OPENING SESSION

COSTELLO: I would like to call the Seventh Air Cleaning Conference to order, and act as chairman for this morning's session. I am from the Environmental and Sanitary Engineering Branch, AEC Headquarters in Washington. There have been some recent changes in our organization. Dr. J. A. Lieberman, who was in charge of our Branch, is now the Assistant Director for Nuclear Safety of the Division of Reactor Development, and Mr. Belter is Chief of our Environmental and Sanitary Engineering Branch.

I want to be the first to welcome you all here this morning. Namely, this includes representatives from all the AEC offices and their contractors, foreign countries and attendees from other government agencies as well as private industry. I believe the total number of representatives will run over 125.

Very briefly, the plan is to have two and a half day technical sessions with a visit around Brookhaven National Laboratory Thursday afternoon. We also have a local visit planned for Friday morning to a high efficiency filter manufacturer's plant. It is planned to put out a revised agenda and a current list of attendees.

About 46 papers were included in the proceedings of the 6th Air Cleaning Conference at Idaho in July 1959, and this year we have reduced the number to about 37 papers, hoping that this will give more time to each speaker. In addition to improving the program by allowing more time for each paper there will be additional time for discussion and also a short review and tie-in with the previous meeting. Further improvements have been the inclusion of a summary and conclusion, together with a session to plan an agenda for future meetings. Mr. Dennis will chair that meeting, so between now and then, if you have any material, you can present it to him.

One of the important parts of this conference is the recording of the technical discussions. We have Dr. Morgan and Mr. Stevenson of V. M. I. under the Johns Hopkins University contract, who will be in charge of the recordings and handling the proceedings of the conference.

I will conclude by thanking Lee Gemmell and his staff, Mrs. Cooper and all the other people at Brookhaven who have provided these excellent facilities and for making all the local arrangements in order to set up this conference. I also want to thank the staff of the Harvard Air Cleaning Laboratory for helping us work up the agenda and other material and, lastly, my own secretarial staff, in Washington, who worked so diligently to get these last-minute papers and notices out to you.

I now have the privilege of introducing the Director of the Brookhaven National Laboratory, who has taken time out of a busy schedule to come here this morning and welcome you to this meeting. Dr. Goldhaber has a long and outstanding scientific career in nuclear physics. He received his early education at the University of Berlin and Cambridge. After receiving his doctorate he came to the University of Illinois and was a professor of physics until 1950. He served as a consultant to the Argonne National Laboratory and joined the Brookhaven Physics Department in 1950. Dr. Goldhaber has received many special honorary awards for discoveries in his field and has served on many scientific committees. It is indeed an honor for me to introduce Dr. Maurice Goldhaber.

GOLDHABER: I see I can start exactly on time to welcome a group, only it happens to be the first group and I am supposed to welcome another group at 10 o'clock. I hope all of you who are new to Brookhaven will have a good chance to see what you find worth seeing here.

You know that Brookhaven is a Laboratory mainly devoted to pure research, as it is called, but no matter what you do today, whether it is pure or applied research, you cannot be entirely pure or entirely applied, if you want to be progressive. So you will find here quite a bit of applied research that is perhaps nearer to your field of research interest than some of our pure research. We have the advantage, on Long Island, of being surrounded by very clean air, though there seems to be always something in the air. I know there are people who suffer from the effects of what little is left and perhaps you will have some ideas on how to remove that. The great amount of grass you see around here has helped to keep the area around the Brookhaven reactor clean, so our filters have not much of a problem in catching what is left. We do have the problem left that Argonne-41 is produced in prodigious amounts in the reactor and out of the chimney that you have probably seen come 15,000 curies a day of Argonne-41. I am assured that this is no health hazard. However, it is a great annoyance to all researchers who have to worry about gamma ray background and if you have any idea on how to get rid of that, I would hope you will let us know. Surely it can be done at some great expense, but it is worth discussing whether there is any practical idea.

I need not tell you about the origin of Brookhaven. You know it started about 15 years ago, when nine eastern universities, some of the big universities, got together and started Associated Universities, Inc., and they are still running Brookhaven as a contractor for the AEC, and this arrangement has been a happy one all around. The university people can come here and do the kind of research they could not do at home because of the lack of special facilities. We have also a very active research staff right here and the happy medium is probably something like 50 percent of the people doing the research here and 50 percent coming in; if you would not have a good, continuing staff, problems would be left hanging in the air and the interaction between people and machines is much poorer when they cannot talk through other people and, in this sense, we feel that we have hit on something like a reasonable formula for keeping a balance between the staff here and the staff which comes from the outside.

I will not take any more of your time, since we are all behind time, and I again say enjoy your conference.

COSTELLO: Thank you very much, Doctor Goldhaber. Next, I would like to introduce Walter Belter from our Environmental and Sanitary Engineering Branch in the Washington office; he has recently been appointed Chief of this Branch.

BELTER: First of all, I would like to express the sincere regrets of Dr. Joseph Lieberman for not being able to be here. He was planning, right up until five o'clock last evening, to be on a plane to get here in time for this morning's session. However, in his new position, schedule changes are required on short notice and at five o'clock he found out that he could not be with us today. I am sure he honestly regrets not being here, since this will be the first Air Cleaning Conference that he has not been able to participate in since these meetings have been held.

As you realize, this is the Seventh Air Cleaning Conference. At present, we are striving to hold these meetings approximately every two years. They started back at Harvard University in June of 1951. Some of you were present at that first meeting.

We certainly have come a long way since then. You could say we have made a complete cycle. During the early days it was considered desirable to remove the particulate material from gas streams by utilizing some type of liquid processing step. It was felt that our liquid waste treatment systems were adequate for handling the resultant waste streams. Now, we are almost in the reverse position - the liquid waste people are striving to convert liquid wastes to a solid form.

As many of you remember, in the meeting we had at Idaho two years ago we discussed the gas cleaning problems that are associated with the high-level waste calcination program, especially our fluidized bed calciner. Mr. Wheeler of Idaho can attest to the many problems that are involved in the removal of gaseous particulates as they are generated in the fluidized bed calcination of chemical reprocessing waste.

I would like to express my thanks to Dr. Silverman of Harvard and his staff for helping to arrange this program and to Lee Gemmell of Brookhaven for making the many arrangements necessary for holding a meeting of this type.

I have also noticed some of our foreign visitors in the audience and would like to extend a special welcome to them.

We are hopeful, too, that in having Dr. Morgan tape these proceedings we are going to be able to cut down on the time lag which is involved in getting out the proceedings for these meetings. We feel that one of the important objectives in holding meetings of this type is to get the information out to the people in the field.

On the agenda we have, as Mr. Costello has indicated, something like 37 papers to present in approximately five sessions. The first 1-1/2 days will be devoted to reports from various installations -- new developments, new problems and also some of the newer equipment developments: Mr. Gilbert will report recent results of our high-efficiency filter testing program. Also on the program is a description and results of the monitoring program which followed the very unfortunate accident at the SL-1 installation in Idaho. We are looking forward to a report on the present status of the Hanford reactor confinement program.

Wednesday afternoon, you will note, is devoted primarily to reports from the Harvard Air Cleaning Laboratory on recent work which has been conducted during the past year. We will have a review of the iodine collection studies and some of the problems involved in the institutional incinerator design and also another further look at the economics of air and gas cleaning within the AEC.

Dr. Silverman will discuss some of the new reactor confinement concepts which are being investigated at Harvard.

Thursday morning we are looking primarily at the meteorological aspects of air cleaning: What happens to our gas streams after they are discharged to the atmosphere? I will not take up any more time now, except to also express my own pleasure in having all of you present here. We hope there will be sufficient time for each of you to contribute actively or participate in the program. We feel that the real value of these meetings is obtained by giving everyone an opportunity to express his ideas and also to discuss some of the problems which are being generated at the various sites.

COSTELLO: We will now turn to Dr. Leslie Silverman of the Harvard Air Cleaning Laboratory who will give us a short review of the last conference and new problems at the sites.

SILVERMAN: On behalf of the Harvard Air Cleaning group, I would like to welcome the attendees. Even though we at Harvard have a perfect attendance record at these air cleaning conferences, about 50 percent new faces appear at these conferences. It appears to be a fact that there are more and more people concerned with air cleaning problems and are now being brought into the program.

In the interest of saving time, I thought I would make my review extremely short, and leave out discussion of Research and Development problems at the sites because we are running far behind schedule.

If you look in the 6th Air Cleaning Conference Report, TID-7593, you will find a rather complete coverage of the subject. Glancing at the table of contents you will note that there have been a number of problems that have already been mentioned this morning. I think it is appropriate to comment that since we had our last meeting, at Idaho, one of our more serious air cleaning problems has been solved simply by means of an Executive Order; the ANP program was cancelled. However, the rest of the nuclear propulsion systems for rockets, Rover, and the Pluto project, and a ram-jet, can become problems of a first order of magnitude and they are not going to be any less severe in the future -- in fact, they will become more severe. Associated with this situation are problems of ground testing of nuclear propulsion devices, which means emission of fission gases and particulates, and the problem of putting package power units into orbit. The SNAP program involves fairly high concentrations of heavy isotopes in thermal devices and small reactors that will be put into orbit. A misfire, or a near miss, may result in radioactive contamination in the vicinity of the launch, These are very real problems that we must be thinking about in the near future.

Associated with air cleaning also are a lot of problems associated with the use of mobile or portable power stations. These include the pressurized water reactors and also the boiling water types for the Antarctic and the Arctic. This problem is one of rare gas and particulate releases. The United States has signed an international agreement that it will not contaminate the Antarctic region above present background levels. We must then make specific confinement of particulates, liquid waste and, we believe, gases as well. At the moment our own people, the cosmic physicists and others that want to measure background at the polar regions are the ones that are demanding no increase in background levels. Well, background, as you know, is something that changes more by politics than it does by scientists, but, nevertheless, these levels that have been proposed are so low that they can't be measured, and this also poses a problem for the people designing and measuring performance in air cleaning. You must then have a device that works so well that you can't even measure what is coming out!

There are other problems that are either on the horizon or bothering us now. At the last air cleaning conference we had a quite a session on the deterioration and damaged condition of filters before they were installed. These problems were thrashed out at the last Idaho meeting and the AEC has followed through by setting up regional testing facilities. There is now quite a bit of concern about whether or not the devices that have been factory tested and have met certain specifications actually have these values when delivered. Field testing will answer this problem; in addition there are always problems in installation. Poor maintaining or handling may make a filter which has met specifications and in testing prior to installation fail to meet requirements. This may be the result of mishandling by some careless workman. This is not a reflection on Atomics International, but I can remember seeing their installation at the SRE several years ago during construction. They had high efficiency filters ready to be installed and the workmen were sitting on them eating their lunch. It is quite likely that these filters would be effective for removing sandwiches, but not much else.

In terms of other problems, we have the rare gas release problem associated with chemical processing and the rare gas problem associated with reactor operation. Problems in relation to iodine removal exist in both cases.

In regard to reactor aerosols I would like to mention one informal meeting on air cleaning that has taken place between the Idaho meeting and this one. This was a meeting on aerosol problems in reactors held at Harwell last December. Mr. Dennis, of our staff was present, as were others that are here, or who have been at our air cleaning conferences. We hope we can needle the British into putting out all the papers presented at that conference. At the moment, we managed to get copies from the individual authors and it could be possible that we can get the AEC to publish or reproduce them if the British cannot see their way clear to do so.

The air cleaning conference reports, even though they have been delayed in release, have presented a tremendous amount of information which has been widely used. For example, the work down at Oak Ridge on iodine and rare gas removal has been used in the design of the facilities going into the Antarctic. This indicates that our current information being developed goes right to work at contractor and licensees' installations. A lot of what will be presented here will likely move into the program.

Our own experience with this rapid utilization has been somewhat dismaying. We found that on the N.S. Savannah they adopted our silver-plated copper filter before we had finished testing it and had not resolved all of our own questions. We are not trying to back away from this as a useful device, but we did feel at the time it would have been useful to have a little early warning of their proposed application.

I simply point this out to indicate that sometimes we are on top of problems when people are pulling the bottom out from under us. Many of you have problems which can be and should be brought up here. One of the obligations of the Harvard Air Cleaning Lab as an AEC contract facility is to try to keep abreast of the many problems in air and gas cleaning.

The last item I wanted to discuss briefly is that since the last AEC Conference several gaseous and particulate releases have occurred as serious problems in air contamination. There is no use in my dwelling on the SL-1 accident. I think all air cleaners can heave a sigh of relief that this reactor had been shut down a few days before the accident happened. Even as it was, the SL-1 iodine release was significant and easily measured at Idaho. We have a plutonium release and other gaseous releases at Oak Ridge, and we have had two criticalities at Idaho. I am not pointing the finger at Idaho, because the reason these projects are there is that the air cleaning problems may be a serious one. Nevertheless, these release incidents should teach us some lessons. We hope to derive information from analysis of these accidents and criticalities that will give us a clue as to how to protect against them.

For many years the Harvard Air Cleaning Laboratory has been trying to get people at the sites to sample their stacks. They all have implicit faith in their design engineers. When a unit is installed with a ninety-nine percent removal as actual performance, we say it is necessary to evaluate. I am happy to say that in at least three places a good testing program is under way. We will get from these studies an idea of how well these devices perform, what the deficiencies are that develop in continued use and what should be done to improve the overall performance.

With these few remarks, I think I have skirted the areas that Mr. Costello has asked me to skirt and we look forward to hearing maybe some answers to some of the questions I have already raised.

<u>COSTELLO</u>: Are there questions about this summary?

<u>SILVERMAN:</u> I would like to comment from the rostrum, that there are a number of these conference reports in existence. These are identified in the list below:

#1. Harvard, June 12 - 15, 1951, (No proceedings issued).

#2. "Air Cleaning Seminar, Ames Laboratory (Ames, Iowa), September 15 - 17, 1952." WASH-147, March 1954, TIS Issuance Date, Technical Information Service, Oak Ridge, Tennessee. (OFFICIAL USE ONLY.)

#3. Los Alamos, New Mexico, September 21 - 23, 1953. (Proceedings not available.)

#4. "Fourth Atomic Energy Commission Air Cleaning Conference Held At Argonne National Laboratory, November (2 - 4), 1955." TID-7513 (Pt. 1),(June 1956, TISE Issuance Date) Available from Office of Technical Services, Department of Commerce, Washington 25, D. C. Price $1.50. (UNCLASSIFIED).

#5. "Fifth Atomic Energy Commission Air Cleaning Conference Held at the Harvard Air Cleaning Laboratory, June 24 - 27, 1957." TID-7551, (Issuance Date April 1958) Available from OTS/ DOC/ Washington 25, D. C. Price $2.00 (NO CLASSIFICATION MARK).

#6. "Sixth AEC Air Cleaning Conference, July 7 - 9, 1959." (Idaho Falls, Idaho.) TID-7593, (Issuance Date October 1960) Available from OTS/DOC/ Washington 25, D. C. Price $4.00.

The first conference did not produce a report, because this was a training conference. The second conference was held at Ames, Iowa. The third conference at Los Alamos resulted in a "classified document" which still remains classified. The fourth conference was held at Argonne National Laboratory. The fifth conference was again at Harvard. In this case, the report did not include the training aspects; the sixth conference was mentioned earlier and is in the list above.

There was an early report which is still classified which covered a stack gas working committee conference which preceded all these conferences. It was held in 1949 at Oak Ridge and, to my knowledge, that report is still classified.

We hope that one of the things that AEC will do is to take the wraps off some of these classified reports because I think they should be subject to declassification review at the earliest possible time.

A DESCRIPTION OF THE BNL RESEARCH REACTOR ENVIRONMENTAL AIR CONTROL, ALARM AND MONITORING SYSTEM

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ABSTRACT

This paper describes the use and functions of the Brookhaven Research Reactor air monitoring and control system. Emphasis is given to the automatic corrective air conditioning control and personnel warning features of the system as a reference in methodology and design for future installations.

The airborne radioactive particulate sample is conveyed from the sampling point to the remotely located instrument where it deposits on a moving filter paper tape. Refering to Figure 1, it is seen that the filter tape passes from the supply reel around the filter grid to the idler rollers and on to the take-up reel where it is stored. Adjustment of the tape speed is provided to suit operating conditions.

A beta-gamma radiation detector is positioned directly above the filter grid to count particulate activity at time of arrival. Delayed alpha counting is an optional feature that is not used in the Research Reactor air system. Alpha detection, when needed, is accomplished by locating a Zinc Sulfide scintillation detector downstream on the filter tape to allow some Radon product decay.

Arrangement of the drive mechanism is shown in Figures 1, 2 and 3. The filter unit consists of a stationary central pipe and a capstan or filter grid. The filter grid fits completely over and revolves about the central pipe with a .001 inch clearance between pipe and filter grid. The central pipe is bolted on and extends through the aluminum mounting plate. A 1.5 inch square hole is provided through the top of the pipe section under the GM tube and inlet air housing. Vacuum connections are made to the other side of the central pipe by way of a flange connection on the rear of the mounting plate. The filter grid consists of a capstan type drum with several rows of peripherally oriented holes. The filter tape passes around the filter grid and is slightly indented by the pressure drop across the paper insuring positive tape drive. Tape speed depends only on the filter grid speed. The tape take-up spool is driven at a faster-than-tape speed rate. A friction slip clutch in the take-up spool hub, maintains constant tape tension regardless of take-up spool loading.

HEALTH PHYSICS AIR SAMPLING FILTER PAPER TAPE TRANSPORT ASSEMBLY

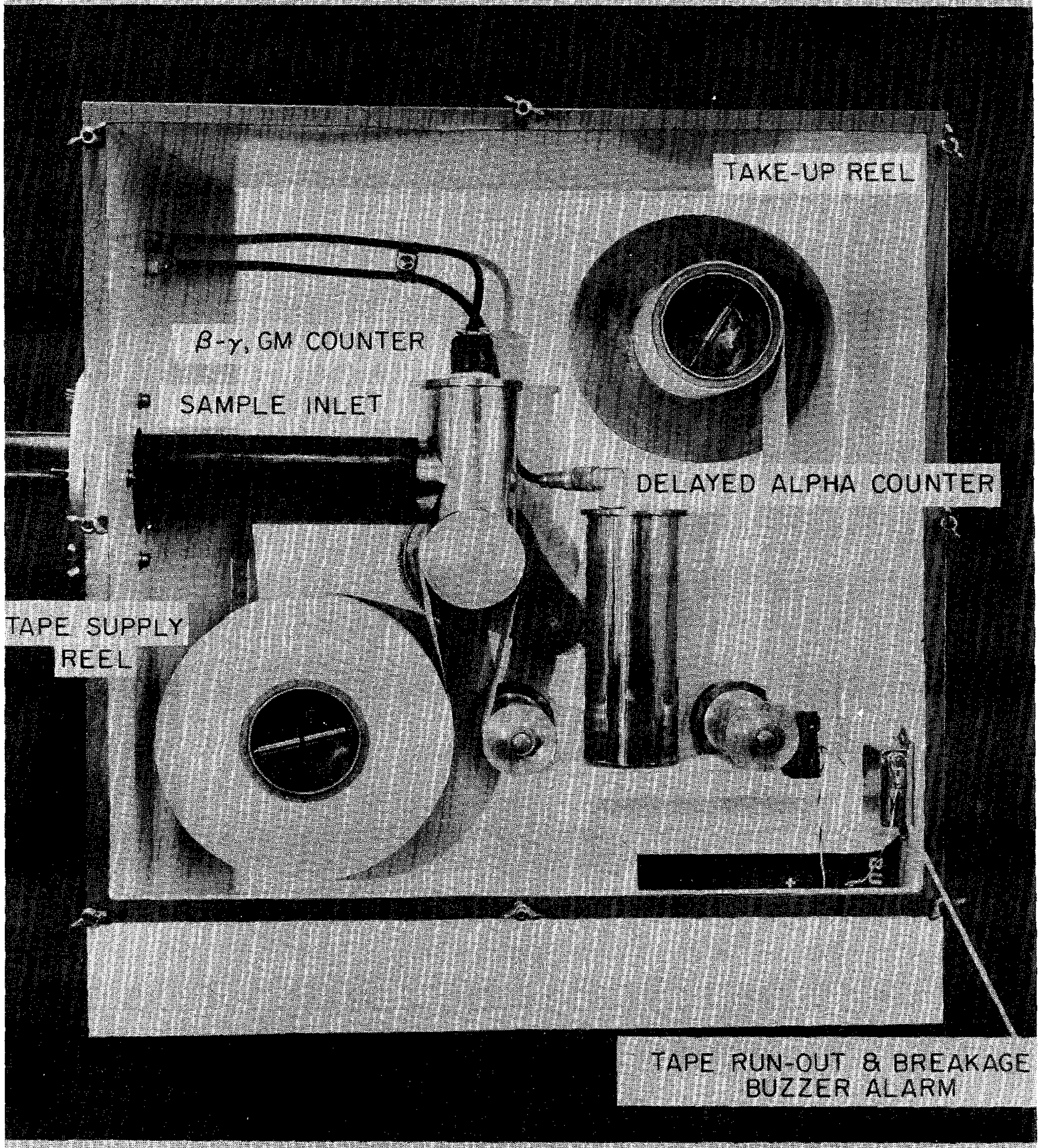
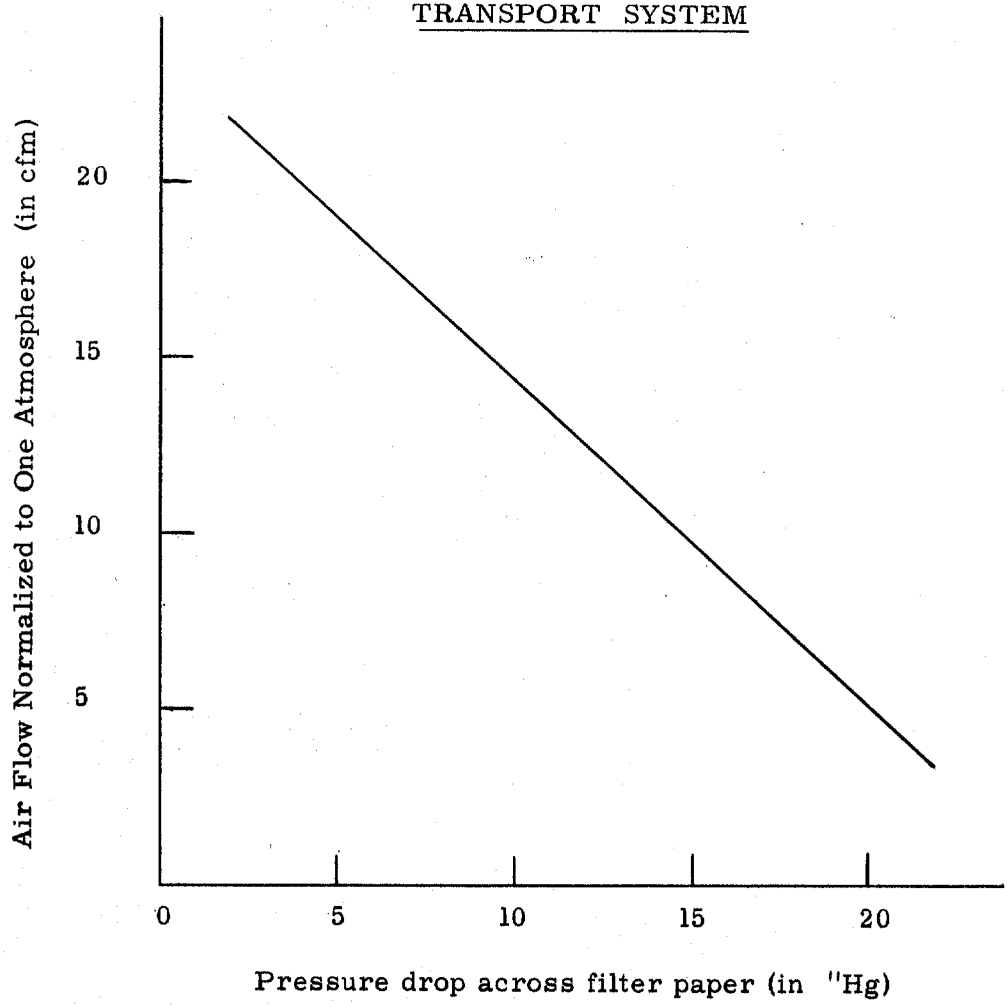


