Session 18

NEW AIR CLEANING TECHNOLOGY FROM EUROPE

WEDNESDAY: October 22, 1980
CHAIRMAN: H.J. Ettinger
Los Alamos Scientific Laboratory

OPENING REMARKS OF SESSION CHAIRMAN:

We plan to hear from each of the speakers on the panel without interruptions from the audience. This will take a little over an hour. Then the session will be open for questions from the audience and an exchange between members of the panel. To minimize interruptions, I would like to introduce each of the individuals at the start in the order shown on the program. We have one substitute. Mr. Dupoux is ill and is not here. Mr. Rouyer, from the French Atomic Energy Commission, is filling in for him and will talk to you about activities in France relative to air cleaning. Mr. Furrer, from the Nuclear Research Center at Karlsruhe, will talk about some of the things going on in Germany. Dr. Collard, from the Center for Nuclear Energy, will talk about current activities in Belgium. Milo Kabat is from Ontario Hydro in Canada and he will talk about activities there. Dr. Michael Marshall is from AERE, Harwell. He will talk to us about some of the activities in the United Kingdom. Mr. Olivier will talk to us about some the activities his organization is involved with that pertain to nuclear energy in Europe. He is from the Organization for Economic Cooperation and Development. Mr. Yoshida is with the Japan Atomic Energy Research Institute in Japan and he will describe recent advances in nuclear air cleaning in his country.

CHALLENGES FOR NUCLEAR AIR CLEANING TECHNOLOGY IN FRANCE

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I. Introduction

In this contribution from France, I shall try to describe the current evolution of nuclear air cleaning research and engineering and the technological developments needed for improving protection and safety.

I shall successively speak of :

- iodine and HEPA filters testing
- cleaning of reprocessing process gas
- plutonium aerosol filtration
- air cleaning in accident situations.

II. Iodine and HEPA filters testing

The TMI accident has claimed attention of our authorities for the necessity, for a good development of our nuclear program, to convince the public opinion that air cleaning systems have been thoroughly studied and tested for facing every conceivable situation.

This will had two consequences :

- on the research and development programs : the range of environments (temperature, R.H., ...) to which air cleaning systems were generally submitted has been enlarged.

As for iodine filter, we still go on testing modular units in our test facility at 2000 $m^3.h^{-1}$ with controlled temperatures and between 40 and 95 % R.H., but we are now installing a bench scale for charcoal testing up to 100°C-100 % R.H. and, for next year, we will start a facility that will allow us to submit filters to extreme environments.

For aerosol filters, we are completing a loop for submitting HEPA filters or prefilters to conditions generated by a fire.

- On officialization of our control action : the technical studies on ventilation and air cleaning for nuclear facilities generally modify the existing standards and regulations or create new ones. This type of action is being officialized by the creation of a Reference Technical Centre within which we will be developing these actions of control, standardization and information.

For example, studies to compare the DF_s of canisters and iodine filters placed in parallel, or safety studies for ventilation networks, or promotion of filter testing by a mobile loop like described in French papers here are in the field of this Reference Technical Centre.

III. Cleaning of reprocessing process gas

With the designers and the operators of reprocessing plants, we are involved in the following developments :

Demisters

In the application of the principles of cleaning gases as close as possible to the source and minimizing the wastes, we are trying to improve the efficiency of demisters for reprocessing process gases. We have developed a parallel plate demister which has a much better efficiency than the existing devices and which is easy to calibrate for different conditions of operations (gas flow rate changing in a wide range of values). The work concerning this demister is completed but it must be followed up new by the development of systems for increasing the size of aerosols before demisting. Several systems are envisioned depending on the location of the demister ; for example, one of them consists of venturis placed within the last plate of a scrubber column.

The total efficiency of the system (increasing the size of aerosols and demisting) is expected to be very good.

Iodine trapping

We are installing in our laboratory a full scale installation described last year by Paul SIGLI in the same panel session, with a AC 6120 Ag impregnated adsorbant and the tests will be completed by the beginning of next year.

In this field, the emphasis is also put on a better efficiency of the alkaline liquid scrubbing of the gas containing iodine and the conditionning of the wastes resulting from the treatment of alkaline solutions.

Ruthenium trapping

We are comparing different possibilities for a complementary trapping of ruthenium after liquid scrubbing and we intend actually to test a polyethylene trap at installations in operation.

Cylindrical HEPA filters

SGN, subsidiary of CEA for reprocessing design, is developping cylindrical filters derived from existing 300 $m^3.h^{-1}$ filters installed in active cells. The outflow rates would range from 1000 $m^3.h^{-1}$ to 3400 $m^3.h^{-1}$ and the main advantage would be to easily telemanipulate filters that are actually replaced horizontally.

IV. Plutonium aerosol filtration

We are concerned with the problem of plutonium aerosol filtration because we have conducted in situ measurements that confirm the results of Mac Dowell, Seeley and Ryan in Oak Ridge. Utilizing filters contaminated by PuO_2 aerosols of a glove box, we have demonstrated the presence of an abnormaly high activity on the filters in succession with the first contaminated filter. This activity tends to reveal a migration with time of the aerosol originally fixed on the first filter.

A more systematic study is starting to quantify more precisely :

- the efficiency of filter media for ultrafine particles
- the migration of alpha and beta contaminated aerosols through the filtering media
- the reentrainment of radioactive aerosols.

The necessary technological developments for plutonium aerosol filtration will be derived from this study. The prospect we are starting to examine in detail is the utilization of electrostatic forces to improve the retention of aerosols in some particular circumstances.

V. Air cleaning in accident situations

We are participating in the CSNI Group of Experts on Air Cleaning in Accident Situations organized by J.P. Olivier of OECD.

Each country member has detailed accident sequences and parameters that are important to air cleaning systems.

We have prepared a working paper summarizing the scenarios for fuel cycle facilities while Dr. Wilhelm has done the same for reactors.

As for fuel cycle, fire, criticality and explosion are the must likely accidents ; fire is certainly the one which affects the most severely the air cleaning system.

By continuing to improve the technology of filtration to respond to these accidents and to test them, let us hope that this group will help to save money by a better coordination of studies and tests.

DEVELOPMENTS IN NUCLEAR AIR CLEANING IN GERMANY

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I. New Filter Designs for Nuclear Power Plants

I. 1. Aerosol Filtration

The limited capacities of repositories for radioactive waste necessitate the development and use of filter elements which can be compacted to the smallest possible volumes after loading.

I. 1.1. Prefilter

Fig. 1 shows a prefilter [1], dimensions 610 x 610 x 54 mm, with a pleated filter medium. The frame remains in the filter housing, while the filter medium is being exchanged and, after having been compacted, will take up very little space. The filter medium is reinforced with a wire mesh so that the filter element, by its inherent stress, generates the clamping load forcing it against the frame, thus providing the necessary sealing action for a prefilter with removal efficiencies of approx. 97 % (for DOP aerosols of 0.3 µm diameter). The filter element proper consists of plastic fiber and has a maximum flow of 3000 m^3/h.

I. 1.2. Drum Filter

The drum filter introduced by Wilhelm [2] at the last Air Cleaning Conference is employed by the following facilities:

(1) For demolition of the Niederaichbach Nuclear Power Station.

(2) In a modified form, in decommissioning and dismantling the "N.S. Otto Hahn" nuclear vessel.

(3) In the reprocessing plant in Brazil.

I. 1.3. High Temperature Filters [1]

Sintered metal fiber webs can be used not only for mist elimination, but also for aerosol separation at temperatures up to 450°C. The aerosol filters built with these metal fiber webs attain removal efficiencies in the sodium flame test which are customary for aerosol filters. Removal efficiencies rise with increasing loading ($c = 12\ mg/m^3$, $v = 0.3\ m/s$) from an initial 70 % to 99.9 % after 10 minutes. After 40 minutes, generation of a filter cake has caused a removal efficiency of 99.997 % to be reached at a differential pressure of 35 mbar. The differential pressure in this case rises in an underproportional function to loading. If Δp becomes too high,

the filters are recleaned by means of pulsating air.

I. 1.4. Electrostatic Precipitators

The precipitators in use in nuclear industry have all been operated with gases of low specific radioactivities $\leq$ 1 Ci/m^3. Accordingly, the question rose up to what specific radioactivities electrostatic precipitators can be used.

At the Institute for Chemical Technology of the Jülich Nuclear Research Center [3], an electrostatic precipitator has been tested with specific radioactivities up to 65 Ci/m^3. The gas enters the cylindrical precipitator (Fig. 2) at the bottom. The discharge electrode has a thin wire at the bottom and two disks at one third and two of the length of the electrode. The length of the precipitator between the gas inlet and the gas outlet is 1 m, the inner diameter is 64 mm. The accumulated deposits are removed from the collecting electrode by washing.

The decontamination factor was measured by introducing cesium aerosols (with Cs-137 as a tracer) into the gas stream and counting the amount of Cs-137 passing the electrostatic precipitator and captured in a HEPA filter. Fig. 3 shows that up to 65 Ci/m^3 the DF does not decrease with increasing specific radioactivity. A DF of 10^4 had been measured earlier in hot experiments with a gas of low specific radioactivity ($\leq$ 1 Ci/m^3).

I. 2. Radioiodine Filters for Nuclear Power Plants

The multi-way sorption filter for radioiodine removal in nuclear power plants introduced at the last DOE-Nuclear Air Cleaning Conference [2] has meanwhile been operated successfully, when required, in a bypass line of the main off-gas conduit of the Stade Nuclear Power Station at a flow of 100,000 m_N^3/h. The necessary minimum removal efficiency to methyl iodide of 99 % is exceeded. The iodine adsorption carbon contained in the filter is moved vertically during replacement, while the layers are exposed to a horizontal flow (Fig. 4). A new development currently being tested is the flow-type sorption filter [1]. The air penetrates through the horizontal layers of activated carbon from bottom to top (Fig. 5). The air loaded with pollutants first reaches the bottom carbon layers which, when attaining their loading limit, can be removed from the air stream by dumping the carbon. In this way, only that carbon needs to be stored in a repository which has been loaded with pollutants up to the limits of its load carrying capacity. Design measures prevent carbon from being stirred up. The two types of filters are expected to contribute to carbon conservation.

Sampling stations in the flow-type sorption filter use the helical and screw type conveyor principle [1]. The sample material used to determine the loading condition is directly taken from the main bed. Sampling points are arranged over the entire depth of the bed.

II. New Filter Designs for Reprocessing Plants

Within the Reprocessing and Waste Treatment Project of the Karlsruhe Nuclear Research Center, an off-gas section for dissolver off-gas cleaning in a reprocessing plant is currently being tested. After testing and optimization, the individual cleaning components will be operated in an integrated system and finally installed in the Karlsruhe Reprocessing Plant as the AZUR dissolver off-gas cleaning section to be tested under radioactive conditions.

Prior to operation and integration of all subsystems, the most important subsystems of the facility are subjected to specific tests involving

- the head end test rig [4] for improving process control of dissolution and NO_x absorption,
- the PASSAT [5] system for the separation of droplets, particles and iodine,
- the catalytic reduction of NO_x and O_2,
- the ADAMO [6] facility for absorption of high-boiling gas components, such as water and carbon dioxide,
- the KRETA facility for cryogenic removal of krypton from nitrogen and xenon.

II. 1. Head End Test Rig

For a domestic reprocessing plant in Germany it has been recommended to limit the I-129 emission from the stack to less than 0.2 Ci/a. With a few modifications, with reprocessing in the head-end, fission product iodine can be confined to the plant area of the dissolver off-gas system. Desorption of the iodine from the fuel dissolution into the DOG is the most important operation. Effective desorption is possible either by distilling a fraction of $\geq$ 20 % from the fuel solution or, more conveniently, boiling under reflux conditions. Additional NO_2-sparging towards the end of the dissolution, or incorporating carrier iodate, were found to be necessary to guarantee residual iodine concentrations well below 10^{-6} mole/l.

The scrubbing acid from the DOG system can be made to contain a fraction of less than 1 to more than 99 % of the desorbed iodine, depending on scrubbing conditions. If high iodine contamination occurs in the scrubbing acid, a desorption step is necessary prior to dissolver recycling.

II. 2. PASSAT Dissolver Off-Gas Filter

The PASSAT dissolver off-gas filter system has been operated for a total of 1.5 years so far under simulated dissolver off-gas conditions. The individual removal components are prototype filters with standardized outside dimensions and lid systems (Fig. 6), which are installed in a cell with remote handling capability. The filter systems and replacement techniques have worked without any failure in more than 50 replacements during trial operation.

