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PROCEEDINGS of the
15th DOE NUCLEAR AIR CLEANING CONFERENCE
held in Boston, Massachusetts
7 - 10 August 1978

Sponsors: U. S. Department of Energy
The Harvard Air Cleaning Laboratory

Editor
Melvin W. First

Published
February 1979

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15th DOE NUCLEAR AIR CLEANING CONFERENCE

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15th DOE NUCLEAR AIR CLEANING CONFERENCE

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Harvard Air Cleaning Laboratory

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15th DOE NUCLEAR AIR CLEANING CONFERENCE

SESSION VII

NOBLE GAS SEPARATION

Tuesday, August 8, 1978

CHAIRMAN: D. Underhill

DYNAMIC ADSORPTION OF RADON ON ACTIVATED CARBON

K. P. Strong, D. M. Levins

DEVELOPMENT OF A CRYOGENIC KRYPTON-SEPARATION SYSTEM FOR THE OFFGAS OF REPROCESSING PLANTS

R. v. Ammon, H. G. Burkardt, E. Hutter,
G. Neffe

OPENING REMARKS OF SESSION CHAIRMAN:

At the earlier Air Cleaning Conferences the primary concern with respect to the noble gases was their release during normal and abnormal reactor operation. These papers go beyond this narrow concern, focusing, first, on the radon problem, and secondly, on the recovery of Kr from spent fuel. The fact that these papers deal with problems that are important from the standpoint of workers and general populations physically quite far from the location of nuclear reactors shows, in my opinion, a maturing of the research in nuclear safety.

One paper in this session will deal with radon produced as a product of uranium before it goes into the nuclear fuel cycle and the other deals with recovery of by-product krypton-85 from the offgases after the fuel has been processed through the reactor.

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DYNAMIC ADSORPTION OF RADON ON ACTIVATED CARBON

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Australian Atomic Energy Commission
Research Establishment, Lucas Heights,
Sydney, N.S.W. 2232 Australia.

Abstract

The adsorption of ^{222}Rn from air onto activated carbon was studied over the range $0-55^{\circ}\text{C}$. A sharp pulse of radon was injected into an air stream that flowed through a bed of activated carbon. The radon concentration in the exit from the column was continuously monitored using a zinc sulfide α -scintillation flow cell. Elution curves were analysed to determine the dynamic adsorption coefficient and the number of theoretical stages.

Five types of activated carbon were tested and the dynamic adsorption coefficient was found to increase linearly with surface area in the range $1000-1300 \text{ m}^2\text{g}^{-1}$. The adsorptive capacity of activated carbon was reduced by up to 30 per cent if the entering gas was saturated with water vapor and the bed was initially dry. If the bed was allowed to equilibrate with saturated air, the adsorptive capacity was too low to be of practical use.

The minimum height equivalent to a theoretical stage (HETS) was about four times the particle diameter and occurred at superficial velocities within the range $0.002-0.02 \text{ m s}^{-1}$. For superficial velocities above 0.05 m s^{-1} , the HETS was determined by the rate of mass transfer. The application of these results to the design of activated carbon systems for radon retention is discussed.

I. Introduction

Three isotopes of radon occur in nature - ^{222}Rn (half-life = 3.8 days) in the uranium-238 decay chain, ^{220}Rn (half-life = 56 s) or thoron from the thorium decay chain, and ^{219}Rn (half-life = 4 s) from the actinium decay chain. Radon isotopes are not a significant radiological hazard in themselves but, because of their mobility as gases, they act as vehicles for dispersal of their alpha-emitting daughters. Radon-222 is the most significant isotope of radon both in terms of occupational exposure in uranium mines and public exposure resulting from its dispersal. The present approach to radon control in underground mines is to reduce the concentration to acceptable levels by forced ventilation. As easily accessible uranium is mined out, there will be a trend towards deeper mines where the technical problems and costs of forced ventilation could be prohibitive. An alternative approach would be to extract the radon from mine air in the vicinity of the working face. An assessment of radon removal processes⁽¹⁾ concluded that adsorption on activated carbon at ambient temperature was the most promising approach in terms of technical feasibility and cost.

This study of radon adsorption on activated carbon was undertaken to obtain more detailed and accurate mass transfer data than are currently available⁽²⁾. Specifically, the results are of value in the design of:

- Large scale equipment to remove radon from underground mine air by adsorption at ambient temperature^(1,3).
- Gas masks or canisters containing activated carbon for short-term use in atmospheres containing very high radon concentrations

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such as unventilated stopes⁽⁴⁾.

- Apparatus used in the analysis of ^{222}Rn and ^{226}Ra by methods that involve the collection of radon on activated carbon^(5,6).
- Systems for delay of ^{220}Rn that is released during reprocessing of high-temperature gas-cooled reactor (HTGR) fuels^(2,7).

II. Experimental Procedure

Figure 1 shows the experimental set-up. Medically pure air was supplied at the required flow rate and humidity to a bed of activated carbon. The temperatures of the carbon bed and the inlet air temperature were controlled to $\pm 1^\circ\text{C}$ by electrical heating. Humidity was adjusted by diverting all or part of the air through a 75 mm diameter column filled with water and packed to a depth of 0.37 m with 5 mm Raschig rings. The moisture content of the air was determined by diverting the air through a silica gel trap and measuring the weight gain. Before commencement of most experiments, the activated carbon was conditioned by passing dry air through the bed for 16 hours. In a few experiments, the bed was heated to 60°C and evacuated overnight. Except where indicated, experiments were conducted at standard conditions:- superficial velocity = 0.087 m s^{-1} , temperature = 25°C , relative humidity = 0%, weight of carbon = 50 g and column diameter = 17 mm.

At the beginning of each experiment, air was momentarily diverted through a bed containing 200 g of high grade (about 30% U) uranium ore, resulting in the injection of a sharp pulse of radon into the carbon bed. Typically, about $0.5\text{ }\mu\text{Ci}$ of ^{222}Rn was extracted from the ore bed with each milking. Zinc sulfide, α -scintillation flow cell detectors located at the inlet and outlet of the adsorption bed monitored both the pulse input and the subsequent elution of radon from the bed. Hold-up times on the activated carbon varied from about 30 min to 24 h. Counts from the outlet detector were integrated using a ratemeter with a time constant of 40 s and graphically recorded. The data from the chart recorder were punched on cards for processing by a computer.

Five types of activated carbon were tested for their efficiency in adsorbing radon. Variables investigated included air velocity ($0.002\text{--}0.3\text{ m s}^{-1}$), temperature ($0\text{--}55^\circ\text{C}$), relative humidity ($0\text{--}100\%$), bed depth ($0.09\text{--}0.9\text{ m}$) and column diameter ($17\text{--}75\text{ mm}$).

III. Flow Cell Detectors

The design of the radon flow cell detectors is shown in Figure 2. The detectors were a modified version of a scintillation cell used in the analysis of ^{226}Ra by emanation of ^{222}Rn . The cells, machined from Lucite (trademark of E.I. du Pont de Nemours & Co. Inc.), had internal volumes of 165 mL , and could be easily dismantled for cleaning or decontamination. A central Lucite baffle directed the flow to prevent short-circuiting and to minimise dead regions within the cell. All internal surfaces including the Lucite top and baffle were coated with a thin layer of high purity ZnS(Ag) scintillant. The coating was applied by greasing all surfaces lightly with vacuum grease and adding a few grams of ZnS(Ag) powder. The scintillant was then tumbled in the cell until a uniform surface coating was obtained. Excess powder was removed from the cell. Besides directing the flow, the central baffle improved the counting geometry by reducing the average path length that an α -particle could travel before striking a surface coated with scintillant. The flow cell was sealed by O-rings and placed, without optical coupling, on top of the photo-multiplier tube. The whole assembly was housed in

a light-tight enclosure. When freshly coated with scintillant, the background of the detectors was less than one count per minute while the efficiency for detection of radon and its α -emitting daughters was over 90 per cent.

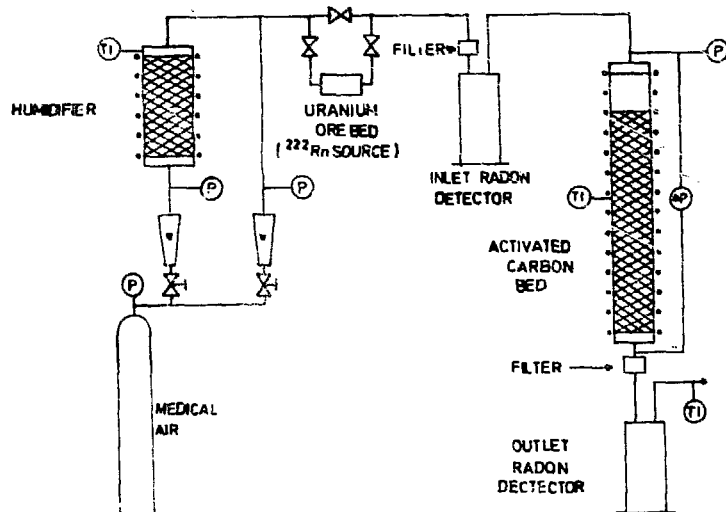


Figure 1. Experimental set-up.

