SESSION XII

NEW AIR CLEANING TECHNOLOGY FROM EUROPE

Wednesday, August 9, 1978
CHAIRMAN: D. W. Moeller

OPENING REMARKS OF SESSION CHAIRMAN:

We have with us for this Session, experts from four European countries who are going to bring us information on new developments in air cleaning technology. These men are: Mr. R. G. Dorman, of the Chemical Defense Establishment, United Kingdom; Dr. P. Sigli, of the Atomic Energy Commission, France; Dr. A. Bruggeman, of the Nuclear Energy Center, Belgium; and Dr. J. G. Wilhelm, Director, Laboratory for Aerosol Physics and Filter Technology, Federal Republic of Germany.

Each speaker will take 5 or 10 minutes to give a short presentation and after each has finished, there will be an opportunity for questions and general discussion.

Although, regrettably, they were unable to be present today, J. Dubé, Organization for Economic Co-Operation and Development Nuclear Energy Agency, and Y. Zabaluev, International Atomic Energy Agency, have submitted for publication in the Proceedings of the 15th DOE Nuclear Air Cleaning Conference their paper entitled "IAEA/NEA Activities in the Field of Gaseous Wastes."

NEW AIR CLEANING TECHNOLOGY IN THE UNITED KINGDOM

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When I was asked to take part in this discussion, I spoke to people in the U.K. and I wrote to others. I looked at the Proceedings of a conference which had been held in France at Aix-en-Provence in late 1976, entitled, High Efficiency Air Filtration in the Nuclear Industry--a Conference at which several Englishmen had contributed papers. Neither the replies to my questions nor the Conference Proceedings indicated that there were any world shaking technological innovations. Rather, the picture was one of relatively modest improvements. I will outline some of the main points.

Some of you may not have heard the paper by Bennett, Elliott, and Lilleyman ("A Review of United Kingdom Work on Gas Cleaning in Fuel Reprocessing Plants"), which was presented yesterday by John Dyment of the AWRE. The areas being studied at Harwell in gas cleaning for fuel processing are reduction of installation size including a study of closed cycle ventilation, increased filter life by prefiltering, and improvements in handling and storage. An interesting development is the use of quick release fasteners for dismantling a filter case--either remotely or directly—in order to reduce storage volume. Another technique for dismantling is also being investigated; one side of the filter case is removed, exposing one edge of the pleated glass fiber paper. This is gripped and the paper is wound onto a spool. Paper and spacers can be compacted or melted. There is also the possibility that cases may be made of resin-glass fiber mixes.

Harwell are also hoping to make use of an electrostatic precipitator as a prefilter. AWRE are mainly concerned with the design of buildings and containment systems for plutonium processing. This will also include attention to ventilation and filtration problems.

In the field of testing, we have sodium chloride equipment which permits us to measure as low as 0.00005% at 2,000 cfm for an upstream concentration of salt of less than 10 milligrams per cubic meter. In the past year or so, we have incorporated a chart recorder and Dautreband atomizers. We hope to make Dautrebande atomizers the European standard instead of the Collision atomizer which requires a higher compressed air pressure and also produces an aerosol of somewhat larger mass median diameter. The aerosol from the Dautreband atomizer is not, of course, monodisperse but it is centered at about 0.45 micrometers mass median diameter.

The thermally generated sodium chloride aerosol equipment which I described at the 13th Air Cleaning Conference is now available commercially. It has shown itself to be reliable and easy to operate. Some of you may recall that a stick or pencil of almost pure sodium chloride may be made by mixing granules of salt with a little magnesium oxide and ethyl hydroxy cellulose. This is passed at a controllable rate through an oxy-propane flame, producing an aerosol composed of all submicrometer particles of salt.

Installation tests on a 2 x 50,000 cubic meters per hour system (30,000 cfm) of HEPA filters in tandem showed an initial penetration of more than 0.1 percent on the lower bank of filters. Holes in the filters and bad seating were found either by simple visual inspection or by scanning with a probe. After rectifying these faults, the penetration measured was 0.01%. When the top bank of filters

was fitted, so the tandem system operated, penetration was less than 0.0007%, the limit at which our detection equipment was capable of accurate measurement.

The generator weighs seven kg and, with two small gas bottles, one of oxygen and one of propane, enough for five tests, the total weight is 13 kgs. The photometer detector is about the same weight. When in operation, you can see the flame coming out, but it's quite a short flame and we don't think there is any fire hazard. We have more than one size. Both generator and detector are completely portable and completely independent of fixed main services such as electricity and compressed air. The penetrations measured with the thermally generated aerosol differ very little from the standard compressed-air generated salt cloud. The aerosol is cubic in shape. I should add that with the thermally generated aerosol, we try to work at a concentration of 7-10 milligrams cubic meter of air for onsite tests so deposition of salt does not cause any rise in filter resistance.

A propos of the first day's discussion on DOP distribution, we found on our own DOP equipment a spread of sizes of 3.5:1, some by a replication method followed by electron microscopy. We are hoping to check absolute sizes from the machine with a new high speed conifuge that we built. We were worried by the spread of sizes and wondered whether we were working the machine properly. We felt a little bit shamefaced at coming and telling you of this sort of spread, but I must say that Monday's two talks relieved my worries considerably.

An investigation into the ASHRAE dust spot correlation with the sodium chloride tests has led us to the view that sodium chloride can replace the very time-consuming dust spot test and, indeed, it is more reliable, as the aerosol is always at a fixed composition and size distribution whereas atmospheric dust may vary considerably in composition, size, and in color. A dust spot and sodium chloride correlation study that included dozens of tests showed at the lower end a transmission percentage for the ASHRAE test of 20% when the sodium chloride test showed about 10%. They went up pretty well together until they became equal at 100%. In general, atmospheric dust penetrates a little more easily than sodium chloride, but there is an extremely good correlation. The scatter was not as severe as I would have thought before the experiments were done. I think this is of interest in the nuclear field because of the need to employ prefilters.

Some work on clogging of filters, mainly of the ventilation type, is being reported next month at a symposium in the U.K. Finally, one of our universities puts in a major effort in filtration and dust sampling. They have, at present, investigations on particle impact on flat surfaces, the effect of aerosol loading on fibrous filters, and aerosol migration in thermal fields, as well as other filtration studies which are, perhaps, of less interest in the nuclear field. There's also been some work, done by Mr. Dyment, on clogging of filters.

DISCUSSION

GILBERT: Was the illustration of sodium chloride aerosol, referred to in the presentation, made from sodium chloride by the thermal method or by droplet evaporation?

DORMAN: It was an electron micrograph of the thermally generated aerosol. However, both the thermally generated aerosol and that produced by evaporation from the atomized solution appear similar.

MOELLER: You mentioned the quick release fasteners for the filters. Do you have data to show how much time you save, or how much money you save, or how much exposure you save?

DORMAN: I don't have figures here. The device saves exposure as well as time but the quick release fasteners are really designed for ease of dismantling and subsequent reduction of filter volume. If the pack can be removed by hand, it can easily be processed and the case can probably be recycled. At higher levels of activity, it may be necessary to go to the second case of the filter where you remove one side and wind the paper onto the spool.

DEMPSEY: Can you measure two filters in series with the sodium chloride method?

DORMAN: I don't think I made it clear that the figure of 0.0007% quoted was the lower limit at which we believe the detector employed could give an accurate answer. The real value was well below this. We certainly could not measure penetration of two intact HEPA filters in series if the DF were greater than, say, 10^7 with our normal upstream concentration of 10 mg/m^3. The 30,000 cfm filter system we tested had filters in tandem, one above the other. Obviously, we couldn't test the top layer without the bottom, so we took off all the top filters and just tested the bottom layer. There, we found faults that gave penetrations of about 0.1%. After rectifying them, we put the top layer back and then tested the two. We didn't measure the penetration. We calculated that it must have been less than 0.0007%, which we thought was about the minimum that we could possibly measure with that particular type of detector. We can do better than that with our big detection device, but it is not easily portable. To test filter combinations with DFs much better than 10^7, it will be essential to identify the penetrating particles. The concentration of the test particle suspension will need to be very low (I assume solid, insoluble particles) to ensure that each droplet sprayed contains only one particle. Without doing calculations, I suspect that particle concentration in the carrier liquid will have to be much less than 0.1% by weight so large quantities of spray (probably greater than 1 liter) will be involved. To detect a truly representative sample of the small number of effluent particles will require a complex mixing device downstream. While we must always strive for greater sensitivity of detection, I have some doubts as to the value--and validity--of quoting figures of DFs much better than 10^7.

SOME DEVELOPMENTS IN NUCLEAR AIR CLEANING IN FRANCE

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I. IODINE TESTING

Before starting up a new plant, we have to check the quality of different elements--charcoal, modular unit, the whole bank installed (in situ test)--and French rules for such tests were defined in a paper (1) presented at the conference on HEPA filtration organised by CEE at Aix-en-Provence.

I.1. Modular unit testing

The problem of testing a modular unit needs facilities permitting reliable control of relative humidity.

During the last two years we have developed such a facility.

The installation has a maximum flow rate of 2,000 $m^3.h^{-1}$ and gives a controlled relative humidity of between 40 and 95%.

In order to obtain these performances this installation comprises:

-a humidifier tower in which the input air reaches a relative humidity of about 95%, its temperature being some degrees more than the ambient temperature;

-this tower is followed by a cooling system so that the gas is cooled to about one or two degrees less than ambient. Just behind this cooling system the air reaches saturation;

-heating the saturated air with a given difference of temperature allows us to reach relative humidity required.

All the temperatures or differences of temperature are controlled and monitored to 1/10th degree centigrade so that the final precision obtained is better than 2%.

