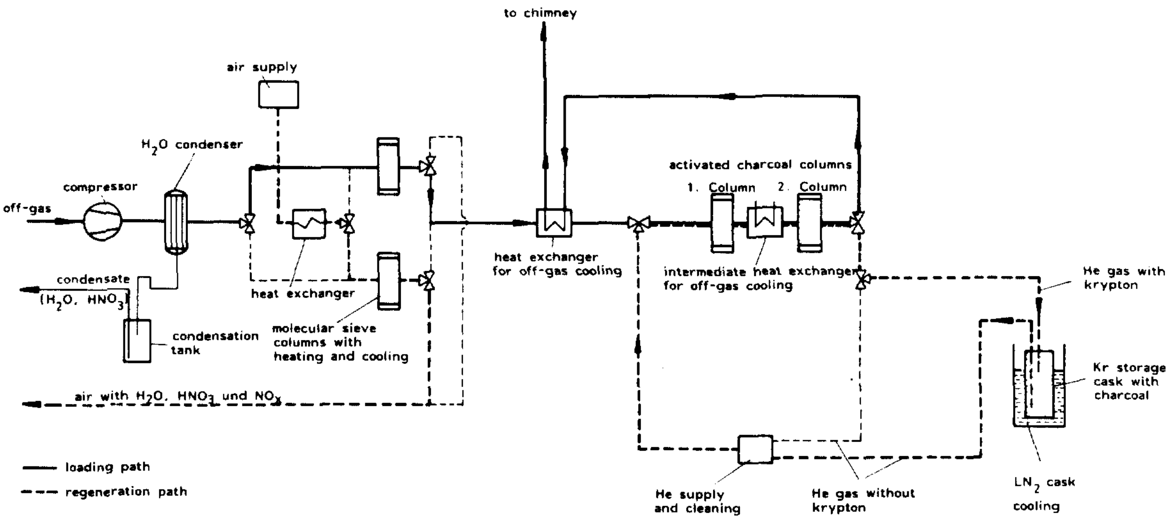


Figure 6: Flow sheet for full-scale NO_x, H_2O and Kr separation for one dissolver off-gas line

Adsorptive H_2O/NO_x separation is carried out by 19 molecular sieve columns connected in parallel with a height of 1400 mm and an internal diameter of 80 mm each. A heating/cooling medium (e.g. steam or helium) is circulated around the columns for cooling and heating. The basic design of the H_2O/NO_x adsorption column corresponds more or less to the Kr adsorption column as shown in Fig. 7. One complete unit is approx. 2 m in height and approx. 0.6 m in diameter.

Two such units are required for continuous NO_x/H_2O separation; they are loaded and regenerated alternately in a 24-hour cycle, i.e. 12 hour for loading and 12 hour for regeneration.

Kr Column Design

The Kr separation from an off-gas flow of up to 50 m^3/h STP could be carried out in a separation unit which is designed similar to the design as shown in Fig. 4. But for even higher off-gas flows parallel arrangements of such units would be necessary. This seems to be too complicated. Therefore for high off-gas flows a parallel connection of columns as shown in Fig. 2 will be more practical. For 100 m^3/h STP dissolver off-gas some 99 columns with an activated charcoal volume of altogether approx. 130 l are required. The dimension of one column is 35 mm ID and 1350 mm length.

The design of the two adsorption units required for one process section is identical and shown in Fig. 7. Helium is fed into the units for cooling and heating, and flows around the activated charcoal columns. So-called baffle plates are provided to improve a regular heat transfer.

The Kr separation units are designed for a 6-hour loading phase followed by 6 hours of regeneration so that continuous operation is possible with 4 units whereas 2 units are connected in series. Fig. 8 shows a diagram for the process sequence of the Kr separation plant. The heating and cooling times selected were checked by

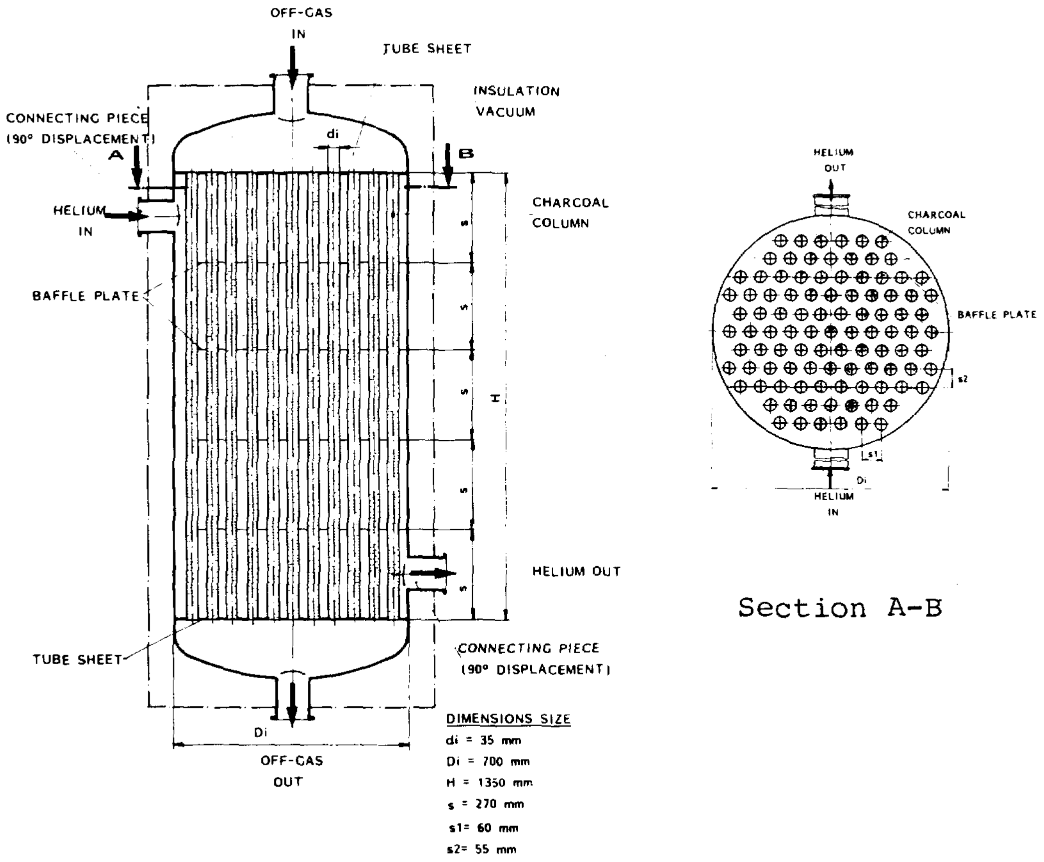


Figure 7: Frontal section and cross section of the full-scale Kr separation units

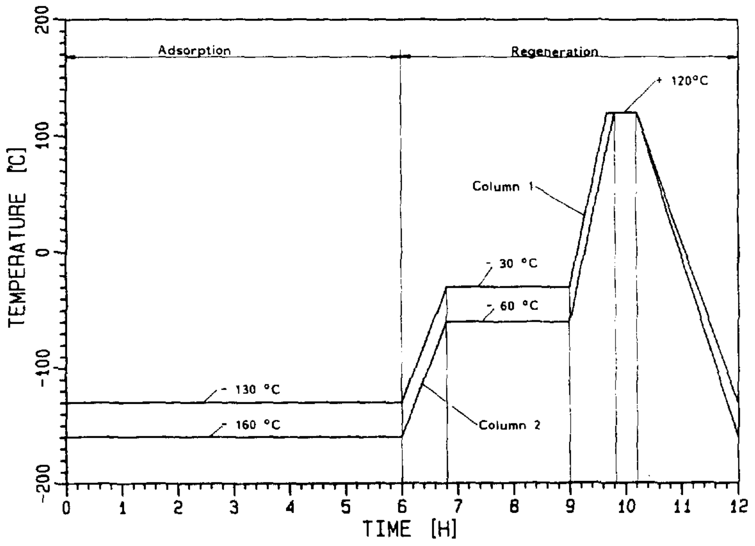


Figure 8: Diagram for process sequence for full-scale separation plant for one dissolver off-gas line

thermal calculations and can be achieved with the selected helium heating/cooling system.

The total Kr-85 activity inventory in the separation columns amounts to approx. 8800 Ci after the 6-hour loading phase. During the column regeneration phase the Kr/He flow is fed into a storage cylinder filled with 50 l of activated charcoal and partly cooled by LN_2, so that the Kr is adsorbed again on activated charcoal. One 50 l storage cylinder is provided for the storage of 200,000 Ci of Kr-85 so that one storage cask is full after approx. 22 loading operations.

Acknowledgments

We would like to thank Mr. Frommelt and Mr. K. Hein for their experimental assistance.

References

(1) Ringel, H., Meßler, M. "Chromatographic Separation of Krypton from Dissolver Off-Gas at Low Temperatures" 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, CONF - 840806, Vol 2, 982-995 (1985)

(2) Meßler, M. "Entwicklung eines Verfahrens zur adsorptiven Abtrennung von Krypton-85" Report: Jül-1988 (1985)

(3) Ringel, H. "Adsorptive Abtrennung von NO_x aus dem Auflöserabgas" Report: JÜl-1930 (1984)

(4) Niephaus, D. "Sicherheitstechnische Untersuchungen und Störfallanalysen zu Beseitigungsmethoden des aus dem Auflöserabgas einer Wiederaufbereitungsanlage für LWR-Kernbrennstoffe abgetrennten Krypton-85" Report: Jül-1765 (1982)

DISCUSSION

JUBIN: You indicated that you were unable to separate N_2 and Kr using a silver exchanged zeolite. Were you able to observe any separation between Xe and Kr using the silver zeolites?

RINGEL: We made no specific experiments concerning this question, but I am sure that Xe and Kr can be separated using silver exchanged zeolite.

GRUNDLER: How much are the costs for a large scale Kr^{85} separation facility?

RINGEL: We made no cost calculation.

HUTTER: We made no cost estimates, but there are cost values from AZUR planning for the WAK reprocessing plant at KfK. Also, the Japan Token Mura reprocessing plant has a cryogenic, retention facility ready to work with Kr^{85}

CONTAINMENT OF KRYPTON-85 BY AN ION-IMPLANTATION/SPUTTERING PROCESS USING RARE EARTH ALLOYS

E.D. McClanahan, R.W. Moss, E.N. Greenwell, E.R. Bradley and E.L. McDonald
Pacific Northwest Laboratory*
Richland, Washington

Abstract

The design of a remotely operable ion-implantation/sputtering process for containment of krypton-85 in a solid metal alloy matrix is described. The process operates at subatmospheric pressure and is compatible with plants for reprocessing irradiated nuclear fuel that range in size up to 2000 metric tons of heavy metal per year. Process capacity is determined by the size, operating power and number of individual krypton-trapping storage devices (KTSDs) that can be operated simultaneously in parallel.

A 65-kW capacity nonradioactive multiple storage device unit was constructed using three unshielded test modules. The KTSDs contained composite targets with copper-yttrium surface area ratios of 1 and 5 to provide the matrix for the krypton product. Two of the KTSDs, with substrate surface areas of 1500 and 1000 cm^2, were operated simultaneously for over 3 h on a common manifold without an auxiliary vacuum pump. The other, a 1000-cm^2 unit, was tested for 1 h.

Krypton was incorporated at rates of 1.5 liters (STP) per hour for the larger and 1.0 and 0.7 liters (STP) per hour for the smaller KTSDs, respectively. A higher yttrium content in one of the smaller KTSD's target was responsible for the greater trapping rate in the smaller units. The energy consumption was 16 kWh per liter for the 1500-cm^2 substrate KTSD and was 20 and 26 kWh per liter for the two smaller units; the unit with the higher energy consumption had the lower yttrium content target.

Radioactive tests, using a total of 382 curies of a krypton mixture containing 4.2% krypton-85, were performed to verify that the β particle flux did not adversely affect the process and to provide representative krypton-85 storage product for long-term elevated-temperature testing. Three bench-scale KTSDs, with a substrate area of 345 cm^2 and containing 80 to 150 curies each of krypton-85 implanted in a copper-yttrium alloy, are being stored at temperatures ranging from 150 to 350°C. Periodic sampling is in progress.

---

* Operated for the U. S. Department of Energy by Battelle Memorial Institute.

I. Introduction

Current U.S. regulations limit the release of krypton-85 to 50,000 Ci/10^9 watt years of electrical energy, or about 15% of the total krypton-85 produced in light water reactor (LWR) fuels.(1) To meet these requirements, storage in pressurized cylinders is the only available method.(2,3) To improve safety and increase economy, the Department of Energy chose to develop and evaluate other concepts for krypton immobilization.

Pacific Northwest Laboratory (PNL) has completed a program to develop ion implantation of krypton in a rapidly growing sputter-deposited metal as the primary method for producing the ultimate waste form for the storage of radioactive krypton. Bench-scale work begun in July of 1976 indicated that this process was well suited for producing thermally stable solids containing an excess of 5 at.% krypton.(4) It was shown experimentally that the highest krypton concentrations and lowest energy requirements were obtained by implanting the krypton in amorphous alloys.(5) A recent report summarizes the selection of alloys for this process.(6)

A study comprising preconceptual engineering design, technical evaluation, and life-cycle costing data was completed for comparing ion implantation/sputtering and zeolite encapsulation of krypton-85.(7) The study results were presented, and a review panel appointed by the DOE Airborne Waste Management Program Office recommended that the ion implantation process be selected for pilot-plant implementation. The main advantages of the process are:

- It operates at subatmospheric pressure.
- It permits high krypton loading.
- It produces a waste form that is thermally stable and has high thermal conductivity.

The original objective of the krypton ion implantation program was to design, construct and demonstrate the operation of a full-scale remotely operable nonradioactive immobilization unit. Later this objective was modified to include design of a smaller KTSD suitable for use with a Breeder Reprocessing Engineering Test (BRET)-scale reprocessing pilot plant (~15 MTHM/yr.) Construction was limited to fabricating three unshielded KTSDs connected to a common manifold to demonstrate process operations. A final task was added to carry out three multicurie laboratory-scale krypton-85 implantation experiments to provide small KTSDs for long-term thermal release tests for a copper-yttrium-krypton matrix.

The work reported here covers three topics:

- design of a remotely operated krypton immobilization station compatible with two sizes of KTSDs

- operation of a multiple-module unshielded nonradioactive krypton immobilization station with a one liter per hour trapping rate
- the results of three fully radioactive bench-scale test runs, in which a total of 382 curies of a 4.2% krypton-85 mixture were immobilized.

The operating principle and design concepts are described in Section II. The design of the remotely operable station is found in Section III. The unshielded multiple-module station and the results of its operation are discussed in Sections IV and V. The radioactive bench-scale tests and provisions for long-term elevated-temperature storage tests are described in Sections VI and VII.

