SESSION 2

REPROCESSING

Monday:	August 24, 1992
Co-Chairmen:	R. T.Jubin
	Y. Kondo

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DISSOLUTION OFF-GASES AT THE MARCOULE PILOT FACILITY: IODINE TRAPPING AND OFF-GAS CHARACTERIZATION UNIT

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Abstract

The Marcoule Pilot Reprocessing Facility (APM) reprocesses spent fuel from light water reactors and fast breeder reactors. A batch dissolution process is used with an annual throughput capacity of 5 metric tons. The off-gas treatment unit is described together with its characterization laboratory in order to highlight the functions and potential of the facilities. The objectives are consistent with the Marcoule site policy regarding diminished iodine release and investigation of the off-gas treatment process. The equipment used to meet these objectives is described from a functional standpoint. The facility implements measurement techniques to allow continuous quantitative measurements of nitrogen oxides, oxygen, iodine and krypton, as well as continuous monitoring of the demister inlet flow by γ spectrometry. Sorbents used for iodine trapping may be tested over a wide range of operating conditions (temperature, flow rate, iodine concentration) with representative dissolution off-gases. An X-ray and γ counting system is used to assess the activity of the adsorbed radionuclides, notably ^{129}I.

Introduction

The Marcoule Pilot Reprocessing Facility (APM*) reprocesses spent fuel from light water reactors and fast breeder reactors, with a throughput capacity of 5 metric tons each year[1]. It is equipped to ensure final iodine trapping on solid media, and to allow investigation and monitoring of the off-gas stream.

The process head-end facility (Building 214) was initially intended to house two dissolution lines, one using a batch process and the other a continuous process. The off-gas treatment unit was therefore designed to accommodate both lines. Currently only the batch dissolution line is operational, with purification of gases from the tank vents and the liquid waste treatment unit vents. A solid-medium trapping unit was recently installed to handle iodine in the process off-gas line. The present configuration of the dissolution off-gas facility is shown schematically in Figure 1. Sampling valves at the inlet of each process device allow a fraction of the gas stream to be diverted to the characterization laboratory, equipped for off-gas monitoring and analysis as well as for testing all types of sorbent media.

This paper describes the iodine trapping unit and the off-gas characterization laboratory, and discusses their role in achieving a fuller understanding of gas purification operations, and in the development of new gas purification systems.
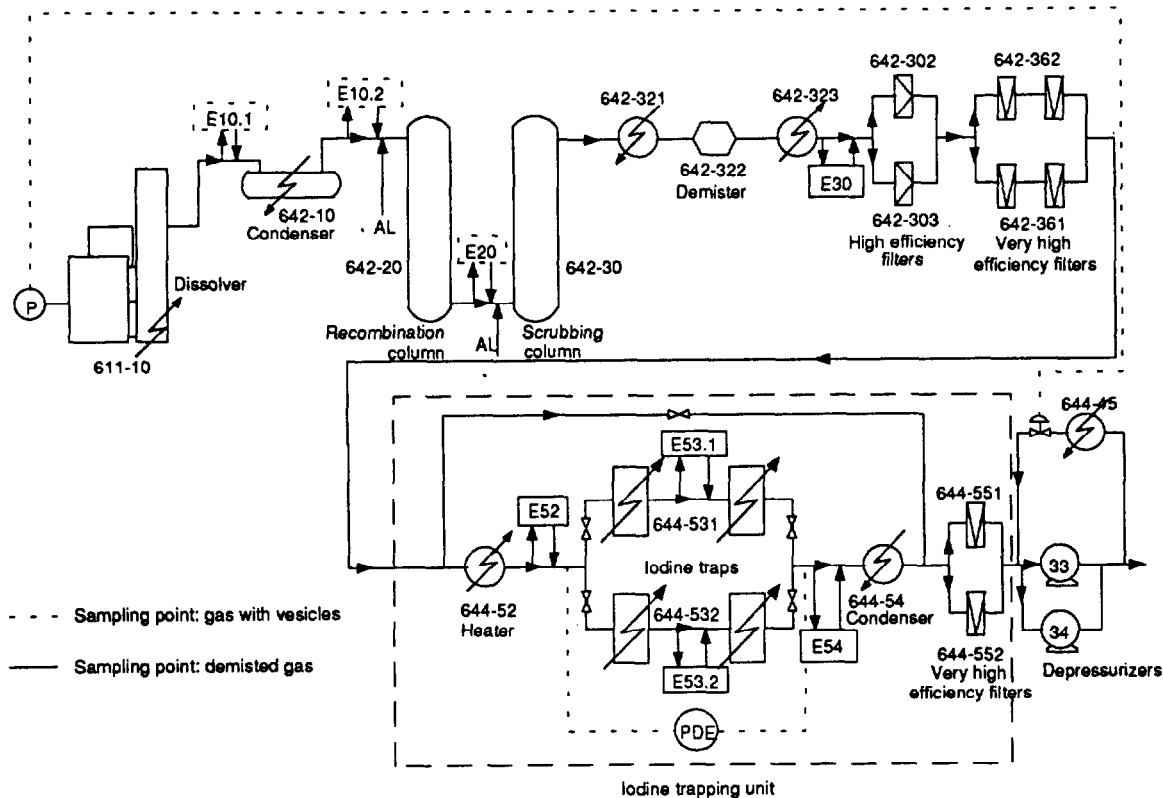
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* *Atelier Pilote de Marcoule*

Figure 1 – Dissolution Off-Gas Treatment Schematic

Objectives

Iodine Trapping Unit

The iodine trapping unit is designed to eliminate iodine from the gas stream by chemisorption on a solid medium. The objectives for this system include:
- checking the efficiency of iodine trapping from the dissolution off-gas stream on various solid media impregnated with silver, under actual media aging conditions;
- characterizing the saturated trap for conditioning and disposal;
- minimizing [129]I release from the APM.

Off-Gas Characterization Laboratory

Analytical facilities available for the dissolution off-gases mainly include the following:
- monitoring of the solid-medium iodine trap by measuring the released iodine quantity;
- off-gas data acquisition, limited to the major fission products (iodine, rare gases and semivolatile fission products) released during the process head-end procedures, and to nitrous fumes generated during fuel dissolution;
- qualification under radioactive conditions of new sorbent materials for iodine trapping;
- qualification of the continuous monitoring methods.

Off-Gas Monitoring and Data Acquisition
Fission products and nitrous fumes are monitored.

Fission Products: Iodine measurements provide data concerning iodine metabolization throughout the head-end units and iodine trap operation in the off-gas line. The released ^{85}Kr is analyzed to validate the computer code, and monitored to determine the proportion of krypton released cutup and dissolution. Aerosol entrainment may result in the presence of other fission products in the off-gas: aerosol formation and behavior mechanisms are investigated to optimize system performance and to estimate the contamination level of process wastes (filters, solid traps).

Nitrous Fumes: The behavior of nitrous fumes may be investigated from the standpoint of the release kinetics and the recombination efficiency. This information is crucial in interpreting the results obtained with the sorbent materials.

Qualification of New Sorbents Iodine trapping on solid sorbents has been selected as the iodine retention method in reprocessing plants. Solid sorbent media are investigated for several reasons: to confirm their sorption capacity and efficiency on a variety of representative off-gases, to assess material aging, and to evaluate sorbent behavior and resistance under actual operating conditions with all the other radionuclides in chemically representative form.

Development of Continuous Monitoring Methods Methods developed in outside laboratories (e.g. continuous gaseous iodine measurement by laser fluorescence) are qualified under actual operating conditions.

Iodine Trapping Unit

Overview

The iodine trapping unit is shown schematically within the overall off-gas system in Figure 1. It includes the facilities necessary for iodine trapping on a solid medium, with inlet and outlet gas treatment and outlet gas filtration prior to recycling through the main circuit, conditioning of the media in containers, and container storage pending disposal. Two traps are placed in series inside a sealed vessel. This is a research & development unit, and may be switched into or out of the dissolution off-gas process stream. The heating system is designed to allow operation at temperatures ranging from 100 to 200°C. The nature of the solid medium may vary, and its dimensions may be modified to a limited extent to cover a range of off-gas flow rates.

The APM facility is used to reprocess fast breeder and light water reactor fuel. Table I indicates the volatile material concentrations in both types of fuel. The trap retention rates were determined from these data, allowing for 10 wt% silver content in the reference sorbent, and considering 40 mg of iodine per kilogram of sorbent as the best tradeoff between the saturation rate and purification efficiency.

The process throughput capacities were determined by assuming there would be no soda scrubbing. The process gas flow rate may range from 10 to 25 $Nm^3 \cdot h^{-1}$; with the standard trap (Figure 2) this corresponds to a flow velocity of 3.8 to 10 $cm \cdot s^{-1}$.

Description

The two iodine trap lines each comprise two stainless steel casings 0.5 m in diameter and 1 m high installed in series in the off-gas system and mounted on separate frames (Figure 2). Because of differential expansion behavior, the casings are not mounted directly on the glove box; leaktightness is provided between each casing and the penetration at the top of the glove box by a lip seal. The leaktight cover of each casing is accessible from inside the glove box. Each casing includes a center-mounted

Table I – Volatile Content of LWR and FBR Fuel (per metric ton of U + Pu)

Fuel Characteristics		LWR	FBR
Burnup (MWd·T^{-1})		33 000	85 000
Fissionable material		3.5% ^{235}U	$\dfrac{Pu}{U + Pu} = 28\%$
Cooling time		3 years	1 year
Krypton ^{85}Kr	(g)	359	588
	(Bq)	2.92×10^{14}	7.365×10^{14}
	(Ci)	7.901×10^3	19.905×10^3
Xenon	(g)	4360	10 332
Iodine ^{129}I	(g)	208	623
	(Bq)	1.106×10^9	3.071×10^9
	(Ci)	2.99×10^{-2}	8.3×10^{-2}
Quantity dissolved per batch (kg U + Pu)		90	63
Number of dissolutions per trap		36	18

temperature sensor accessible from the outside. The gas temperature is monitored by a sensor at the outlet of each casing.

The outer wall of each casing and the off-gas lines are insulated. Six insertion heaters are located around the periphery of the gas inlet manifold to provide uniform heating. The cover of each casing
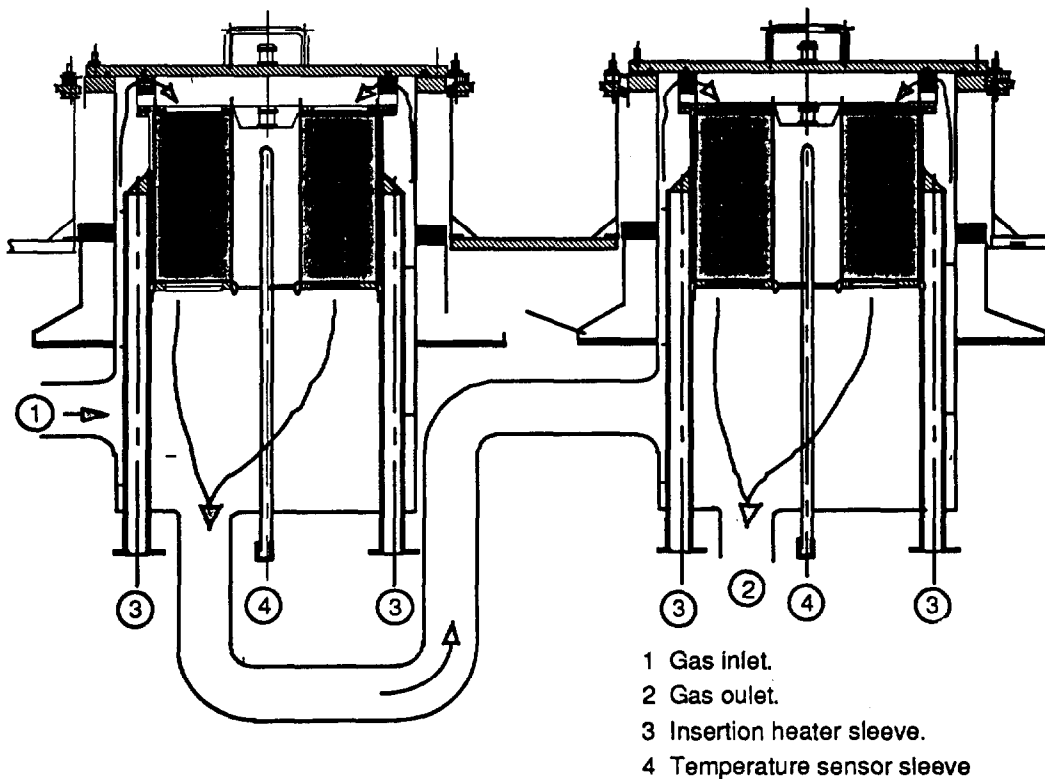

1 Gas inlet.
2 Gas oulet.
3 Insertion heater sleeve.
4 Temperature sensor sleeve

Figure 2 – Iodine Traps

includes two plugs providing access to the interior of the casing for R & D devices, such as gas samplers to determine the nitrogen oxide content in the immediate vicinity of the sorbent. The standard trap (Figure 3a) comprises a stainless steel filter cartridge 371 mm in diameter and 290 mm high with grip provisions. As mentioned above, the trap geometry may be modified. The trap configuration used for LWR fuel (Figure 3b) is designed to approximate the flow velocity of the La Hague reprocessing plants and to determine the concentration profile, and thus qualify the sorbent behavior according to the degree of saturation.

The unit includes handling provisions to install and remove the traps and other process equipment. The saturated traps are conditioned in stainless steel containers and placed in dry interim storage pits.
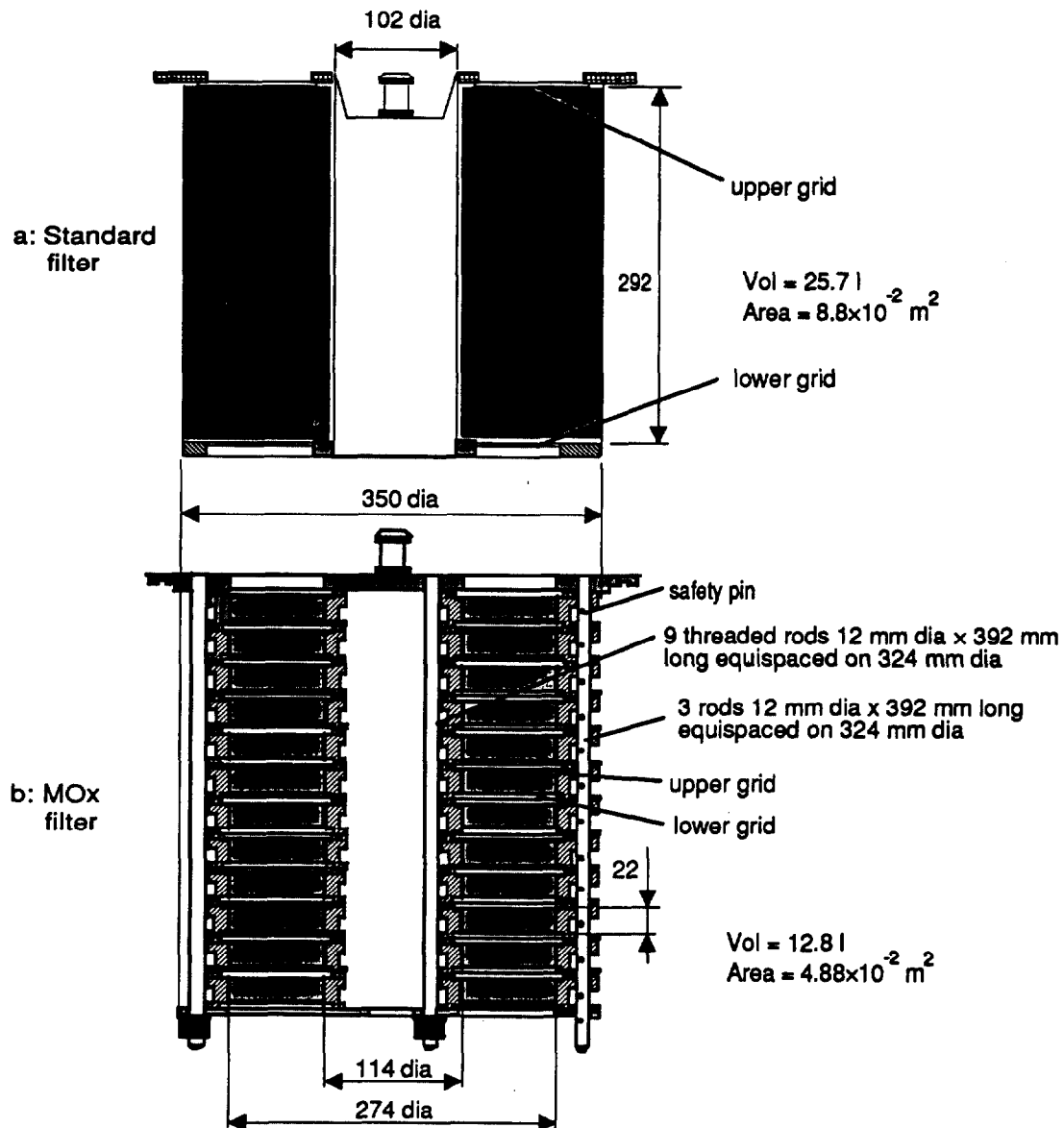

Figure 3 – Iodine Trap: (a) Standard Filter, (b) MOx Filter

Off-Gas Characterization Laboratory

Overview

From the main unit (Figure 1) gas samples may be transferred to the off-gas characterization laboratory. Because of the large number of sampling points, two glove boxes are required to distribute the flow lines and to ensure maximum sampling flexibility (Figure 4). Each glove box accommodates a separate category of sample inlets:

- category A (glove box 643.10) includes the sampling points upstream from the demister. Because of the potentially high activity level, these samples are first filtered and the activity is monitored in a shielded vessel. The samples containing vesicles are distributed to two specific measurement channels reserved for the most active samples (A1 and A2).

- category B (glove box 643.11) includes the sampling points downstream from the demister. The demisted samples are distributed to two specific channels (B1 & B2). An analysis channel (C) is reserved for continuous measurement of iodine leakage from process traps.

The specific gas flows (A1, A2, B1, B2, C) are routed through several glove boxes for characterization and investigation of iodine trapping on different media. After flow rate measurement, the gases are pumped back to the unit via the two above-mentioned glove boxes: the most active samples (category A) are routed via glove box 643.10, while categories B and C transit through glove box 643.11. The analyzed gas streams are reinjected 0.3 m downstream from the sampling point.
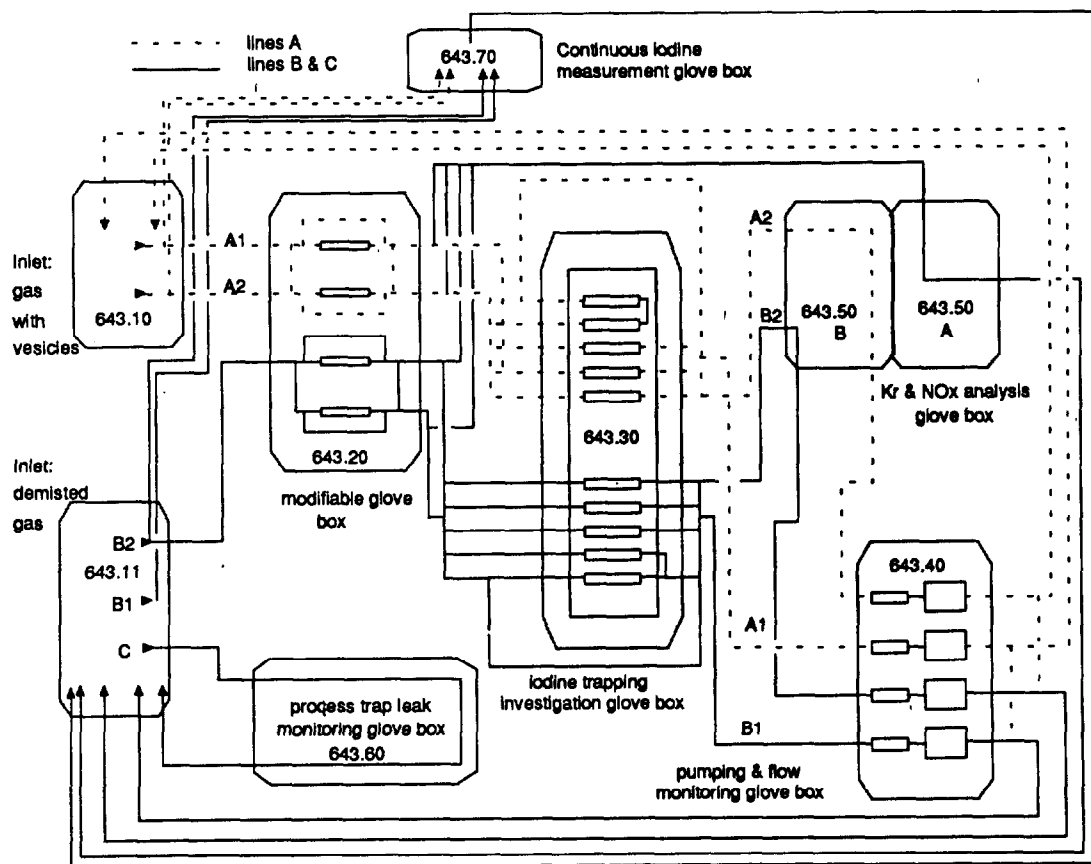

Figure 4 – Characterization Laboratory Flowsheet

Table II – Characterization Laboratory Flow Properties

Group	Sampling position	Ref.	Composition	Flow rate	Pressure	Humi-dity	Temp. (°C)
A	Batch dissolver outlet	642 E 101	NO_x-I_2-Ru-RG	$1\ m^3 \cdot h^{-1}$	Neg	Humid	90
	Condenser outlet	642 E 102	NO_x-I_2-Ru-RG	$1\ m^3 \cdot h^{-1}$	Neg	Humid	25
	Recombination column outlet	642 E 20	I_2-RG-Ru	$1\ m^3 \cdot h^{-1}$	Neg	Humid	30
	Column 644-40 inlet	644 E 30	NO_x-I_2-Ru-RG	$1\ m^3 \cdot h^{-1}$	Neg	Humid	30
	Column 642-40 inlet (Vent treatment)	642 E 401	I_2-RG-Ru	$1\ m^3 \cdot h^{-1}$	Neg	Humid	30
B	Column 642-40 outlet (Vent treatment)	642 E 402	I_2-Ru-RG	$1\ m^3 \cdot h^{-1}$	Neg	Dry	25
	Column 642-30 outlet (H_2O)	642 E 30	I_2-RG-Ru	$1\ m^3 \cdot h^{-1}$	Neg	Dry	25
	Inlet, internal and outlet	644 E 52	I_2-RG	$1\ m^3 \cdot h^{-1}$	Neg	Dry	150
		644 E 531	RG	$1\ m^3 \cdot h^{-1}$	Neg	Dry	150
	(Solid-media trap)	644 E 532	RG	$1\ m^3 \cdot h^{-1}$	Neg	Dry	150
		644 E 54	RG	$1\ m^3 \cdot h^{-1}$	Neg	Dry	100
	Dissolution off-gas treatment outlet	642 E 33	I_2-RG	$1\ m^3 \cdot h^{-1}$	Pos	Dry	25

All the gas aliquots contained β and γ emitters
RG: Rare gases

Description

Flow Control Glove Boxes The vesicle-laden gas arrives in glove box 643.10 (Figure 4) and the demisted gas in glove box 643.11 (sample sources are indicated in Table II). These glove boxes distribute the gas flow from process vents to and from the off-gas characterization laboratories via specific channels A1, A2, B1, B2 and C (channel C is reserved for monitoring of iodine leakage from process traps). Glove box 643.10 provides safety and detection functions by means of filters and scintillation detectors.

The activity is monitored and trapped to limit the activity concentration in the gases supplied to the other glove boxes and thus avoid any irradiation hazard in the remainder of the laboratory, and to quantify radionuclide entrainment from each process step. Two NaI scintillation detectors (Figure 5) are installed in a lead chamber in each line. One is located in a gas flow line and designed for continuous gamma spectrometry; the crystal is machined to a maximum thickness of 0.3 mm. The other detector is located above the demister to assess the vesicle irradiation level; if the background level reaches 50 $mrad \cdot h^{-1}$ the feed pump and inlet valve are automatically secured. The vesicle traps are removable and may be transferred in lead-shielded containers to a γ and X-ray spectrometry unit to measure the cumulative vesicle activity due to each element for correlation with the scintillation monitoring.

Trapping Glove Boxes The system includes two glove boxes for investigation of new iodine trapping sorbent materials. Glove box 643.20 (Figure 6) is a modifiable unit that can also be used for other purposes. The preheated inflowing gas stream is directed to the traps through a flexible stainless steel duct fitted with Leybold couplings. Each trap consists of twelve stages 50 mm in diameter and 15 mm thick, heated by a 1500 W thimble and operating in the 100–200°C range within ± 2°C.
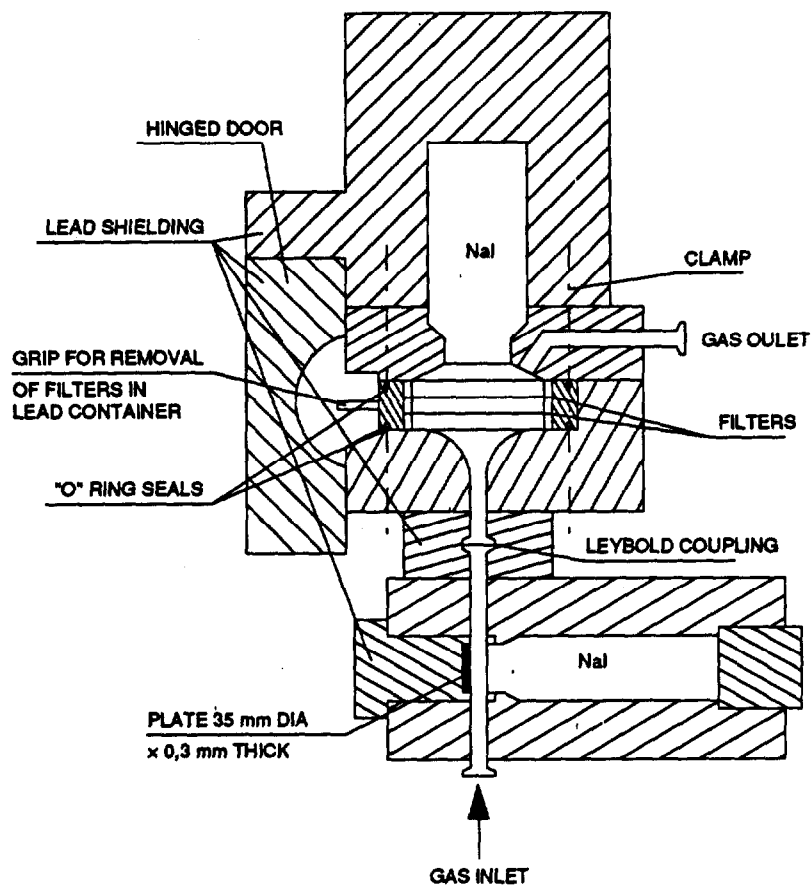
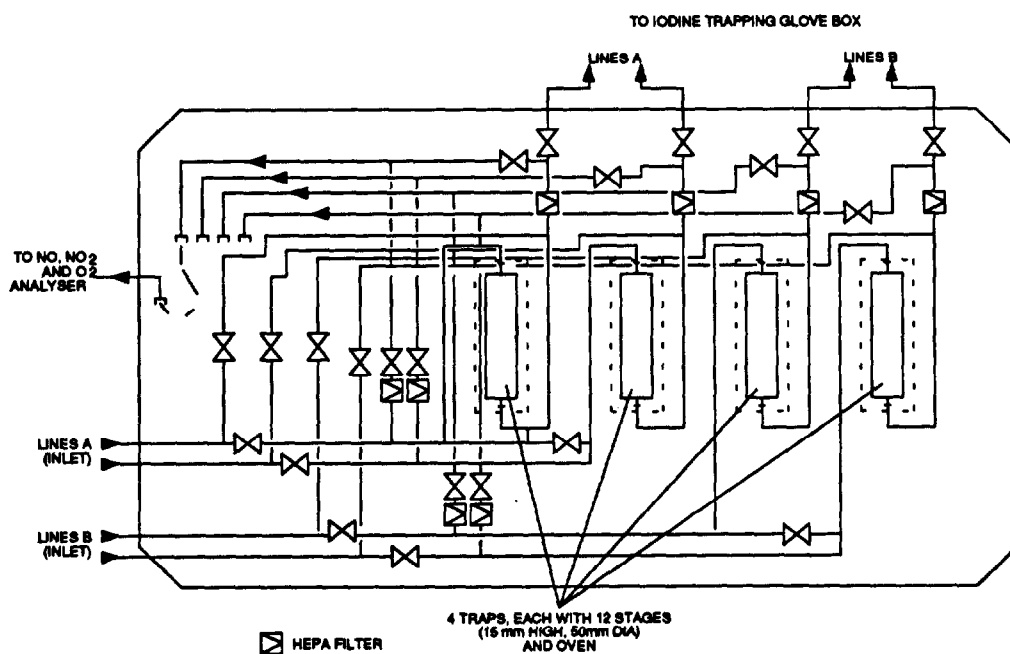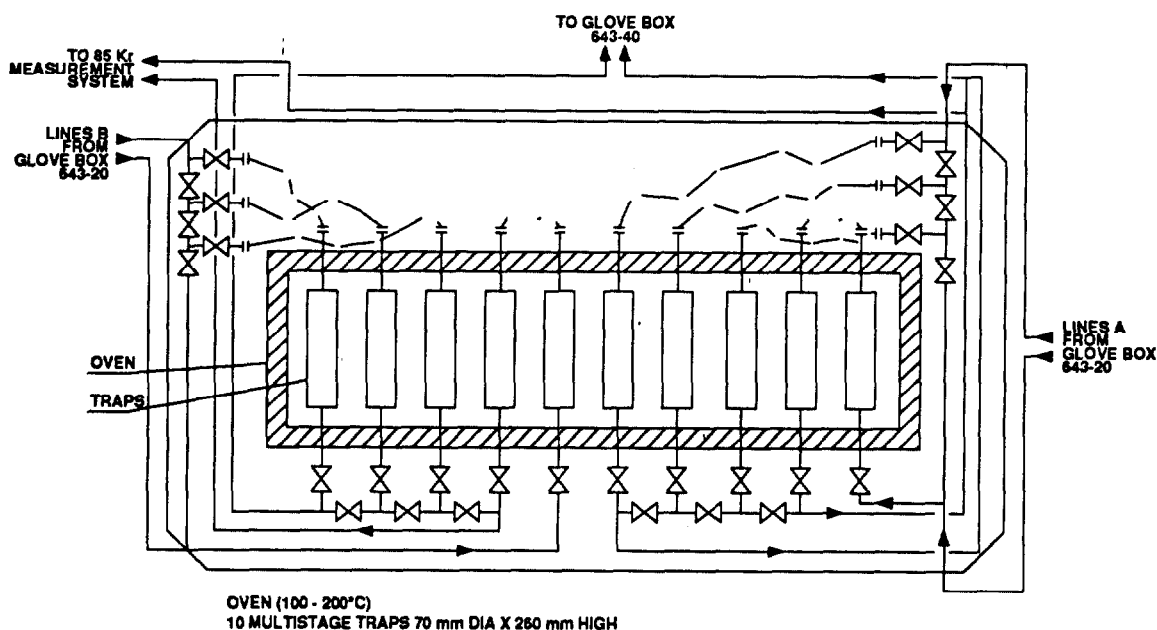Figure 5 – Scintillation Detector Assembly

HINGED DOOR

LEAD SHIELDING

Nal

CLAMP

GAS OULET

GRIP FOR REMOVAL
OF FILTERS IN
LEAD CONTAINER

FILTERS

"O" RING SEALS

LEYBOLD COUPLING

Nal

PLATE 35 mm DIA
x 0,3 mm THICK

GAS INLET

Figure 6 – Modular Glove Box Layout

TO IODINE TRAPPING GLOVE BOX

LINES A

LINES B

TO NO, NO₂
AND O₂
ANALYSER

LINES A
(INLET)

LINES B
(INLET)

4 TRAPS, EACH WITH 12 STAGES
(15 mm HIGH, 50mm DIA)
AND OVEN

HEPA FILTER

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Figure 7 – Iodine Trapping Glove Box Layout

Glove box 643.30 (Figure 7) is specifically designed to investigate the properties of new sorbent media. It includes an oven to heat all the iodine trap stages to the 100–200°C range within ± 3°C by means of six insertion heaters with a total rating of 4200 W. The traps are 260 mm high and 70 mm in diameter, and may be sectorized as necessary for testing. The unit is now equipped with four 12-stage traps, but can accommodate up to ten traps for sequential trapping by modifying the flow channels.