During a test, a modular unit is installed and upstream and downstream samplings are made with the same charcoal--same grade, same quality, same batch--as that filling the cell under test. This permits comparison of the efficiency of a modular unit with that of the charcoal itself, in exactly the same conditions of velocity, temperature and relative humidity.

Using this facility it has been possible to show that the efficiency of the cell with regard to methyl iodide is strongly dependent on a shape factor taking into account some partial internal leaks and reducing the real bed depth of the charcoal bed.

We are now developing a computer program which will be able to predict values of efficiency for any relative humidity and velocity, starting from an experimental value at given relative humidity and velocity (limited for one charcoal and for one

manufacturer). It is believed that this method could be extended to in situ tests, permitting estimation of performance in accident conditions.

I.2. Iodine in situ testing

Before the new plant starts, the whole bank installed is tested in situ by a labelled methyl iodine test and periodic in-service checks are requested.

Principles of the experimental method used in France were given elsewhere (2).

Due to aging of the charcoal we have initiated a program to follow the weathering of a bank in a nuclear plant. The initial test took place some weeks ago and a three or four weeks test frequency is foreseen.

II. IODINE TRAPPING IN A REPROCESSING PLANT

Up to now, iodine from dissolver off-gas is trapped by washing in a caustic scrubber. A large part of iodine is in solution. A process for ^{129}I recovery from the scrubber solution by precipitation as an insoluble metallic iodide is actually in progress (3). This iodide will be ultimately included in a low melting point glass.

Concerning ^{129}I contained in gas after the scrubber, a program of investigations is just starting. This program mainly includes the construction of a full scale installation with an inorganic adsorber, zeolite-Ag exchanged, or AC 6120-Ag impregnated, or another equivalent adsorbent.

This installation, with a flow rate of 300 $m^3.h^{-1}$, must improve the behavior of the filter at temperatures of 120-150°C. It will permit, also, testing of handling systems, irradiation and contamination problems linked to handling, loading effects, and so on.

Located after the caustic scrubber, this solid filter will be preceded by a demister, a HEPA filter and, eventually, a cooler and a heater in order to have a dry gas at 120-150°C.

Conditioning of the adsorber, mainly in a glass, is examined for long term storage of ^{129}I.

III. KRYPTON RECOVERY (3)

Krypton eighty-five is present in gaseous wastes with inert xenon. We have therefore examined the possibility of recovery of both gases and their final separation, krypton for ultimate storage and xenon for possible commercial purposes.

In France, cryogenic distillation and separation were investigated on inactive gas. The process was based on a concentration of noble gases in liquid oxygen before distillation. Separation needs previous elimination of iodine, hydrocarbons, and ozone. Fundamental research on physical and chemical properties of noble gases and air component mixtures is now under investigation.

A basic flow sheet of a krypton recovery installation is given in fig. 1.

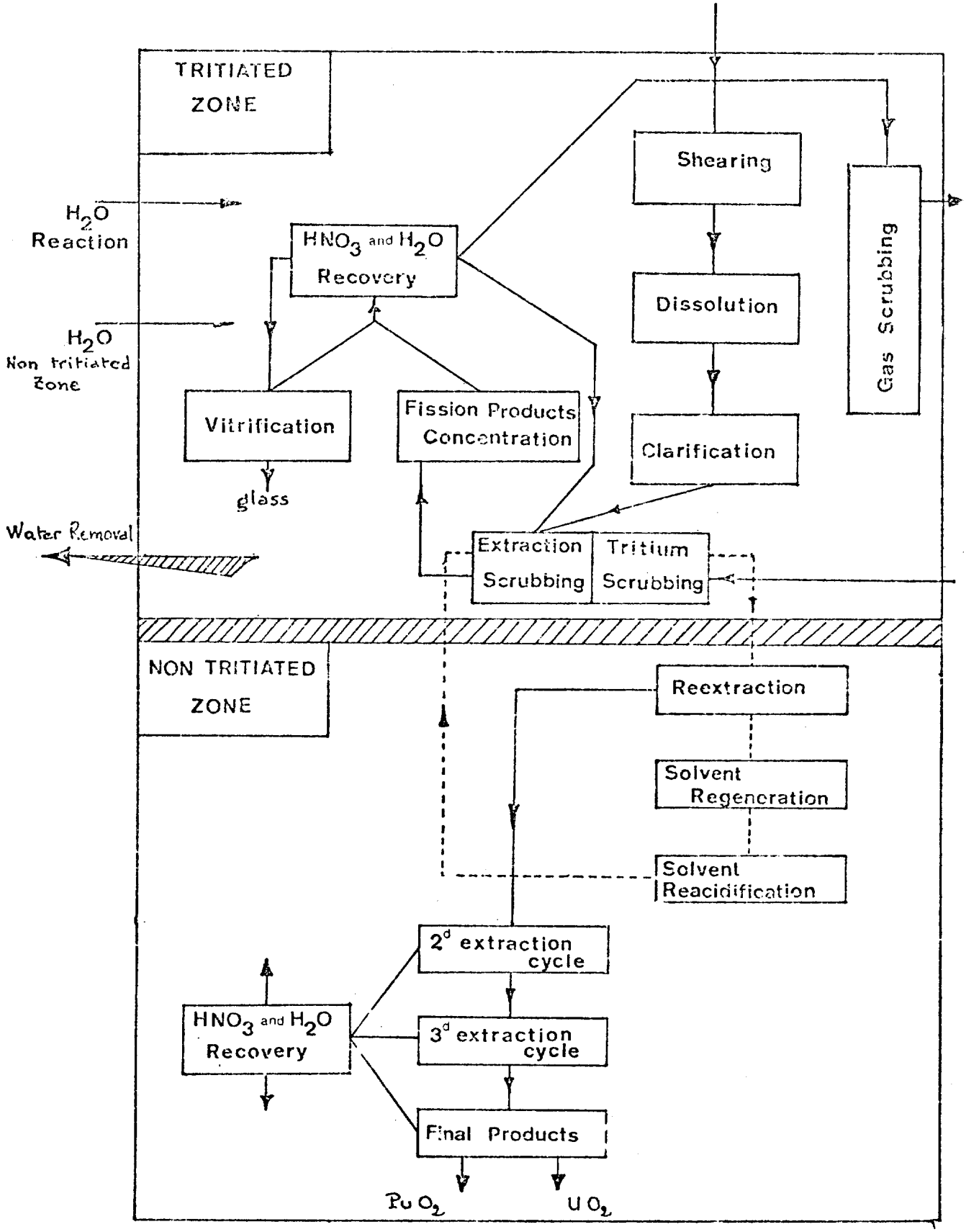

Fig. 1 - TRILEX process

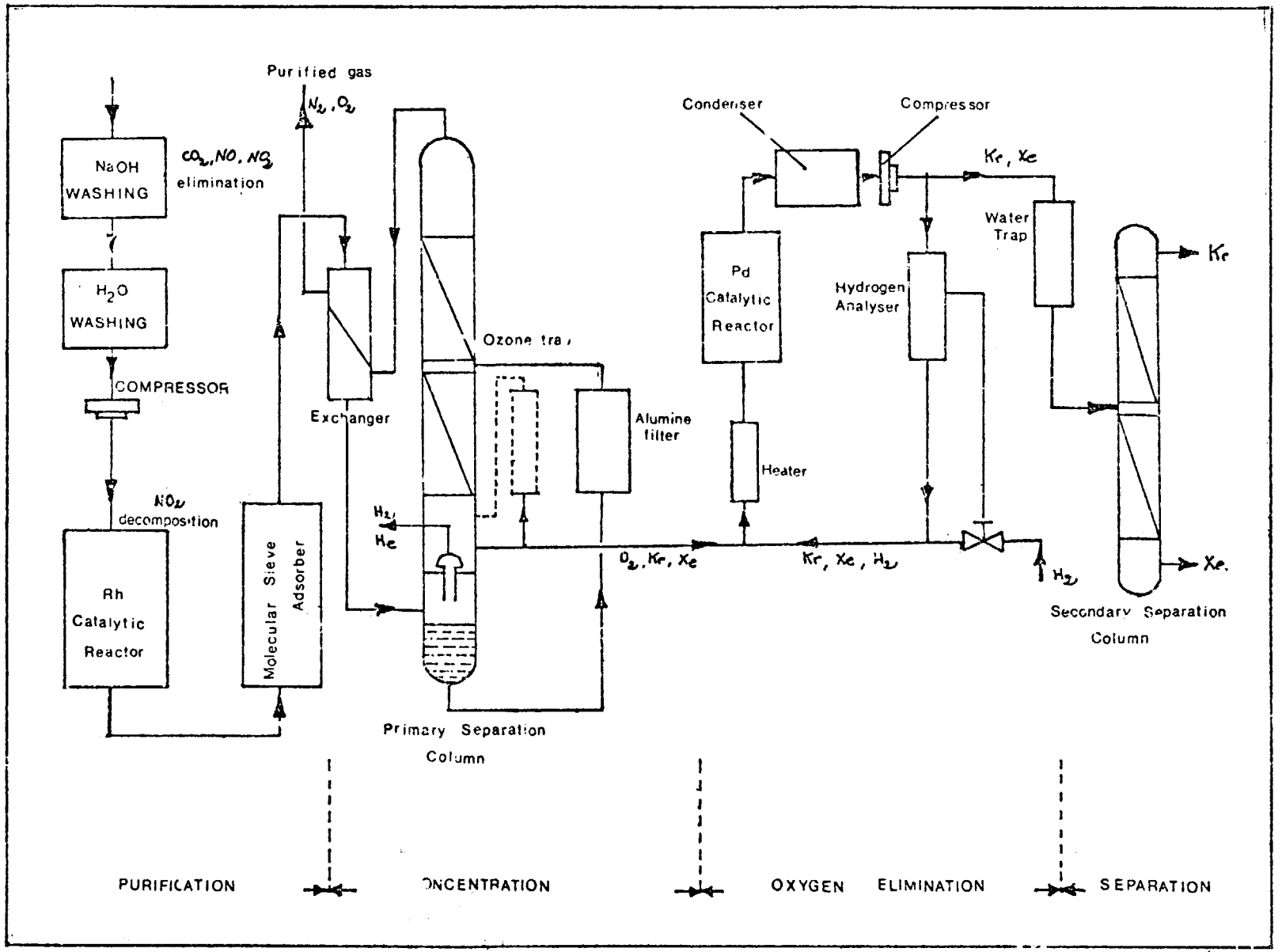

Fig. 2 - Krypton Recovery flow-sheet

IV. TRILEX PROCESS (4)

During reprocessing, tritium which is contained in the fuel is exchanged very quickly with hydrogen, principally during the dissolution step. This provides about 10^6 Ci of tritium in aqueous wastes coming from a reprocessing plant of 1,500 T/year capacity.