II. Operating Principles and Design Concept

Because krypton does not form stable compounds with other elements, successful isolation of large quantities of krypton-85 in a stable solid requires an efficient process that "glues" the krypton atoms to a surface long enough to cover them with other atoms.

Trapping gases in metals exposed to a gas discharge is almost as old as sputtering.(8) Commercially manufactured ion pumps are widely used in industry for achieving an oil-free ultra-high vacuum. Unfortunately, these pumps are unsuited for immobilizing krypton-85 recovered from irradiated nuclear fuel because their design was optimized to pump active gases in a very low pressure regime ($<1 \times 10^{-3}$ Pa).

Krypton can be "glued" to a substrate by first ionizing it and then implanting it with the aid of an electric field. If the substrate is a growing film of suitable material, concentrations of krypton exceeding several atomic percent can be trapped in this solid matrix.(9) Resputtering and reemission of the krypton can be minimized if the implantation energy can be kept low. Since this form of "glue" requires ionized krypton atoms, it makes sense to use the krypton ions to sputter-deposit the matrix atoms also. This may be conveniently accomplished by placing the collecting surface (substrate) and the sputtering target (matrix atom source) in a concentric arrangement. Figure 1 provides a simplified depiction. A reasonable collection efficiency is obtained for the sputtered matrix atoms if the gap between them is not too large. The gap, however, must be at least several centimeters to produce the discharge. Concentric cylinders and hemishells or pipe caps make strong, convenient shapes for substrates.

The simplest way to produce krypton ions in the space between the substrate and the target is to establish a glow discharge in the annulus filled with krypton at a pressure of ~10 Pa by applying a minus 3 to 5 kV to one of the electrodes. Whitmell in the United Kingdom demonstrated a half-scale pilot plant for immobilizing krypton in sputtered copper.(10) The U.K. process works by alternately implanting krypton in the substrate (outer electrode) and sputtering metal from the target (inner electrode) to coat the implanted layer. Trapping

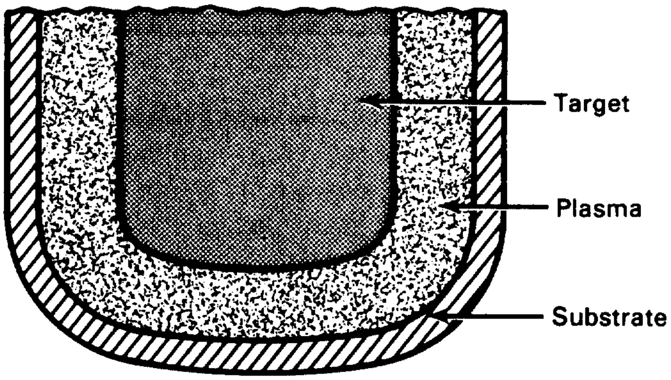


FIGURE 1. Concentric Arrangement of Collecting Surface and Sputtering Target

rate is controlled by a special thyratron switch controlling the relative times and voltages used for each phase.

A more efficient way to produce positive ions, and the scheme chosen by PNL, is with a low-voltage, thermionically supported discharge. This scheme has several advantages. First, the ion density can be controlled independently of the accelerating voltages placed on the target and the substrate. But more important, the ion density can be increased by a factor of ten or more over that obtained in the glow discharge. Also the presence of a "full"-time anode means the negative bias or ion-accelerating potential on the substrate is continuous, hence more efficient. The thermionically supported or so-called triode discharge is slightly more complicated by the need for additional electrodes (anode and filament) and their respective power supplies. This complexity makes it harder to build and easier to short out by film buildup or deposit flaking.

Regardless of the discharge scheme used, the object is to come up with an economical long-lived disposable trapping chamber with a high krypton trapping rate, capacity, and stable product. The filling process would involve the following steps: 1) evacuating of the chamber, 2) backfilling the chamber with inactive krypton and establishing the discharge, 3) ion etching the substrate, 4) establishing full-power pumping conditions, 5) switching to active krypton-85 and continuing to full capacity, and then 6) switching back to inactive krypton to apply a final closeout layer. For redundancy, ease of operation and flexible capacity, several small units should be operated in parallel rather than a single large unit. A krypton-85 immobilization plant would consist of one or more banks of the multiple-module KTSD stations. With appropriate biological shielding, it should be possible to maintain most of the equipment by direct contact.

Once filled, the KTSD would be disconnected and a protective pipe cap placed over the electrode flange and welded. After helium leak-testing, the unit would be stored for a time and then transported probably to a SURFF-type (spent unreprocessed fuel facility) dry well.(11)

III. Description of Full-Scale Remotely Operable Ion-Implantation/Sputtering Station

At PNL the objective was to develop a dual design which would immobilize the krypton-85 from a full-scale commercial fuel reprocessing plant (2000 metric tonnes of heavy metal per year) or a demonstration pilot plant (e.g., BRET) with a capacity of ~15 metric tonnes of heavy metal per year. The krypton is assumed to be 6% krypton-85 with an overall purity of 95%. To meet this range in capacity, two sizes of KTSDs were designed, with substrate areas of 3000 and 1000 cm^2. Each has a diameter of 30 cm and lengths of 30 and 10 cm, respectively.

Figure 2 is a cutaway drawing of the 1000-cm^2 substrate KTSD, a size which was built and used experimentally. Essential to the device is the centrally located water-cooled target made of copper or of a transition metal and filled with press-fitted plugs of yttrium, zirconium, or lanthanum. The composition of the alloy matrix is determined by the surface area and the sputtering yields of the two metals at the target-operating potential (~2400 V). The substrate surrounds the target concentrically. A single-turn tungsten filament, supported by six tantalum posts, and a cylindrical tantalum anode, located adjacent to the filament, serve as the auxiliary electron source and collector. The upper flange of the apparatus completes the vacuum envelope and supports the electrically insulated feed-throughs for the various electrodes. The upper flange and substrate, like the target, are water-cooled, while the other electrodes are uncooled. Allowing the anode and filament supports to run near the vaporization temperature for the matrix alloy minimizes the amount of sputter-deposited material available for shorting the closely spaced electrodes. The krypton gas is fed into the KTSD opposite the manifold port connection to give good control of pressure in the chamber. It is necessary to electrically isolate the krypton feed line from the substrate.

Figure 3 illustrates how the individual KTSD can be placed into a shielded carrier. The carrier can be moved laterally on rails to a location from which it can be raised in order to mate with the shielded service connector. In a single motion the KTSD is automatically connected to the pumping manifold, the krypton feed line, and all the necessary utility connections.

The critical parameters for scaling the size of the plant and providing the data for estimating the equipment, materials, and utility requirements are the KTSD pumping speed, energy consumption rate, and total krypton capacity. Formulas for calculating KTSD performance are given below. The estimated performance data for the 1000- and the 3000-cm^2 substrate KTSDs for $Ni_{.81}$-$La_{.09}$-$Kr_{.10}$ and $Cu_{.67}$-$Y_{.25}$-$Kr_{.08}$ are given in Table 1. It is assumed that each KTSD contains a uniform deposit 1 cm thick. The dimensions of the KTSDs, the sputtering parameters, and other pertinent data are found in the Appendix.

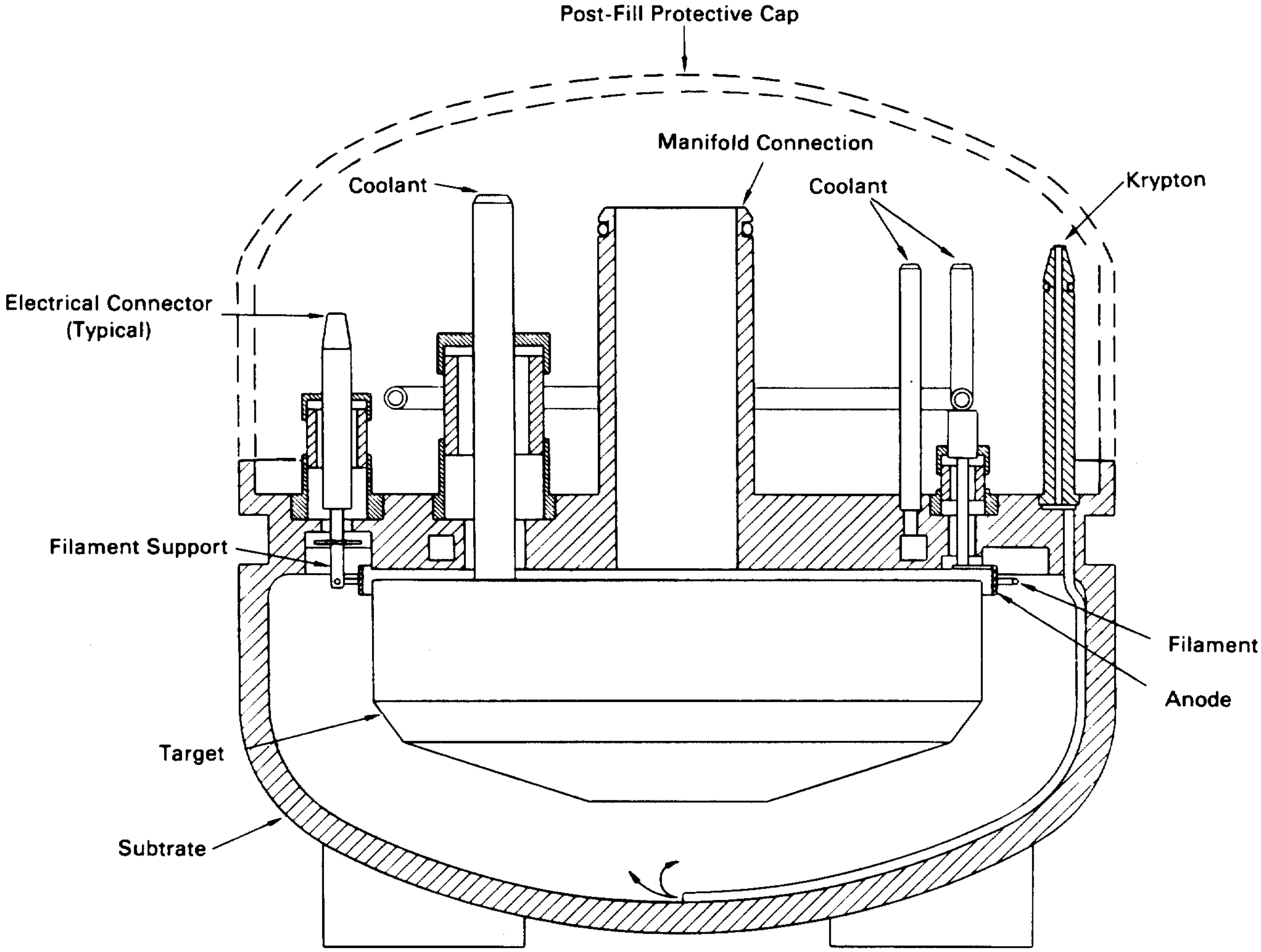


FIGURE 2. 1000-cm^2 Substrate Disposable KTSD

The trapping rate, S, of the KTSD is given by the following equation:

$$S = \frac{22.4\ j_+\ A_t\ Y\ C_{kr} K}{84} \quad \text{(L/h) (at STP)}$$

where:
j_+ = positive ion current density
A_t = area of the target
Y = net sputtering yield (grams/A h)
C_{kr} = weight percent of Kr trapped in the deposit
K = net fraction of the sputtered deposit collected by substrate.

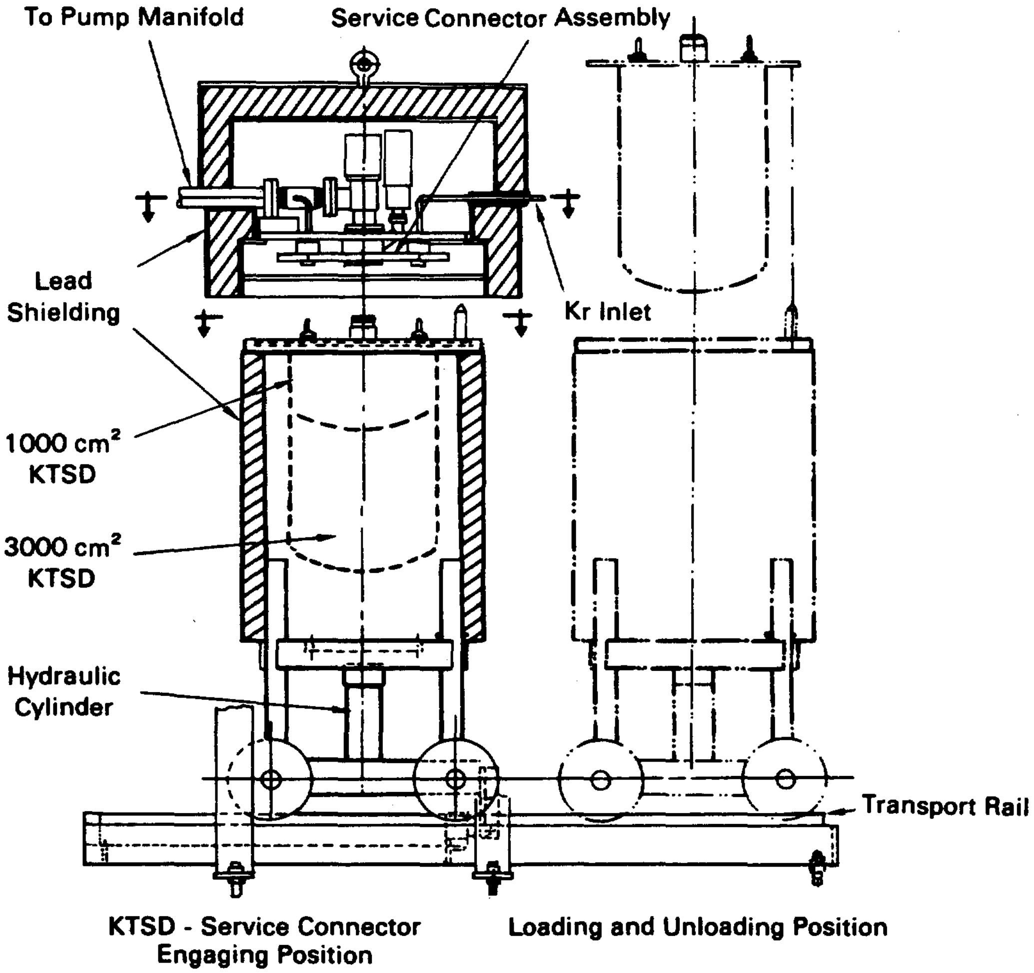


FIGURE 3. Sputtering Station, Showing Shielded Carrier, KTSD, and Shielded Service Connector

TABLE 1. Estimated Performance Data for KTSD.