FIG. 1



MEASURED AIR FLOW
THROUGH THE BGRR
FILTER PAPER
TRANSPORT SYSTEM

Figure 1A

HEALTH PHYSICS AIR SAMPLING FILTER PAPER TAPE TRANSPORT ASSEMBLY

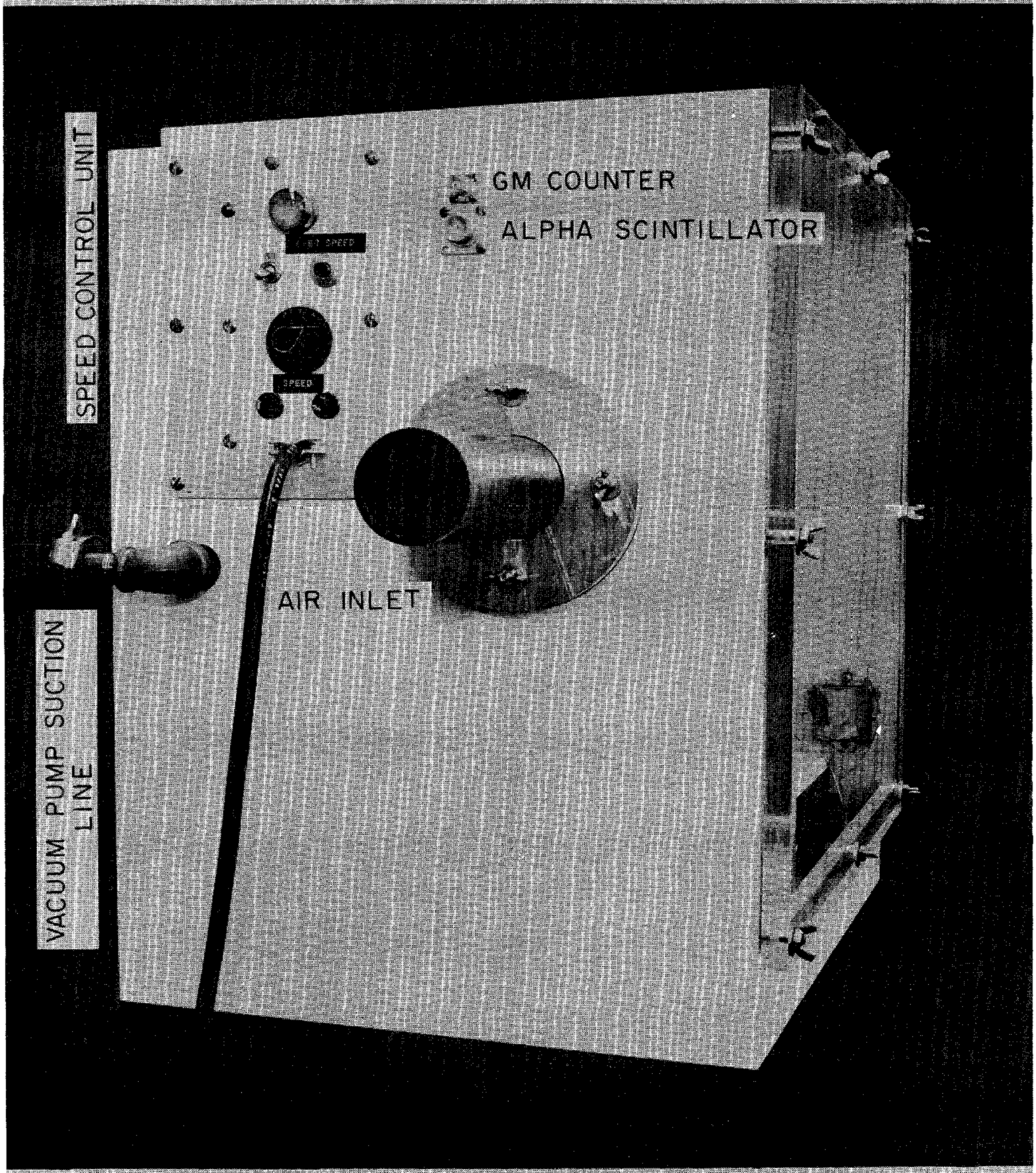


FIG. 2

HEALTH PHYSICS AIR SAMPLING FILTER PAPER TAPE TRANSPORT ASSEMBLY

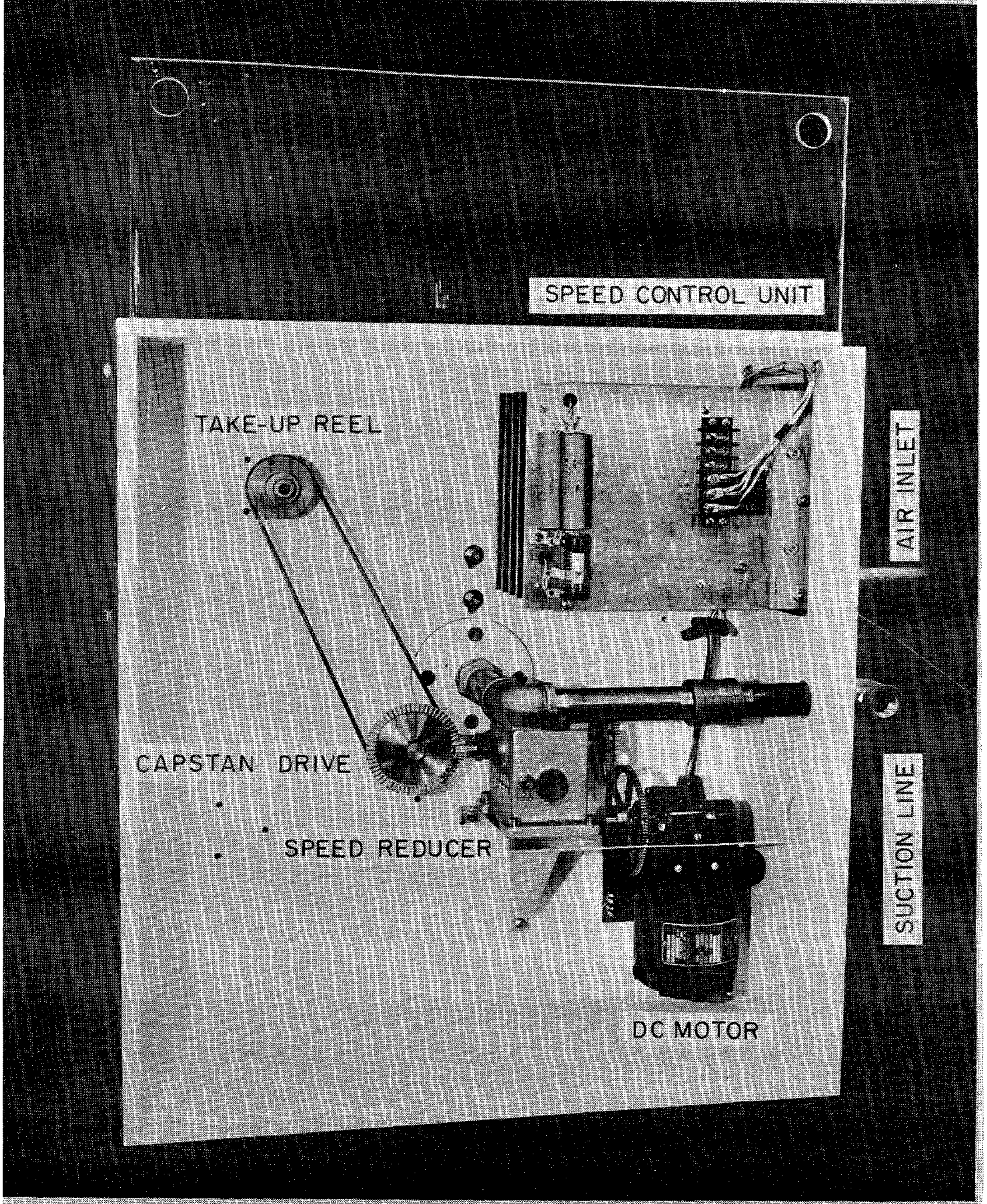


FIG. 3

Positive and reproducible speed control is accomplished by supplying a 115 VDC shunt wound Bodine instrument motor with Zener diode regulated field and armature potentials. A Zener diode potential divider supplies the several fixed voltages necessary for armature speed control. Stable speed control is insured by small load changes and very low Zener diode dynamic impedances.

The complete particulate air collecting and counting section of the instrument (Figure 1) is vacuum tight to prevent any dilution of the sampled air. The rear cover of the instrument is designed to be moderately resistant to dust.

A Gast Manufacturing Corporation Model 3040, 1 HP carbon vane vacuum pump establishes a flow rate of approximately 10 CFM at 14 inches of Hg through 9 mil Hollingsworth and Voss Number HV-70 filter paper. Figure 1A describes the air flow characteristics of the above mentioned system with any type of pumping equipment.

Two sampling mechanisms, as mentioned above, are used in the air monitoring and control system associated with the Research Reactor. The units sample from the North and South sides of the inhabitable air space surrounding the biological shield.

Figure 4A is a block diagram of the more important functions of the air monitoring and control system. Reference will be made to Figures 4 through 10 as an aside and the description will be solely based on Figure 4A.

Starting with the North side and pursuing from top to bottom, it is seen that the sampled air is drawn through a filter paper tape transport mechanism. (Figure 4) The resultant radioactive sample is directly viewed at time of arrival by a halogen quenched end window GM counter that is connected to a log count ratemeter. (Figure 5 and 6) Multiple ratemeter outputs are provided for activity level recording in the Health Physics Office, instantaneous level indications at the reactor console, level information to the Working Level Alarm Unit, and finally to an alarm trip signal to the Main Control Unit.

The Working Level Alarm Unit is a device with adjustable trips that are set to a level that corresponds to maximum permissible continuous airborne activity concentration (MPC) for unidentified Beta emitters. Trip level information is conveyed to the Grab Sample Controller and to the Main Control Unit. When activated by an alarm condition of the North Working Level Alarm Unit, the Grab Sample Controller initiates the collection of a high volume particulate air sample to provide physical and or radiochemical analysis without interrupting the main monitoring and control assembly. The sample is collected on a stationary Whatman 41 filter that is viewed by an end window GM counter connected to a linear ratemeter, recorder and alarm. The length of sample is limited to either a full scale count rate deflection or 10 minutes of sampling, depending on whichever occurs first.

Progressing from North to the South sampling unit an identical filter paper tape transport unit, GM counter, and log counter ratemeter are provided, (Figure 7 and 8). Similar and second channel use is made of the Log Count Ratemeter

BGRR NORTH AIR MONITORING FACILITY

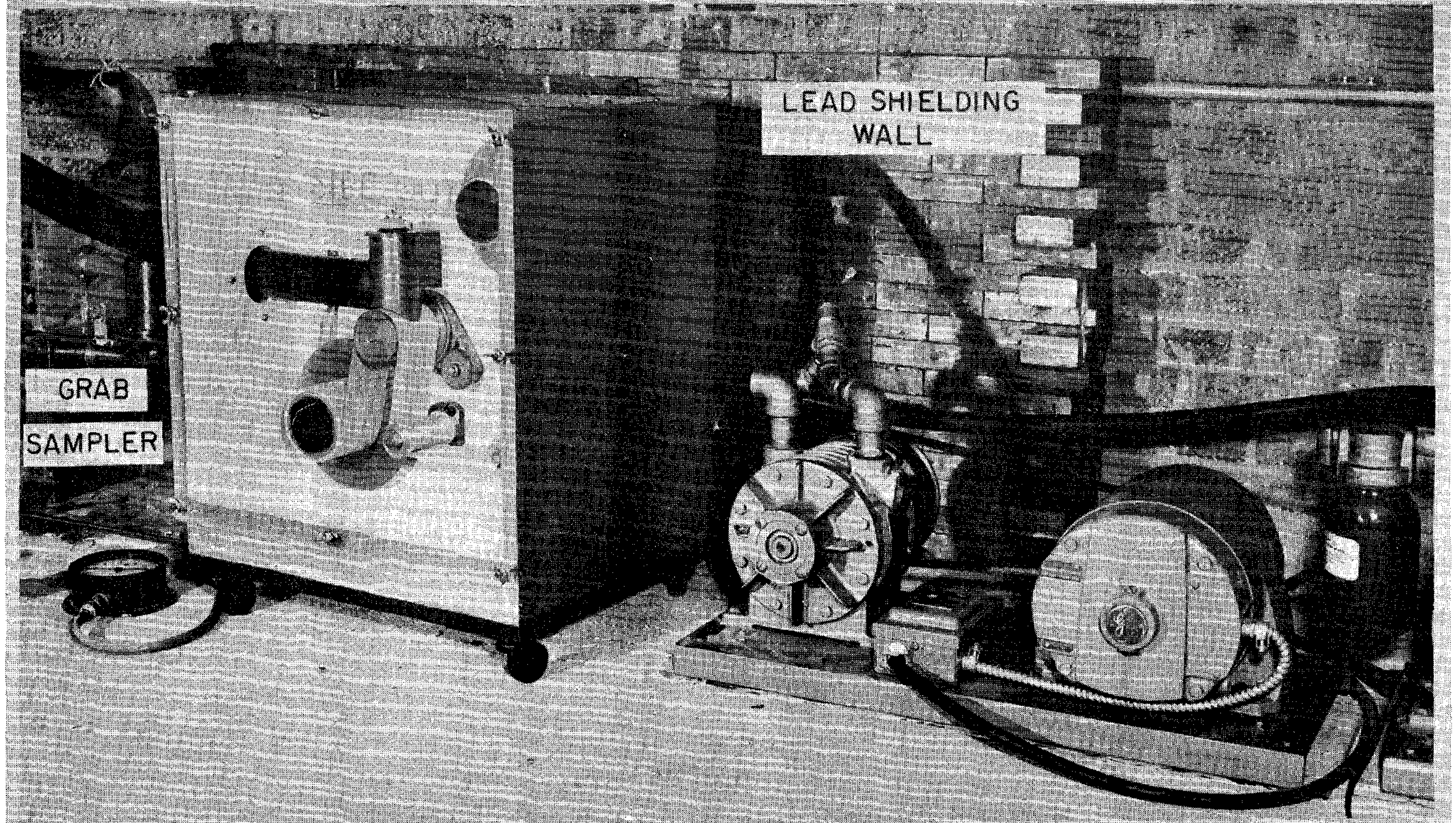


FIG. 4

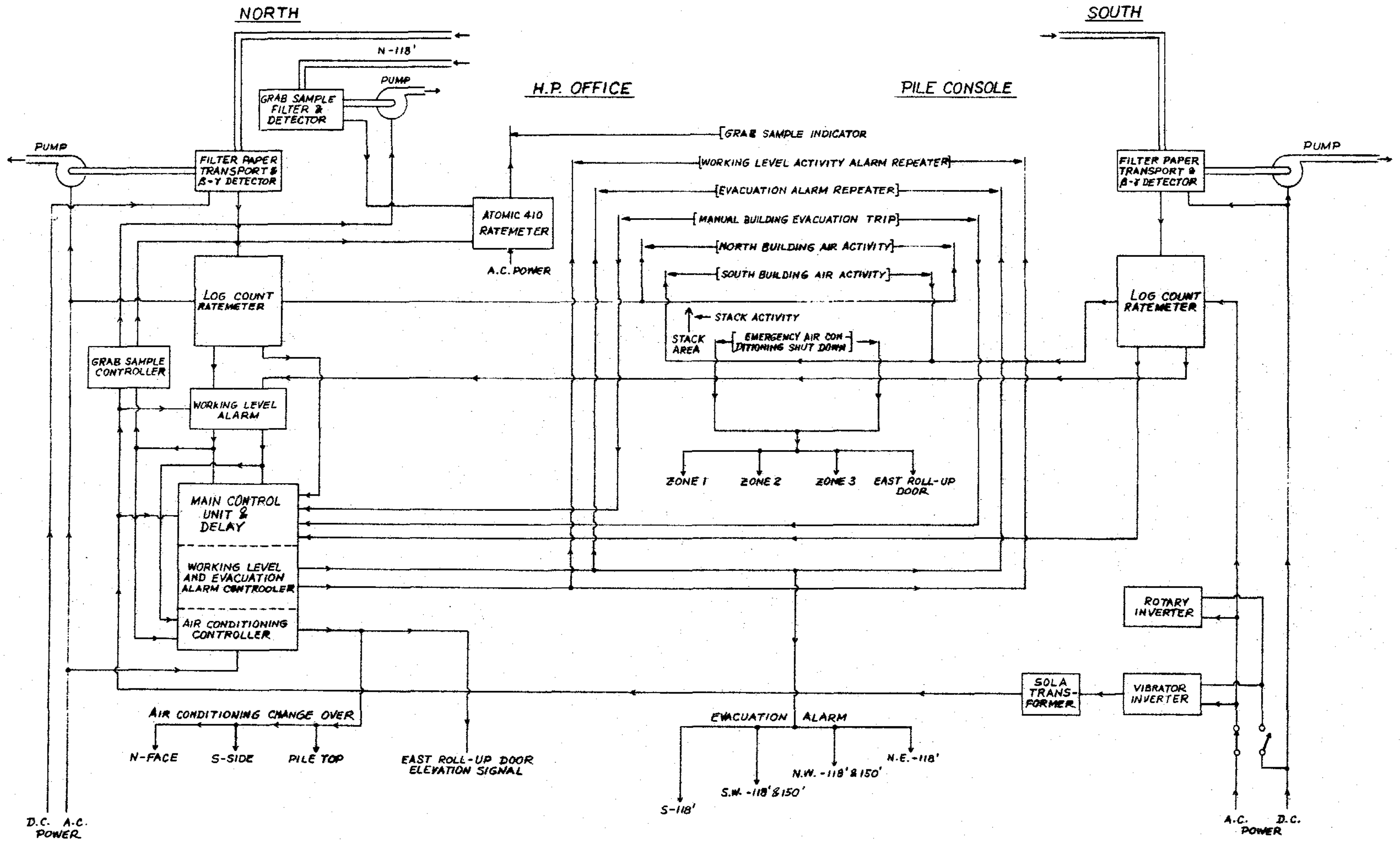

NORTH
N-118'
PUMP
GRAB SAMPLE FILTER & DETECTOR
H.P. OFFICE
PILE CONSOLE
SOUTH
PUMP
FILTER PAPER TRANSPORT & β-γ DETECTOR
GRAB SAMPLE INDICATOR
WORKING LEVEL ACTIVITY ALARM REPEATER
EVACUATION ALARM REPEATER
MANUAL BUILDING EVACUATION TRIP
NORTH BUILDING AIR ACTIVITY
SOUTH BUILDING AIR ACTIVITY
PUMP
FILTER PAPER TRANSPORT & β-γ DETECTOR
ATOMIC 410 RATEMETER
A.C. POWER
LOG COUNT RATEMETER
LOG COUNT RATEMETER
STACK ACTIVITY
STACK AREA
EMERGENCY AIR CONDITIONING SHUT DOWN
GRAB SAMPLE CONTROLLER
WORKING LEVEL ALARM
ZONE 1
ZONE 2
ZONE 3
EAST ROLL-UP DOOR
MAIN CONTROL UNIT & DELAY
WORKING LEVEL AND EVACUATION ALARM CONTROLLER
AIR CONDITIONING CONTROLLER
ROTARY INVERTER
SOLA TRANSFORMER
VIBRATOR INVERTER
AIR CONDITIONING CHANGE OVER
N-FACE
S-SIDE
PILE TOP
EAST ROLL-UP DOOR ELEVATION SIGNAL
EVACUATION ALARM
S-118'
S.W.-118' & 150'
N.W.-118' & 150'
N.E.-118'
D.C. A.C. POWER
A.C. D.C. POWER