To avoid dispersion and dilution of tritium in natural waters, it becomes necessary to confine tritium in a limited part of the reprocessing plant, to remove the tritium from the process in order to transport it to a final disposal after enrichment by a conventional system.

For reaching this objective, a new process, the TRILEX process, was investigated at the Centre d'Etudes Nucléaires de Fontenay-aux-Roses.

Mainly, this process consists of (fig. 2):

- establishing a tritium barrier located at the same level as the first plutonium and uranium extraction cycle by washing the loaded solvent with natural water and making use of the property of tritium to be not irreversibly fixed on TBP diluted in an alkane;

- limiting the entrance of new water at the plant inlet so that dilution of tritiated water will be avoided.

This process needs to have two different nitric acid recovery systems. One of them is located in the tritiated zone; it receives liquids coming from the first extraction cycle and from fission product treatment. The second one, located in the non-tritiated zone, receives all the non-tritiated acid of the plant.

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DISCUSSION

R. A. BROWN: What rate of aqueous waste processing will be required for tritium recovery?

SIGLI: About 500 to 1000 kg per uranium metric ton. The water is coming from the reaction of the solution coming from the extraction. If you limit the quantity of water entering in the first part of the plant, you reduce the amount of water to be removed at the end.

BROWN: You spoke of the use of conventional processes for concentrating the tritium. What process will be used for separation and enrichment of the tritium?

SIGLI: Principally isotopic exchange (water-tritiated water) or electrolysis followed by H_2-HT separation.

BROWN: You mean electrolysis and then a catalytic exchange?

SIGLI: Yes.

BROWN: That might be a very costly process applied to a fuel reprocessing plant.

SIGLI: Yes, it is.

BROWN: Will the radioactive and chemical contaminants interfere with the enrichment process?

SIGLI: Due to the low level of radioactive contaminants in the solution at the removal point, we don't expect any particular problem.

VON AMMON: I am surprised that you did not mention the French program of krypton retention which is carried out at Fontenay-aux-Roses, as far as I know. Since rare gas retention is a topic of this conference, I would ask you to say a few words.

SIGLI: You are right, but I prefer not to speak about it because I do not have enough information.

HULL: Are there governmental regulations in France that require the removal of ^{129}I or tritium from reprocessing effluents?

SIGLI: Up to now,the retention of iodine and tritium was not specifically requested. However, it is foreseeable that in the near future regulations will become more stringent and we are planning corrective actions when necessary.

GAS CLEANING RESEARCH AND ENGINEERING IN BELGIUM

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S.C.K./C.E.N., Mol, Belgium

I. Introduction

The Belgian electro-nuclear plan in execution foresees a total nuclear power capacity of 5246 MWe available in 1982, i.e. 35% of the electrical power capacity at that time.(1) On a long term the Belgian nuclear program will be restricted to a maximum of about 10,000 MWe. The latter figure has been used in the assessment of the off-gas cleaning conditions in a potential reprocessing plant localized on Belgian territory.(2) This assessment indicated the necessity to trap the iodine compounds with an over-all decontamination factor of 181, to retain krypton with a D.F. of 6.3 and to remove the actinides with a D.F. of 7.10^7. For discharge of tritium into the Scheldt river as HTO, a D.F. of 6.2 is required.

II. Survey of the Belgian R & D Program on Gas Cleaning

These requirements for the reprocessing of LWR-fuel are the basic incentive for the Belgian R & D program on the cleaning of nuclear off-gases. Other incentives are:

- the long term program on the reprocessing of fast reactor fuel;
- the need for gas cleaning units in the nuclear waste treatment facilities;
- the optimization of the off-gas treatment in pressurized water reactors.

Hence, the Belgian R & D program on nuclear gas cleaning has a broad application field.(3) It concentrates on the following items:

- the delay of xenon in PWR off-gas;
- the trapping of iodine compounds on various solid m-terials;
- the trapping of iodine compounds by mercuric nitrate solutions;
- the removal of krypton by cryogenic distillation and the gas preconditioning steps related to it;
- the behavior of semi-volatile ruthenium compounds in gaseous effluents;
- the isotopic enrichment and separation of tritium from aqueous effluents by electrolysis and water hydrogen isotopic exchange, and the catalytic oxidation of tritium.

The various test loops available to investigate these items will be described and their present state of development will be commented.

III. The Pilot Delay-Bed

The experimental data obtained in a test rig consisting of a 15 cm diam. column(4), have been used to design a pilot unit for the delay of xenon released in a nuclear power station. This unit consists of two columns 0.5 m in diameter and 5 m in height interconnected for parallel or in series operation. The unit has been installed on the off-gas system of the pilot nuclear power station BR-3 (a PWR of 10 MWe) in Mol. After a few start-up difficulties, 6 months of effective operation with a close measuring campaign are yet realized. The statistical treatment of the experimental data obtained during the variations of gas flow rate, relative humidity and radioactive back-ground is underway.

IV. The Laboratory Set-Up for Iodine Sorbents

To check the performance of solid material on its iodine trapping capability a laboratory set-up working at a nominal flow rate of 1 m^3 h^{-1} is available. As illustrated in Figure 1, the carrier gas can be artificially loaded with ^{131}I traced iodine species in the absence or presence of humidity. The way these iodine compounds are trapped on a 5 cm diameter column of sorbents is determined by the measurement of the γ activity of the effluent gas. These measurements result in a complete break-trough curve of a long duration run or in an initial decontamination factor of a short run.

The ventilation line of the hood in which this laboratory set-up is mounted contains a filter case in which any commercial iodine filter can be tested.

This installation has been used in an intercomparison program organized by E.E.C. and remains operational for quality tests of various sorbents' lots.

V. The Integrated Gas Purification Test Loop

Several years ago, the S.C.K./C.E.N. started the construction of an integrated gas purification test loop for dissolver off-gases. This loop whose flow-sheet is schematized in Figure 2 has a nominal through-put of 25 m^3 h^{-1} and has been designed as a demonstration rig equivalent to a reprocessing capacity of 1 ton a day, hence his name "GAS-TON." At the moment, the various processes applied in this loop have been developed separately. For instance, the Mercurex process has been investigated in the scrubbing columns filled with Rashig or Pall rings. Decontamination factors of 500 have been measured even when only iodomethane was used as iodine compound. The various data have been treated into an over-all mass transfer model which allows proper design for any inlet conditions.

Also a new demister made of stainless steel fiber has been developed. The data of the laboratory set-up for iodine sorbents were used to specify the silver sorbents for the removal of the last traces of iodine compounds. The commissioning of these surbent beds put in series after the mercurex scrubber indicated a guaranteed decontamination factor of 10,000 for any iodine species present. The selective NO-removal by catalytic reduction with ammonia will be checked this month as mentioned in the paper submitted to this conference. The low dewpoint drying is ready for operation at a working pressure of 8 bar.

The cryogenic distillation unit could be mastered in the spring of this year. A decontamination factor of 1,000 for the krypton present in the carrier gas at a level of 100 ppm can be guaranteed in industrial conditions.

In the future, it is aimed to control the promising results obtained in the separate units. Therefore, long duration tests will be started this year at the nominal through-put of 25 m^3 h^{-1} while the carrier gas is artificially loaded with nitrous vapors, iodine species, krypton and xenon.

VI. Test Rigs for the Removal of Particulates

In the past, the trapping of dust on a layer of sand has been investigated in a laboratory set-up and in a technical unit with a filtering area of 1 m^2.[5] The results obtained have been applied to design a high temperature dust filter for the waste incinerator under development at Mol. During the commissioning of this incinerator with a complete gas purification train it became clear that salt condensation phenomena occur in the off-gas line. In this way fume with a particulate