Pump Glove Box This unit is designed to circulate the gas flow and monitor the flow rate. It includes four diaphragm pumps rated at 3 $Nm^3 \cdot h^{-1}$ with a needle valve regulating the gas flow between the pump discharge and suction sides. A mass flowmeter is used to monitor the gas flow rate.

Process Trap Leakage Monitoring Glove Box This unit was initially designed to monitor iodine leakage from the process traps via line C, but was modified for R & D test purposes. The preheated gas flow from line C is supplied to glove box 643.60 (Figure 8) which includes two parallel traps that may contain either activated charcoal impregnated with potassium iodide or a special sorbent medium for specific testing. The glove box is equipped with a self-contained gas pumping and monitoring system and a chemiluminescence nitrogen oxide analyzer.

Off-Gas Analysis and Characterization Online analysis is provided for nitrogen oxide, oxygen, krypton and iodine. The laboratory includes a γ and X-ray spectrometry line with a 4-channel analyzer, and two nitrogen oxide measuring devices: a chemiluminescence system is installed in glove box 643.60, and a radiation absorption system is installed in glove box 643.50a (Figure 4). Nitrogen monoxide is measured in the infrared region, and nitrogen dioxide in the ultraviolet region.

Oxygen: The oxygen analyzer located in glove box 643.50a is based on the paramagnetic properties of the oxygen molecule.

Krypton: The krypton analyzer in glove box 643.50b is supplied by lines A2 and B2: one line bypasses the analyzers while the other is ducted through the ionization chamber. Considering the nature of the
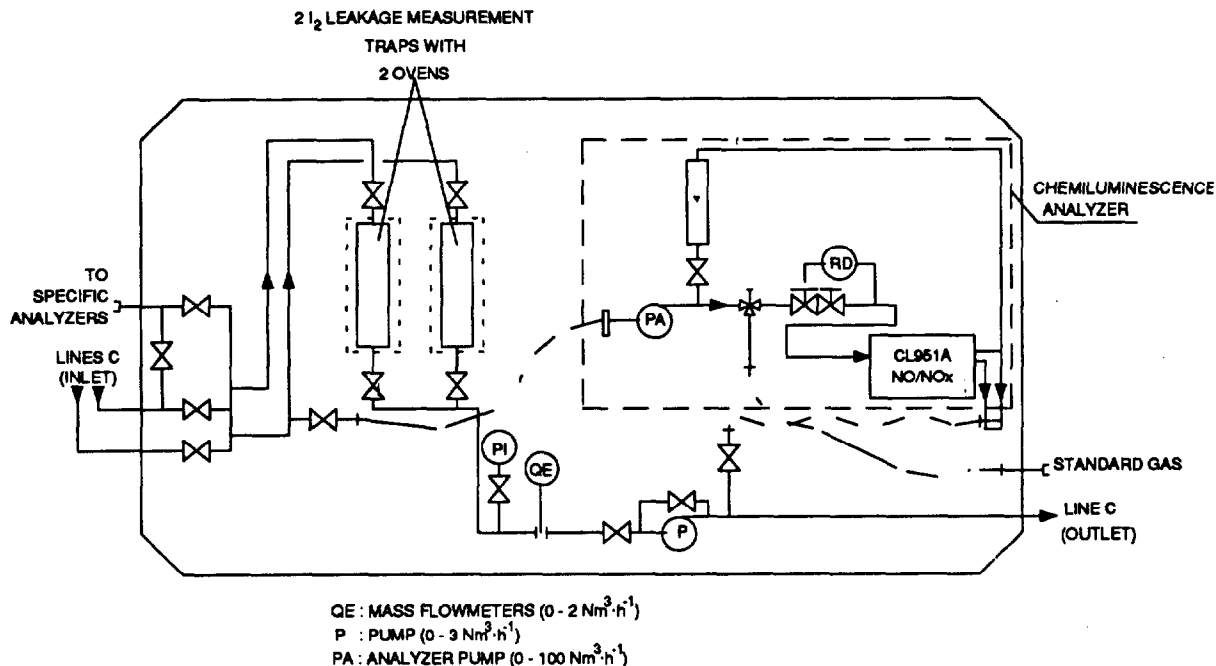
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Figure 8 – Process Iodine Leakage Monitoring Glove Box Layout

fuel reprocessed in the APM facility, the measurement range extends from 10^4 to 10^{13} Bq·m^{-3}. Two units are installed to cover the full range: two 10-liter differential ionization chambers are used to compensate for background γ radiation between 10^4 and 10^{10} Bq·m^{-3}; a single 100 cm^3 ionization chamber covers the range from 10^8 to 10^{10} Bq·m^{-3}.

Iodine: When excited by a laser beam, an iodine molecule produces a fluorescent emission. The selected optical transitions are the 526 or 532 nm lines after monochromatic excitation at 514 nm. This laser-induced fluorescence allows highly sensitive determination of the total molecular iodine concentration without isotopic discrimination. The technique was nuclearized as indicated in Figure 9. The glove box contains the process and standard gas inlets, a glass and optical grade quartz measurement cell, a pyrolysis cell on a bypass line, a process gas circulation pump and various pressure sensors and flowmeters. The analysis system basically comprises a 514 nm argon laser coupled with a single-mode optical filter, a signal collector lens and filter, a 600 mm focal length spectrometer, a photomultiplier, and signal acquisition and processing circuitry. One of the advantages of this technique is that it includes a calibration system connected in parallel with the process sampling line. The calibration system, based on the vapor pressure of the iodine crystals in a temperature-regulated heat exchanger, is used to determine the detection limit (currently 10^{-4} g·m^{-3}) under actual operating conditions.

γ *and X-ray spectrometry line*: This line is designed to determine the ^{129}I concentration trapped on the solid media, as well as any other products that may be fixed there, and to determine the vesicle activity entrained in the off-gas stream. A conventional hyperpure germanium detector is used in the counting system. A 4-channel analyzer is used in conjunction with the scintillation detectors as described above.
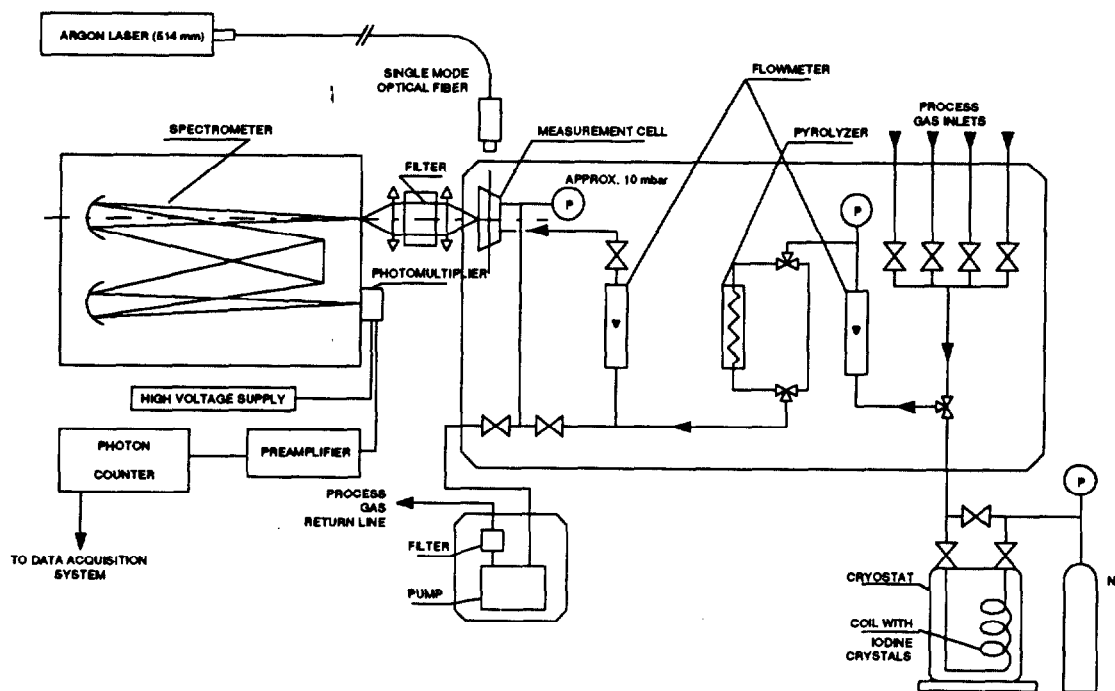
Figure 9 – Laser Fluorescence Iodine Measurement System Schematic

Conclusion

The Marcoule Pilot Facility is equipped to eliminate iodine from the dissolution off-gas stream by adsorption on solid media, and includes a laboratory to analyze and characterize the off-gases from each process step (condensation, recombination of nitrous fumes, soda scrubbing, iodine trap inlet and outlet stages). The system is consistent with the policy of reducing gaseous iodine release[2] and with the investigation of dissolution off-gas treatment.

Program objectives include the following:
• assess the efficiency of each step in the off-gas treatment process;
• develop continuous monitoring techniques for vesicular activity;
• investigate iodine metabolization;
• investigate iodine trapping behavior depending on the sampling point;
• prepare sorbents with various iodine concentrations approaching saturation to develop a suitable disposal matrix.

An LWR fuel reprocessing campaign was recently completed. Throughout this campaign the following points were investigated:
• testing of AC 6120 and AgA sorbents with dissolution off-gases after soda scrubbing and HEPA filtration (AC 6120 sorbent was used for all the process off-gas stream, with AgA sorbent in a bypass line (C) to the analysis and characterization laboratory);
• testing of four sorbents (AC 6120, AgA, JFM1 and JFM4) under identical geometry, flow velocity and temperature conditions;
• determination of the nitrogen oxide concentrations;
• determination of the krypton release during cutup and dissolution;
• qualification of the continuous laser fluorescence iodine monitoring system.

References

[1]   Fabre, J.J. and Rouches, F., International Conference on Fast Reactors and Related Fuel Cycles, *Proceedings*, vol. II, pp. 15.1.1–11 (November 1991).

[2]   Mulcey, P., 21st DOE/NRC Nuclear Air Cleaning Conference, vol. II, pp. 978–979.

DISCUSSION

MOELLER: Would you please explain the role of the glove boxes in your sampling and analytical system?

VIGNAU: The glove boxes are installed to prevent a gas leakage at the coupling tube with the traps and the analyzers. Outside the glove boxes there is no possibility of contaminating systems because the coupling tubes are welded.

TRITIUM EMISSIONS REDUCTION FACILITY (TERF)

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Abstract

Tritium handling operations at Mound include production of tritium-containing devices, evaluation of the stability of tritium devices, tritium recovery and enrichment, tritium process development, and research. In doing this work, gaseous process effluents containing 400,000 to 1,000,000 curies per year of tritium are generated.

These gases must be decontaminated before they can be discharged to the atmosphere. They contain tritium as elemental hydrogen, as tritium oxide, and as tritium-containing organic compounds at low concentrations (typically near one ppm). The rate at which these gases is generated is highly variable. Some tritium-containing gas is generated at all times.

The systems used at Mound for capturing tritium from process effluents have always been based on the "oxidize and dry" concept. They have had the ability to remove tritium, regardless of the form it was in. The current system, with a capacity of 1.0 cubic meter of gas per minute, can effectively remove tritium down to part-per-billion levels.

A new, more efficient, larger capacity (2.5 cubic meters per minute), system has recently been built. It is called the Tritium Emissions Reduction Facility (TERF), and it incorporates a considerable number of improvements over the current system. These include: 1) replacement of the easily plugged sintered metal filters with fiber bed filters, 2) installation of condensers off-line rather than on-line to reduce the refrigeration load, 3) larger, variable-speed compressors, 4) increase of the system pressure from 1.5 to 4.5 atmospheres absolute, to minimize the size of process components, 5) increase the maximum temperature of the oxidizing catalyst to $600^\circ C$, 6) higher temperature regeneration to more completely regenerate the molecular sieve desiccant, 7) use of activated carbon and ion exchange resins to purify the product water for tritium recovery, 8) use of more corrosion-resistant materials, 9) use of a process control computer, and 10) secondary containment of the system inside of gloveboxes. These improvements are discussed.

I. Introduction

Mound conducts a wide variety of multi-gram tritium operations for the U.S. Department of Energy (DOE). These include research, development, testing, weapon component evaluation, loading of microballoons, preparation of gas standards for mass spectrometers, loading tritium into weapon components, commercial sales, scrap recovery, and isotope enrichment. Several support functions, such as mass spectrometry, analytical chemistry and materials analysis, also involve significant quantities of tritium. To protect the workers and the environment, these operations are performed in nitrogen-purged gloveboxes which are connected to an effluent capture system. This capture system also decontaminates all process effluents, such as vacuum pump exhausts, raffinates from isotope enrichment operations, gaseous contaminants separated from tritium streams, and exhausts from analytical instruments.

At Mound it is very important to contain tritium and to minimize effluents. Continuous systems to capture process effluents have been utilized at Mound for 30 yr. Currently, Mound's tritium emissions to the atmosphere are less than 2500 Ci/yr; this is less than 1% of the releases in 1969. A desire to further reduce these emissions is one of the principal reasons that the Tritium Emissions Reduction Facility (TERF) was designed and built.

Background

The system in use at Mound for removal of tritium from concentrated gaseous effluents is the Effluent Removal System (ERS)[1]. It has been on-line for the past 20 yr and has a capacity of 35 scfm. Tritium is removed by the oxidation to water of tritium and tritium-containing compounds over platinum catalyst at 450°C, followed by the collection of the water on molecular sieve drying beds. The gases processed by the ERS include nitrogen, air, argon, helium, hydrogen, water vapor, and various organic compounds, such as solvent vapors and vacuum pump oil mists. The introduction of halogen-containing compounds is strongly discouraged because of the corrosive materials that are formed in the high temperature oxidizing reactors. Single-pass decontamination factors of 1,000,000 and 10,000 have been achieved for HT and tritiated organics, respectively.

Need for Improvement

There are several reasons why the improved capture system, which is called the Tritium Emissions Reduction Facility (TERF), was designed and built: 1) the ERS is aging and, hence, becoming less reliable, 2) the flow capacity of the ERS is too low, 3) the amount of tritium being released to the atmosphere needs to be reduced, and 4) there is a need to become less dependent upon the availability of skilled, experienced operators to run the tritium capture system.   Age was a concern because of the unavailability of crucial spare parts, the cumulative effects of 20 yr of corrosion, inconvenient location of instruments (it was not "user friendly") and an outmoded control system. The limited capacity (35 scfm) of ERS was not enough to support the current workload. TERF should reduce effluents because it will be more efficient and because it will be entirely secondarily contained. Also, the larger capacity of TERF will permit the processing of low concentration streams that must now be released directly to the stack.

The operation of the TERF will be less dependent on the skill of the operator.   It will be easier to monitor because the instruments are located in much more accessible locations.   The TERF equipment is also much more accessible, being built in a large area which does not contain other processes or items of equipment whose presence constricts the equipment arrangement.   Because the TERF is continually monitored by a process control computer, necessary adjustments are made automatically, reducing the work load on the operator and keeping the system components at their optimum settings a greater percentage of the time.

II. System Description

A simplified system flow diagram of the TERF is shown in Figure 1. The system has redundant components, for additional gas processing capacity and for backup, which are not shown for the sake of simplicity.

The gas collection subsystem is comprised of several branch lines which service the laboratory and production areas. Each branch line has its own gas flowmeter and ionization chamber to monitor the quantity of tritium entering the system. The branch lines will be maintained at a pressure of approximately 84.5 kilopascals (12.2 psia). This pressure will normally be maintained by a control valve and generated by a compressor that moves the gas through the TERF.

Abnormally high gas flows and temporary system outages will be handled by an evacuated surge tank with a volume of approximately 142 STP cubic meters (5,000 STD cubic feet).

A mist eliminator will be used to remove any entrained particles and mist in the incoming gas prior to entering the compressor. Pressure drop across the elements will be kept to a minimum in order to maintain rated gas processing capacity at maximum flow. Pressure drop across the inlet system is limited to a maximum of 15.5 kilopascals (2.2 psi) by the compressor inlet vacuum at maximum capacity.
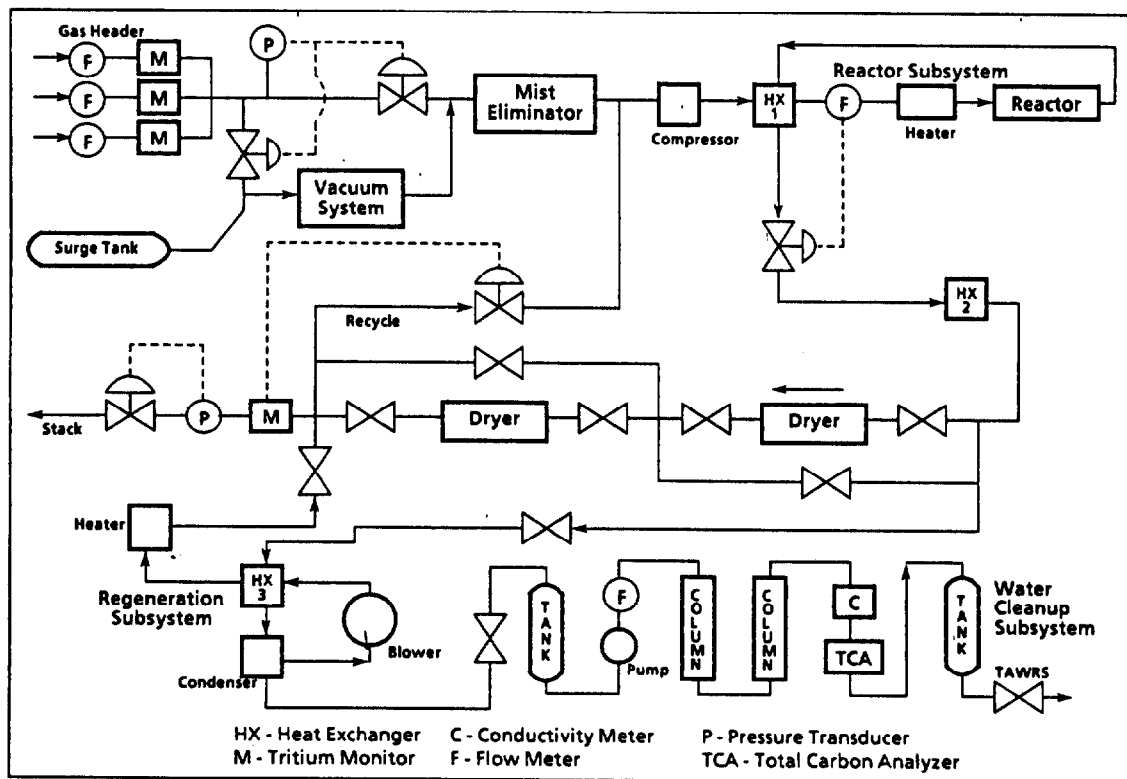

FIGURE 1

SIMPLIFIED FLOW DIAGRAM FOR TRITIUM EMISSIONS REDUCTION FACILITY

The two compressors are positive-displacement, oil-less, two-stage reciprocating compressors with an intercooler to control second stage seal temperatures. The intercooler, head, and motor are water-cooled to reduce heat release to the glovebox atmosphere. The variable speed motors are controlled to follow the gas load.

The reactor subsystem consists of a conservation heat exchanger, preheater, and reactor containing a platinum-on-alumina catalyst (Englehard A16, 648). Reaction temperatures will be maintained up to 600°C in order to oxidize any tritiated organic molecules, primarily methanes, as well as elemental hydrogen. Flowmeters and control valves will control gas flow through the reactor systems. The conservation heat exchanger will recover approximately 80% of the heat in the gas exiting the reactor system.

Water-to-gas heat exchangers will be required after the reactor system to cool the process gas from 230°C leaving the conservation heat exchangers to 27°C before entering the dryers. The heat exchangers are constructed with a double wall to prevent contamination of the cooling water in case of a tritium leak in the inner wall.

Each dryer tower contains 72.6 kg (160 lb) of type 4A Molecular Sieve (trade name of the Union Carbide Corporation). One dryer bed will hold approximately 14.5 liters (3.8 gal) of water when saturated. Filters are located downstream of the dryers to contain the spread of Molecular Sieve dust.

Gas exiting the dryer system will be monitored for tritium content prior to release to the atmosphere, and will be recirculated if the tritium content is above allowable discharge limits.

The regeneration system is designed to remove water from the dryer towers by recirculating hot, dry gas through them. The saturated tower will be taken off-line, and dry nitrogen will be heated to 315°C and circulated through the bed at 1.4 cubic meters per minute (50 scfm) for 16 hours. The direction of flow of the regeneration gas will be opposite to that of the normal process flow. The exiting gas will be heat-exchanged with the incoming stream, then cooled to 4°C and the condensate collected. The next heat exchanger is also refrigerated and will freeze out additional moisture. The final element is a Molecular Sieve bed which will reduce the moisture content of the gas to the low parts-per-million range. Then the gas stream enters the blower for another cycle.

Water collected in the regeneration system will be sent to the water cleanup system. When a batch has been collected, it will be purified by processing through filters, activated carbon beds for organic impurity removal, and mixed ion exchange resin beds for ionic impurity removal. Water that is certified pure by means of analysis with a conductivity meter and total carbon analyzer will be transferred to the Tritium Aqueous Recovery System (TARS) for the first step in the tritium recovery and enrichment process.

III. Use in Maintenance Operations

One of the ways the increased capacity of the TERF will be used is in supporting operations which generate large volumes of gas, such as maintenance operations which cannot be performed through the glovebox gloves. Some maintenance operations require the opening of gloveboxes, tritium-contaminated lines, or contaminated equipment, which exposes previously confined, tritium-contaminated volumes to the surrounding environment. During these operations, the release of tritium to the atmosphere is inevitable without some type of containment and decontamination of effluents.

At Mound, the item to be opened is enclosed in a flexible plastic sheet supported on a metal frame. The enclosure is exhausted to the capture system (ERS or TERF) through the open glovebox or through plastic tubing connected to one of the inlet lines to the capture system. Sufficient air flow is maintained through the enclosure to prevent tritium migration to the room air. An enclosure large enough to allow operating and maintenance personnel to work inside can be used. While inside the enclosure, a worker wears a plastic bubblesuit with supplied breathing air so that all physical contact with tritium is avoided.

Several methods are used to decontaminate or reduce the quantities of tritium in lines and equipment if they must be opened for maintenance. If liquid could be present, the item is fitted with a specially designed piercing valve. This valve makes a small hole, and the liquid drains into a container without any release of tritium. If welding is involved in the maintenance operation, external heat is applied to the area while a small flow (0.05 m^3/min) of air is flushed through the line. This vaporizes moisture and liberates sorbed tritium in the vicinity of the repair and carries it to the capture system.

During the maintenance operation, air flow from the enclosure to the ERS is limited to only that necessary for tritium containment. At times, this type of operation requires a major fraction of the capacity of the ERS for several hours. This is one example where the increased capacity of the TERF is needed. Not only is the enclosure ventilated but, whenever possible, open contaminated lines are directly purged to the capture system. The intent is to move air from the room, through the enclosure, into the open glovebox or process lines, and then into the capture system.

These simple but very effective methods have helped considerably in reducing tritium releases to the atmosphere and have contributed to reducing personnel exposures to tritium by containing and capturing tritium that would otherwise be released from maintenance operations.

IV. Design Features

Control System

The TERF has been designed for automatic operation and process control by means of a distributed process controller with computer supervisory control and system monitoring. This significantly reduces the operator's experience and capabilities as major factors in system reliability, availability, and operation.

TERF has been designed as a separate Material Balance Account with computer integration of incoming tritium in gas streams, the capability of accurately determining system holdup, and accounting of tritium quantities transferred out of the system.

Dryer Regeneration

The molecular sieve beds will be regenerated when moisture monitors located after the dryers indicate that breakthrough has occurred. Once bed saturation has been detected, the loaded tower will be taken off-line to be regenerated. Studies have shown that the molecular sieve holds water tenaciously, and a considerable effort is needed to thoroughly remove it. Therefore, the process gas in the saturated dryer will be circulated at a rate of 1.4 m^3/min (50 scfm) and heated to 315°C to regenerate the molecular sieve. It will flow in the opposite direction to the process gas. We expect that it will take 16 hr to complete the regeneration cycle.

Water Purification

Mound has the capability of recovering tritium from water by the sequential use of three processes: 1) the Tritiated Aqueous Recovery System (TARS), which enriches tritiated water and uses electrolysis to produce hydrogen, 2) the Tritium Recovery Column (TRC), which uses cryogenic distillation for the bulk stripping of protium, and 3) the Hydrogen Isotope Separation System (HISS), which is a three-column cryogenic distillation system for tritium enrichment. The TERF product water stream, which is transferred to TARS, must contain less than 10 parts per million of organics and have an ionic purity such that the resistivity is greater than 1 megohm cm. This is accomplished by the use of activated carbon and ion exchange columns.

Double Containment

The Mound philosophy of containment requires that the entire TERF system be doubly contained, with the exception of process lines on the vacuum side of the system between components, equipment, and instrumentation. As a result, all instruments, valves, and equipment are enclosed in gloveboxes or secondary enclosures, and all pressurized process lines and all liquid transfer lines are doubly contained. Careful consideration has been given to the design of the gloveboxes and to the location of equipment in the gloveboxes so that the maximum amount of maintenance can be performed through the gloves.

V. Differences Between the TERF and the ERS

With a design capacity of 80 scfm, the TERF will handle more than twice the volume of gases handled by the ERS. The two TERF compressors have capacities of 60 and 100 scfm, considerably more than those of the ERS compressors. One way in which this additional capacity is processed without a commensurate increase in the size of the equipment is to use higher pressures (50-100 psig in the TERF vs. 10 psig in the ERS). The higher pressures reduce the actual gas volume significantly, thereby allowing a reduction in the size of the piping needed to transport the larger gas flows and a reduction in the size of the valves to control these flows. The higher pressures also provide a longer residence time of the gases over the catalyst inside reactors of the same size.

The TERF will utilize reactors constructed of Inconel 625 and operated at temperatures as high as 600°C to assure the total destruction of all tritiated compounds, including tritiated methanes. A more complete description of these new reactors is given in a paper by Wieneke.[2]

The TERF will use countercurrent heat exchange equipment to recover process heat. The recovery of 80% of the heat put into the gases by the preheater and reactor is expected. Also, approximately half of the heat created in the gases by the compressors will be conserved by using insulated lines to keep these gases hot while they are being transported to the reactor systems.

In the TERF, all of the water from the process gas streams will be removed in molecular sieve dryers following the oxidation reactors. The collection of much of the water in the on-line condenser in the ERS required that all of the ERS process gases be cooled to -50°C and then reheated – a considerable thermal inefficiency. The ERS also cooled the gases to this low temperature before the gases were passed through the reactors. This results in freezing out some additional materials – such as vacuum pump oil vapors and other fine mists – which cause the water melted out of this condenser to be contaminated with these organic materials. In the TERF all of the organics contained in the entering gas stream will be oxidized in the reactors, leading to a considerably reduced load on the water clean-up system.

The TERF will use larger dryers than are installed in the ERS. The amount of water adsorbed in the dryers will be continuously calculated to determine how much unused capacity remains in a TERF dryer. Also, each dryer is composed of two towers, and the moisture content of the process gas will be monitored between the towers. This will allow us to regenerate these dryers as needed (which will be approximately weekly), instead of regenerating every day as in the case of the ERS dryers.

When the molecular sieves in the TERF dryers are regenerated, this will be done at a higher temperature and will use a recirculating gas volume for a longer time in order to more completely remove the absorbed water. Lowering the amount of water left in the molecular sieves will lead to lower concentrations of water in equilibrium with them at the dryer exit, and this should result in lower tritium oxide emissions. The regeneration gas flow will be in the opposite direction to that of the normal process flow, so that the absorbed water will be "backed out" of the dryer instead of having to be pushed through the entire column of molecular sieve. The higher temperatures for drying the molecular sieves will be achieved by a combination of external electric heaters and preheated hot gas continuously recirculated through the molecular sieves.

The TERF is designed to operate unattended for long periods, such as overnight and over weekends. Control during these periods is maintained by the use of a distributed process controller from the Foxboro Co. This system, referred to as Intelligent Automation (I/A), continuously monitors the key process variables that include temperature, pressure, flow, and tritium and moisture concentrations throughout the process. The system has been made fault-tolerant by the use of redundant control processor modules so that failure of one computer module will not cause system shutdown. This system

is configured to make decisions during normal operations and also in the case of deviation from normal operation. The logic and decision trees for control of the process are programmed into the Foxboro process control system. It automatically switches to a back-up in case of component failure. If the whole system should fail, it will shut down and preserve the process in a fail-safe condition. The additional control instrumentation in the TERF will increase system reliability. This system is more fully described in a paper by Kissner.[3]

Key components in the TERF which will be exposed to corrosive conditions are constructed of materials which are more corrosion-resistant that those used in many parts of the ERS. The TERF reactors and the product water tanks are constructed of Inconel 625, as opposed to Type 304H stainless steel for the ERS reactors. Most of the other parts of the TERF are constructed of type 316L stainless steel.