FIG. 4A

BGRR BUILDING AIR CONDITIONING CHANGE-OVER &
DUAL AIR MONITORING ALARM CONTROL ASSEMBLY

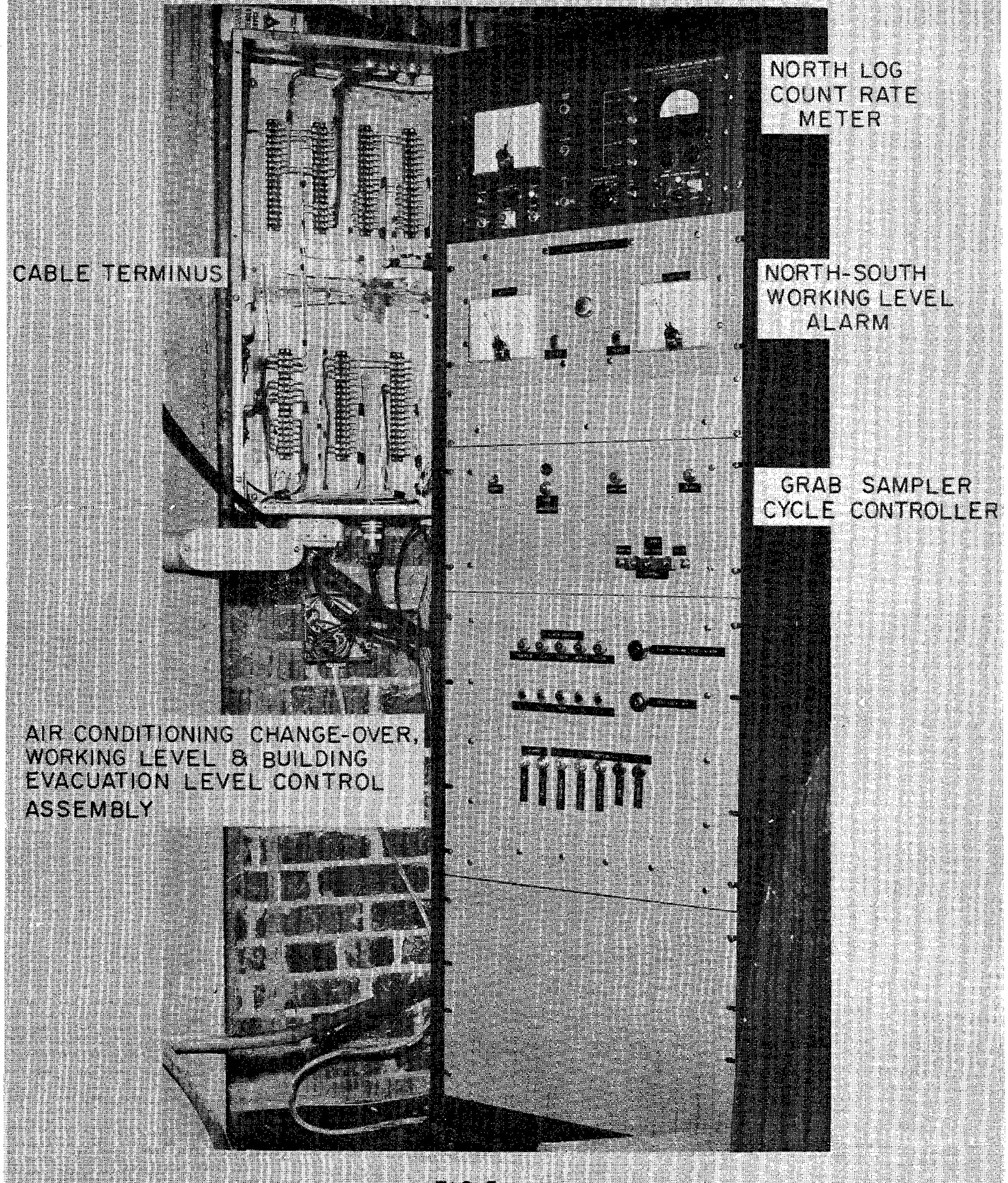


FIG. 5

BGRR BUILDING AIR CONDITIONING CHANGE-OVER & DUAL AIR MONITORING ALARM CONTROL ASSEMBLY

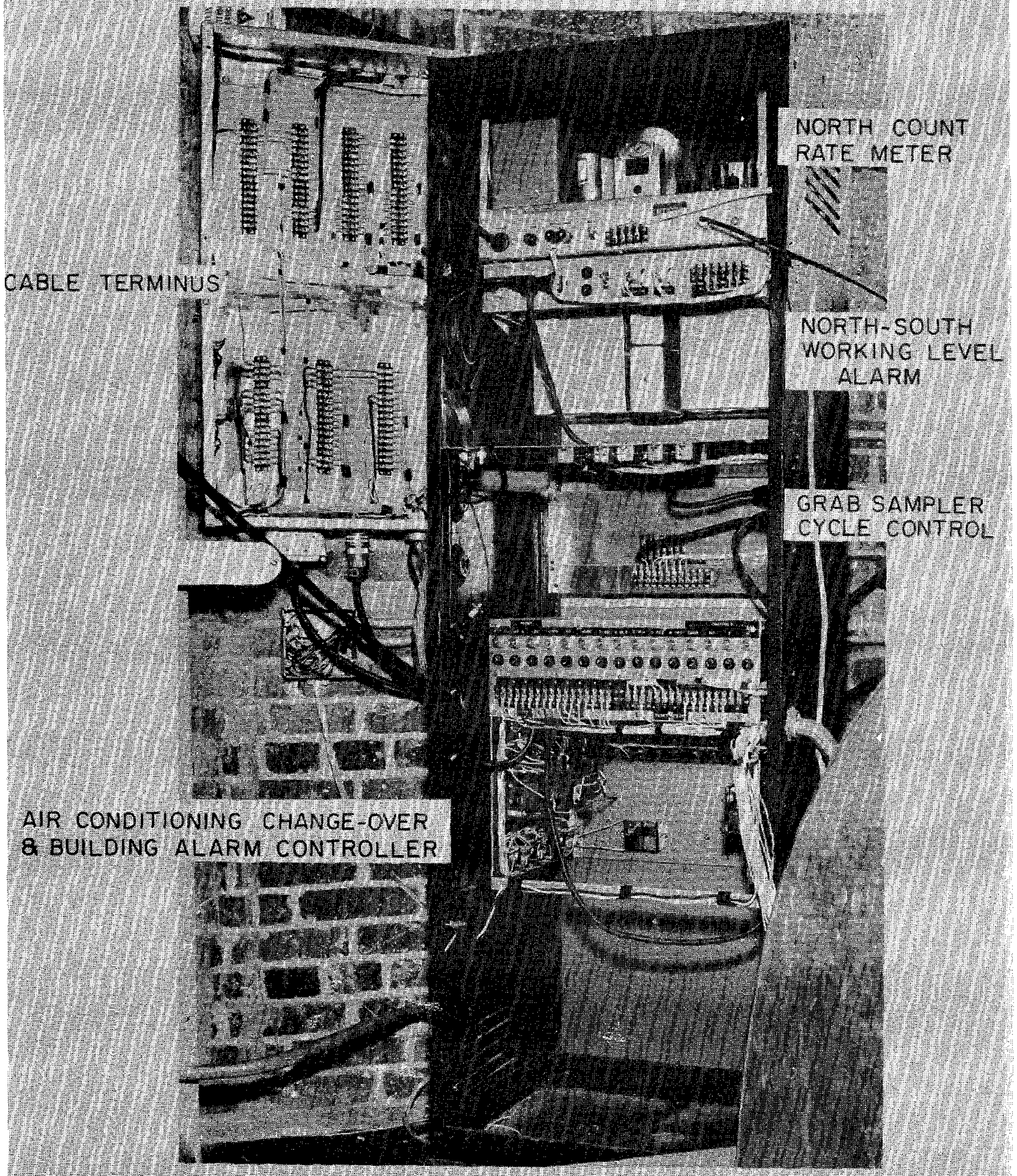


FIG. 6

BGRR SOUTH AIR MONITORING FACILITY

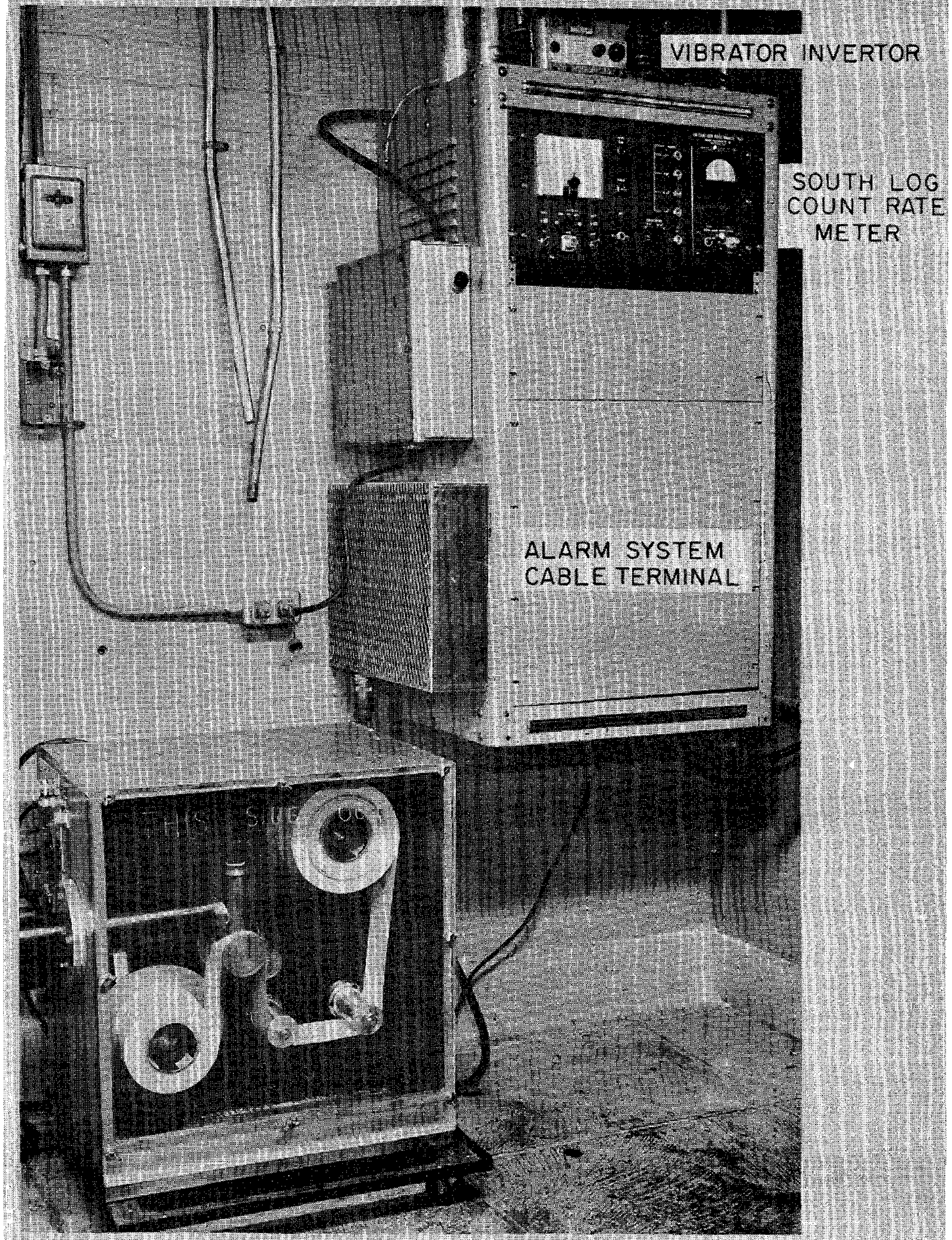


FIG. 7

BGRR SOUTH AIR MONITORING FACILITY

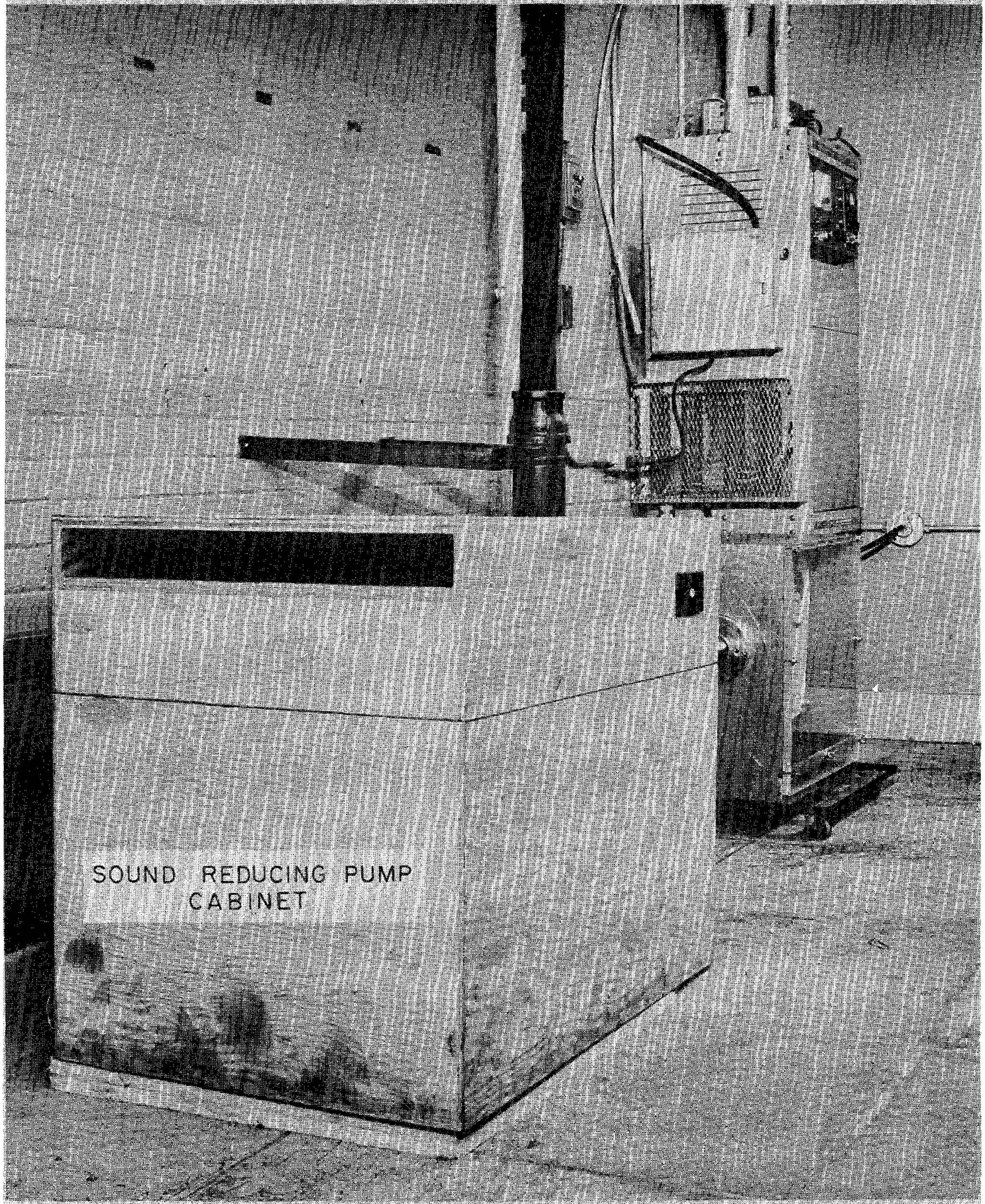


FIG. 8

BGRR HEALTH PHYSICS AIR MONITORING CONSOLE

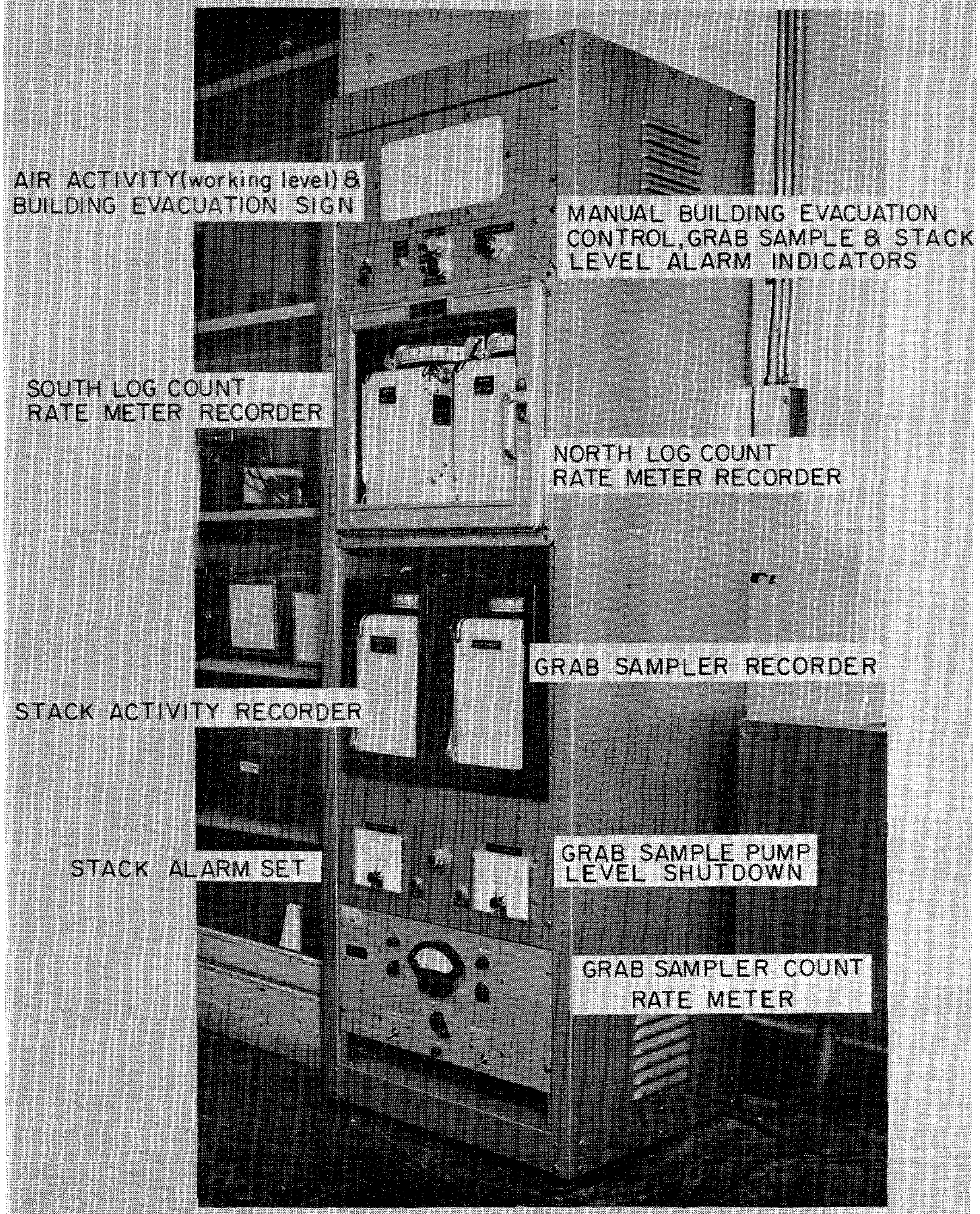


FIG. 9

BGRR BUILDING EVACUATION SIGN & HORN

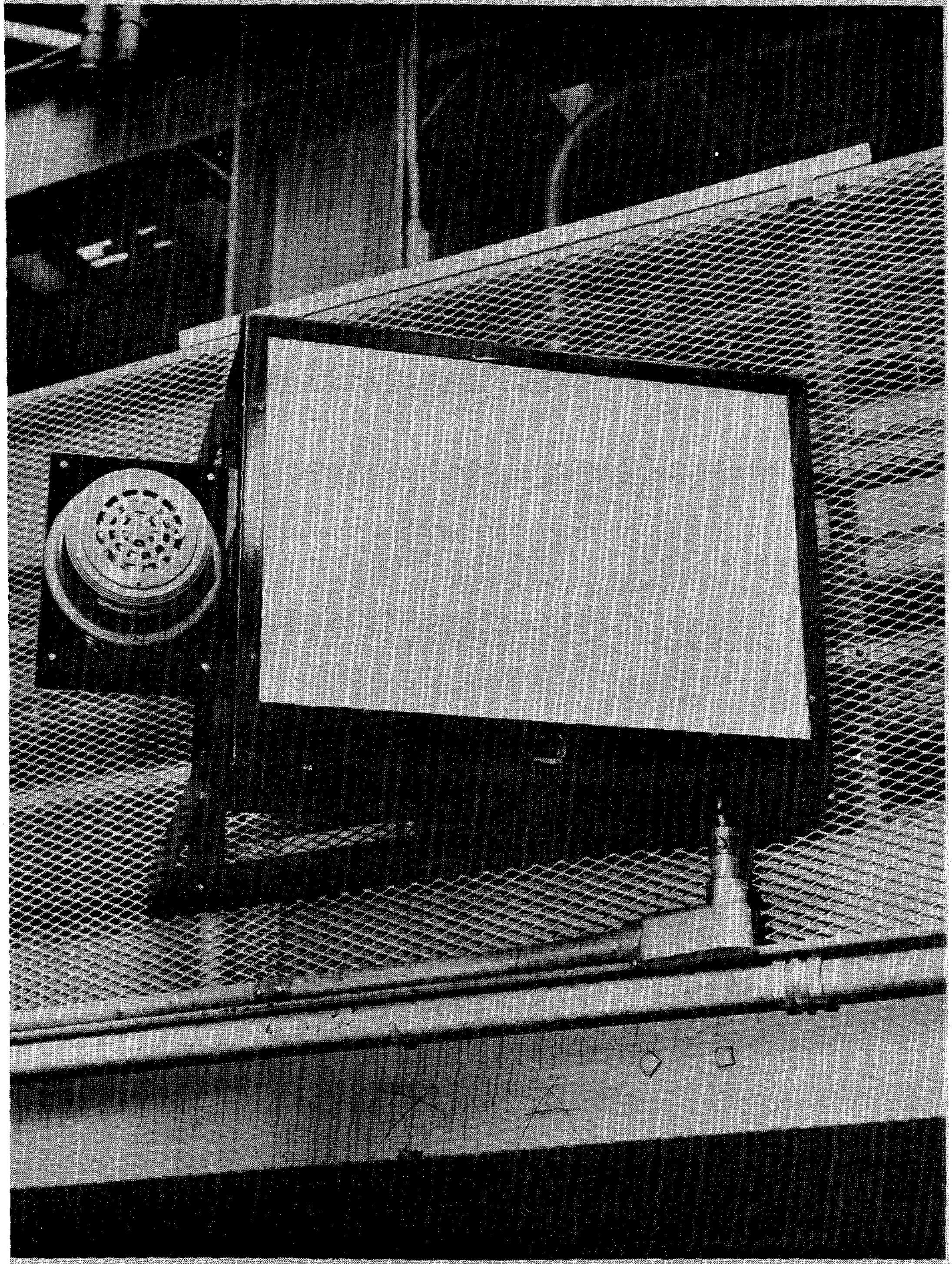


FIG. 10

outputs for purposes of recording in the Health Physics Office, indicating activity levels at the Reactor console, Working Level Alarm Unit information, and a high level alarm trip to the Main Control Unit. The separate high level alarm trip is identical to that provided by the North unit, and is adjusted to about 1000 times MPC levels.

Sufficiently high activity levels to cause the Working Level Alarm Unit to trip on either North or South channels causes several remedial changes. The three zones of building air conditioning automatically change over to 100% outside air; the East roll-up door elevates to prevent pressurization and ensuing spread of air activity to the remainder of the Pile complex; and the Pile console and Health Physics Office are notified by use of an audio-visual signaling device (Figure 9).