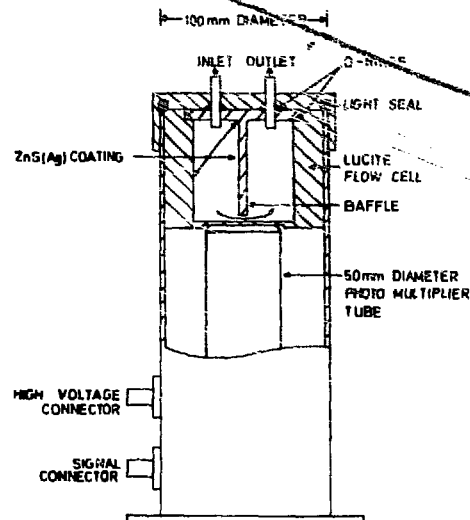


Figure 2. Radon flow cell detector

The inlet of each flow cell contained a glass fiber filter (Whatman GF/B) to remove any radon daughters from the gas stream. Nevertheless, ^{218}Po (RaA) was formed during passage of radon through the flow cell and some was deposited within the cell. The extent of deposition of radon daughters was determined by measuring the rate of decrease in the count rate following passage of a pulse of radon through the inlet detector. The percentage of ^{218}Po deposited varied from 60 per cent to almost 100 per cent and increased as flow rate was decreased. A computer program was written to convert the count rate from the detector to the radon concentration by taking account of the continual deposition and decay of ^{218}Po and the subsequent ingrowth of ^{214}Po . The efficiency of detection of the β -emitter ^{214}Bi was assumed to be zero.

IV. Mathematical Analysis

There are two conventional methods of analysing chromatographic adsorption data. The first approach, borrowed from distillation theory, assumes that a column can be divided into a number of stages of equal volume in which equilibrium always exists between the fluid and the adsorbent. The second approach acknowledges that equilibrium is never actually reached at any point in the column and explains performance in terms of the rate of diffusion and other mass transfer processes. Most descriptions of chromatographic separation rely on the equilibrium stage concept and accordingly this approach is adopted here. The response of N equilibrium stages to a change in concentration of adsorbing species can be determined by solving N first order differential equations that describe the mixing on each stage. For a pulse input in the adsorbing species and assuming a linear isotherm, the elution curve is^(8,9)

$$C = \frac{N! N^{N-1} e^{-NT}}{(N-1)!} \quad (1)$$

where T is dimensionless time normalised to the mean residence time, τ ; $T = t/\tau$; and C is dimensionless concentration normalised so that $\int_0^\infty C dT = 1$. The mean residence time is given by

$$\tau = \frac{k_a m}{F} \quad (2)$$

where F = flow rate ($\text{m}^3(\text{STP})\text{s}^{-1}$), k_a = dynamic adsorption coefficient ($\text{m}^3(\text{STP})\text{kg}^{-1}$) and m = mass of adsorbent in bed (kg). Equation (1) is asymmetric for small N and the maximum in the elution curve occurs at⁽²⁾

$$T_{\max} = \frac{F t_{\max}}{k_a m} = \frac{N-1}{N} \quad (3)$$

For large N , Equation (1) approaches the Gaussian distribution⁽¹⁰⁾

$$C = \sqrt{\frac{N}{2\pi}} e^{-\frac{N(T-1)^2}{2}} \quad (4)$$

and the models based on equilibrium stage theory and diffusion rates coincide.

In practice, the dynamic adsorption coefficient can be determined from the mean residence time or, more simply, from the maximum in the elution curve using Equation (3). A number of methods have been proposed to determine the number of theoretical stages including moment analysis, calculation of the variance, the ratio of peak height to area and the method of tangents^(11, 12). One of the simplest yet accurate methods is to measure the width of the elution curve half-way up the peak ($\Delta t_{1/2}$). Peak widths were determined numerically from Equation (1) and found to satisfy the following relation,

$$\left(\frac{t_{\max}}{\Delta t_{1/2}}\right)^2 = 0.180 N - 0.194 \quad (5)$$

Since the number of theoretical stages depends on column length (L) and the diameter of the packing (d_p), the HETS and the reduced height of a theoretical plate (h) are frequently quoted as indices of column performance⁽¹³⁾. These terms are defined by the equation,

$$h = \frac{\text{HETS}}{d_p} = \frac{L}{N d_p} \quad (6)$$

V. Results and Discussion

Shape of the Elution Curves

The radon elution curves were processed by computer to determine the dynamic adsorption coefficient from Equation (3) and the number of theoretical stages from Equation (5). For the packed beds used in these experiments, the number of theoretical stages generally varied between ten and one hundred. The theoretical elution curves were calculated from Equation (1) and compared with the experimental curves.

Figure 3 compares a series of experiments using varying bed depths of Pittsburgh PCB activated carbon. In general, the experimental elution curves fitted the theoretical curves quite closely even when the number of theoretical

stages was so low that the asymmetry in the elution curves was obvious. A slight degree of tailing was noted in the experiments using Sutcliffe-Speakman type 207C activated carbon.

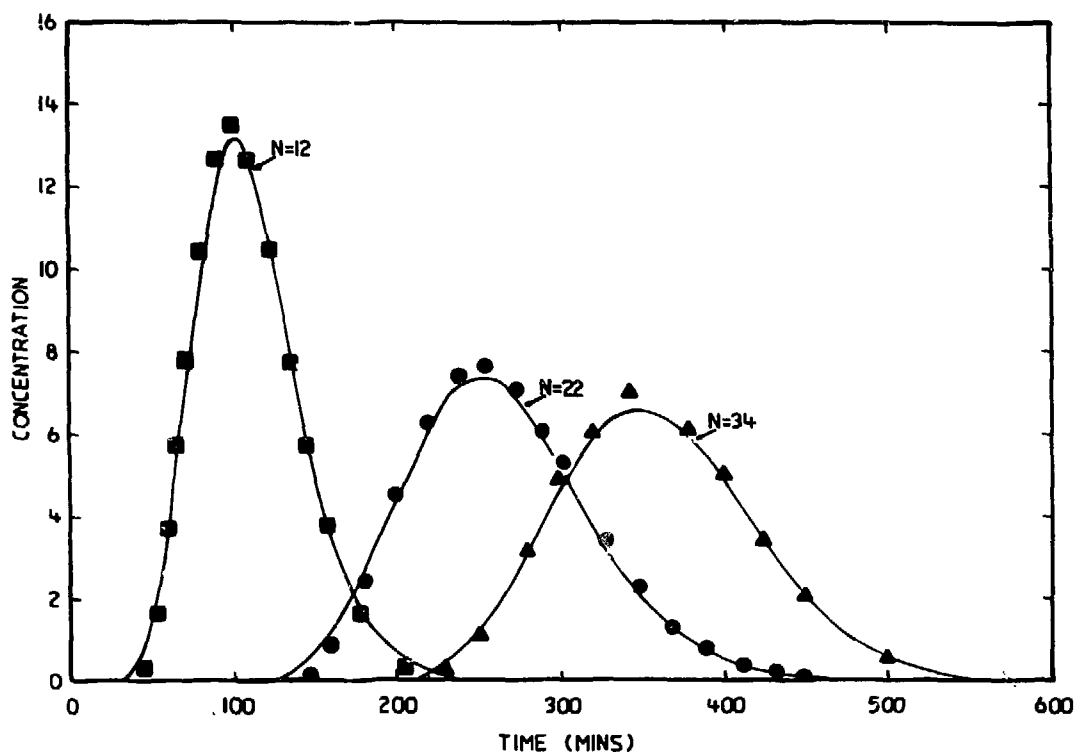


Figure 3. Shape of elution curves.

Air Velocity

As expected, the dynamic adsorption coefficient was found to be independent of air flow rate. Figure 4 shows the effect of superficial air velocity on the HETS for Sutcliffe-Speakman type 207C activated carbon. The minimum HETS was about four times the particle diameter. For velocities above 0.05 m s^{-1} , the HETS increased linearly with velocity.

The shape of Figure 4 agrees with the van Deemter equation for the effect of carrier gas velocity on HETS⁽¹⁴⁾.

$$\text{HETS} = A + \frac{B}{u_s} + C u_s \quad (7)$$

where u_s is the superficial gas velocity. A, B and C are, respectively, the coefficients due to eddy diffusion, longitudinal diffusion and resistance to mass transfer which occurs primarily within the adsorbent particle. The value of the constants in Equation (7) that give the best fit to the experimental data are $A = 2.7 \times 10^{-3} \text{ m}$, $B = 7.3 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$, $C = 0.12 \text{ s}$. For Sutcliffe-Speakman 207C carbon, Equation (7) can be expressed in dimensionless form as,

$$h = \frac{\text{HETS}}{d_p} = 2.2 + \frac{0.73}{\text{Pe}} + 0.84 \text{ Pe} \quad (8)$$

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where Pe = Peclet number based on superficial velocity ($d u_s/D$) and D = diffusion coefficient of adsorbate ($D \sim 10^{-5} \text{ m}^2 \text{ s}^{-1}$ for radon). The constants in Equations (7) and (8) are similar to published data for other noble gases^(2, 12, 13, 15).