	$Ni_{.81}La_{.08}Kr_{.10}$		$Cu_{.67}Y_{.25}Kr_{.08}$	
Substrate Area, cm^2	1000	3000	1000	3000
Trapping Rate, S (L/h) (at STP)	1.4	5.4	1.0	3.9
Energy Consumption, E (kWh/L)	12	11	16	15
Storage Capacity, C (L)	199	616	117	414

The energy consumption, E, for the KTSD is given by the following equation:

$$E = \frac{V_tI_t + V_sI_s + V_pI_p + V_iI_i + V_fI_f}{S} \quad \text{(kWh/L)}$$

where: V_t, V_s, V_p, V_i and V_f are the voltages applied to the target, substrate, plasma, ignitor, and filament, respectively.

I_t, I_s, I_p, I_i, and I_f are the currents flowing through the target, substrate, plasma, ignitor, and filament, respectively.

The capacity for the KTSD is given by the following equation:

$$\text{Capacity, } C = \frac{22.4}{84} C_{kr}\, v_D\, \rho_D \text{ (L) (at STP)}$$

where: C_{kr} = weight percent of Kr trapped in the deposit
v_D = volume of the deposit
ρ_D = density of the deposit.

Using the results given in Table 1 for $Cu_{.67}$-$Y_{.25}$-$Kr_{.08}$ and the figures of 300 working days per year and operating three shifts per day with 70% equipment utilization, it can be shown that at least two banks of four KTSD modules each would be required to immobilize the estimated 210 m^3 at STP, or about 17 MCi of 6% krypton-85 released during the processing of 2000 metric tonnes of irradiated fuel per year.(12) (Figure 4 depicts a bank of four KTSD modules.) In contrast, only one 1000-cm^2 substrate KTSD would be required to immobilize the estimated 1.8 m^3 of Kr released during the processing of 15 metric tonnes of irradiated FFTF fuel per year (60,000 MWd/t cooled 180 days). The actual number of KTSDs cannot be determined for either size plant until a reliability factor is obtained by filling a meaningful number of units to full capacity, including the cap-welding and leak-testing steps.

IV. Description of Unshielded, Multiple-Module KTSD Station

It was considered very desirable to eliminate the need for continuous recirculation of untrapped krypton in the ion-implantation/sputtering process. This was primarily to avoid problems in maintenance of the krypton-85-contaminated pumps resulting from radiolytic decomposition of pump fluids and seals. Early prototype units with auxiliary pumps required recirculating up to 10% of the krypton feed stream. Three methods were considered for eliminating this necessity. The simplest was to find a way to operate the KTSD over a wider pressure range. The second method considered was the addition of a large ballast reservoir to the pump. And the third was to demonstrate that one or more KTSDs could be made to operate in parallel connected to a common manifold in close proximity without disruptive electrical interference.

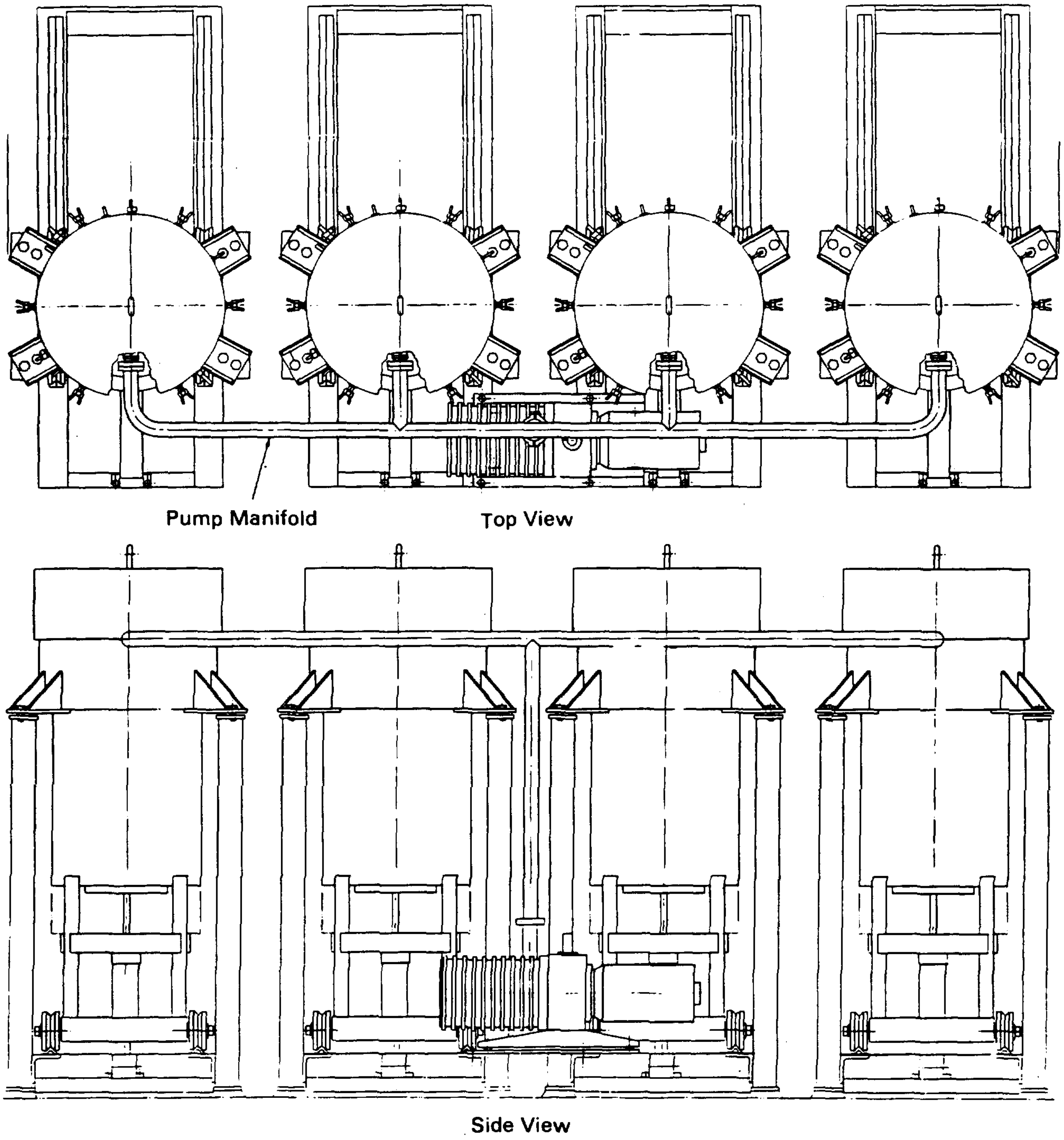


FIGURE 4. Bank of Four Remotely Operable KTSDs for a Commercial-Scale Krypton-Immobilization Plant

It was discovered during preparation for the bench-scale radioactive experiments, discussed in Section VI, that the KTSDs could be made to operate over a much wider pressure range than previously thought.(13) Instead of 0.3 to 1 Pa they could, at least with the 345-cm^2 substrate unit, be made to operate over the range of 1 to 10 Pa. However, there was a penalty. When the pressure exceeded 1 Pa, the pumping speed decreased by more than a factor of 10. The frequency and magnitude of pressure bursts (caused usually by internal electrical arcs) which would raise the KTSD pressure above its most efficient trapping range would reduce the overall trapping speed. The addition of a ballast reservoir KTSD would handle most of the gas bursts observed in operating KTSDs with substrate areas up to 1500 cm^2. The ballast volume would require more space and shielding. For this reason the third method, use of multiple KTSDs in parallel on a common manifold, was chosen as the most practical for eliminating recirculation of krypton.

The main purposes for the multiple-module operation experiment were to:

- demonstrate the stable operation of a 1000-1500 cm^2 substrate area KTSD without the need of a continuously operating auxiliary pump
- demonstrate stable side-by-side KTSD operation
- evaluate the design of the 1000-cm^2 substrate area disposable KTSD using the copper-yttrium matrix alloy.

Three KTSDs were connected to a common manifold but insulated electrically by a short section of alumina tubing as shown in Figure 5. Each unit was equipped with its own fast-acting servo control valve for admitting krypton. This valve was located as close as possible to the KTSD and also insulated electrically by a short length of nonconductive tubing. The krypton feed rate was measured by individual mass flow meters. The power supplies and current interrupters are not shown, but a typical schematic wiring diagram is shown in Figure 6. The composite sputtering targets for each unit were fabricated from forged OFHC copper bar stock fitted with cast yttrium metal plugs pressed tightly into an evenly spaced array of holes.

Figure 7 is a cutaway view of Unit 1, a demountable version of the 1000-cm^2 substrate disposable KTSD (seen in Figure 2). A demountable version was chosen to simplify post-run examination as well as making any electrode modifications. The Cu-Y surface area ratio for the target was 1.0. Unit 2 was a 1500-cm^2 substrate pump with a target Cu-Y surface area ratio of 1.0. Unit 3 was a 1000-cm^2 substrate early-model pump with a Cu-Y surface ratio of 5. The internal electrode geometries of Units 2 and 3 were very similar to that of Unit 1. All three KTSDs used a one-turn, 50-mil (1.3 mm) diameter tungsten filament for the electron source. Units 1 and 3 were heated with 3-phase and 6-phase power, respectively, and Unit 2 was heated with dc.