Whenever working level alarms are experienced on both channels simultaneously, the Main Control and Building Evacuation Control Units are electrically unlocked. At this level, the main control unit remains dormant until a manual evacuation alarm is ordered by controls in the Health Physics Office or Pile console or if one of the log count ratemeter upper level trips, operates and causes an automatic building evacuation alarm. The building evacuation alarm consists of six 18 by 24 inch signs that flash an "Evacuate Air Activity" message (only when illuminated) in unison with horns emitting 100 db at 10 ft. (Figure 10). The signs and horns alarm together at a three-quarter second "on" and half second "off" basis for the first minute of a two minute cycle. The horns are muted during the remainder of the cycle and the entire sequence is repeated as long as the upper level trip remains in effect after an attempt at automatic reset.

To add a higher measure of reliability during power failures, the South unit, Working Level Alarm Unit, and portions of the Main Control Unit are DC or inverter supplies. The skeletal system then is capable of notifying the Pile console and Health Physics Office of working level alarms.

An air conditioning shutdown and roll-up door lock out feature was also incorporated within the system to contain migration of gross contamination if deemed necessary. Close control of this feature is assured by use of a key switch with limited key issue.

DISCUSSION

THAXTER: Because of the rather notorious inefficiency of air filters for picking up volatiles, such as some halogenated compounds and rare gases, is it possible that you could have an MPC in those situations?

DISTENFELD: Yes, it is possible.

SCHWENDIMAN: I was wondering how many minutes of sampling is required before 1 MPC is detected?

DISTENFELD: Positive indications of one MPC would probably require at least a half hour of sampling. The tape moves at one inch per hour and the rate of air flow is in the neighborhood of ten cubic feet per minute. The background count rate is the difficulty, as far as determining MPC.

PATTON: Have you had any trouble with false alarms in the system?

DISTENFELD: No. The system is not a fail safe system. If something malfunctions it does not alarm. It takes both working-level alarms on both channels for evacuation to be available. If both are not provided, there is no signal. The system is reliability tested on a daily basis, and an electrical lockout is provided for external alarms. External alarm testing is kept to an absolute minimum as we try to have people think the alarm they hear is not a test.

COSTELLO: Captain Conway, from the Army Chemical Center, has asked me to have some of you gather in the corner here during any of the breaks to discuss inhalation hazards associated with fallout. He has come up here especially to pick up some information on this problem.

SUMMARY OF FISSION PRODUCT ADSORPTION WORK AT BNL

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ABSTRACT

A summary is presented and references given on work completed and under way at Brookhaven National Laboratory on the adsorption of fission products on graphite and activated charcoal. Results on the dynamic behavior of xenon and krypton on small charcoal adsorption beds is described. The behavior of xenon on graphite at high temperatures is discussed and some preliminary data on the adsorption of iodine on graphite is presented.

INTRODUCTION

Current interest in the field of high temperature gas-cooled reactors has focused attention on the difficulties of designing high temperature fuel elements. Leakage of fission products into a reactor coolant stream is one problem reactor designers must face since it may not be economical at present to design fuel elements for use at high temperatures which will completely contain all the fission products produced. Since the presence of large quantities of fission products in a reactor coolant system will considerably complicate the maintenance of the reactor and thus add to the cost of the power produced, it becomes necessary to develop information that reactor designers can use to develop the most efficient and economic fission product clean-up systems. Such information would also be useful in design of fission gas recovery systems in fuel element processing plants. Also important to high temperature reactor technology is information on the chemical and physical behavior of the fission products in the various high temperature materials under consideration.

IMPORTANT FISSION PRODUCTS

Although there are some 300 fission products, they are not all equally important from the point of view of contamination of a coolant loop of a gas-cooled reactor or release to the atmosphere. On the basis of yield, half-life, method of disintegration, probable release rate, and chemical and physical properties, Lane et al. (1) considers the chains' mass numbers 84, 78-95, 129 and 131-143 to be important in regard to the problem associated with fission product release from ceramic fuels. The decay scheme for these fission product chains is given by J. O. Blomeke. (2)

From the point of view of deposition the relatively nonvolatile and long-lived fission products such as strontium-90, barium-140, and cesium-137 are important members for which suitable techniques must be developed to prevent their deposition or methods must be developed to facilitate their removal.

Iodine is also an important fission product which is relatively volatile and a serious physiological hazard since it is rapidly adsorbed into the body where it concentrates in the thyroid gland. The relative ease with which iodine can be removed from gas streams has been established by several investigators. (3, 4) It has been determined by F. Hudswell et al. (5) that iodine will adsorb on a variety of materials. Thus, even though iodine is a relatively volatile material, it may be retained to a large extent on such a material as graphite. An important question is, if fission product iodine is released from a fuel element in a reactor core, what portion will remain in the core and how much will find its way into the coolant stream to be removed in a trapping system?

The noble gases, xenon and krypton, are inert chemically and are relatively volatile; therefore, one would expect little or no retention on reactor materials at high temperatures, but because of the high neutron adsorption cross section of xenon-135 one must have accurate information on its adsorption behavior on reactor graphites.

Although the noble gases will not contribute greatly to the maintenance problems of a gas-cooled reactor system, they could present a problem due to possible inhalation by maintenance workers or if large amounts are released to the atmosphere. In order to avoid release of activity to the atmosphere, it will be necessary to collect or hold up these gases until they have decayed to safe levels of activity. A convenient way of accomplishing this is by means of an adsorption bed which can be either used to delay or permanently hold up the noble gas fission products.

A good discussion of the many problems associated with the release of fission products from reactor fuel in gas-cooled reactors is given by J. A. Lane et al. (1)

The object of this paper is to present a summary of the results of some recent work done on the dynamic adsorption behavior of xenon and krypton from helium on small charcoal adsorption beds and also some information obtained on the adsorption behavior of xenon and iodine on graphite.

XENON-KRYPTON ADSORPTION ON CHARCOAL

A series of experiments has been carried out at Brookhaven in which the dynamic adsorption of xenon and krypton from helium has been investigated on a small charcoal bed by measuring the shape and position of breakthrough curves as a function of flow rate, concentration, and bed temperature. The material studied, the column dimensions, the concentration ranges, the flow rates and bed temperatures investigated are listed in Table I. This work will eventually be

TABLE I

Adsorption of Xenon and Krypton on Charcoal from Helium at 1 Atm. Total Pressure

Material - Columbia Activated Carbon

HCC 12/28
CXC 6/8

Column Dimensions

Inside diameter	1.38 in.	
Length	10.5 in.	
Weight of Charcoal	HCC 12/28	141.25 g
	CXC 6/8	149.85 g

Xenon

Flow Rate Range	500 $\frac{cm^3}{min}$ to 3000 $\frac{cm^3}{min}$
Concentration by Volume	0.02% to 1%
Bed Temperature	25° to 100°C

Krypton

Flow Rate Range	1500 $\frac{cm^3}{min}$ to 5000 $\frac{cm^3}{min}$
Concentration by Volume	0.025% to 0.1%
Bed Temperature	25 to 100°C

Xenon - Krypton Mixtures

Flow Rate	2000 $\frac{cm^3}{min}$	
Concentration by Volume	Xe	.025% to 1%
	Kr	.025% to 0.1%
Bed Temperature	25°C	

reported as three separate reports. The first one by Kenney and Eshaya (6) describes the work on the adsorption of xenon from helium. The second and third by Eshaya et al., are yet to be published and describe the work done on the adsorption of krypton from helium and the adsorption of xenon and krypton from xenon-krypton-helium mixtures, respectively.

The object of these studies is to provide performance data useful for engineering design and operation of an activated charcoal adsorber suitable for the removal of radioactive contaminants from helium streams.

CHOICE OF VARIABLES INVESTIGATED

Because of the heat generation in an adsorption bed due to the decay heat of the radionuclides, it will be necessary to supply coolant to such an adsorption bed in a reactor application. Since water is an inexpensive and readily available coolant it will be good engineering practice to operate such a bed at, or near, ambient temperatures; thus, the temperature range studied was 25° to 100°C.

The actual concentration of radioactive contaminants from a reactor coolant stream will probably be in the parts per million range. Because of the experimental difficulties of working at such low concentrations, it was necessary to compromise and work at concentrations higher than might be expected in a reactor application. The actual concentration ranges studied are indicated in Table I.

LITERATURE REVIEW OF XENON-KRYPTON ADSORPTION

An extensive bibliography is given by Kenney and Eshaya (6) of the various investigators which have dealt with the mathematics of the adsorption wave in chromatography, dehumidification, and other similar processes. The equations of Glueckauf (7) and Jury (8) are of particular interest and involve the concept of the number of theoretical equilibrium stages. Cantelow (9) presents breakthrough curves for xenon on "Cliffchur" 4-10 mesh and "B C Adsorbate" 6-14 mesh. His data are compared with the equations of Glueckauf (7) and Jury. (8)

Amphlett and Greenfield (10) have measured the equilibrium adsorption of xenon and krypton on Sutcliffe Speakman 208C (-8+14 mesh) at 20° and 100°C in the pressure range of 0.1 to 10 mm Hg.

EXPERIMENTAL EQUIPMENT AND PROCEDURES

A schematic drawing of the experimental equipment, which was located in a conventional chemical laboratory hood, is shown in Figure 1. A metered amount of the fission product gas containing radioactive tracer was mixed with the helium

ADSORPTION OF FISSION PRODUCT GASES ON CHARCOAL BEDS

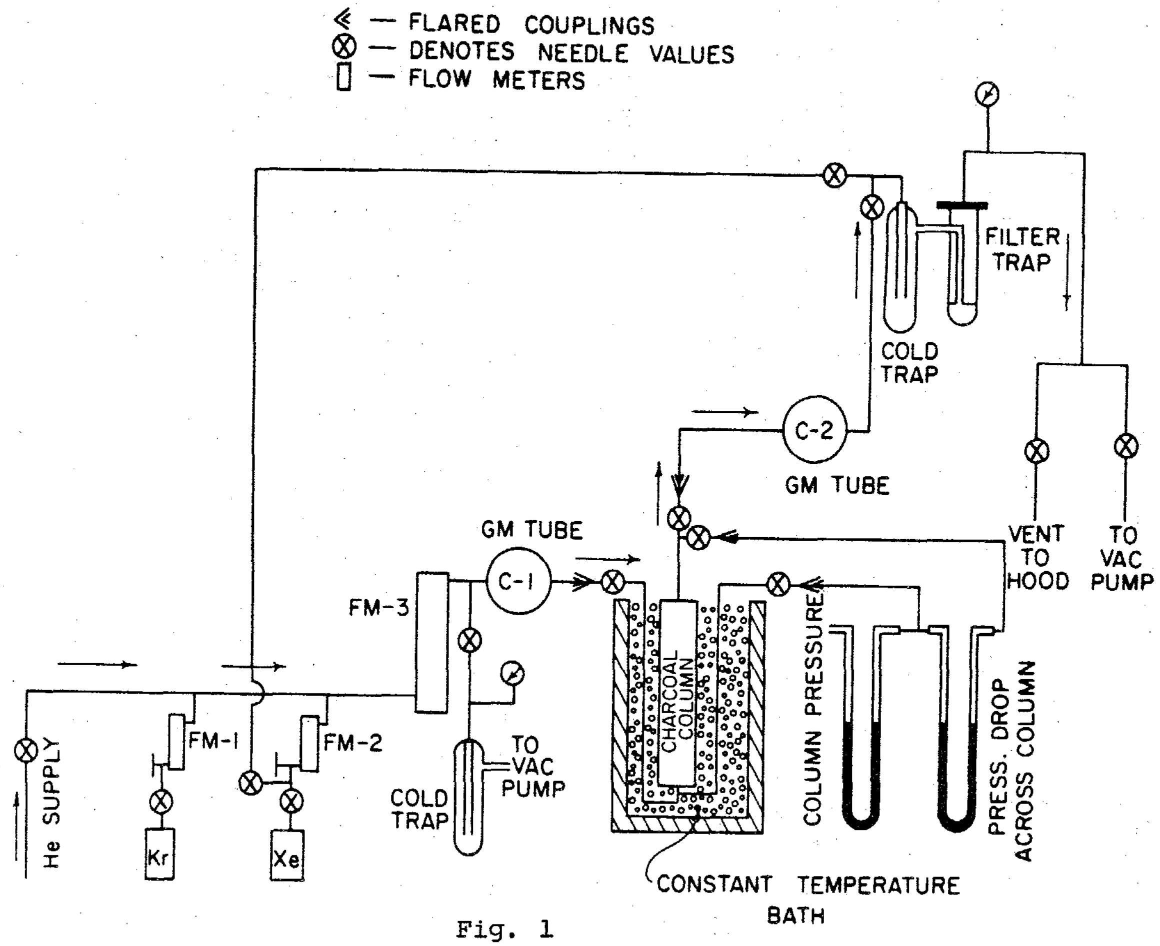


Fig. 1

stream, and the mixture was allowed to flow through a GM counter, the charcoal column, another counter tube, and a liquid nitrogen cold trap before exhausting into the hood ventilation system. The ratio of the net counting rate at the exit of the column to that at the inlet is the same as the ratio of the xenon concentrations in the outlet and inlet streams and is used directly in the determination of the breakthrough curves.

FISSION PRODUCT HANDLING SYSTEM

The design of the fission product handling system was based on the fact that both xenon and krypton will condense at the temperature of liquid nitrogen (-196°C). Each supply vessel was equipped with a pressure gauge, a needle valve, and a pressure regulator on its exit and was placed inside a Dewar flask within a lead shield. When the Dewar flask was filled with liquid nitrogen, the vessel could be evacuated, and the desired amounts of both radioactive and natural xenon (or krypton) could be condensed into it quite conveniently. When a run was to be made, the liquid nitrogen was removed from the Dewar, and the xenon or krypton evaporated and thus created the required pressure for flow. The gas was removed from the exit stream by a cold trap at the column exit and could be returned to the supply vessel through a bypass line by warming the cold trap and freezing the supply vessel in order to conserve radioactive gas.

ADSORPTION COLUMN

The column is 1.38 in. i.d. and 10.5 in. long and had an approximate capacity of 150 g of charcoal, which was supported on a stainless steel screen and perforated plate at the inlet to the column. The entire bed and the inlet line were submerged in a thermostated mineral oil bath that could be automatically maintained at any temperature up to 100°C. The column could be sealed off with needle valves and removed from the system.

COUNTERS

The Geiger tubes were thin-walled (20 mg/cm^2), all glass cylindrical counters (Nucleonic Corporation of America, Model 2A) with the organic coating removed to prevent adsorption of xenon or krypton. The tubes were sealed into a glass envelope connected to the flowing stream. The inlet counter was read on a conventional scaler, but the effluent was connected to a combination scaler-ratemeter (Nucleonic Corporation of America, RCR-3), the output of which was recorded on a strip chart recorder (Minneapolis-Honeywell, 0-10 mv) to provide a permanent and continuous record of the effluent concentration during a run.

MISCELLANEOUS EQUIPMENT

The flowmeters were Brooks rotameters that had been calibrated with the working fluid. The manometers and cold traps were glass, and the valves were Hoke bellows-sealed needle valves.

PROCEDURES

The operating procedure for an experiment consists of four phases: preparation, operation, shutdown, and desorption. Each phase is described in detail below.

a) Preparation

The entire system was evacuated and checked for leaks, the pressure in the gas supply autoclave was raised to operating level by warming to room temperature, the exit cold trap was filled with liquid nitrogen, and the counters checked for proper operation. The vacuum system was shut off, and helium admitted to the system. When the system pressure reached atmospheric, the exhaust valve was opened and the helium flow was set at the desired value as measured on flowmeter FM-3. The final step was to establish background counting rates for both counters. The chart recorder on the effluent counter was started during the background readings.

b) Operation

The run began when gas flow was started and adjusted to the desired flow. The scaler portion of the exit counter-ratemeter was then started and allowed to run continuously during the run to provide a record of the duration of the run and a periodic check of the ratemeter readings. The flowmeter readings, inlet and outlet counting rates, column pressure and temperature, and column pressure differential were monitored and recorded periodically. Care was taken to keep the exit cold trap properly refrigerated to prevent the escape of xenon.

The run was continued in this manner until the net counting rates at the inlet and outlet of the column were identical, at which point the system is shut down.

c) Shutdown

The initial step in the shutdown procedure is to stop the gas flow and the exit counter. The helium flow was stopped and the exhaust valve closed simultaneously. The gas supply vessel was frozen with liquid nitrogen. The adsorption column was isolated from the exit cold trap and the latter evacuated while still under refrigeration to remove the helium present. The cold trap was then isolated and warmed to room temperature. The gas trapped there was returned to the supply vessel by opening the valves in the bypass line and allowing the gas to be condensed into the cold supply vessel. The final step in the procedure was to seal off the vessel and warm it to room temperature again to measure the pressure of the xenon it contained. An independent estimate of the amount of gas adsorbed could be obtained by comparing this pressure to the pressure in the vessel at the start of the run.

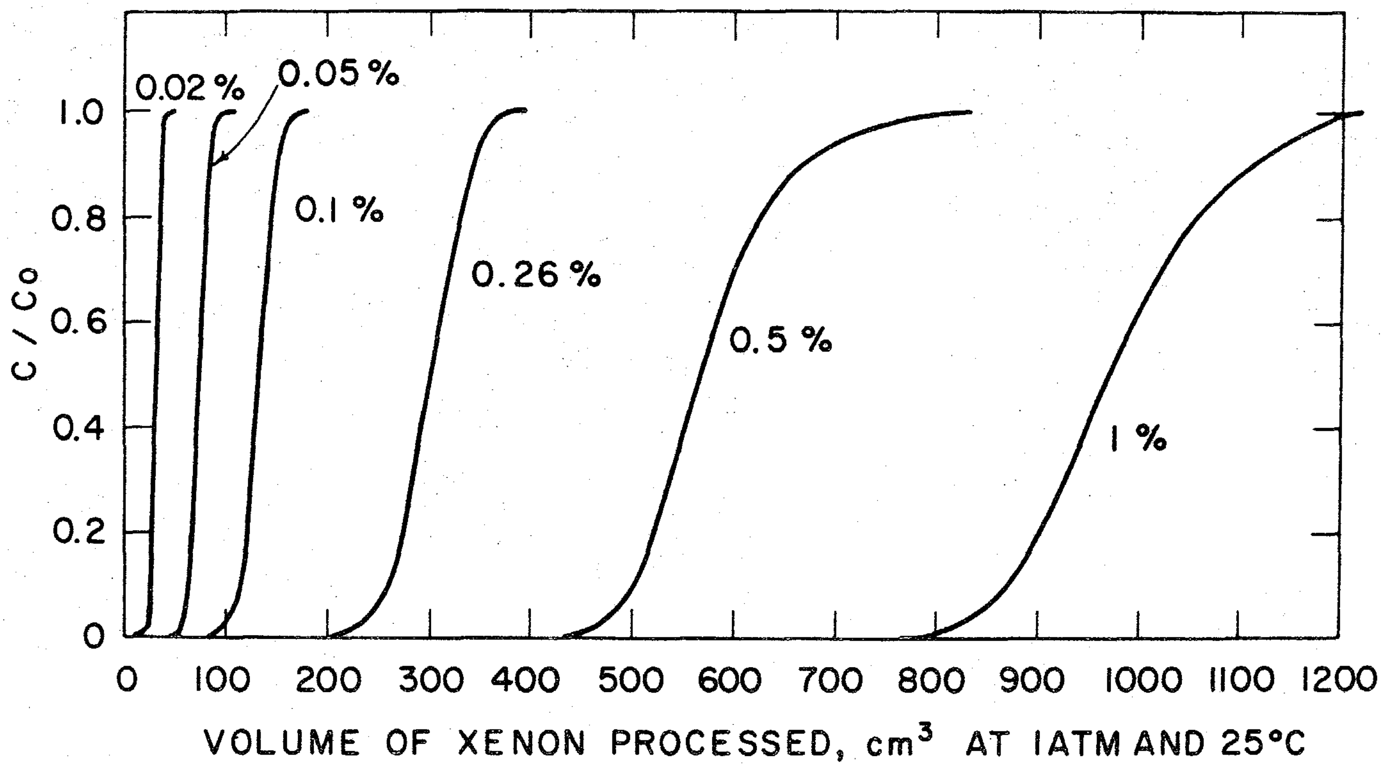
d) Desorption

After all the gas caught in the exit cold trap was returned to the supply vessel, the cold trap was refrigerated once more and the gas adsorbed on the charcoal was removed by heating the column to 100°C and evacuating through the cold trap. This procedure could be interrupted at any time and the gas removed was returned to the supply vessel to provide information on the rate of desorption. When no more gas was being removed from the column, it was cooled to room temperature with continuous evacuation in preparation for another run.

RESULTS AND DISCUSSION

In Figures 2 and 3 are shown some typical breakthrough curves obtained for the adsorption of xenon and krypton from helium at a flow rate of 2000 cm^3/min and a bed temperature of 25°C. The data are plotted as C/C_o vs. the volume of adsorbate, at 1 atm and 25°C, put through the bed. C is the outlet concentration at any time and C_o is the inlet concentration to the bed. On the plots are shown data for a range of concentrations for xenon (0.02 to 1% by volume) and krypton (0.025 to 0.1% by volume). The breakthrough curves show the symmetrical "S" shaped curve which is typical of a breakthrough curve. It can be shown that, because of the symmetry of these curves about the point $C/C_o = 0.5$, the quantity of adsorbate which represents an equilibrium adsorption is the volume processed at $C/C_o = 0.5$ and is designated as $\overline{V}$. It should be noted that the slope of the breakthrough curves at $C/C_o = 0.5$, in both the case of xenon and krypton, is decreasing with concentration and the curves deviate more and more from the ideal square wave as the concentration increases. A complete presentation of all the data for the system xenon-helium and discussion of the shapes of these breakthrough curves as a function of all the variables is given by Kenney and Eshaya (6) in terms of the rate of adsorption in the bed and rate of introduction of the adsorbate to the bed, from which it is concluded the rate-controlling mechanism for the adsorption process is diffusion into the charcoal particles.

In Figure 4 is shown a typical breakthrough curve for a mixture of equal proportions of xenon and krypton in helium (0.025% by volume). The separate C/C_o values for xenon and krypton are plotted against the individual volumes of xenon and krypton put through the bed. It should be noted that a very distinct separation is obtained between xenon and krypton. From these curves the $\overline{V}$ values are obtained at $C/C_o = 0.5$.