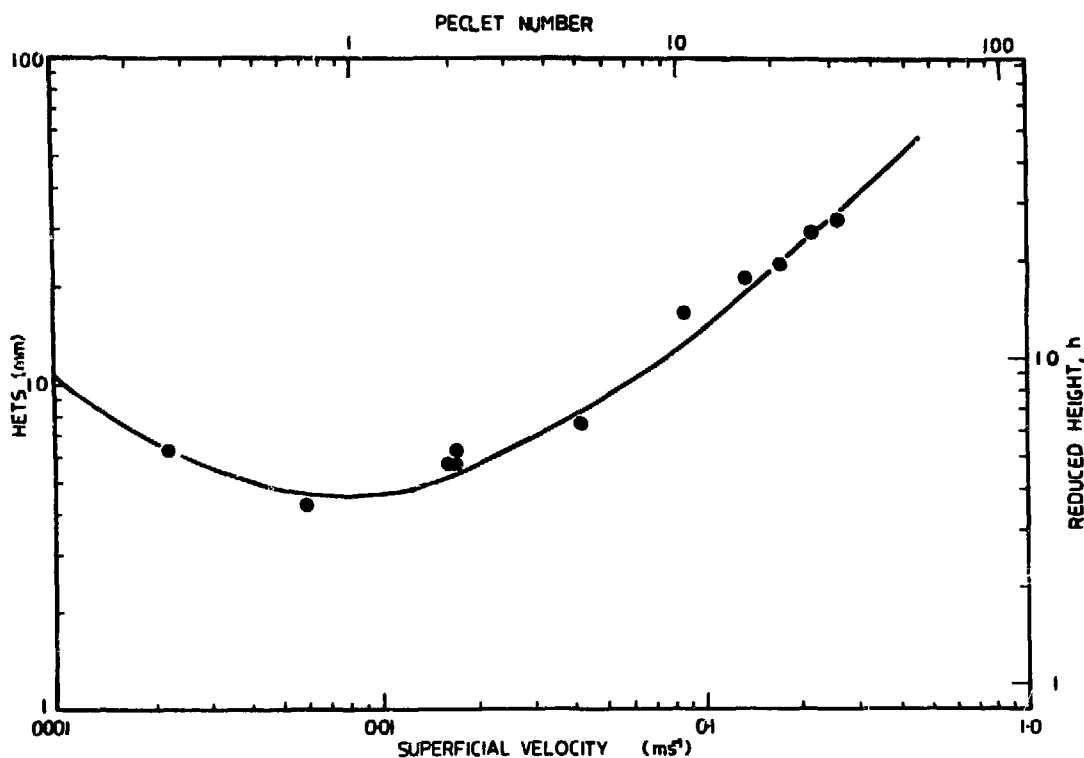


Figure 4. Effect of superficial velocity on HETS (temperature = 25°C)

Temperature

Figure 5 shows that within the temperature range 0-55°C the dynamic adsorption coefficient follows the Arrhenius equation. The calculated heat of adsorption is $6.9 \text{ kcal mol}^{-1}$ (29 kJ mol^{-1}) which compares well with values in the range $6.1\text{--}7.1 \text{ kcal mol}^{-1}$ ($26\text{--}30 \text{ kJ mol}^{-1}$) reported in the literature^(1, 2).

Figure 6 shows the effect of temperature on HETS for Sutcliffe-Speakman type 207C carbon at constant molar flow rate. The HETS increased with temperature in a similar way to that previously reported for adsorption of krypton and xenon on activated carbons⁽¹³⁾. The theoretical effect of temperature on HETS is complex since the effects due to changes in the diffusion coefficient, gas velocity and the dynamic adsorption coefficient often act in different directions⁽¹¹⁾. The data in Figure 6 correspond to the case in which mass transfer makes the major contribution to HETS. In this case, for a rise in temperature, the diffusion coefficient increases thus tending to decrease the HETS, whereas the increase in gas velocity and the fall in the dynamic adsorption coefficient tend to increase the HETS. The biggest effect is caused by the change in dynamic adsorption coefficient since this affects the relative times that the adsorbing species spends in the adsorbed state and undergoing mass transfer. Overall the performance should improve (i.e. the HETS should fall) as temperature is decreased⁽¹¹⁾.

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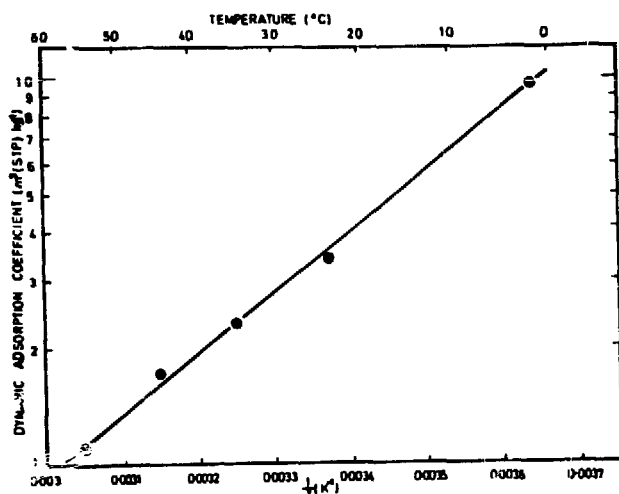


Figure 5. Temperature dependence of the dynamic adsorption coefficient

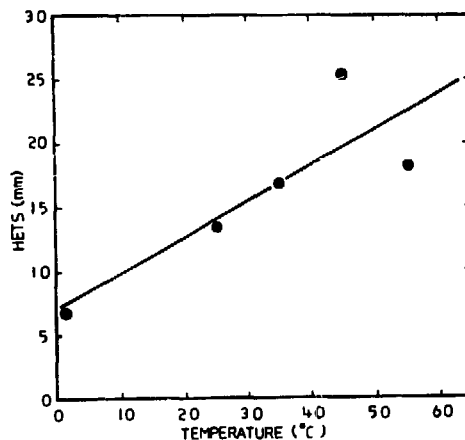


Figure 6. Effect of temperature on HETS

Type of Carbon

A comparative study was made of five commercial grades of activated carbon at the same superficial velocity (0.087 m s^{-1}) and otherwise identical conditions. There was a wide variation in performance as shown in Table 1. The measured dynamic adsorption coefficients ranged from $3.53\text{--}5.69 \text{ m}^3(\text{STP})\text{kg}^{-1}$ compared with values ranging from $0.90\text{--}7.50 \text{ m}^3(\text{STP})\text{kg}^{-1}$ in the published literature^(1,2). Figure 7 shows a positive correlation between the dynamic adsorption coefficient and specific surface area. This is in contrast with the results of Kovach⁽¹⁶⁾ for adsorption of krypton and xenon on activated carbon where the dynamic adsorption coefficient passed through a maximum at a surface area of $900 \text{ m}^2\text{g}^{-1}$.

Table I. Properties and performance of activated carbons

Type	Source	Shape	Mass Mean Diameter (mm)	Surface Area (N_2 , BET) (m^2g^{-1})	k_a $\text{m}^3(\text{STP})\text{kg}^{-1}$	HETS (mm)	h
Sutcliffe-Speakman 207C	Coconut shell	flakes	1.2	1000	3.53	14.9	12.4
Norit RFL3	Peat	extruded cylinders	4.7	1150	4.61	66.9	14.2
Norit RL111	Peat		4.7	1200	4.66	67.9	14.4
Ultrasorb SC2	Coconut shell	chips	0.7	1200	5.00	5.6	8.0
Pittsburgh PCB	Coconut shell	flakes	2.5	1300	5.69	22.9	9.2

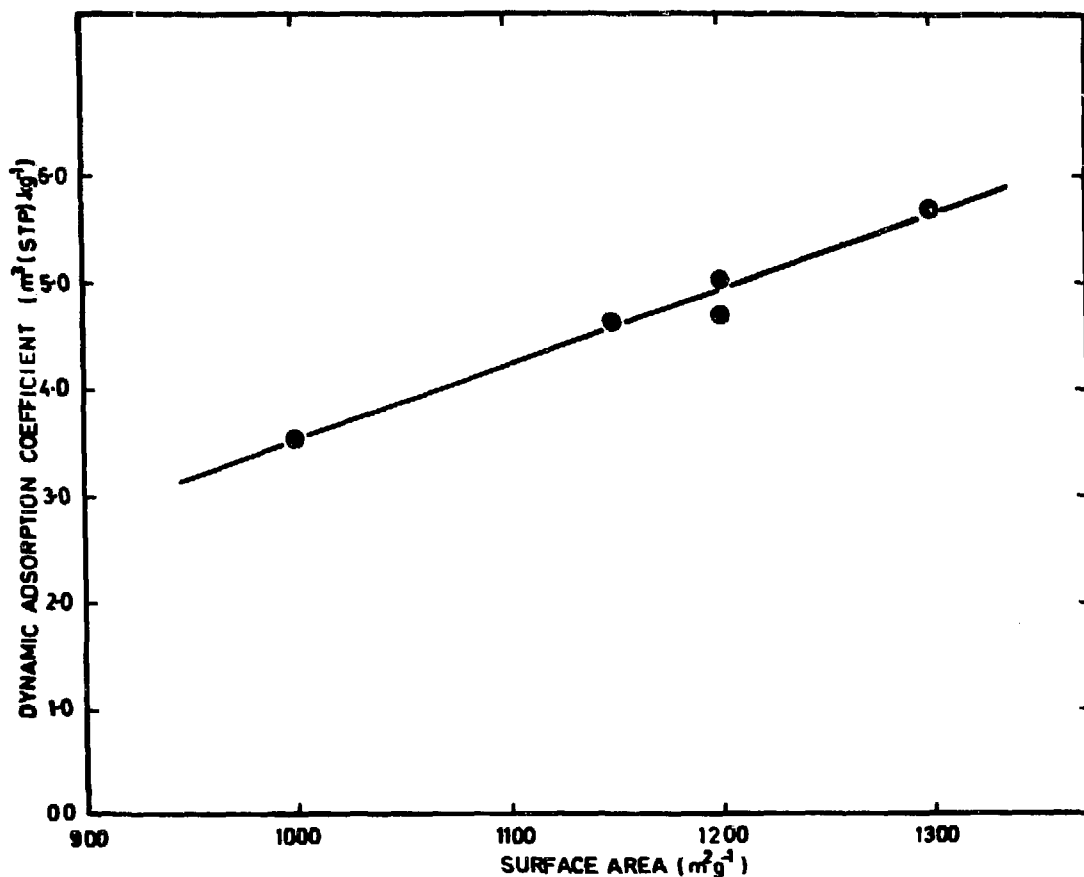


Figure 7. Relation between surface area and dynamic adsorption coefficient

Particle size is the most significant factor affecting the HETS. There appears to be a residual correlation between reduced height of a theoretical stage and both particle diameter and k_a but these effects are difficult to unravel. Prediction of k_a using Equation (8), which strictly only applies to Sutcliffe-Speakman 207C carbon, gives reasonable agreement with the experimental data except for the two Norit carbons. For these extruded cylindrical carbons it is difficult to define an effective diameter for use in the Peclet number, and the predicted values of h using the mass mean diameter are about twice the experimental values.