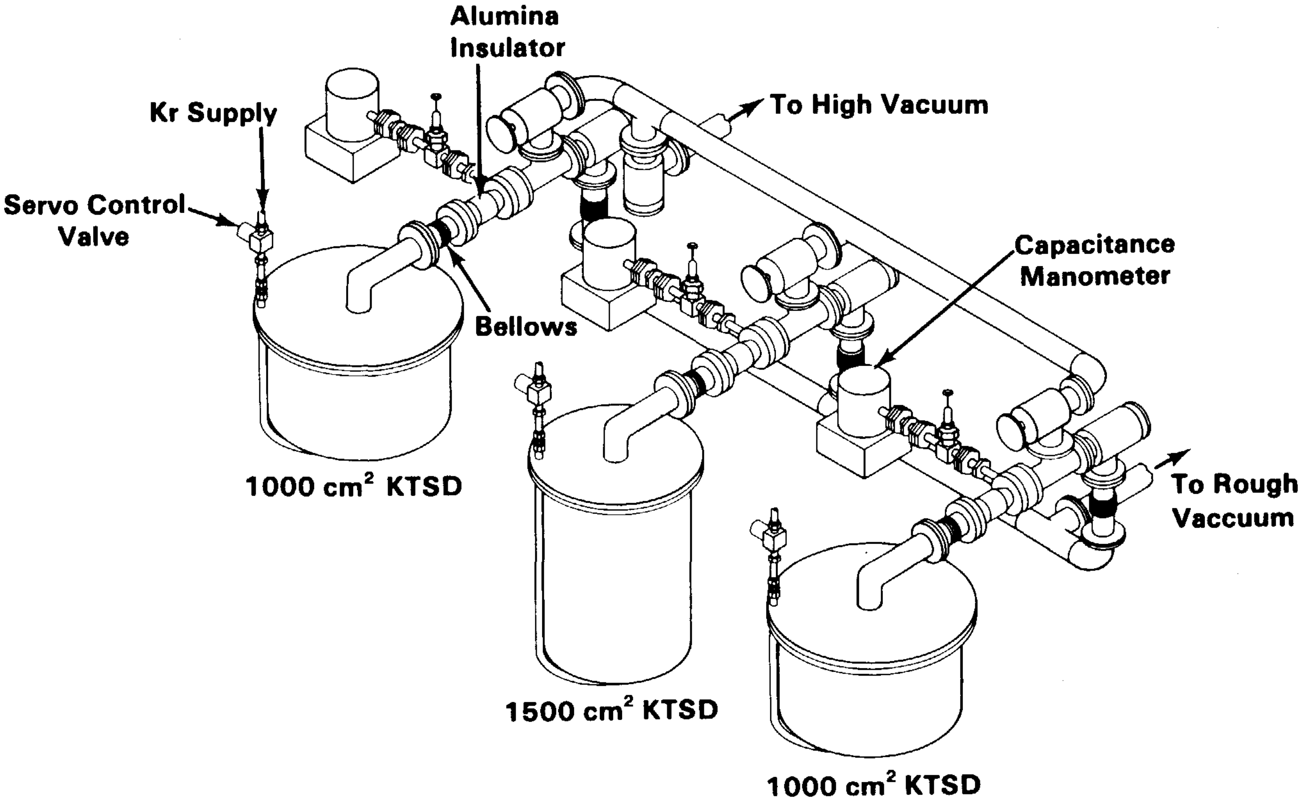


FIGURE 5. Vacuum Piping Arrangement for Multiple-Module Krypton-Trapping Tests

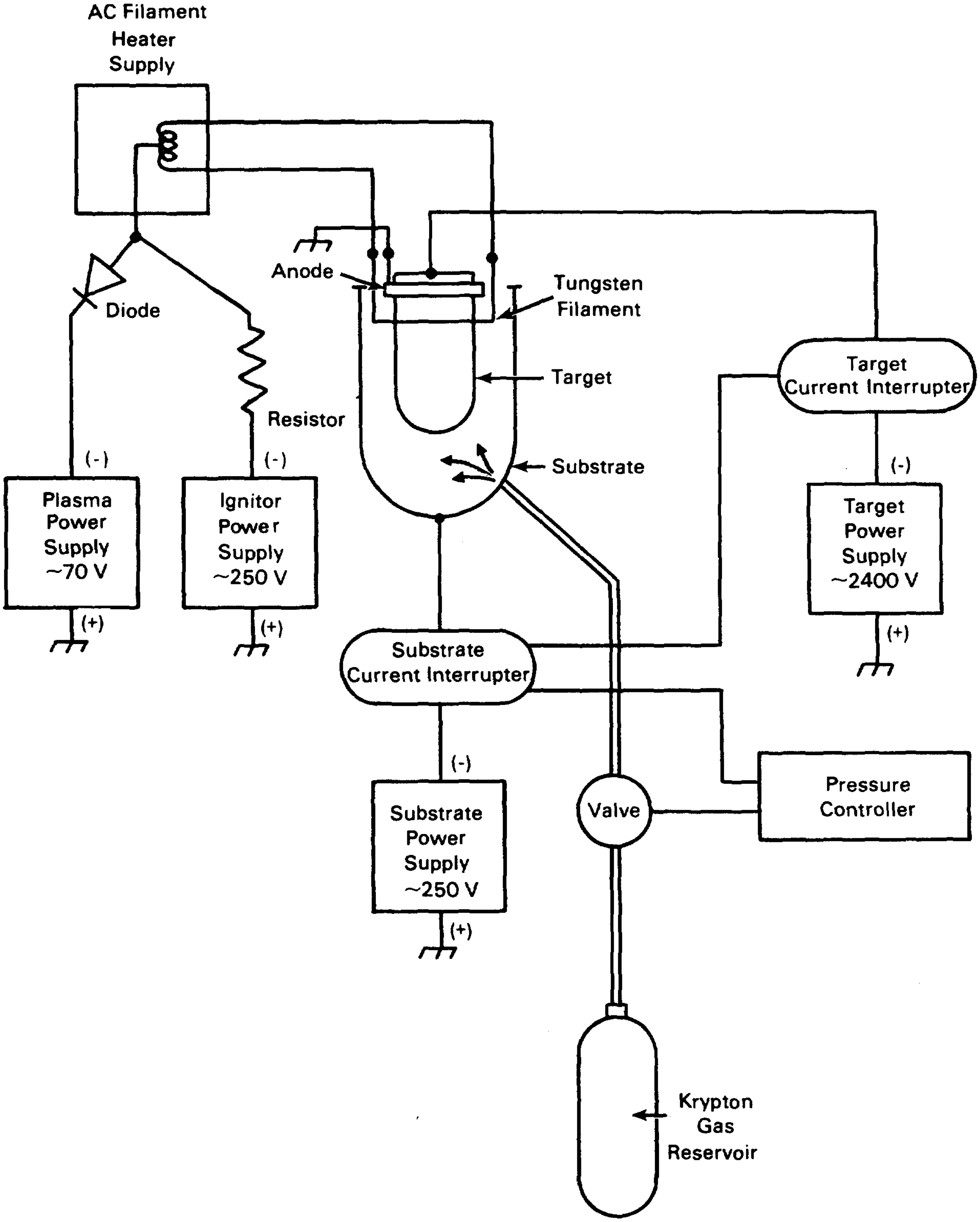


FIGURE 6. Schematic Wiring Diagram for KTSD Operation

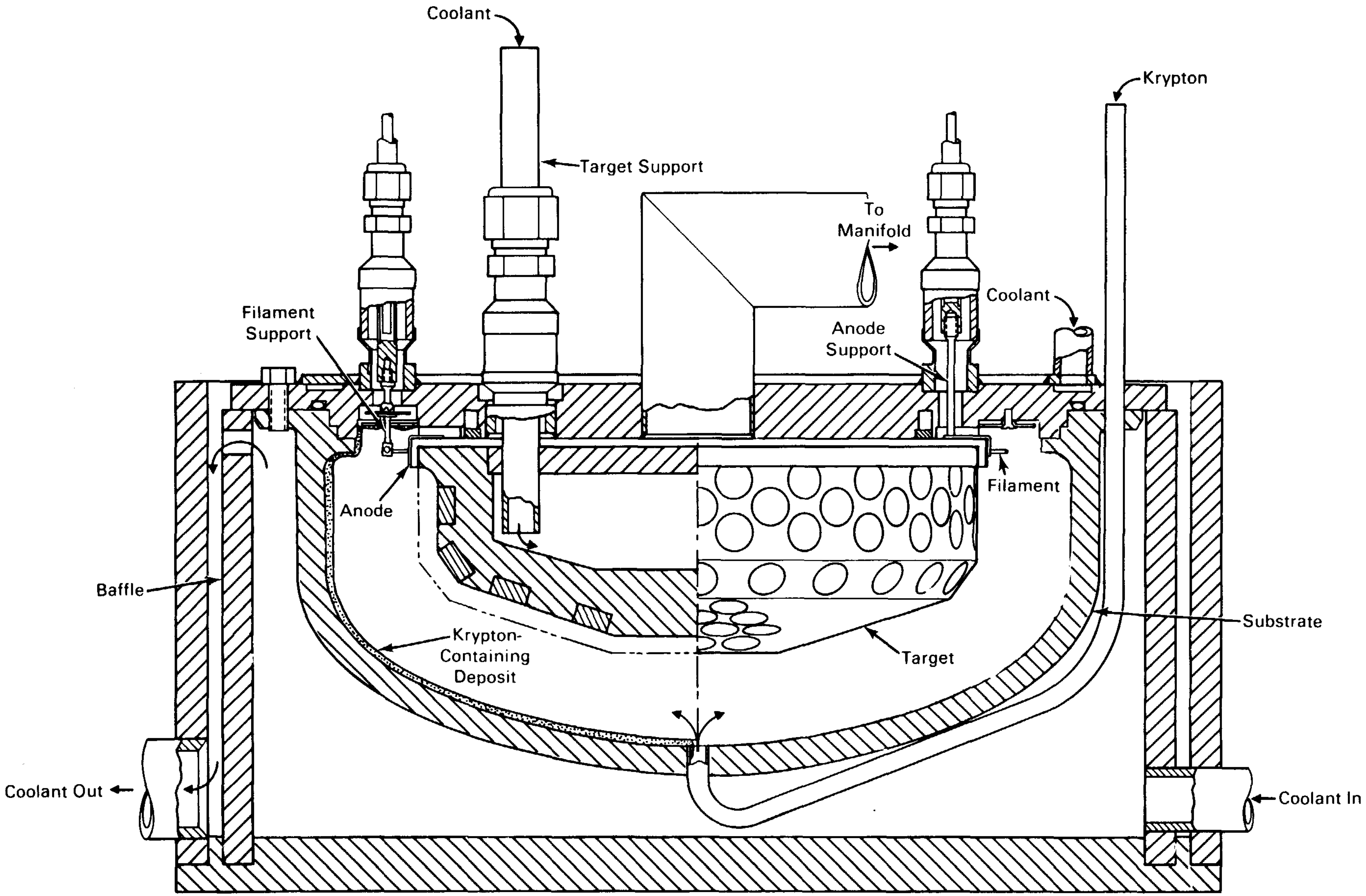


FIGURE 7. Unshielded, Demountable 1000-cm^2 Substrate KTSD

Lead shielding required for radioactive operation was not justified since only natural krypton was used. By eliminating the lead, some economy in fabrication was realized, but more important, the chance for electrical interference was increased. This produced a more rigorous test since the units were placed closer together and the lead was not there to act as an electrical ground shield.

V. Results and Discussion of the Multiple-Module Experiment

Before the multiple-module experiment was attempted, each unit was assembled and tested individually, first using an auxiliary pump and then closing the valve between the KTSD and the manifold. Typical operating parameters and pumping speeds are given in Table 2. All three units operated satisfactorily at their rated power when isolated from the auxiliary pump. Units 2 and 3 were restarted in sequence and run concurrently for a period of 3.5 h with no external pumping. While the units worked independently of one another, it was noted that pressure fluctuations during arcing were reduced.

Three conditions were found necessary to promote stable operation during arcing:

1. Both the target and substrate current interrupters must have "off" periods of less than ~10 ms.
2. Following an arc, the substrate current interrupter must signal the krypton feed control valve to close completely as rapidly as possible, and the krypton control valve must return to its original position just prior to the arc when the arc is extinguished.
3. In the case of an arc causing a loss of pressure control, normal operation is regained more rapidly if the substrate supply voltage is lowered to ~100 V, then raised slowly to the set point until the excess krypton has been trapped and the pressure is under control again.

The first condition was met by making a one-time adjustment in the timing circuits of the power supply current interrupters for Units 2 and 3. The power supply current interrupter timing circuits for Unit 1 were of a type that could not be adjusted to the required off period, so no attempt was made to operate this unit in the simultaneous mode. The second condition was met by modifying the circuits of a commercial pressure control unit. The third condition was met in this test by manually adjusting the substrate voltage, but control could easily be automated. The absence of disruptive electrical interference was fortuitous.

Three likely sources for electrical interference are the power supply current interrupters during cycling, poor grounding, and the nature of the triode discharge itself. There are three fast-acting current interrupters placed in series or built into the target, substrate and plasma power supplies

Table 2. Operating Parameters Multiple-Module Experiment.

				Target		Substrate		Plasma		Ignitor		Filament		Total	Trapping	Power Con-
	Substrate	Target	Area Cu	V_t	I_t	V_s	I_s	V_p	I_p	V_i	I_i	V_f	I_f	Power,	Rate,	sumption,
Unit	Area, cm^2	Area, cm^2	Area Y	V*	A*	V	A	V	A	V	A	V	A	kW	L/h	kWh/L
1	1000	547	1	2400	5.5	250	10	65	6	190	2	3	230	17	1.0	17
2	1500	981	1	2400	9	220	13.9	65	9.4	190	2	2.8	250	26	1.5	17
3	1000	547	5	2400	6.3	270	11	65	12.7	190	2	7.4	93	20	0.7	29

---

* V = volts; A = amps

for operating each KTSD. These interrupters serve to prevent damage to the individual power supplies and to the internal electrode structure of the KTSDs. The substrate power supply interrupter plays a special role in minimizing re-emission of krypton from the growing deposit caused by arcs or by KTSD control logic. These switches are capable of turning off up to tens of kilowatts of power in microseconds and hence could easily become powerful oscillators. Good grounding and low-inductance connections between the interrupters and the KTSDs, no doubt, were important in eliminating electrical noise. Under certain conditions, which are not well understood, noise in the kilohertz range, probably generated in the triode discharge, was observed superimposed on the dc supply voltages. This noise can cause the current interrupters to cycle unnecessarily, reducing momentarily the pumping capacity of the KTSD. This noise can be filtered by adding capacitance to the output circuits, but it is also important to minimize this capacitance to minimize the energy available for arcs.

VI. Radioactive Bench-Scale Experiments

The 450 Ci of krypton-85 available was divided equally for loading three Cu-Y samples for long-term stability tests at 150, 250 and 350°C. Since the specific activity of the krypton-85 feed (4.2%) was representative of a commercial reprocessing plant stream, the experiments themselves were expected to reveal operational problems attributable to the high beta activity. Prior to the radioactive runs, a nonradioactive krypton implantation run was carried out to simulate conditions and to identify procedural problems and the need for additional radiation shielding. Later the loaded KTSD with nonradioactive krypton was used for accelerated thermal stability tests to identify the approximate temperature at which krypton would be released at a rate >1% in 10 years.

A 345-cm^2 substrate KTSD, with a composite target having a Cu-Y surface area ratio of 1.4, was surrounded with a minimum of 2.5 cm of lead shot, as seen in Figure 8. A schematic piping diagram, including the krypton reservoirs, pumps, KTSD, valves and miscellaneous sensors, is shown in Figure 9. This apparatus and its operation are fully described in Reference 13.

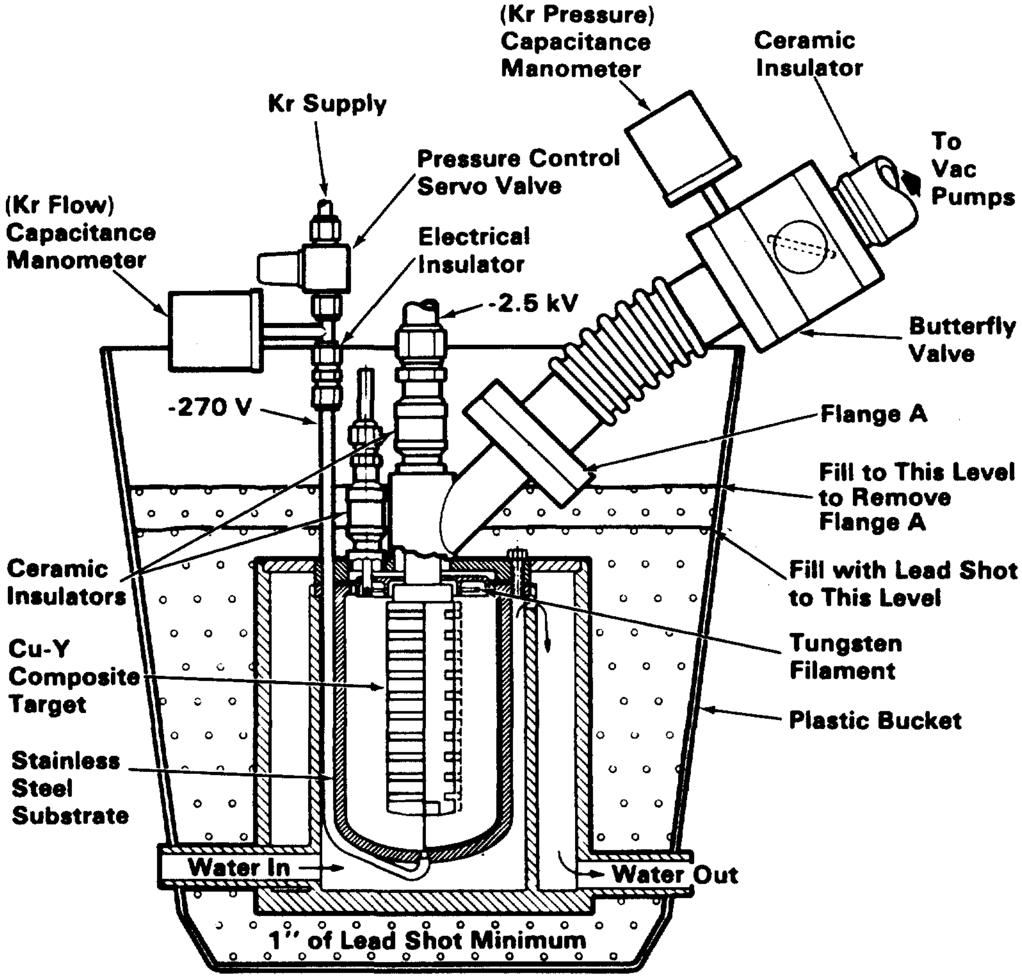


FIGURE 8. Multicurie Bench-Scale KTSD

One important feature of this KTSD apparatus was the exclusion of elastomer seals due to its intended use as a high-temperature storage vessel for up to 10 years. Viton and Teflon seals, however, were used extensively in other parts of the apparatus.