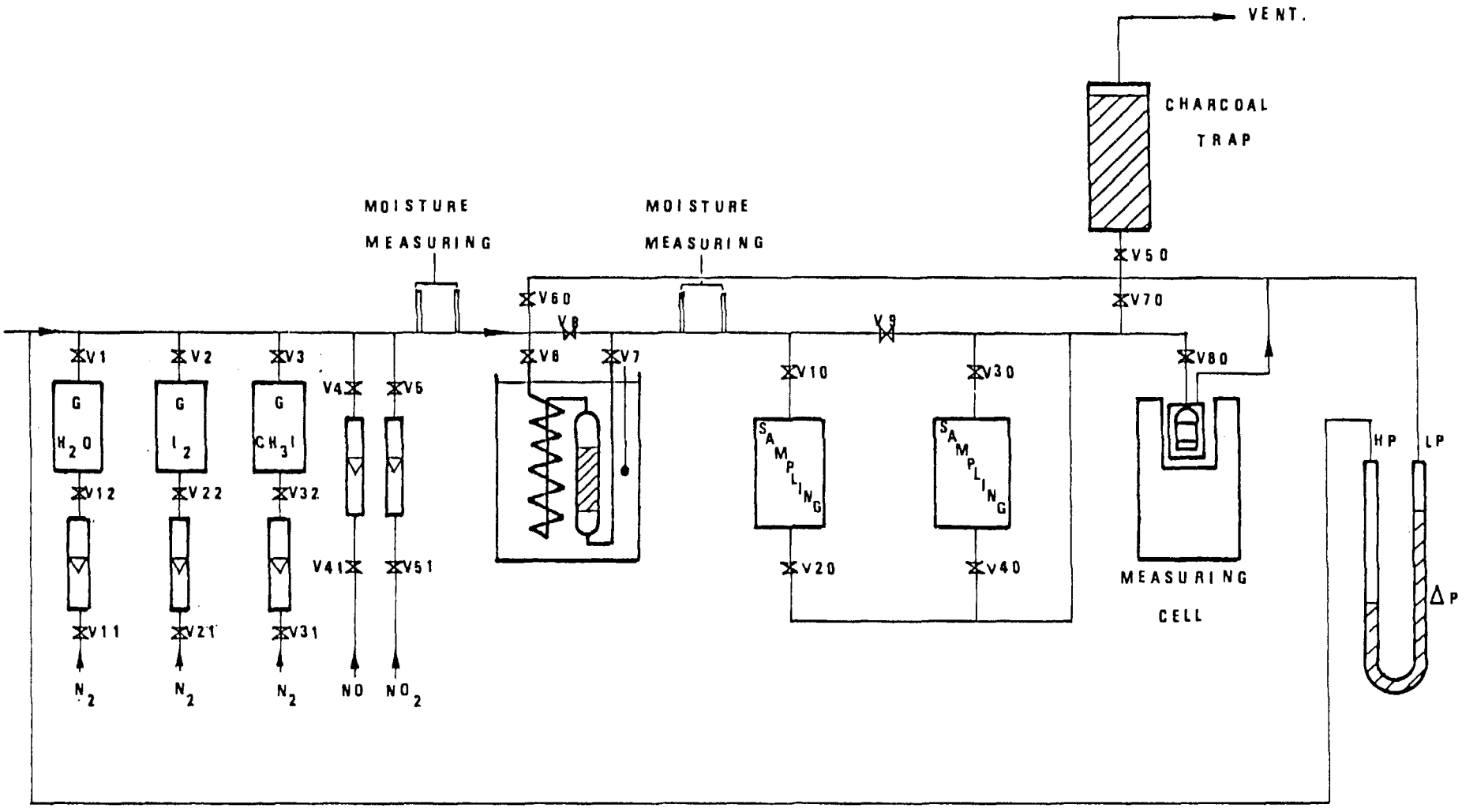


Fig. 1 : SCHEME OF 1 m^3 h^{-1} LOOP

Fig. 2 : INTEGRATED GAS PURIFICATION TEST LOOP
25 Nm^3h^{-1}

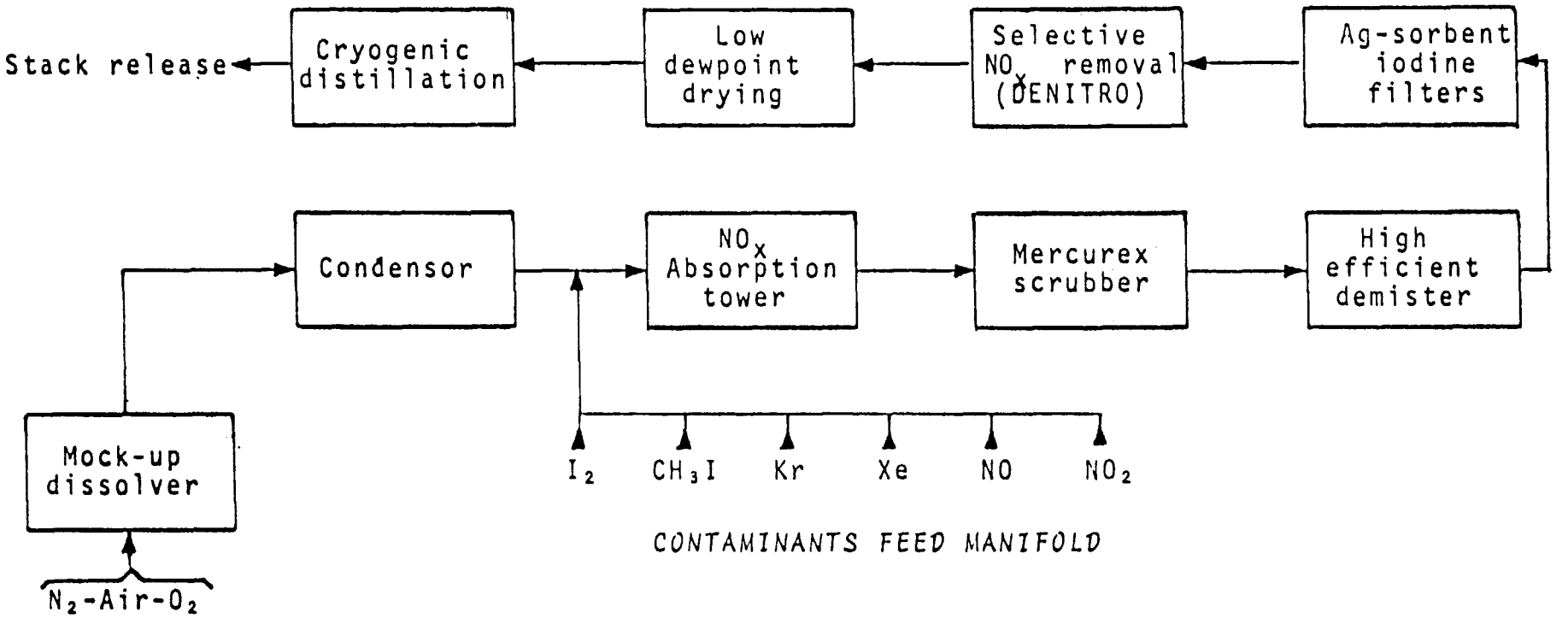
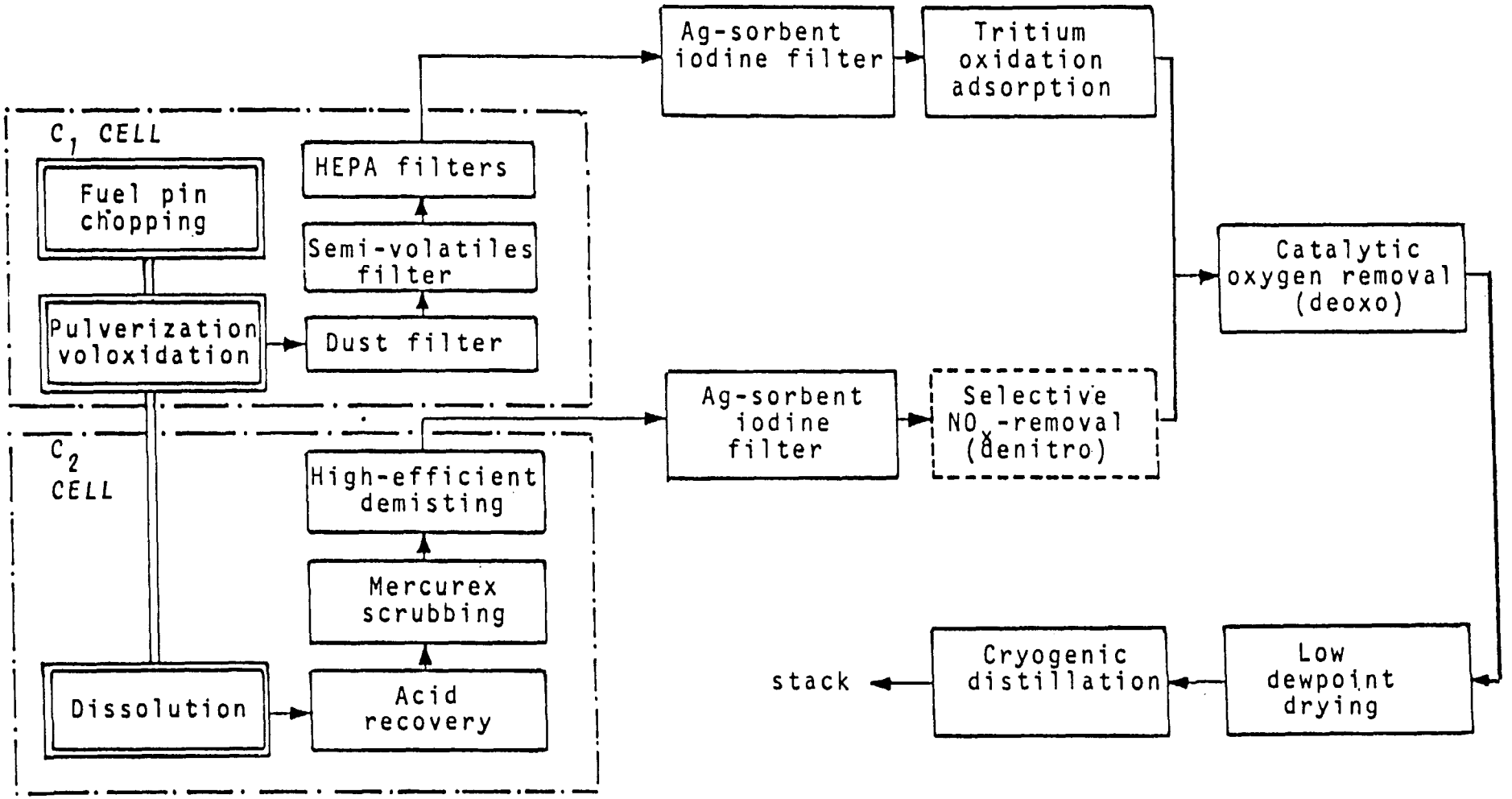


Fig. 3 : BASIC OFF-GAS PURIFICATION SCHEME (HERMES)



size of about 0.5 μm is formed. The trapping of this fume will be studied in a laboratory test rig now under construction with a nominal gas through put of 5 m^3 h^{-1}. It is planned to study also the behavior of semi-volatile ruthenium compounds in this test rig.