Bed Height

Four experiments were carried out with bed heights ranging from 90-900 mm but with otherwise identical conditions. The dynamic adsorption coefficient was constant but, as shown in Figure 8, the HETS increased slightly as height was raised. This is a well-known phenomenon in chromatographic columns which has generally been explained in terms of the change in velocity through the bed due to the pressure drop (10, 11). In the present case, this does not appear to be the explanation since the pressure drop was only 1 kPa per meter of packing. A more plausible explanation for the deterioration in performance as bed height is raised is the progressive increase in channeling in the area of high voidage near the wall. This effect has been observed in other mass transfer equipment and can be remedied in very deep beds by installation of redistributors at regular intervals along the column.

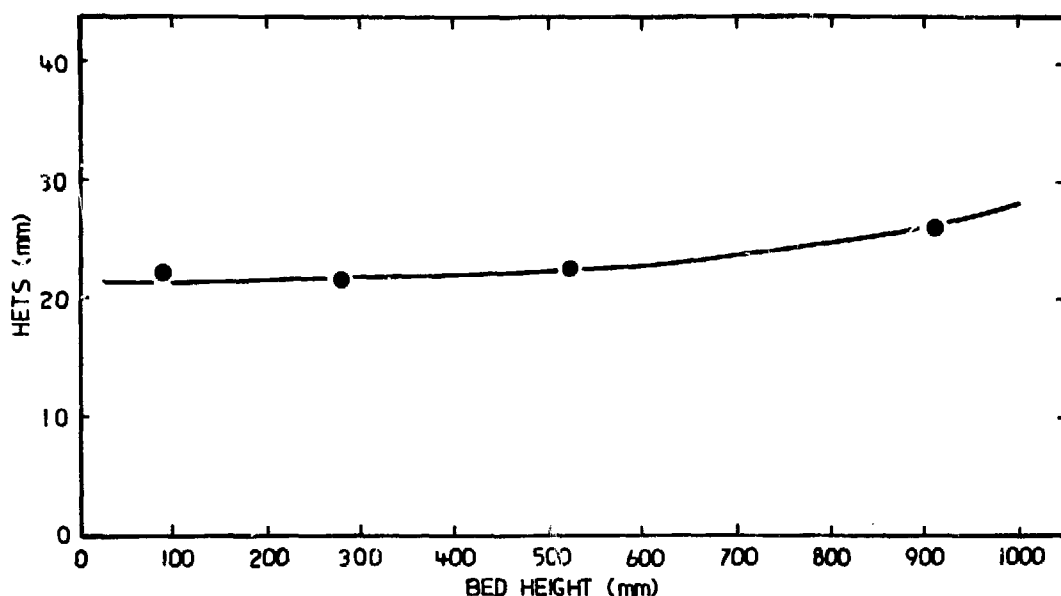


Figure 8. Effect of bed height on HETS (Pittsburgh PCB carbon, temperature = 25°C, superficial velocity = 0.087 m s⁻¹)

Column Diameter

A reduction in performance with increase in diameter of chromatographic columns has been reported by many workers and is usually attributed to an uneven flow profile across the column⁽¹¹⁾. To determine the effect of column diameter on performance, three experiments were conducted with column diameters ranging from 17 to 75 mm at constant superficial velocity. Each column was carefully packed to the same bulk density (0.47 kg l⁻¹) with Sutcliffe-Speakman 207C activated carbon. The experimentally measured HETS was 5.7 mm for both 17 mm and 38 mm diameter columns and slightly higher (6.4 mm) for the 75 mm column. This variation in HETS was probably within experimental error and these experiments do not indicate a significant effect of column diameter on performance.

Humidity

The effect of water vapor on radon adsorption was studied in two series of experiments. In the first series, the activated carbon bed was first equilibrated with water vapor by passing humidified air through the bed at the test conditions for 16 hours. When equilibrated in this way, the performance of the bed was satisfactory at low relative humidities (<15%) but deteriorated rapidly for higher humidities. The dynamic adsorption coefficient was reduced to 50 per cent of its initial value at 30 per cent relative humidity. The reduction in k_a was matched by a corresponding rise in HETS. A few experiments were carried out at 100 per cent humidity but the retention time was too low to be of practical use. There was considerable scatter in the results of these experiments and local condensation of water was suspected.

In the second series of experiments, humidified air was passed into a dry bed at a temperature of 25°C and a superficial velocity of 0.087 m s⁻¹. Figure 9 shows that the dynamic adsorption coefficient decreased by 30 per cent for an increase in relative humidity from 0-100 per cent. This relatively small effect is explained by the observation that the radon penetrates the bed at a faster rate than water

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vapor. Hence, for most of its travel, the radon passes through a dry bed. Near the inlet, the combined effects of temperature rise due to the heat of adsorption of water and the competition with water vapor for active sites tends to reduce the dynamic adsorption coefficient for radon. A computer program is being developed to solve the equations for heat and mass transfer during adsorption of water vapor on activated carbons. Initial results suggest that the effect of temperature rise on k_a is more important than co-adsorption of water vapor.

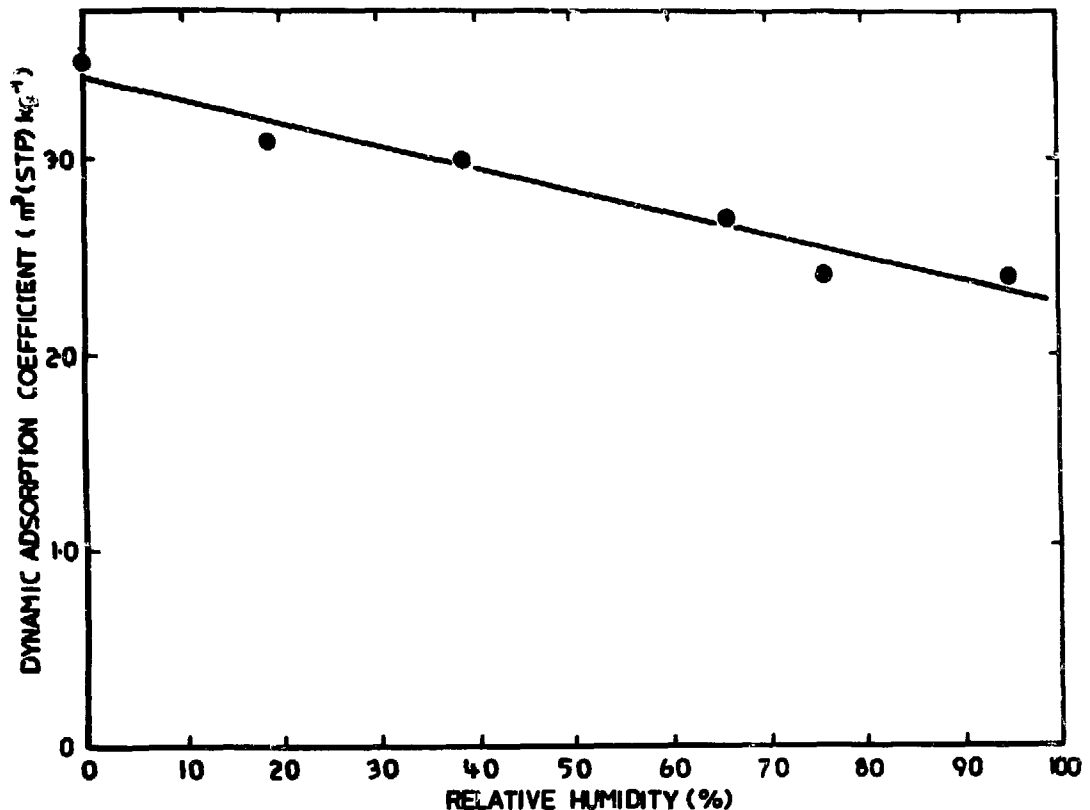


Figure 9. Effect of humidity on the dynamic adsorption coefficient (Sutcliffe-Speakman 207C carbon)

The results reported here are in contrast to the work of Thomas⁽⁴⁾ who reported that deterioration in performance of gas mask canisters was most marked when the activated carbon was initially dry. The canister tested by Thomas had a diameter of 0.1 m and a depth of only 31 mm so inlet effects, particularly the sharp initial rise in temperature, would be expected to dominate under these conditions.