The TERF is more strongly protected against environmental disasters than is the ERS. The TERF is installed in a heavily reinforced, underground building; hence, it is much more resistant to damage from tornados and earthquakes. All of the equipment items are designed to withstand an earthquake with 0.15 g acceleration.

All of the TERF process equipment which operates above one atmosphere pressure is doubly contained. The equipment items are enclosed inside of stainless steel gloveboxes equipped with safety plate glass windows and rubber gloves. Pipes outside of the gloveboxes are welded within a larger pipe, and the annulus is maintained at vacuum. In the event of a leak, the tritium-containing gases are confined. This is not true of the ERS. The TERF gloveboxes are purged with dry nitrogen, in keeping with the standard practice at Mound, and exhausted to the inlet header of TERF.

Finally, the TERF has been designed so that its capacity can be expanded. Space has been left which could accommodate an additional large compressor, another reactor, and two new dryer trains. This equipment would provide redundancy and could increase the capacity of the present TERF unit by more than 50%.

VI. Conclusion

The TERF has been designed on the same overall "oxidize and dry" principle as the very successful ERS, and it uses many of the same types of components. However, several significant engineering modifications in the design of the TERF will increase its capacity, efficiency, reliability, and ability to contain tritium, while reducing emissions below those which are achievable by the ERS.

The new TERF will be a much "smarter" system than its predecessors. It will be capable of automatically handling problems normally encountered in system operation; e.g., equipment malfunctions, instrument failures, and variations in the gas load. It will also note trends in operation and recommend routine maintenance.

Operating experience with previous systems has been incorporated to improve maintenance and reliability of equipment. Lower emissions limits and concern for personnel safety have placed increased emphasis on maintenance operations. Double containment and ease of replacement and changeout of equipment have been incorporated in the design to reduce the possibility of release and/or personnel exposure.

The results of experimental work (catalyst comparisons, higher operation pressures, increased instrument accuracy, and better molecular sieve regeneration efficiency) have been incorporated into the new system to increase processing efficiency. This experimental work, and lessons learned from operating the existing Effluent Removal System, have contributed to an increased understanding of system function.

Mound's goal was to incorporate the latest technical advances, state-of-the-art control technology, and twenty years of operating experience with a tritiated gas capture system to make the new Tritium Emissions Reduction Facility as safe and efficient as possible without sacrificing reliability. This system will be Mound's primary capture system for tritiated gaseous process effluents into the twenty-first century.

References

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DISCUSSION

JUBIN: What is the magnitude of cost savings expected by the implementation of automation? Was this a factor in the selection of automation?

LAMBERGER: Cost saving was not an important driving function in deciding to automate. No significant cost savings are expected.

FURRER: I have three questions; first, I would like to know the space velocity at the catalyst? Secondly, the reaction rate of the catalyst for methane at 400°C? and thirdly, what was the impregnation?

LAMBERGER: In answer to your first question, the residence times is 2 sec. The space velocity is 0.1 sec^{-1} at 400°C and 50 psig and 0.45 sec^{-1} at 600°C and 100 psig. Secondly, the reaction rate constant for methane over this catalyst is K = 6.0 sec^{-1} at 400°C and 15 psig. It is K = 14 sec^{-1} at 500°C and 15 psig and third, platinum on alumina. We used Englehard catalyst A16648.

DEMETRIA: 1) How large is the facility housing the 35 cfm-capacity "TERF"? 2) What kind of ventilation is provided to this room?

LAMBERGER: 1) Approximately half of this room x 3 times the height (30' x 60' x 30' high). 2) Once through system through fume hoods and discharge to the outside without any treatment. Face velocity is 150 ft/min.

DUVALL: Out of 2,000 CiT released to the environment, what fraction is HTO?

LAMBERGER: 50%.

MINIMIZING OF IODINE-129 RELEASE AT THE
KARLSRUHE REPROCESSING PLANT WAK

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Abstract

From 1971 until 1990 the Karlsruhe Reprocessing Plant WAK reprocessed 207 tons of uranium from oxide fuel. During the fuel dissolution 94 % of the iodine-129 inventory were expelled together with the dissolver offgas (DOG) and fixed on iodine filters. About 5 % reached together with the fuel solution the PUREX process and spread there among the various gases and liquid streams. About 1 % remained in the dissolver residues. With regard to minimize iodine-129 release, iodine retention devices were tested in all vessel offgas (VOG) lines of the process and the vessel offgas of the waste treatment facilitiy. If only the dissolver offgas is passed through iodine filters, 2,5 % of the iodine input is emitted with the total offgas.

If the dissolver offgas and the offgas from the high active waste storage tanks is cleaned, 1,8 % of the iodine input is emitted. This was the actual state at WAK in 1988 until 1990.

It could be demonstrated that by iodine retention in all relevant offgas lines, besides offgas of the head end cell, only 0.3 % of the iodine input with the fuel is emitted.

Testing of iodine sorption materials has shown that the removal efficiency of the $AgNO_3$-impregnated materials depends on the iodine concentration in the WAK offgas. The causes of the observed dependence of the decontamination factor (DF) on the iodine-129 concentration may be not only organic iodine compounds difficult to remove but also the high dissociation pressures in the $AgNO_3/AgI$ system. Impregnation with elemental silver improves iodine removal considerably.

I. Introduction

After dissolution of the fuel, fission product iodine is preferably present as volatile elemental iodine. According to its distribution coefficient, it is distributed in the fuel solution, in the scrubber solutions of the dissolver offgas section and in the offgas.

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The distribution of iodine among various process streams should be minimized because such a distribution, considering present restrictive requirements, would imply that up to the stage of waste conditioning iodine retention systems in the vessel offgas lines would have to be provided.

Therefore, it has been the goal pursued in most of recent studies to transfer iodine-129, if possible quantitatively, from the dissolver solution into the gas phase where it is fixed on appropriate sorption materials to make it suitable for repository storage.

For the Gorleben site, a site selection upheld until 1979, the limit value of iodine emission applied for and recommended was 7.4×10^9 Bq/a (200 mCi/a); with a throughput of 1400 metric tons fuel per year and a burnup of 35 GWd/t (57 Ci/a iodine inventory) this corresponds to an emission of 0.35 % of the iodine-129 input with the fuel.
In order to be sure that this limit is not exceeded, the German licensing authorities in their expert opinion dated 20th October, 1977 underlined that even with 99 % iodine expelling from the dissolver, iodine filters would have to be provided in the vessel offgas lines.

The scene completely changed with the selection of the new site at Wackersdorf in the mid-eighties and with values of iodine-129 release of as low as 50 mCi/a (1.85×10^9 Bq) imposed by the licensing authorities for planning that plant.

DWK (Deutsche Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen), which was to assume the responsibility for the German industrial scale reprocessing plant, urgently requested from WAK validated results on the iodine release from a reprocessing plant.

- The first goal had been to show how much iodine can be expelled from the dissolver.

- Besides, an iodine balance was to be elaborated for the plant.

- Moreover, it was necessary to test iodine filters in the vessel offgas.

This had been the reason of an extensive iodine program in the Karlsruhe reprocessing plant which had been performed during the period from 1986 until 1990. The investigations have been continued up to now in the order of KEWA to gain further knowledges which may be applicated usefully in other reprocessing plants.

II. Iodine Distribution in WAK

Figure 1 shows the process units and offgas streams which are relevant to iodine distribution in WAK.

Iodine which is not expelled into the dissolver offgas during dissolution remains in the feed from which it is carried togehter with the liquid streams into the extraction process and into the medium

active waste solutions. Some of it is subsequently detected in the vessel offgases of the PUREX process and in the waste stores for medium and high active wastes (MAW and HAWC).

After four to five years duration of the iodine program quantitive statements can be made today on the iodine distribution in WAK. They will be discussed in more detail in this paper.

It should be mentioned here that iodine-129 which had been released with the offgas during "none production" periods and after definitive plant shut down end 1990, has to be taken into account to establish the iodine balance. For this reason, slight deviations are noticed of the iodine-129 fractions in the offgas relative to the values communicated at the previous Air Cleaning Conference [2].

II.1 Iodine in the Dissolver Solution (Feed)

After dissolution of fuels from KKS* and MZFR**, the residual iodine was determined in the dissolver solution in order to obtain more accurate values about the iodine input into the WAK extraction process.

A method developed at KfK/IHCh*** allows to analyse iodine which is difficult to detect by means of pressureless tempering at approx. 100 °C, subsequent extraction and gamma spectrometric determination [3] or mass spectrometric one [5] of the residual iodine content.

It appears from the determinations of residual iodine that even after 8 to 9 hours of dissolution and distillation (about 5 %/h) of the feed up to 5 % of the iodine input is left in the feed and may reach the PUREX process.

Additional iodine expelling by addition of iodine-127 (natural iodine) and NO_2 sparging of the fuel solution reduces the residual iodine content in the feed only by the factor of 2 to 3.

Therefore the vessel offgases have to be purified by iodine filters in order to achieve a nearly complete iodine retention in the plant.

II.2 Iodine in Liquid Process Streams of the PUREX Process

Quite a number of investigations performed with the solvent in the first extraction cycle from June 1986 until December 1988 yielded iodine contents between 16,000 and 26,000 Bq/l.

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*    KKS      = Kernkraftwerk Stade (Stade Nuclear Power Plant)
**   MZFR     = Mehrzweckforschungsreaktor (Multipurpose Research
                Reactor Karlsruhe)
***  KfK/IHCh = Kernforschungszentrum Karlsruhe/Institut für Heiße
                Chemie (Hot Chemistry Institute of the Karlsruhe
                Nucleare Research Center)
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In early June 1989 the organic solvent of the first extraction cycle was replaced. This allowed to observe the increase in iodine-129 concentration in the organic solvent as a function of the iodine input by the fuel solution during the third KKS and the ninth MZFR campaigns.

From June 1989 until March 1990 it was possible for the first time to follow the buildup of iodine concentration in the organic solvent versus time (Fig. 2).

After feeding of 5 KKS batches, the iodine concentration increased to 4800 Bq/l. This increase in iodine concentration from 1900 to 4800 Bq/l, with a solvent inventory of about 3.5 m^3, implies that 1% of the iodine inventory of five batches has been transferred from the fuel solution into the solvent.

The iodine concentration in the organic solvent increased to as much as 22,000 Bq/l after feeding another 25 batches of fuel solution from MZFR and KKS into the process.

This means that after replacement of the solvent 70 MBq iodine-129 had been input into the "fresh" solvent. This is equivalent to 2.5 % of the iodine contained in the dissolved fuel elements.

Scrubbing with sodium carbonate of the organic solvent lowers the iodine content only slightly and is not taken into account here.

Table 1 gives a survey of the iodine concentrations in some characteristic WAK process streams.

III. Iodine in Offgas Streams

Long-term measurements performed in all offgas streams from WAK show the distribution of iodine among the individual offgas lines.

The following offgas and vent air streams were recorded (Fig. 3):

- dissolver offgas   (since 1975)

- vent air from dissolver cell   (since 1988)

- vessel offgas of the PUREX process   (since 1988)

- vessel offgas of medium active waste storage
  tanks (MAW)   (since 1988)

- vessel offgas of high active waste storage
  tanks (HAWC)   (since 1988)

- vessel offgas from waste treatment   (since 1988)

III.1 Dissolver Offgas (DOG)

To control iodine loading and to determine the decontamination factor DF* of the two filters in series in the dissolver offgas the iodine concentration in the dissolver offgas is monitored at the sampling points S1, S2 and S3 using iodine sampling filters (Fig.3).

The annular cylindric iodine filters (inside dia. = 106 mm, outside dia. = 400 mm, height = 570 mm) were operated at 100 to 130 °C.

In 1989 and in 1990 approx. 95 % of iodine input with the fuel was expelled from the dissolver offgas and fixed on the iodine filters with a DF > 1000. In this way, about 0,05 % of the iodine-129 input with the fuel was released with the dissolver offgas.

III.2 Vent Air from the Dissolver Cell

The vent air flow rate of the fuel element dissolving and dismantling cell is 7300 m_3/h.

The measurements performed during several months in 1989 make evident rather strong variations. The iodine-129 concentration in the vent air is between 0.02 and 0.2 Bq/m^3.

According to the measurements related to 1989, 10.7 MBq of iodine were released from the dissolver cell. This corresponds to 0.15 % of iodine-129 input with the fuel.

III.3 Vessel Offgas of the PUREX Process

In the course of investigations performed under the iodine measuring program it was repeatedly found that the iodine discharged with the vessel offgas of the PUREX process undergoes considerable variations which are associated with the process (Fig. 4).

The maximum iodine-129 concentrations were measured when reprocessing was under way. The values amounted up to 60 Bq/m^3, the average being 30 Bq/m^3.

The lowest iodine-129 concentrations of 1 to 5 Bq/m^3 occured during the intervention and outage periods.

The contribution by the vessel offgas, related to the iodine inventory input, is about 1 %.

Testing of an iodine filter in a 10 % partial stream of the vessel offgas (35 m^3/h) of WAK had been carried out successfully since October 1987[1] (Fig. 3).

Iodine measurements in the raw and clean gas for determination of the DF were made at the sampling points S6 and S7 located upstream and downstream of the 10 % iodine filter (Fig. 3).

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* DF = quotient obtained by dividing the iodine concentration entering the filter by iodine concentration leaving the filter.

Testing extended over 4.5 years. The sorption materials used were AC 6120/12 % Ag and AgA/12 % Ag (Al_2O_3 carrier material). The DF measured with AC 6120 was $\geq$ 20. With the new material AgA/12 % Ag a DF of up to 400 has been observed. The Ag consumption in AC 6120 was less than 0.2 %.

III.4 Vessel Offgas of the Medium Active Liquid Waste Tanks

During recent years 10 to 15 m^3 of organic medium active waste containing 10,000 to 20,000 Bq iodine-129 per liter and 15 up to 75 m^3 aqueous medium active waste with 160 Bq iodine-129 per liter have been permanently stored in the tanks. Due to its elevated iodine content, the organic MAW should primarily account for the iodine discharge together with the MAW offgas.

Following termination of processing operation in February 1991 about 12 m^3 of organic MAW and 28 m^3 of aqueous MAW were still stored in the tanks.

Iodine emitted together with the MAW offgas amounted to roughly 0.4 % of iodine input with the fuel.

III.5 Vessel Offgas of the High Active Liquid Waste Tanks

At present, approx. 70 m^3 high active waste concentrate (HAWC) are stored in the tanks of the HAWC store of WAK. The iodine-129 and iodine-131 are regularly measured in the offgas of these tanks at the iodine sampling points S8 und K2 (Fig. 3).

Since the measurement in the HAWC offgas started in mid-1988 an iodine-129 release of about 140 MBq from the HAWC tanks to the iodine filter was measured until mid-1992. (The iodine-129 content of HAWC amounts to approx. 1000 Bq/l which corresponds to a total inventory of 70 MBq in the liquid).

The monthly discharge with the offgas system attained about 3 MBq iodine-129, but 1.5 years after shutdown of WAK as much as 1 to 3 MBq iodine-129 are still released from the tanks and fixed on the iodine filter every month.

The iodine filters were replaced about every two years.

It is conspicuous that the DFs measured for the filter in the HAWC vessel offgas are relatively high; for instance ,in 1989 they attained values between 46 and 460. This high DF can be seen in the connection with "easier removable" iodine in the high active solutions.

III.6 Vessel Offgas of the Waste Treatment Facility

In the vessel offgas of the waste treatment facility and during the waste treatment operation of medium active and low active wastes (MAW and LAW) up to 0.3 % of iodine-129 related to iodine inventory in the fuel must be expected. Also here an iodine filter installed in the offgas line of the MAW evaporation unit has minimized the amounts of iodine released[4].

IV.   Iodine Discharges with the Vent Air and Waste

Table   2 gives a survey of the amounts of iodine-129 discharged with the vent air and waste from WAK during the years 1986 until 1990.

WAK   produced every year more than 250 m^3 of aqueous MAW, approx. 10 m^3 of organic MAW and 1200 to 1500 m^3 of aqueous LAW. These volumes were   processed at the waste treatment facility of the Karlsruhe Nuclear   Research Center. The aqueous MAW contains   about 0.5 % of iodine-129 inventory in the fuel, the organic MAW contains about 1.7 % of   iodine-129 and reaches values up to 3 % of the iodine inventory. The   aqueous LAW contains about   0.3 % of iodine-129   input with the fuel   into WAK. This means   that on   a   yearly average   2.5 % of the iodine   inventory introduced   with the   fuel   into   WAK reach   the waste processing system.

V. Testing Iodine Filter Materials in the Dissolver offgas and Vessel offgas of WAK

Minimizing   iodine release in a reprocessing plant is closely linked to the effectivity of the iodine filters.

In   an attempt   to   optimize the iodine   filters, alternative iodine sorption materials placed in the bypass streams of the dissolver and vessel   offgases of WAK and in a   prototype filter element have been examined.

These are the results from the investigations performed at WAK:

- The DF of the iodine sorption material generally depends on the B.E.T. surface of the carrier material.

- For all sorption materials an improvement of the DF with rising temperature was found.

- All materials exhibited higher DF with increasing silver content.

- Particularly in the vessel offgas of WAK all materials with Ag contents $\geq$ 20 % were characterized by signs of exsudation which is attributable to Ag   or $AgNO_3$ growing out   of the   pores of   the carrier material.

-   Testing   of   the   iodine   sorption materials   has shown   that the removal   efficiency of the $AgNO_3$-impregnated   materials depends on the iodine concentration in the WAK offgas (Fig. 5). The causes of the   observed dependence of the DF on the iodine-129 concentration might be organic iodine compounds difficult to remove.

- It has appeared that impregnation with elemental silver improves iodine   removal compared with $AgNO_3$-impregnation, with the carrier material   remaining the same (Fig. 6). This   finding has been confirmed by thermodynamic computations[2].

The investigations of the sorption materials in the dissolver and vessel offgases of WAK have shown that the sorption materials with sintered glass (B.E.T. surface 0.15 m^2/g) and α-Al$_2$O$_3$ (B.E.T. surface 7 m^2/g) as carrier materials are not suited because of their insufficient iodine retention capability.

VI. Minimizing Iodine-129 Release

Starting from experience accumulated two means are available, besides the selection of a suited iodine sorption material, in order to minimize the iodine-129 releases with the liquid waste and with the offgas.

VI.1 Minimizing Iodine-129 Release with the Liquid Waste

As already mentioned, 1.7 to 3 % of the iodine-129 input with the fuel reaches the solvent at the extraction stage. By optimization of the process the discharged amounts of spent solvent can be minimized such that the following values are obtained:
80 to 90 % of the solvent can be returned into the plant after purification by distillation of the organic MAW. During treatment by distillation about 70 % of iodine-129 contained in the organic MAW reaches the offgas. It can be fixed on iodine filters.

VI.2 Minimizing Iodine-129 Release with the Vent Air and Offgas

As already stated, the iodine-129 release can be markedly reduced by iodine filters which have been installed in the dissolver offgas and in the vessel offgas lines of the PUREX process since 1975.

If only the dissolver offgas is passed through iodine filters, 2.5 % of iodine input with the fuel can be expected to be set free by iodine release with the vent air and offgas under the conditions prevailing in WAK process control.
When filters are installed in the relevant offgas sections, the iodine release can be minimized as follows:

- vent air from cells not provided with an iodine filter 0.15 %
- dissolver offgas passing through iodine filter   0.05 %

- vessel offgas of the PUREX process with iodine
  filter installed   0.05 %
- vessel offgas of the HAWC tanks with iodine
  filter installed   0.03 %
- vessel offgas of the MAW tanks with iodine
  filter installed   0.02 %
- vessel offgas of MAW/LAW-treatment facility   0.03 %

Making conservative assumptions one can state that in iodine filtration of the major offgas lines 0.3 % of the iodine-129 input with the fuel is released. Vitrification is not taken into consideration in this statement.

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Process stream	Iodine-129 contents [Bq/l]
dissolver solution	up to 20 000
organic solvent (1st extraction cycle)	up to 26 000
aqueous washing solution of the organic solvent (1st extraction cycle)	600 to 6 000
high level liquid waste concentrated	≈ 1 000
organic medium active waste	≈ 20 000
aqueous medium active waste	≈ 150
low active waste	≈ 30

Table 1 : Iodine-129 contents in liquid streams of the PUREX-
-Process in the WAK plant.

Path of discharges	discharges of I-129 in % of fuel inventory
gaseous and liquid discharges	
gaseous emission via stack	1,8
liquid discharges with medium and low active waste	2,2
solid discharges	
iodine filter material (dissolver off-gas line)	94 - 95
iodine filter material (HAWc off-gas line)	0,6
dissolver residues	0,8 - 1,0

Table 2 : Average discharges of Iodine-129 from the WAK plant
during the period from 1986 until 1991.
Fuel inventory : 41 GBq I-129

Iodine Balance in a Reprocessing Plant
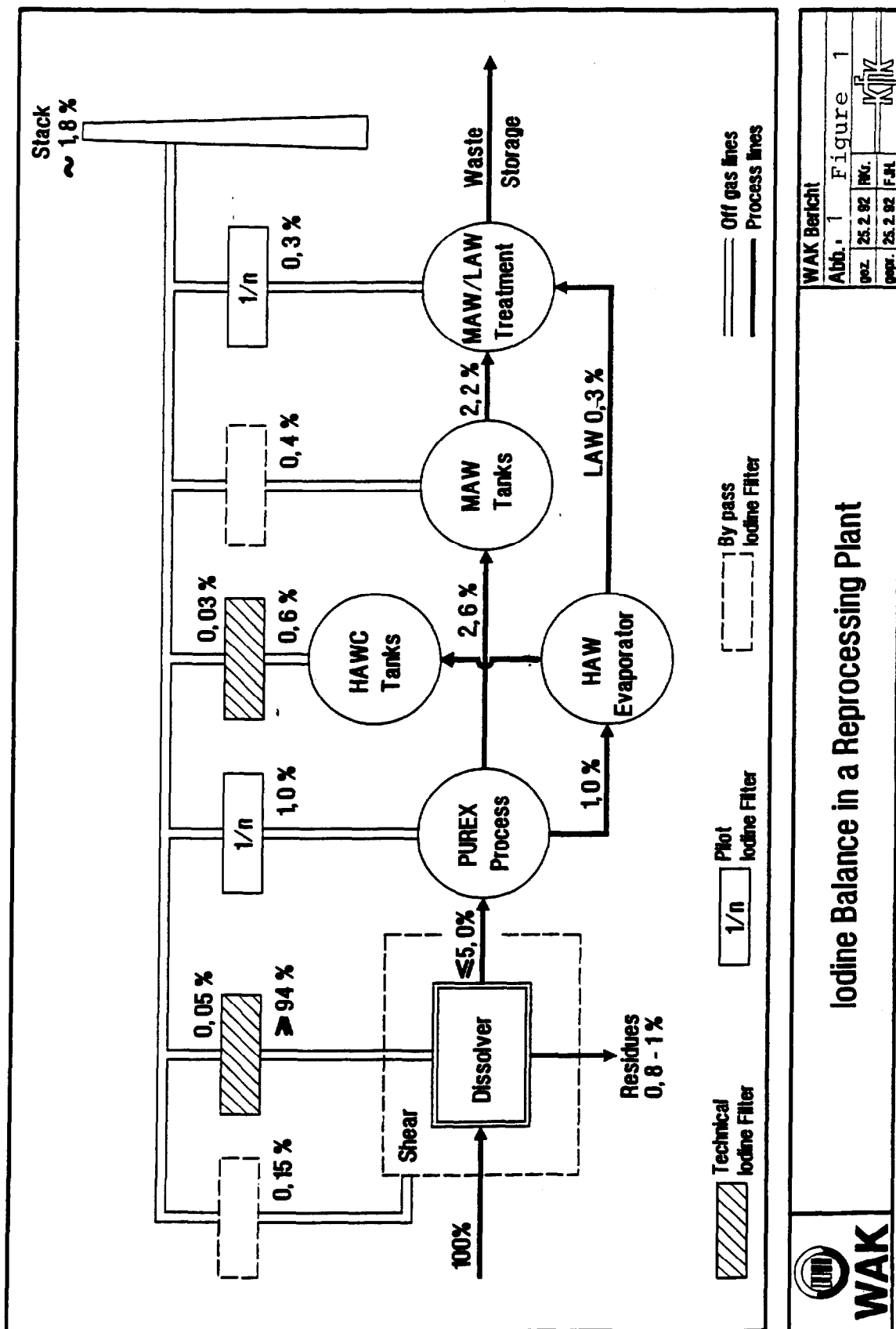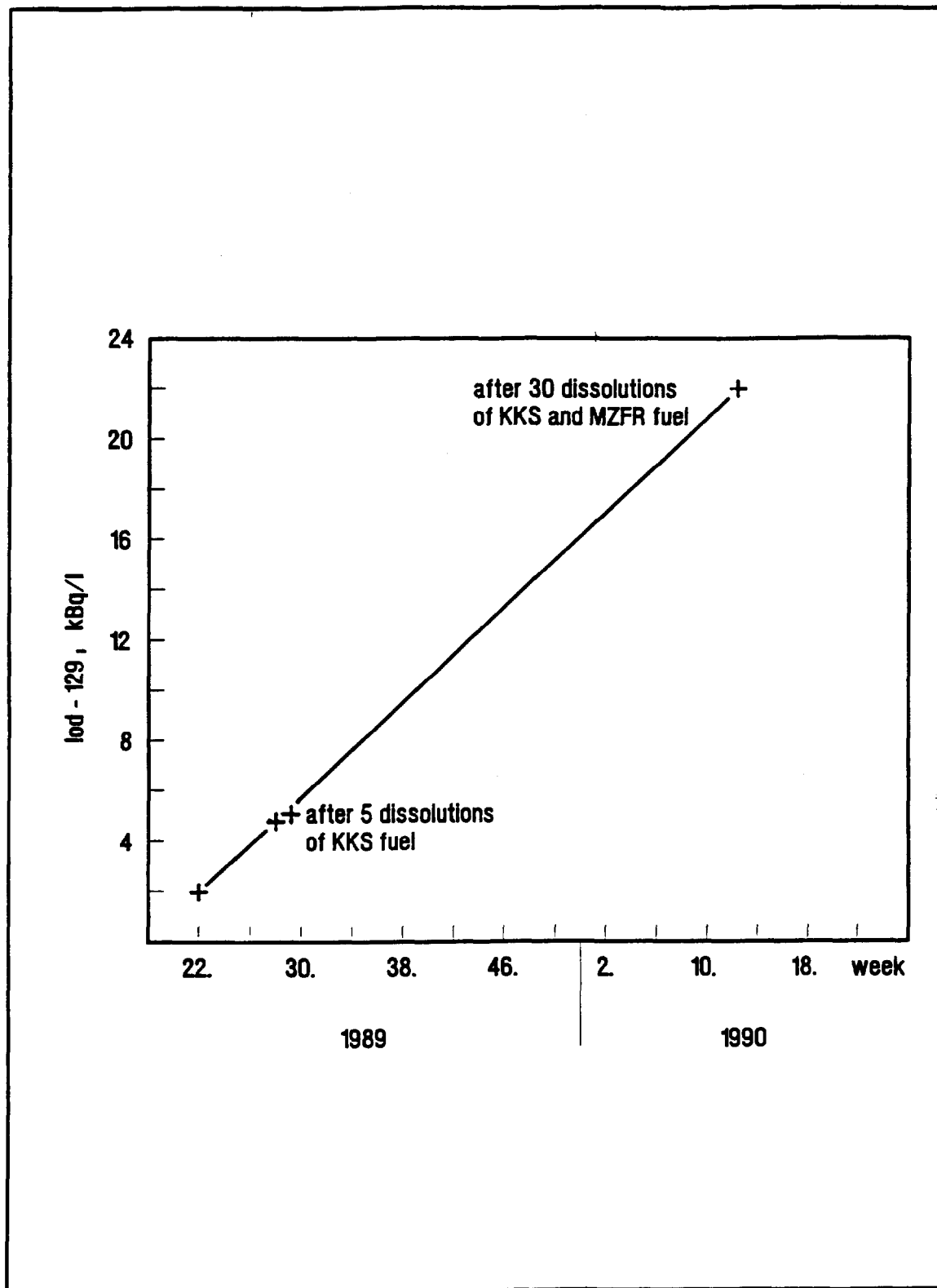
	Iodine-129 Concentration Built up in the Organic Solvent	WAK Bericht		
		Abb. 2		Figure 2
		gez. 1.6.92	RKr.	
		gepr. 1.6.92	F.JH.	
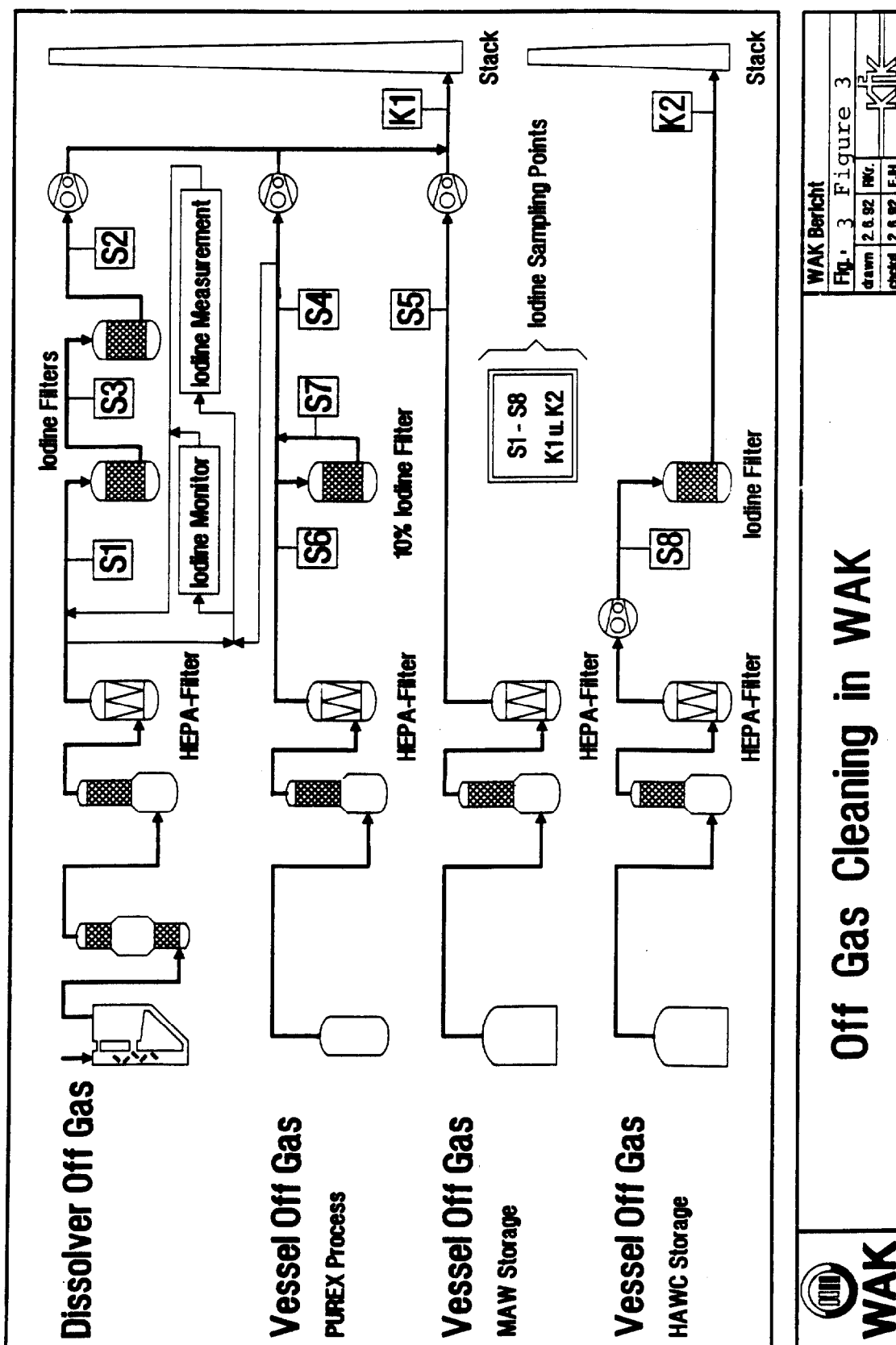
Off Gas Cleaning in WAK