Briefly, the procedure for filling the KTSD was to evacuate and backfill with natural krypton. The substrate was then ion-etched before applying full power to the substrate and target. After ~1 h of operation the natural krypton supply was closed and krypton-85 of 98% purity was admitted. The major impurity was air, and the minor impurities were ~0.06% each of He, A, CO_2 and H_2. When all the krypton-85 was used up, the radioactive reservoir was closed and a closeout layer sputtering with natural krypton was added. The three layers were ~0.1, 0.5, and 0.04-0.1 mm thick in the order that they were deposited. A practice run was made with natural krypton, and a dummy reservoir

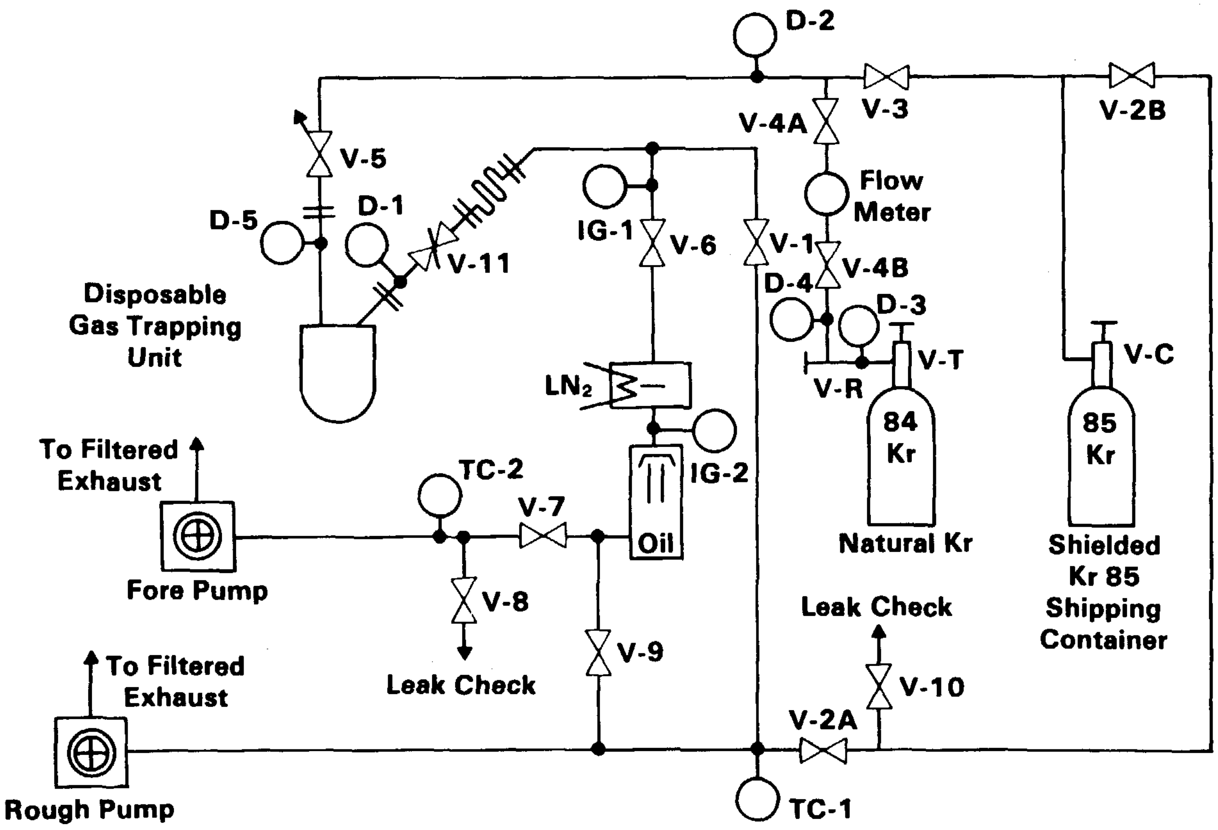


FIGURE 9. Schematic Piping Diagram for 150-Ci ^{85}Kr Bench-Scale Experiments

also was filled with natural krypton at ~0.5 atmosphere to simulate the procedure for the radioactive runs. The product from this run was saved for accelerated testing of thermal stability. Two radioactive runs were completed, immobilizing 150 Ci each. A target-to-anode short a little over halfway through the third run prevented trapping more than 82 curies in that deposit as well as adding the closeout layer.

The results from these runs showed no observable difference when radioactive instead of natural krypton was used. This is not surprising since the number of 40 eV electrons emitted by the filament is a factor 10^{12} times greater than the 0.7 MeV beta particles present in the space between the target and substrate apparatus at any one time. The small number of rubidium atoms produced in the space between the target and substrate would not be expected to cause any unusual effect either. Equally important, no radioactive particulate material was detected during disassembly of the KTSDs, even the third one without the benefit of the nonradioactive closeout layer. Each KTSD operated the entire radioactive portion of the run without the aid of auxiliary pumping, and thus allowed 100% recovery of the krypton. The trapping rate of the 345-cm^2 substrate KTSD could be varied from 0 to greater than 0.5 L/hr. The whole-body exposure received by the operator was low, ~25 mr/run.

VII. Long-Term Elevated-Temperature Storage Tests on Cu-Y-^{85}Kr

Long-term stability of the sputter-deposited krypton-metal matrix is important in evaluating the ion implantation/sputter-deposition process for immobilizing krypton-85 gas. Laboratory experiments using nonradioactive krypton trapped in a Ni-La sputter-deposited matrix have shown that krypton gas is released from the metal matrix at low rates by a diffusion process.(14) Gas release rates and the stability of the metal matrix may, however, be affected by the radiation field and transmutation products in an actual storage container of krypton-85. Consequently, experiments were started to measure the release of krypton-85 gas from multicurie-loaded Cu-Y-Kr deposits and to ascertain that the alloy is stable at elevated temperatures that bracket the anticipated long-term storage conditions.

Three multicurie Cu-Y-Kr deposits that are contained in the KTSDs are being used to determine the stability of the metal matrix during 5 to 10 years' storage at elevated temperatures. Temperatures of 150, 250, and 350°C were selected to bracket anticipated storage conditions. The three units are currently being stored at 150°C in lead-shielded casks, and the amount of krypton released from each unit will be measured after 3 months' storage. One unit will be returned to 150° while the temperature of the others will be increased in 100°C increments to reach their final storage temperatures, 250 and 350°C, respectively. Gas release will be measured at 3-month intervals during the initial heat-up periods and at 6-month intervals thereafter. One nonradioactive Cu-Y-Kr deposit prepared identically to the multicurie deposits will be subjected to accelerated thermal testing to provide an early warning of any major release of krypton unrelated to radiation damage or growth of the rubidium daughter into the Cu-Y-Kr product.

A diagram of the storage cask configuration is shown in Figure 10. The cylindrical cask was fabricated from stainless steel sheet in a double-walled configuration. Lead shot was used to fill the 3.8-cm-thick wall cavity and, combined with a solid lead lid (3.8 cm thick), provides the necessary radiation shielding. Radiation levels were reduced from about 50 R/hr at contact with the KTSD to less than 25 mR/hr at the cask surface.

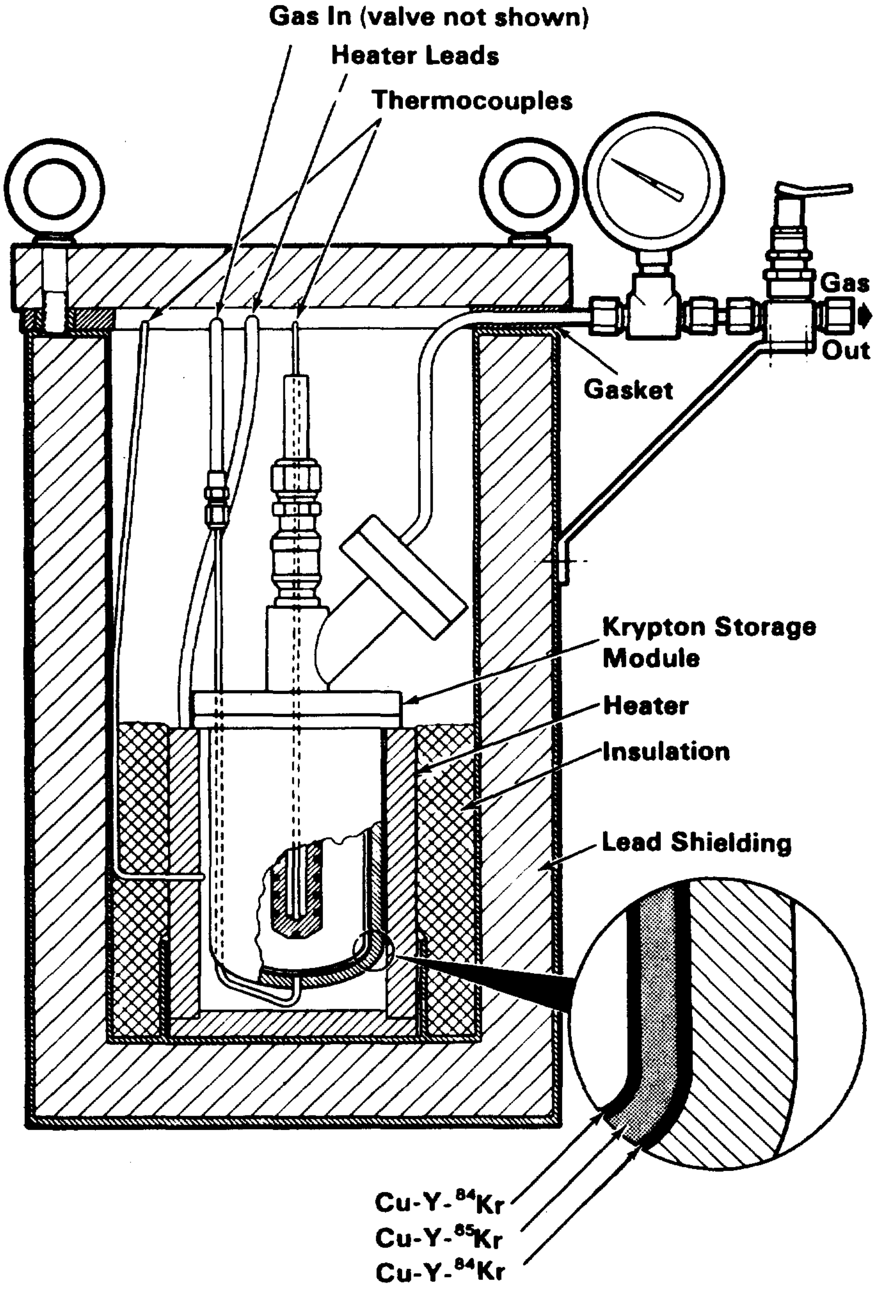


FIGURE 10. Bench-Scale KTSD Storage Cask and Heater

The KTSD is positioned within a clamshell heating element centered near the bottom of the cylindrical cask. Inlet and exhaust lines with valves outside the cask are provided for measuring release of krypton gas. The internal pressure of the unit is measured with a pressure gauge attached to the exhaust line. This gauge is monitored weekly and would provide early warning of an unexpectedly large krypton release from the metal matrix or of a leak in the containment. Thus, a significant increase or decrease in pressure would indicate a problem with the unit and the experiment would be terminated.

Temperature of the units is measured with chromel-alumel thermocouples and controlled with variable transformers. The temperature-power characteristics of each storage cask were experimentally determined by spot-welding thermocouples directly to the side of a nonradioactive KTSD and measuring the wall and furnace temperatures as a function of power level. The data from these experiments were used to establish the proper furnace temperature and power level to obtain the desired KTSD wall temperature for long-term storage.

VIII. Summary and Conclusions

A preconceptual design was completed for a dual-scale pilot ion-implantation station for immobilizing krypton-85 in a growing sputter-deposited matrix. Plant capacity is determined by the number of units, size and matrix alloy selected for the individual krypton-trapping storage device (KTSD). The calculated capacity for a 1000- and 3000-cm^2 substrate KTSD loaded with a Cu-Y-Kr alloy 1 cm thick would contain the equivalent of 100 and 400 L of krypton at STP, or about 8,000 and 32,000 Ci of 6% krypton-85, respectively. The calculated trapping rate for the 1000-cm^2 KTSD operated at 17 kW would be 1.0 L/h. For the 3000-cm^2 KTSD the trapping rate would be 3.9 L/h operated at 58 kW total input power. An experimental reliability factor needs to be obtained before both the exact number of units for a given plant capacity and reliable cost figures can be determined.

After evacuation to <5 Pa, KTSDs of up to 1500 cm^2 substrate area will operate satisfactorily without the need of an auxiliary pump. Multiple KTSDs can be operated close to each other. Operating KTSDs on a common manifold improves the overall trapping efficiency of the combined units.

Tests using up to 150 Ci of 4.2% krypton-85 behaved similarly to identical nonradioactive krypton using a target with a Cu-Y surface ratio of 5. It was possible to disconnect the KTSD semiremotely once it was filled with 150 Ci of krypton-85, with minimal radiation exposure. No detectable radioactive particulate was found in the manifold piping.

Long-term tests in progress will yield information about the stability of the Cu-Y-Kr alloy and the construction materials used in fabricating the KTSDs.

Acknowledgments

The authors gratefully acknowledge the U.S. Department of Energy, Office of Nuclear Energy, for financial support of this work under Contract DE-AC06-76RLO 1830. We also acknowledge J. W. Johnston for technical support, J. E. King for mechanical design, and J. A. Powell for editing the manuscript.

References

1. "Environmental radiation protection standards for nuclear power operations," Fed. Regist. 42 (9)(1977) Title 40, Part 190, January 13.

2. U.S. Energy Research and Development Administration, Alternatives for managing wastes from reactors and post-fission operations in the LWR fuel cycle, Vol. 2, ERDA-76-43 (April 1976), pp. 14.1 to 14.7.

3. Bendixsen, C. L. and Knecht, D. A., "Separation and storage of krypton," Proceedings of the international symposium on the management of wastes from the LWR fuel cycle, CONF-760701 (1976) p.343.

4. Tingey, G. L., McClanahan, E. D., Bayne, M. A. and Moss, R. W., "Entrapment of krypton in sputter deposited metals--a storage medium for radioactive gases," PNL-2879/, Pacific Northwest Laboratory, Richland, 1979.

5. Bayne, M. A., Moss, R. W. and McClanahan, E. D., "Krypton entrapment in continuous-biased sputter-deposited glassy metals," Thin Solid Films, 63:137-141 (1979).

6. Knoll, R. W., McClanahan, E. D., Tingey, G. L. and McDonald, E. L., "Development and selection of a matrix alloy for ^{85}Kr encapsulation," PNL-5926 Pacific Northwest Laboratory, Richland, Washington, 1986.

7. "Kr encapsulation preconceptual design," Report No. 6154-3-1, Prepared for U. S. Department of Energy by the Ralph M. Parsons Company, 1981.

8. Plücker, J., Poggendorff's Annalen (Annalen der Physik, Series 2) 103:88 (1858).

9. Blodgett, K. B., and Vanderslice, T. A., "Mechanism of inert gas cleanup in a gaseous discharge," J. Appl. Phys. 31:1017-1023 (1960).

10. Whitmell, D. S., Nelson, R. S., Williamson, R., Smith, M. J. S., and Bauer, J. S., "A half-scale pilot plant for the immobilization of krypton in a metallic matrix by combined ion implantation and sputtering: final report 1980," AERE-R 10204.

11. "Krypton-85 disposal program conceptual design phase: final report," Klett, R. D., editor. Prepared for U.S. Department of Energy by Sandia National Laboratories, SAND 81-1957, 1981.