Ageing and Regeneration

The activated carbon bed was usually regenerated between experiments by passing dry air through the bed at a superficial velocity of 0.1 m s^{-1} for 16 hours. In a few cases the bed was heated to 60°C and evacuated overnight. This did not improve the adsorption properties of any of the carbons with the exception of Sutcliffe-Speakman type 207C where a slight increase in k_a was observed. Many experiments were conducted with the same carbon bed over several months without deterioration in performance due to ageing.

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VI. Application of Results

Since the mass of radon adsorbed on an activated carbon bed for all practical cases is infinitesimal, the performance of a bed is independent of radon concentration and its capacity is determined by the total volume of air that passes through it. In most practical instances, the radon enters the bed as a step change in concentration rather than as the radon pulse used in these experiments. The response to a step change can be simply calculated by integrating the elution curve for a pulse input (Equations (1) or (4)).

For optimum performance, a radon adsorption bed should be sufficiently deep (at least 0.2 m is suggested) so as to contain a large number of stages and minimise broadening of the breakthrough front. The superficial velocity should ideally be in the range $0.002\text{--}0.02\text{ m s}^{-1}$, which will minimise the HETS (see Figure 4) and ensure a low pressure drop. Unfortunately these requirements are not always consistent. This can be seen by expressing Equation (2) in the form,

$$\tau = \frac{T k_a \rho_b L}{273 u_s} \quad (9)$$

where ρ_b = bulk density of the carbon bed, typically about 450 kg m^{-3} . Substituting $k_a = 5\text{ m}^3(\text{STP})\text{kg}^{-1}$ at $T = 298\text{ K}$ in this equation gives

$$\tau = \frac{2500 L}{u_s} \quad (\text{s}) \quad (10)$$

For the recommended limits, $L = 0.2\text{ m}$ and $u_s = 0.02\text{ m s}^{-1}$ then $\tau = 7\text{ h}$. This means that, unless the bed is designed for a retention time of 7 h or longer, some departure from optimum conditions must be made. In practice, the velocity should be increased rather than the column length reduced because this results in a larger number of theoretical stages.

The design of an activated carbon adsorption process to remove radon from underground mine air was considered by Arthur D. Little Inc.⁽¹⁾. This design was based on $k_a = 4\text{ m}^3(\text{STP})\text{kg}^{-1}$ which is consistent with the results reported here after allowing for the effects of humid air on performance. A sharp breakthrough front was assumed and no allowance was made for broadening due to a finite number of stages. The equipment was designed to process $2.36\text{ m}^3\text{s}^{-1}$ (5000 SCFM) at a bed superficial velocity of 0.4 m s^{-1} . For these conditions, the HETS would be approximately 50 mm (see Figure 4) and the bed depth of 0.82 m would be equivalent to only 16 theoretical stages. Broadening of the breakthrough front would be significant and the capacity of the bed would be reduced accordingly. Even with optimistic assumptions, the study found that removal of radon from underground mine air was not competitive with conventional forced ventilation on economic grounds. The major cost was not the carbon adsorber itself but that of ancillary equipment such as blowers, heat exchangers and heating costs associated with thermal regeneration to desorb radon from the exhausted bed. If a simple and cheap method of bed regeneration were devised, adsorption on activated carbon could become a feasible method of removing radon from mine air.

Thomas⁽⁴⁾ has demonstrated that portable canisters containing activated carbon can provide short-term protection (up to 1 h) from exposure to very high radon concentrations such as might occur in unventilated mine stopes. The breakthrough data presented by Thomas have been reanalysed to determine the dynamic

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adsorption coefficient and values ranging from 3.8-5.7 m³(STP)kg⁻¹ at room temperature have been estimated for the most effective carbon tested^(1, 2). The Mil Army gas mask canister has a depth of only 31 mm which violates the design criteria discussed above. As a consequence, some breakthrough of radon occurs after a very short interval and this canister is not recommended. The Scott-Acme canister has a greater capacity, and being longer (15 cm), it is a more satisfactory design.

Activated charcoal traps are frequently used to collect radon for analytical purposes. Two common applications are atmospheric sampling for ²²²Rn in the vicinity of uranium mines and mills, and determination of ²²⁶Ra in solutions by ingrowth and subsequent de-emanation of ²²²Rn^(5, 6). In most applications it is recommended that the activated carbon traps be cooled with liquid nitrogen or dry ice. Calculations of adsorber capacity based on the mass of carbon and the volume of air that is passed through the trap frequently show that it is not essential to cool the traps, although in some cases the geometry of the bed needs to be modified. Adsorption of radon at ambient temperatures would simplify procedures particularly in situations that require field work.

Another possible application for activated carbon is the removal of ²²⁰Rn from the off-gas stream during reprocessing of HTGR fuel^(2, 8). Since ²²⁰Rn has a half-life of only 56 s, only 20 minutes is sufficient for essentially complete retention. Activated charcoal has 10-20 times the adsorptive capacity of molecular sieves and about 200 times that of silica gel^(1, 2). As with other applications for activated carbon, the major disadvantage is the fire hazard.

VII. Conclusions

Over the temperature range, 0-55°C, the dynamic adsorption coefficient for radon on activated carbons with specific surface areas (S) in the range 1000-1300 m²g⁻¹ can be estimated from the equation

$$k_a(\text{m}^3(\text{STP})\text{kg}^{-1}) = (0.0070S - 3.51)e^{-\frac{6.9 \times 10^3}{RT}} \quad (11)$$

The presence of water vapor in air decreases the dynamic adsorption coefficient. For an initially dry bed of activated carbon at ambient temperature, the dynamic adsorption coefficient decreases by about 30 per cent for an increase in relative humidity from 0-100 per cent. If the bed is allowed to equilibrate with saturated air, the adsorptive capacity is too low to be of practical use.

The minimum height equivalent to a theoretical stage (HETS) occurs at superficial velocities in the range 0.002-0.02 m s⁻¹. This minimum value is typically about four times the particle diameter. For superficial velocities above 0.05 m s⁻¹, the HETS is governed by the rate of mass transfer.

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DISCUSSION

TADMOR: Dr. Levins, as you rightly mentioned, radon is a problem in the mining of uranium, but it may be also a long term problem from the long term release of radon from tailings and other residual materials from mining. Do you have any data, or would you venture to make some predictions, on the adsorption of radon on soil and underground material which may be used to cover the tailings produced in mining or milling of uranium?

LEVINS: We haven't a lot of evidence on the adsorption of radon on soil materials. My general observation would be that it wouldn't be very strong. There is one exception; silica, that may appear in tailings, may adsorb the radon weakly, but I don't think it would be a significant factor in retarding the release of radon from a mine site or tailings pile.

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DEVELOPMENT OF A CRYOGENIC KRYPTON-SEPARATION SYSTEM FOR THE OFFGAS OF REPROCESSING PLANTS

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Abstract

The concept of cryogenic rare gas separation from reprocessing offgas pursued at present at the Kernforschungszentrum Karlsruhe (KfK) is discussed and compared with other offgas purification flowsheets. The KfK concept includes: separation of O_2 and residual NO_x by catalytic reduction with H_2 , adsorptive retention of H_2O , CO_2 , NH_3 etc. and cryogenic distillation of first N_2 -Kr-Xe, then Kr-Xe mixtures. Some features pertinent to this flowsheet which were studied experimentally either on a laboratory or semiworks scale, are described including the following: desublimation of Xe in the first column, coadsorption of Kr at and its selective desorption from the adsorption bed, purification of the Xe product, poisoning of the reduction catalyst and methanization of CO_2 . It is shown that all of these features, despite being capable of causing malfunctions of the process, can be controlled by proper process design and operational conditions in a way not to impair with a good Kr decontamination factor.

I. Introduction

Plans for retention of the fission gas Kr from the offgas of the future large German reprocessing plant as well as of the existing small plant WAK (Wiederaufarbeitungsanlage Karlsruhe) prefer a cryogenic distillation scheme. The reason is the long experience with such a process⁽¹⁾ in the industrial production of rare gases in air liquefaction.

Two essential differences between the rare gas-oxygen mixtures in air liquefaction plants and reprocessing offgases demand for modifications of the process:

- the mole ratio Kr/Xe in fission gas (0,103) which is almost reciprocal to the ratio in air (13,1); the resulting higher Xe-concentrations enhance the problem of Xe freeze-out;
- the radioactivity of Kr-85 resulting in the radiolytic ozone formation from O_2 .

Depending on the risks one places on these differences the existing or planned Kr retention units show different flowsheets (fig. 1)⁽²⁾. At the I.N.E.L. (U.S.A.)⁽³⁾ and at the C.E.A. (France)⁽⁴⁾ the complete offgas including O_2 is liquefied in the first column. Here the risk of ozone formation is accepted, but freeze-out of Xe is decreased because of improved solubility in O_2 compared to N_2 . Further enrichment of Kr is achieved either by batch distillation of the three-component mixture O_2 -Kr-Xe in the second column (I.N.E.L.) or by catalytic reduction of O_2 with H_2 between the two columns and

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subsequent distillation of Kr-Xe (C.E.A.).