VII. The Experimental Program on Tritium

The catalytic oxidation of the elementary hydrogen on copper oxide catalysts has been studied in the past. This reaction is now applied together with the low dewpoint drying in the 15 m^3 h^{-1} safety trap behind a 1.4 kW electrolyser for tritiated water. This installation has just been put in operation and the measuring program will start in the near future. It is foreseen to expand this installation with a three phase chemical reactor in order to determine the isotopic exchange possibilities of various hydrophobic catalysts.

VIII. Conclusion

From the short survey given it can be concluded that S.C.K./C.E.N. at Mol is well prepared to investigate any nuclear gas cleaning problem that may arise in the future. A complete set of laboratory and technical units are available for this purpose. In the future, the Belgian technical possibilities in this field will increase further (see Figure 3) as soon as the Head-End Research facility on the Mockup Engineering Scale (HERMES) of 10 kg irradiated fuel per batch will be operational in 1982.

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DISCUSSION

R.A. BROWN: What do you plan to use as a secondary waste treatment for the Mercurex process?

BRUGGEMAN: We have tried to process the wastes from the Mercurex process, but without great success. At the moment, we are thinking about using copper instead of mercury for the process. We may abandon mercury because it causes a lot of difficulties in disposing of the secondary waste.

R. A. BROWN: Considering the technical problems associated with secondary treatment, have you evaluated the alternative of a solid sorbent system such as silver zeolite or silver nitrate impregnated silicic acid (AC 6120)?

BRUGGEMAN: We will use a solid sorbent after the Mercurex process. At the moment, there's a trend to use only the solid sorbent and maybe this is a good solution. The Mercurex process works very well, but for about a year we have worked on the secondary waste to recuperate the mercury and iodine and we have had a lot of difficulties. We will now try not to recuperate but to use copper instead of mercury.

Developments in Nuclear Air Cleaning in Germany

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I. New Filter Designs for Nuclear Power Plants

The recent developments of nuclear filters in the Federal Republic of Germany were prompted by the intention of a German nuclear power plant manufacturer to simplify the handling of contaminated nuclear filters and minimize the space requirement for the radioactive waste accumulating.

I.1 HEPA-Filters

Fig. 1 shows a HEPA filter element of 24 x 24" size, which can be used for volumetric flow rates up to 1770 cfm and temperatures up to 80°C. The resistance of the new HEPA filter is 250 Pa at rated air flow capacity.

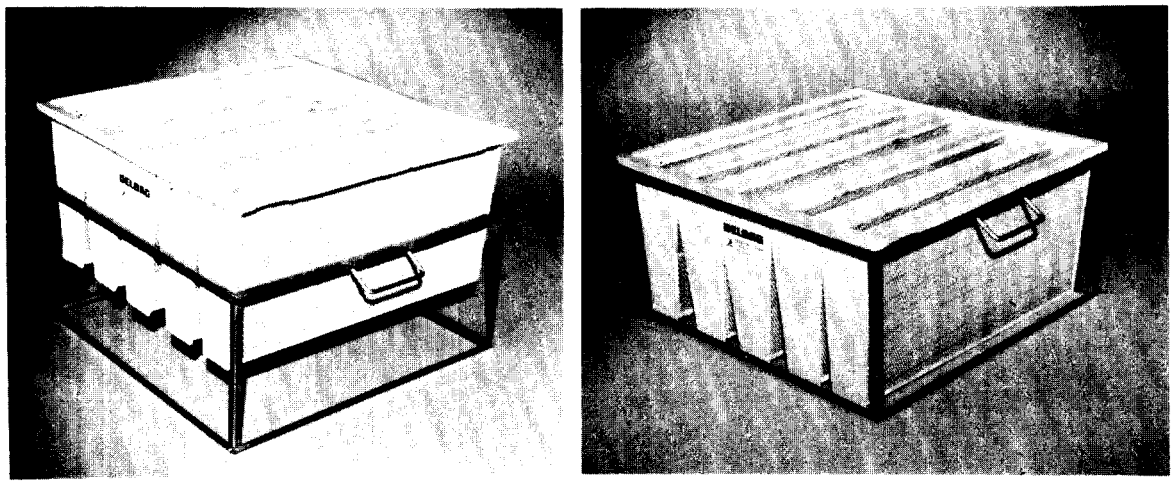

Fig. 1

The complete filter element consists of a rigid frame for transmitting the contact pressure forces and an exchangeable filter insert placed in the frame.

The exchangeable filter insert consists of various V-shaped filter bags which, including the sealing surfaces against the filter housing, form an exchangeable unit by means of a permanently resilient synthetic material. The filter papers in the V-shaped filter bags are provided with a protection handle and, at the same time, offer the advantage of reinforcing the filter element under air loading conditions.

When changing the filter elements it is not necessary any longer to change the complete filter element, but only the exchangeable filter insert. The rigid frame remains in the filter housing. The latter is reusable and takes up the new filter inserts after changing.

The exchangeability of the filter insert permits that only by manual pressing the outer shape of the filter can be adjusted to the inner shape of the drum without crushing the filter in advance. It is possible to reduce the volume of the filter insert in the drum to a third by using a suitable press under vacuum conditions. This will allow the storage of several filter inserts in one drum of 200 liters (Figs. 2 - 5.

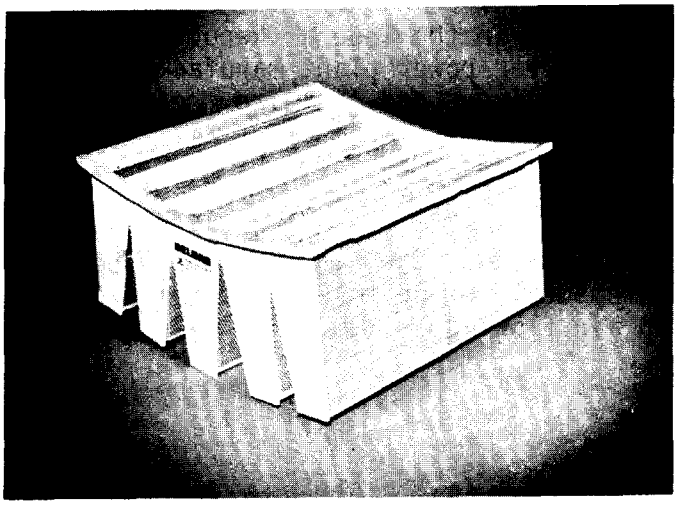

Fig. 2

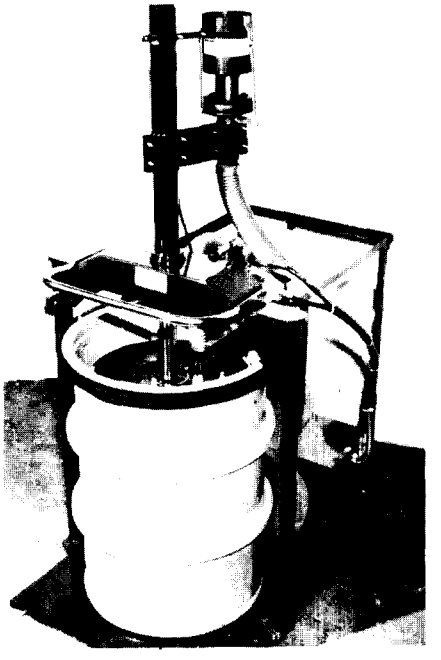

Fig. 3

Fig. 4

Fig. 5

Another recent development is a filter element which consists of a standard 200 l waste drum with integrated HEPA filters. The drum at the same time acts as a filter housing. The special design of the filter elements and the sealing relative to the wall of the drum at the diagonally opposed edges splits the gas stream into unfiltered and filtered gas streams within the drum. The maximum volume flow through such a HEPA filter may be up to 5000 m^3/h. The filter is connected directly to the ventilation ducts. When replaced, the drum with the integrated filters is disengaged and sealed. This work can easily be performed by remote handling equipment.

I.2 Iodine Sorption Filters

Because of the high contents of filter pollutants in nuclear power plants [1] and the resultant loss of removal efficiency of radioiodine filters the iodine sorption material, in German nuclear power plants presently only impregnated activated carbon, must be replaced at relatively short intervals. Aside from the consumption of high grade filter carbon this produces considerable amounts of contaminated waste whose disposal requires expenditures by far exceeding the costs of filter carbon proper.

Since the usual iodine sorption filters in nuclear power plants are not loaded with major quantities of radioiodine, the loss of removal efficiency is practically all due to loading of the activated carbon with pollutants and to aging and weathering phenomena. Activated carbon in an iodine sorption filter protected from pollutant loads by a pre-filter will largely retain its loading capacity relative to other, more easily adsorbed materials for the time of its use in an iodine sorption section of a Multiway Sorption Filter (MWS filter). Hence, it is obvious to reuse this carbon, after removal from the iodine sorption section, in an activated carbon pre-filter for pollutant removal.

In order to avoid refilling of the contaminated activated carbon a filter housing has been developed in which the activated carbon is contained in a shaft type filter chamber through which the exhaust air stream to be cleaned can be passed many times as a result of deflection (Fig. 4).