12. U. S. Department of Energy. 1980. Management of commercially generated radioactive waste environmental impact statement, DOE/EIS-0046-F, Department of Energy, Washington, D.C.

13. McClanahan, E. D., Moss, R. W., and Greenwell, E. N., "Operation and control of an ion-implantation/sputtering storage device for ^{85}Kr," PNL-SA-13239, presented at the 32nd National Symposium and Topical Conference, American Vacuum Society, Houston, Texas, November 19-22, (1985).

14. McClanahan, E. D., et al. "Krypton ion implantation," In Nuclear Waste Management Semiannual Progress Report, April 1983 through September 1983., pp. 6.10-6.16. Pacific Northwest Laboratory, Richland, Washington, 1984.

APPENDIX

Estimated Sputtering Parameters and Dimensions for KTSDs

Substrate Area, cm^2	Target Area, cm^2	Deposit Vol. (1 cm thick), cm^3	V_t V*	I_t A*	V_s V	I_s A	V_p V	I_p V	V_i V	I_i A	V_f V	I_f A	Collection Efficiency (K), %
1000	547	765	2400	5.5	250	10	65	6	190	3	3	230	95
3000	2029	2685	2400	20	250	30	65	10	190	3	3.1	250	98

Properties of Matrix Alloys for KTSDs

Alloy	Net Sputtering Yield, Y, g/Ah	Estimated Density, ρ, g/cm	Krypton Concentration, C, wt. %
$Ni_{.81}La_{.09}Kr_{.10}$	7.9	6.8	12.7
$Cu_{.67}Y_{.25}Kr_{.08}$	7.6	6.2	9.4

---

* V = volts; A = amps

DISCUSSION

BASTIN: Will the planned long-term testing, if successful, qualify the packages for near-surface burial?

McCLANAHAN: Long-term testing of the three bench scale KTSDs will help qualify the copper-yttrium alloy as a suitable package, or waste form, for near-surface burial.

HUTTER: Can the presented Kr-85 sputtering system be connected to any Kr separation system directly without an intermediate buffer tank for Kr?

McCLANAHAN: Yes, the KTSDs described in this work can be connected to any separation system producing reasonably pure krypton with the help of a pressure reducing valve. You simply have to adjust the voltage on the power supplies to go from zero to full pumping capacity. I did not list that as a main advantage, but I don't see the need for a very large krypton reservoir if the head end of the process is large enough in the first place.

RINGEL: I have a question concerning the material loaded with Krypton-85. Isn't there a chance that this material may eventually become brittle with time and you will end up with a radioactive dust or aerosol problem?

McCLANAHAN: That's an excellent point. Of course, that is the purpose of long-term testing. The krypton-loaded rare earth alloys are all brittle but, as-deposited, form a monolithic solid. After each radioactive bench-scale experiment we checked the connecting piping for contamination with negative results. Whitmell, in the U.K., improved the mechanical and thermal stability of the krypton-containing metals he tested by raising the substrate temperature.

CROFF: Could you tell us what beneficial uses are being considered for krypton-85?

McCLANAHAN: Krypton-85 has been considered as a substitute for tritium for lights used in remote locations. Krypton-85 has also been considered as a low temperature heat source, e.g., oil pipelines.

CONDITIONING OF RADIOKRYPTON USING ION SPUTTER PUMPS

J. Römer, E. Henrich, T. Fritsch
Kernforschungszentrum Karlsruhe
Fed. Rep. Germany

Abstract

Krypton ion implantation into sputtered metals seems to offer a safe conditioning and storage of fission product krypton separated in a spent fuel reprocessing plant. Various crystalline and amorphous metal matrices have been investigated in 1.0 kW laboratory scale ion sputter pumps. An amorphous Cu70/Zr30 alloy is favoured, due to its high efficiency and material properties.

Radial scale up of the laboratory pumps is simple. Longitudinal scale up was investigated in a dummy device by visible observation of the gas discharge. A relatively homogeneous discharge has been obtained for inner to outer cathode voltage ratios greater than four. One anode, which is front side earthed, is sufficient for such operating conditions.

The design of 10-15 kW pumps has been made in view of technical application. The total length is about 1.0 m with a sputter surface of 0.3 m^2.

Optimal operating conditions are sputter voltages of 2 to 3 kV and implantation voltages of 0.3 kV for Fe/Zr and 0.4 kV for Cu/Zr. Pumping rates between 2.0 to 4.0 L (STP) per square meter of sputter electrode surface have been reached. Effectivity has been as low as 13.0 kWh per 1 L (STP) of Kr. These results are in good agreement with previous laboratory data. Aspects for technical applications in a reprocessing plant are discussed.

I. Introduction

During the last 10 years an integrated system for treatment of the dissolver offgas was developed in the Institut fuer Heisse Chemie. This system, which exists in a semi-technical size, consists of three plate columns, which perform the nitric acid recovery and iodine desorption. In connection, a pressureless cold washing (Fig. 1) with freon 12 removes and separates the noble gases xenon and krypton.(1) Finally, the radioactive krypton is continously stored by ion sputter pumps preventing a high unconditioned inventory in the separation equipment and resulting in increased safety against leakage, as the system works without high pressure. This paper provides more details on the development of these pumps.(2,3,4,5)

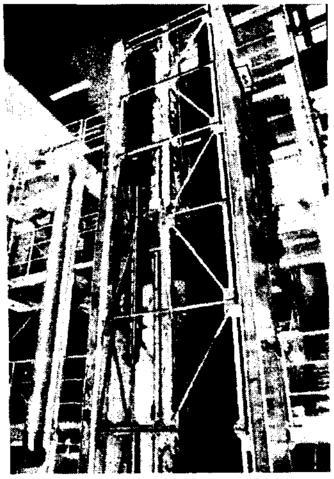
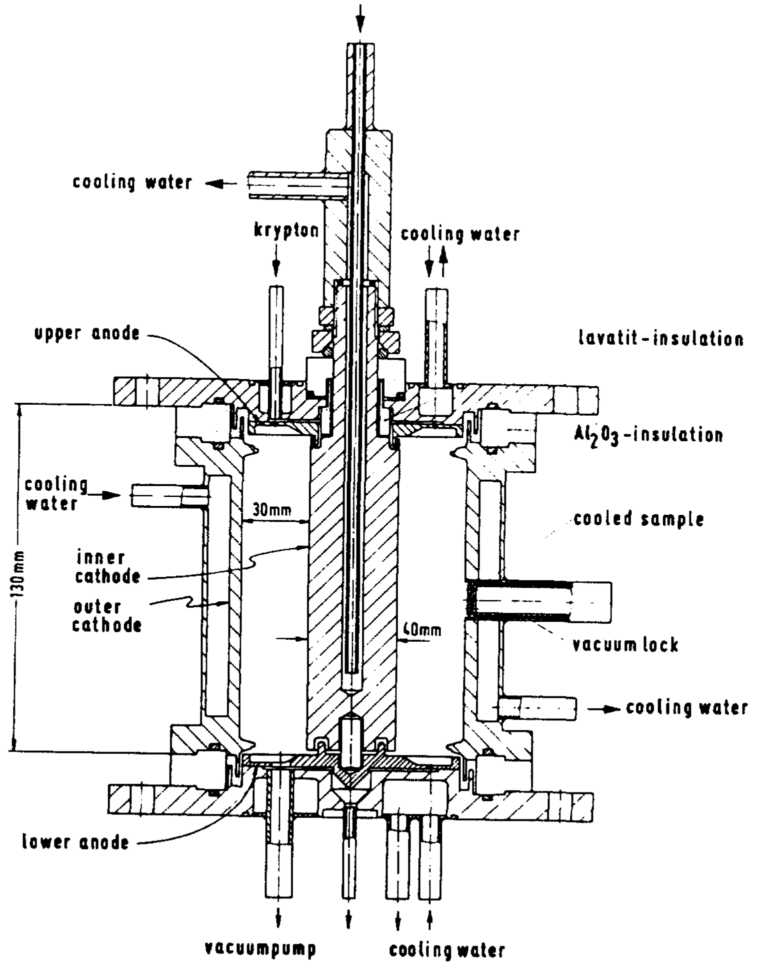


Fig.1 Noble gas scrubber

II. Experimental

1. The Experimental Pumps

In a first step some experimental pumps were constructed for fundamental testing.(6) Figure 2 gives the details of such a pump, which have an electric consumption of 1.0 kW. The inner cathode had a diameter of 40 mm and was fabricated of the testing sputter material. A voltage of -2500 V is applied to the this cathode to obtain a self-sustained krypton discharge in the annular gap, which has a distance of 30 mm, by a pressure of about one Pascal for sputtering the cathode material. The krypton is implanted by a voltage of -300 V to -1000 V into the continuously growing metal coating, which is collected on the inside of the outer cylinder. The upper and lower anodes are grounded. They are insulated against the cathodes by a gap of one millimeter preventing a discharge due to the Paschen Back law.



IHCH

Fig. 2 Design of the laboratory pump

2. Crystalline Material

First we used crystalline materials such as copper or titanium. These metals result in stable substrates with good thermal and mechanical properties. In Fig. 3 the left part gives a functional diagram of the pumping rate against the implantation and sputter voltage and the right part shows the power consumption. Here we find a minimum at 65 kWh/L(STP) Kr with an implantation voltage of -900 V and a sputter potential of -2.5 kV.

In Fig. 4 electron microscopic photos are shown of copper and titanium substrates containing up to 200 L(STP) Kr per liter metal in the upper row and the same material after a thermal treatment up to 1000 degrees C. Especially after heating the bubbles of krypton are more obvious.

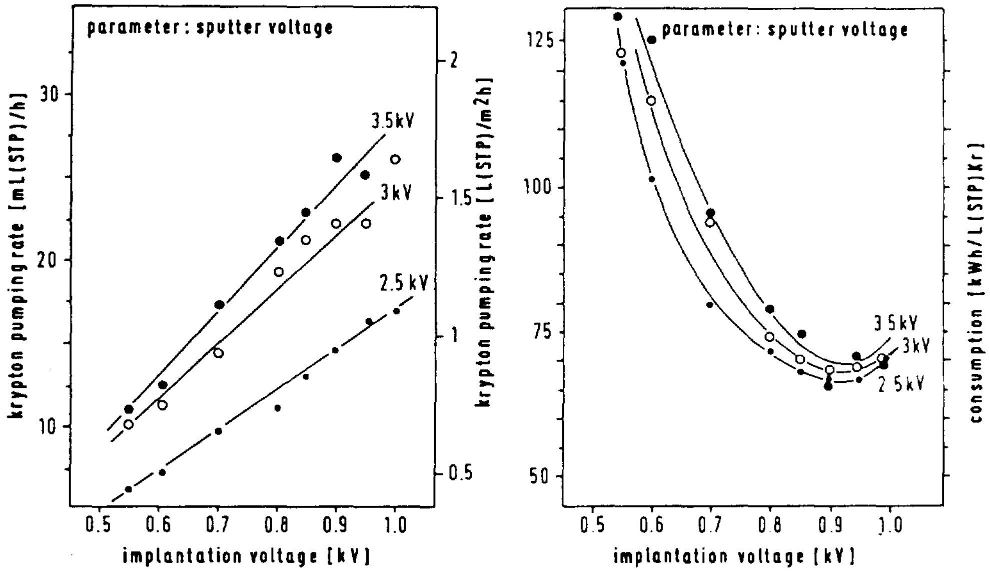


Fig.3 Data of the Ti laboratory pump

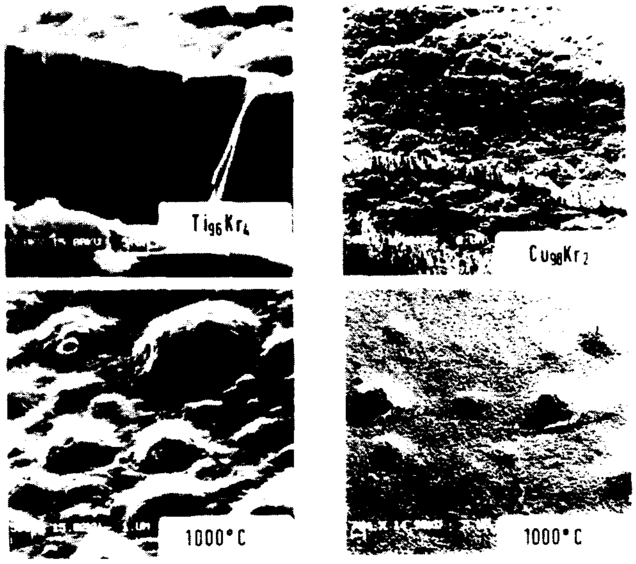


Fig. 4 Electron microscopic photos of crystalline Kr-metals

3. Amorphous Material

By sputtering it is possible to produce amorphous alloys using two or more components with different atom diameter. Fig. 5 gives an example of a suitable element choice.

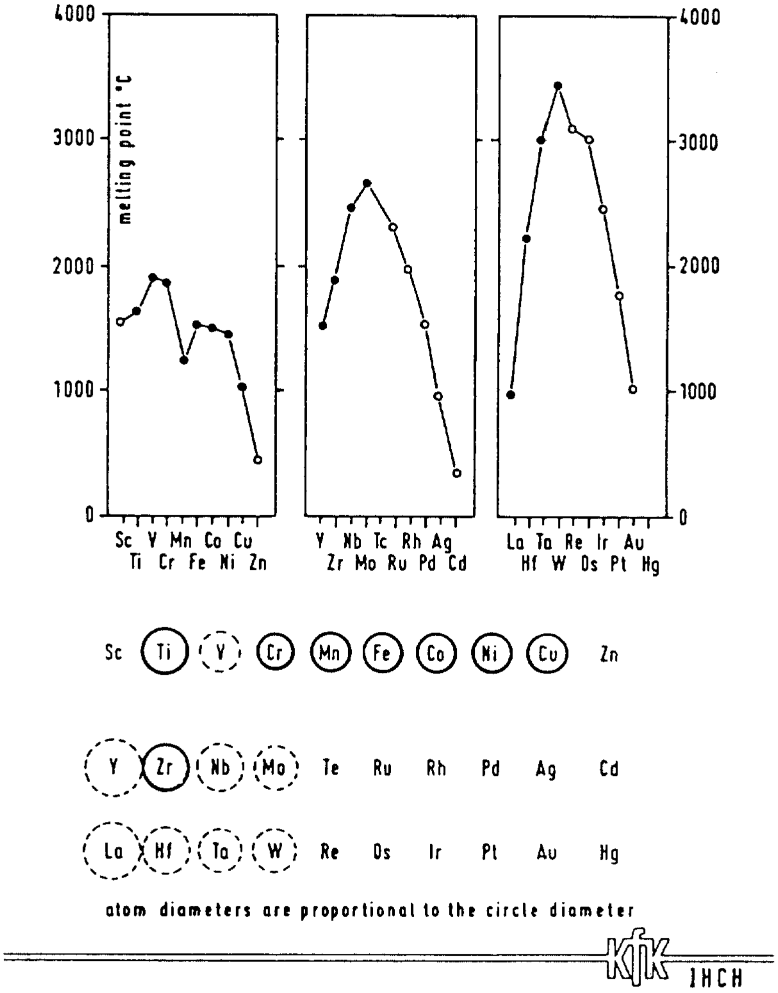


Fig.5 Melting points of suitable metals

We selected Fe and Cu with 30% of Zr. Fig. 6 gives two examples for constructing such cathodes. One is fabricated by setting plugs of Zr into holes and the other by inserting strips of Zr into slots of the cathode. The amorphous deposits shown in the operation diagram (Fig. 7), emphasise the lower power consumption of 15 kWh/L(STP) Kr at the optimal implantation voltage of -350 V and a sputter potential of -2.5 kV.