Figure 4

Monthly Iodine-129 Release by VOG of the WAK-Plant
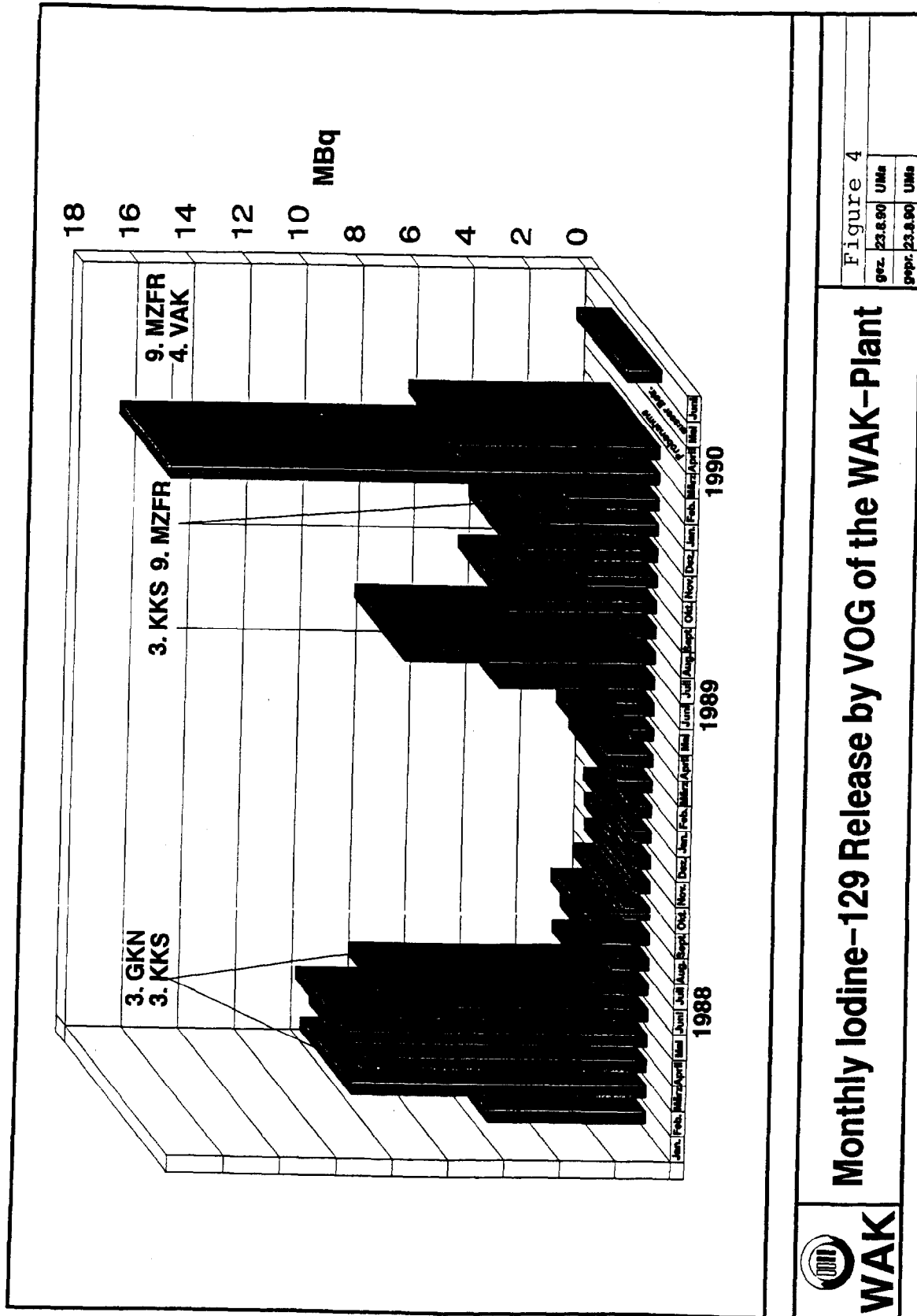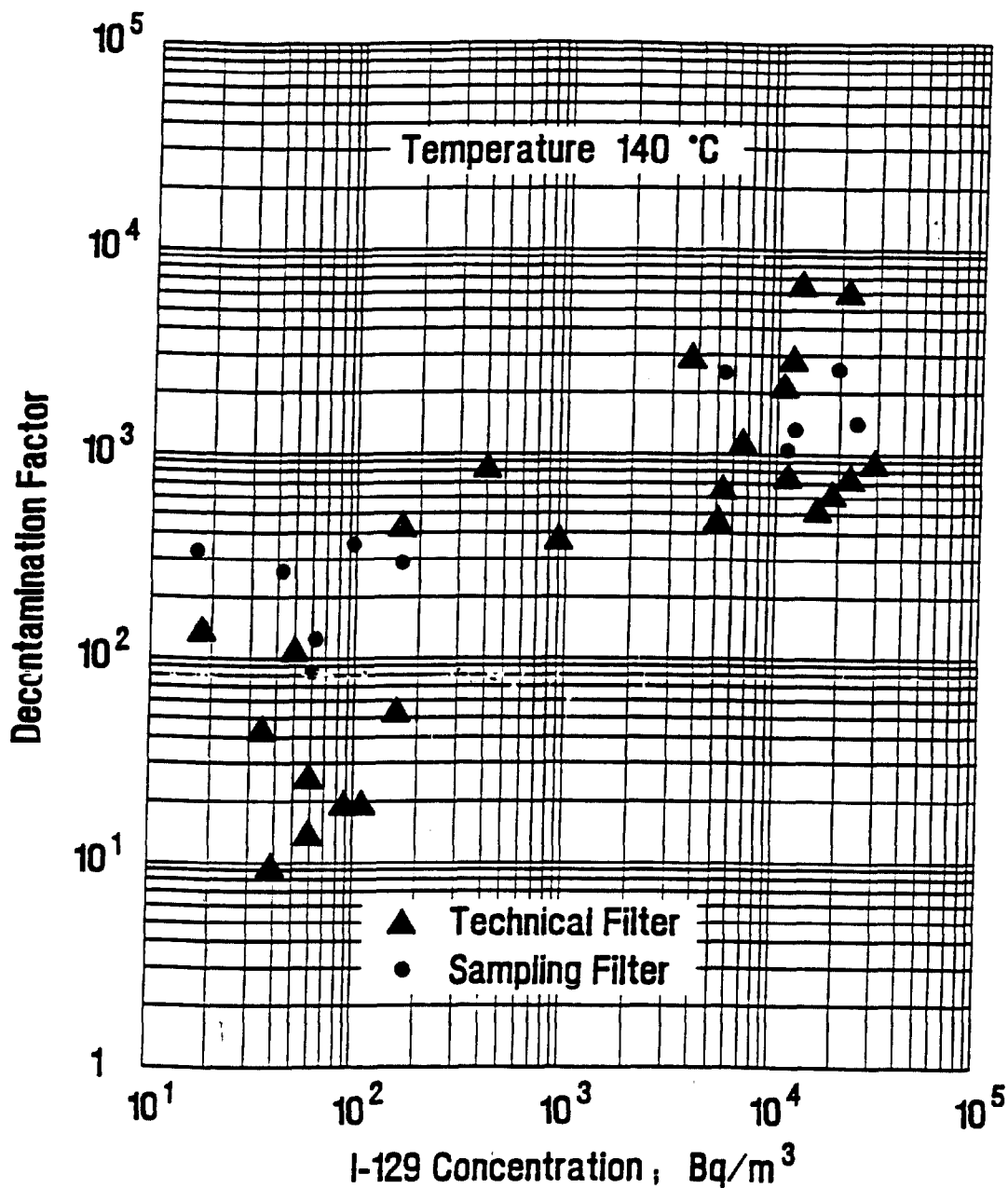
Decontamination Factor of Iodine Filter in DOG for different Iodine Concentrations
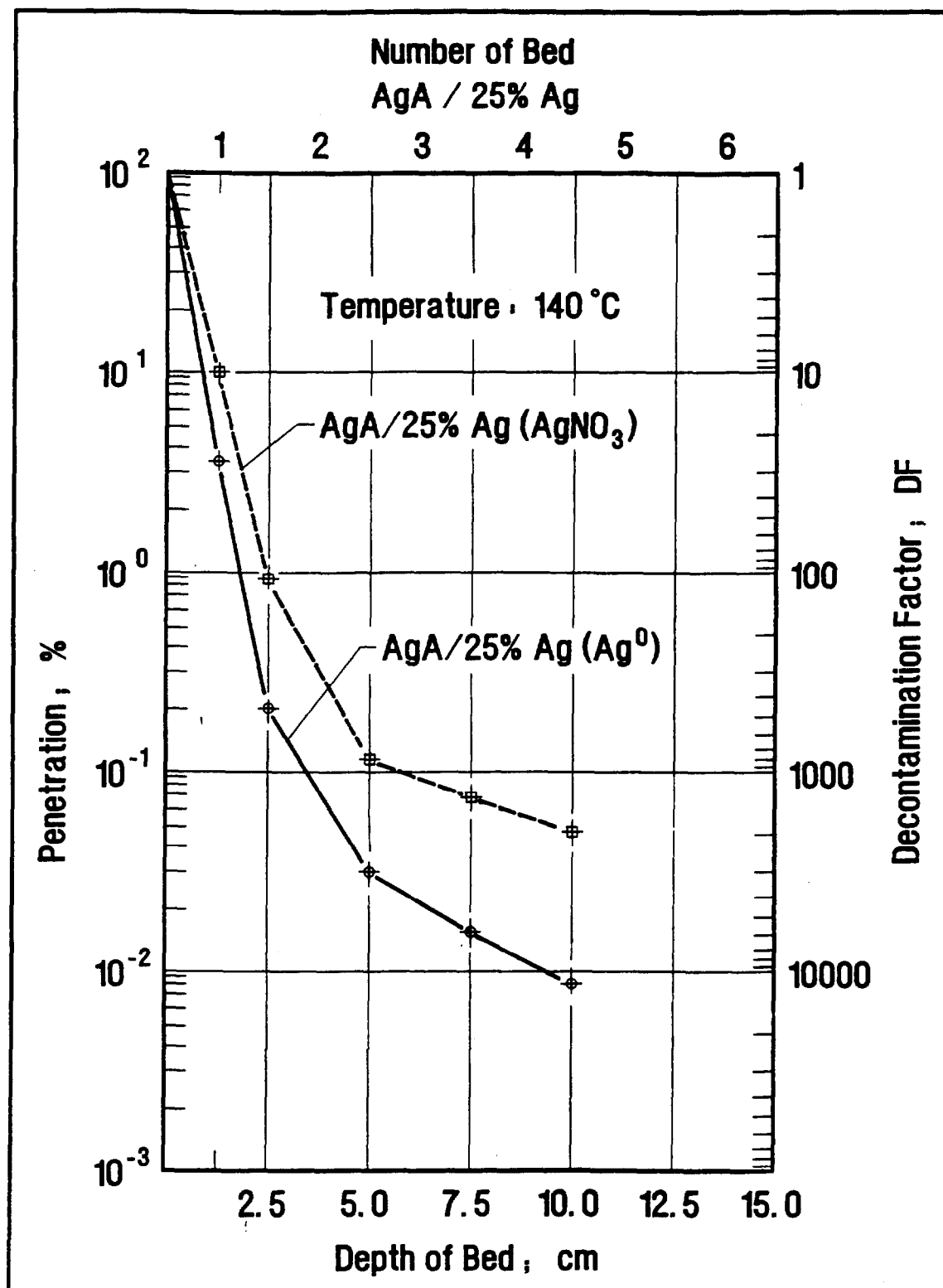
Decontamination Factor as Function of Bed Depth for AgA/25% Ag — Figure 6

IODINE STRIPPING FROM NITRIC ACID SOLUTIONS IN IATEMA

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Abstract

When fuel elements are dissolved 95 to 98 % of iodine present in the fuel elements as I-129 and I-127 are stripped from the dissolver solution into the dissolver offgas. In order to avoid that the admissible limits of I-129 release are exceeded, either a large number of vessel offgas lines must be equipped with iodine filters or iodine release must be optimized in such a manner that more than 99.6 % of iodine gets into the dissolver offgas.

The results of experiments carried out in an industrial scale iodine desorption facility (IATEMA) showed values of residual iodine in the dissolver solution of less than 0.4 %. The iodine can be removed from the dissolver solution into the dissolver offgas after addition of carrier iodate, thermal treatment of the solution, and reduction of the iodate by NO_2 in a stripping column.

I. Introduction

The iodine-129 (I-129) fission product with its long half-life of 1.57×10^7 a is a serious problem in reprocessing plants. In recent years highly effective iodine filters have been developed for iodine compound retention in the offgas [1,2]. With the AC 6120 iodine sorption material used in the iodine filters a decontamination factor (DF) of ≥ 1000 is achieved for iodine released with the dissolver offgas; this material has been applied in the dissolver offgas of the Karlsruhe Reprocessing Plant since 1975. Requirements imposed later by the authorities on the formerly planned German reprocessing plant at Wackersdorf, specifying a very low emission value of 50 mCi/a = 1.85×10^9 Bq/a for I-129 have led to a new strategy to cope with iodine-129.

According to that strategy iodine was to be released almost quantitatively in a controlled manner via a defined release path. The preferred path has been the dissolver offgas. It has been possible by this approach to reduce the highly expensive iodine filtration in the vessel offgas (volume flow rate in the vessel offgas 5 - 10 times higher than in the dissolver offgas) and to bind iodine directly to sorption material suited for repository storage. This very economical solution that more than 99.6 % of iodine is removed from the feed solution has been associated with the problem of providing the technical means of releasing such a high fraction of iodine.

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Experiments on the release from nuclear fuel solutions in the Karlsruhe Reprocessing Plant (WAK) yielded residual contents of 2 - 5 % of I-129 originally present in the fuel elements [3]. Simulation experiments carried out on a laboratory scale and in industrial plants yielded residual iodine contents in the simulant solutions of 1 - 2 % which resulted from boiling at reflux and sparging with nitric oxides (NO_x). Additional studies were performed in the WAESCHE-PASSAT dissolver offgas simulation facility where residual iodine contents in the simulant solution of 0.2 - 0.5 % were obtained [4]. After an accurate method of analysis had been elaborated for I-129 in nuclear fuel solutions the residual iodine content after dissolution of a high burnup fuel and treatment of that solution was recorded quantitatively for the first time [5,6]. These experiments were carried out in the hot cells. By addition of 10^{-4} mole/l carrier iodine in the form of KIO_3, sparging with air or oxygen, and thermal treatment at > 100 °C, and following sparging with NO_x and filtration a high degree of stripping was obtained. Besides, it has been demonstrated that the fraction of residual iodine in a nitric acid solution, molarity between 3 and 7, is nearly independent of the history of the acid, except for the dependence on the concentration of iodine originally contained in the acid. The presence of uranyl nitrate, fission product simulants, recycled acid or only 5 mole/l of reagent grade nitric acid yielded with the same initial iodine concentration residual iodine fractions on the same order after iodine stripping by boiling and sparging with air, nitrogen or oxygen. In order to transfer the findings from many laboratory tests to the industrial scale, the IATEMA iodine stripping facility was planned, built and put into service by Hauptabteilung Ingenieurtechnik (Central Engineering Department) in cooperation with Laboratorium für Aerosolphysik und Filtertechnik (Laboratory for Aerosol Physics and Filter Technology). The facility has been designed for a throughput in a reprocessing plant of 2 metric tons per day of spent fuel. After feed clarification the nuclear fuel solution will be further treated in IATEMA with a view to minimizing the iodine fraction.

II. Objective and Description of the IATEMA Facility

It is proposed to demonstrate with the IATEMA iodine desorption facility that by iodine treatment and desorption the residual iodine fraction in the nuclear fuel solution can be greatly diminished. The target are I-129 contents of less than 0.4 % of the amount of iodine originally present in the fuel. As 2-3 % residual iodine fraction was measured after dissolution at WAK following several hours of boiling an additional DF of up to 10 has to be achieved.

II.1 Description of IATEMA

IATEMA comprises a storage tank of 2 m^3 volume (B1), a preconditioning column (K1) and an iodine desorption column (K2), a cooler and a pump (Fig. 1). The components and pipework were made of stainless steel 1.4306, the column inserts from 1.4301.

In the first step of the process, the preconditioning phase, an iodine isotope exchange is to be achieved by addition of inactive iodate in excess. It is the purpose of this treatment to replace the inactive iodine by the radioactive iodine present in the heavily volatile compounds. This treatment can be intensified by heating up

to boiling the dissolver solution in the storage tank, circulating it in the conditioning column, and sparging with nitrogen and synthetic air, respectively, in a countercurrent flow. Moreover, the high operating temperature promotes the degradation of iodine containing compounds. Oxygen, nitrogen and nitrogen oxides can be passed through the conditioning column.

In a second process step the radioactive and the rest of inactive iodine are desorbed in the iodine desorption column (Fig. 2).

Before the preconditioned dissolver solution is fed into the head of this column, nitric acid is brought to boiling in the column sump by means of a steam heated jacket heating. The rate of boiling is adjusted to approx. 10 %/h of the throughput of the dissolver solution feed. Iodine stripping is promoted by heating the dissolver solution to approximately the boiling point in the storage tank, metering the feed into the column head at a rate of 100 to 600 l/h, and supply of up to 2 m^3/h nitrogen dioxide together with nitrogen or synthetic air at a rate of 3 m^3/h at the foot of the column.

The iodine containing offgas is supplied to a heat exchanger which cools the offgas down to 60. °C. At this temperature a major part of the acid vapors condense. The amount of condensate serves to determine the boiling rate in the column sump while iodine is fed into the dissolver offgas section. The dissolver solution freed from iodine is discharged at the overflow and further treated in the extraction cycle.

In the IATEMA facility the process can be controlled in a batchwise or continuous mode. Consequently, the functions to be fulfilled by the IATEMA test facility include:

- taking over the iodine containing nuclear fuel solution from the dissolution/clarification/balancing (WAESCHE-PASSAT) functional group;

- reducing the residual iodine inventory in the nuclear fuel solution by continuous processing;

- providing the nuclear fuel solution conforming to the specifications for the purpose of extraction;

- maintaining normal operation during plant induced failures of preceding functional units;

- release of the iodine loaded offgases to the dissolver offgas system.

II.2 Sampling

Partial streams were passed through the iodine sorption material (AC 6120-12 % Ag) and their iodine contents determined in order to be able to determine the iodine concentration in the offgas streams of the columns. (Fig. 1: sampled gas = SG). Taking into account the partial gas volume flow rates and the sampling durations it was possible

to calculate the iodine concentrations and hence the aggregate amount of iodine entrained in an experiment.

Samples were collected at the outlet of the storage tank, between the two packing materials of the iodine desorption column, in its sump and at the outlet, in order to be able to determine the iodine concentration in the liquid phase (Fig. 1: sampled liquid = SL). Partial streams of the dissolver solution were delivered by a pump through a silicone rubber-sealed glass vessel and returned into the storage tank (Fig. 3).

By means of a syringe 10 ml solution each were collected and filled into a rubber-sealed glass vessel nearby. This work was done in a ventilated glovebox. The amounts of iodine were determined by γ-spectrometry (I-131) and the amount of iodine contained in the dissolver solution was determined by extrapolation.

II.3 Conditioning of the Dissolver Simulant Solution

The feed solution for iodine treatment in IATEMA is conditioned in the dissolver simulator of the WAESCHE and PASSAT facilities which are operated in combination with IATEMA [2]. The offgas produced during the experiments is cleaned in the PASSAT facility (aerosol and iodine retention). As radioactively tracered iodine is used in the experiments and temporarily NO_2 is supplied, the facility is operated as a closed loop for safety reasons.

In the dissolver simulator 300 l of 3 - 5 molar nitric acid are supplied and heated to about 105 °C. 300 g of I-127 are metered in over five hours as NaI (60 g/h) and 3×10^8 Bq I-131 as NaI. The feed tank is installed in an α-tight glovebox and is evacuated by means of an air jet pump. The air from the air jet pump is used to mix the solution. While iodine is metered in and for another hour the solution is kept boiling. The rate of evaporation is 10 %/h of the acid volume of the dissolver simulator. The mean temperature of the condensate rising in the offgas downstream of the dissolver is adjusted to the optimum working point of 60 °C. The condensate produced is returned into the dissolver. The NO_2-scrubber connected downstream is a bubble tray column of 400 l liquid volume. The iodine containing offgas is carried to the series-connected iodine filters passing through the removal components of wave plate demister, high efficiency mist eliminator (HEME), high efficiency particulate air (HEPA) filter . The iodine containing offgases of IATEMA are introduced upstream of the NO_2-scrubber.

After five hours of iodine dosing the solution in the dissolver is kept boiling for another hour in order to complete the first iodine stripping. The carrier iodine is then metered into the dissolver solution in order to achieve a sodium iodate concentration of 2×10^{-4} - 8×10^{-4} mole/l. Then the solution is quickly transferred into the storage tank B1 where the dissolver solution is subjected to thermal treatment.

III. IATEMA Pilot Plant Experiments

The following parameter variations should be studied in an attempt to investigate and optimize residual iodine stripping from the dissolver solution which had been transferred into IATEMA:

- addition of carrier iodine as potassium iodate;

- duration of thermal treatment of the solution in the B1/K1 conditioning loop;

- volume flow rate of the solution in the head of the K2 desorption column;

- number of desorption cycles in K2;

- for sparging with gas during desorption: amounts of NO_2 and N_2 as well as additional aeration with synthetic air;

- amount of residual iodine at the head end of IATEMA.

A total of 43 experiments of one week duration each were referred to for statistical evaluation.

III.1 Addition of Carrier Iodine

At the end of simulation of the dissolution carrier iodine is admixed in addition to the dissolver solution. By dosing iodine concentrations between $2x10^{-4}$ and $8x10^{-4}$ mole/l are set. For the experiments to be performed 2.5 %, 5 % and 10 % of the iodine originally present in the nuclear fuel solution are added as iodate. In this way, the goal is reached of improving the exchange of isotopes with the radioactive iodine present in the solution and to ameliorate stripping at a later stage.

When carrier iodine at the specified concentration of 2.5 - 10 % was added no noticeable influence was found on the fraction of residual iodine in the solution (Fig. 4).

III.2 Conditioning Time and Amount of Feed

The presence of very different types of iodine compounds in the iodine-nitric acid-nitric oxide system has been reported in the literature [7,8].

In all systems investigated organic impurities and organo-iodine compounds have been found and it has been supposed that inorganic iodine compounds are present in addition. The authors have demonstrated that HOI, H_2OI^+, IO_3^-, IO_4^- and I^- do not occur in the residue solutions. The compounds identified have been pentyl iodide and n-undecane iodide. Besides, quite a number of other iodides are present which, considering their small concentrations, could actually not be identified individually.

In order to enhance isotope exchange, to optimize mixing of the nitric solution, and to degrade or convert the mostly unknown iodine compounds, the iodate containing solutions were subjected to a ther-

mal treatment in the conditioning loop of IATEMA. Thermal treatment lasted 3, 4.5, 6 and 9 hours, respectively. The solutions were heated to boiling in the storage tank and circulated in the loop by a pump wh. e passing through the K1 column. At the end of the scheduled duration of treatment the solutions were treated in the K2 iodine desorption column as described in II.1. The hot solutions were supplied at a rate of 150 l/h and 300 l/h into the column head. The evaporation of about 10 % of the fractions of solution supplied in the sump of the column ensured the necessary vapor volume flow which moved as a countercurrent flow and was used to strip volatile iodine fractions from the solution. Simultaneously with the vapor, nitrogen oxides are introduced in a countercurrent flow in order to reduce the amount of iodate present. The supply of the energy of evaporation is controlled by measurement of the amount of condensate. 24 m^3 and 40 m^3 of vapor, respectively, per hour were made to counterflow 150 l and 300 l liquid, respectively, in the column. Iodine stripping takes place in the packing material of the column. The liquid is passed through the overflow into the storage tank underneath and measured to detect any residual iodine fractions present. In order to increase the time of holdup in the column, all the liquid can be pumped a second time through the column. Figure 5 shows the dependence of iodine stripping on the duration of conditioning. If only one stripping cycle is run in the desorption column, the percentage of residual iodine increases with the duration of conditioning. In case of two stripping cycles more than 85 % of the residual iodine has already been stripped during conditioning times of 4.5 hours.

Examination of the dependence of iodine stripping on the amount of liquid supplied yielded a much lower release of iodine at 300 l/h than at 150 l/h during the first cycle in case of batch operation in the column. During the second cycle the amounts stripped of about 88 % were roughly similar upon supply of 150 and 300 l/h liquid via the column head (Fig. 6).

III.3 Influence of NO_2 Sparging

During iodine stripping NO_2 (1 - 2 m^3/h) is continuously fed into the column sump. In addition, nitrogen as the carrier gas (1 - 3 m^3/h) as well as synthetic air (1 - 3 m^3/h) can be metered in. Nitrogen oxides are added to reduce the iodates into elemental iodine, and, besides - together with the oxygen - keep the redox potential so low that no new iodate is generated.

Dosing 1 m^3 NO_2/h into the vapor phase is sufficient to reduce the amount of iodate when the liquid circulates twice through the desorption column. No influence has been detected of nitrogen or synthetic air.

III.4 Amount of Residual Iodine at the Head End of IATEMA

It was possible to confirm in the industrial scale experiments the dependence of iodine stripping on the residual iodine content of the processed solution as found in the laboratory scale experiments (6). Figure 7 shows the residual iodine stripping, expressed in %, as a function of the amount of head-end iodine supplied to IATEMA. At residual iodine contents of 1 % further 95 % were stripped, whereas

at 0.1 % residual content at the head end only about 80 % were stripped.

IV. Conclusions

Residual iodine contents of dissolver solutions as might be present after dissolution of spent fuel elements can be considerably reduced. The experiments performed in IATEMA yielded at 0.1 - 1 % residual iodine contents depletions in iodine which ranged from 80 to 95 %, depending on the value of the head-end iodine concentration. However, this calls for several hours of intensive thermal treatment with iodate of the dissolver acid subsequent to feed clarification and transfer into a facility similar to IATEMA. In IATEMA experiments a DF ≥ 3000 was achieved, related to the head-end iodine concentration in the dissolver simulant solution. A DF of 250 was considered sufficient to do without iodine filtering in the vessel offgas in case of the planned German reprocessing plant.

The experiments carried out in IATEMA have allowed to validate the findings from laboratory scale studies in an industrial scale facility. The results obtained on a laboratory scale, in experiments conducted in hot cells with spent fuel, and on an industrial scale in IATEMA using simulant solutions, while varying quite a number of parameters, allow an extrapolation to be made to industrial scale reprocessing.

If the residual iodine is stripped in a reprocessing plant characterized by low values of residual iodine in the dissolver solution of < 0.3 % of the amount of head-end iodine, one can possibly fully dispense with iodine filtration of the vessel offgas. This means that iodine filtration of the dissolver offgas achieving a DF ≥ 1000 would be adequate.