No wear in the mechanical parts or corrosion in the filter drums and filter housings was detected. Extensive test runs with a packed fiber mist eliminator designed for prefiltering particles and droplet aerosols and having recleaning capability, which was operated in the function of a Brink filter, were carried out in PASSAT. For droplets with diameters between 1 and 6 µm, removal efficiencies >> 99 %, for particles with the critical diameter of 0.12 µm, removal efficiencies of 99.9 % were obtained. Consequently, an extension of the service life of the downstream HEPA filter can be expected. Iodine experiments in PASSAT on an iodine concentration of 1.1 g I_2/m^3 did not generate any contamination in the filter drums and housings. The iodine adsorption material, AC 6120, was utilized up to 95 % of its absorption capacity with a residual removal efficiency of 97 %.

II. 3. Reduction of Oxygen and Nitrogen Oxides

The reduction of oxygen and nitrogen oxides will be included in test operation from 1981 onwards. Poisoning [6] of the oxidation and reduction catalysts (Pt and Ru, respectively) by tri-n-butyl phosphate and iodine is determined in a laboratory experiment. This phenomenon will be relatively insignificant; accordingly, long service lives can be expected in real off-gas.

II. 4. Precleaning Device

The ADAMO adsorption facility [6] serves for the removal of carbon dioxide and water from the dissolver off-gas by means of silicagel and molecular sieves. It was operated in connection with the KRETA low temperature rectification system. It was shown in several weeks of routine operation that the adsorber bed loaded with co-adsorbed krypton could have the krypton removed to a level of $\leq$ 0.1 % by a recleaning step at room temperature installed upstream of regeneration (Fig. 7).

II. 5. Krypton low Temperature Rectification

The KRETA low temperature rectification system [7] for Kr-85 removal from the dissolver off-gas consists of a sieve tray column with an internal pressure of 5 bar and a packed column with 2 - 3 bar. In the first column, the nitrogen is separated, while in the second column krypton is separated from xenon and collected for final storage in a repository.

After some initial difficulties in operation caused by xenon freezing out of the gas phase in the region of the feed inlet, the gas flow has now been changed (feed to the area of higher column temperature of 120 K), so that the simulated reprocessing off-gas containing up to 5000 vpm of xenon and above is being processed in continuous operation without any defects.

Studies of the behavior of O_2 and CH_4 impurities in the off-gas resulted in an equilibrium accumulation of the oxygen of 100 vpm as a maximum and a maximum of methane of 6 % in the gas phase. The sump liquid (10 % Kr, 90 % Xe) contained 200 vpm of CH_4, which remains in the krypton product, unless other measures are taken.

II. 6. Solutions for Dissolver Off-Gas Cleaning in a Reprocessing Plant

A dissolver off-gas system with the following features is under investigation [4] as a backup solution:

- The DOG flow is reduced to a reasonably small volume of about 100 m^3/t U.
- All process operations are conducted at atmospheric pressure, including noble gas removal by selective absorption in freon-12.
- Iodine is removed from the fuel solution by boiling under reflux conditions.
- Scrubbing the simulated DOG with dilute nitric acid using a two part plate column removed more than 99.9 % of NO_x and more than 99.9 % of the iodine under fumeless dissolution conditions.
- Aerosols were removed by a packed fiber mist eliminator followed by a HEPA-filter.
- The noble gases will be selectively absorbed at atmospheric pressure in two subsequent packed columns using recirculating R 12 fluorocarbon solvent.

III. References

1) P. Hayn, Fa. Delbag Luftfilter, W-Berlin: Private Communication (1980).

2) J.G. Wilhelm: " Developments in Nuclear Air Cleaning in Germany". CONF-780 819, p. 963 (1978).

3) H. Barnert-Wiemer, KFA-Jülich, FRG: Private Communication (1980).

4) E. Henrich et.al: "I-129, Kr-85, C-14 and NO_x-Removal from Spent Fuel Dissolver Off-Gas at Atmospheric Pressure and Reduced Off-Gas Flow". Paper 8.5 of this conference.

5) J. Furrer et.al: "Results of Cleaning Dissolver Off-Gas in the PASSAT Prototype Dissolver Off-Gas Filter System", paper 8.3 of this conference.

6) R.v.Ammon, E. Hutter: Kernforschungszentrum Karlsruhe, FRG: Private Communication (1980).

7) R.v.Ammon et.al: "Steady State Operation of the First Cryogenic Column in a Krypton Separation System", paper 3.4 of this conference.

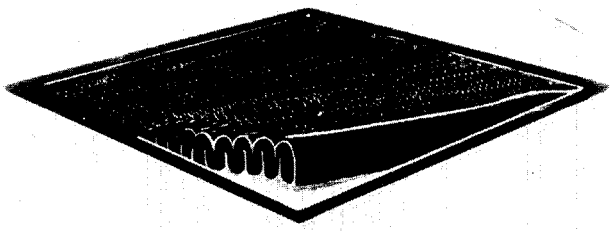


Fig. 1 Prefilter

with exchangeable filterelements

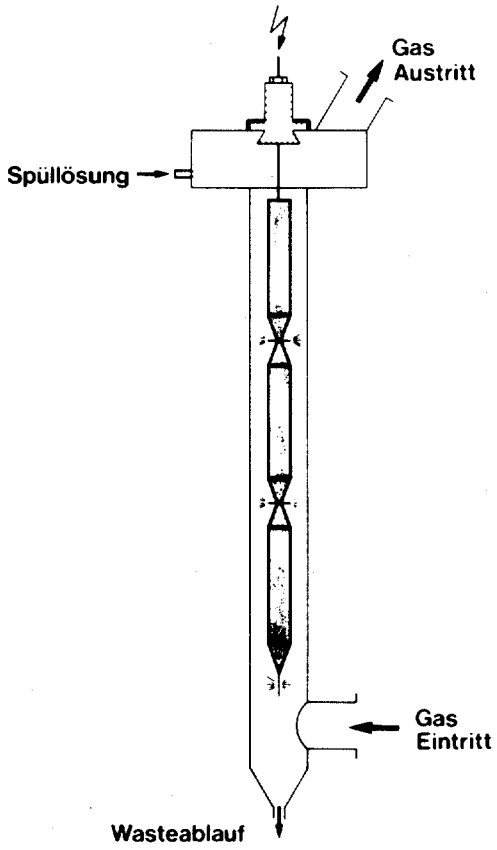


Fig. 2 Electrostatic Precipitator (KFA Jülich)

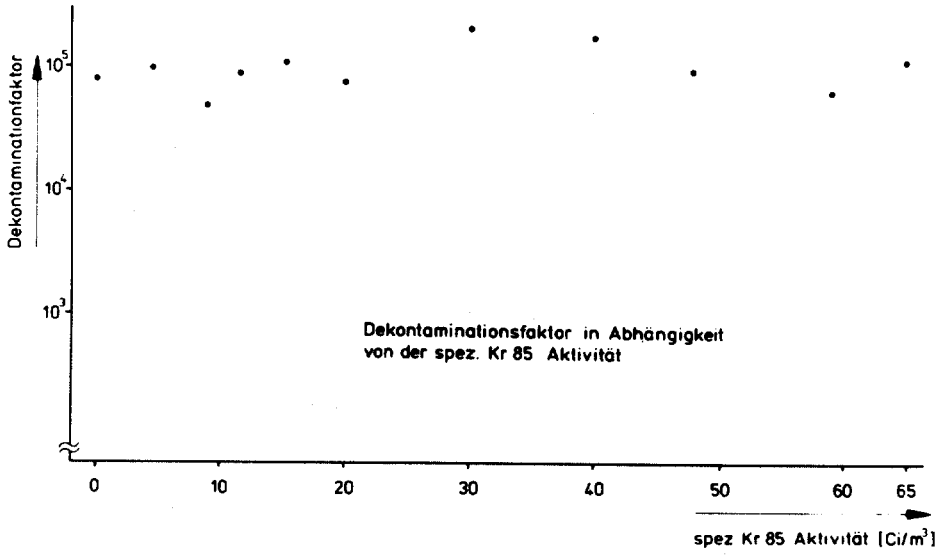
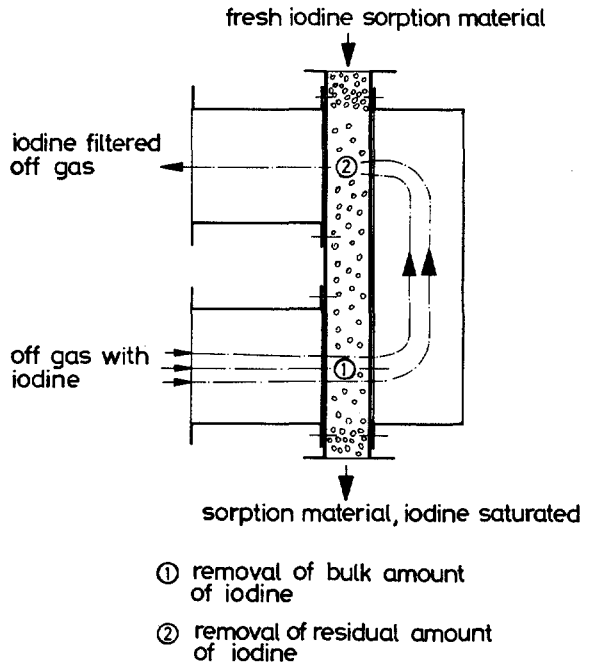


Fig. 3 Decontamination factor for aerosols as a function of the spec. Kr-85 activity



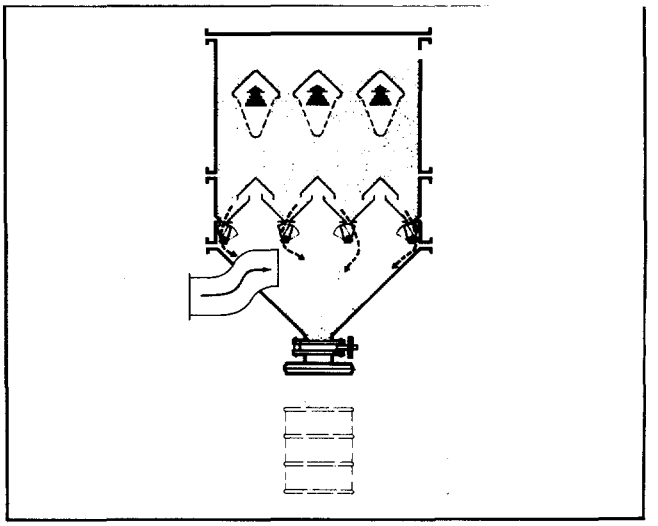


Fig. 5 Flow-type sorption filter

Fig. 4 **Multiway iodine sorption-filter**

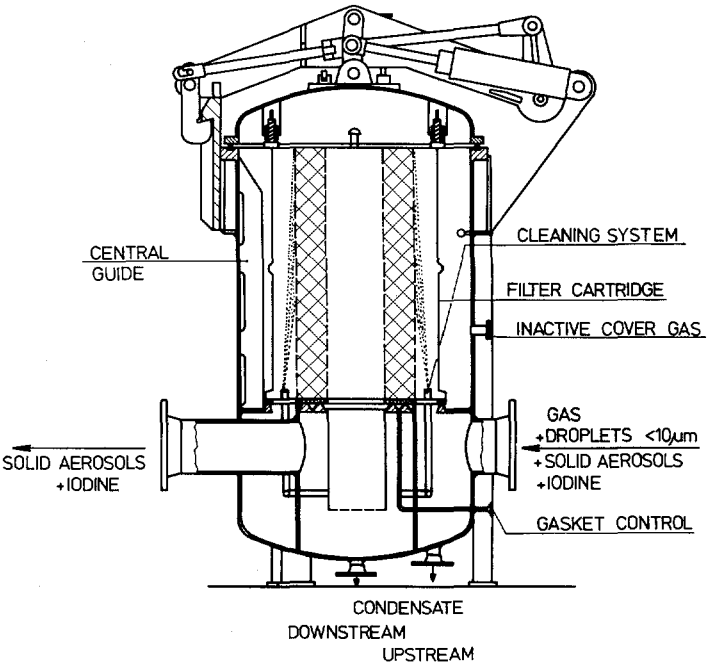


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FIG. 6 PASSAT packed fiber mist eleminator (PFME) for droplet removal <10µm