XENON ON CHARCOAL

BREAK-THROUGH CURVES AT CONSTANT FLOWRATE

HELIUM FLOW 2000 cm^3 / MIN

149.85 gms COLUMBIA CXC 6/8 MESH CHARCOAL

Fig. 2

KRYPTON ON CHARCOAL
BREAK-THROUGH CURVES AT CONSTANT FLOWRATE
HELIUM FLOW 2000 cm^3 / MIN
141.25 gms COLUMBIA HCC 12 / 28 MESH CHARCOAL

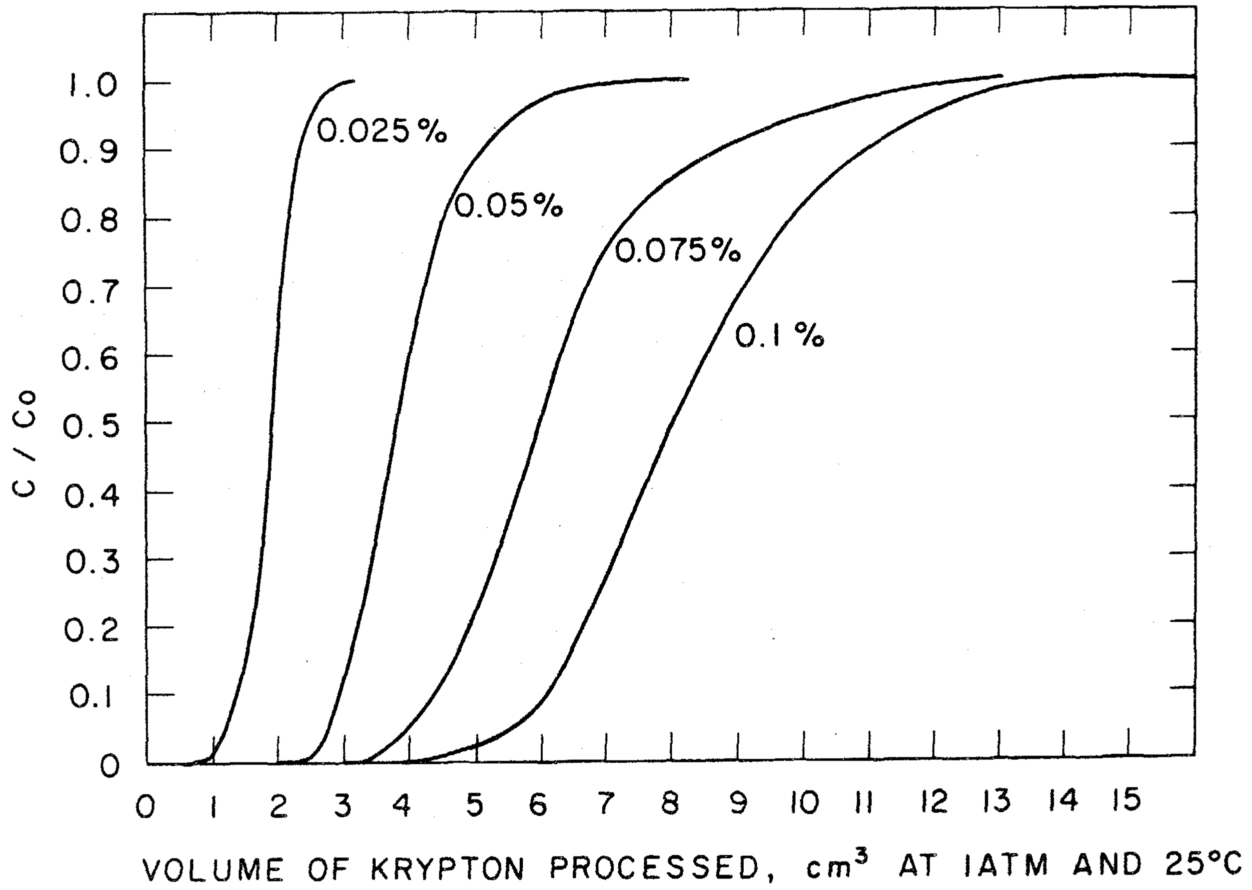


Fig. 3

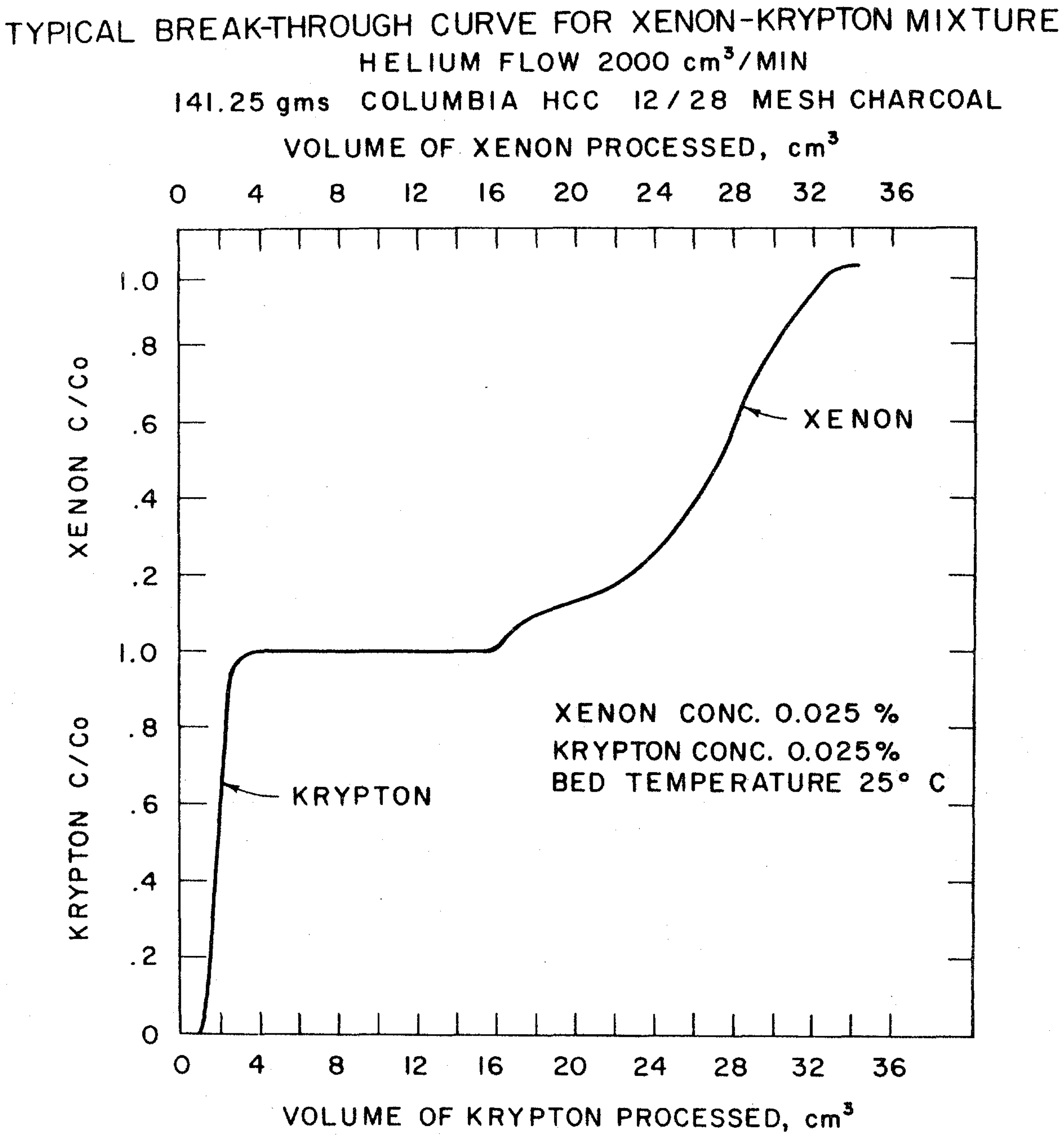

TYPICAL BREAK-THROUGH CURVE FOR XENON-KRYPTON MIXTURE
HELIUM FLOW 2000 cm³/MIN
141.25 gms COLUMBIA HCC 12/28 MESH CHARCOAL
VOLUME OF XENON PROCESSED, cm³
0
4
8
12
16
20
24
28
32
36
XENON C/Co
1.0
.8
.6
.4
.2
KRYPTON C/Co
1.0
.8
.6
.4
.2
0
XENON
KRYPTON
XENON CONC. 0.025 %
KRYPTON CONC. 0.025%
BED TEMPERATURE 25° C
0
4
8
12
16
20
24
28
32
36
VOLUME OF KRYPTON PROCESSED, cm³

Fig. 4

In Figure 5 are plotted all the $\overline{V}$ values, in cm^3/g of charcoal at 25°C and 1 atm, obtained in all the single component xenon and krypton runs, as well as the xenon-krypton mixtures, against the partial pressure in mm mercury for each adsorbate. The $\overline{V}$ values obtained in the single component runs show good agreement, within the limit of experimental error, with the $\overline{V}$ values obtained in the case of the xenon-krypton mixtures, from which it is concluded that there was no detectible interference between the adsorption of xenon and krypton.

The isotherms obtained are the Freundlich type, as are data of Amphlett and Greenfield (10) whose data for 20°C is shown as a dotted line in Figure 5 for both xenon and krypton. The agreement between this data (6) obtained in a dynamic system and the data of Amphlett and Greenfield (10) obtained by measuring the static equilibrium adsorption is very close.

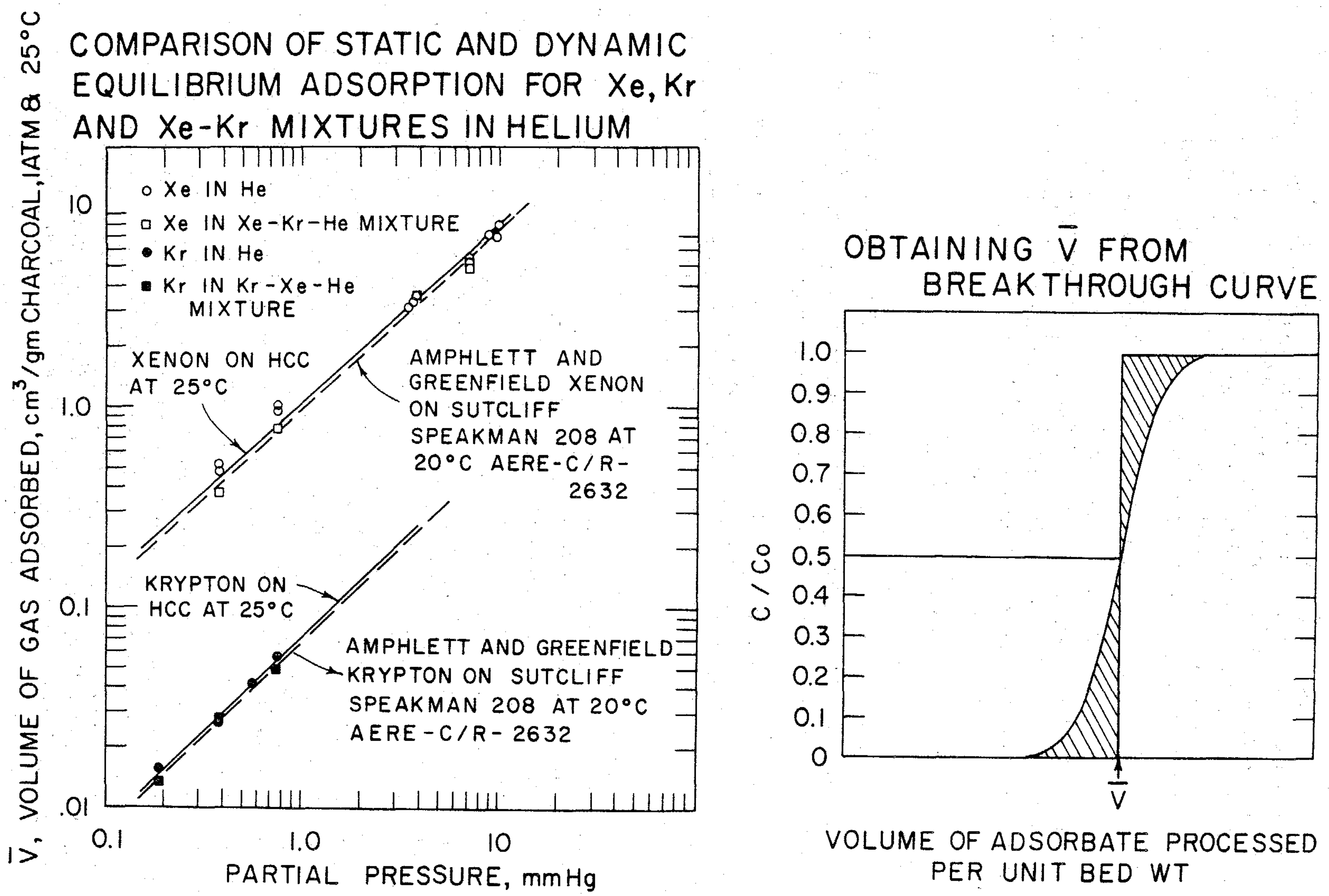
XENON ADSORPTION ON GRAPHITE

Another program recently completed at Brookhaven by Salzano and Eshaya (11) was a study of adsorptive behavior of xenon in graphite at high temperatures (above 500°C).

Since it is extremely difficult to make direct measurements of the adsorption of xenon on graphite at high temperatures, an indirect approach was used whereby the total xenon taken up by the graphite at high temperatures was measured.

Samples of R-4 graphite with a density of 1.92 g/cm^3, supplied by the Graphite Specialties Company, Niagara Falls, New York, were machined into 1/2-in.-diam spheres. These spheres were outgassed at 1100°C and then equilibrated with radioactive xenon-133 tracer at various pressures, at temperatures of 750° and 1000°C. At the end of a 60-min equilibration period the hot graphite was covered over with cold mercury and the mercury frozen with liquid nitrogen. The xenon sorbed into the graphite was thus trapped in the solid mercury ingot. After pumping the excess xenon from the system, partly melting the ingot to free it from the walls of the equilibration cell and flushing with helium to remove all traces of active gas adsorbed on these walls, the mercury was melted off and the sorbed xenon desorbed into a measured amount of inactive carrier xenon. Aliquots were then taken of this desorbed gas plus carrier and counted in a 4 π proportional counter. By comparing the activity in a sample of the original equilibration gas and the desorbed gas plus carrier, the original quantities of xenon sorbed were determined.

The results of these experiments carried out at 750° and 1000°C in the pressure range of 60 to 700 mm mercury indicate clearly that the major sorption mechanism of xenon in graphite at high temperatures is that of pore entrainment. Above 500°C the quantity of xenon held by surface adsorption is probably less than 1% that held in the voids. The large sorptions of xenon on graphite measured by Eshaya (12) can be explained by the proposed pore entrainment mechanism since



COMPARISON OF STATIC AND DYNAMIC EQUILIBRIUM ADSORPTION FOR Xe, Kr AND Xe-Kr MIXTURES IN HELIUM

OBTAINING $\bar{V}$ FROM BREAKTHROUGH CURVE

Fig. 5

a comparison of the calculated sorption shows good agreement with experimental results. These data are in agreement with the extrapolated results of M. C. Cannon et al., (13) who measured the adsorption of xenon on graphite at -79°, 0°, 20°, 30°, and 80°C.

IODINE ADSORPTION ON GRAPHITE

Presently under way at Brookhaven is a study of the adsorption of iodine on graphite at high temperatures. The program is in its early stages and only a limited amount of experimental data is available. The main effort to date has been the development of a suitable and sensitive enough technique. One of the accomplishments to date is a series of measurements of the adsorption of iodine on type R-4 graphite at 1073° and 1273°K and 23 mm mercury pressure. These measurements were made by equilibration of graphite samples with iodine vapor and subsequent analysis of the graphite by a wet chemical analytical technique. It was determined that at 1000° and 800°C the adsorption of iodine on type R-4 graphite is in the order of 150 and 270 ug iodine respectively, per gram of graphite.

A more sensitive and efficient technique has been developed which makes use of a sodium iodide scintillation probe and iodine-131 tracer for the direct measurement of the iodine concentration on the graphite. With this technique a series of six isobars have been obtained in the pressure range of 1 to 40 mm mercury and in the temperature range of 475° to 725°K.

The apparatus in which the experiments were carried out is shown in Figure 6 and consists of a sealed quartz vessel in which a graphite sample (1/2 in. diam by 1 1/2 in. long) is suspended on a quartz thermocouple well which is 1/8 in. diam by 1/2 in. long. The graphite and iodine vapor are initially separated by a break-seal. The sample is heated with a semi-circular induction coil, while the vapor pressure of the iodine is controlled by immersing the end of the quartz vessel in a thermostated oil bath. The upper end of the quartz vessel, which is above the oil bath, is heated by a hot air flow from a high temperature blower, the temperature of which is maintained a few degrees above the bath temperature to prevent condensation of the iodine.

The concentration of the iodine on the graphite is measured by counting the full gamma spectrum of iodine-131 in the graphite with a sodium iodide scintillation crystal, sighted on the graphite through a 1/4 in. diam by 2 in. long hole in the 2 in. lead shielding which surrounds the crystal. The face of the crystal is 4 in. from the center line of the sample. An absolute calibration of the system is made by counting an aqueous solution of iodine-131 of known specific activity in a cellulose nitrate tube in which the length of the liquid column and the inside diameter of the tube are the same as the graphite sample.

APPARATUS FOR MEASURING I_2 ABSORPTION ON GRAPHITE

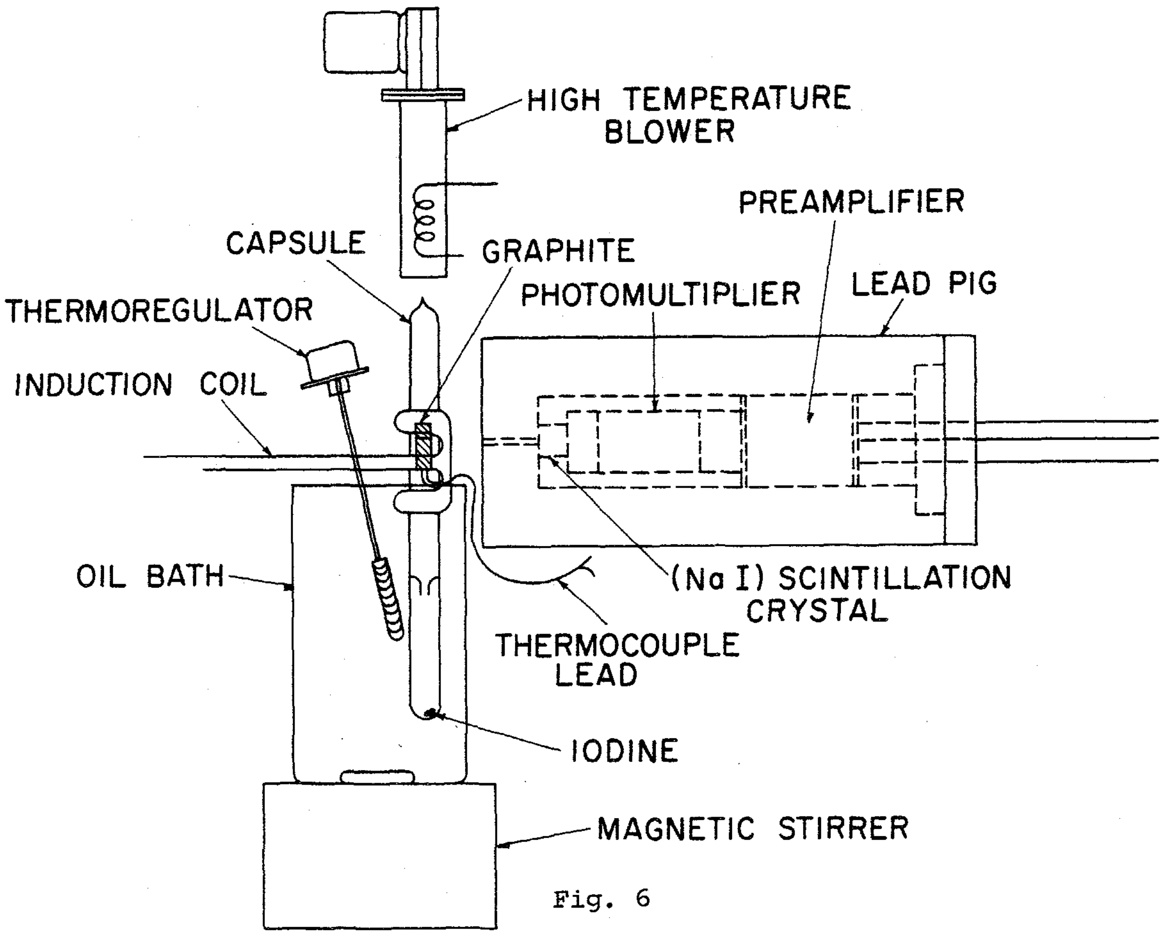


Fig. 6

The graphite sample was first outgassed at 1073°K and the top of the quartz capsule sealed. 0.2 g carrier iodine-127 containing 40 $\underline{\mathrm{uc}}$ iodine-131 was distilled under vacuum into the bottom of the quartz vessel which was then sealed off. After breaking the seal between the iodine and the graphite, the first equilibration was carried out with the graphite heated to 725°K, where the iodine could be detected at the lowest experimental pressure, 1.03 mm mercury. The temperature of the graphite was subsequently reduced in approximately 50°-intervals and the iodine concentration measured. Equilibrium was assumed to be the point where no detectable increase in concentration occurred over a 60-min interval. When an isobar had been completed the graphite was heated to 925°K and the iodine vapor pressure reduced essentially to zero by cooling the iodine with liquid nitrogen. The equilibrium concentrations were always approached from a lower concentration.

Six isobars were measured in the temperature range 475° to 725°K, in the pressure range of 1 to 40 mm mercury by approximately doubling the pressure between each isobar. All of the data are presented in Figure 7 as micrograms of iodine per gram of graphite vs. the absolute temperature.

An extrapolation of the isotherms interpolated from this data could be used to estimate iodine concentrations on type R-4 graphite at lower partial pressures of iodine.

While carrying out the experiments the following observations were made: 1) The time to reach equilibrium was found to be long, in the order of 5 to 6 hr at the lowest temperatures, and seemed to be strongly temperature dependent, i.e., equilibrium occurring more slowly at low temperatures. In addition, the rate of desorption was seen to be strongly temperature dependent, i.e., the desorption occurring rapidly at high temperatures. 2) A small fraction of the iodine put on the graphite could not be easily desorbed even at high temperatures; in fact, the effect seemed cumulative. This work will continue with the object of going to temperatures as high as 1000°C.