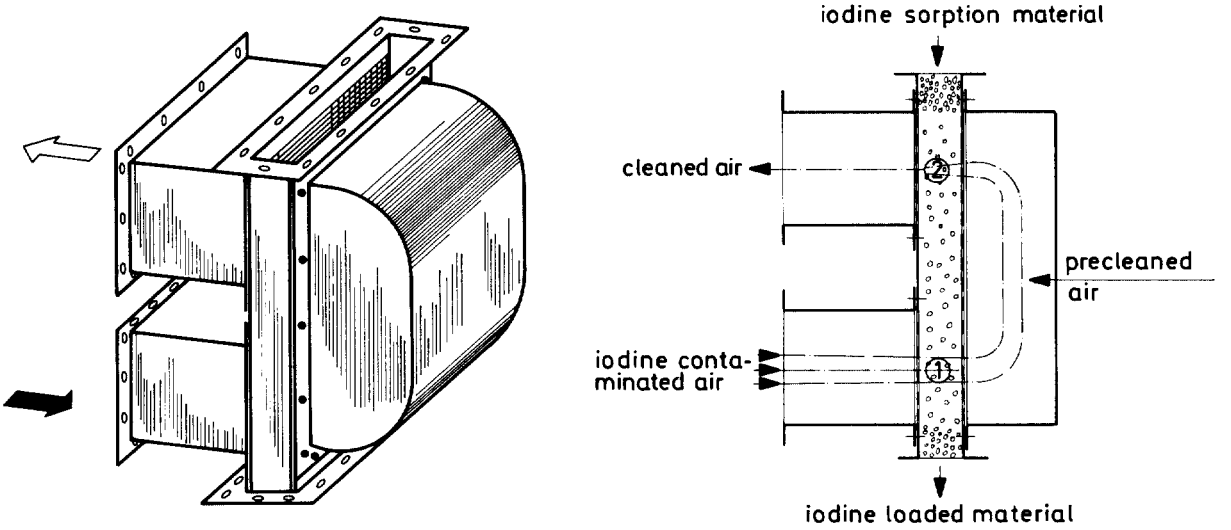

Fig. 4 Multiway iodine sorption filter

The activated carbon contained in the lower part of the filter chamber is used for pre-adsorption of pollutants. The upper part of the filter chamber represents the iodine sorption filter proper. After a the first operating period in which the removal efficiency of the activated carbon in the upper section of the filter chamber has dropped to the lowest acceptable level, the carbon is moved from the top into the lower part of the chamber where it serves for pre-adsorption of pollutants.

This technique is based on a plane parallel withdrawal of the carbon and a suitable flow distribution in the filter. These characteristics have been verified by experiments in a prototype filter for nuclear power plants. Presently two nuclear power stations are equipped with the MWS filter described above. The volume flows to be filtered amount to 53,000 and 2 x 10,300 cfm, respectively.

In the MWS filter the depth of the filter bed is not influenced by pebble bed and withdrawal cones; complete utilization of the sorption material can be achieved together with maintaining a given removal efficiency. Mechanical leakages can be excluded with high reliability. Moreover, a design can be realized in which there is only a relatively slight pressure drop over the filter layer.

II. Testing Iodine Sorption Filters

In the Federal Republic of Germany, iodine sorption filters installed to control the consequences of accidents must be examined for their removal efficiencies prior to startup of the respective nuclear power plant and at regular intervals. These checkups are increasingly becoming the rule also for iodine sorption filters used for exhaust air cleanup in normal operation of a nuclear power plant. As a rule, the examination includes an in-place test and testing of activated carbon samples in the laboratory at specific time intervals. Both the in-place test and the laboratory examination have so far been carried out with radioactively labeled methyl iodine as the test medium. Now that iodine species samplers [2] have been developed which allow a clear distinction to be made between elemental iodine and organically bound iodine (methyl iodide, as a rule) it is possible, provided that there is a sufficient concentration of ^{131}I in the filter intake air, to measure iodine sorption filters used in normal operation of a nuclear power station without additional feeding of a radioactive test agent directly by way of their removal behavior relative to these iodine components. For an activity concentration of 10^{-12} Ci/m^3 in the filter intake air a decontamination factor of 1000 can be proved by means of a solid state detector for sample counting. The measured results obtained are always much more precise than measurements of activated carbon samples in the laboratory, because the true loading condition of the activated carbon with pollutants during the measuring period remains unchanged; there is no need for conditioning of the activated carbon in respect of the uptake of water vapor prior to determining the removal efficiency.

III. Off gas Treatment in Reprocessing Plants

III.1 Reprocessing Plants for LWR Fuel

The treatment of the dissolver off gases of these plants is being studied in a comprehensive program ranging from laboratory experiments to cold and hot operation of test facilities on a technical scale. In the sequence of treatment components listed here the following systems are being developed and tested at the Karlsruhe Nuclear Research Center:

- Iodine vaporisation from the feed solution and from the recovered acid (HET test rig, flow 50 m^3/h at STP, cold and tracer trial operations).

- Separation of large and fine droplets, removal of the bulk quantity of iodine accumulated in the solid AC 6120 iodine sorption material and removal of particulates in HEPA filters (PASSAT test rig [3] with 1 : 1 components, flow 250 m^3/h at STP, cold and tracer trial operation, tests of remote handling systems).
- Removal of krypton [4] including the separation of O_2 and residual NO_x by catalytic reduction with H_2, adsorptive retention of H_2O, CO_2, NH_3 etc., and cryogenic distillation in two columns, first the N_2-Kr-Xe three-component mixture, then the Kr-Xe two-component system (cold pilot plants for gas flows of 50 m^3/h at STP called ADAMO and KRETA).

A program for studies of the final storage of the Kr product has also been started. This includes corrosion tests of various steels by Rb, a filling station built to minimize Kr losses and allow safe handling of the fillied steel cylinders, a concept of final storage and alternative storage methods.

At the Karlsruhe reprocessing pilot plant (WAK) a facility will be built for testing the newly developed removal components for the dissolver off gas under hot conditions.

III.2 Reprocessing of HTR Fuel

At the Jülich Nuclear Research Center systems are being developed for off gas cleaning of the head end of an HTR fuel reprocessing plant [5]. This includes

- burning the graphite in a fluidized bed combustion reactor. For off gas cleaning a series connection of two different centrifugal force dust collectors is being tested:

  one cyclone for particles with diameters $\geq$ 5 µm.

  one rotational flow centrifugal separator for particles $\geq$ 1.8 µm.

  Improvements are to achieve removal down to particle diameters of approx. 0.6 µm.
- the removal of aerosols, I and Kr from the burner off gas (AKUT II test facility with a flow of 10 m^3/h at STP). Aerosols are removed in an electrostatic precipitator with downstream HEPA filters; tritium, which is present as tritiated water, is adsorbed onto molecular sieves.

  CO is transformed into CO_2 by oxidation with oxygen on a palladium catalyst. The off gas consisting of CO_2, 0.25 % O_2 and the fission noble gases, is compressed and liquefied. Kr is separated from the bulk CO_2 by distillation. The distillation column is designed for a decontamination factor of 1000 and a product of 0.1 % Kr in CO_2.
- removal of the Xe and Kr noble gases from the dissolver off gas. Two cryogenic separation units have been developed:

  (1) the KRYOSEP I facility (flow 5 m^3/h at STP) equipped with demister, HEPA filters, iodine filters, O_2 and NO_x removal by catalytic reaction with H_2, and removal of the water by means of molecular sieves. In a subsequent cryogenic separation unit Xe is frozen out and separated from residual Kr by distillation. The liquefied N_2-Ar-Kr mixture is distilled and the Kr-rich fraction is removed and filled in pressurized cylinders;

  (2) the KRYOSEP II facility (flow 1 m^3/h at STP) for the removal of Kr and Xe from the off gas of a continuous dissolver with a closed loop helium purge gas cycle. In this facility Xe and Kr are frozen out separately after the same preliminary treatment as described under (1) above. Xe is cleaned of the residual Kr in a batch distillation column.

References

(1) J. Furrer, R. Kaempffer, J.G. Wilhelm, Ageing and poisoning of iodine filters in nuclear power plants, Kerntechnik Vol. 18, No. 7 (1976).

(2) H. Deuber, J.G. Wilhelm, Determination of the Physico-Chemical ^{131}I Species in the Exhausts and Stack Effluent of PWR Power Plants, paper 6-7 of this conference.

(3) J. Furrer, J.G. Wilhelm, K. Jannakos, Aerosol and Iodine Removal System for the Dissolver Off Gas in a Large Fuel Reprocessing Plant, paper 6-2 of this conference.

(4) R.v. Ammon, H.G. Burkhardt, E. Hutter and G. Neffe, Development of a Cryogenic Krypton-Separation System for the Off Gas of Reprocessing Plants, paper 7-2 of this conference.

(5) H. Barnert-Wiemer, M. Heidendael, E. Merz, H. Vygen and G. Schröder, paper 6-1 of this conference.

DISCUSSION

BURCHSTED: The Delbag filter concept is interesting. I would like to know what the air flow rating is; what the medium is made of, its thickness and area; and how much pressure drop it can withstand without rupture.

WILHELM: I think I will transfer all these questions to the manufacturer, Dr. Hayn.

HAYN: The normal airflow rating is 1,000 to 2,500 cfm. The normal pressure drop is 1 in.w.g.

BURCHSTED: And the area of the medium?

HAYN: 8 sq. meters.

BURCHSTED: Is the medium fiberglass?

HAYN: Yes.

BURCHSTED: What is its thickness?

HAYN: The thickness is 0.015 in.

BURCHSTED: What is the maximum pressure drop in normal service?

HAYN: Maximum pressure drop in normal service is 4 in. w.g. but in Europe and especially in Germany we have to change the filters after three years whether the pressure drop has reached 4 in. w.g. or not.

BURCHSTED: We do the same thing in many places. How much can it withstand before rupture?

HAYN: At least 10 in. w.g.

GEER: How is this filter sealed to the housing seal-surface?