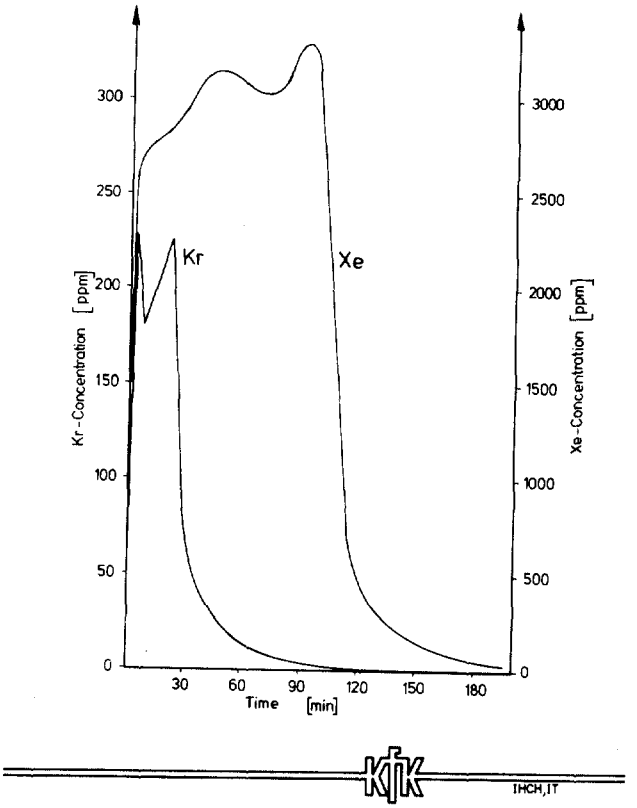

Fig. 7

Desorption of Kr and Xe from the silica and molecular sieve beds in the pilot plant ADAM0 with 5 Nm^3/h N_2 at 1bar and 25°C

NEW TRENDS IN THE BELGIAN PROGRAMME ON NUCLEAR AIR CLEANING TECHNOLOGY

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Abstract

In the Belgian Programme on nuclear air cleaning technology the Mercurex process has been developed to trap iodine compounds from dissolver off-gases. Krypton is removed with the help of a cryogenic distillation unit. The various gas cleaning units have been integrated in a gas purification test loop for dissolver off-gas at a through put of 25 m^3gas h^{-1}.

The separation of tritium from liquid reprocessing effluents is being developed according to the ELEX-process. New research is started on the capture of semi-volatile ruthenium compounds.

Introduction

The nuclear air cleaning research and engineering in Belgium has been pursued according to the lines set forward in the presentation of last nuclear air cleaning conference (1). The main aim of this experimental program remains the development of nuclear air cleaning technology in support of the Belgian nuclear schedule that foresees for 1983 an installed nuclear power capacity of 5246 MWe reflecting 35 % of the total electrical power capacity at that time. Thereby, special attention has to be given to the in-land location characteristics of the different nuclear installations related to this schedule.

The new trends obtained during the last two years in the experimental programme will be reviewed

Situation of the actual experimental program

In general terms, the Belgian R & D program on the cleaning of nuclear off-gases is now focused on the development of a suitable gas purification circuit for dissolver off-gases of a reprocessing plant. Therefore, an integrated gas purification test loop "GAS-TON" has been constructed and tested under non-irradiated fuel conditions while a technical demonstration loop connected to a batch type dissolver for irradiated fuel is under design. The trapping of iodine compounds and the retention of krypton are the specific objectives of this investigation.

Additionally, the separation of tritium from aqueous effluents of nuclear installations is studied taking mainly into account the inland localisation of a potential small reprocessing plant. For this purpose, an electrolysis -catalytic exchange process with the trade name ELEX- process is developed.

Finaly, the potential application of vitrification processes for the solidification of high level liquid waste drove us to start a new investigation aimed at the retention of semi-volatile ruthenium compounds by solid materials.

Before describing these program items in more details, I like to mention two other subjects which have been treated in the earlier Belgian program. Firstly, the delay of short-living Xenon isotopes on charcoal beds was tested in a pilot unit installed on the off-gas system of the Belgian pilot pressurised water reactor BR3 in Mol. The performance of these charcoal beds under real conditions was satisfactory and the experimental design model was confirmed during the tests. Hence, this successful investigation was stopped.

Secundly, the development of a granular type dust filter for the removal of dust from the off-gases of a high temperature waste incinerator has been dropped from the earlier program as soon as it was clear that not dust but fume with a particulate size of about 0.5 µm was the main filtration problem.

The trapping of iodine compounds

The investigation on the retention of iodine compounds was started in the past in a 1 m^3h^{-1} laboratory set-up equipped to determine the performance of various solid materials on their trapping capability for elementar iodine and for methyliodide. The relative humidity of the carrier gas could be accurately controlled from o to 98 %. This laboratory set-up is now used to check the quality of charcoal applied in Belgian nuclear power stations. In the ventilation line of the hood in which this laboratory set-up is mounted, the performance of full size charcoal filter cases is determined.

The experimental effort related to the retention of iodine compounds was concentrated on the Mercurex process, of which the performance was checked for transient conditions in the gas released during the dissolution of non-irradiated fuel batches traced with iodine. Also the conditioning of the iodine trapped in the Mercurex solution and the recycling of the mercury content in spent Mercurex solutions are investigated. The results of these efforts are reported in the paper of this conference entitled "Iodine trapping and conditioning in the Mercurex system "[2].

The trapping of krypton

The removal of krypton from a carrier gas has been investigated in a packed distillation column. This column highly instrumented has continuously being in operation during 13 000 hours with an average availability factor of 99.70 %. Transient peak conditions in the inlet gas of this column have been tested too. As reported in another paper of this conference [3], large krypton decontamination factors can be acquired without any operation problem for an inlet gas containing mainly nitrogen and up to 1 % argon, 2000 ppm krypton and 8000 ppm xenon.

For the conditioning of the gases entering the cryogenic distillation the catalytic destruction of nitrogen oxides with ammonia and also the removal of water vapour and CO_2 on molecular sieves has been studied on an engineering scale. In this way, it has been possible to realise the flow sheet shown schematicly in Fig. 1, by integration of the various units tested earlier separately. This integrated gas purification test loop for dissolver off-gases has been designed as a

demonstration rig with a nominal through-put of 25 m^3gas h^{-1}, equivalent to a reprocessing capacity of 1 ton a day, hence its name "GASTON" (4). Short integrated runs did not show any operation problems. Also longer runs performed earlier on a combination of the iodine section with the conditioning section or on a combination of the conditioning section with the cryogenic section showed an excellent reliability of this integrated demonstration rig. It should be emphasized that the experiments in this rig were performed during dissolution of non-irradiated uranium dioxide.

Anorganic and organic iodine species traced with I-131 were artificially added to the carrier gas or to the nitric acid solution. As indicated on the flow sheet, other gaseous contaminants could be added artificially as well. The technical problems encountered during the more than seven years old program were related to internal leaks of the various equipment parts of the drying unit working under pressure and temperature swing conditions.

In the near future, the flow sheet shown in Fig. 1 will be modified a little bit by replacing the selective NO_x removal step by a combined oxygen elimination and NO_x-destruction step. A suitable catalytic reactor with hydrogen as reagent is under construction. Earlier experiments at a pressure of 0.5 MPa and a temperature of 300 °C have shown that it is possible to remove oxygen by this catalytic reaction down to at least 5 vpm at a relative gas flow rate of $8.3\ 10^{-4}$ $kg_{gas}\ kg^{-1}_{catalyst}\ s^{-1}$. It is this modified flow sheet that will be applied in the future to clean the off-gases of a dissolver where batches of 10 kg LMFBR fuel will be dissolved.

The separation of tritium

The tritium retention studies were initially focused on the removal of tritium from gaseous reprocessing effluents. If tritium can be released from the spent fuel into the gaseous phase before any aqueous operation, adsorption on molecular sieves after some isotopic dilution with hydrogen and after complete conversion to (tritiated) water is the most practical collection method. A once-through 15 m^3h^{-1} oxidation-adsorption unit with a closed regeneration system and with a decontamination factor of 1000 at total (tritiated) hydrogen and water inlet concentrations down to 1000 vol. ppm has been constructed and tested with success.

Further tritium retention studies treat the separation of tritium from liquid reprocessing effluents. Indeed, an isotope separation process becomes necessary to reduce the volume of tritiated aqueous effluents even when an appropriate liquid management inside the reprocessing plant has reduced this volume already to about 3 m^3 per ton of LWR fuel processed. Within the frame-work of the indirect action program of the European Communities, S.C.K./C.E.N. is therefore developing the ELEX process (5). As schematized on Fig. 2 , this ELEX process is a combination of water Electrolysis and tritium EXchange between hydrogen and water, the exchange being promoted by a hydrophobic catalyst. Until now the single constituent steps of this process have been investigated. For electrolysis under normal conditions an elementary tritium separation factor of 11.6 with a standard deviation of 6 % was obtained. As to the exchange step a hydro-

phobic catalyst has been developed which yields an over-all exchange rate constant of 9 mol $s^{-1}m^{-3}$ in a countercurrent trickle-bed reactor. At the moment an integrated bench scale detritiation unit is being built for further tests and for a dynamic demonstration of the ELEX process.

The capture of semi-volatile ruthenium compounds

The capture of semi-volatile ruthenium compounds by solid materials is studied using rutheniumtetraoxide as representative semi-volatile product. Therefore, a ruthenium tetroxide generator has been developed to get a stable production rate in the range from 3 to 50 mg h^{-1}. This artificial semi-volatile ruthenium source is used to test the performance of sintered stainless steel porous discs, silica-gel and ferric-oxide silicagel catalysts on their ruthenium retention capability under different conditions of temperature, flowrate, moisture and NO_x concentration(6). Radio-active tracing is foreseen to get able to determine decontamination factors as high as 10^4. A demonstration unit with a gas throughput of 10 m^3h^{-1} is in the design phase. This research on ruthenium capture is performed under contract with DWK as a part of the German HAW-technological program for the development of the vitrification process PAMELA.

Conclusive remark

This review of the S.C.K./C.E.N. experimental program on nuclear air cleaning technology has indicated that in Belgium a technical infrastructure is available to tackle any nuclear gas cleaning problem that might arise in the future.

References

(1) W.R.A. Goossens, L.H. Baetslé, A. Bruggeman, G. Collard, "Gas cleaning research and engineering in Belgium", 15th DOE Nuclear Air Cleaning Conference, Conf.-780819, Vol. 2, p.956-962 (1979).

(2) G.E.R. Collard, D. Hennart, J. Van Dooren, W.R.A. Goossens, "Iodine trapping and conditioning in the Mercurex system" 16th DOE Nuclear Air Cleaning Conference.

(3) G.E.R. Collard, L.P.M. Geens, P.J. Vaesen, W.R.A. Goossens, " Experimental development and design aspects of a krypton removal distillation unit" 16th DOE Nuclear Air Cleaning Conference.

(4) G.E.R. Collard, P.J. Vaesen, W.R.A. Goossens, L.H. Baetslé, "Operational experience with a 25 m^3h^{-1} simulated dissolver off-gas purification loop" IAEA-SM-245/49 (1980).

(5) A. Bruggeman, W. Doyen, R. Harnie, R. Leysen, L. Meynendonckx, M. Monsecour, W.R.A. Goossens, L.H. Baetslé "Separation of tritium from reprocessing effluents" IAEA-SM-245/52 (1980).