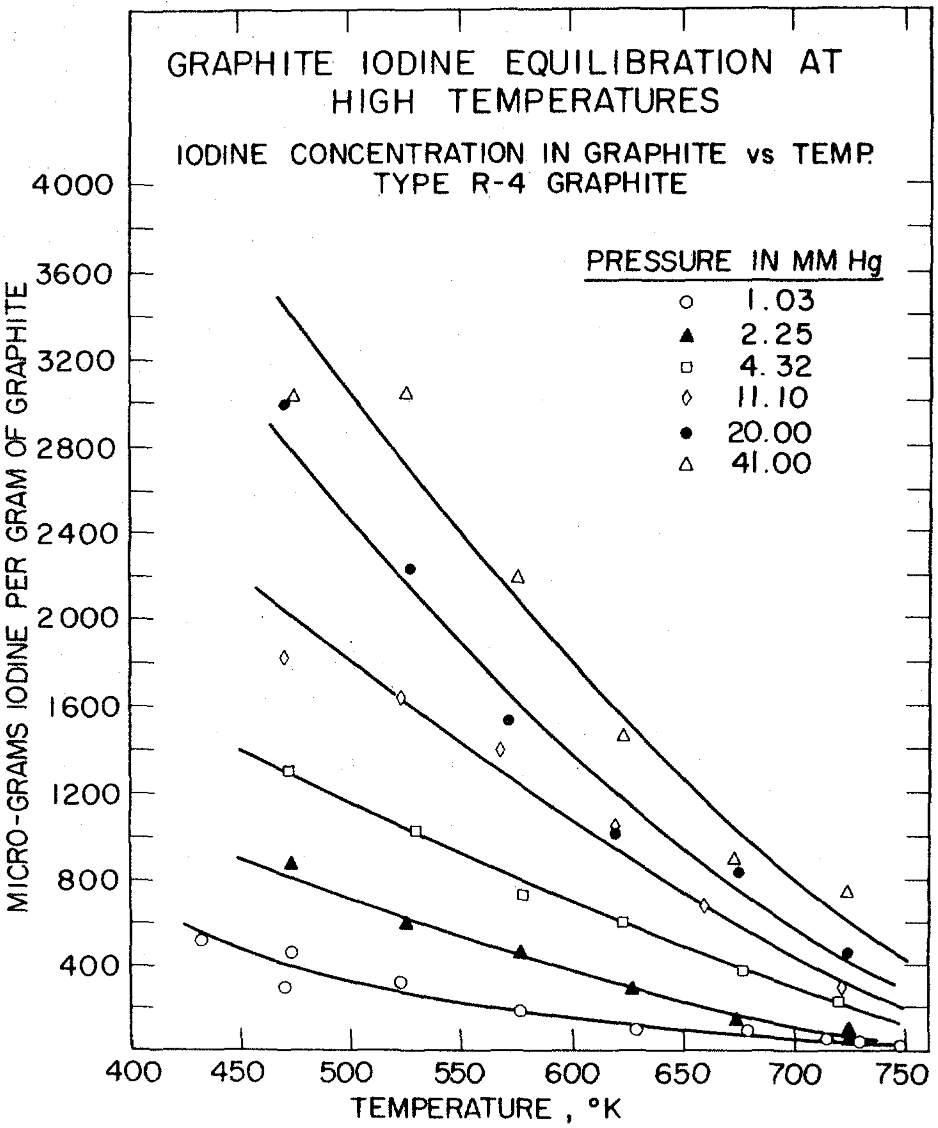

GRAPHITE IODINE EQUILIBRATION AT HIGH TEMPERATURES
IODINE CONCENTRATION IN GRAPHITE vs TEMP.
TYPE R-4 GRAPHITE
PRESSURE IN MM Hg
○ 1.03
▲ 2.25
□ 4.32
◊ 11.10
● 20.00
△ 41.00
MICRO-GRAMS IODINE PER GRAM OF GRAPHITE
4000
3600
3200
2800
2400
2000
1600
1200
800
400
400
450
500
550
600
650
700
750
TEMPERATURE , °K

Fig. 7

REFERENCES

1. Lane, J. A., et al., ORNL-2851, October 1959.
2. Blomeke, J. O., ORNL-1783, September 1959.
3. Adams, R. E. and Browning, E. W., Jr. ORNL-2872, March 1960.
4. Arthur D. Little, Inc., HW-65587, March 1960.
5. Hudswell, F., et al., AERE-R-3183, January, 1960.
6. Kenney, W. F. and Eshaya, A. M., BNL-689.
7. Glueckauf, E., Trans. Faraday Soc. 51, 34 (1955).
8. Jury, S. H. and Licht, W., Chem. Eng. Prog. 48, 103 (1952).
9. Cantelow, H. P., UCRL-8755, May 1959.
10. Amphlett, C. B. and Greenfield, B. F., AERE C/R 2632 (1958).
11. Salzano, F.J. and Eshaya, A. M., BNL-633, September 1960.
12. Eshaya, A. M., Adsorptions of Xenon and Iodine on Graphite Surfaces, LMFR Monthly Progress Reports, February 1958 through May-June 1959.
13. Cannon, M. C., Ward, W. T., Watson, G. M. and Grimes, W. R., ORNL-2955, December 1960.

DISCUSSION

SILVERMAN: I would like to ask if you have done any work on trying to make equivalents of activated graphite?

SALZANO: Similar to charcoal?

SILVERMAN: Yes.

SALZANO: No.

FIRST: Why do you use the midpoint for your V-bar value? Are you not more interested in the point at which it begins to come through? What is the value of the 50 percent?

SALZANO: The V-bar value is the value that you would measure in an equilibrium adsorption. The value you get at C/Co = 0.5 should be comparable to the value obtained in a non flow system at equilibrium.

FIRST: Why not use the point at which it rises from zero?

SALZANO: We are interested in making a comparison between a static absorption and dynamic adsorption and at the C/Co = 0.5 position and they should be equivalent.

AIR SAMPLING FILTER PAPER RETENTION STUDIES USING SOLID PARTICLES

Samuel Posner
Health and Safety Laboratory
U. S. Atomic Energy Commission

In 1953 the A. D. Little Company published a report entitled "Properties of Various Filtering Media for Atmospheric Dust Sampling" which discussed the performance of most of the air sampling filter papers then in use. Using di-octyl phthalate (DOP) mist as the test aerosol, the filters were tested for penetration as a function of velocity through the filters. Measurements of resistance to air flow as a function of air velocity were also included in the report. For many years this report has been our most comprehensive source of information regarding the characteristics of air sample filter media.

At approximately the same time Adley, Scott, and Gill were performing somewhat similar work at Hanford, Washington. Their results were reported in a paper entitled "A Study of Efficiencies and Pressure Drop Characteristics of Air Filter Media." In their study of filter efficiencies these investigators used U_3O_8 fume having a mean particle size by count of 0.07 micron. Their study was limited to fewer papers than those covered in the A. D. Little report, but, interestingly, for similar types of papers the two studies in some instances showed different results. This is not surprising in view of the fact that dissimilar aerosols were used in measuring efficiency, the one being an oil droplet and the other a solid fume particle. Other investigators have measured filter penetration using still different methods. However, for several reasons, the DOP test became widely regarded as a standard assessment technique despite the fact that filter sampling in practice is usually used for the collection of solid particles. It should be noted that in Great Britain solid methylene blue particles constitute the standard test aerosol for filter evaluation.

There are several reasons for questioning the validity of DOP aerosol penetration as a standard evaluation. In the 1953 A. D. Little report, previously mentioned, it was stated that with regard to membrane filters collection appears to be almost entirely at the surface. It is perhaps for this reason that they fill up rapidly in an oil smoke like DOP and so may not show up to the best advantage in this test. The report went on to say that all fiber materials fatigue when exposed to DOP. After running the DOP for several minutes, penetration values increase. One possible explanation is that the electrostatic effects in the filter body are neutralized due to the accumulation of liquid. It is known that filters depending upon electrostatic effects fail quickly when used on oil smokes, so there is some basis for suggested explanation.

For these and other reasons it appeared that to better understand and document the collection characteristics of filter media as they are used for air sampling, measurements should be made using a solid particle of some substance measurable to very small concentrations. Uranine, a dye that was found to be measurable to about 5×10^{-11} grams in a fluorimeter, was selected. HASL has to date performed penetration and resistance measurements on approximately three dozen air sampling filters. This paper reports the preliminary results of those tests and compares the efficiencies measured by uranine dye with the efficiencies reported by A. D. Little in 1953 based on the DOP aerosol. The HASL program is continuing and will include the remeasurement of the same types of filters with DOP apparatus so that changes in the composition and manufacturing of filters that have occurred since 1953 will not be reflected in any differences that show up under the two types of testing.

The aerosol generating system is illustrated in Fig. 1. It is patterned after a system developed by Professor K. Whitby, of the University of Minnesota. In his original design he used a Collison generator and impactor to skim off the larger particle sizes. He followed these with a deionizer and mixing tank. The HASL modification of this system included the substitution of a Dautrebande $D_{30}1$ generator for the Collison generator and the inclusion of a dry ice trap followed by a reheater in the by-pass line. As a primary particle generator, the Dautrebande $D_{30}1$ generator has been described previously in the literature. The impactor described by Whitby consisted of #60 drill holes separated from an impactor plate by 8/1000 of an inch. The impactor plate has several large holes into which the impacted aerosol can be passed to the aging tank. It also has a drain so that the uranine-containing liquid can be drained off as the level in the impactor builds up. One HASL modification in the impactor was to substitute #80 drill holes for the #60 holes and to reduce the spacing between the holes and the backing plate to 5/1000 of an inch. This lowers the cut-off size of the particles. The aerosol which emerges from the impactor goes into one side of the T which is connected to the mixing tank.

On the other side of the system compressed air is first sent through to a filter, then through a dry ice trap, where the temperature is maintained at approximately -80°C. When the air has been thoroughly dehumidified, has left the dry ice trap and is refiltered, it is sent to a long tube upon which is wrapped a heating tape maintained at 100°C.

From this heater the air passes to the deionizer, on the far side of which is a 1/16" opening drilled in a brass plate. A needle is placed within the chamber in front of, and approximately 1/8" from, this opening. 3.5 KV rms is maintained between the needle and the brass plate. It is presumed that this alternating current completely ionizes the air with a charge of both signs. From this device the air is immediately discharged into the other side of the T previously cited, and is then passed into a 4" diameter by 4' long mixing tank. The dried, ionized air mixes with the aerosol, dries it, and neutralizes the charge which has been built up on the particles during the generation process.

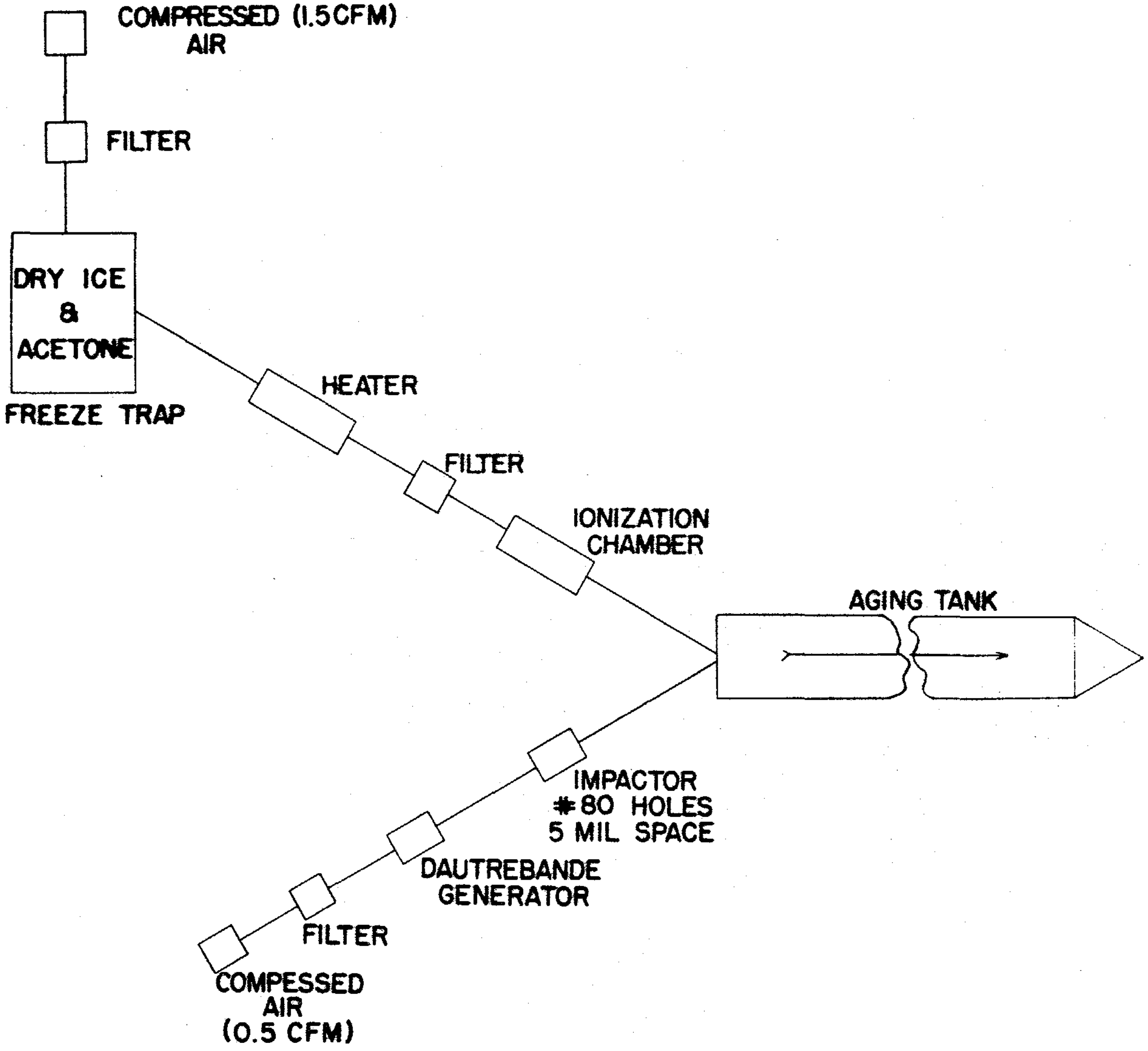

COMPRESSED (1.5CFM) AIR
FILTER
DRY ICE & ACETONE
FREEZE TRAP
HEATER
FILTER
IONIZATION CHAMBER
AGING TANK
IMPACTOR
#80 HOLES
5 MIL SPACE
DAUTREBANDE GENERATOR
FILTER
COMPESSED AIR (0.5 CFM)

FIG. 1

From the end of the mixing tank aerosol samples are collected on electron microscope grids mounted in a special electrostatic precipitator. It appears that the particles are dry and discrete. Approximately 1% of the particles appear to be agglomerated.

Aerosols of two sizes were used. One size was prepared using 0.1% uranine solution in the generator and #80 holes in the impactor. The particles had a count median of 0.008 micron and a mass median of .025 micron, with a standard deviation of 1.9. The second size was prepared using 1% uranine and #60 holes in the impactor. These particles had a count median of 0.07 micron and a mass median of 0.27 micron, with a standard deviation of 1.7. This size was generated to simulate the DOP mist (0.3 u) normally used for testing. Each time a new solution was prepared, electrostatic precipitator samples were studied and counted to confirm the consistency of the aerosol. Particle size distribution curves are shown in Fig. 2 and Fig. 3.

A Leiman type C-3 vacuum pump was used to draw air from the aerosol generator and through the filter under test. The test filter was followed by a back-up filter (1106B fiber glass paper) selected on the basis of high retention quality (see Fig. 4). As a check on this backup filter, in several tests a 1" diameter Millipore VF filter was inserted downstream from the 1106B paper. In no case was uranine detected on the Millipore.

The pressure drop across the various filters was determined using Dwyer Magnehelic pressure gauges. In the filter testing apparatus the pressure drop across the filter being tested was measured with the Dwyer Magnehelic pressure gauge, and flow rate was determined from curves of flow rates vs. pressure drop.

Most filter media were checked at face velocities of 5, 10, 25, 50, 100, 200, and 300 ft/min. The IPC paper was checked at 100, 200, 300, 400, and 500 ft/min. All papers were tested with both the 0.025 micron particles and the 0.27 particles. Some of the papers (see Tables I, II, and III) were compared with the A. D. Little DOP tests. For better visualization of the comparison of results of the DOP and the uranine tests, the penetration data for Whatman #41 are shown graphically in Fig. 9. For some of the newer papers there are no data available on the DOP penetration. HASL is now in the process of assembling a DOP generator in order to further study and test all of the papers.

Table IV shows pressure drop characteristics of some of the filter media tested at HASL.