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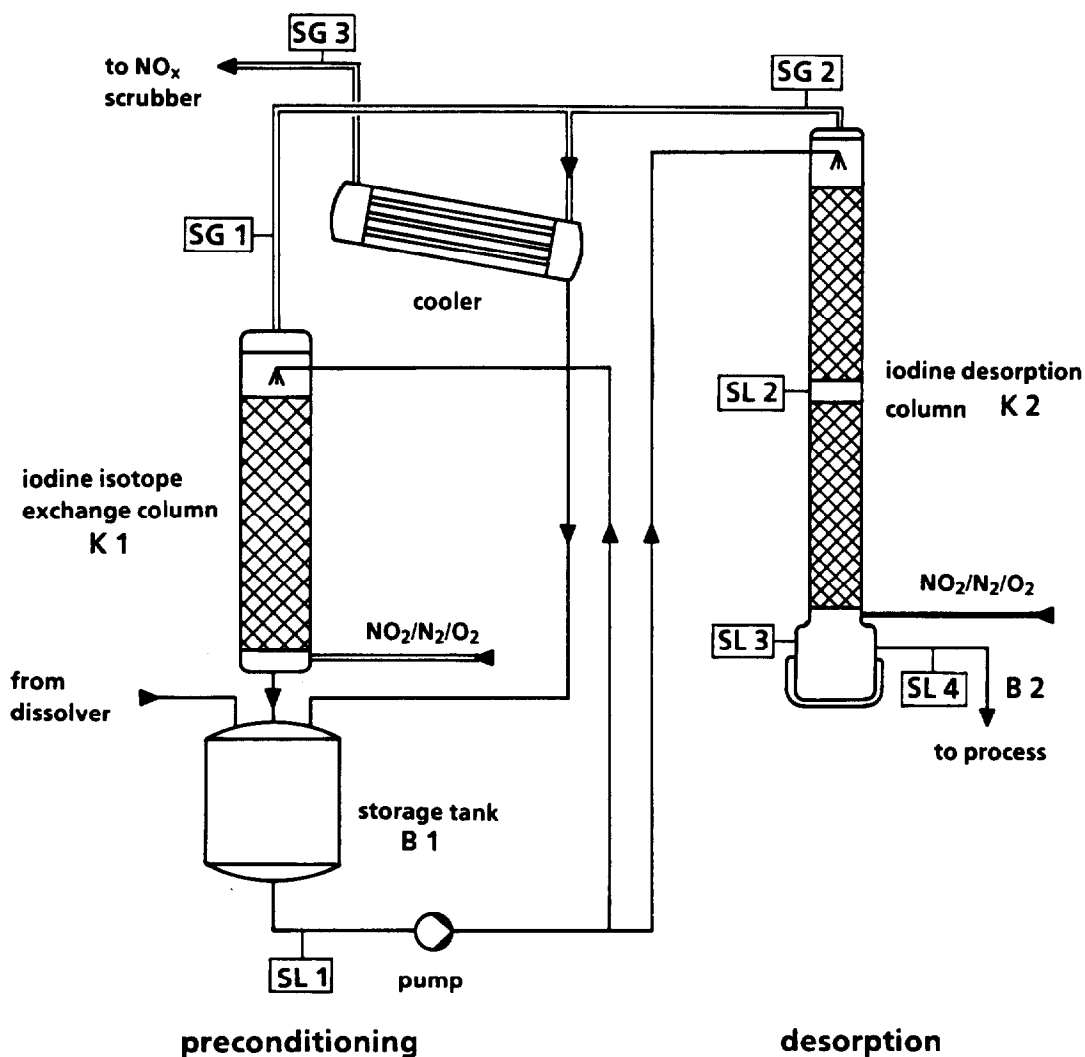
Figure 1 IATEMA: Iodine Stripping Facility

Figure 3   IATEMA : Iodine Desorption Column
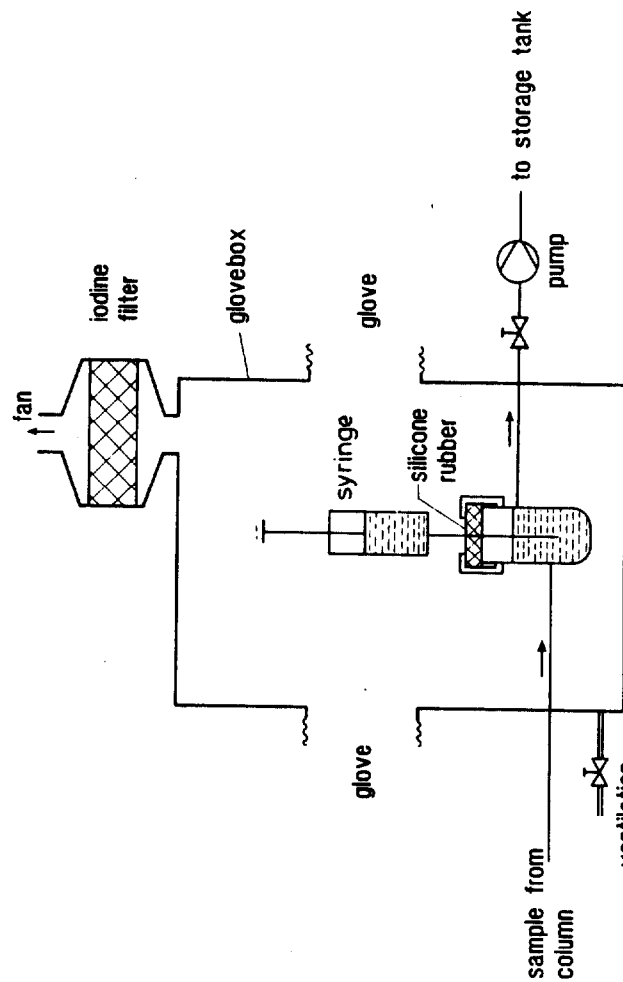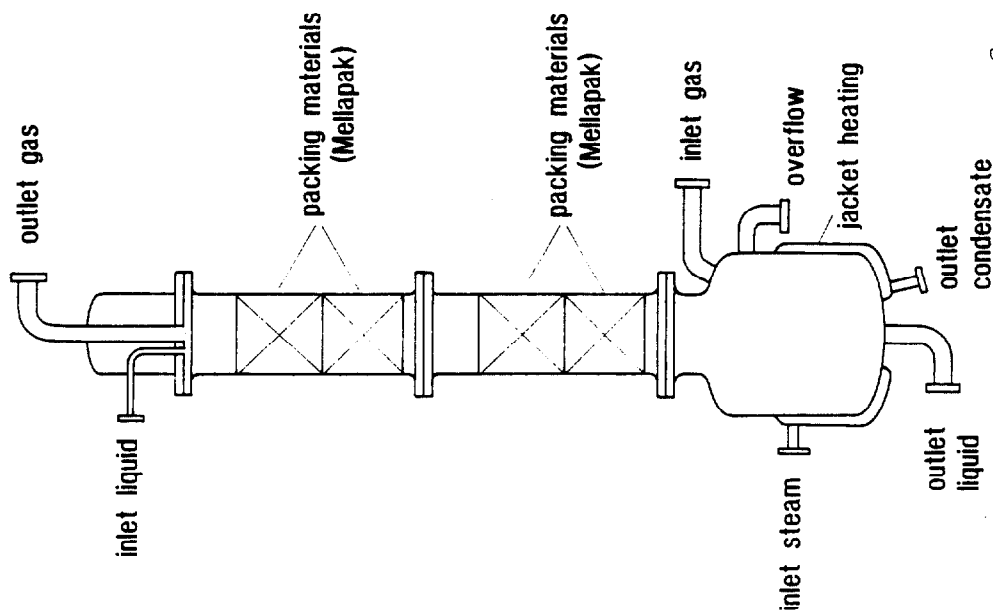
Figure 2   IATEMA : Liquid Sampling Device

IODINE STRIPPING IN % OF INPUT

DURATION OF ISOTOPE EXCHANGE [h]

o 12.6 g KIO3   ▽ 25.3 g KIO3   * 50.5 g KIO3
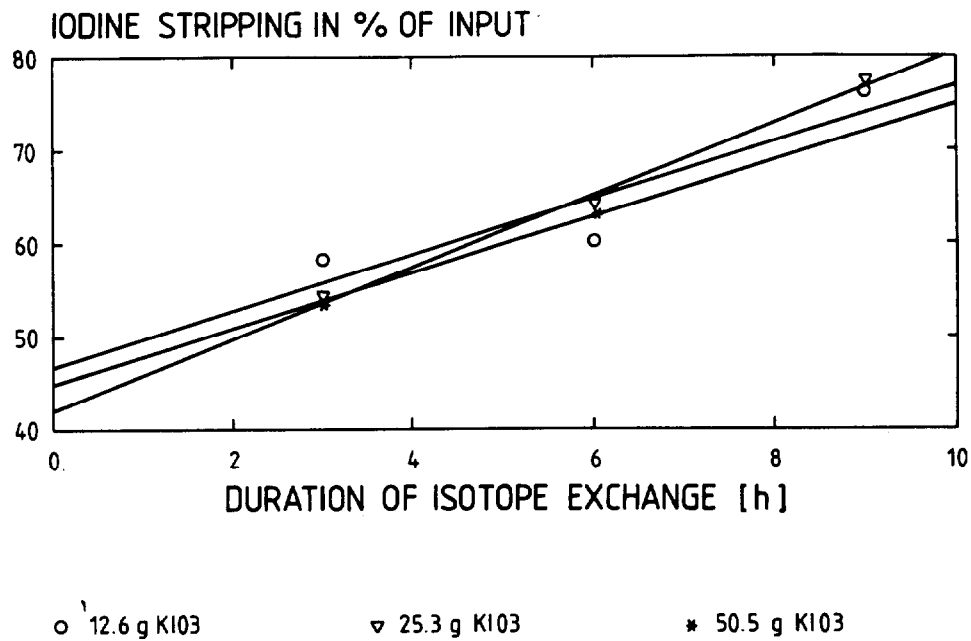
Figure 4   IATEMA - Optimization

Variation : Duration of Exchange / Amount of Carrier Iodine

IODINE STRIPPING IN % OF INPUT

DURATION OF ISOTOPE EXCHANGE [h]

■ 1 cycle in K2   * 2 cycles in K2
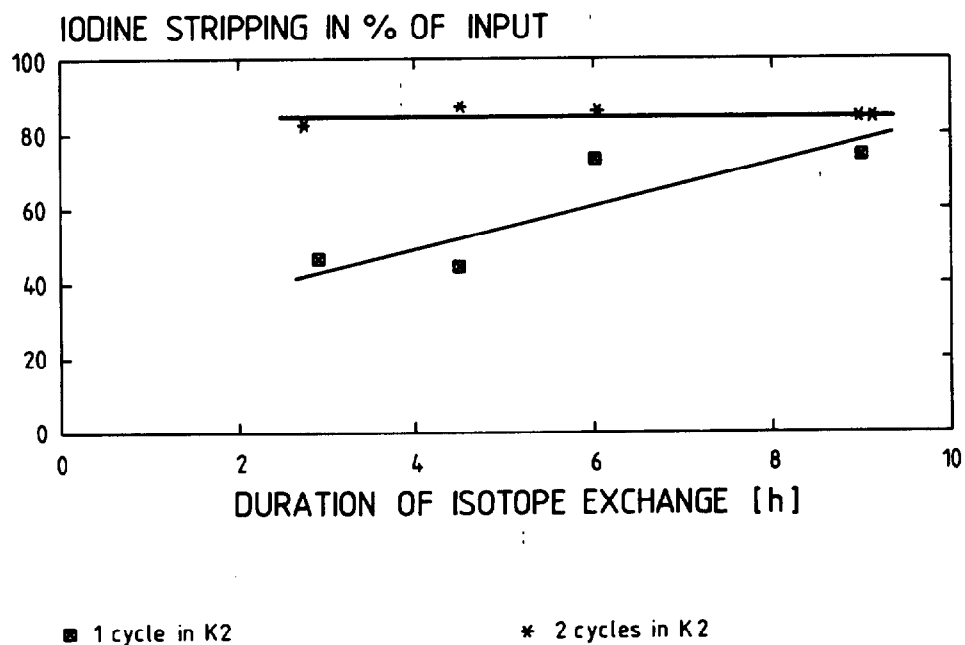
Figure 5   IATEMA - Optimization

Variation : Duration of Isotope Exchange

IODINE STRIPPING IN % OF INPUT

AMOUNT OF FEED IN K2 [l/h]

o 1 cycle in K2   * 2 cycles in K2
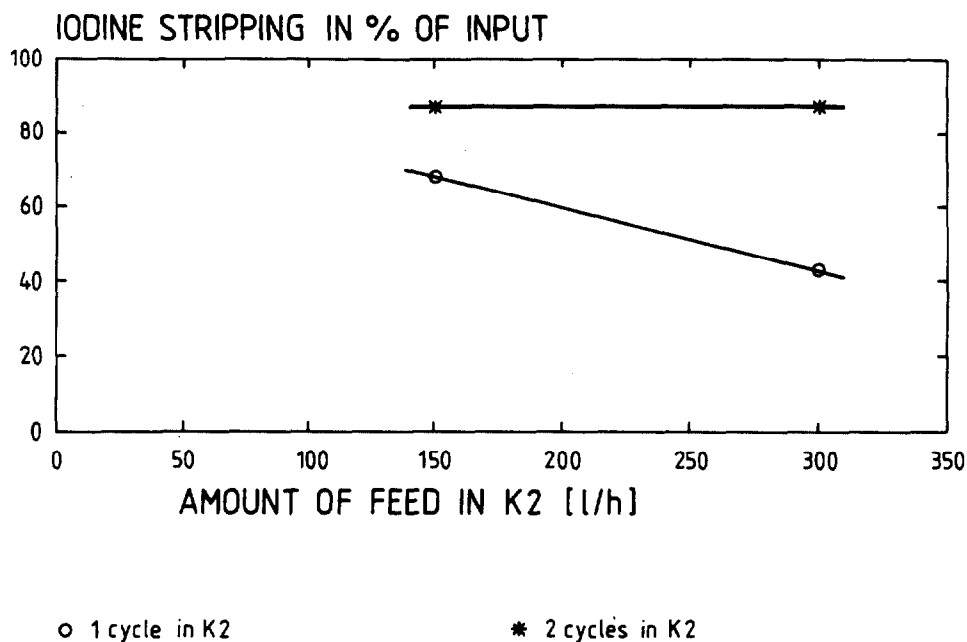
Figure 6   IATEMA-Optimization

Variation: Amount of Feed in the Desorption Column K2

IODINE STRIPPING IN % OF INPUT

IODINE INPUT IN % OF INVENTORY

* 2 cycles in K2
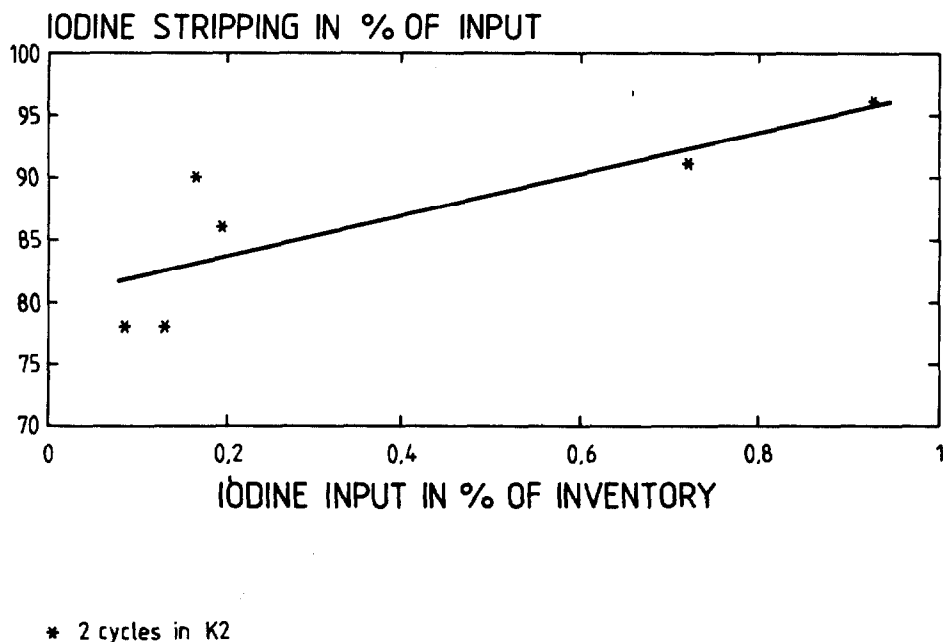
Figure 7   IATEMA-Optimization

Variation: Input of Residual Iodine

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The Development of Corona Discharge for Iodine Removal From Nuclear Fuel Reprocessing Plant Off-Gases

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Abstract

Studies on single, wire-in-tube corona discharge experimental rigs have been carried out to examine the behaviour of molecular iodine, organic iodide and oxides of nitrogen (NO_x) in both glow and streamer corona discharges. The parameters investigated include electrode potential, electrode geometry, gas composition and throughput, and for the streamer corona, pulse length and repetition frequency. Operations with continuous irrigation of the ground (tube) electrode for both glow and streamer discharges are also discussed.

Organic iodine DFs in excess of 10^4 (>99.99% removal) have been readily achieved, and the use of streamer coronas has significantly reduced the interfering effects of NO_x and moisture which have been established in earlier work (1).

Two outline process options are presented, one based on a single stage irrigated streamer corona and the second on a two stage scheme utilising an irrigated glow discharge unit as the first stage for NO_x removal, followed by a second stage dry streamer unit for iodine removal.

The estimated power consumption and unit size of corona units for treating dissolver off-gases from a reprocessing plant with a nominal 5te/day fuel throughput are presented.

1. Introduction

Of the gaseous species released during nuclear fuel reprocessing from fuel shearing and dissolution in nitric acid, iodine - 129 is of particular importance due to its long half-life (1.6×10^7 years). Around 99% of the total iodine is volatilised into the Dissolver Off-Gas (DOG) stream. Traditionally this stream is passed through nitric acid and caustic scrubbers to remove NO_x (predominantly NO and NO_2) and iodine.

Some of the iodine released is present as organic iodides. Although their concentration is small relative to molecular iodine, they provide a limit on the total iodine decontamination factor (DF) obtainable by conventional caustic scrubbing (typically <100) as they are not absorbed efficiently. In investigating alternative methods for improved iodine removal, the behaviour of organic iodides is, therefore, particularly important.

The use of corona discharge for the removal of iodine was originally proposed by Dickson et al in 1985 (2) for the removal of iodine from containment air in the event of a reactor incident.

Studies carried out in relation to the treatment of fuel reprocessing off-gasses were first undertaken by AEA Technology in 1989 (1). These single tube experimental studies were carried out using a continuous glow discharge corona and resulted in the publication of a Patent for a two stage wet and dry corona system (3). The wet first stage uses a continually irrigated ground electrode to provide high efficiency removal of NO_x which otherwise interferes with iodine removal. The second dry stage can effect iodine DFs of $>10^4$.

The use of steamer coronas have been investigated by other workers (4-8) primarily for the treatment of NO_x and SO_x from flue gases. The potential advantages of using streamer coronas are associated with lower power consumption and a more efficient utilisation of the inter-electrode reaction zone within the corona unit. The use of streamer coronas for fuel reprocessing off-gas treatment have been investigated by AEA Technology (AEA D&R) under contract for BNFL. In this paper the results of these experimental studies are presented and compared with earlier glow discharge data.

2. Nature of the Corona

2.1 Corona Formation

A glow discharge is produced when an electrical potential is applied between two electrodes forming a non-uniform electric field. Electrode geometries which provide a suitable non-uniform electric field include wire and plate, and wire-in-tube. The glow discharge is produced as the potential is increased by electrical breakdown at the surface of the wire. Coronas may be positive or negative depending on the polarity of the central discharge electrode. Negative coronas were studied exclusively in the earlier glow discharge studies (1) due to their reduced tendency to spark-over and the higher ozone production rates that are achievable.

Operation at potentials close to spark-over is the basis for the production of streamer coronas. Observation of spark formation shows that prior to complete breakdown, filamentary streamers cross the inter-electrode space. If the potential is applied for a time period so short that the streamers form, but sparking is prevented, typically < 1 microsecond, then this is described as a streamer corona discharge.

Pulses of power may be repeated at, typically, around 100 Hz energising much of the gas in the reactor volume with each pulse. This is different to a glow discharge where only the reactor volume local to the discharge electrode is highly energised.

For streamer coronas, a positive discharge produces current densities approximately 100 times greater than a negative discharge. This was confirmed in the early stages of the experimental programme and subsequent studies concentrated on positive discharge operation.

2.2 Chemical Reactions in the Corona

The reactions occurring in the corona are dependant on the nature of the corona, i.e. whether positive or negative, glow or streamer, and on the composition of the gas.

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Oxygen concentration is an important parameter, the oxygen tending to produce anionic species, atomic oxygen and ozone. These species may be in excited states which react readily with other gaseous molecules in the gas stream.

Oxides of nitrogen (NO_x) are important pollutants, and their behaviour in the corona has been studied by a number of workers (3, 5, 7 and 8). In air the reaction of NO with oxygen species generated within the corona occurs readily, e.g.:

$$NO + O_3 \rightarrow NO_2 + O_2 \tag{2}$$

In addition, NO_2 or NO can also give N_2O_5

$$2NO_2 + O_3 \rightarrow N_2O_5 + O_2 \tag{3}$$

Under certain conditions, i.e., at high current, atomic nitrogen may be formed and react with either oxide, however, in air the oxygen reactions are expected to predominate. Earlier studies (1) have confirmed that the presence of moisture assists in the removal of NO_x from the corona.

Methyl iodide (MeI) will react efficiently with free electrons to give Me· and I⁻. However, in air, reactions with oxygen species are likely to dominate (2), e.g.:

$$O^- + MeI \rightarrow Me· + IO^- \tag{4}$$
$$O· + MeI \rightarrow ·CH_2I + ·OH \tag{5}$$
$$O_2^- + MeI \rightarrow Me· + I^- + O_2 \tag{6}$$

For iodine, similar reactions take place. The radicals and ions produced react further with oxygen species eventually forming deposits of I_4O_9 on the anode and I_2O_5 on the cathode. In the presence of moisture the acidic oxides may also be hydrated, and are all water-soluble.

<div align="center">3.   Experimental Equipment</div>

3.1 General Description and Glow Discharge Operation

This can be segregated into three main areas: gas mixing and flow control; the corona unit and associated pulsed power supply; and the gas analysis system. A flow diagram of the system is given in Figure 1.

Gases, with the exception of nitrogen and air, were supplied from standard gas cylinders, mixed where appropriate by varying flow rates of two or more streams and/or using a gas divider (Signal 821). For MeI, diffusion tubes of various rates were used with a small (200 cm^3 min^{-1}) proportion of the flow to the corona passed over them at ambient temperature. In the case of iodine, a similar system was used with the entire flow passed over a number of tubes thermostatted at temperatures up to 100°C.
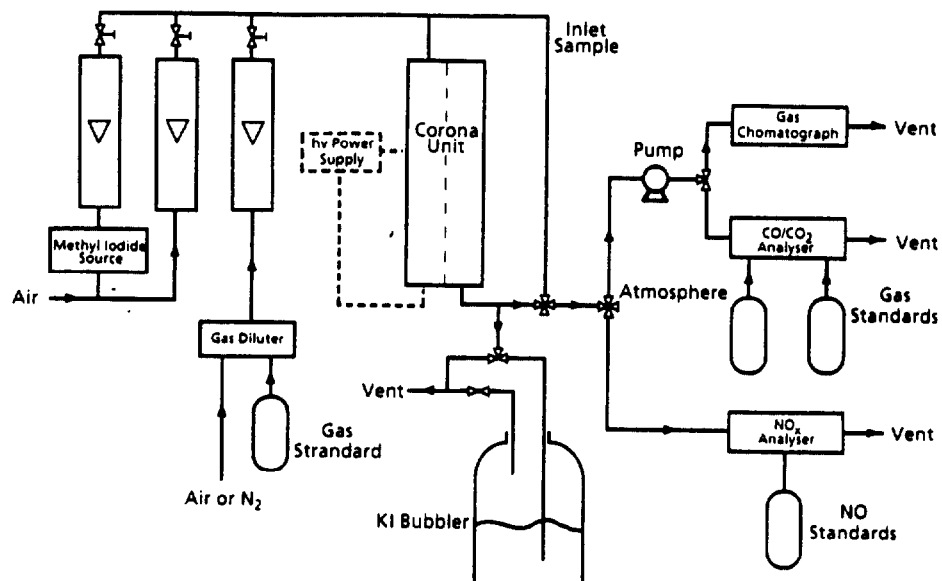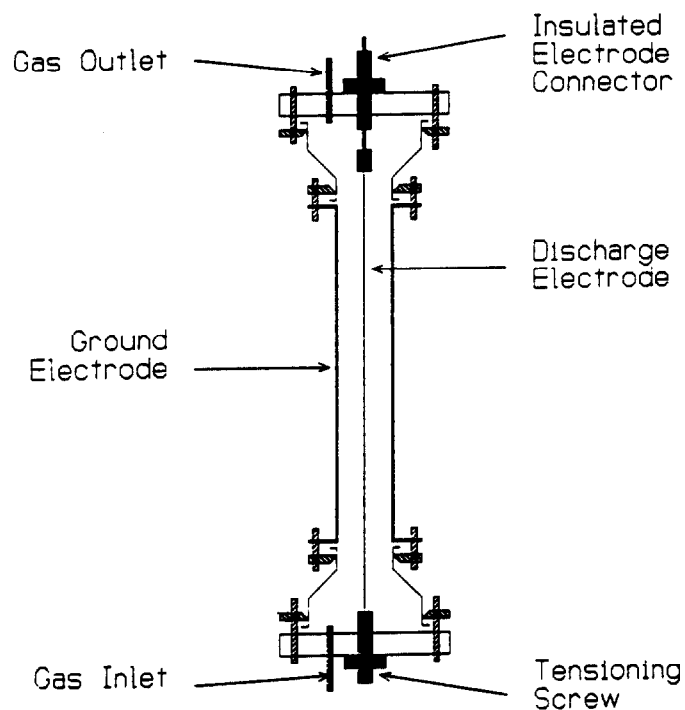
Figure 1

Flow Diagram of Experimental Rig

Figure 2

Corona Discharge Assembly

The standard corona unit, illustrated in Fig 2, consists of a cylindrical ground (+ve) electrode with wire corona electrode held taut by a tensioning screw.

Methyl iodide was analysed by gas chromatography (Varian 3400 GC) with DB-1 non-polar megabore column at 30°C. A flame ionisation detector was used to analyse concentrations above 200 vpb (parts per billion by volume), whilst an electron capture detector (ECD) was used for lower concentrations. The lower detection limit (DL) varied according to the other constituents present between 0.1 and 10 vpb.

For iodine, the same system was used with the column at 60°C and using an ECD detector. In this case the DL was of the order of 0.01 vpm (parts per million by volume), i.e. 10 times less sensitive.

When NO_x was included in the gas stream this was analysed using a chemilumnescence NO_x analyser (Signal Series 4000). This gave readings both for total NO_x and NO and the difference between the two was quoted as NO_2, though any other species (e.g., HNO_3) which could be converted to NO on the heated platinum catalyst would also have been detected.

For irrigated electrode experiments the ground electrode (38 mm id.) was irrigated by the use of a weir arrangement. The liquor passed out of the system via a U-tube to be recycled by peristaltic pump. In this case, gas flow was downwards, i.e. co-current, to prevent condensation on the electrode insulator and hence spark-over.

3.2 Pulsed Power (Streamer) Operations

These experiments used the same wire-in-tube corona unit used in the earlier glow discharge experiments. The pulsed power supply to generate the streamer corona was a purpose-built unit designed and built by AEA Industrial Technology. No commercial pulsed power supply units were available which would provide the desired pulsing characteristics.

The design was based around a 10 kV, 150 mA power unit (Wallis Hivolt OL150) which was used to provide power for the pulses and a 15 kV, mA supply (Brandenburg Gamma range) which provided a dc bias for some experiments.

The pulsar unit used a network of resistors and capacitors to charge the system and this was discharged across a spark gap using a trigger generator operating at frequencies between 1 and 100 Hz. A second spark gap cut the pulse short, to produce a close approximation to a square wave. The pulse length was varied, discretely, by using different length delay coils between the two gaps.

Following initial operation of the pulsed corona unit in dry conditions, continually irrigated operations were also carried out. The pulse generator was modified to accommodate this and permit reversal of the electrode polarity for both negative and positive corona generation. As a result of these modifications changes in the pulse rise time were noted. The effects of this are discussed in section 4.2.2.

An important consideration in the operation of pulsed coronas is the generation of radio-frequency (RF) interference. To screen against this, the whole system including corona unit and pulse generator network were incorporated into a RF screened metal cabinet.

4.   Results and Discussion

4.1 Glow Discharge Performance

The results from earlier glow discharge studies published previously (1) can be summarised as follows:-

i)   DFs for molecular iodine and methyl iodide of $>10^3$ can be achieved in the absence of water vapour and NO_x. NO_x above ~ 100 vpm reduces the iodine DF. The iodine oxide deposits on the ground electrode are highly soluble and can be readily washed off.

ii)   Nitric oxide is very efficiently oxidised to NO_2 and higher oxides of nitrogen in the corona. These are efficiently removed by using a continuously irrigated ground electrode. Separate experiments to measure the ozone generation rate confirm a linear relationship between this and the NO oxidation efficiency.

iii)   A minimum oxygen concentrations of 10% is required to ensure efficient iodine and NO_x removal.

iv)   Low concentrations of CO_2 and CO (up to 100 vpm) in the influent gas stream have no effect on iodine removal. There was no evidence, however, of oxidation of CO to CO_2.

v)   The electrode geometry and influent gas concentrations also have a significant influence on the decontamination efficiency.

vi)   To remove both NO_x and iodine efficiently from a gas stream using a glow discharge corona, a two stage system is necessary. The first continuously irrigated stage removes the bulk of the NO_x and the second dry stage removes the iodine. Dehumidification of the gas stream is required between the two stages.

For a gas containing ~ 0.1% NO_x, 10 vpm I_2 and 1vpm MeI, at a nominal flowrate of 100 m^3h^{-1}, the primary corona power requirement would be ~ 25kW. The second dry unit would have a power requirement of <10 kW. Each unit would occupy a volume of approximately 1m^3 (approximately square dimensions).

4.2 Streamer Corona

4.2.1   Comparison of Positive and Negative Streamers

Initial experiments to establish the characteristics of the corona showed that a positive corona could produce currents at least 10 times higher than a negative corona. This is consistent with results reported by Urabe (5).

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Ozone production rates were also measured and an approximately linear relation to electrical power was noted for both positive and negative operation as illustrated in Figure 3. It has been shown that although higher currents can be generated for positive coronas more power is required to generate the same ozone concentration.

On balance, however, from measurements of methyl iodide and NO_x removal efficiencies it was found to be more advantageous to achieve higher ozone concentration from the positive streamer discharge than to maximise power utilisation efficiency, and thus positive discharges were used in most of the subsequent experimental studies.

For comparison, the ozone generation from a glow discharge on the same corona unit are plotted on the same graph. It should be noted that the values of electrical power are at least ten times higher for the glow discharge to produce similar ozone concentrations.

4.2.2   Effect of Pulse Parameters

Pulse Rise Time

As stated in section 3.2, in modifying the pulsar unit for irrigated operation the pulse rise time was increased from 35 to 70 ns. This had an unexpectedly profound effect on the removal efficiency of methyl iodide. For example two experiments were carried out under identical conditions apart from the rise time change, i.e. MeI feed concentration of 7 vpm, 8 lpm gas flow, 150 ns pulse length, 5Hz pulse frequency and 70A average pulse current. The experiment using the 35 ns pulse rise time gave a MeI DF of ~550, compared with only 10 for the 70 ns pulse rise time.

Figure 3

Ozone Concentration as a Function of Power
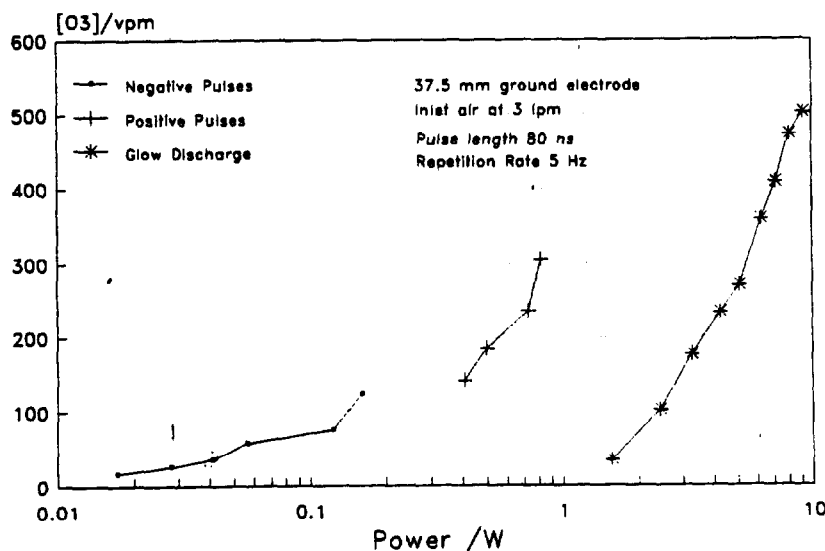
Pulse Length

Three different pulse lengths, 80, 120 and 150 ns were examined for two different tube diameters (37.5 mm and 50 mm). For both tube sizes it was found that the ozone concentration produced and MeIF DFs increased only slightly with increased pulse length (Figure 4).