(6) M. Klein, M. De Smet, W.R.A. Goossens, L.H. Baetslé, "Filtration and capture of semi-volatile nuclides", IAEA-SM-245/51 (1980)

FIG. 1 - INTEGRATED GAS PURIFICATION TEST LOOP
25 Nm^3h^{-1}

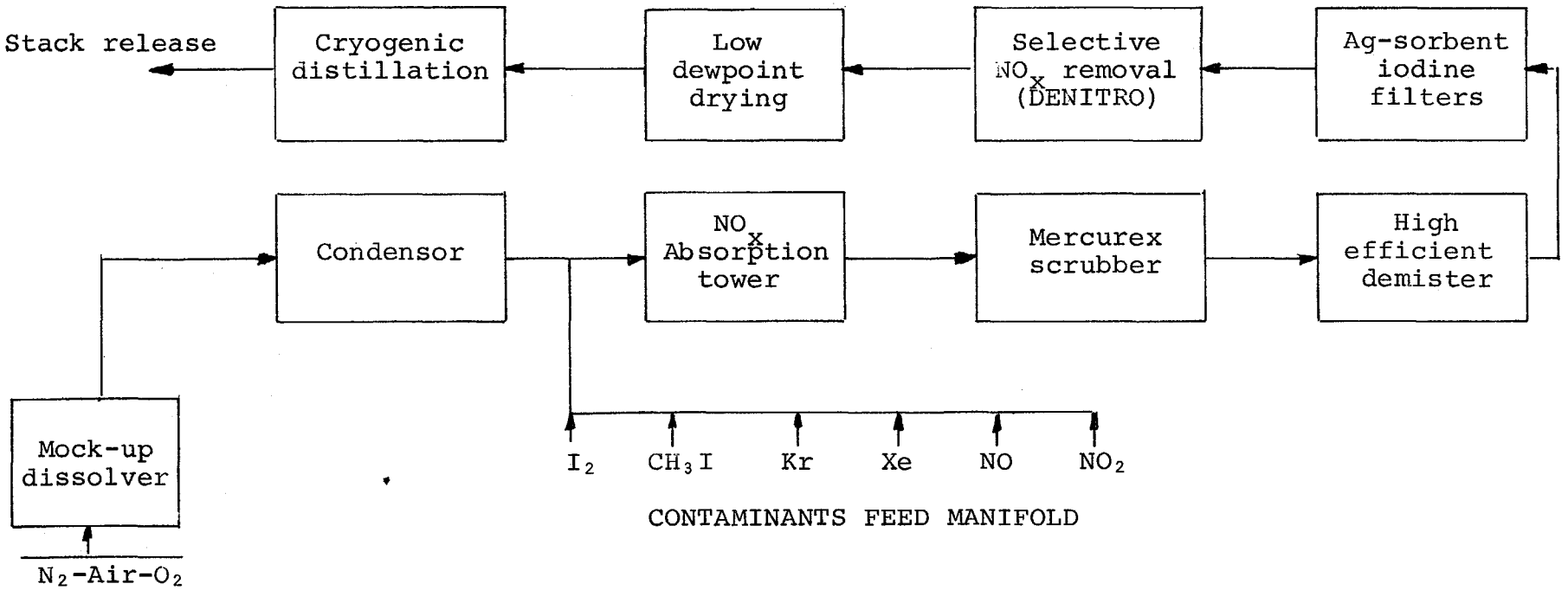
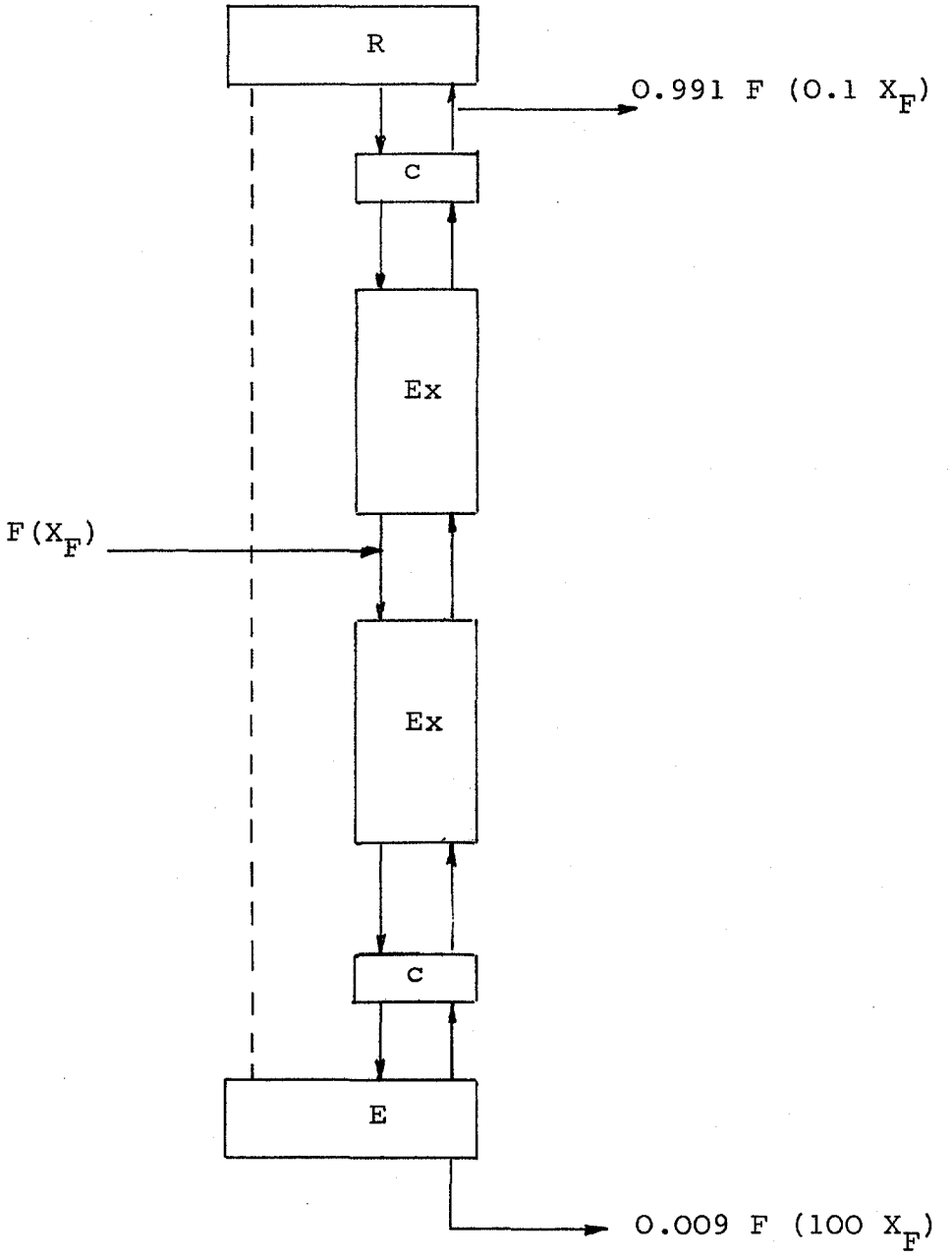


FIG. 2 - SCHEMATIC REPRESENTATION OF THE ELEX PROCESS FOR SEPARATION OF TRITIUM FROM AQUEOUS EFFLUENTS



LEGEND

C : conditioning step
E : electrolyser
Ex : tritium exchange catalyst column
F : tritium contaminated water feed
R : recombiner
X_F : tritium concentration of the feed

RECENT DEVELOPMENTS IN THE TESTING OF ABSORBERS IN VENTILATION SYSTEMS

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A centralized filter testing service was established in Ontario Hydro nuclear power stations in 1976. This includes acceptance and surveillance testing of the integrity of air cleaning assemblies and determination of absorber efficiency for the removal of airborne iodine species including hypoiodous acid.

Integrity Testing of Absorbers

The accuracy and reproducibility of the previously used freon method was not adequate. A new procedure was developed with stable methyliodide as the testing gas and a portable gas chromatograph with Chromosorb W + 30% metatritolyl phosphate column and an electron capture detector, to measure the concentration of methyliodide upstream and downstream of the absorber.

This method is much more sensitive and reproducible than the freon method with a heated detection diode and the testing period is not limited by the adsorbate breakthrough when testing absorbers filled with TEDA impregnated charcoal.

Decontamination factors of $> 10^4$ can be accurately measured. The method has been used since 1979 in routine testing of absorber integrity in Ontario Hydro nuclear power stations.

Testing of Absorbers for Iodine Species Removal Efficiency

Test conditions and penetration limits listed in Table 1 were proposed for "normal operation" filters, controlling radioiodine releases within regulatory emission limits, where a food chain is the major exposure pathway. Originally prepared for internal use in Ontario Hydro, the limits have been recommended for the proposed Canadian Standard CSA-N288.3 "Design, Installation and Testing of Air Cleaning Systems". The penetration limits for CH_3I and elemental iodine are in agreement with values in the ASTM D 3803 standard.

Experimental Evaluation of Iodine Species Deposition on Grass

The value for HOI penetration was derived from results of our evaluation of iodine species deposition on grass. Grass samples in a glass chamber were exposed for approximately two hours to pure iodine species, dispersed in air continuously flowing through the chamber at 5 lpm flow. Iodine species were then collected with a selective sampler downstream of the chamber to measure their airborne concentration. The activity of ^{131}I, retained on 100 cm^2 of grass, was also measured. The deposition velocity values were then calculated from

^{131}I on grass, the airborne concentration and the exposure times. Results in Table 2 show that the deposition velocity of HOI is approximately 60 times greater than its value for methyliodide and approximately 10 times lower than elemental iodine deposition velocity. Our results of CH_3I and elemental iodine deposition are in agreement with previously published data.

Absorbers Testing Procedure and Equipment

Charcoal samples are routinely tested with CH_3I, HOI and occasionally with elemental iodine. Both acceptance and surveillance testing of charcoal samples is performed in an environmental chamber (Figure 1) which is equipped with automatic control and recording of both temperature and relative humidity. Test rings, illustrated in Figure 2, are filled with tested absorbent to the depth of 50 mm. Two 25 mm rings are stacked for methyliodide testing. However, five 10 mm rings are used for HOI and elemental iodine penetration testing, in order to obtain adequate number of experimental points in the penetration graph.

Methyliodide ^{131}I is produced in our laboratory by isotopic exchange of ^{131}I from sodium iodide solution with stable methyliodide. It is then filled into a pressure bomb with nitrogen carrier gas and released at a controlled rate into the test column through the challenge period.

Elemental iodine is generated from highly acidic solution of $Na^{131}I$ with KIO_3 and stripped with air into the test column.

The procedure for HOI generation, described in the 14th Air Cleaning Conference, paper "Selective Sampling of Hypoiodous Acid", has been used in our charcoal testing program.

Teflon coated aluminum tubing is used to transfer elemental iodine and HOI from the generators into the test columns.

From the results of this program it has been concluded that HOI penetrated impregnated charcoal less than CH_3I, but it is more penetrative than elemental iodine under standard testing conditions. Most of our samples were at least 10 times more efficient for HOI than for CH_3I removal and at least 2 times less efficient for HOI than for elemental iodine removal. Long term desorption tests are presently in progress.

Proposed Standard CSA - N288.3	TEST CONDITIONS	ADSORBER DEPTH	50 mm
		TEMP.	25°
		RH	95%
		AIR VELOCITY	0.2 $m \cdot s^{-1}$
	PENETRATION LIMITS	I_2	0.05%
		HOI	0.1%
		CH_3I	3.0%

Table 1

Proposed test conditions and penetration limits for absorbers in nuclear air cleaning systems.

DEPOSITION VELOCITY (grass)	
	Vg – $m \cdot s^{-1}$
I_2	7.6×10^{-3}
HOI	6.9×10^{-4} 7.2×10^{-4} 7.0×10^{-4} 6.5×10^{-4}
CH_3I	1.2×10^{-5}

Table 2

Velocity of iodine species deposition on grass at 25°C, 60% RH.

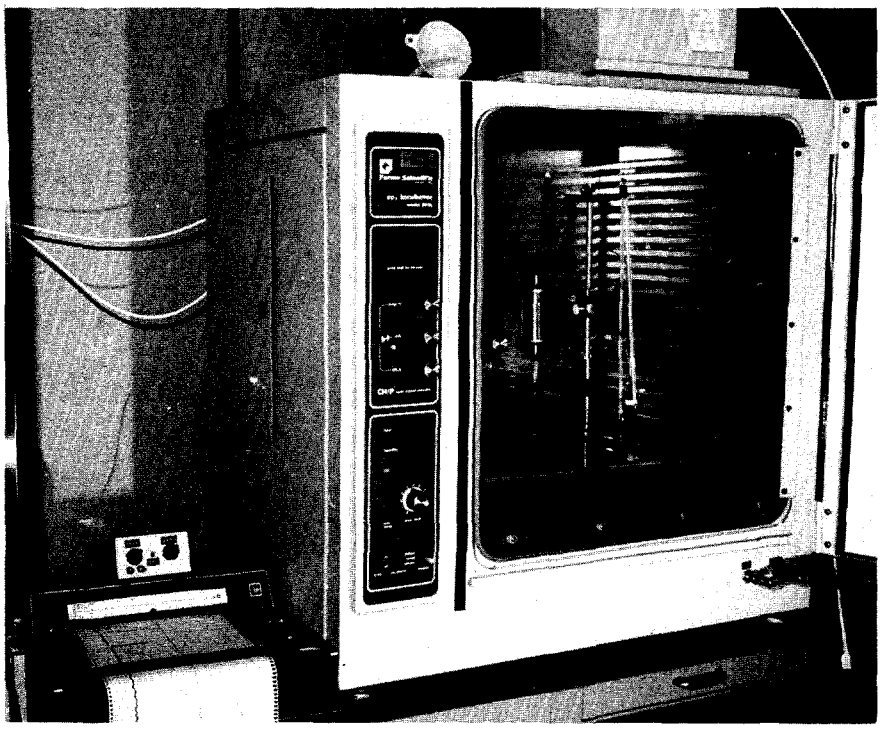

Figure 1
The environmental chamber for absorbent testing.