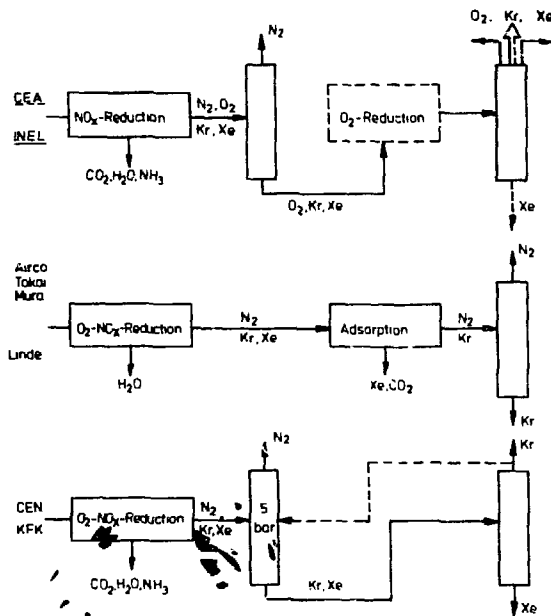


FIGURE 1

Different flowsheets for the retention of Krypton from the offgas of LWR-reprocessing plants

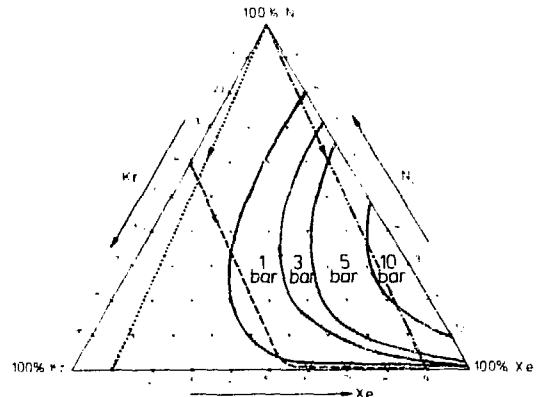


FIGURE 2

Pressure dependence (ideal behavior) of the two-phase area solid-liquid in the phase diagram N₂-Kr-Xe and course of bottom product concentrations during start-up operation of column
 --- without additional Kr
 --- additional Kr before operation
 --- Different operation modes with Kr recycle

Both risks are circumvented by proposals of Linde (Germany)⁽⁵⁾ and by Airco (U.S.A.) for the Japanese Tokai Mura plant⁽⁶⁾: both O₂ and Xe are separated from the offgas before the cryogenic part by catalytic reduction and by adsorption, respectively. Workers at KfA Jülich (Germany) prefer a separation of Xe by freeze-out instead of adsorption⁽⁷⁾. After separation of Xe the cryogenic distillation is simplified to the two-component system N₂-Kr.

In the present concept of KfK⁽⁸⁾, similar to the plans of the C.E.N. (Belgium)⁽⁹⁾ O₂ is also removed by catalytic reduction, but adsorption of Xe is omitted. Thus the three-component system N₂-Kr-Xe must be distilled in the first column. Whereas N₂ is driven off at the head, Kr and Xe are collected in the bottom of the still. After discontinuous transfer to the second column Kr and Xe are separated into pure products and filled into steel cylinders. The cryogenic part of this concept together with a preceding molecular sieve bed as drying unit was installed on a "cold" semiscale basis (gas throughput 30-50 m³/h at S.T.P.). These units which bear the names KRETA (Krypton-Entfernungs-Tieftemperatur-Anlage) and ADAMO (Adsorption an Molekularsieben) have been in operation now for 4000 hours. The

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design of these units has been described elsewhere⁽¹⁰⁾. Some of the results gained are reported in the following sections.

II. Features of the KfK Flowsheet and Results

1. Desublimation of Xenon

1.1 Solid-Liquid Equilibrium

The phase diagram of the three-component system N_2 -Kr-Xe has a region of limited solubility on the N_2 -Xe side. Solid Xe is in equilibrium with N_2 -Kr-Xe liquid (fig. 2). This two-phase area decreases on increasing the pressure and thus the temperature. One could therefore prevent Xe from freezing out completely by operating the column at 17-20 bar⁽¹¹⁻¹³⁾. Because of safety aspects in plants containing large inventories of radioactivity we chose an intermediate pressure of 5 bar which is also familiar practice in air liquefaction.

Fig. 2 also shows the problem of Xe freeze-out during the start-up period of the column: if the fission gas mixture is fed into the column cooled down with liquid nitrogen (at 95 K) one would pass straight through the critical area (dot-dashed line). This situation can be avoided, if the column is filled with a N_2 -Kr mixture before the fission gas feed is opened. In this case the stationary bottom concentration is approached first on a straight line, then, after opening the bottom product transfer line to the second column, on a curved line (dashed line). The critical area is not intersected. Alternatively, one can also raise the Kr/Xe ratio to or near that of the air by means of an additional Kr feed. In the second Kr separation unit at Karlsruhe being in the planning state now for the offgas of the WAK plant a Kr recycling loop from the top of the second column to the first column possibly will be installed (see fig. 1)⁽¹⁴⁾. In this case one would be on the safe side in all parts of the column (dotted line in fig. 2).

We have tested all three versions in the KRETA campaigns and confirmed the expectations: whereas in the first version we had serious malfunctions of the column because of Xe freeze-out, we have attained Xe concentrations up to 80 % in the bottom product in the second version without having detected any precipitation of solid Xe.

During unperturbed operation of the column the Kr decontamination factor (DF) at the head was $\geq 10^3$, the upper limit set by the sensitivity of the He ionization detector of our gaschromatographic analysis (100 ppb) and the normal Kr feed concentration of 160 ppm by vol.

1.2 Gas-Solid Equilibrium

More essential than the solid-liquid equilibrium is in our column the gas-solid equilibrium. Due to the construction of the column which is a sieve plate column containing 37 plates with the feed point above the 18th plate (overall height: 8 m) and due to the temperature profile within the column (gas temperature at the feed point: 135 K) we observe desublimation of Xe at the first plate above the feed point (temperature: 95 K) under certain conditions. There solid Xe is not dissolved completely by liquid N_2 , but accumu-

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lates until the sieve holes are plugged. According to the sublimation pressure curve of Xe (fig. 3) the limiting Xe-concentration in the feed gas at 95 K should be only 80 ppm. In practice we observe a limiting Xe concentration which is higher by almost an order of magnitude. We conclude therefore that the difference is either dissolved by liquid N₂ or carried down the column mechanically in solid form. The aim of our present work is to increase this concentration by increasing the temperature profile in the vicinity of the feed point and by altering the construction of the column interior in that area.

What are the consequences of this result for the rare gas separation from a real offgas? The offgas most pertinent to our development at the moment is the offgas of the WAK. This offgas consists of 80 - 120 m³/h at S.T.P. of essentially air containing a maximum of 1600 ppm of Xe at the peak of a dissolution. If continuously fed to a column like ours, it should result in serious plugging after a short time. However, a dissolution of fuel in WAK is essentially completed after two hours followed by a day until the next dissolution starts. We simulated such a feed gas in KRETA (fig. 4) and showed that such a rare gas concentration peak does not lead to any signs of plugging, even if another rare gas peak simulating a dissolution is fed to the column two hours after the end of the first run.

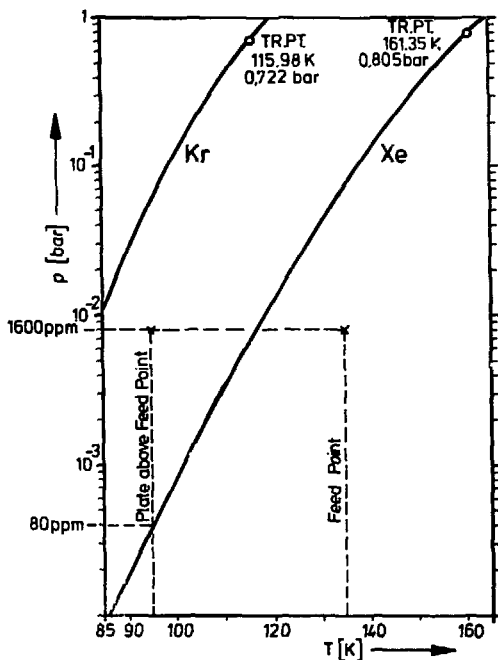


FIGURE 3
Sublimation pressure of Krypton and Xenon

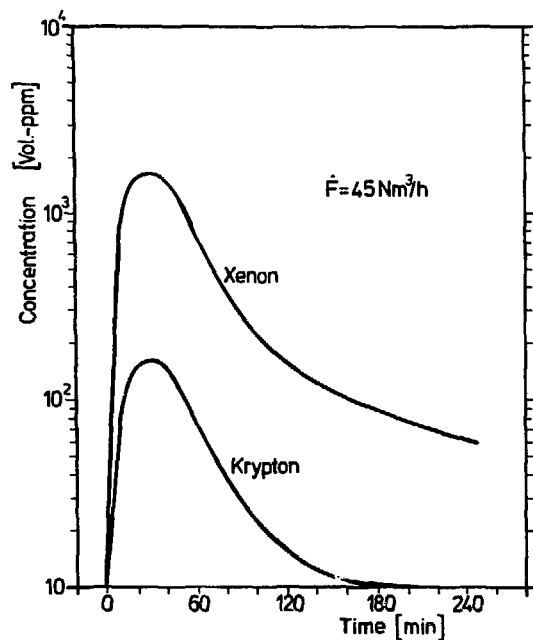


FIGURE 4
Kr- and Xe-concentrations in a simulated offgas of a dissolution at WAK

In the case of a large reprocessing plant where rare gas concentrations probably will be higher and on a more continuous level, this concept would have to be changed, however, unless we can avoid plugging by desubliming Xe by means of modifying the column structure.