HAYN: The filter consists of a number of banks of micro-pleated fiberglass paper sealed into an elastomeric casing which forms several V-cross-section "major" pleats. The upper section of this assembly is contiguous with the sealing flange of the filter. A neoprene gasket is glued to the sealing flange, similar to conventional U.S. filters, and forms the seal between filter and housing. The flange of the steel support frame fits under the filter flange. Bolts (4 or 8) clamp the support frame to the housing, or a pressure frame (similar to one used in a caisson) may be employed.

BROWN: What do you plan to do with the large quantities of krypton that you would recover from a 1400 metric ton per year operation?

WILHELM: When we have a large reprocessing plant, I think we will put it into pressurized steel containers.

BROWN: As a temporary measure? Or do you think that will be your chosen method of storage?

WILHELM: There is thought to fill the inside of the cylinder with a sorbent which will take up krypton and lower the vapor pressure, but at this time, we have not made any decision what it will be.

BROWN: Over the lifetime of a reprocessing plant, 30-40 years, you may collect more than 200,000,000 curies of krypton in one place.

WILHELM: We know this. We expect to have an engineered storage facility. That is the idea we have at the moment.

BROWN: Do you have any plans for tritium recovery?

WILHELM: The tritium activity in the offgas will be controlled by reducing offgas temperature below its dew point. Cooling the offgas to 5°C or below is intended. The temperature required will be determined by calculating the total amount of tritium that will be allowed by the German radiation protection commission for a large reprocessing plant. We will recirculate the water as we recirculate the acid and, by this means, try to have very low offgas rates. No final decision has been made for tritium recovery.

BROWN: If you recirculate the acids, won't there be a build-up of other chemical and radioactive components to interfere with other parts of the process?

WILHELM: This is under investigation.

DEMPSEY: Have you checked the new *in situ* charcoal test method, described earlier by Dr. Deuber, against your standard method wherein you release substantial quantities of iodine? I am enthralled with Dr. Deuber's method as it seems to me to be a very valid way to go. But I'm wondering if you have checked it by your standard technique.

WILHELM: Yes, *in situ*, 95% removal efficiency was measured. The standard test (with charcoal from a bypass test canister) showed 90% removal efficiency in the laboratory test under the same conditions. But I would not give too much credit to the test canister method because the charcoal from the test canister has to be preconditioned in the test apparatus to the relative humidity of the air and, when preconditioning with a polar medium like water, the solvent gets driven off.

I can give you the results from a test of a pressurized water reactor. It was one of the filters for the offgas from the equipment rooms. They run under 40°C and around 70% RH. The test inside the plant, performed 4 weeks after the canister test, showed penetration of 10% methyl iodide. This was a second unit. The first with the test canister showed 95%, which is not too bad. We have given the figures in the talk by Dr. Deuber. With respect to testing accuracy, I would absolutely rely on the test inside the plant with a real iodine species under real conditions with the original packed filter itself, and not on a test canister. I feel that is better by far, because you can only test a filter when it operates under normal conditions. You could not apply the same results to a filter used after an accident because the filter would see high temperature and high relative humidity, which you don't have during a test. So the results apply only to the continuously working filter units we have for the exhaust air of equipment and operating compartments of our pressurized water reactors.

DEMPSEY: And you still test them in the laboratory for accident conditions?

WILHELM: Yes, because it is a new method we are trying to standardize. I have spoken to the licensing people and it is absolutely perfect. It seems that they agree with this method.

HAAG: I noticed that you failed to comment on carbon-14 releases in Germany in the reprocessing of LWR and HTR fuel. I would like to refer you to our work in which packed beds of $Ba(OH)_2.H_2O$ have been found capable of CO_2 removal from dilute and high CO_2 content feed streams. What is the present and future status of reprocessing nuclear fuel in Germany?

WILHELM: In the offgas cleaning process developed for a large reprocessing plant, CO_2 will be adsorbed on molecular sieves and, after desorption, may be finally transformed into a carbonate. No final decision is made for the total release of C-14 from the reprocessing plant. At the moment, a release of 1000 Ci per year of C-14 is assumed, but we asked for more research and development. Until the plant is built, research shall continue and we will have space to bring in a unit in case one is available at that time. In a cryogenic system, carbon-14 will remain in the form of CO_2 in the molecular sieves used for the drying of the offgases. We plan to strip it from the molecular sieves and wash it out with barium hydroxide or something like that. This is the intention of the moment.

IAEA/NEA ACTIVITIES IN THE FIELD OF GASEOUS WASTES

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Introduction

The protection of man and his environment from harmful concentrations of radionuclides has been the primary objective of radioactive waste management activities since the beginning of the nuclear industry. The treatment of gaseous waste produced by nuclear facilities is a key safety element during both normal operation and under accident conditions. The design and operation of off-gas cleaning systems are therefore extremely important, notably in view of the possibility of requirements to remove and retain radioiodine, krypton-85, tritium, carbon-14 and other long-lived radionuclides from nuclear facilities.

In this connection, a considerable expansion of national activities concerning the radioactive gaseous effluents control has taken place over the past several years in the countries that are using nuclear power or embarking on nuclear programmes.

IAEA and NEA Activities and Programmes in the field of Gaseous Waste Management

Questions concerning gaseous radionuclides releases and their retention have become international in scope and are now a matter of international co-operation. The International Atomic Energy Agency (IAEA) and the OECD Nuclear Energy Agency (NEA) have established programmes in this area. In order to avoid overlapping and duplication in international activities, these programmes have been discussed and to a certain extent co-ordinated.

In August 1976, the NEA Group of National Nominees for the Exchange of Information on Gaseous Wastes, in which the IAEA was also represented, recommended that the following questions be investigated:

(a) Sampling and measurement technologies of off-gases from nuclear facilities.

(b) Conditions, methods and installations for testing aerosol filters.

The first question was covered in the enquiry on the "state-of-the-art" of sampling and measurement technologies, which has been set up jointly by the two Agencies. The results of this enquiry are discussed below (see Section following).

The enquiry on conditions, methods and installations for testing aerosol filters carried out by NEA showed that the test methods used in different countries were defined for normal conditions of use. Their reliability was not well known outside their normal environment. Certain methods could be chosen for a specific parameter. There did not seem to exist a test method that could be used for the study of the combined effects of the test parameters.

Developing the results of this enquiry, the IAEA launched a co-ordinated research programme on the comparison of existing aerosol filters testing methods. As a result of this programme, standardisation of testing procedure is envisaged.

The NEA Committee on Radiation Protection and Public Health is preparing a study on Effluent Releases from the Nuclear Fuel Cycle. The purpose of the study is to develop criteria and mechanisms to optimise limitation of effluent releases into the environment in relation to the four most important radionuclides at regional and worldwide levels: tritium, carbon-14, krypton-85, and iodine 129, based on the new ICRP-26 recommendations, and to offer recommendations for the policy objectives required in relation to the possible future retention of the radionuclides.

IAEA is taking part in the preparation of this study. It submitted to the NEA Radioactive Waste Management Committee the recent IAEA publication "Removal, Storage and Disposal of Gaseous Radionuclides from Airborne Effluents".

Finally, a joint IAEA/NEA Symposium on the Management of Gaseous Effluents from the Nuclear Fuel Cycle is scheduled for 1980.

The Enquiry on the State-of-the-Art in the Field of Sampling and Measurements of Off-Gases from Nuclear Facilities

In May 1977, NEA in co-operation with the IAEA undertook an enquiry on the state-of-the-art in the field of sampling and measurement of off-gases from nuclear facilities. The enquiry was intended to cover both occasional and on-line sampling and measurement techniques at the stack or other points of the specified installations.

1. Objective

The present document summarises the results of the enquiry in order to define what the actual problems are. It aims at getting a comprehensive study on currently available and acceptable techniques and possible further developments.

In the last few years the measuring and monitoring requirements for different types of nuclear facilities have changed in different countries as a result of changes or anticipated changes in radioactive release rate requirements. The types of effluent information required for assessing the environmental impact of certain airborne releases are also different and the effluent measurement requirements vary depending on the process involved. At the present time, airborne effluent measuring and monitoring "state-of-the-art" in these industries is not well defined. Because this information is necessary for design of new facilities and for regulatory considerations, clarification of available technology and the identification of limitations are needed.

This paper describes some of the present and planned radioactive airborne measuring and monitoring practices in some IAEA/NEA Member countries' nuclear facilities. Special attention has been given to taking into account the behaviour of air cleaning systems under accidental conditions, as this was of particular importance as demonstrated notably during the Browns Ferry Fire. This information is the basis for defining applicable "state-of-the-art" methods, advanced technology, and development needs.

2. Scope

Measuring and monitoring systems and practices are described for power reactors, reprocessing, waste solidification, uranium and plutonium fabrication facilities and for incinerator-compactor waste facilities. This paper is based on the information received from Argentina, Australia, Belgium, Canada, Czechoslovakia, Federal Republic of Germany, Hungary, India, Iran, Italy, Japan, Netherlands, Poland, Sweden, Switzerland, United Kingdom and United States.

Operating experience and problems of monitoring systems at these facilities is outlined and some future development requirements defined. As some answers provided new developments in monitoring technology during the last few years, these have been included.

3. Requirements

System designs are determined by three factors:

(a) Government regulations and guidelines;

(b) Process control needs;

(c) Effluent stream characteristics.

Government regulations and guidelines. Various regulatory guides, national standards and regulations exist in different countries, but these are not discussed or summarised in this document.

Process control needs. Another factor in a monitoring system design is process control requirements which are determined by the

choice of monitoring points. Monitoring instrumentation will indicate how well a plant's off-gas treatment process is functioning.

Off-gas stream characteristics. In order to determine monitoring requirements in nuclear facilities, it is necessary to characterise the various effluent off-gases. Rough estimates of concentrations of gaseous species and particulates are made. Such estimates indicate the required levels of sensitivity and possible sources of interference.