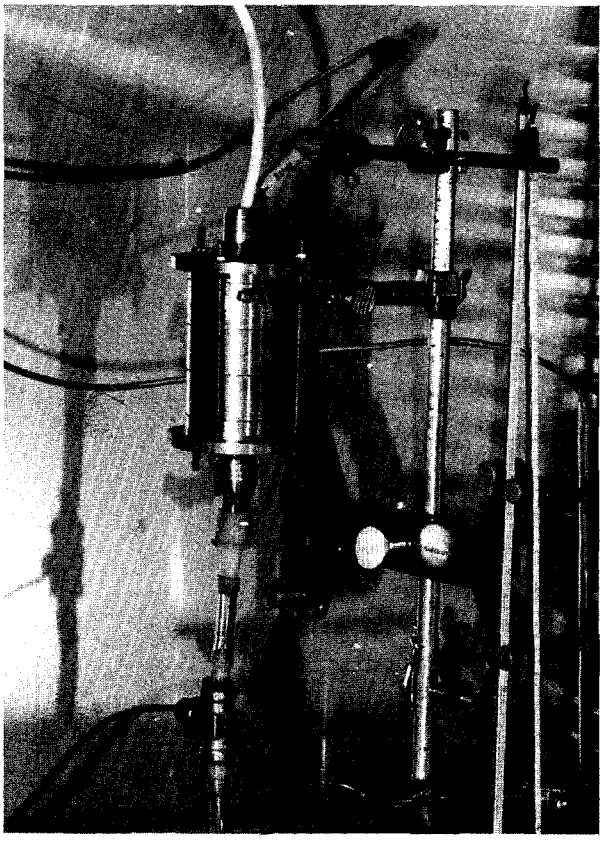

Figure 2
Assembled test rings filled with absorbent.

ADVANCES IN AIR CLEANING TECHNOLOGY IN THE U.K.

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Introduction

The following is an attempt to summarize advances in the field of nuclear air cleaning technology in the U.K. Although I have obtained information from many sources, I apologise for any bias towards work in the UKAEA.

Filtration

Standards

The British Standard sodium flame bench test for HEPA filters (BS3928) is to be updated. The ASHRAE test is to be adopted in Europe for lower efficiency filters.

Prefilters

A cleanable prefilter could considerably enhance the life of HEPA filters. Potentially an electrostatic precipitator (ESP) is such a device. A programme to develop a safe, reliable and rugged ESP which will meet the special requirements of the nuclear industry has been initiated at Harwell. This involves computer modelling to optimise parameters and the development of plates with optimum resistivity to limit the energy available for discharge.

Cylindrical Filter Development

Axial-flow circular filters are being developed. They should have a much higher flow rate than conventional circular (radial-flow) filters which require a 90° change in flow direction and considerable space for air-flow passages. It is believed that circular filters will offer significant advantages since they are compatible with current posting and storage geometries and reduce sealing problems for posting and remote change systems. Filters up to 450 mm diameter by 200 mm deep have been produced and work is progressing on optimising the spacing between paper layers to increase the flow rates. It is hoped that a 450 mm x 200 mm unit will operate at a flow rate comparable to that of a standard HEPA filter (1700 m^3/hr).

Sealing and Change Techniques

A paper by E. Lillyman at this conference describes developments in ventilation and filtration systems in the UKAEA, particularly at Dounreay.

The UKAEA at Risley and Dounreay are developing double-lidded transfer systems for conventional 600 x 600 x 300 mm HEPA filters. Results are encouraging but the housings are somewhat large and cumbersome. Change systems incorporating cylindrical filters (currently the radial-flow type) are being developed at Harwell. In one a rotatable magazine carries four filters in a common duct. Filters are loaded and unloaded via transfer chutes. Gravity feed alone is not sufficient and further developments will require positive feed and withdrawal mechanisms. The other system is a push through system where filters are stacked end-to-end and separated by spacers. The laboratory model has shown that this simple system should be trouble free and a version is being installed on a new suite of cells.

The UKAEA at Risley have developed a remote change filter system using

conventional HEPA filters and have shown that for their system wooden-cased filters are sufficiently fire resistant to replace the metal framed filters normally used in the U.K. The filters can therefore be incinerated to reduce the volume for disposal.

Filter Loading

A paper has been presented at this conference by R.P.Pratt describing dust loading tests on conventional and high capacity filters. The dust-loading capacity at the design flow rate of 3400 m^3/hr was not twice that of the conventional filter at 1700 m^3/hr. Significant differences were found between different manufacturers.

Storage and Disposal

Much of the work described above has as one of its main objectives a reduction in the volume of waste for storage and disposal.

A technique for dismantling and separation of the components of HEPA filters has been developed at Harwell but in practical tests there can be problems due to the weakness of the filter paper after service in some circumstances. The emphasis has therefore been on crushing the complete (metal cased) filter and the filter pack without case. A 4:1 reduction in volume can be achieved. Cases with quick release fasteners have been produced for ease of dismantling. Plastic filter cases have also been developed which meet the BNFL/UKAEA temperature specifications (500°C for 10 min) and are capable of being incinerated.

Filter Testing

R. G. Dorman has presented a paper to the conference on a comparison of methods for particulate testing of HEPA filters. Another paper (M. Marshall) has compared Condensation Nuclei, DOP and Sodium Flame test methods for in-situ filter testing. For unfiltered leaks (e.g. gasket leaks) all methods agree when used properly. For filters with low efficiency, DOP and Sodium Flame methods are in reasonable agreement with the British Standard filter test rig while condensation nuclei do not penetrate the filters and show a very high efficiency. Methods based on DOP are generally the most satisfactory for operational use.

A hot, dynamic filter test rig is to be built at Harwell to look at the performance of filter systems at high temperatures.

Gaseous Wastes

There is considerable interest in the storage and disposal of gaseous wastes. Dr. H. A. C. McKay of Harwell presented a paper on background considerations in the immobilisation of volatile radionuclides at the IAEA symposium held earlier this year on the Management of Wastes from Nuclear Facilities. Joint assessment studies, between the NRPB and the UKAEA in Britain and the CEA in France, are in progress on the waste management of iodine-129 and there is a proposed study on carbon-14.

A method of krypton immobilisation by combined ion implantation and sputtering is being developed at Harwell. The krypton is stored as bubbles in a metallic matrix and therefore does not rely on a pressure vessel. Methods of tritium immobilisation in inorganic hydrides, hydroxides and hydrates are also being studied. Work on liquid metal embrittlement (by rubidium) is also in progress as this could cause catastrophic failure of pressure vessels containing krypton.

At Windscale, where there is a fuel storage and reprocessing plant, there is

an active programme to quantify minor discharges. For iodine they have developed a sampler containing a heated bed of AC6120 (high fired silica spheres impregnated with silver nitrate). This is not affected by moisture, traces of organic vapours or oxides of nitrogen.

Work on the ageing of charcoals, presented at Vienna by Mr. R. D. Collins of UKAEA, Windscale, suggests that impregnation with TEDA is somewhat better than with potassium iodide in preventing long term deterioration of coal based charcoal in the presence of water vapour and oxygen.

Conclusions

Progress is being made over a wide front but it is generally 'slow and steady' with no obvious breakthroughs. The main objective in the field of filtration at present is to minimise the quantity of waste for storage or disposal.

Acknowledgements

My thanks to all those throughout Britain who so readily supplied information at short notice for this review, particularly R. P. Pratt and M. J. S. Smith at Harwell.

REVIEW OF THE ACTIVITIES OF THE OECD NUCLEAR ENERGY AGENCY ASSOCIATED WITH AIR CLEANING TECHNOLOGY

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The Nuclear Energy Agency of the OECD (NEA) is an intergovernmental organisation whose primary objective is to promote co-operation between its 23 Member countries on various aspects of nuclear development, notably on safety and regulation matters. NEA does not conduct any research or development work, its role being essentially to assist Member countries with regard to safety, radiation protection and waste management problems, by collecting and disseminating technical information, discussing and evaluating R & D results and when possible, coming up with broadly accepted views and guidance on specific issues.

NEA's programme in the field of safety and regulation is governed by the three following committees:

- the Committee on Radiation Protection and Public Health (CRPPH);
- the Radioactive Waste Management Committee (RWMC);
- the Committee on the Safety of Nuclear Installations (CSNI).

Most of the work of these committees is done through expert groups, specialist meetings and workshops on specific topics leading sometimes to the publication of state-of-the-art reports. Wider meetings, such as symposia, are also organised by NEA and in this respect, NEA co-sponsored with IAEA the International Symposium on the Management of Gaseous Wastes from Nuclear Facilities, which took place in Vienna last February.

Some of the work carried out by NEA has already been reported at the Vienna Symposium; this concerned essentially the preparation of a report on the Radiological Significance and Management of Tritium, Carbon-14, Krypton-85, Iodine-129, Arising from the Nuclear Fuel Cycle*. This report was prepared by a group of experts for the Committee on Radiological Protection and Public Health and represents what is probably the most advanced attempt to implement the optimisation principle of ICRP Publication 26. The main objective of this study was to provide OECD Member countries with information on which an optimum, uniform, management strategy for each of the four nuclides could be based. An optimum strategy was defined as the strategy that would result in the lowest combination of cost and detriment to society consistent with an acceptably low risk to the maximally exposed individual.

*Published by the OECD in April 1980. [ISBN 92-64-12083-17]

Based on a number of assumptions regarding source terms in normal operations from reference reactors and fuel reprocessing facilities, the report took into account:

- distribution pathways on local, regional and global scales for each of the nuclides;
- maximum dose rates to the most exposed individuals from particular local nuclear facilities and from regional and global distribution of a representative mixture of nuclear facilities;
- appropriate collective dose commitments for each of the nuclides and for each of the nuclear facilities studied;
- overall ranges of uncertainty for maximum dose rate and cost effectiveness.

No attempt was made to consider radiation exposure received by operating staff involved in the retention, storage and disposal of the nuclides, nor the consequences of possible accidents resulting from these practices. The overall cost benefit analysis is from that point of view not really complete. However, without attempting to make specific recommendations for the management of the nuclides concerned, the group of experts has come up with extremely interesting results concerning:

- the maximum effective dose equivalent rates to the most highly exposed individuals from local, regional and global sources;
- the values of marginal cost effectiveness of reducing the collective dose commitments from the various radionuclides (for given decontamination factors).

The results are presented in a matrix form, including the range of uncertainties for the various nuclides and plants considered and it is therefore not possible to enter into details in this paper.

Another activity of NEA under the Committee on the Safety of Nuclear Installations is an assessment of the various codes used to model the consequences of nuclear accidents, which are necessarily related to the role played by air cleaning systems as far as source terms are concerned. It is being carried out as an international standard problem exercise which is designed to enable a comparison and validation of the various codes used in different countries for modelling the consequences of accidents.

The Committee on the Safety of Nuclear Installations also sponsors various groups dealing with airbourne contamination and reactor containment response in case of accidents. A State-of-the-Art Report on the Role of Nuclear Aerosols in Reactor Safety was published in 1979*. This report deals with aerosol forms and

*Published by OECD in 1979 /ISBN 92-64-11977-9/

characteristics, aerosol processes inside containments which govern aerosol transport behaviour after the aerosol source has been released to the reactor containment building, and finally the computer modelling of this behaviour. The report puts the emphasis on available aerosol computer codes, their comparison with test data and evaluation of the importance of uncertainties in aerosol processes described by the codes. It covers mainly the aerosol behaviour in LMFBR systems. The publication of this report was followed last April by the organisation of a Specialist Meeting on the subject of Nuclear Aerosols, held in Gatlinburg, Tennessee, with the co-operation of the Oak Ridge National Laboratory and the US Nuclear Regulatory Commission. As a result of this meeting, which enabled a detailed review of the technical problems involved, further work was identified concerning the use of computer codes and their validation with regard to aerosol modelling.