2. Ozone Formation

The primary reason for the separation of O_2 in our concept was to prevent excessive ozone formation. However, even during normal operation, not to speak of malfunction of the reduction catalyst traces of O_2 will reach the cryogenic column. Ozone formation rates therefore have to be known. In addition, use of liquid O_2 as solvent for the rare gases in the first cryogenic column is an interesting process alternative in many respects⁽³⁾. If one considers this alternative, the safety risks of ozone formation have to be assessed. We have therefore carried out a calculation of the ozone concentration to be expected in the first column considering three process alternatives (table I):

- 1) The first case is the alternative presently investigated by us in the KRETA pilot plant; it is assumed that 10 ppm O_2 pass the reduction unit.
- 2) The second case was proposed for the offgas purification system at the WAK⁽¹⁴⁾, called project AZUR. Here the ratio Kr/Xe is raised to that of air by a Kr recycling loop; O_2 again is reduced down to 10 ppm.
- 3) In the third case, O_2 which is present in the offgas in air concentration (20 %) is not reduced; it is partially (50 %) liquefied thus acting as solvent for the rare gases; Kr is not recycled.

The O_2 and Kr concentration profiles along the column were calculated by a tridiagonalmatrix method⁽¹⁵⁾. Stationary column operation has been assumed which is attained 700 hours after startup.

The ozone formation rate was calculated according to the equation shown in fig. 5, where an average G-factor of 10 was assumed⁽¹⁶⁾, where ϵ_{O_2} is the O_2 electron fraction, where the Kr concentration C_{Kr} takes into account the abundancy of Kr-85 in overall Kr (6 %) and where the radiation dose rate is the product of the specific activity of Kr-85 and its average β -energy. The dimensions of the column considered are typical of the KRETA pilot plant (liquid volume of bottom product: 15 l, and of each of 10 theoretical plates: 1,2 l).

The calculated ozone concentrations in the bottom product are given in table I. Surprisingly the highest value is not obtained in the case of O_2 operation (case 3), but with 10 ppm O_2 in the feed gas (case 1), whereas in the Kr recycle mode (case 2) it is lowest. The explanation is found in the different transfer rates of the bottom product; whenever this rate is high, the residence time of O_2 and Kr in the column is short and thus the ozone formation rate is low. This is particularly true for cases 2 and 3. In case 2 the effect of high Kr concentration is even overcompensated.

Generally, the calculated ozone concentrations are very low and presumably beyond a critical concentration^(17,18). The calculation

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Flowsheet	1 "KRETA"	2 Kr-Recycle	3 O ₂ -Liquefaction
Feed Composition (N ₂ Carrier Gas)	Kr 500 Vol.-ppm Xe 5000 Vol.-ppm O ₂ 10 Vol.-ppm	5 Vol.-% 5000 Vol.-ppm 10 Vol.-ppm	500 Vol.-ppm 5000 Vol.-ppm 20 Vol.- %
Transfer Rate of Bottom Product [l/h]	0,56	3,98	8,41
Krypton-Inventory [l] [Ci]	2,24 1,14 · 10 ⁵	21,2 108 · 10 ⁶	0,06 3,06 · 10 ³
Ozone-Concentration [Mol-ppm] (Bottom)	170	8,2	53

Table I

Ozone formation in the first cryogenic column in different process flowsheets

$$\frac{d[O_3]}{dt} = \frac{G [eV^{-1}] \cdot \epsilon_{O_2} [Mol/Mol] \cdot C_{Kr} [Mol/Mol] \cdot \text{dose rate} [eV \cdot l^{-1} \cdot s^{-1}]}{N_L [Mol^{-1}]}$$

$$\frac{d[O_3]}{dt} = \frac{10 \cdot 10^{-2} [eV^{-1}] \cdot \epsilon_{O_2} \cdot 0,06 \cdot C_{Kr} \cdot 8,51 \cdot 10^5 [Ci \cdot l^{-1}] \cdot 3,7 \cdot 10^{10} [Ci^{-1} \cdot s^{-1}] \cdot 2,49 \cdot 10^5 [eV]}{6,02 \cdot 10^{23} [Mol^{-1}]}$$

$$\frac{d[O_3]}{dt} = 7,8 \cdot 10^{-5} \cdot \epsilon_{O_2} \cdot C_{Kr} [Mol \cdot l^{-1} \cdot s^{-1}]$$

FIGURE 5

Ozone formation rate in cryogenic rare gas mixtures

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shows that ozone formation is not a serious problem in cryogenic Kr separation, since it can be controlled by proper design of the process. It must be kept in mind, however, that further build-up of the ozone level may take place in the second column, especially in the Xe product, if an ozone and oxygen reduction step between the columns is not considered (4, 17, 19).

Table I indicates another inherent safety aspect: the inventory of Kr and thus of the activity. It varies by almost three orders of magnitude and is lowest in the O_2 operation mode and highest in the Kr recycle mode.

3. Points of Krypton Leakage Inherent to the Process

The head of the first column is the most important, but not the only point, where Kr is released to the atmosphere. Two additional points are discussed in the following sections.

3.1 Adsorption Unit

A conventional adsorber unit is provided in our pilot plant to dry the feed gas and, later on, to retain all offgas components which, if present, would freeze out in the cryogenic part, i.e. CO_2 , NO_x and NH_3 . Some Kr is also adsorbed during this step. Although this coadsorption of Kr is weak at room temperature (20), it would be sufficient to spoil the good column decontamination factor, if it would be released to the atmosphere on regeneration of the adsorber. Therefore a purging step is included in the operation mode of the adsorber bed, where the coadsorbed Kr is selectively desorbed and recycled. This concept led to the design of three adsorber lines each consisting of two beds. The first one is presently filled with 70 kg of silicagel, the second one with 100 kg of molecular sieve type 10A.

Breakthrough₃ (at $c/c_E = 0,5$) of Kr (160 ppm by vol.) and Xe (1600 ppm) in $31 \text{ m}^3/\text{h}$ (S.T.P.) at 5 bar and room temperature takes place after 4.2 and 26.0 minutes, respectively, through the two beds of a line. Desorption curves for Kr labelled with Kr-85, and Xe using a purge gas stream of $5 \text{ m}^3/\text{h}$ (S.T.P.) at 1 bar and room temperature are shown in fig. 6. The bulk amount of Kr has been desorbed already after 30 minutes, after 2 hours only 1 % of the Kr originally adsorbed remains on the beds. If one would stop the purging step at this point, this residual Kr would be released on heating the bed in the regenerating step. It can be shown that a release of 14 % instead of 1 % residual Kr would still limit the release to the same amount which is released at the head of the first column at a column decontamination factor of 10^3 .

3.2 Xenon Product

The bottom product of the first column after transfer to the second column (packed bed) is separated by distillation into Kr (head product) and Xe (bottom product). This process step is only sensible, if storage volume of radioactive product can be saved. This is possible only, if the inactive Xe can be purified from Kr-85 to such an extent that it can be released to the atmosphere (or utilized commercially). If we postulate again that the amount of Kr-85 released with the Xe product should not exceed the amount released at the head

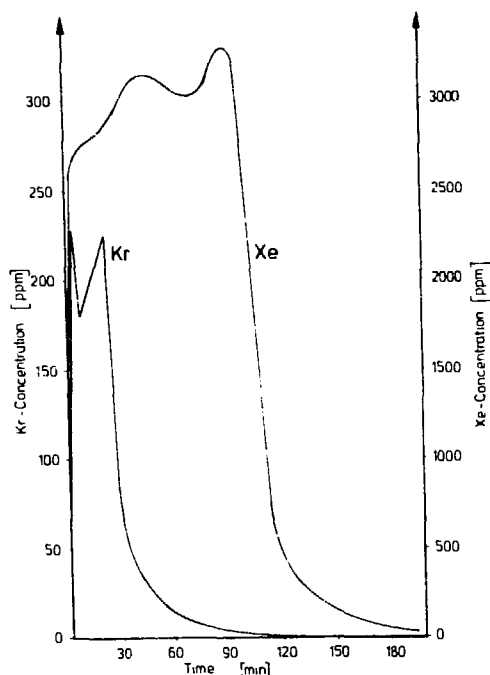


FIGURE 6

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

head of the first column at a $\text{DF} = 10^3$, then the Kr content of the Xe must be limited to 100 ppm by vol.

This postulate could be fulfilled almost routinely. At the moment, the Kr and Xe products are both collected in steel cylinders by means of a membrane compressor. Later on, transfer of the products into the cylinders by way of cryogenic pumping will be applied.

4. Catalytic Reduction of O_2 and NO_x

4.1 Poisoning of Noble Metal Catalysts

On the basis of laboratory experiments a Ru catalyst on Al_2O_3 carrier of relatively low specific surface area ($9 \text{ m}^2/\text{g}$) has been selected as catalyst for the reduction of O_2 and residual NO_x with H_2 . From all noble metals investigated, this catalyst is most resistant to poisoning by iodine and organic phosphorus compounds which are both trace components of reprocessing offgases. The poisoning effect of I_2 was studied with respect to activity and selectivity on a series of Pt, Pd and Ru catalysts. Some of the results are shown in Fig. 7 and 8. The activity of Pt and Pd is definitely reduced by increased loading with I_2 . As can be expected, the activity of catalysts of high specific surface is more strongly affected than of

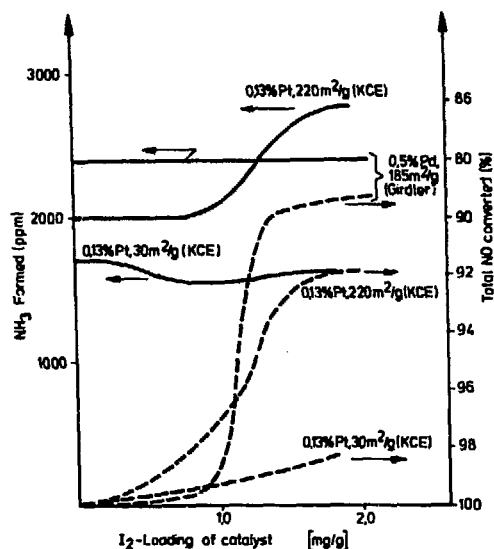


FIGURE 7

Poisoning effect of I_2 on Pt- and Pd-catalysts with respect to activity and selectivity of NO-reduction.