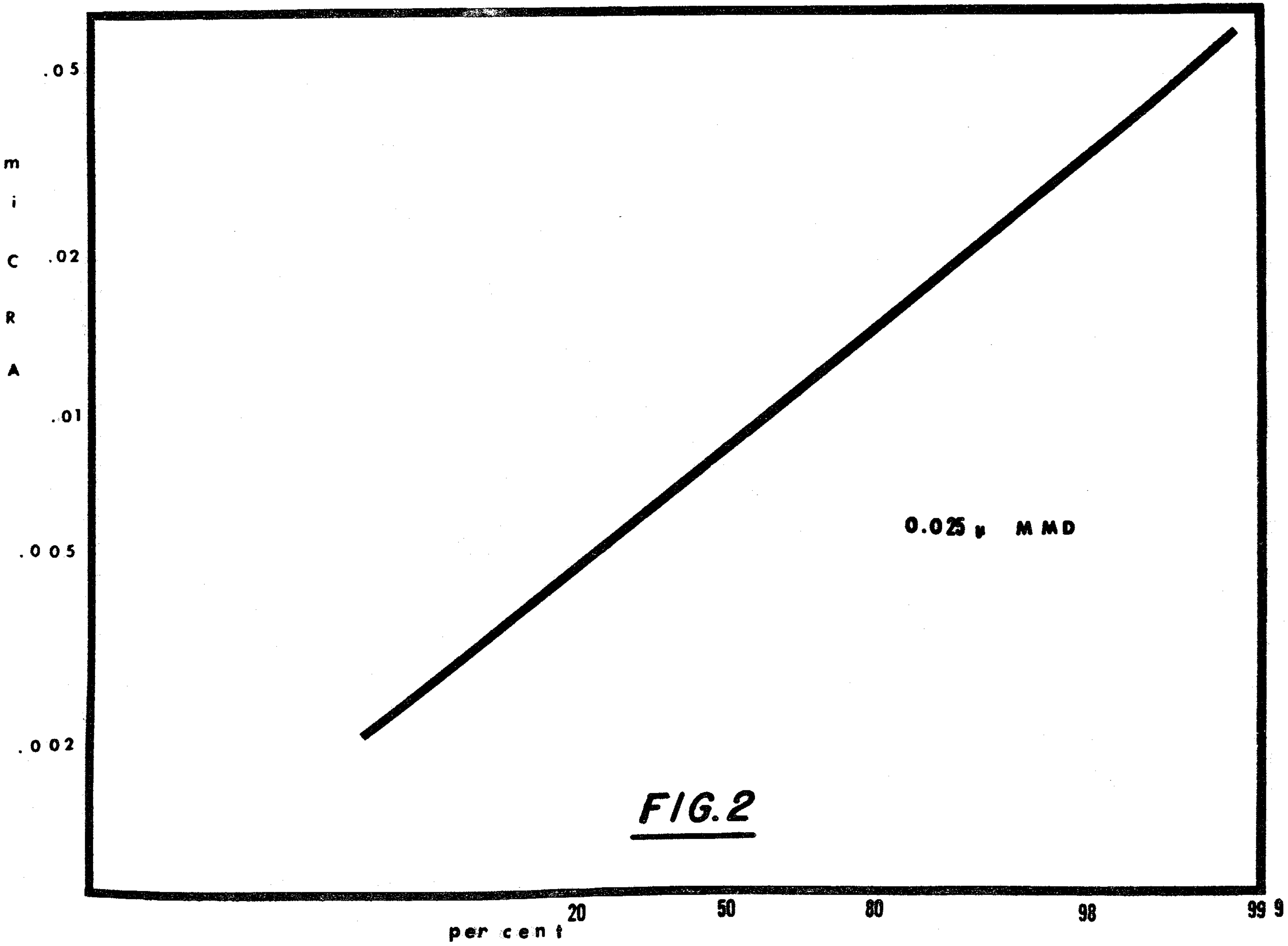


FIG.2

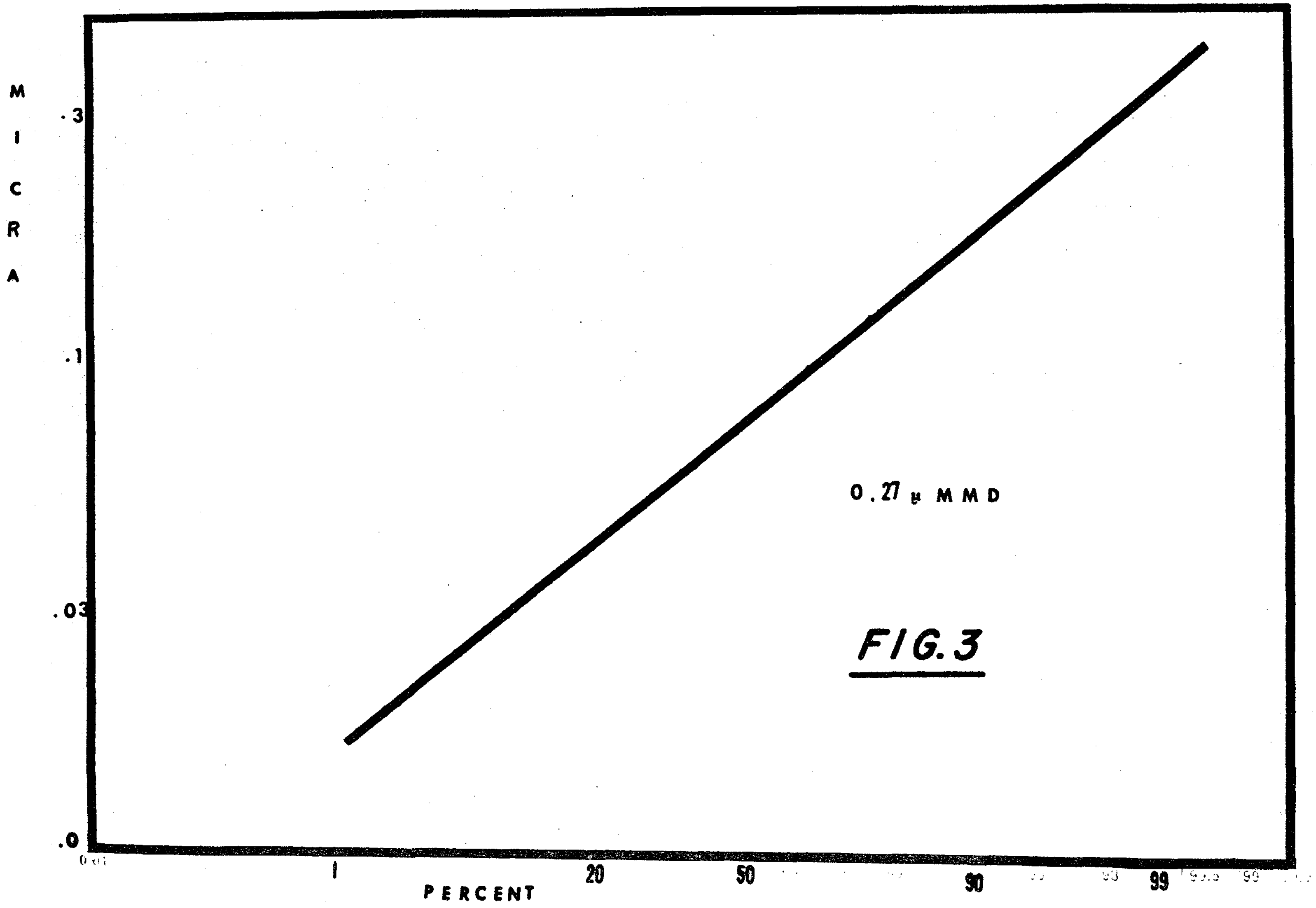
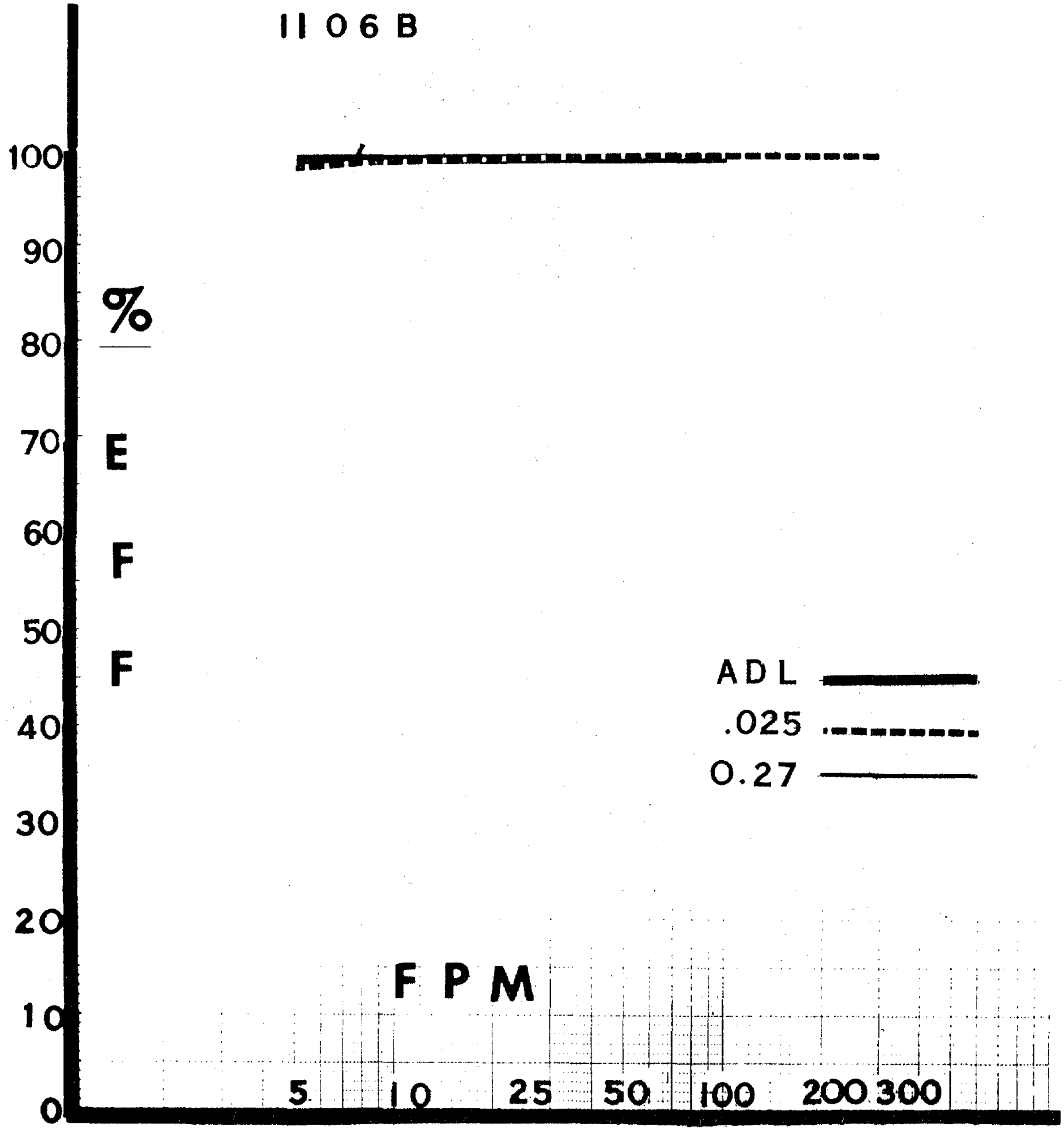


FIG.3



11 06 B
100
90
80
70
60
50
40
30
20
10
0
% EFF
ADL
.025
0.27
F P M
5
10
25
50
100
200
300

FIG. 4

TABLE I

EFFECT OF FLOW RATE ON URANINE AND DOP GENERATOR

	HV 70 9 mil			Whatman 41		
		HASL			HASL	
FPM	ADL	.025 μ	0.27 μ	ADL	.025 μ	0.27 μ
5	95.0	99.1	98.7	11.0	84.0	68.8
20	96.5	-	-	23.0	-	-
25	-	91.73	97.2	-	65.0	80.1
28	98.0	-	-	25.0	-	-
50	98.3	95.2	98.2	33.0	82.0	87.2
100	99.8	97.6	97.2	66.0	75.0	91.0
200	-	98.9	96.5	85.0	82.0	94.5
300	-	98.6	99.7	-	88.0	88.8
Pressure drop at 100 FPM inches water	21.8	17.0		8.1	12.0	

FIG. 5

TABLE II

EFFECT OF FLOW RATE ON URANINE AND DOP GENERATOR

	1106B			S and S 604		
		HASL			HASL	
FPM	ADL	.025 μ	0.27 μ	ADL	.025 μ	0.27 μ
5	99.9+	99.0	99.6	7.0	53.9	71.0
20	99.9+	-	-	15.0	-	-
25	-	99.9+	99.9+	-	70.3	84.4
28	99.9+	-	-	21.0	-	-
50	99.9+	99.9+	99.9+	33.0	86.1	95.1
100	99.9+	99.9+	99.9+	61.0	91.8	96.3
200	-	99.9+	99.9+	93.0	91.0	97.6
300	-	99.9+	99.9+	-	92.9	96.7
Pressure Drop at 100 FPM inches water	19.8	10.8		8.5	17.0	

FIG. 6

TABLE III

EFFECT OF FLOW RATE ON URANINE AND DOP GENERATOR

	Millipore AA			Whatman #4		
		HASL			HASL	
FPM	ADL	.025 μ	0.27 μ	ADL	.025 μ	0.27 μ
5	99.9+	99.9+	98.5	16.0	32.1	92.7
20	99.9+	-	-	23.0	-	-
25	-	99.9+	99.9+	-	56.6	90.4
28	99.9	-	-	27.0	-	-
50	99.9+	99.9+	99.9+	38.0	62.0	93.4
100	99.9+	99.9+	99.9+	75.0	74.6	97.9
200	-	99.9+	99.5	-	86.8	95.2
300	-	-	-	-	88.3	94.7
Pressure drop at 100 FPM inches water	39.0	67.0		11.5	8.4	

FIG. 7

TABLE IV

HASL RESULTS
PRESSURE DROP OF INCHES WATER

FPM	HV-70 9 Mil	Whatman 41	Millipore AA	1106B Fibroglas
5	0.7	0.4	2.6	0.68
25	3.9	2.5	15.0	3.5
50	8.0	5.5	31.0	10.2
100	17.0	12.0	67.0	11.2
200	36.0	26.5	114.5	32.0
300	56.0	42.0	225.0	48.0

FIG. 8

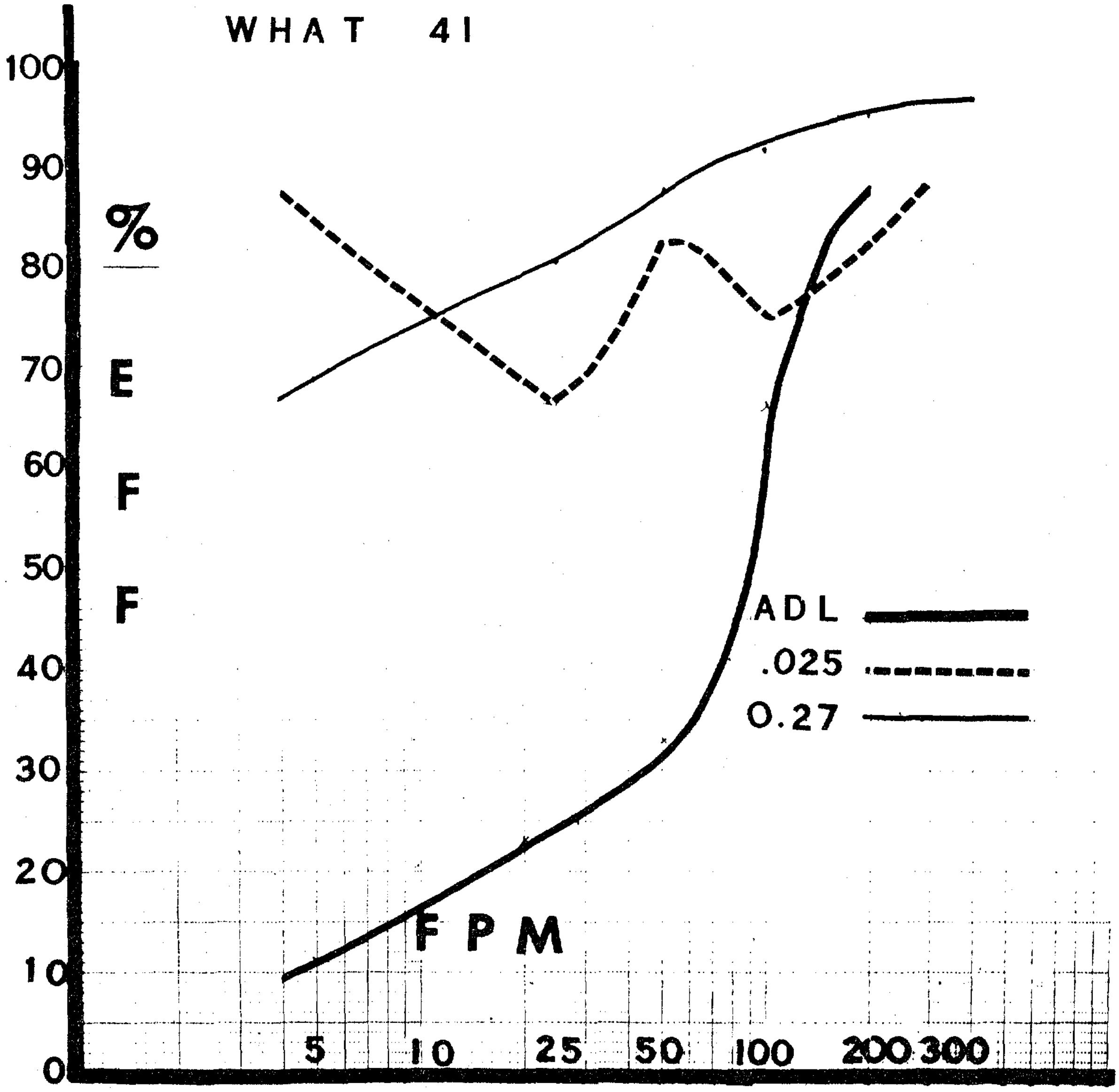

WHAT 41
100
90
%
80
70
E
60
F
50
F
40
30
20
10
0
ADL
.025
0.27
FPM
5
10
25
50
100
200
300

FIG. 9

DISCUSSION

COSTELLO: Are there questions concerning this paper?

SILVERMAN: I think you have left out some other related filter paper investigations. Detweiler at KAPL did a lot of work on particles in your size range. I am also a little puzzled by your need for the de-ionizer if you are trying to be realistic. To backtrack on that statement, Jess Thomas at Oak Ridge used a P-32 source for this purpose almost eight years ago in order to take the charge off his aerosols. He studied both DOP and methylene blue aerosols with this technique, as well as utilizing polystyrene latex of controlled sizes.

If you are trying to be realistic as to what is encountered in practice, I would like to know why you insist on taking the charge off.

POSNER: The reason for removing the charge from the generated aerosol was to eliminate the frequency of agglomeration. We are interested in a solid, spherical discrete particles, which could be produced and reproduced continuously and efficiently.

SILVERMAN: Then the charge on the aerosol is not critical. In your opinion, you don't have an aerosol without a charge?

POSNER: That is correct Dr. Silverman. The De-ionzer was solely employed to limit agglomeration and give us an aerosol of constant M. M. D., and not to remove all charge on the particles so as to neutralize them.

STERN: You have a plate containing 80 tiny perforations backing up the generator. Have you calculated the velocity through each individual hole?

POSNER: I have not done that.

LIEBERMAN: Have you calculated the velocity profile at the low face velocities in your system?

POSNER: This report is very preliminary and we expect to publish in the relatively near future where all this data will be included. I find it impossible to try to include everything we did to measure the aerosols at this particular meeting.

COSTELLO: Mr. A. L. Breslin from the Health and Safety Laboratory intended to present a complete paper as outlined on the program. All his data did not come in and in view of this he will present a very short summary, using the same title, "Simple Apparatus for the In-Place Testing of Unit Filters."

BRESLIN: We have addressed ourselves to the problem which Dr. Silverman mentioned in his summary earlier, that of testing absolute filters or, for that matter, any unit filter when installed in the ventilation system.

Nominally, this is a progress report, but in point of fact, progress has been very limited because of delays in the receipt of certain components. However, I should like to describe the work that has been done.

It is evident that the AEC's absolute filter testing program only partially answers the question of whether absolute filters perform properly when they are installed. We were considering methods of testing filters in place, that is, after installation in the ventilation system, when we heard a paper by Dr. Young, of the Naval Research Laboratory, at the AEC Safety and Fire Protection Meeting in Germantown earlier this year. He described test apparatus consisting of a DOP generator and measuring equipment that he had used to evaluate the performance of absolute filter installations. I note that he is on the program later on, and I assume he will discuss this work. His paper prompted us to look for a comparable system, but one more readily adaptable to the needs of New York Operations Office contractors. More about that later.

We had in the shop the uranine generator which Mr. Posner just described and we thought that with suitable modifications it could be made to produce an acceptable aerosol, but with considerably less equipment than was being used in the laboratory measurements. As modified, the generator consisted simply of the Dautrebande nebulizer and the impaction plate. Actually, with regard to the original equipment, a third component, the aging tank, was retained, but this was in effect the duct into which the aerosol would be injected. The generator fits compactly in a standard attache case along with the associated equipment. The total weight is in the order of ten pounds.

The aerosol produced by this simplified uranine generator is larger and has a wider size distribution than that produced in the laboratory, but is considered to be satisfactory for field tests. The mass median diameter is 0.7 micron and approximately 90% of the particles are less than 1 u.

The test procedure that we visualize consists of introducing the aerosol in the duct several feet upstream from the filter to be checked, and collecting simultaneously samples upstream and downstream from the filter with conventional duct sampling equipment.

We had hoped to be able to report actual field use of this equipment, but there has been time only for a few laboratory tests. We mocked up a duct section containing an absolute filter, and after a few shakedown tests we made three sets of measurements which appear to be valid. There we hit a snag.

The measured collection efficiency of the absolute filter was 99%. The reason for this is still uncertain. There was insufficient time to subject this absolute filter to the standard DOP test and, therefore, we don't know whether or not the unit was faulty.

Another possibility relates to our test aerosol. Something like 10% of the particles are less than 0.3u in diameter. We don't know what the efficiency of the absolute filter material is for these small particles -- perhaps there are persons who can tell us.*

That is the status of our program. The generating apparatus is simple and compact. This meets a paramount objective. Characteristically, our contractors utilize numerous, small ventilation systems, each serving limited areas rather than large systems. Each system, then, rarely contains more than two filters, and most usually only one. For this application, a testing arrangement must be simple and rapid so that a series of measurements can be made in a short period of time, preferably by one person.

As soon as the meaning of the unexpected result mentioned before is resolved, field tests will be performed.

GILBERT: What kind of a detection font are you using?

BRESLIN: The same as Mr. Posner described in the analytical part of it. We are taking samples with probes from the duct at a rate of one CFM upstream from the filter and about 20 CFM downstream. The reason for the greater flow downstream is to give us enough sensitivity to measure a collection efficiency of 99.9 plus.

GILBERT: How do you read it, then?

BRESLIN: The filter is macerated in water and read in a fluoimeter.

SILVERMAN: It doesn't have to be chopped, you just wash it off.

MURROW: Did you try this at rated-flow capacity or more or less?

BRESLIN: Of the absolute filter?

MURROW: Yes.

BRESLIN: It was operated at about rated capacity. This was a small MSA-150-CFM unit and we were running 140 to 145-CFM.

CHEEVER: About how much did it cost you to put this equipment together?

BRESLIN: I expected this question and I don't have a ready answer. I think I can make an estimate and see you later.

---

* Later penetration measurements on absolute filter material using the same aerosol confirm the 99% value.

THE REMOVAL OF GASEOUS CONTAMINANTS FROM AIR WITH A SORBANT INJECTION SYSTEM

R. Lopes Cardozo and P. Dejonghe
CEN Laboratories, Mol, Belgium.

SUMMARY

It is shown that a sorbant injection system can be used as a means of cleaning gases from contaminants when low or intermediate decontamination factors are allowed, for instance in closed circuits.

The sorbant injection system, in its simplest form, is composed of a certain pipe length with a sorbant feed system at the head-end and a cyclone, to remove the sorbant from the gas stream, at the tail-end.

It is shown that decontaminations of the order of 99% are easily obtainable.

INTRODUCTION

Many methods are known to remove gaseous contaminants from air, roughly to be divided into wet and dry systems.

Wet systems - packed absorption columns, venturi scrubbers, etc... - when used in nuclear systems contain certain disadvantages such as the production of certain volumes of effluent, the risk of carry-over of water mist that may cause clogging of filters, corrosion, etc...

In these cases dry filtering systems are more suitable to use if available. Most common is the use of static filters, e.g. to remove gaseous iodine and rare gases with activated charcoal, fluorine compounds with slag wool and many other applications. These filters can be designed to obtain the high efficiency usually required for open circuits, e.g. 99.9% and more.

In closed circuits however, a much lower efficiency can be tolerated, e.g. 98-99%. For this range of efficiencies a simple gas cleaning system was designed and tested, based upon the principle of injection of a powdered sorbant into the gas stream. After a certain contact time, the sorbant is filtered off from the gas stream.

In its simplest form, the system is composed of a certain pipe length with a sorbant feed system at the head-end and a cyclone, to remove the sorbant from the gas stream, at the tail-end. If desired the collected sorbant can be recycled.

The system offers many advantages over conventional systems:

1) It is simple and cheap to construct,
2) It has a very low pressure loss,
3) It is easily adaptable to existing installations.
4) It removes the contaminant from the gas circuit whereas, with static filters, the contaminant remains on the filter and thus continuously in contact with the gas stream. This may cause difficulties, for instance in the case of rare gas adsorption at low temperatures on activated charcoal filters. If these filters are heated up for one reason or another, the total of the rare gas adsorbed is again brought into the gas circuit.
5) If a high amount of radioactive matter is present in the gas, the sorbant can be bled off continuously at a predetermined rate in order to facilitate the handling of the active matter, that can now be done in parts.
6) It is thus quite possible to recover valuable sorbed matter; if, for instance, the system is used to adsorb rare gases at low temperatures, easy continuous recovery can be effected by warming-up the bled-off part of the sorbant. This can be done by using two cyclones in series. If the first one has a somewhat lower efficiency than the second one, the sorbant collected from the first cyclone is recycled. The fraction collected with the second cyclone is used for recovery, etc...

Inherent in the system is the fact that it shifts the difficulty of gaseous contaminant filtration to solid particle filtration. But, since the efficiency aimed at is rather low, it is believed that an adequate solid particle filtration system can have an efficiency as great as or even greater than the efficiency of the sorption. However, the efficiency can never be greater than that of the sorbant removal system. Of course, it is also possible to use methods other than cyclones such as bag filters or electrostatic filters.

The sorbant injection method was studied for the system activated charcoal/gaseous iodine/air at room temperature and pressure.

EXPERIMENTAL

Apparatus and procedure.

The following Pyrex glass ducts were used:

1) A helical tube with an internal diameter of 4 cm, a length of 10 m and a helix diameter of 54 cm.

2) Two straight ducts with an internal diameter of 2.5 cm and a length of 9.5 m (ABCDE Figure 1), respectively 4.5 m (ABCE).

Two small cyclones in parallel collected the charcoal that was mostly recycled by means of a Sigma pump (finger pump). A container with two litres of charcoal, about 1.3 kg, was placed between the cyclones outlet and the Sigma pump. Together with the Sigma pump, the container prevented blow-back of the gas up to gas velocities of 10 m/sec. For the once-through runs, a metered quantity of the charcoal was blown into the gas stream.

An absolute filter was placed in front of the induced draught fan. Normal laboratory air was mixed with gaseous iodine spiked with I-131 as is indicated in Figure 1. Samples were taken at the sampling points S just before the charcoal inlet and after the cyclones. A sampler is shown in Figure 1. It consisted of about 1 cm AAF FG 100 glass wool followed by 5-8 cm of granular activated charcoal. The efficiency of the sampler is known to be more than 99.99% (1). The charcoal used in the samplers was the CEBCA SA 1073 variety of 16-20 mesh size. As injected sorbant, the same variety was used, but with a mass median diameter of 136 micron.

Calculation of the decontamination factors.