Concerning the impact of accident situations on air cleaning and filtration systems, NEA organised in October 1979 a Specialist Meeting at the Karlsruhe Nuclear Research Centre, in the Federal Republic of Germany. During this meeting the problems concerning accident situations in reactors and fuel cycle facilities were reviewed, notably the behaviour of charcoal bed filters and HEPA filter performances. Following that meeting a group of experts was set up, which will have its first meeting in December this year. This group will first review accident scenarios in various types of nuclear facility as a starting point to identify what the actual impact is on filtration systems, and what would be the minimum requirements for these systems to withstand accident conditions. The experience which is available from the TMI accident will be very valuable for this exercise, which will also include consideration of other accidents, such as criticality, fires, explosions, in fuel cycle facilities.

The monitoring of airborne radioactive effluents is also important, particularly in accident situations, and the NEA Radioactive Waste Management Committee has recently carried out an enquiry on this subject. The problems of sampling and measurement were reviewed and suggestions were made notably concerning the need for discriminating between noble gases, halogens, and other contaminants such as aerosols, and the improvement of monitoring systems.

Concluding Remarks

It is obvious from this brief review of NEA's activities associated with air cleaning technology, that more emphasis is being placed on accident situations and the important role which the air cleaning systems will be expected to play to mitigate the consequences of accidents. Routine situations seem to be properly controlled, at least at the level of reactors. With regard to reprocessing plants, a great deal of work will still be necessary concerning the practical application of the retention, storage and disposal techniques for gaseous or volatile radionuclides which are being developed. One of the elements of discussion will be the safety of storage facilities, which will have to be taken into account in the definition of optimum control systems.

NEW AIR CLEANING TECHNOLOGY IN JAPAN

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I. Introduction

In Japan, application of the new techniques and improvements in air cleaning systems have been made to reduce release of radioactive materials from nuclear facilities based on the ALARA concept. For example, the reduction of release of radioactive gaseous effluents has been made by installation of a charcoal gas hold-up system and a clean steam supply system for a turbine gland seal in a BWR and of a gas decay tank system in a PWR.

In connection with the effort for reduction of releases in plants, research and development on air cleaning technology have been also made. Some activities are introduced in the present paper.

Removal of particulates

In relation to the research on collection efficiency of multi-stage HEPA filters, the collection characteristics of HEPA filters for submicron particles were investigated, as parameters of particle size and face velocity[1].

Multi-layer type filters with geometrically distributed holes in filter media were developed for the LMFBR's air filters, because loading of sodium oxide aerosols on HEPA filters may cause the sharp increase of pressure drop.

Investigation was made on operational performance of ceramic filters incorporated with the incinerator for low level radioactive wastes[2][3][4], which were constructed based on the Karsruhe design[5] with some modifications.

Removal of airborne radioiodine

In addition to the studies on iodine-removal performance of charcoals, the effect of weathering on impregnated charcoals was extensively investigated for establishing the criteria for design and maintenance of the charcoal filter systems, mainly for emergency in reactors[6][7]. Removal performance of the other adsobers was also studied[8].

Containment spray tests have been carried out with the JAERI Model Containment ($708m^3$) since 1977, to demonstrate the effectiveness of the spray systems installed in containment vessels of PWRs and BWRs for the removal of radioiodine in accidents.

Removal of noble gases

A removal system using cryogenic selective adsorption-desorption process was developed as an alternative technique for a noble gas

hold-up system[9][10].

A pilot facility to recover radioactive krypton and xenon gases by a cryogenic technique has been under construction in the spent fuel reprocessing plant, Power Reactor and Nuclear Fuel Development Corporation[11].

Removal of tritium

Fundamental studies have been made on removal of tritiated water by adsorption[12] and on tritium gas removal with the catalystic oxidation and adsorption using D_2 gas in relation to fusion research[13]. Based on these results, in addition to the experiences mainly in the U.S.A., a tritium gas removal system is designed for the tritium handling facility[14].

In the above research activities, the followings are described in detail.

II. Penetration characteristics of submicron DOP aerosol for HEPA filters

The penetration characteristics of submicron particles for HEPA filters were investigated through the particle concentration measurements of the polydisperse DOP aerosol with a laser particle spectrometer as a function of particle size and face velocity[1].

Figure 1[1] shows a flow diagram of the experimental apparatus for the test of HEPA filters. Three kinds of HEPA filters (A, B and C) were tested, which have two filter sizes of 200mm × 200mm × 150mm and 200mm × 200mm × 75mm.

The laser particle spectrometer (Hitachi DECO TST-500) is equipped with a He-Ne laser of a beam power of 8mW at the wave length of 0.6328μm and with a photocell to introduce an air sample at a high flow of 2.8ℓ/min. This spectrometer can measure the aerosol particles of sizes ranging from 0.1 to 10μm in the concentrations as high as about 10^5particles/cm^3 with less than 5% coincidence error.

In order to measure the high particle concentrations at the upper stream of a test filter without significant coincidence error in particle counting, the dilution equipment was used as shown in Fig.1, whose typical dilution ratio was 50.

The size distribution of the DOP aerosol was log-normal in the count median diameter of 0.15μm with the geometric standard deviation of 1.5.

In order to obtain the penetration of particles, particle concentration measurements were made at the upper and down streams of a test filter at the face velocities from 0.5 to 10cm/sec (20 ~ 400% of the standard face velocity of 2.5cm/sec).

Figure 2[1] shows the penetration of the HEPA filter A against face velocity as a parameter of aerosol particle size. The penetrations were about 1×10^{-5} in the particle size range of 0.3 ~ 0.4μm at

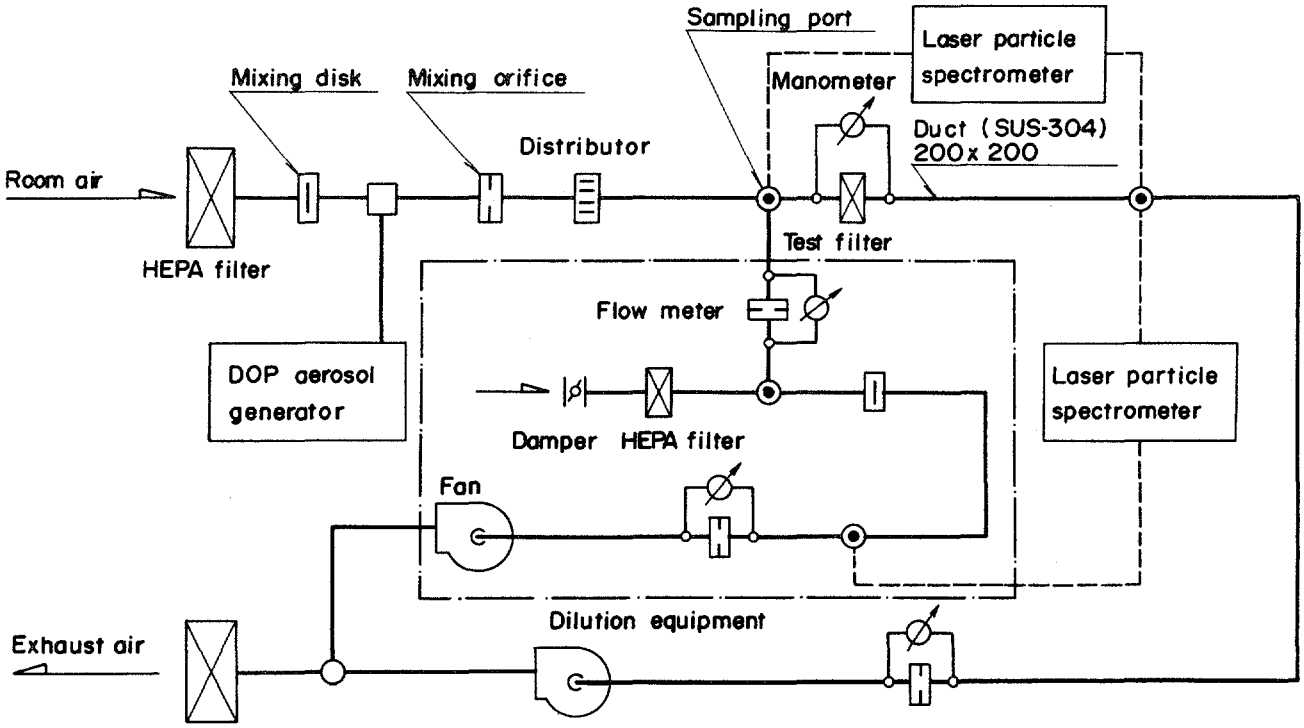

Fig. 1 FLOW DIAGRAM OF TEST APPARATUS FOR DOP AEROSOL PENETRATION OF HEPA FILTERS(1)

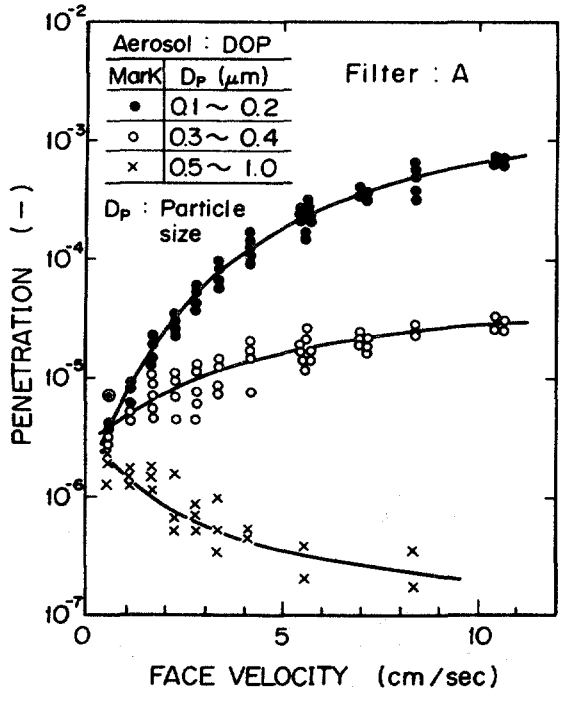

Fig. 2 PENETRATION vs. FACE VELOCITY OF HEPA FILTER A(1)

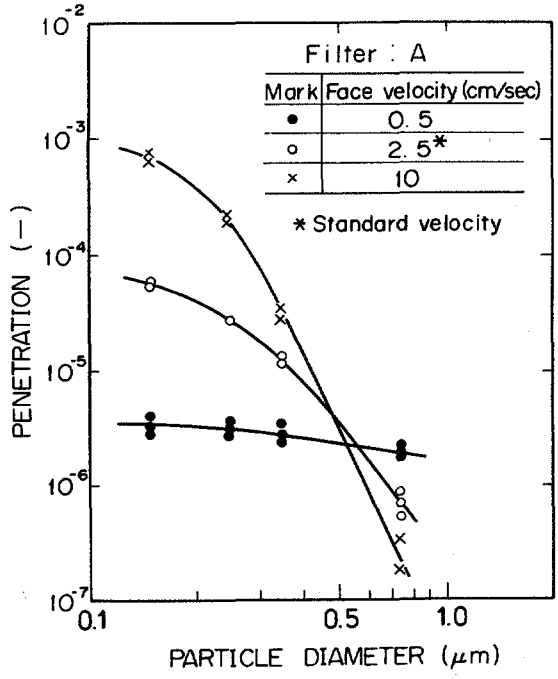

Fig. 3 PENETRATION vs. PARTCLE DIAMETER FOR HEPA FILTER A(1)

the standard face velocity of 2.5cm/sec and about an order of magnitude less than the specified value of 3×10^{-4}, that is, the collection efficiency of more than 99.97%.

As shown in Fig.2, the penetration for the HEPA filter A increases with decrease of particle size in the experimental conditions, and also increases with increase of face velocity in the particle sizes below 0.4μm and decreases with increase of face velocity in the particle sizes over 0.5μm. These results show that the diffusion mechanism is more effective at the sizes smaller than 0.4μm, and that the inertial impaction mechanism is more effective at the sizes larger than 0.5μm. The HEPA filters B and C showed nearly the same penetration characteristics as the HEPA filter A.

Figure 3[1] shows the penetration of the HEPA filter A against mean particle size as a parameter of face velocity. It is clear from Fig.3, that the penetration increases with decrease of particle size in the range of 0.1~1.0μm and the tendency become more remarkable with increase of face velocity.

From these experimental results, it can be concluded that a most penetrating particle size for HEPA filters is smaller than 0.1μm at the face velocities from 0.5 to 10cm/sec, which is similar to the other reports such as the Sinclair's[15].