The highest pumping rate was 4.0 l(STP) Kr/hm^2. Fig. 8 presents electron microscopic photos of such deposits before and after the annealing. Here the glassy edges show mussel-shaped fractures and demonstrate the amorphous structure which remains distinct up to 400 degrees C thermal annealing temperature. Above this definite temperature, which is a function of substrate composition, the deposit becomes porous in structure and the release rate of Kr increases. Neutron scattering and Mößbauer measurements suggested that the Kr atoms are distributed on single sites in the amorphous lattice.(7)

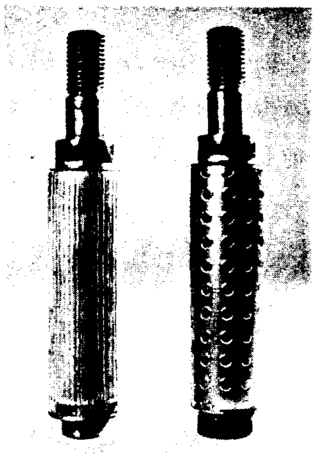
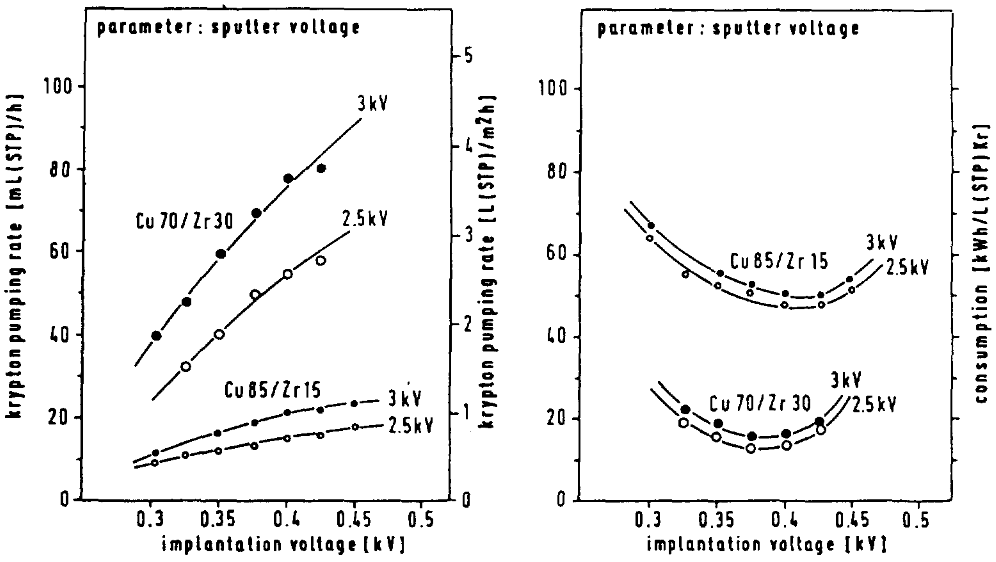


Fig. 6 Cathodes for sputtering amorphous Zr-alloys



IHCH

Fig.7 Data of the Cu 85/Zr 15 and Cu 70/Zr30 laboratory pump

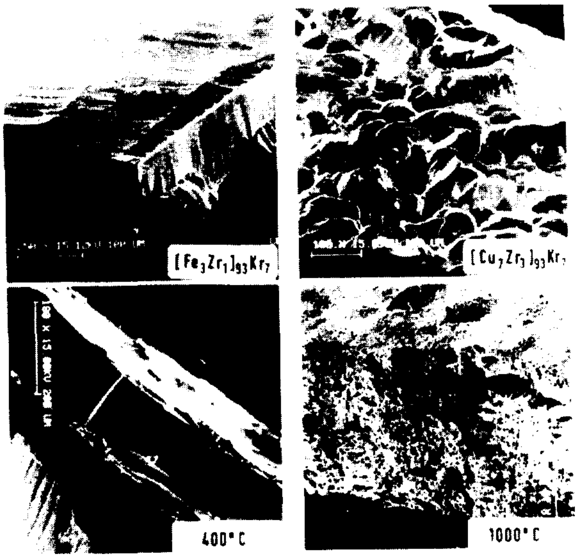


Fig. 8 Electron microscopic photos of amorphous Kr-Zr-alloys

4. Summary of the Experiments

Fig. 9 gives an overview of the pumping rates and power consumption of different material combinations as function of the implantation and sputter voltage. The lowest consumption was reached with the Cu70/Zr30-alloy.

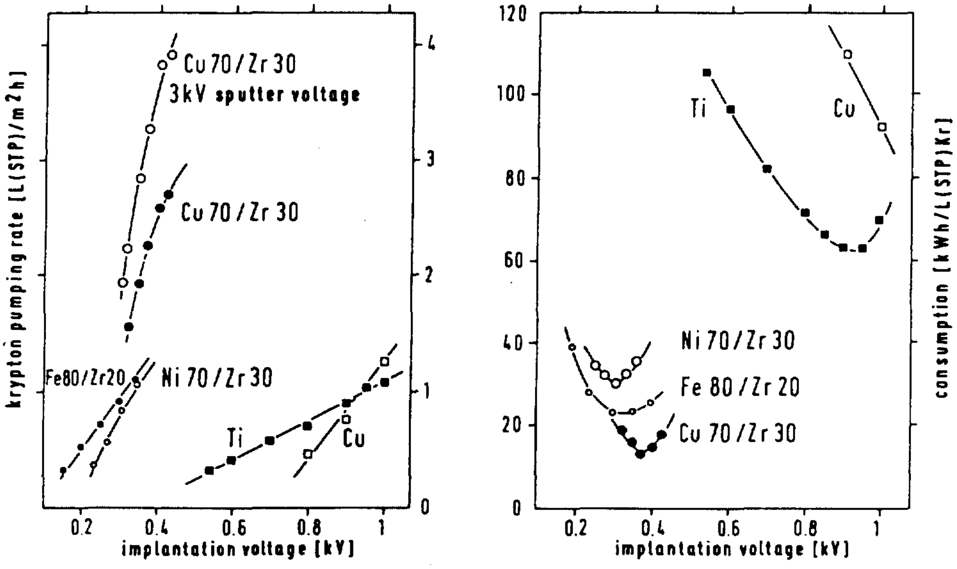


Fig.9 Pumping rates and power consumption of different materials at 2.5 kV sputtering voltage

The comparision of the quasi steady Kr release rates given in % per year of crystalline and amorphous deposits in Fig. 10 shows; that these rates are in relation to the melting point of the used material. For the Cu-, Ti- and Cu70/Zr30-curves thin samples getting with the sample catcher were used. The Fe75/Zr25- and two Cu70/Zr30-measurements were performed with thick samples coming out of a spent pump. We suggested surface effects as reason for the big difference between the different Cu70/Zr30-results. These thermal release measurements show a very low release up to 400 degrees C. The total Kr amount released below this temperature up to steady state is less than 0.1 %.

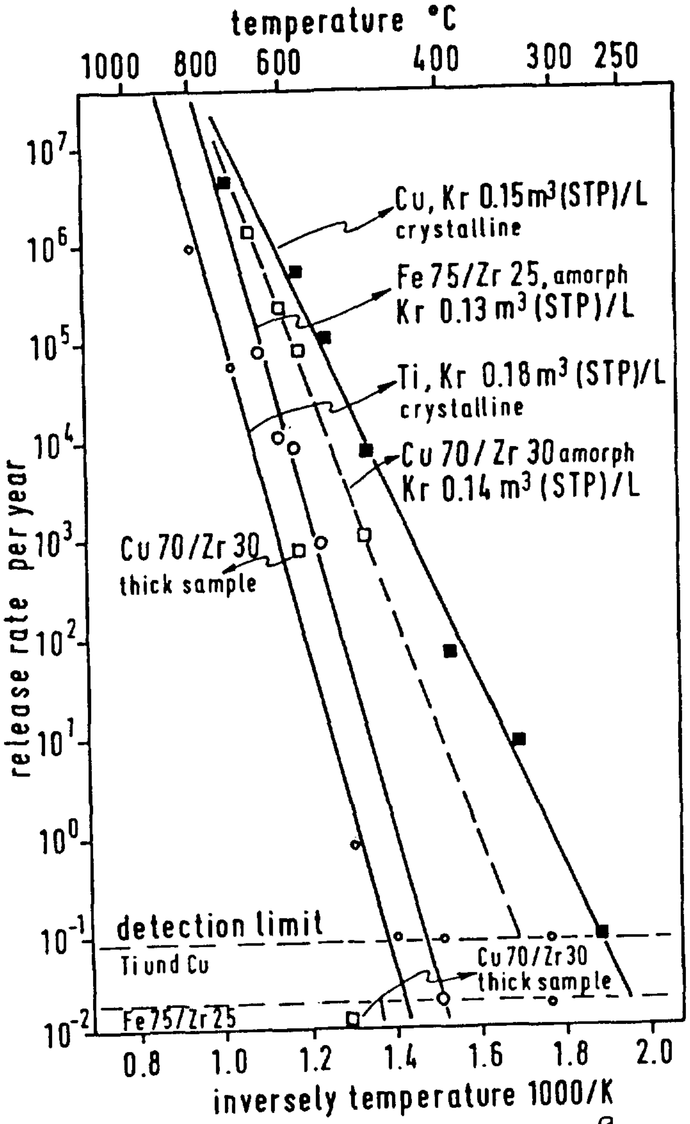


Fig.10 Quasi steady Kr-release rates of different products

The following Fig. 11 shows a consumed Ti- and Cu70/Zr30-cathode. The different plasma density of the Ti-cathode during sputtering operation is in remarkable contrast to the uniform consumption of Cu/Zr-cathode due to its lower implantation voltage. Fig. 12 shows a sectional view of the Ti-pump after 700 h operation.

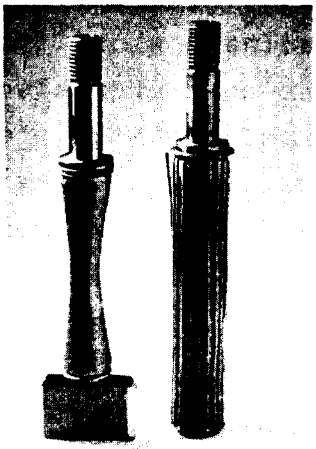


Fig. 11 Comparison of a spent Ti- and Cu/Zr- cathode

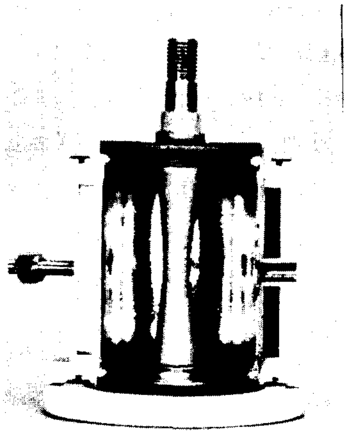


Fig. 12 A sectional view of a spent Ti-lab. pump

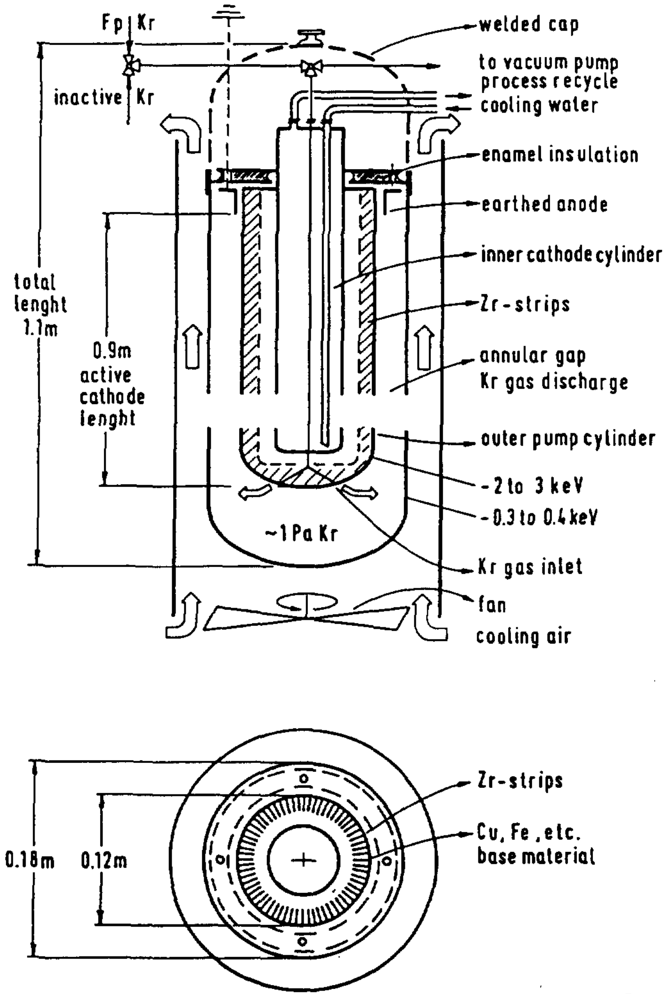
III. Development of a Technical Pump

1. Fundamental plasma research

The scale up of the pump diameter seems to be without problems, but increasing the length is an unsymmetrical scale up. For plasma research, mainly by visible inspection, a glass tube with an identical electrode arrangement ebables rapid checking of the uniformity of the discharge under different potential conditions. We found only a uniform discharge with sputter to implantation potential ratios of greater than four. In this case no changes of the uniformity are detectable with only one grounded anode. Therefore, a lengthened pump with one anode works using amorphous material successfully only due to its low implantation voltage.