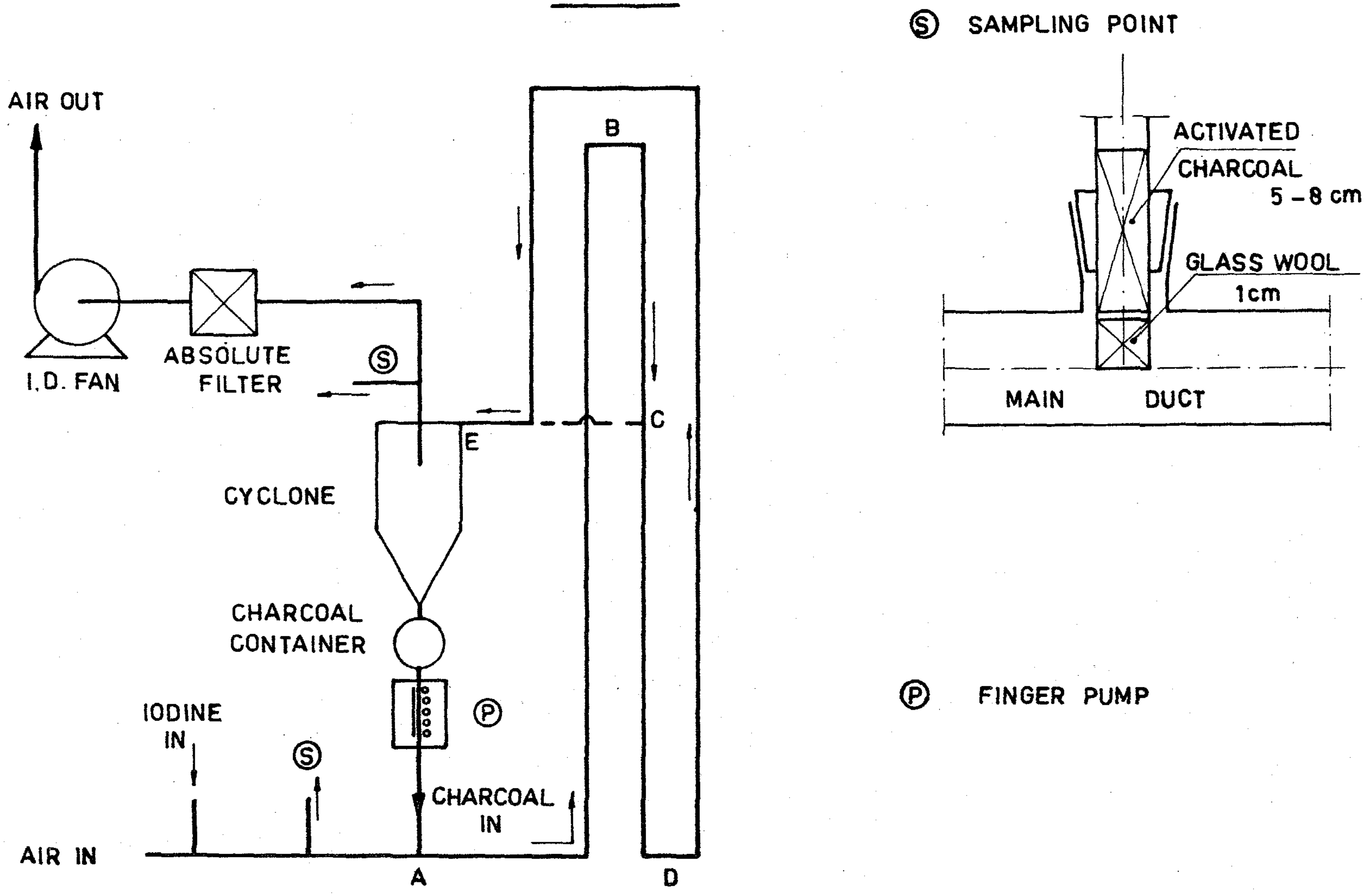
The over-all decontamination factor δ_1 was calculated from the total activity found on the samplers. It was observed that some charcoal was always present on the glass wool of the tail-end sampler. This is possibly due to abrasion of the charcoal whilst passing the total system. These very small particles were apparently not captured by the cyclones. It was therefore impossible to know whether the activity found on the glass wool came from gaseous iodine sorbed on these charcoal fines *in situ* or whether the activity arrived at the sampler already sorbed on the fines during the contact time in the duct.

For the once-through runs, the given δ_1 is thus a minimum value, since it is calculated from the total activity on the samplers.

For the recycle runs, however, it was possible to obtain a correction for δ_1 by continuing each run after stopping the addition of gaseous iodine. After a certain time lapse for the removal of all gaseous iodine still present, samples were taken again. The activity now found on the glass wool came from abraded charcoal fines and was subtracted from the original quantity. The corrections amounted to about 10% (absolute) for the low decontamination factors and to about 30% for the high ones.

The δ_1 -values, corrected in this way, thus correspond with factors obtainable with a sorbant removal system more efficient than the cyclones used here. The cyclone efficiency here was better than 99%, but lower than

FIGURE 1.



FLOWSHEET SORBANT INJECTION SYSTEM

that of the so-called high efficiency cyclones.

From the experimental data, another decontamination factor was calculated. This factor, δ_2 , was calculated from the activity found on the head-end sampler and on the charcoal only of the other sampler. It represents the decontamination of a system consisting of a contacting duct, followed by a cyclone and a glass wool prefilter.

RESULTS

Helical duct.

The decontamination results obtained with the helical tube are given in Table 1.

TABLE 1

DECONTAMINATION RESULTS WITH A HELICAL DUCT

Iodine concentration: 1 to 2 mg/m^3

N°	$\bar{v}$ m/sec	τ sec	H hr	K kg/m^3	Recycle	$\delta_1 \pm [\sigma(\delta_1)\ \%]$	Samples	δ_2
1	5.2	1.92	–	1×10^{-3}	–	6.9 ± 7	2	7.6
2	5.2	1.92	–	6×10^{-3}	–	13.3 ± 7.5	2	39
3	5.2	1.92	–	7.5×10^{-3}	–	13 ± 38	4	–
4	5.2	1.92	–	16×10^{-3}	–	13.3 ± 1.5	2	300
5	5.2	1.92	–	28×10^{-3}	–	9 ± 19	2	300
6	15.5	0.65	–	4×10^{-3}	–	12.2 ± 3.3	2	160
7	9.7	1.03	3.5	0.3	+	10 ± 20	2	–
8	9.7	1.03	7.1	0.3	+	9.2 ± 22	3	2800
9	9.2	1.19	10	0.3	+	14.5 ± 9	2	280
10	8.1	1.23	13	0.3	+	26 ± 40	3	400
11	8.1	1.23	21.3	0.3	+	22 ± 27	3	500

In this table the face velocity $\bar{v}$, the period spent by the gas in the duct τ , the charcoal concentration K and the total of hours the charcoal was recycled H are given.

The results (δ_1) are under all circumstances low. It was therefore decided to replace the helix by another contacting duct of smaller diameter, in order to obtain a higher charcoal concentration with the same face velocities. Since the flow in the helix, owing to its helical form, was still laminar, the friction factor f having a value between 0.05 and 0.09, it was decided to have a straight duct.

Straight ducts.

The results obtained with straight ducts are represented in Table 2.

After several runs it became apparent that decontamination increased with the charcoal recycling time. This can be understood by assuming that abrasion of the charcoal causes a decrease of its mean particle diameter, thus increasing the effective surface per weight. Abrasion may take place both during the time the charcoal flows through the ducts and in the feed pump. The limit of the recycling time is set by the cyclone efficiency or by the sorption capacity of the charcoal. In this case about 50 mg of iodine was introduced per hour. This makes, on a total inventory of 1.3 kg of charcoal and at 80 hr recycling, about 3 mg per gram; this value is much lower than the sorption capacity, of the order of several hundred mg per gram charcoal (1). It is noted that the sorption of iodine on the CEBCA SA 1073 charcoal is of a chemical nature, owing to the copper present on the charcoal (1).

In order to check the assumption that abrasion is the cause of the improved decontamination, grain size distributions were analysed of several charcoal loads.

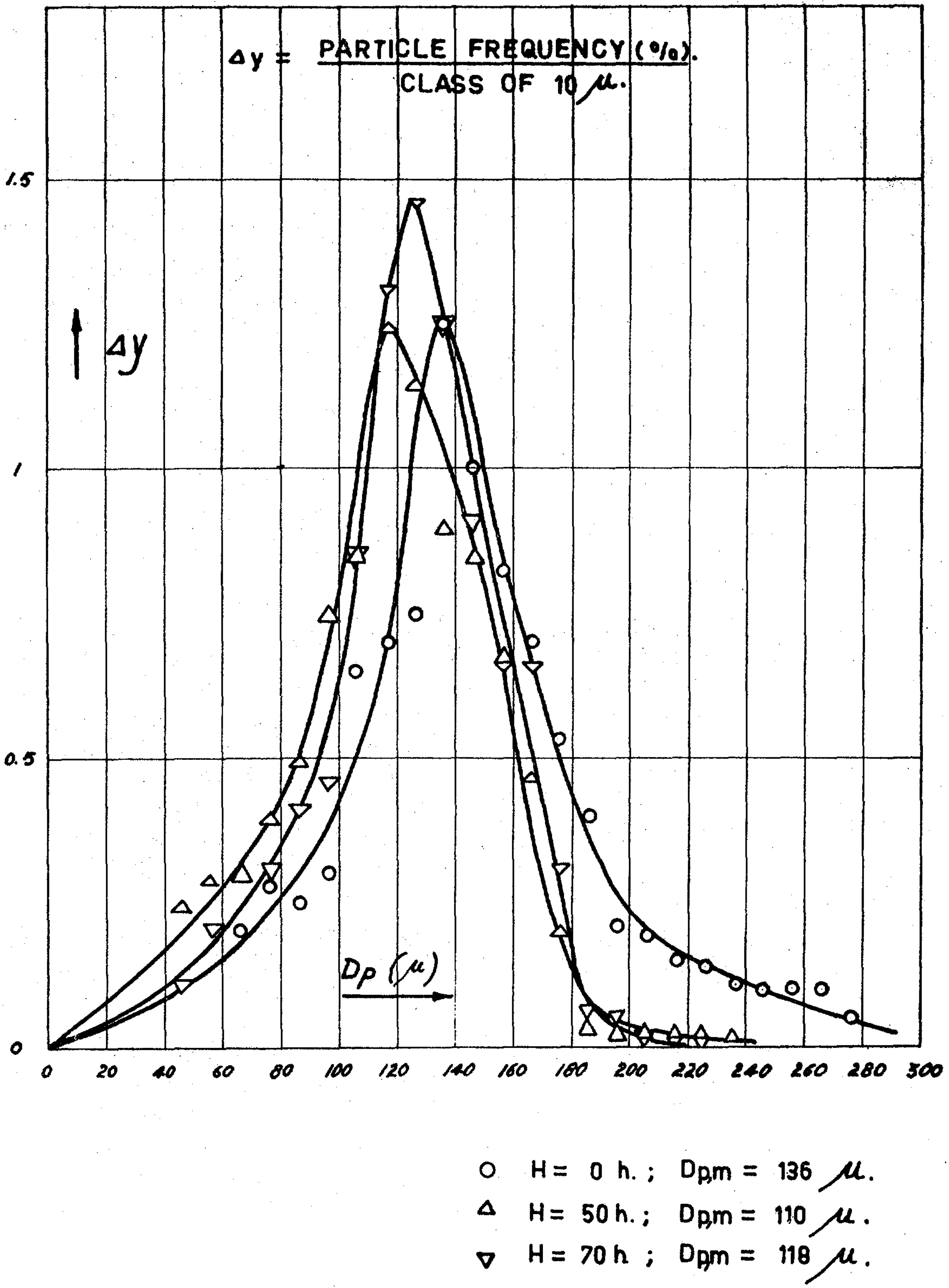
The method used was a sedimentation analysis in ethylene glycol according to the method of Bergshoeff (2), in a slightly modified Andreasen-Esenwein pipette. The results are given in Figure 2 as frequency distributions of the various samples. It can be seen that the smaller particle sizes are more frequent in the charcoals that were recycled for 50 and 70 hr, whilst the diameters of more than 200 micron disappeared almost completely as compared to the original charcoal. It is noted that the 50 and 70 hr-charcoals were from two different batches and since the nature of the abrasion is not exactly known, the irregularity in the decrease of the mass median diameter $D_{p,m}$, cannot be explained.

The increase in the charcoal surface is about 1.5 times for the 50 hr-charcoal as compared to the initial charcoal, assuming that the particles are spherical.

A representation of the experimental data in a generalized form can be given by reasoning that the decontamination factor δ_1 is increased by longer use of the charcoal. In Figure 3 some data of Table 2 are given.

FIGURE 2.

PARTICLE SIZE FREQUENCY DISTRIBUTIONS.



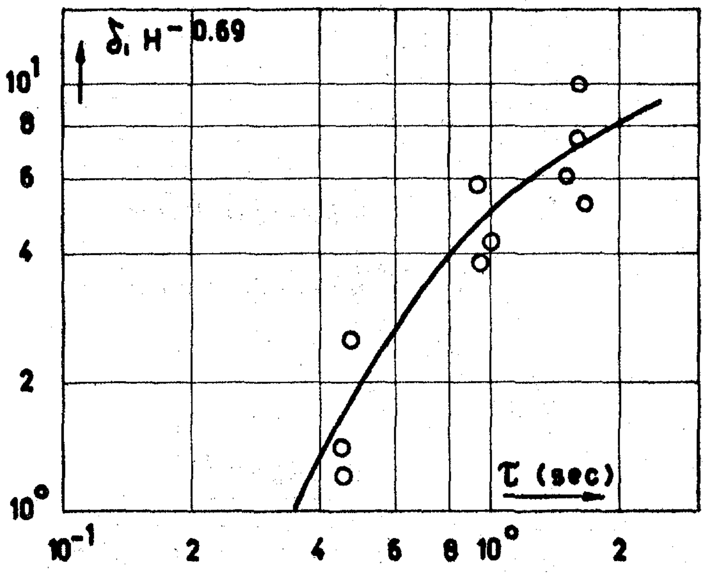
At a charcoal concentration of 0.8 kg/m^3 and a residence time of 1.6", it appears that

$$\delta_1 = 8.3\ H^{0.69},$$

with a standard deviation of 25%.

Another factor influencing the decontamination factor is the residence time τ, this being the case until this period is sufficient to obtain equilibrium.

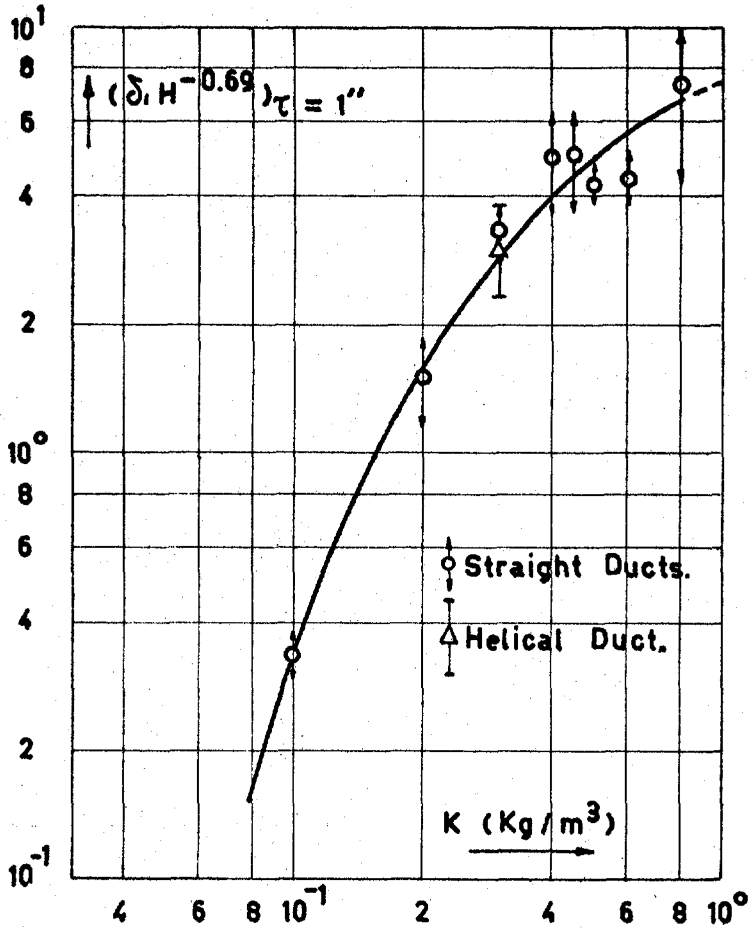
Data at K $\doteq$ 0.4 $\longrightarrow$ 0.5 kg/m^3 are given in Figure 4, where the factor $\delta_1\ H^{-0.69}$ is plotted against the residence time τ. It can be seen that equilibrium is not yet established at $\tau \leq 1.6$ sec.



FACTOR $\delta_1\ H^{-0.69}$ VERSUS RESIDENCE TIME τ AT K = 0.4 - 0.5 Kg/m^3.

FIGURE 4

FIGURE 5.



NORMALIZED VALUES OF $\delta_1 H^{-0.69}$ VERSUS CHARCOAL CONCENTRATION K (Kg/m^3).

It is thus possible to account for the influence of τ by standardizing all $\delta_1 \ H^{-0.69}$ - values to a value of τ = 1 sec with the help of Figure 4. The corrected values of $\left[\delta_1 \ H^{-0.69}\right]_{\tau = 1 sec}$ are represented in Figure 5, plotted against the charcoal concentration K for both helical and straight ducts. For purposes of clarity, only the averaged values with standard deviations are given.

It is noted that the data obtained with the helical duct in recycle fit surprisingly well into the generalized form of Figure 5, although the flow there was laminar. In the straight duct runs, the flow was slightly turbulent, the friction factor having a value between 0.007 and 0.008.

The cyclones used were only home-made medium efficient ones. The charcoal losses amounted to about 0.2 g/m^3. This value can be made smaller if high efficient cyclones are used.

The over-all decontamination factor δ_2 varies for all runs between 30 and 20,000, but it has mostly a value around 2500. This means that the decontamination is about 99.96% for the sorbant injection system, if the cyclones are followed by a glass wool filter. In this case about 99% of the iodine is retained in the recycle system, the rest on the glass wool.

CONCLUSIONS

It is shown that it is in principle possible to use the sorbant injection system as a means of removing contaminants from gases when low or intermediate decontamination factors are allowed.

About the experimental conditions the following can be said:

1. Face Velocity

The lower limit is set by a satisfactory transportation of the injected sorbant, the upper limit by several factors, like blow-back through the feed system, pressure loss, abrasions, etc... In general, the face velocity can have a value high enough to obtain slightly turbulent air flow.

2. Residence Time

This time cannot be fixed, since sorption depends on the characteristics of sorbant and contaminant. In the case described here, a residence time of 1-2 sec seems to be sufficient.

3. Charcoal Concentration

In recycle, the concentration is of minor importance. It is felt that a concentration between 0.5 and 1.0 kg/m^3 of finely divided sorbant will in general be sufficient.

4. Charcoal Particle Size

The results depend to a great extent on the particle size of the sorbant. The smaller the size, the better the decontamination. The lower limit is set by the efficiency of the particle removal system, the abrasion rate and the feed system. The recycled sorbant should flow easily in the feed system, in order to obtain troublefree performance.

5. Recycling Time

The time a sorbant can be recycled depends on the contaminant concentration and the capacity of the sorbant.

In the case described here, a decontamination factor of 100 or more was obtained under the following conditions:

1. Face Velocity : 6-8 m/sec
2. Residence Time : 0.8 - 1.6 sec
3. Charcoal Concentration : 0.8 kg/m^3
4. Charcoal Particle Size : around 100 micron
5. Recycling Time : up to 80 hr

NOTATION

D_p	particle diameter, (micron)
$D_{p,m}$	mass median particle diameter, (micron)
f	friction factor, (-)
H	recycling time charcoal, (hr)
K	charcoal concentration in gas, (kg/m^3)
L	length of contact duct, (m)
$\bar{v}$	gas face velocity, (m/sec)
Δ_y	frequency of particle size group
δ_1	decontamination factor of contact duct only, (-)
$\sigma(\delta_1)$	standard deviation of former, (%)
δ_2	decontamination factor of contact duct and glass wool, (-)
τ	residence time of gas in contact duct, (sec)

REFERENCES

1. Cardozo, R. Lopes and DeJonghe, P. : Glg 53 - C.E.N. Mol, Belgium. November 1960.

2. Bergshoeff, G.: Chemisch Weekblad 56, 709 (1960).

DISCUSSION

SCHNELLER: Do you have any figures on the pressure drop?

DEJONGHE: Pressure drop is just the pressure drop of the ducts and cyclones. It depends on the design of the tube and can easily be calculated.

SILVERMAN: I don't want to monopolize the question period, but I had a doctoral candidate who did a fluidized bed study of carbon in such a system for SO_2. It would work equally well for iodine, but we got enough attrition in the fluidized bed to indicate that you are transferring the situation, from an adsorption problem to a filter problem. This fine particulate will deposit on the filter. As far as I know, I have not heard engineering advantages of this system defined. I know enough disadvantages now to raise a question about it in terms of net performance. In fluidized contacting there will be attrition and instability problems. I think they have already found such problems at Idaho. I would raise a question here too as to the amount of iodine to be handled. Wouldn't a static system have material advantages over this particular combination with its cyclone performance and all the rest of the system? It still cannot be good enough to prevent fine particulate from reaching the filter and giving a very rapid build-up of resistance.

DEJONGHE: Here we have no fluidized bed but merely a concurrent flow of air and charcoal. Attrition is less than in a fluidized bed since the charcoal concentration is smaller. Besides, ducts can be designed to give minimum attrition. The fines are note so small, about 10 u, so an inexpensive glass wool filter can do the job.

Net losses amounted in this work to about 200 mg per m^3 of air treated, using the low efficiency cyclone. A high efficiency cyclone would of course be much better to use.

SILVERMAN: Of course, the iodine problem and the relative volumes of carbon, the amount of actual iodine you are collecting in terms of micrograms is pretty small because of its high specific activity. If you can prove that the carbon deteriorates in place, then all the static beds we have we are in trouble.

DEJONGHE: Saturation is of course a function of the quantity of iodine present. In this work we had about 1% saturation after 80 hours of operation.

SILVERMAN: Even with carbon, you get a chemical reaction, as far as we can tell. What I am trying to point out is that the life of a carbon bed is tremendous in terms of the work it will ever have to do.

DEJONGHE: Yes. You get a saturation of a tenth of a percent, or something like that, at the end of the operation. That is true.

GILBERT: Did you do any studies on the relative collection efficiency of just activated charcoal versus impregnated charcoal which you are using?

DEJONGHE: No. It has been done on filters, as far as I can recall, but I couldn't give you any results. Results are given in BLG-53.

ROBINSON: What range of iodine concentration did you work with for initial iodine concentrations. Did the DF stay essentially the same over this range?

DEJONGHE: It is rather unfortunate that I didn't do this work myself and I have to present it here. I can find this out and if you can ask me this question somewhat later in the day, I will tell you. I think it is something of the order of a milligram of iodine per cubic meter, but I am not sure.

COSTELLO: I understand these papers are translated?

DEJONGHE: That is in English. Whether it is good English or not, that is another question.