III. Radioiodine removal from air exhausts

One of the essential problems for trapping radioiodine by adsorption is the deterioration in performance due to weathering of activated charcoal impregnated with chemicals such as potassium iodide, TEDA and stannous iodide.

Decrease in removal efficiency of impregnated charcoal due to weathering was investigated for two kinds of charcoals, BC-727 ($KI-I_2$ impregnated) and HCA (SnI_2 impregnated) using a weathering monitor equipped in the air cleanning system of the JAERI Model Containment building[7].

Figure 4[7] shows the flow sheet of the ventilation system used for the weathering. The iodine sampler for the weathering monitor consists of five test charcoal beds of 1cm in depth and 5cm in inner diameter in series. The test charcoal beds were exposed to the weathering air at a flow velocity of 25cm/sec. The temperature and humidity of the air exhaust were continuously monitored.

The change of $CH_3{}^{131}I$ removal efficiency of the test charcoal beds was measured under a condition of 30°C and 90 ± 5% R.H.. The test results are shown in Fig.5[7]. The broken lines in the figure are the curves estimated from the data of the last three downstream beds. It is estimated from the figure that 10cm and 15cm thick impregnated charcoal beds have removal efficiencies of more than 90% and 95%, respectively, after one year's continuous service in the air exhaust system.

The poisoning of silver zeolite, silver silica-gel and silver alumina was investigated for the application to the removal of

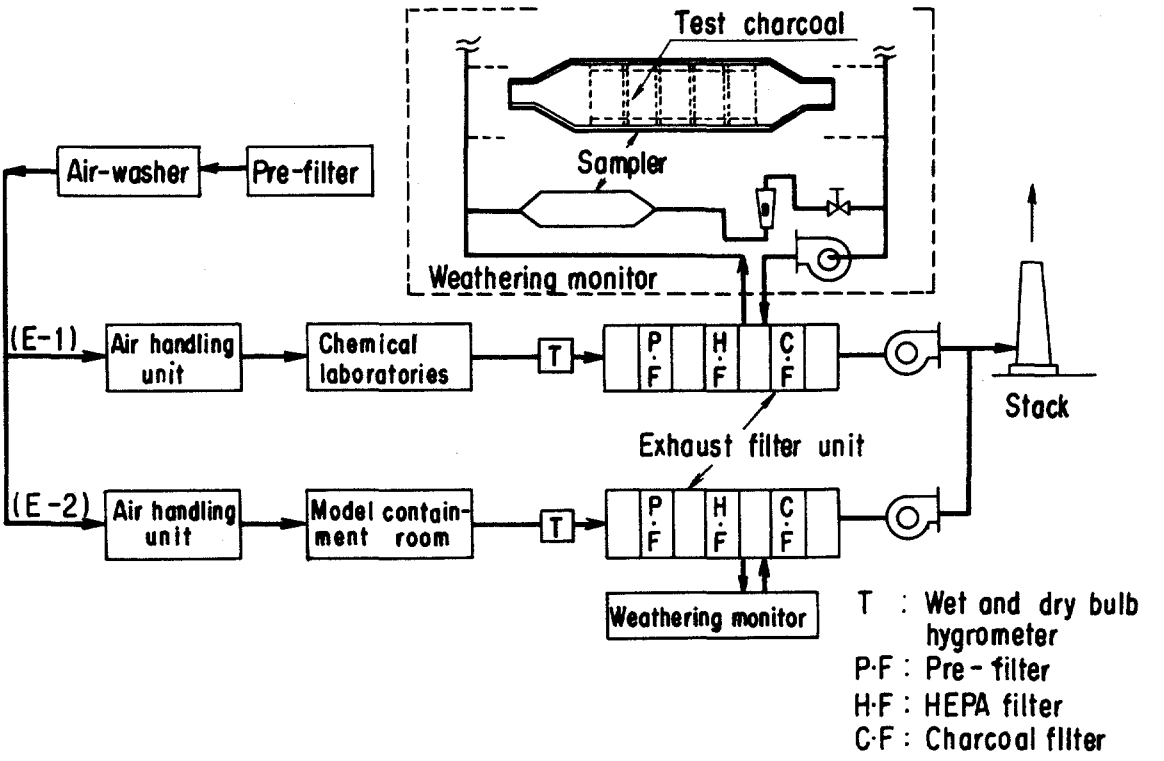

Fig. 4 FLOW SHEET OF WEATHERING TEST APPARATUS[7]

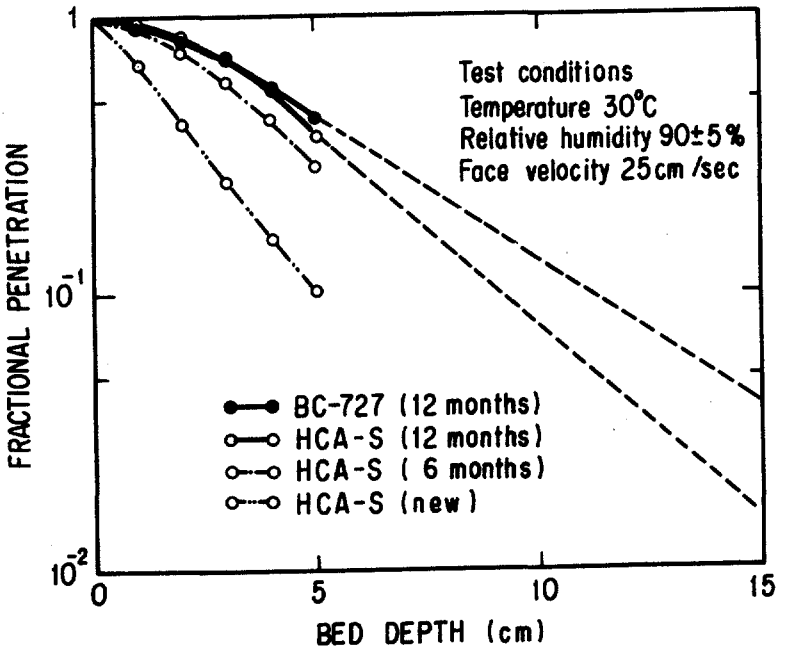

Fig. 5 $CH_3{}^{131}I$ TRAPPING CAPABILITY OF CHARCOAL BEDS WEATHERED IN CONTINUOUS AIR FLOW[7]

radioiodine from the off-gas of reprocessing plants[8]. The size of test adsorber was 2.2cm in inner diameter and 10cm in depth. The air conditioned with temperature, humidity and NO_2 concentration was passed through the adsorbers at 20cm/sec for 3 hours and the removal efficiency for $CH_3{}^{131}I$ vapor was measured. In the condition of 40°C, 70% R.H. and the flow velocity of 20cm/sec, the efficiency remarkably decreased when exposed to the air containing NO_2 of more than 100ppm. It was considered that the deterioration of adsorbers was due to HNO_3 formation by the reaction of NO_2 adsorbed and water trapped on silver impregnated inorganic adsorbers. Under the condition of 50°C and 37% R.H., silver zeolite, silver silica-gel and silver alumina indicated the removal efficiencies of 99.2~96.3%, 99.2% and 99.4%, respectively.

IV. Operational performance of the incineration plants using ceramic filters

The incineration plant with ceramic filters for low level radioactive wastes was originally developed at the Karlsruhe Nuclear Research Center, West Germany[5].

In Japan, experimental tests were made using a test incinerator with Japanese ceramic filters in the JAERI in corporation with the NGK Insulators, Ltd., in order to obtain data on operational characteristics of the system[2]. It was estimated from the experimental results that an overall decontamination factor of the test incinerator was $10^5 \sim 10^6$ for the simulated wastes contaminated with ^{58}Co, ^{85}Sr, ^{132}Cs and ^{32}P.

Based on the experimental results in addition to the Karlsruhe design, several incineration plants using ceramic filters with some modifications in the system have been constructed for incinerating low level radioactive wastes in Japan[3][4]. As an example of these plants, Fig.6[4] shows a flow diagram of the plant in the JAERI which has been operated since 1979.

Main design specification of the JAERI's plant is as follows; incineration capacity: approximately 100kg/h, operating temperature of the primary- and secondary- ceramic and HEPA filters: 550-950°C, 650°C and 250°C, respectively and each ceramic filter is composed of 92 candle type filter elements.

Main features of these plants are improvement in the filter element suspension and material of ceramic filters, use of the back-flow system in the primary ceramic filter to decrease pressure drop through removing soot trapped on the elements. These are useful for extending life and decreasing penetration of ceramic filters. The incineration plant in Tsuruga Power Station, the Japan Atomic Power Co. Ltd. (completed in 1977), for example, has shown good performance on decontamination factor and pressure drop without changing ceramic filter elements for more than 2000 hours operation (two years)[3].

In the JAERI's plant, the decontamination factor, that is, the ratio of radioactivity in a burned waste to that in the outlet of the secondary ceramic filter was 3.4×10^6 for a simulated waste contaminated with ^{137}Cs of about 75mCi (580kg)[4]. The decontamination factor measured in the plant at Tsuruga Power Station was $5 \times 10^5 \sim 10^6$ for

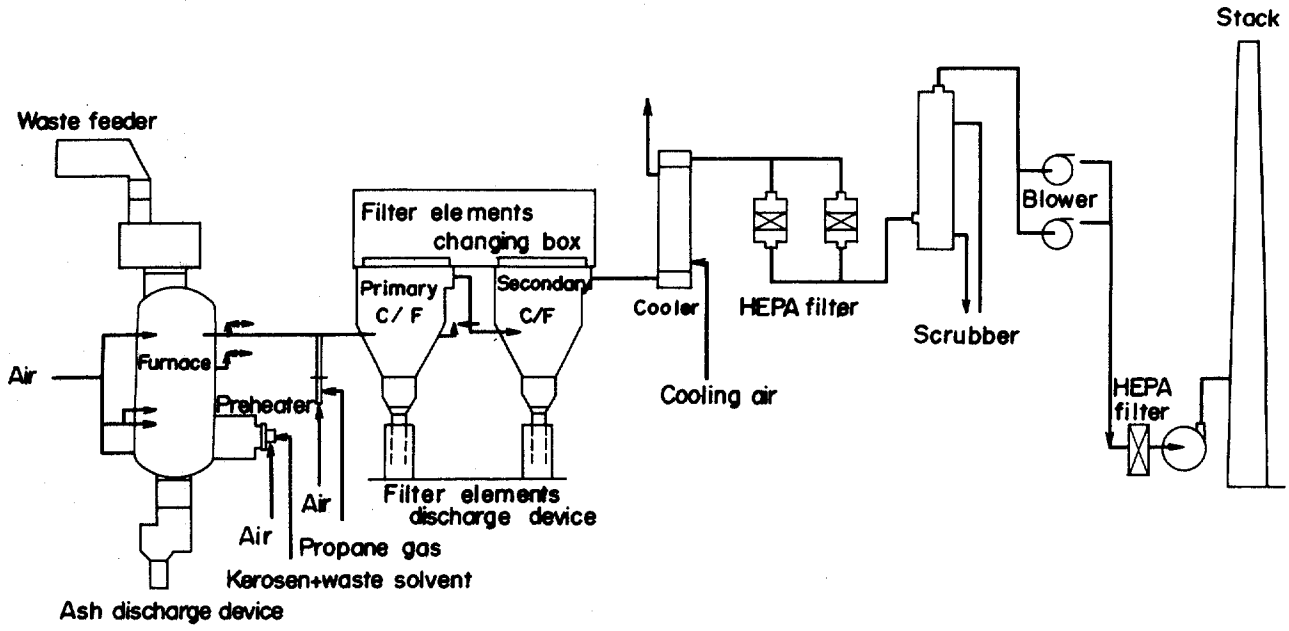

Fig. 6 FLOW DIAGRAM OF THE INCINERATION PLANT WITH CERAMIC FILTERS IN THE JAERI(4)

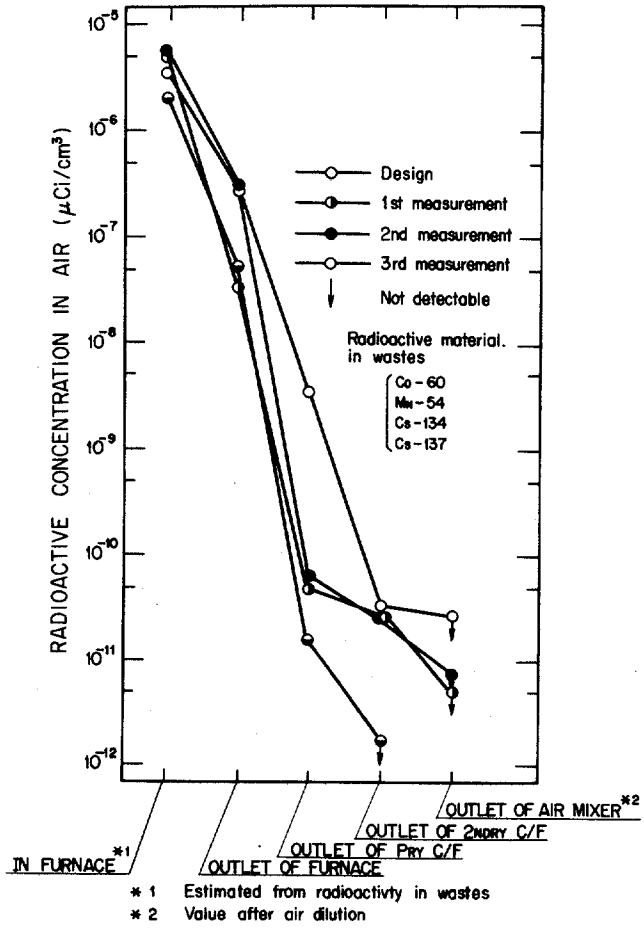

Fig. 7 DECONTAMINATION FACTORS OF THE INCINERATION PLANT USING CERAMIC FILTERS(3)

the wastes contaminated with ^{60}Co, ^{54}Mn, ^{134}Cs and ^{137}Cs as shown in Fig.7[3]. It was proved from these results that the decontamination factor of these incineration plants is more than 10^5 for nonvolatile radioactive materials in low level wastes although it depends on operating conditions.