4. Description of Present Systems

Information on monitoring of off-gas systems for different types of nuclear facility received from all countries was summarised and analysed. Some negative aspects of present monitoring systems were found as follows:

(a) Representative sampling rarely occurs due to non-homogeneous gas streams, non-isokinetic sampling, and line losses due to plate-out, adsorbtion, impaction, temperature changes, proper location of probes, etc.

(b) Monitoring of low-energy beta emitters are difficult to detect when high energy radiation from other radionuclides is present.

(c) Real-time monitoring is necessary in order to allow operating personnel to minimise abnormal releases of radioactive effluents. No facility surveyed has continuous I-129, H-3, Ru-106 or C-14 monitors. Continuous particulate monitors are generally gross monitors.

(d) Alarm monitors are not able to detect an increase of "x" counts per minute immediately after installation of a new sample collector, but this same "x" counts per minute could trigger the alarm monitors just before removal by the collector.

(e) The reliability of monitoring data is difficult to evaluate due to the numerous possible interferences in the complex gas streams. Calibration of monitoring equipment with pure radionuclides only adds to the difficulties as it does not take into account interferences from other components of the off-gas stream. Monitoring reliability under abnormal conditions is also not fully understood.

(f) The reliability and sensitivity of Kr-85 monitors in some complex and corrosive environments is unknown.

(g) The problem in particulate monitoring is essentially that of representative sampling. In addition, sampling filters have varying efficiencies, depending on particle size, gas stream conditions, flow rates, etc.

5. "State-of-the-Art"

The "state-of-the-art" monitoring outlines the different

practices relating to sampling and measurement technologies for different radioisotopes. The following remarks can be considered after analysing the data received:

(a) The results of the enquiry are complete enough to permit an approximation of the "state-of-the-art" monitoring of off-gases for different radioisotopes as well as those isotopes that are currently measured at different types of facilities.

(b) The different monitoring techniques cannot be compared without making specific value judgements on sensitivity, range or specificity of the method and these criteria may not be applicable to a specific type of installation.

Therefore, Tables I-VI give a list of the "state-of-the-art" monitoring techniques for the most currently measured radioisotopes, that is tritium, carbon-14, noble gases, iodine-129, iodine-131, particulates. Each table contains information on the sampling method and measurement technology.

It has been noted that representative sampling can be best achieved if the following methods are taken into consideration:

(a) Design of sampling systems should be made before construction of facilities.

(b) Use of multiple isokinetic samples probes.

(c) To avoid condensation problems, use of heated sample lines, collectors and adsorbers, and filters.

6. Advanced Monitoring Methods

As some countries provided new developments in monitoring technology during the last few years, these have been included in Table VII for different radioisotopes.

The elimination of some problems related to representative sampling is accomplished by direct analysis of the entire off-gas stream. This would, of course, require development of appropriate monitoring technology.

7. Research and Development Recommendations

Proper design of monitoring systems requires a knowledge of the species and concentrations in an off-gas as well as possible sources of interference. The characteristics of off-gas effluents will change, depending on the type of treatment system. The magnitude of the changes is in many cases unknown (voloxidation, fluorocarbon adsorption and rapid cycle adsorption).

Technical R & D. An adequate continuous tritium monitor for reprocessing plants is not available. It is recommended that laser and other tritium monitoring methods be investigated.

A continuous I-129 monitor is needed to monitor iodine adsorber

TABLE I

State-of-the-Art Sampling and Measurement Techniques for Tritium

Sampling	Measurement
1. Collection of sample as tritiated water on a molecular sieve, silica gel, or other adsorbent.	1. Liquid scintillation counting in the laboratory.
2. Liquid condensate from off-gas with separation from interfering radionuclides.	2. Anticoincidence counting.
3. Bubbling off-gas in water column.	
4. Oxidation unit and cold trap.	

TABLE II

State-of-the-Art Sampling and Measurement Techniques for Carbon-14

Sampling	Measurement
1. Collection in basic solution and adsorption by ascarite or a molecular sieve and separation from interfering radionuclides.	1. Liquid scintillation analysis in laboratory.

TABLE III

State-of-the-Art Sampling and Measurement Techniques for Noble-Gases

Sampling	Measurement
1. Flow-thru flask, chamber or vessel.	1. Gamma spectrometry with sodium iodine or Ge-Li detectors with single and multi-channel analysers and rate meters. 2. Proportional counters and Geiger Muller tubes. 3. Plastic scintillator. 4. Ionization chamber.

TABLE IV

State-of-the-Art Sampling and Measurement Techniques for Iodine-129

Sampling	Measurement
1. Adsorbtion of iodine on silver zeolite, charcoal, silver impregnated material.	1. Liquid scintillation counting. 2. Neutron activation analysis. 3. X-ray spectrometry.

TABLE V

State-of-the-Art Sampling and Measurement Techniques for Iodine-131

Sampling	Measurement
1. Adsorbtion on charcoal or silver zeolite filters or cartridge.	1. Sodium iodine and germanium lithium gamma detectors with single or multichannel analysers.
2. Continuous flow chamber.	2. Geiger Muller detector.

TABLE VI

State-of-the-Art Sampling and Measurement Techniques for Particulates

Sampling	Measurement
1. Continuous or stepwise moving filter.	1. Sodium iodine detector with multichannel analyser for gamma spectrometry.
2. Glass fiber and charcoal filter.	2. Zinc sulfide coated plastic scintillator for alpha monitoring.
3. Millipore and cellulose filters.	3. Geiger Muller counter.
4. Polyethylene absorbers.**	4. Gross alpha, beta, gamma analysis.
	5. Germanium-lithium spectrometry.
	6. P-M tube (Cerenkov-light).*
* for Sr-90	
** for Ru-106	7. Barrier layer alpha detector.

TABLE VII

Advanced Monitoring Methods for Different Radioisotopes

Isotope	Monitoring methods
Tritium	1) Differential monitor 2) Condensation followed by liquid scintillation counting 3) Water or ethylene glycol bubbler and large surface area plastic scintillator (continuous) 4) Possible laser monitoring techniques
Carbon-14	1) Possible laser monitoring techniques 2) Adsorption in a liquid media followed by liquid scintillation counting
Krypton-85	1) Silicon surface barrier detector after removal of interfering radioisotopes 2) Laser monitoring possible if ultraviolet lasers become available
Iodine-129 Iodine-131	1) Laser induced fluorescence 2) Intracavity absorption laser spectroscopy 3) Molecular beam quadrupole mass spectrometry
Particulates	1) Surface ionization mass spectroscopy 2) Annular impactor for alpha particulates 3) "AIRTRACE" laser techniques 4) Moving filter tape monitors for transuranic particulates

"Ramp alarm monitors

performance and for alarm purposes for use at a "hot" plant.

Continuous particulate monitors with the necessary sensitivity and background discrimination are needed, especially for alpha-transuranium monitoring. A complete study of size/nuclide distribution of particulate is recommended to properly define monitoring system requirements. An investigation of the molecular structure of ruthenium compounds in off-gases would be desirable for monitoring purposes as well as off-gas treatment system design.

The development of alarm monitoring instrumentation incorporating "ramp" type alarms is recommended.

It is recommended that any instrumentation developed in the laboratory be tested under conditions duplicating those in the field as closely as possible.

General R & D. There seems to be a need for an international standard guide treating the complete sampling and measuring station for any radioactive component. Technically this would result in the development of an integrated stack monitoring system that could function satisfactorily under all and for all off-gases of interest.

8. Conclusions

The ultimate approach to the various problems of monitoring is continuous real-time measurement of all individual nuclides in an effluent by direct analysis. This is not feasible at present, and does not appear to be in the foreseeable future. Therefore, the approach to monitoring should be based on the following points:

(a) Three types of monitor are actually needed - inventory monitors, alarm monitors, and process control monitors.

(b) The particular nuclides that should be monitored, and the methods for monitoring them, should be mentioned in the regulations.

(c) Specific removal devices should be continuously monitored at the clean-up device for process control and alarm purposes.

(d) Inventory monitoring at present is most reliably accomplished with fixed collectors which are removed periodically and brought to a laboratory for analysis.

CLOSING REMARKS OF SESSION CHAIRMAN:

We have heard interesting presentations on recent developments in air cleaning technology in Europe. Although there were similarities in the reports of the work underway, the paper from Belgium was unique in that it included a review of the process by which officials, on the basis of permissible environmental radionuclide levels and attainable effluent decontamination factors, have set a maximum for the quantity of electricity that can be generated in that country by nuclear power. This would appear to be an interesting exercise to conduct worldwide.

In a general sense, the papers in this session addressed three areas of technological development. First was the matter of the increasing emphasis on air cleaning systems that can be operated for longer periods of time. In this regard, the use of prefilters to protect the primary air cleaning units was emphasized. Also stressed in these programs was the use of components that can be maintained with a minimum of occupational exposure and which can be disposed of more readily. Second was the effort to develop facilities for improved testing of air cleaning systems under a wider range of parameters (temperature, pressure, humidity). This includes the application of a range of test aerosols, and evaluations of removal efficiencies for specific radionuclides in specific chemical forms at concentrations normally anticipated in operating facilities. Third was the increasing attention being directed to the long range need for the removal and retention of selected radionuclides such as ^{14}C, ^{129}I, ^{3}H, and ^{85}Kr. This last effort is being addressed primarily to air cleaning problems associated with spent fuel chemical processing plants. This is an area in which scientists in the European countries are moving forward rapidly and I believe it is clear to all that they are ahead of similar research and technological developments within the U.S.

Although the pressure of time has made it mandatory that I keep my summary comments brief, I would not want to close without thanking our speakers for a most interesting session. Individually, and collectively, their presentations were excellent.