This implies that the initial 'burst' of power is the most effective phase of the pulse cycle. For all experiments, the larger diameter tube gave a higher MeI DF. This was probably due to the increased gas residence time.

Pulse Repetition Frequency

Ozone generation rates were measured for both positive and negative coronas and were found to give an approximately linear relationship to pulse repetition frequency (Figure 5). Methyl iodide DFs measured over a range of other conditions, tube size, NO_2 concentration etc, also exhibited a similar relationship. From this, it is clearly preferable to operate at the highest frequency that can be practicably achieved.

Figure 4

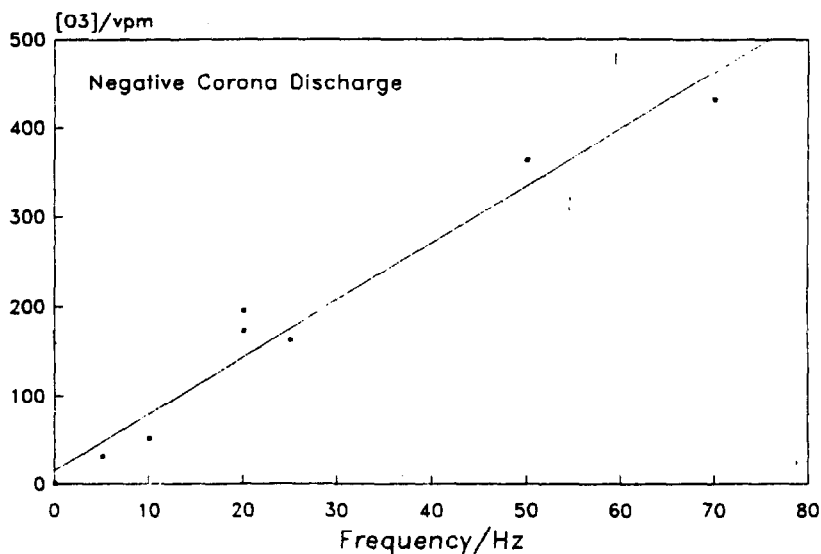### Effect of Pulse Length on Iodine Removal

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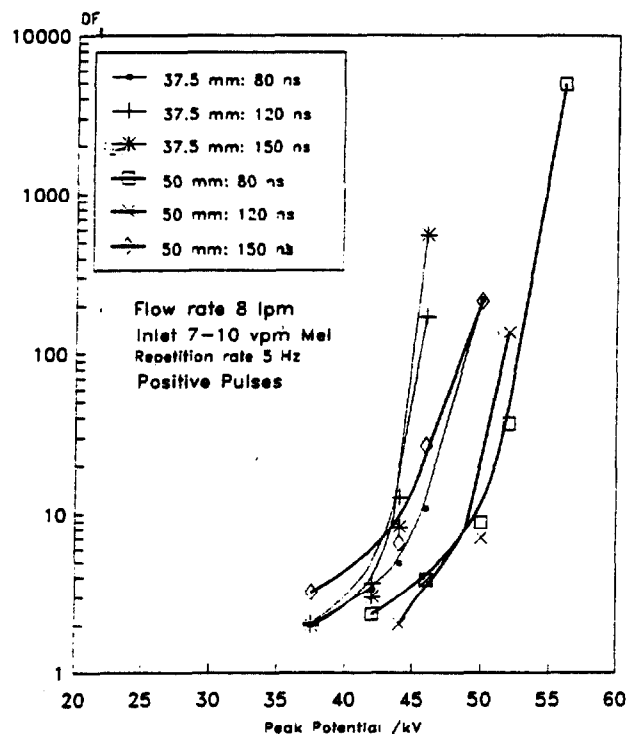
Figure 5

Effect of Pulse Frequency on Ozone Production

4.2.3   Irrigated Operation

Experiments carried out at low and high humidity showed that using a pulsed corona, water vapour had much less of an interfering effect than for a glow discharge. Subsequent studies using pulsed corona with a continuously irrigated ground electrode confirmed this. Results of some of these experiments are given in Figure 6. The data presented in this graph also shows the NO_2 at lower concentrations, i.e. ~ 100 vpm, has little or no effect on the MeI DF.

From these experiments and earlier glow discharge data two points emerged:

i)   That by using an irrigated glow discharge for the first stage, bulk NO_x could be effectively removed typically from 1000 vpm to around 100 vpm. By using a streamer corona second stage, only crude dehumidification between stages would be required to ensure effective iodine removal.

ii)   The data on pulsed corona operations were sufficiently encouraging to offer the possibility of a single stage streamer corona.

This second option was investigated in practice using a gas feed containing ~ 900 vpm NO_2 and an inlet MeI concentration of 8-9 vpm.

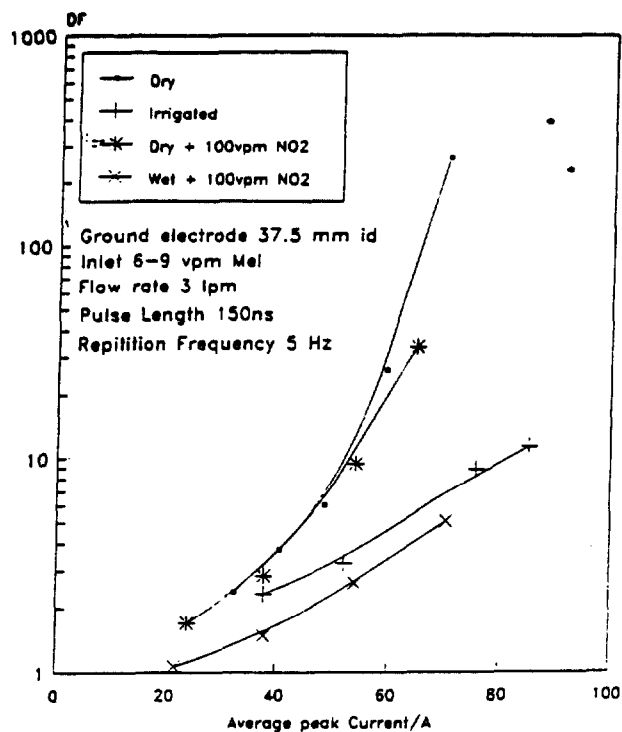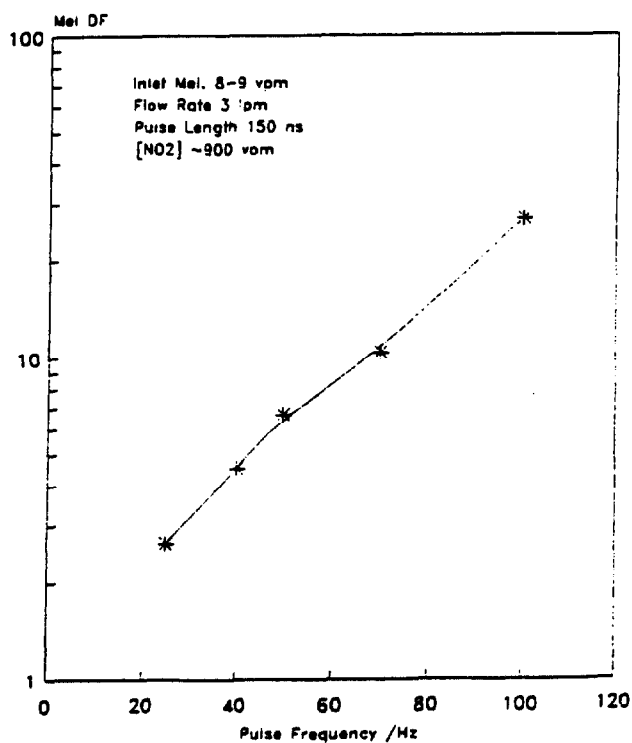110

Figure 6

Effect of NO2 and Irrigation on MeI Removal

Ground electrode 37.5 mm id
Inlet 6-9 vpm MeI
Flow rate 3 lpm
Pulse Length 150ns
Repitition Frequency 5 Hz

Legend:
— Dry
+ Irrigated
:—*— Dry + 100vpm NO2
—x— Wet + 100vpm NO2

Average peak Current/A

Figure 7

Effect of Pulse Repetition Rate on MeI Removal in a Pulsed Irrigated Corona with NO2

Inlet MeI. 8-9 vpm
Flow Rate 3 lpm
Pulse Length 150 ns
[NO2] ~900 vpm

Pulse Frequency /Hz

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The data on MeI DF plotted against increasing pulse frequency is given in Figure 7. Although the MeI DFs are reduced by the presence of high NO_x concentrations at 100 Hz, a DF of 30 was achieved. Under these conditions, approximately 60% NO_2 removal was achieved down to a residual 300 - 400 vpm. At lower NO_x concentrations, e.g. ~ 100 vpm, NOx can be removed down to < 10 vpm (see Figure 8).

In the absence of NO_x, very high MeI DFs can be achieved even for an irrigated system. Figure 9 presents data for two different tube diameters, 37.5 mm and 75 mm at the same gas flow rate giving residence times of 8 and 24 seconds respectively. Note that when the gas flow rate was increased to maintain the same residence time (8 seconds) the DF was drastically reduced for the 75 mm diameter tube. Re-plotting these data for the irrigated experiments as moles of MeI removed per second against power consumption shows that although the DFs were lower, more methyl iodine is removed per unit power consumed. (See Figure 10.)

Figure 8
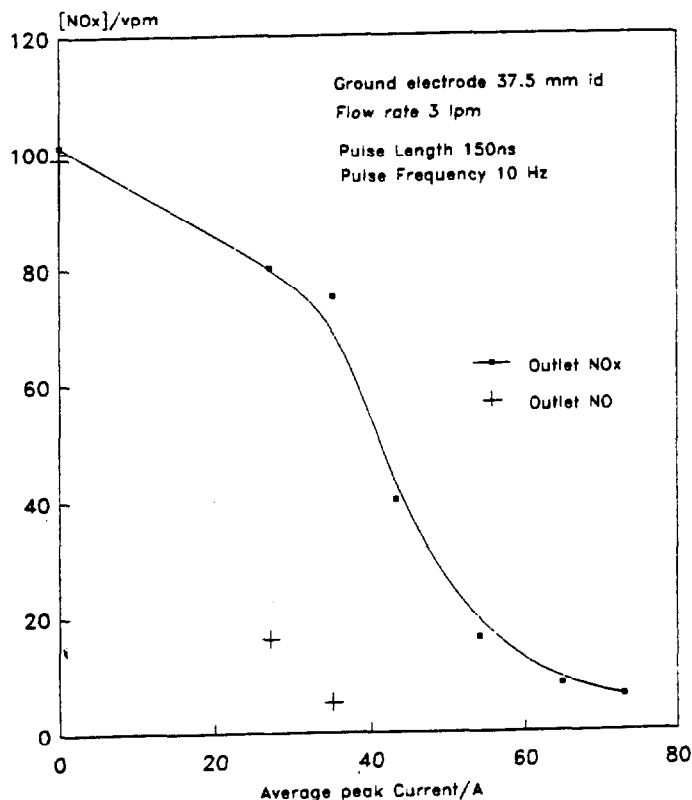
NO_x Removal From a Pulsed Irrigated Corona

Figure 9

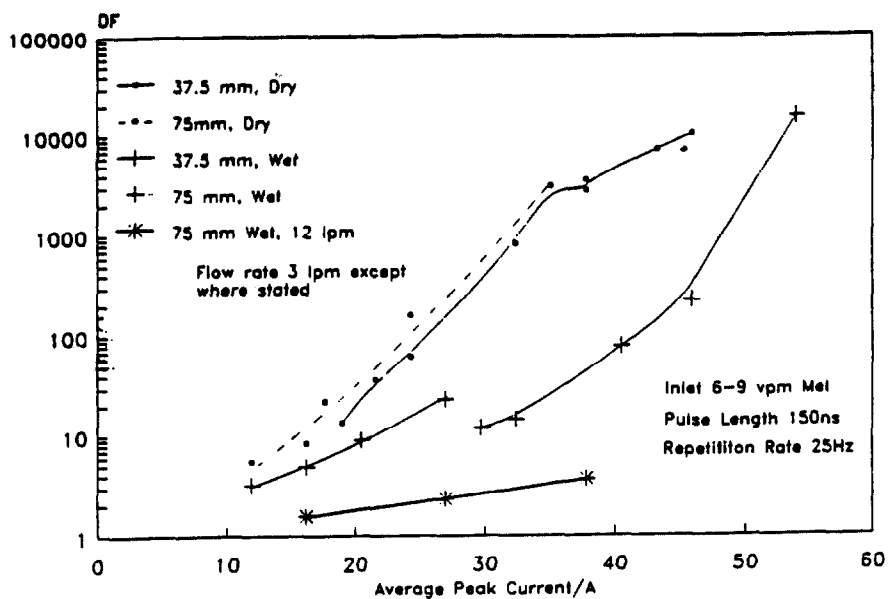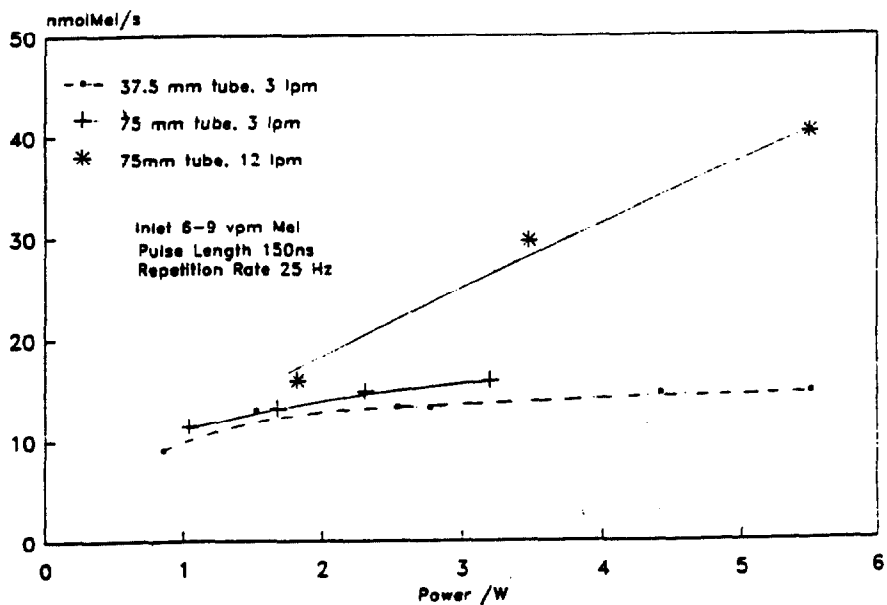## MeI Removal as a Function of Peak Current

Figure 10

Molar MeI Removal as a Function of Power

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5.   Process Considerations

The data obtained at this stage of development for the streamer corona is sufficient only to make approximations as to the size of a full scale unit, for example for the treatment of a dissolver off-gas (DOG) stream.

There are two options as discussed in 4.2.3 above; one using a two stage system utilising a first stage continuously irrigated discharge, followed by a second stage streamer discharge. The use of a streamer corona as a second stage offers advantages over a glow discharge with respect to reduced sensitivity to moisture and lower power consumption. This means a simple, coarse packed demister device would probably suffice between stages, compared with the high efficiency dehumidification required prior to a glow discharge second stage.

The first stage irrigated glow discharge would remove bulk NO_x from around 1000 vpm (from a modified primary acid scrubber) to approximately 100 vpm with little iodine removal. The iodine would be removed in the dry streamer second stage to DFs of around 10^4 or greater. The iodine deposited in the dry streamer unit would be removed by intermittent washing in the same way proposed for the two stage glow discharge system (1).

For a gas throughput of 100 m^3 h^{-1} comprising 1000 vpm NO_x, and 10 vpm I_2 ($\sim 10\%$ as organic iodine), the first stage unit would require approximately 300-500, 50 mm diameter, 1 m long tubes occupying a volume of 1 m^3. The power unit would be around 25 kW. The second stage streamer unit would be a similar size, but would require a much smaller power unit of around 2kW with a minimum pulse repetition frequency of 25Hz.

The alternative approach of using a single stage pulsed irrigated system provides a simpler plant, but there is still uncertainty with regard to the maximum iodine DFs which are likely to be achievable. Based on the best data achieved in the single tube experiments, a corona unit consisting of approximately 500, 37.5 or 50 mm diameter tubes would provide a DF of around 30, ($\sim 97\%$ removal). The power unit would be somewhere between 10 and 30 kw.

Other aspects of the process which require further consideration include the behaviour of other species which occur in dissolver off-gases, including CO, CO_2, volatile ruthenium and particulates and aerosols. Some experiments have been carried out with CO, and no conversion to CO_2 has been observed consistent with thermodynamic considerations (6).

The behaviour of ruthenium, particulates and aerosols has not been investigated, but certainly effective particulate and aerosol removal would be anticipated, similar to conventional Electrostatic Precipitators (ESPs).

6.   Plant Hardware

Comparisons of the anticipated process requirements with available industrial Electrostatic Precipitation technology confirm that much of the plant requirement is available on an industrial scale. There are significant differences in the design of the electrode system, but essentially the technology is very similar, even the use of continuously and intermittently irrigated electrodes. Further consideration should be given as to whether wire-in-tube is the most favourable geometry for a larger system. A wire-in-plate geometry may be more economic from engineering/construction considerations, providing the performance is not compromised.

On the pulsed power supply side, there is still some development work required to demonstrate the reliability and robustness of a larger power unit capable of generating the necessary pulsing characteristics.

7.   Conclusions

The use of corona discharge for the treatment of nuclear fuel reprocessing plant off-gas offers significant potential for the economic reduction of gaseous emissions, particularly iodine - 129, which may be present as both molecular and organic iodine. The corona unit is very compact compared with conventional wet scrubbing units.

Two process options are possible, one using a two stage system utilising a wet continuously irrigated first stage glow discharge for bulk NO_x removal, followed by a second stage dry pulsed streamer corona for high efficiency iodine removal. The use of a streamer corona as a second stage overcomes problems previously encountered with moisture interference and permits only crude water removal between stages.

The alternative single stage continuously irrigated (wet) pulsed corona may offer a more compact and cheaper option, however, so far the iodine DFs have not been as high as for the two stage approach, i.e. 30 compared with 10^4.

8.   Acknowledgements

9.   References

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DISCUSSION

GOOSSENS: May I invite you to comment first, on the iodine compound formed by corona discharge, and secondly, on the secondary waste treatment.

BARLOW: Initial studies on the corona reactions of MeI are preliminary in nature, but it has been suggested that I_2O_5 and I_4O_9 are formed dependent on polarity of corona. Being that both are water soluble, secondary waste treatment would need to take account of the oxidizing properties of the compounds. Treatment of the liquid to precipitate the iodine, followed by encapsulation, might be a suitable way forward.

HYDER: What is the temperature of the effluent gas from the corona discharge?

BARLOW: On this particular system, it has been very close to room temperature.

JUBIN: How does this corona discharge process compare with solid sorbent methods for iodine removal?

BARLOW: To my knowledge, a comparison has not been made as the corona work is considered a novel system to demonstrate the feasibility of MeI/NO removal in a single compact unit.

VENDEL: Do you think all the particles created in the corona discharge are trapped in the scrubber?

BARLOW: Deposits of oxides of iodine are retained on the electrode surfaces, which necessitates either off-line periodic cleaning or continuous irrigation to maintain efficient operation.

VENDEL: Do you think it is possible that new particles could be created downstream of the scrubber?

BARLOW: To my knowledge the formation of particles after the unit has not been investigated and the possibility of aerosol formation would need to be examined. In the presentation, I have a slide showing schematic treatment, including post corona HEPA filters, to prevent any particulates being released. Further work would need to be carried out to investigate the post-corona formation of particulates/droplets which might occur, possibly by gas phase reaction or condensation.

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REMOVAL OF IODINE-129 FROM DISSOLVER OFF-GAS OF REPROCESSING PLANT BY SILVER IMPREGNATED ADSORBENTS

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Abstract

The removal characteristics of I-129 by the silver impregnated adsorbents were evaluated using the dissolver off-gas sampling line of Karlsruhe Reprocessing Plant (WAK) in Germany. This study aimed at confirming the applicability of the new adsorbent, AgA (silver alumina which contains 24% Ag), to the actual reprocessing plant.

Distribution profile of iodine in a test adsorption column and decontamination factors (DFs) at each bed were determined by measuring I-129 activities at each bed after certain operating times. The DF values of over 10^3 were obtained at 10cm bed depths with various experimental conditions.

It was revealed that AgA adsorbent could get high DF value for iodine with NOx and water vapor of the actual reprocessing plant.

I. Introduction

Iodine is released to a great extent during dissolution of spent nuclear fuel and follows the dissolver off-gas. As the permitted amount of iodine-129 release to the environment is rather small, the removal of iodine is considered from the off-gas stream in reprocessing plant.

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Two kinds of methods have already been developed for iodine removal, that is dissolution of iodine in an alkaline solution (wet method) and adsorption of iodine on a material containing silver (dry method). Both methods have quite a good ability to remove iodine. For the dry method, the adsorbent AgS (silver silica gel which contains 12 % Ag) has been applied in German facilities and shown the high iodine removal efficiencies.[1] On the other hand, the authors have developed silver impregnated alumina (AgA) and tested with the simulated reprocessing off-gas under various conditions in laboratories.[2] The parameters such as temperature, iodine form, iodine concentration, NOx concentration, humidity, off-gas flow rate and adsorbent thickness were changed including the predicted off-gas conditions of actual reprocessing plant.

Test results showed the high performance of AgA for the removal of not only elemental iodine but also organic iodine compounds.[3] [4] [5]

The aims of this study are to determine the retention capability of AgA under exposure of actual dissolution off-gas at the WAK plant during extended time and to compare its ability to that of AgS.

II. Experimental

The specification of silver impregnated adsorbents used in this work is shown in Table 1. The silver silica gel, AgS, is called as AC 6120 in this work. The silver alumina, AgA, has larger pore size, higher silver content, higher bulk density, and smaller surface area compared with AC 6120.

Investigations of iodine adsorption materials in the WAK dissolver off-gas were carried out during two campaigns at the WAK plant. Campaign I was carried out with MZFR and VAK fuels and campaign II with MZFR and KKS fuels. Table 2 shows the summarization of each campaign.Total amounts of spent nuclear fuels dissolved and iodine-129 introduced to the test section were 8.7t and 3.5GBq, respectively.

The constitutions of dissolver off-gas treatment system in WAK plant and the test section (sampling cabinet) are schematically shown in Figures 1 and 2, respectively. Sample gases are drawn from the dissolver off-gas line after nitrogen oxides scrubbers and HEPA filters as shown in Figure 1 and then treated with HEPA filter, cooler and demister as shown in Figure 2. This system enables the sufficient removal of NOx, particles and water vapor from the off-gas. Test column of 8 adsorption beds was kept at constant temperature of 140°C. Each bed had 2.5cm thickness and 2.5cm diameter. In order to quantify the slope of separation profile, the first 2.5cm bed is subdivided into two 1.25cm parts. Flow rate of the sampled off-gas was 353 ℓ／h (140°C, ～956mbar) and linear flow rate was 21cm／s. The material weight of each 2.5cm bed was 17g AgA or 8.1g AC 6120／12 % Ag.

The calculated iodine concentration in the sample off-gas during dissolution is in the order of about 0.1-10ppm and the caluated average iodine concentration in the sample off-gas is about 0.1ppm. A maximum of 5 vol% and 3.5 vol% in average of NOx are contained in the dissolver off-gas in addition to air. The ratio $NO_2／NO$ is about 3／1.

Furthermore, the dissolver off-gas containes about 3-4 vol% of water vapor.

Iodine-129 adsorbed in the beds was measured by the Ge detector. The detection limit for 40keV γ-ray of ^{129}I in a bed was 1 ± 1 Bq／g.

III. Results and Discussion

Decontamination factor for AgA

The iodine retention capacity of a bed is determined from the ^{129}I activity measured after the bed, and the sum of the ^{129}I activities of all beds. It is expressed by means

Table 1   The Specification of silver impregnated adsobents

	AgA	AgS (AC 6120)
Carrier	Activated Alumina	Silica Gel
Pore Size of Carrier (Å)	600	100
Specification Surface Area (m^2/g)	~10	~70
Silver Content (wt%)	24	12
Adsorbent Size (mm)	1 − 2	1 − 2
Bulk Density (g/cm^3)	~1.5	~0.7

of the decontamination factor DF. For a series of beds (1, 2, 3···, n) in an adsorption tube containing a total amount of n beds, the DF for bed X is defined as follows :

$$DFx = \cfrac{\sum\limits_{1}^{n} {}^{129}I \text{ activities}}{\sum\limits_{X}^{n} {}^{129}I \text{ activities}} \qquad (1)$$

The DF and separation profile of the adsorption material AgA were determined at 140°C and three residence time of the adsorption tube (35, 60 and 95 days). Figure 3 shows the dependence of the decontamination factor and the penetration against the thickness of the bed at campaign I. The penetration P is calculated by equation (2).

$$P (\%) = 1/DF \times 100 \qquad (2)$$

The single values of each curve are the mean value of two parallel measurements. The deviation of the actually measured values from the mean value are indicated.

Table 2 Conditions for dissolved fuels for campaigns I and II

Fuels	Campaign I		Campaign II		
	MZFR*1)	VAK*2)	LWR*3)	KKS*4)	MZFR
Burn Up (MWD/t)	≦ 15,000	≦ 12,000	≦ 12,000	≦ 40,000	≦ 13,000
Dissolved Amount (t)	1.3	2.1	0.9	1.0	3.4
Iodine Content (Bq) (Calculated Value)	12.4×10^8		22.8×10^8		
Maximum Operation (days)	95		118		

* 1) Multi - Purpose Research Reactor
* 2) Experimental Reactor Kahl
* 3) Light Water Reactor
* 4) Nuclear Power Reactor Stade
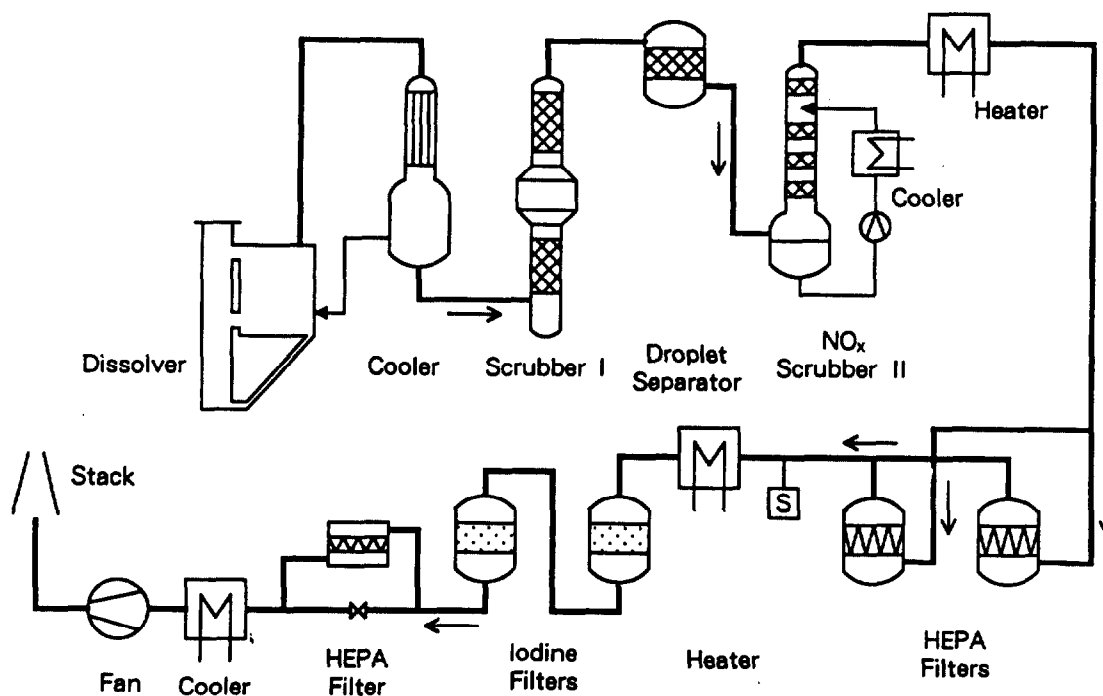
Figure 1 Dissolver off-gas treatment system in the WAK and iodine sampling station S
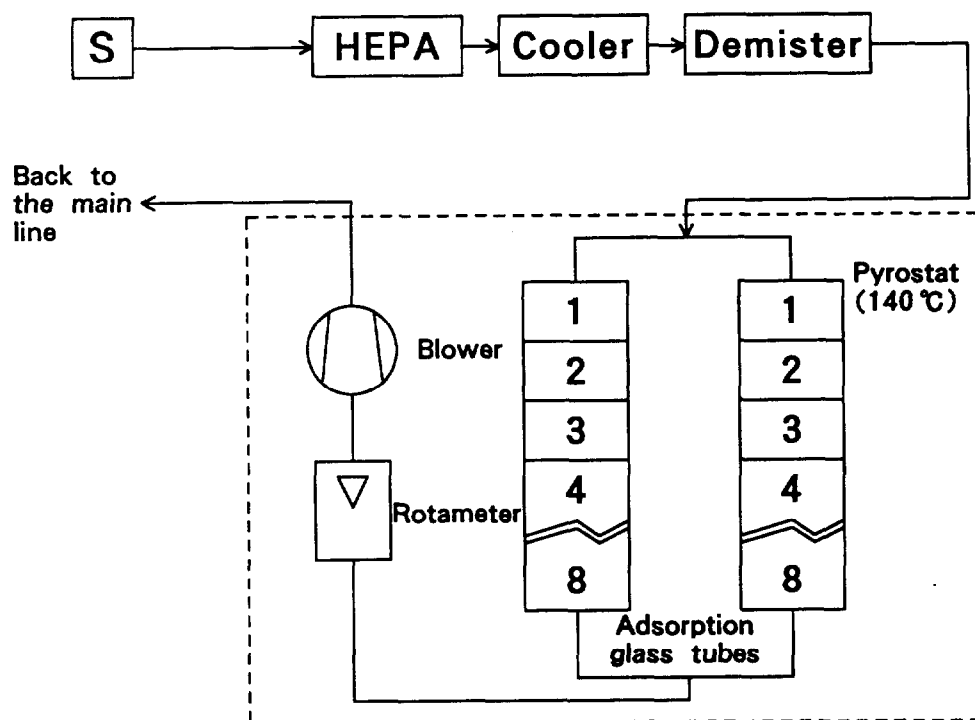
Figure 2 Flow diagram for sampling cabinet

For a bed thickness of 5.0cm, the average DF is 600 to 800 for 35 to 95 days operations. At a bed thickness of 10.0cm, the DF for the three experiments lies between 10^4 and 2×10^4.