References

(1) Nuclear Fuel Facility Filters Committee of JACA (S.Kitani,et al) : Studies on Safety of High-Efficieney Air-Filter in Nuclear Facilities, J. Japan Air Cleaning Association,18 (4), 1 (1980).

(2) C. Machida, K. Kato, Y. Karita : Development of an Incinerator Using Ceramic Filters for Low Level Radioactive Solid Waste Treatment, JAERI 1244, (1976).

(3) Y. Meguro, et al : Operational Experience and Performance of New Radwaste System at Tsuruga Power Station, The Thermal and Nuclear Power, 31, 417 (1980).

(4) K. Hirayama, K. Kamei, et al : Construction and Test Operation of the Inceneration Flant for Low Level Radioactive Wastes, Proc. 1980 Fall Meeting of the Atomic Energy Society of Japan, Sendai, October (1980).

(5) Bähr, et al : Survey of Operating and Development Work for Treatment and Disposal of Radioactive Waste at the Karlsruhe Nuclear Center, KFK 1455, p.9 (1971).

(6) Y. Ikezawa, Y. Yoshida, S. Kitani and M. Kozuka : Studies on Weathering of Impregnated Charcoals Used in Reactor Filter Systems, J. Japan Air Cleaning Association, 10 (2), 71 (1972).

(7) S. Kitani, et al : Weathering of Iodine Adsorbers by Continuous Air Flow, Proc. 1980 Annual Meeting of the Atomic Energy Society of Japan, Nagoya, Mar. (1980).

(8) T. Hoshino, et al : The Poisoning of Nitrogen Dioxide to Iodine Adsorbers, Proc. 1979 Fall Meeting of the Atomic Energy Society of Japan, JAERI, Oct. (1979).

(9) Y. Yuasa, et al : Selective Adsorption-Desorption Method for the Enrichment of Krypton, Proc. 13th AEC Air Cleaning Conf. p.177 (1974).

(10) T. Kanazawa, et al : Development of the Cryogenic selective Adsorption-Desorption Process on Removal of Radioactive Noble Gases, Proc. 14th ERDA Air Cleaning Conf. p.964 (1976).

(11) K. Nakajima, et al : Removal Processes for Radioactive Rare Gas of Fuel Reprocessing Plant, J. Atom. Energy Soc. Japan, 22 (8), 463 (1979).

(12) S. Tanaka, et al : Removal of Tritiated Water Vapor by Adsorption, J. Nucl. Sci. Technol. 13 (5), 251 (1976).

(13) H. Yoshida, et al : Mock-up Test of a Tritium Removal System Using D_2-Gas, Proc. 1980 Fall Meeting of the Atomic Energy Society of Japan, Sendai, Oct. (1980).

(14) Y. Naruse, et al : A Conceptual Design of Tritium Handling Facility, ANS Topical Meeting, Tritium Technology in Fission, Fusion and Isotopic Applications, Dayton, U.S.A. (April 29 - May 1, 1980).

(15) D. Sinclair : Penetration of HEPA Filters by Submicron Aerosols, J. Aerosol Sci., 7, 175 (1976).

DISCUSSION

ETTINGER: I will summarize what the seven speakers have said by commenting that, what we have just heard is really a microcosm of the entire meeting over the past few days. Perhaps more accurately, the topics which have been covered by the different speakers read like a table of contents from Mr. Burchsted's index of all the past Air Cleaning Conferences. They cover topics ranging from in-place testing, iodine, sintered filters, electrostatic participators, noble gases, methyl iodide, laser particle spectrometer, et al. If I sat down and tried to check off all the topics covered in this session, it might match the program that has been given at the Conference. To me, this indicates the importance of communication between researchers and operating people in the United States with people doing similar tasks in Europe, Asia, Canada, and other parts of the world. That, actually, is very much an integral part of this meeting. What the Program Committee should be proud of is that a significant fraction, approximately one-third of the papers presented here, have come from foreign countries. With those few words, I would like to open the meeting for a general discussion with our panelists.

DEUBER: I have a question addressed to Mr. Rouyer. What is the experience with your ruthenium filter consisting of polyethylene?

ROUYER: We have no experimental results now. We have conducted a bibliographical survey.

GILBERT: I would like to address a question to Dr. Yoshida. Was the face velocity for the charcoal weathering, 25 cm/s or 2.5 cm/s?

YOSHIDA: 25 cm/s.

GILBERT: I assume that you accelerated the aging of your

charcoal by using a greater than normal air flow. That is, you aged it faster than you would have had the charcoal been in normal service?

ETTINGER: As a velocity of 25 cm/s is higher than normal, was it your intention to accelerate the results?

KITANI: I believe that the velocity of the flow rate through the carbon bed was 20 cm/s. The comment of Mr. Gilbert is very interesting, as it confirms that the relationship of the rate of aging is connected with such a velocity.

WILLIAMS: Dr. Furrer, are you able to say anything about iodine corrosion within the PASSAT facility?

FURRER: Yes, when we worked at higher iodine concentrations of about 1.1 g/m^3, we found corrosion in our PASSAT test rig. Now we intend to use another steel so that we will have no more corrosion. We tried many steels and now we have found one which shows no corrosion against this high iodine concentration in an NO_2 atmosphere.

WILLIAMS: Are you trying to work at iodine concentrations where you are not going to get plate out?

FURRER: We had plate out. If you have temperatures between 30° and 150° and a wet path, there will be a condensation of droplets and plate out of iodine. After 5 hours of operation, the iodine will have passed to the filters.

WILLIAMS: Are you willing to say what steel you are using?

FURRER: Yes, it is a German specification, number Inconel 625.

HENRICH: In a real plant about 10% of the fission product iodine is fission product bromine; concerning halogen corrosion problems, this should not be neglected.

ANON.: Dr. Yoshida, with regard to the tests of silver impregnated gel, I did not understand your comment that 100 ppm, or less, of NO_2 reduced the efficiency of the adsorbers, but above that, in a saturated atmosphere, the efficiency of the material did not decrease. Is that correct?

YOSHIDA: When the air contained 100 ppm of NO_2 and the relative humidity was 70%, after 3 hours of continuous operation, efficiency decreased very much. But when the air contained 100 ppm of NO_2 and a relative humidity of 40%, efficiency did not decrease after 3 hours of continuous air flow.

ANON.: At the lower humidity, was the efficiency better?

YOSHIDA: Yes.

ANON.: I have a question for Mr. Olivier of OECD. Do you have a management policy concerning which of the radioisotopes, iodine-129, carbon-14, tritium, krypton, etc., needs to be handled first? What order of importance does OECD place on these?

OLIVIER: I have a lot of details in the report I have with me if you would like to see it. It is difficult for me to give you an idea of priorities because, first of all, there are several aspects which have not been considered in the overall cost benefit analysis, such as the safety of storage. This is, perhaps, more critical with regard to krypton-85 than it would be for tritium. The dose to operators has not been taken into account either. This is why the group that wrote this report did not come up with precise recommendations with regard to which isotopes should be retained first. If you look at the report of the study, you can arrive at your own conclusions, because it is presented in a way which gives the maximum dose rate to the most exposed individuals, on one side, and on the other side, the minimum cost figures per man-rem saved. Therefore, from that you can really get some idea which should come first.

ORTH: With respect to the Helex process for the detritiation of water, can you give us any idea of the size of a unit or the capacity of the unit, in terms of the size of a typical plant that would be adequate for a one metric ton per day or a five metric ton per day processing plant?

COLLARD: It is difficult for me to answer, as I am not directly connected with this work. To give you an idea, the electrolizer used up to now has a power of 1.4 kw. A pilot is being built with an electrolizer of 80 kw so that the extrapolation can lead to the design of an installation needed by a 60 ton per year reprocessing plant. Perhaps you may wish to refer to a better document, which is a summary of the presentation of Mr. Bruggeman in Vienna this year. The number of the document is IAEA SM-245/52. I think you will find the latest public data in this document.

FIRST: I would like to address this question to the panel in general. At the 14th and 15th Air Cleaning Conferences, Dr. Moeller presented papers on Licensee Event Reports and, more recently, the US Regulatory Commission issued a report on plant failures. My question is, are there similar requirements in other countries for prompt reporting of nuclear power plant failures and, if so, what is the experience?

OLIVIER: In our Committee on Safety of Nuclear Installations, we have recently set up an Incident Reporting System which is called the IRS. It is a system which, in principle, works between all the OECD countries. They are supposed to report promptly all incidents that happen in their power stations. I should say that at the start, it was a bit difficult to implement, because some countries with particular regulations were sometimes reluctant to do that. The system has been in existence for about a year now and things are improving. We met about two weeks ago in Paris and agreed on a standard format. Something is going on in this field, and hopefully with time, we will increase the contact between the various countries in this respect, and this should be done for the entire fuel cycle.

YOSHIDA: In Japan, the incidents are reported to the government. But after the TMI accident, statistical analysis is now recommended by the Committee in the government. Now all incidents are reported. But how we are to analyze and use the information effectively is now under discussion in our country, and this is the

question for the nuclear industry.

MARSHALL: In Britain also, of course, we have to report an incident to the Nuclear Installations Inspectorate. But I do not have any specific first-hand knowledge of this. I wonder if any of my colleagues would like to add anything.

WILLIAMS: I am not going to give a lot of facts and figures, because I cannot remember them all, and I am not a health physicist. We have a series of reporting requirements, depending upon the seriousness of the accident. There are various pre-defined features of any given accident which will decide the level to which it has to be reported and the time scale over which it has to be reported. The big things, if ever they came about, would have to be reported within hours to government bodies. These are all defined in advance.

FIRST: My next question is directed in particular to Mr. Olivier. We heard your description of OECD's activities and also that you cooperated with the International Atomic Energy Agency in the seminar in Vienna in February; you obviously cooperate in other areas. My question is, does OECD duplicate the work of IAEA? What is its special function that is not covered by the international organization?

OLIVIER: That is a good question, and we are used to giving an answer to it. The IAEA, as you know, covers the entire world, and you may have noticed that in the last few years the developing countries have gotten more and more power within IAEA. IAEA certainly has a strong influence on their programs and, therefore, the IAEA program is more and more reoriented toward assistance to developing countries, preparation of guidebooks and safety codes in general. IAEA has a specific program called Nuclear Safety Standards, which is not of major interest to developed countries. On the contrary, as far as we are concerned within OECD, we represent a group, the biggest part of which is composed of developed countries like North America, Japan, and many of the European countries. We tend to focus on issues which are of direct interest to these countries, like aerosols, for instance, which I have mentioned and for which we recently produced a state of the art report. Mr. Hilliard could talk about it, as he is a member of the group responsible for that work, and which is really at the forefront of knowledge in this field. So, from that point of view, although we cooperate on some specific activities like the organization of this meeting in Vienna that you mentioned, we are essentially trying to concentrate on the most advanced problems, if I may use that expression, whereas the IAEA in Vienna is more and more helping the developing countries.