2. Construction of the Semi-technical Pump

A design of 10-15 kW pumps has been made in view of technical application. The total length is about 1.0 m with the outer diameter 0.18 m. This amounts to a sputter surface of $0.3\ m^2$. Fig. 13 gives an impression of the whole assembly. The cathode cylinder was made of thick Cu or Fe tubes with longitudinal slots containing Zr strips. The annular gap between the inner and outer electrode cylinders is about 3 cm. Gas, electric power and cooling water is supported only from the upper anode front side, where electrical insulation and vacuum tightness is provided by a 1.0 mm enamel layer. A cap welded to this side of the spent pump provides a second containment for Kr.



IHCH

Fig.13 KR ION SPUTTER PUMP DESIGN PRINCIPLE

Matrix material peeling off in case of malfunction can be collected without disturbance at the lower spherical front side. The inner cathode cylinder is watercooled and the outer cylinder is aircooled by a fan to get better matrix properties at higher implantation temperatures of about 100 degrees C.

3. Pump Operation

The semi-technical pump is easier to operate due to its greater plasma volume. The electrode assembly must be fabricated very carefully because of the thin insulation gaps and the surfaces have to be smoothed.

At the beginning we used the Fe80/Zr20-cathode. The operation diagram, Fig. 14 is in good agreement with the data of laboratory pumps. Especially the pumping rates are remarkably higher.

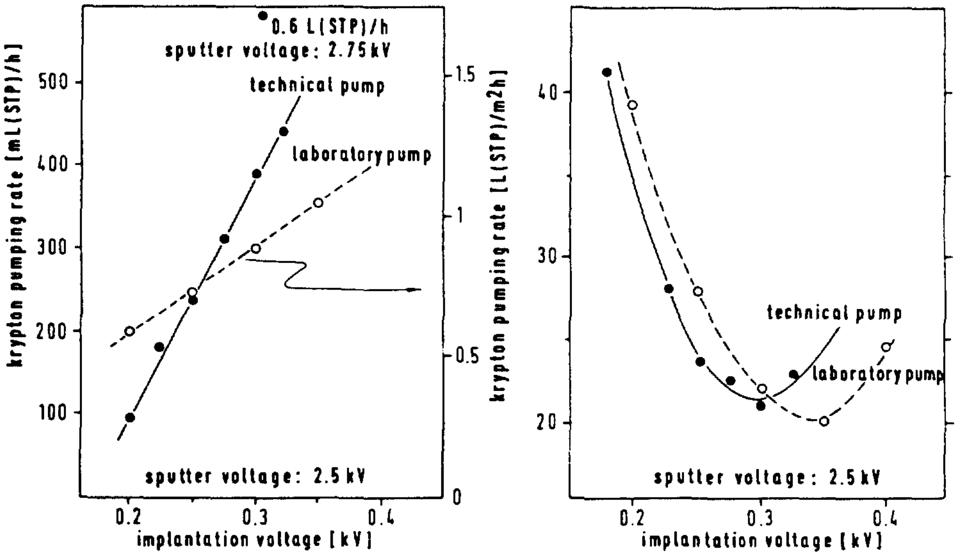


Fig.14 Data of the Fe 80 / Zr 20 technical pump

Fig. 15 gives the operation diagram of a Cu70/Zr30 cathode for a relative low sputtering voltage. We did not succeed until now to use higher sputter voltages and resulting in higher pumping rates because of extension cracks in the insulation enamel. These small crevices arose during the fabrication process. The pumping efficiency however corresponds very well to the laboratory scale pump.

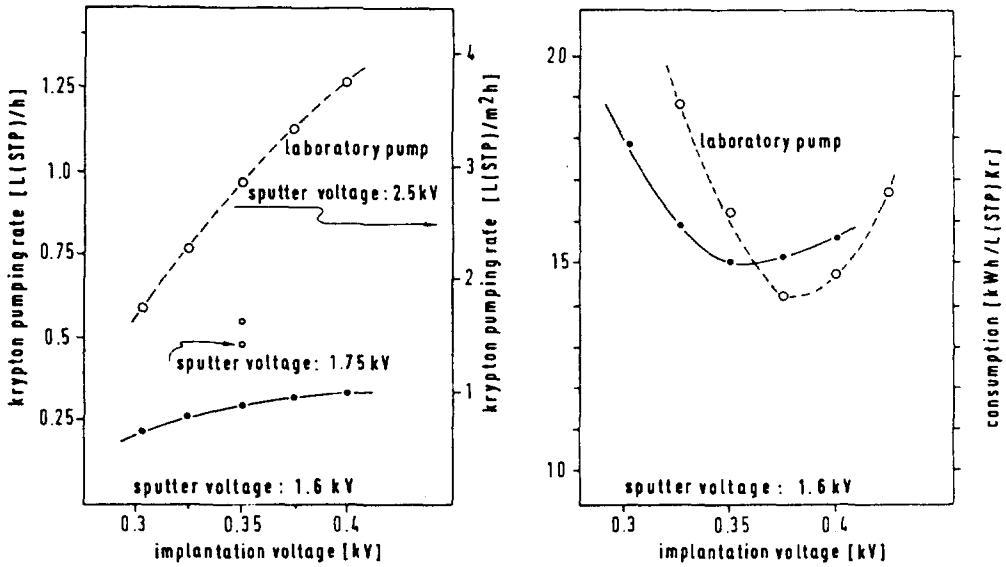
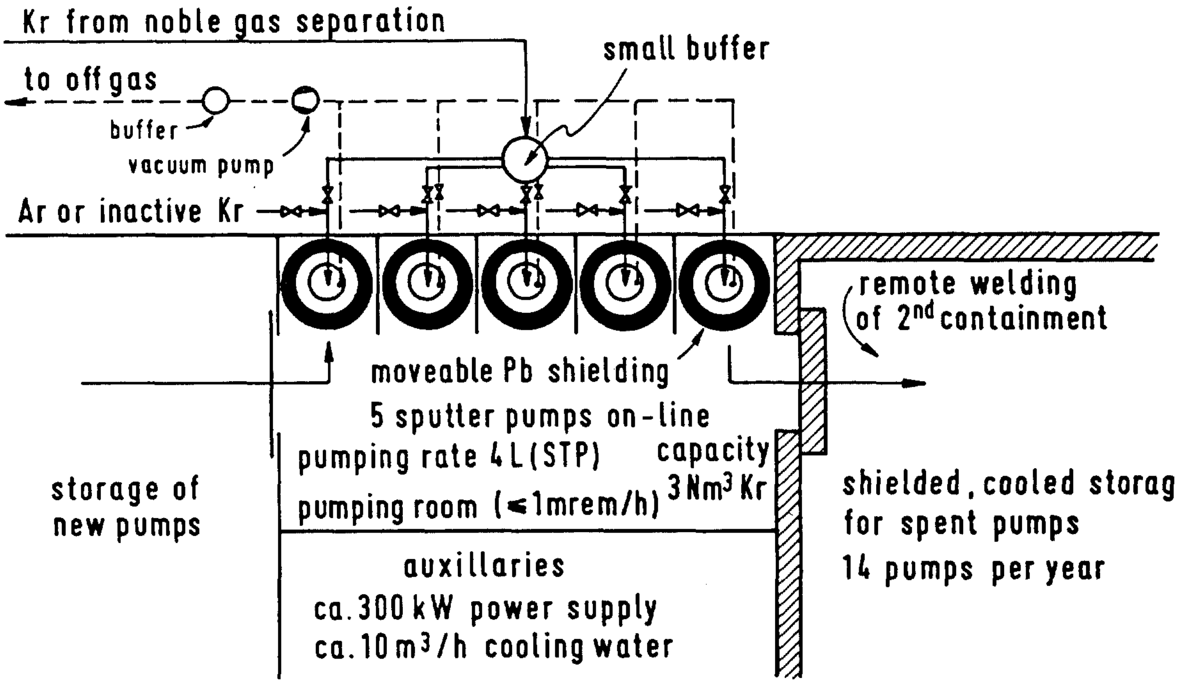


Fig.15 Preliminary Data of the Cu 70 / Zr 30 technical pump

IHCH

IV. Design of an Application

The practical application of such a sputter pump for a reprocessing plant is given schematically in the Fig. 16. The technical pump has a sputter cathode diameter of 0.3 m and a length of 1.2 m, which results in an active surface of 1.0 m^2 with a capacity of 3000 L (STP) of Kr. With a pumping rate of 4.0 L(STP)/h the daily rate of one pump is equivalent to dissolution of 700 kg spent fuel per day with a burnup of 30000 MWdt. During operation and handling the pumps stand in a moveable lead shielding. In a nonshielded cell



KfK IHCH

Fig. 16 Design of a reference concept
(350 tons spent fuel per year)

with separate ventilation some of these pumps operate simultaneously. The total pumping rate is in accordance with the produced Kr plus a reserve for changing a spent or a damaged pump. Small buffers reduce the peak value during the dissolution of spent fuel.

The operation time for a pump is about one month. Damaged pumps will be changed without overhaul. For changing the pump it is operated finally with inactive Kr for flushing the pipes and burying the active Kr. After this operation, the gas, cooling water, and current supply will be removed and the frontside will be closed by remote welding of the end cup. Now the pump case is the second containment for a safe storage.

References

(1) Entwicklung integrierter Auflöserabgassysteme
E. Henrich, U. Bauder, F. Weirich, T. Fritsch;
Europ. Konf. Behandlung der Abluft in Kerntechn. Anlagen
Luxemburg, 14-18.10.1985

(2) D. W. Whitmell;
18th DOE nucl. Airborne Waste Management and Aircleaning Conf.
(1984) Proceedings S. 667

(3) G. L. Tingey, E.D. McGlanahan, M.A. Bayne, W. Gray;
IAEA-SM-345/31 (1980)

(4) H.J. Schmidt, E. Henrich, F. Baumgärtner;
16th DOE Nucl. Air Cleaning Conf., Proceedings S. 1019 (1980)

(5) G. L. Tingey, E.D. McGlanahan, J. M. Lytle, N. R. Gordon,
R. W. Knoll; in "Methods of Kr-85 Management" Vol. 10
(1983) /7/ S. 229

(6) H. J. Schmidt; Dissertation Univ. Heidelberg (1980)

(7) H. J. Schmidt, E. Henrich, T. Fritsch, F. Gompf, B. Renker,
E. Mohs;
Proc. Int. Conf. On Liquid and Amorphous Metals, Grenoble
(1980)

CLOSING COMMENTS OF SESSION CO-CHAIRMAN MOTES

The papers presented here have been concerned with the recovery and treatment of Kr-85. It is notable that this was the only radionuclide addressed, although papers concerning iodine and tritium will be given in Session 8. Work concerning krypton recovery is apparently reaching a relatively mature developmental stage. With work being largely focussed on final details and optimization, a continued decline in this work is to be expected. Ion implantation is also well along in development, with scale-up and optimization being considered, although testing of long-term waste form adequacy remains, and differences in the metal matrix composition are noteworthy. While some work remains to be done on krypton and iodine, and considerably more on tritium and carbon, the lack of an identifiable need results in very modest support and the likelihood that the required technologies would not be available if reprocessing were to move forward; in particular, the absence of generally accepted disposal technologies for each radionuclide. Attendant acceptance criteria uncertainties regarding allowable releases of tritium and carbon, and the lack of integrated recovery and treatment flowsheets, may well become limiting constraints.

SESSION 7

VENTILATION AND AIR CLEANING IN REACTOR OPERATIONS

TUESDAY: August 19, 1986
CHAIRMEN: R.R. Bellamy
U.S. Nuclear Regulatory Commission
A.G. Evans
E.I. DuPont de Nemours

DESIGN APPLICATIONS FOR PROPOSED NEW SOURCE TERMS
R.G. Eble, Jr.

CONTROL ROOM HABITABILITY STUDY - FINDINGS AND RECOMMENDATIONS
J.W. Driscoll

CAPTURE OF IODINE IN SUPPRESSION POOLS
P.C. Owczarski, W.K. Winegardner

ABSORPTION OF AIRBORNE MOLECULAR IODINE BY WATER SPRAYS
M.F. Albert, R.P. Wichner, P.K. Baumgarten,

LOWERING COSTS AND ENHANCING SAFETY WITH THE NUCLEAR-KARLSRUHE FILTER SYSTEM
M. Ohlmeyer, W. Stotz

SIMULATION OF VENTILATION NETWORKS: PRESENTATION OF THE SAFETY CODE PIAF COMPLEMENTARY WITH FLUID MECHANICS CODES
Ph. Perdriau, S. Raboin, Ph. Mulcey, G. Martin, Ph. Pruchon

NRC FORM 335
(2-84)
NRCM 1102,
3201, 3202

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

SEE INSTRUCTIONS ON THE REVERSE

1. REPORT NUMBER *(Assigned by TIDC, add Vol. No., if any)*

NUREG/CP-0096, Vol. 1
CONF-860820

2. TITLE AND SUBTITLE

PROCEEDINGS OF THE 19th DOE/NRC AIRBORNE WASTE MANAGEMENT AND AIR CLEANING CONFERENCE

3. LEAVE BLANK

4. DATE REPORT COMPLETED

MONTH	YEAR
April	1987

5. AUTHOR(S)

Melvin W. First, Editor

6. DATE REPORT ISSUED

MONTH	YEAR
May	1987

7. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*

Harvard School of Public Health
655 Huntington Avenue
Boston, MA 02115

8. PROJECT/TASK/WORK UNIT NUMBER

9. FIN OR GRANT NUMBER

Grant No.
NRC-04-86-304

10. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*

Division of BWR Licensing
Office of Nuclear Reactor Regulation
Nuclear Regulatory Commission
Washington, DC 20555

11a. TYPE OF REPORT

Conference Proceedings

b. PERIOD COVERED *(Inclusive dates)*

12. SUPPLEMENTARY NOTES

13. ABSTRACT *(200 words or less)*

This document contains the papers and the associated discussions of the 19 DOE/NRC Nuclear Air Cleaning Conference. Sessions were devoted to (1) fire, explosion and accident analysis, (2) adsorption and iodine retention, (3) filters and filter testing, (4) standards and regulation, (5) treatment of radon, krypton, tritium and carbon-14, (6) ventilation and air cleaning in reactor operations, (7) disolver off-gas cleaning, (8) adsorber fires, (9) nuclear grade carbon testing, (10) sampling and monitoring, and (11) field test experience.

14. DOCUMENT ANALYSIS – a. KEYWORDS/DESCRIPTORS

Adsorption	Filtration
Air Cleaning	Ventilation
Carbon Testing	Waste Management

b. IDENTIFIERS/OPEN-ENDED TERMS

15. AVAILABILITY STATEMENT

Unlimited

16. SECURITY CLASSIFICATION

(This page)
Unclassified
(This report)

17. NUMBER OF PAGES

18. PRICE

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE, $300