Figure 4 shows the dependence of the DF and the penetration on the bed depth during campaign II. For a bed of 5.0cm depth, the average DF is about 2.0×10^5 after 76 days and about 2.3×10^5 after 118 days. For a bed of 10.0cm depth, the average DF lies within the value of 10^4 to 2×10^4 for both periods.

The data shown in Figures 3 and 4 demonstrate not only the high DF value for AgA but also stable DF value among different operation times. The DF values of each bed show no decrease with increasing the operation times.

Thus the adsorbent AgA was revealed to get the high and stable performance within 3 months test in the WAK plant.

Comparison of DF between AgA and AgS with the plant and laboratory tests

Figure 5 shows a comparison of decontamination factors of AgA and AC6120 obtained for each campaign. The content of Ag for AC 6120 was up to 20% for campaign II. The DF of AgA at a 10cm thick bed is higher by about an order and half an order of magnitude than the DF of AC 6120/12% Ag and AC 6120/20% Ag, respectively. These results can be explained by the dependency of DF on the silver content of the adsorbents.

Figure 6 shows the results of a laboratory test using the same adsorbents AgA and AC 6120 as the WAK plant test. [2] Iodine used in the laboratory test was elemental iodine I_2 and its concentration was 4 orders higher than that of plant test. Such high iodine concentration (750 ppm) caused the short operation times (10 hours for AgA and 4 hours for AC 6120) and the saturated iodine adsorption at several top beds (1-3 beds for AgA and 1-4 beds for AC 6120). The depths of each bed were 1 cm. Here unsaturated part of Figure 6 is discussed.

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(a) 35days Operation   (b) 60days Operation   (c) 95days Operation
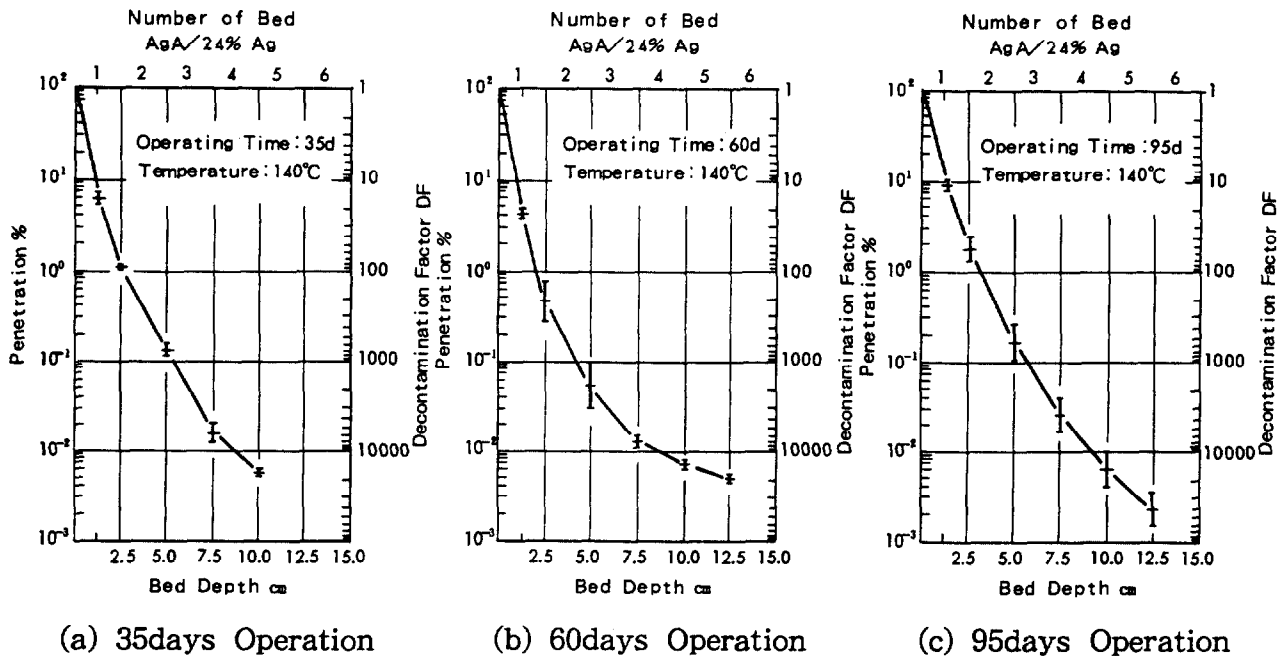
Figure 3   Decontamination factor as a function of bed depth for AgA (campaign I)

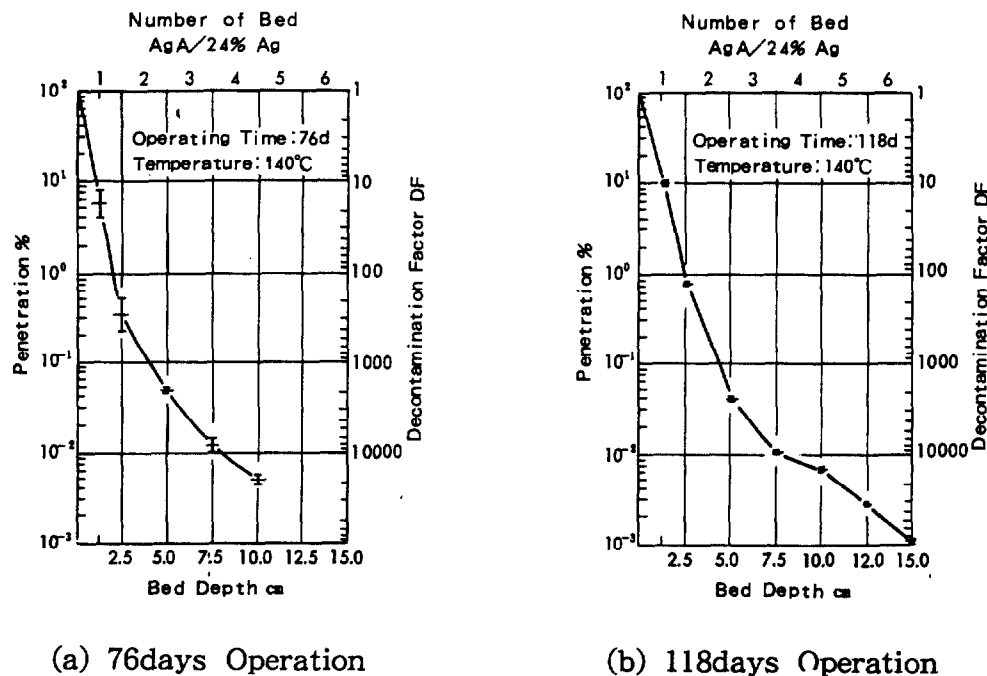(a) 76days Operation   (b) 118days Operation

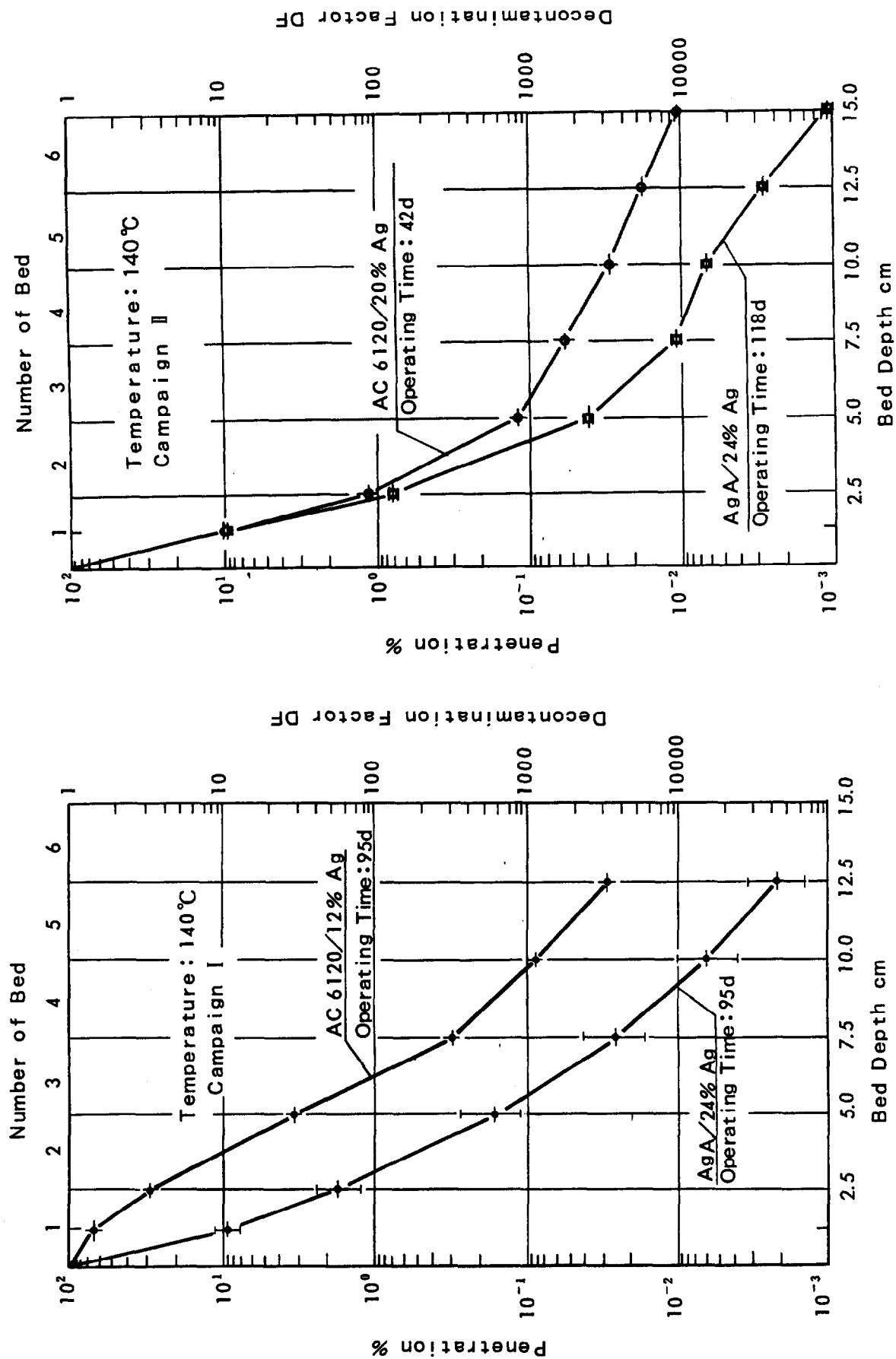Figure 4   Decontamination factor as a function of bed depth for AgA (campaign II)

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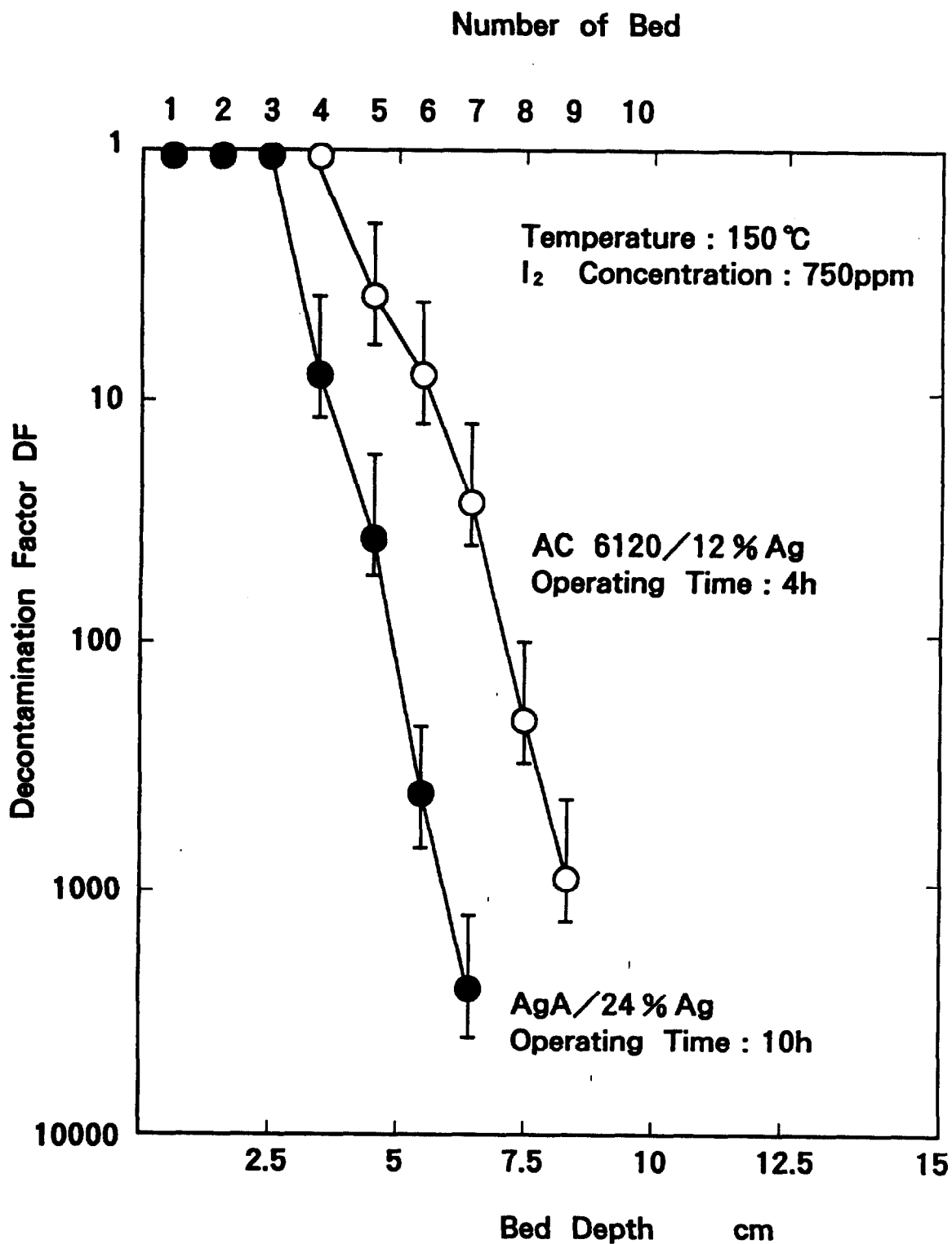Figure 5 Comparison of decontamination factor of AgA and AC 6120

Figure 6 Comparison of decontamination factor of AgA and AC 6120
obtained in a laboratory test

Comparison of Figures 5 and 6 indicates the different tendency of DF dependence on bed depth with each other. For AgA initial slopes of both curves are the same but the slope of downward beds (bed numbers 3 and 4) in Figure 5 decreases, while no slope decrease is observed in Figure 6. The same tendency for AgA is also observed for AC 6120 as shown in Figures 5 and 6. The decrease of the absolute slope value at downward beds means the deterioration of retention ability for iodine in those beds. This may be caused by the difference in iodine forms and/or iodine concentrations between the top and the downward beds.

In WAK plant, iodine species other than elemental iodine should exist and behave in a different way from elemental iodine. Iodine concentrations at the downward beds should be lower than those at the top beds because iodine in the off-gas stream was adsorbed mostly at the top beds.

The DF data like Figures 3 and 4 exist in other papers, [6] and relatively low DF data is obtained in the vessel off-gas system of WAK plant where the ratio of organic iodine forms is higher and iodine concentration is lower than in the dissolver off-gas system. [7] [8]

But on the other hand several data were obtained for the DF dependency on iodine concentration, which shows the constant DF values among different iodine concentrations. [9] Dissolver off-gas should include the iodine species which are difficult to remove and in the vessel off-gas their content is larger than in the dissolver off-gas. [1]

Thus the dependency obtained here may be attributable to the existence in the off-gas of the composition hardly removed by the adsorbent and the increase of difficult-to-remove iodine speices ratio at the downward beds and not to the iodine concentration itself.

Anyway, from the slope of DF versus bed depth, an average DF value of 10 can be obtained in the bed depths of about 1 cm for AgA and about 2 cm for AC 6120 12% Ag.

Futhermore, the DF at 10cm for each operation period obtained are more than 10^3 and revealed that AgA adsorbent could get high DF values for iodine with NOx and water vapor of the actual reprocessing plant.

IV. Conclusions

The following results were obtained from the test results of actual dissolution off-gas at the WAK Plant.

(1) For a bed of 10 cm depth, the DF value of more than 10^3 was obtained for AgA.

(2) AgA shows relatively high DF values compared with AgS. This was explained by the higher silver content of AgA than that of AgS.

(3) The data obtained from the actual dissolution off-gas of a reprocessing plant with NOx and water vapor showed that AgA could get high DF values.

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DISCUSSION

MOELLER: In your paper you say that you achieve a decontamination factor greater than 10^3 for a 10 centimeter bed depth. As I look at your curves, it appears that you achieved more than 10^4. Why did you not say greater than 10^4?

KONDO: As the needed DF values in our assessment were no more than 10^3, we summarized the conclusion by citing that value.

GOOSSENS: I would like to discuss a policy question on I-129. From a global point of view, there is no need to trap I-129. Recent assessments indicate that in practice the trapped I-129 cannot be isolated from the environment longer than 10^5 years, or fraction of the half-life time of I-129. Thus, there is no a need for trapping I-129. Why have we been making all this effort?

HERRMANN: The question must be addressed to the licensing authorities who have fixed the low admissible values for Iodine 129 emission.

SCHOLTEN: In answer to Goossens' question, I can tell you that in the Netherlands an individual mortality risk of 10^{-8} per year is considered negligible.

EFFECTS OF THE PARTICLE PENETRATION INSIDE THE FILTER MEDIUM ON THE HEPA FILTER PRESSURE DROP

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Abstract

Filter pressure drop modelisation as a function of the deposited aerosols mass requires to know the penetration profile of the particles inside the filter medium and to take into account the evolution of the internal structure of the filter.

These two parameters can be determined by combining two models:

- a filter efficiency modelisation to calculate the mass deposited in the filter,
- a filter pressure drop model to evaluate the change of the internal filter structure.

Different hypothesis are made concerning the shape of the penetration profile. It seems, according to experimental results, that to take into account an unlike evolution of the internal filter structure according to the depth inside the medium leads to a satisfactory description of the pressure drop.

I. Introduction

High Efficiency Particulate Air (HEPA) filters are successfully used in a multitude of applications such as nuclear, pharmaceutical or semiconductor industries where the dust concentrations are very low. HEPA filters are now used in industrial processes that involve higher dust concentrations; under such conditions a rapid increase in pressure drop occurs to a point where the filter replacement is necessary.

So to extend the working life of a filter becomes a challenge of considerable practical and economic importance.

Removal efficiency and pressure drop of clean HEPA filters have been widely studied but only few publications are devoted to the study of the filtration efficiency evolution during the service time as well as the effects of aerosol penetration inside the filter on the pressure drop increase.

This paper is concerned by the filter pressure drop modelisation according to the mass of aerosol deposited in the filter. First we present the experimental set-up and results on the change in pressure drop for two kinds of filter as a function of different aerosol sizes and various filtration velocities. Then two models, which take into account the penetration profile of aerosol inside the filter medium, are proposed and compared to experimental results.

II. Experimental

II.1. Test facility

The test rig is shown on Figure 1.

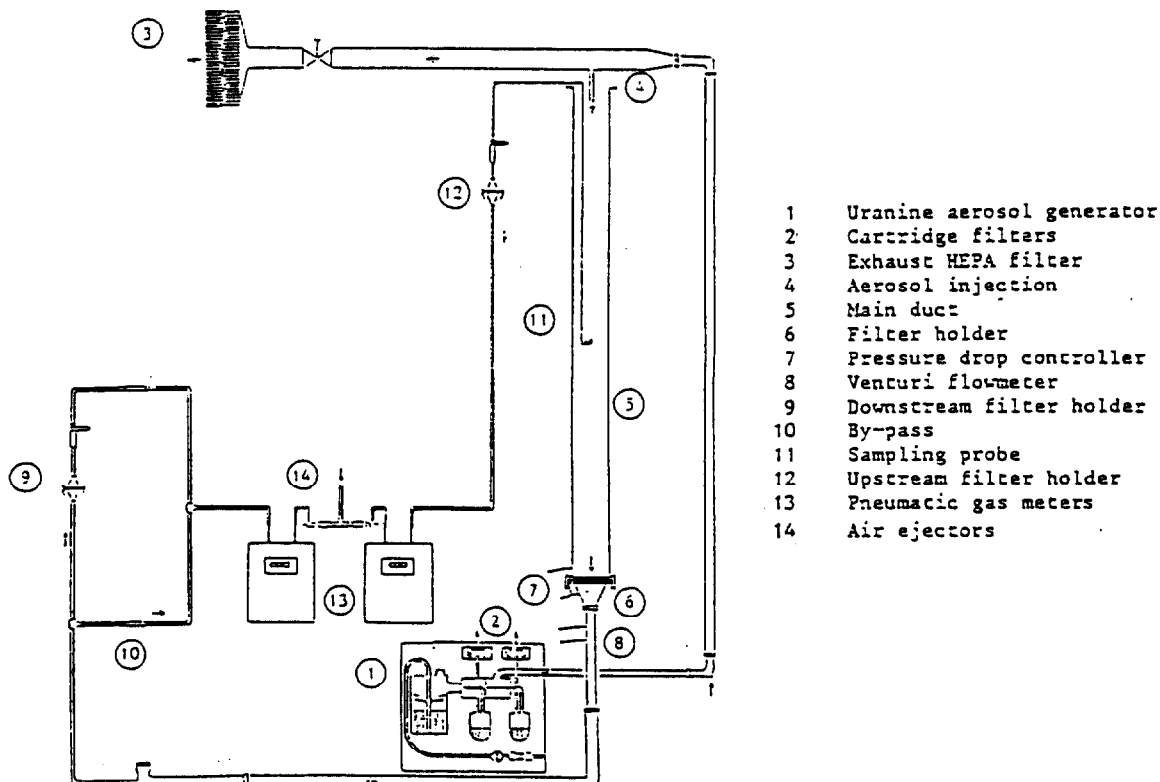|	
1	Uranine aerosol generator
2	Cartridge filters
3	Exhaust HEPA filter
4	Aerosol injection
5	Main duct
6	Filter holder
7	Pressure drop controller
8	Venturi flowmeter
9	Downstream filter holder
10	By-pass
11	Sampling probe
12	Upstream filter holder
13	Pneumatic gas meters
14	Air ejectors

Figure 1 - Scheme of the test rig

The different test aerosols are injected in the main duct (5) where the plane filters are installed to be tested (6). The air flow rate is kept constant during the test via a sonic nozzle (14) and the filter pressure drop is continuously monitored (7).

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Upstream (12) and downstream (9) aerosol concentrations are measured to evaluate the filter removal efficiency.

II.2. Test aerosols

Three uranine aerosols (Soda fluorescein) with different diameters have been used.

One is produced by a standardised NFX 44-011 generator (Sg) filled with a 10 g/ℓ soda fluorescein solution. The two others are produced from a 100 g/ℓ solution; the first one is generated by a standardised generator (Sg) and the second one is produced by a modified standardised generator (Sgm).

The aerosol characteristics have been measured with a Differential Mobility Particle Sizer (DMPS TSI 3071) associated with a Condensation Nucleus Counter (CNC TSI 3020). These aerosols follow log-normal size distributions whose mean diameters (Dp) and geometric standard deviation (σg) are indicated in Table 1

Table 1 - Aerosol characteristics

Uranine concentration (g/ℓ)	Generator	Dp (μm)	σg
10	Sg	0.15	1.6
100	Sg	0.25	2.0
100	Sgm	0.41	2.0

II.3. Filter media

Two types of filter media made of glass fibers have been used and their characteristics are given in Table 2.

Table 2 - Filter media characteristics

Reference	D306	B345
Supplier	B. DUMAS	B. DUMAS
Shape	Plane	Plane
Diameter	10.8 cm	10.8 cm
Thickness	370 μm	370 μm

GSM	87.4 g/m²	81 g/m²
Packing density	9.45 10⁻²	8.2 10⁻²
Fiber diameter*	1.4 μm	3.1 μm

* Fiber diameters have been calculated from the Davies'equation [1] :

$$dP = 16 \; \mu \; V_f \; Z \; \frac{\alpha^{3/2}}{R_f^2}$$

Where :

dP :	filter pressure drop (Pa)
μ :	dynamic viscosity (Pa.s)
V_f :	filtration velocity (m.s⁻¹)
Z :	filter thickness (m)
α :	packing density
R_f :	fiber radius (m)

III. Experimental results

III.1. Experimental conditions

Investigations in pressure drop evolution according to the mass deposited in the filter have been carried out at various filtration velocities, particle sizes and type of filter. The main experimental conditions are given in Table 3.

Table 3 - Experimental conditions

	Aerosol diameter Dp (μm)	Filtration velocity V_f (cm.s⁻¹)
D306	0.15	5
		10
		20
	0.26	3
		6
B345	0.15	5.3
	0.25	6
		3
	0.41	5.3

IV. Discussion of results

A previous study [2] has shown, using a peeling method, that the Bi nan's model [3] can be modified by taking into account an exponential profile for the aerosol penetration inside the filter medium.

Experimental results have revealed a satisfactory correlation with this modified model so called "M.L.V.". The limitation in this approach is the experimental determination of the penetration factor (K).

IV.1 - Penetration factor modelisation

To evaluate the K factor two models are combined:

-the "M.L.V." pressure drop model to determine the filter structure evolution according to the surface mass (M/S) of filtered aerosol,

-an efficiency model [4] to evaluate the penetration factor K as a function of the filter characteristics.

From the initial filter characteristics (α_0, dP_0 and R_{f0}) the efficiency model is used to calculate the penetration factor Ko. This value of Ko is introduced in the "M.L.V." model to calculate the pressure drop dP_1 associated with the first step of aerosol mass deposited per unit of aera (the surface mass step is equal to 0.25 g/m^2). Then the dP_1 value is used to determine the new characteristics of the filter from Davies'equation as:

$$\alpha_1 = \alpha_0 + \frac{M}{S} \frac{1}{\rho o \, Z}$$

$$R_{f1} = \left[\frac{16 \, \mu \, V_f \, Z \, \alpha_1^{3/2}}{dP_1} \right]^{1/2}$$

where ρo is the particle density.

With these new characteristics (α_1, dP_1 and R_{f1}) a K_1 value is calculated from the efficiency model.

This procedure is repeated by increasing step by step the surface mass until its maximal value.

For D306 filter experimental results obtained in the conditions described in Table 3 are compared to the model.

Figures 6, 7 and 8 show the change in pressure drop according to the surface mass at three filtration velocities for 0.15 μm particle diameter.
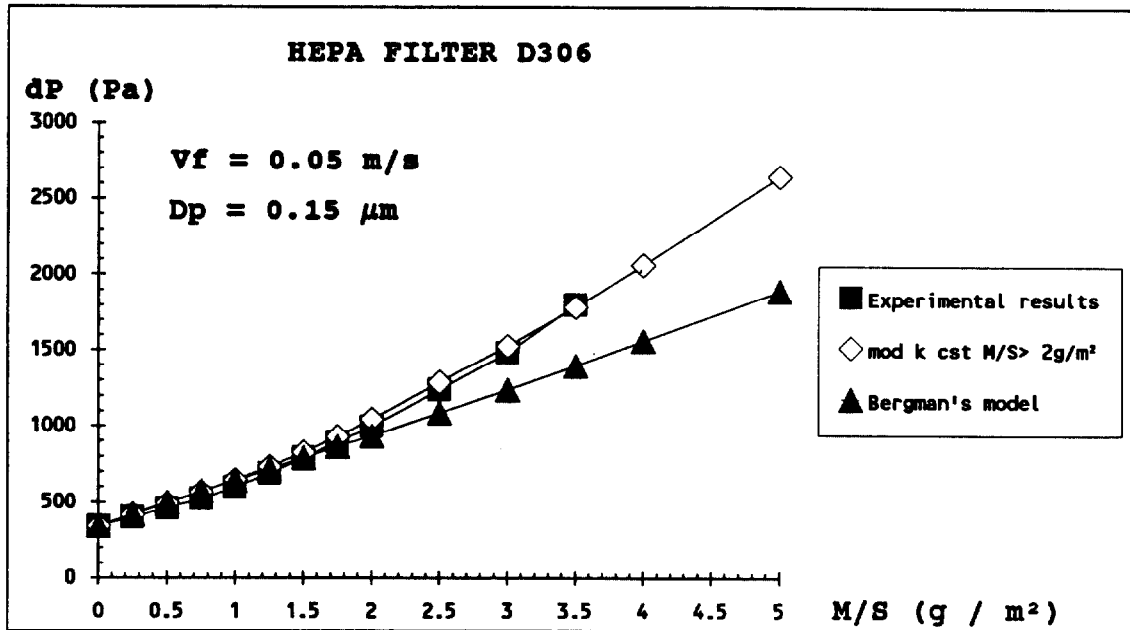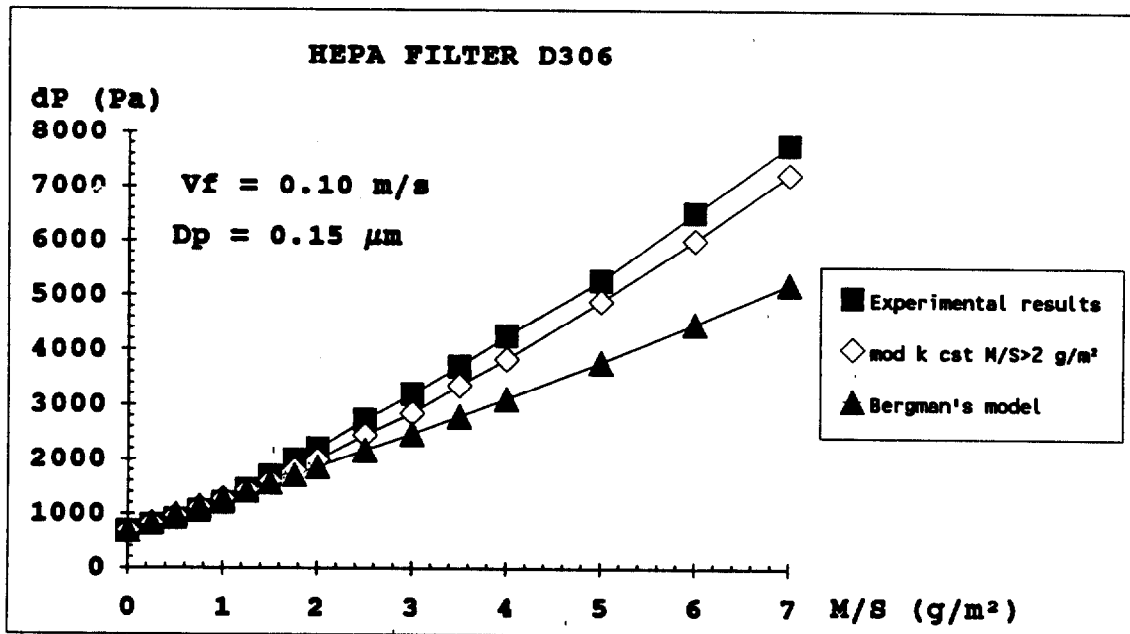

Figure 6 : Comparison of pressure drop models with experimental results (Vf = 0.05 m/s)

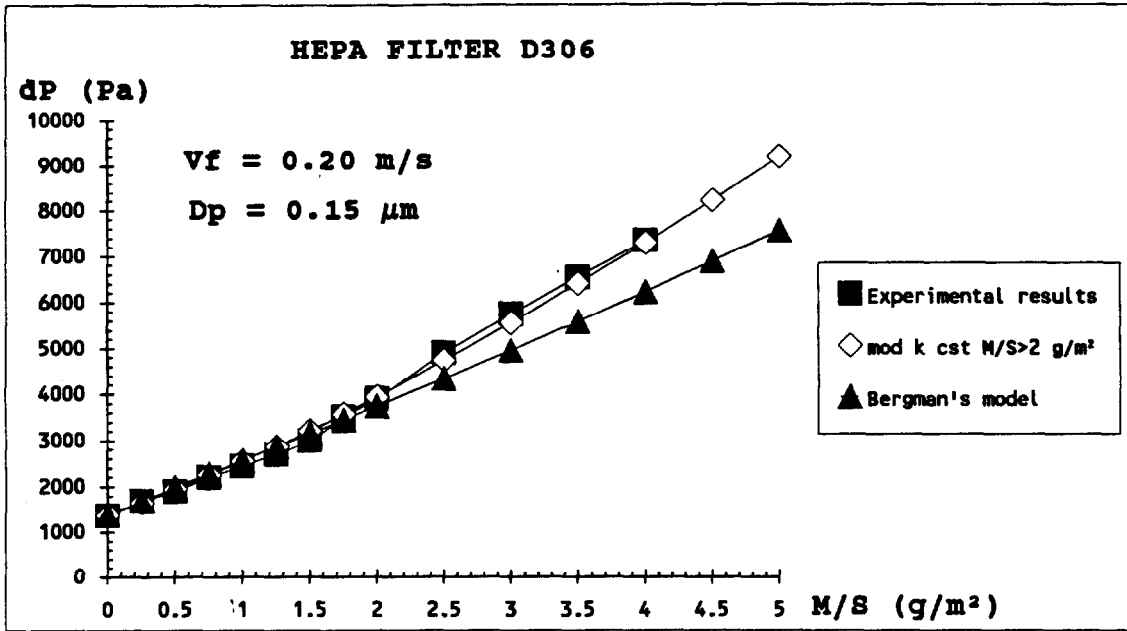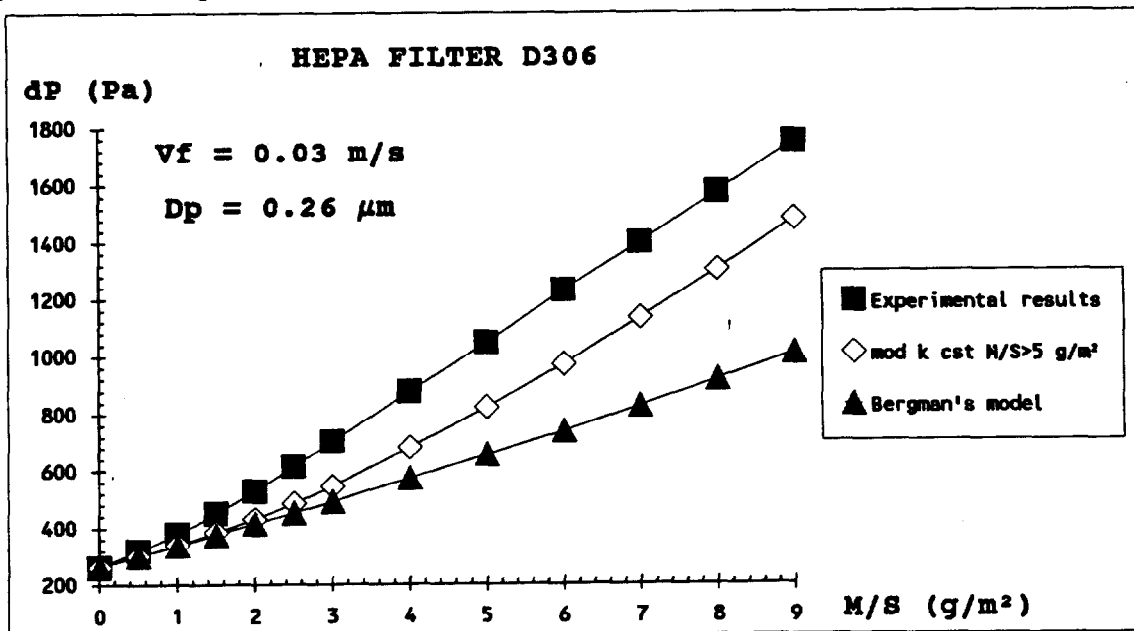Figure 7 : Comparison of pressure drop models with experimental results (Vf = 0.10 m/s)

HEPA FILTER D306

Figure 8 : Comparison of pressure drop models with experimental results
(Vf : 0.20 m/s)

A good agreement between experiences and model is noted if we consider that the filter characteristics remain constant when the surface mass has reached 2 or 2.5 g/m^2. These maximal values have been determined by a peeling method [2]. On these figures the Bergman's model results are also indicated.

Figures 9 and 10 present results for 0,26 μm particle diameter at two filtration velocities.

HEPA FILTER D306

Figure 9 : Comparison of pressure drop models with experimental results
(Vf = 0.03 m/s)
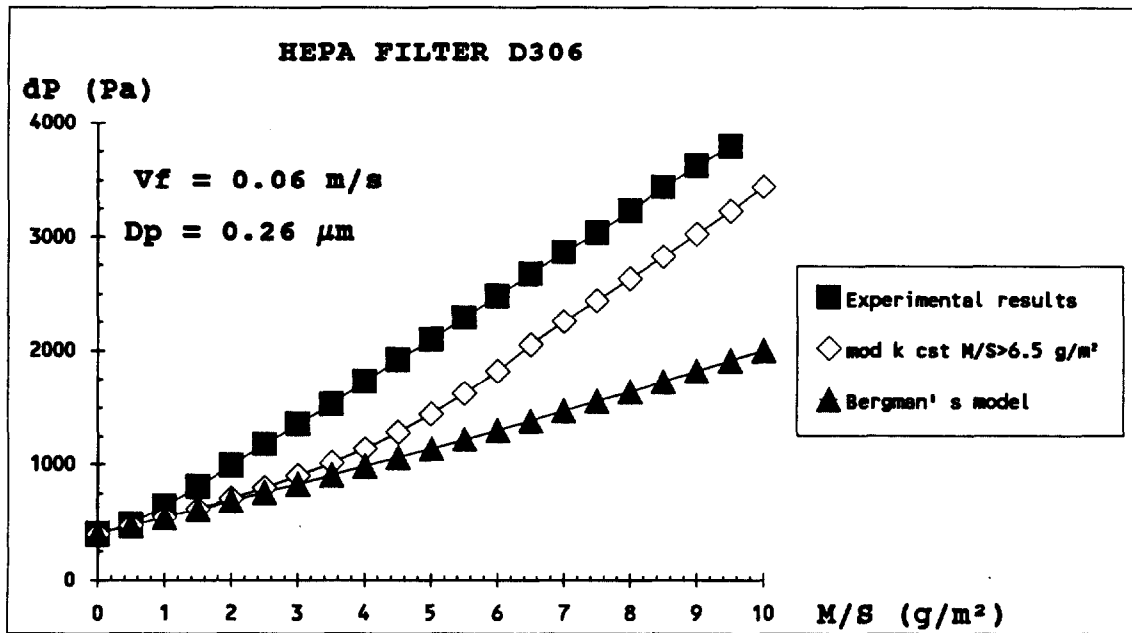
Figure 10 : Comparison of pressure drop models with experimental results (Vf = 0.06 m/s)

Model and experimental results are close together if we consider a surface mass around 5 or 6.5 g/m².

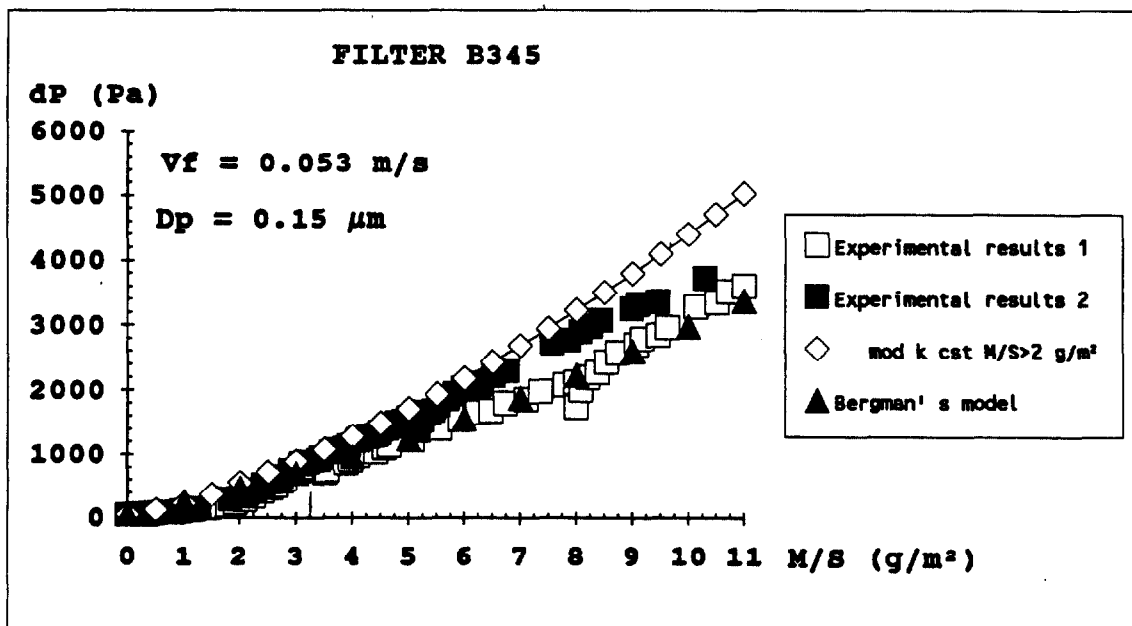For B 345 filter the change in pressure drop is indicated on Figures 11, 12 and 13 for three particle diameters.

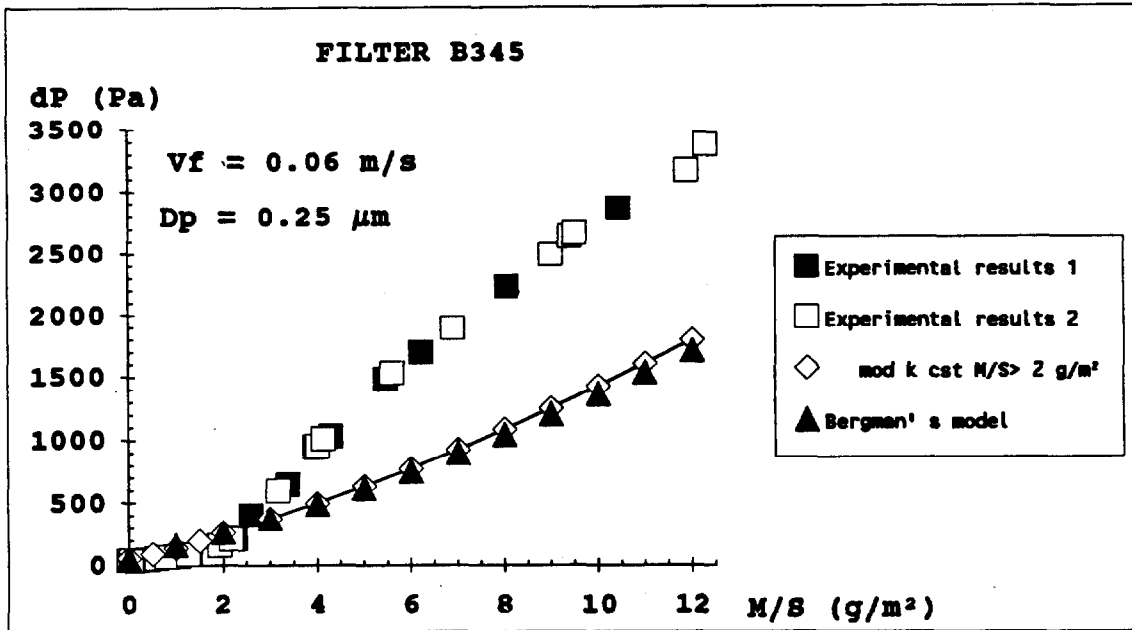Figure 11 : Comparison of pressure drop models with experimental results (Dp = 0.15 μm)

FILTER B345

dP (Pa)

Vf = 0.06 m/s
Dp = 0.25 μm

Legend:
- ■ Experimental results 1
- ☐ Experimental results 2
- ◇ mod k cst M/S> 2 g/m²
- ▲ Bergman's model

M/S (g/m²)

Figure 12 : Comparison of pressure models with experimental results
(Dp = 0.25 μm)

FILTER B345

dP (Pa)

Vf = 0.053 m/s
Dp = 0.41μm

Legend:
- ☐ Experimental results 1
- ■ Experimental results 2
- ◆ Experimental results 3
- ◇ mod k cst M/S>3 g/m²
- ▲ Bergman's model

M/S (g/m²)
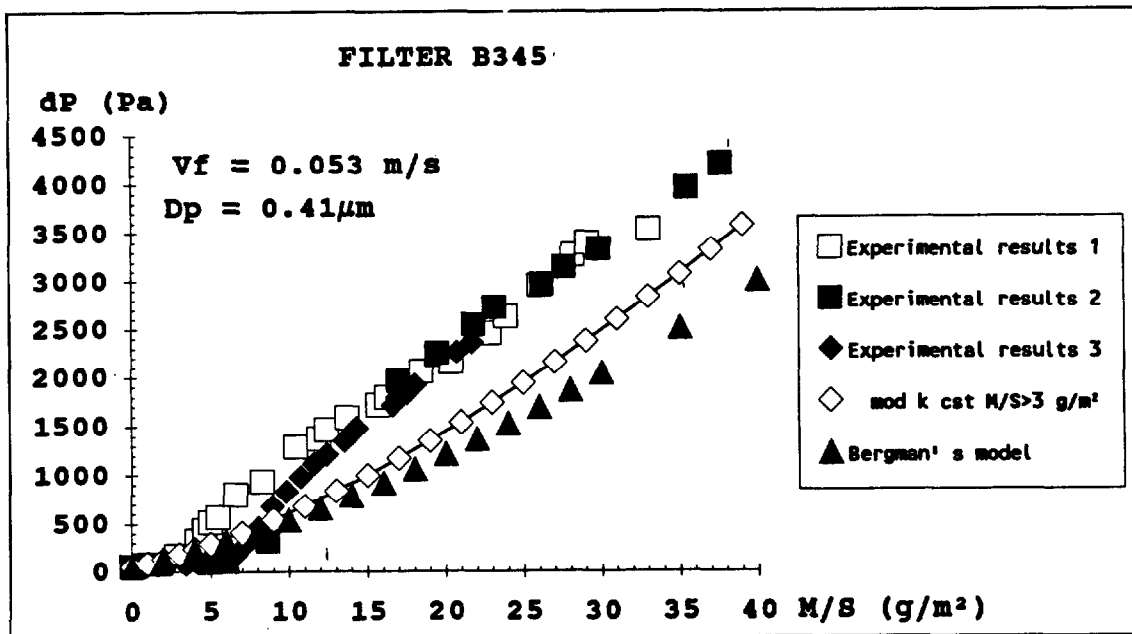
Figure 13 : Comparison of pressure drop models with experimental results
(Dp = 0.41 μm)

For 0.15 μm particle diameter, the penetration factor and the Bergman models give a good estimate of the pressure drop, but for 0.25 μm and 0.41 μm particle diameters these two models give a 50 % underestimate pressure drop compared to experimental results.

This discrepancy can be attributed to a cake formation on the upper face of the filter which have a large contribution in the increase of pressure drop.

In conclusion, models in which only the penetrated particle mass is considered to modify the internal filter structure, give good results for the most penetrating particles (0.15 μm) but underestimate the pressure drop for the less penetrating aerosols.

To only take into account the penetration profile without to consider the cake formation seems to be insufficient to describe the change in pressure drop during clogging.

IV.2. Profile penetration modelisation

In the previous approach (IV-1), the filter characteristics change for the whole filter. In this new model [5] we consider the filter as an assemblage of n filters of Z/n thickness where the each thickness characteristics are modified according to the filtration time and the mass deposited on each elementary filter thickness.

To evaluate the filter characteristics and pressure drop evolution, two models have been used:

-the Bergman's model to determine the pressure drop of each elementary thickness due to the mass deposited. The pressure drop value allows to calculate, from the Davies'equation, the new fiber diameter of each thickness,

-an efficiency model to calculate the mass deposited on each thickness.

As in the previous approach, we use an iterative procedure to calculate the evolution of each elementary filter thickness characteristics and to determine the filter pressure drop.

D306 filter results are shown on Figure 14, they are presented in term of flow resistance and they are in good correlation with this modelisation for 0.15 μm particle diameter.
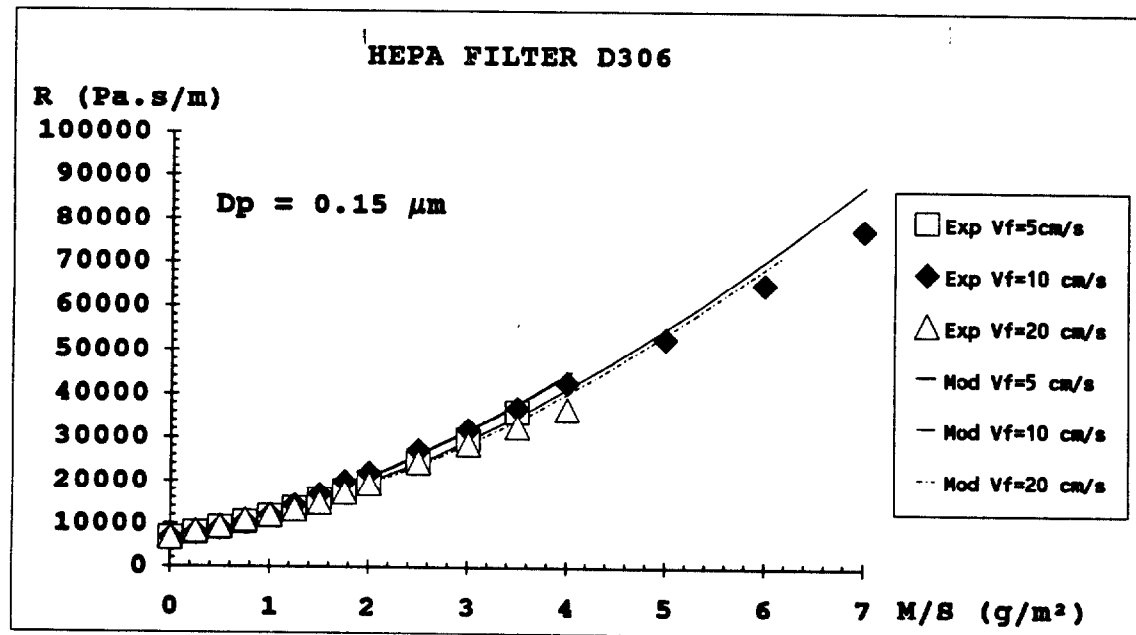

Figure 14 : Flow resistance increase with clogging

B345 filter results are presented on Figures 15, 16 and 17 for three particle diameters. We can see that this model surestimates the pressure drop for the most penetrating aerosols (0.15 μm) and is in good agreement for others particle diameters (0.25 μm and 0.41 μm). On these figures, calculatings are presented for two numbers of elementary thicknesses ; a better agreement with experimental results is noted when this number is increased.
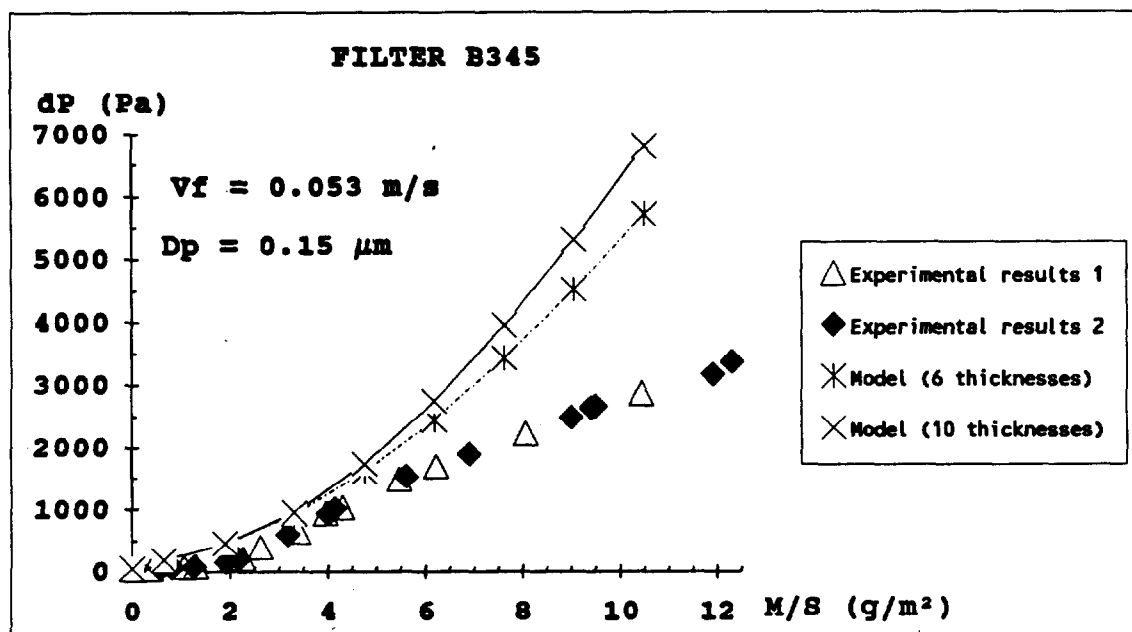
Figure 15 : Comparison of pressure drop models with experimental results (Dp = 0,15 μm)

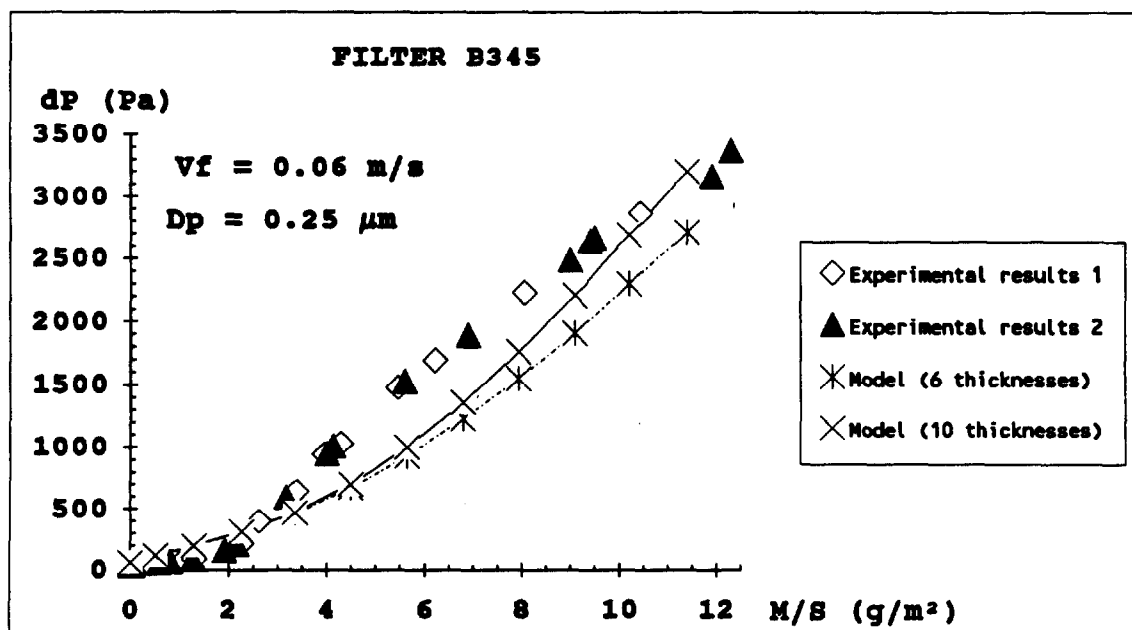Figure 16 : Comparison of pressure drop models with experimental results (Dp = 0.25 μm)

FILTER B345

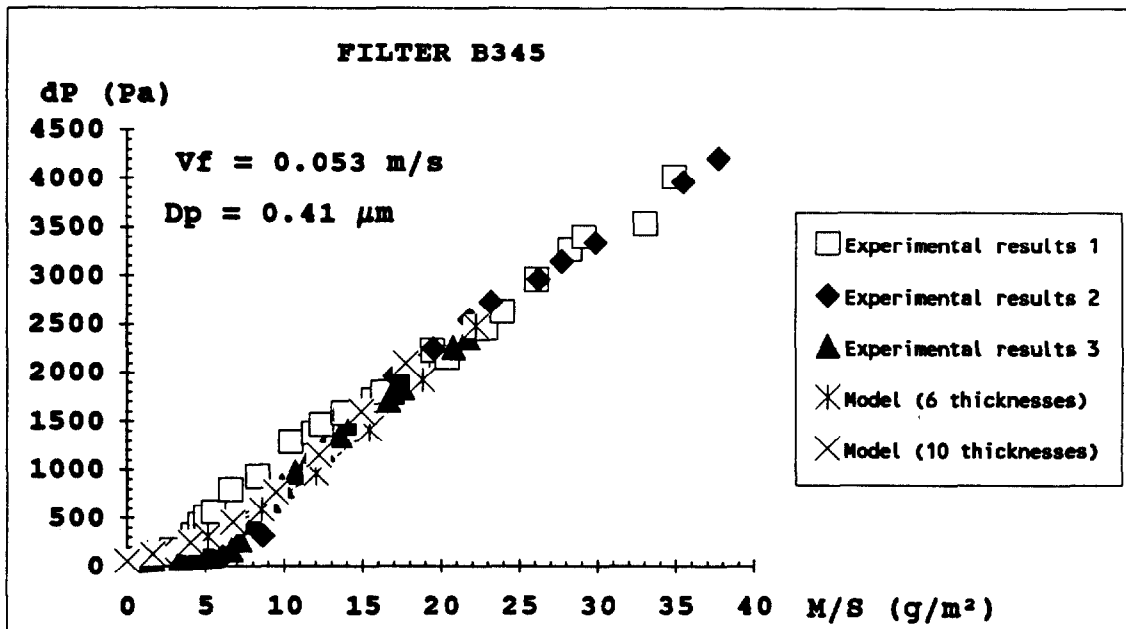Figure 17 : Comparison of pressure models with experimental results
(Dp = 0.41 µm)

V. CONCLUSION

Modelisation of the pressure drop evolution according to the surface mass of deposited aerosol requires to know the particle penetration profile inside the filter medium.

A first approach consisting to consider an exponential penetration profile and to make the internal filter structure modify in function of the mass deposited is unfit to well describe the change in pressure drop except for the most penetrating aerosols.

The last model which takes into account the influence of the mass deposited on the efficiency, pressure drop and internal characteristics of each elementary filter thickness, allows to predict the pressure drop evolution without make any hypothesis on the particle penetration profile. This model well takes into account the most important clogging of the first filter thickness and its impact on the whole filter pressure drop.

In the case of the aerosol amount is known, this model can be applied to predict with a good accuracy the service life of a filter.

Now futur studies will must extend this modelisation to the filter clogging by liquid aerosols.

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DISCUSSION

ANDREAS: How do you take into account the distribution of particles between the surface and pore structure?

VENDEL: The distribution of the particles between the surface and the pore structure is not taken into account. The particles on the surface of the filter are considered in the first stage of the filter.

DYMENT: 1) In making modelling calculations of particle deposition within the filter, have the authors taken into account the change in efficiency with particle size and its effect on a nominally exponential profile?
2) Have the authors considered the influence of "slip flow" correction on pressure drop incrementscaused by deposition of very small particles?

VENDEL: 1) Yes, we consider size distribution of the particles and efficiency is calculated as functions of this distribution.
2) We have not considered a slip flow correction for the deposition of very small particles.

TSAL: What is your experience in predicting filtration efficiency? Is it less with lower velocity or higher?

VENDEL: The theory of filtration shows that the filtration efficiency is better when the filtration velocity is decreasing.

CLOSING COMMENTS OF SESSION CO-CHAIRMAN JUBIN:

In this session there have been a number of papers that focused on iodine recovery. In many senses, this is a fairly mature technology. What we are seeing in this Conference is a focus more on the evaluation of plant performance, larger test facilities, and tests looking at the distribution of iodine through the process pathways of a nuclear fuel reprocessing plant. At the same time, we heard a paper that looks at a fairly novel technique for recovering iodine. This process is in the early stage of development and its prospects are not certain at this time. What we do hear in the area of iodine recovery and treatment will, I think, play an important role, at least for the U.S., in the future. However, this will most likely not be in the applications originally intended. I would expect to see the applications of traditional technologies for the treatment of gaseous effluents from reprocessing plants and reactors to the area of waste recovery and treatment. I would expect to hear a great deal more on this application in future conferences.

We heard one paper on tritium recovery that exemplifies a proactive response to today's ES&H requirements. As the plants throughout the world are faced with more stringent emissions requirements, new approaches will be required. In this case, the methods include improvements in capacity, efficiency, and reliability. In addition, the operations are provided assistance through the application of automation.

The final paper in this session was somewhat different from the other technologies that were discussed today. We were focusing on chemistry, this paper focused on modeling of the pressure drop across HEPA filters through the combination of filter efficiency models and changes in internal structure.

I would again like to acknowledge all of the authors for their efforts and papers. I would also like to thank the audience for their attention and contributions to the discussions. I certainly feel that all have made significant contributions to this conference.