Inlet concentration: 0.75 vol-% O_2 ; 0.5 vol-% NO; 2.75 vol-% H_2 ;
Space velocity: $5000 h^{-1}$; $400^\circ C$.

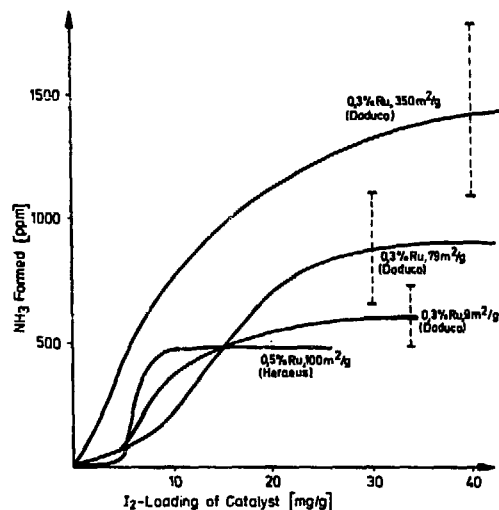


FIGURE 8

Poisoning effect of I_2 on Ru-catalysts with respect to selectivity of NO-reduction.

Inlet concentrations: 0.75 Vol-% O_2 , 0.5 Vol-% NO, 2.75 Vol-% H_2 ;
space velocity: $5000 h^{-1}$; $400^\circ C$.

types of lower specific surface area. The selectivity which is not good at the beginning (about 40 - 50 % NH_3 formed, the rest N_2), is not affected significantly, however (21).

On the contrary, the activity of Ru catalysts is not influenced even by very high I_2 loadings: under the experimental conditions, NO is reduced throughout to a residual level of ≤ 1 ppm. A poisoning effect can only be observed in the selectivity: formation of NH_3 which is very low in the unpoisoned state (22), increases more or less strongly on increasing I_2 loading (fig. 8). Again, the influence of the specific surface area is obvious. The NH_3 formation never reaches the amount produced at Pt and Pd.

Thus, on the basis of these data, further work will be carried out with the Ru catalyst of low specific surface area. It can be shown that in a real offgas the time of operation of such a catalyst bed should not be limited below about one year by iodine poisoning, if an iodine filter with a DF of 10^3 or better is installed before the catalyst.

4.2 Formation of Methane

It is well known that Ru is an active catalyst for the reduction of CO_2 to CO and CH_4 (23). CO_2 is an essential component of the

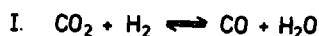
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offgas if air is the carrier gas. In addition, CO_2 is formed, if organic substances are burned on an oxidation catalyst, which is conventional practice in air liquefaction plants. Also, C-14 is set free from the fuel elements predominantly as $^{14}\text{CO}_2$. CO and CH_4 , if formed at the reduction catalyst, will not be retained at the adsorption bed and will reach the cryogenic column. According to their boiling points (81.7 K and 111.6 K at 1 bar, respectively) they will attain a certain equilibrium concentration there. For two reasons CH_4 formation must be avoided:

- it is a safety hazard in the presence of O_2 (or O_3);
- retention of C-14 will be inhibited.

The reduction of CO_2 by H_2 is governed thermodynamically by the equilibria given in table II (23). Whereas CO is formed preferably at higher temperatures ("water gas" reaction), CH_4 is formed preferably at lower temperatures ("Fischer-Tropsch" reaction).

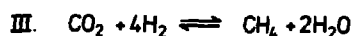
Table II: Reduction of CO_2 by H_2



$$K_p^{\text{I}} = \begin{cases} 0,067 & \text{at } 350^\circ\text{C} \\ 0,017 & \text{at } 250^\circ\text{C} \end{cases}$$



$$K_p^{\text{II}} = \begin{cases} 2,62 & \text{at } 350^\circ\text{C} \\ 3,04 & \text{at } 250^\circ\text{C} \end{cases}$$



$$K_p^{\text{III}} = K_p^{\text{I}} \cdot K_p^{\text{II}} = \begin{cases} 0,18 & \text{at } 350^\circ\text{C} \\ 0,05 & \text{at } 250^\circ\text{C} \end{cases}$$

The overall equilibrium constant is the product of both K_p^{I} and K_p^{II} shifting reaction III to the right with increasing temperature. The experimental results show that at temperatures above 400°C substantial amounts of CO, but little CH_4 is formed, whereas at lower temperatures high yields of CH_4 are obtained depending strongly on the H_2 -concentration. Thus one requirement for the minimization of CO_2 -reduction is to keep the overstoichiometric H_2 amount with respect to

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the reduction of O_2 and NO_x as low as possible.

As a matter of fact, the presence of O_2 suppresses the formation of CH_4 drastically, the overall atmosphere still being reducing. If O_2 is added to a gas mixture containing only CO_2 and H_2 in N_2 , the formation of CH_4 immediately drops down below the level of detection (20 ppm by gas chromatography). Equilibrium III (table II) is shifted to the left by a decrease of the H_2 - and an increase of the H_2O -concentration.

It can be concluded from these results that in a realistic offgas formation of CH_4 at the reduction catalyst can be controlled to such an extent that it poses no serious problems from a safety standpoint.

Acknowledgements

We acknowledge gratefully the assistance of the following co-workers and colleagues carrying out the experiments and calculations: W. Bumiller, G. Franz, E. Hauß, G. Kimmig, G. Knittel and K. Schulz. We also thank E. Henrich, C.H. Leichsenring, R.-D. Penzhorn and W. Weinländer for stimulating discussions.

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SESSION VIII

DAMAGE CONTROL

Tuesday, August 8, 1978

CHAIRMAN: W. L. Anderson

IN-DUCT COUNTERMEASURES FOR REDUCING FIRE-GENERATED-SMOKE-AEROSOL EXPOSURE TO HEPA FILTERS
N. J. Alvares, D. G. Beason, H. W. Ford

AIR CLEANING SYSTEM ISOLATION DURING TORNADO CONDITIONS
W. A. Pysh, T. A. Clements

EVALUATING NUCLEAR POWER PLANT VENTILATION SYSTEM ADEQUACY IN REDUCING AIRBORNE RADIOACTIVITY EXPOSURE
W. E. Jouris

AIR CLEANING SYSTEMS ANALYSIS AND HEPA FILTER RESPONSE TO SIMULATED TORNADO LOADINGS
W. S. Gregory, R. W. Andrae, K. H. Duerre, H. L. Horak, P. R. Smith, C. I. Ricketts, W. Gill

OPENING REMARKS OF SESSION CHAIRMAN:

This session will address several aspects of damage control of particulate filtration devices utilized in nuclear installations. Four papers are included on the agenda; however, these will address only two of the many threats for potential system damage. Two of the papers will describe the determination of pressure differential under tornado conditions and the response of the various filtration/protective components to these conditions. A third paper addresses potential countermeasures against fire conditions and fire generated aerosols, while the fourth will describe a total evaluation of system adequacy for reducing airborne exposure.

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Dup

IN-DUCT COUNTERMEASURES FOR REDUCING FIRE-GENERATED-SMOKE-AEROSOL EXPOSURE TO HEPA FILTERS *

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Abstract

An experimental program was conducted to assess the endurance and lifetime of HEPA filters exposed to fire-generated aerosols, and to reduce the aerosol exposure by installing engineering countermeasures in the duct between the fire source and HEPA filters.

Large cribs of wood and other potential fuels of interest were "forcefully burned" in a partially ventilated enclosure. In a "forceful burn" the crib of fuel is continuously exposed to an energetic premixed methane flame during the entire experimental period. This tactic serves two purposes: (1) It optimizes the production of smoke rich in unburned pyrolyzates which provides severe exposure to the filters, and (2) It facilitates the ignition and enhances the combustion of cribs formed with synthetic polymers.

The fuel cribs vary in size but have similar weight (~ 200 kg). The crib elements were natural and synthetic polymers; i.e., polyvinyl chlorides, polymethyl methacrylate (fire retarded), polycarbonate, dense fiberboard, wood, or combinations thereof. These elements are approximately 16 cm^2 in cross sectional area by 90 and 120 cm in length. The elements are either closely packed where the spacing dimensions are identical to the element cross section, or relatively loosely packed where the elements are supported by a metal framework. Some of these materials required a metal framework with a wire screen "floor" at alternate layers to prevent sagging. Wood cribs supported by the same type of framework burned at a faster rate, indicating a somewhat greater lateral crib porosity.

The experiments were conducted in an enclosure specifically designed and instrumented for fire tests. The test cell has a volume of 100 m^3 and includes instrumentation to measure the internal temperature distribution, pressure, thermal radiation field, flow fields, gas concentration, particulate size distribution and mass, fuel weight loss, inlet and exit air velocities, and smoke optical density.

The countermeasure techniques include the use of passively operated sprinkler systems in the fire test cell, of fine and dense water scrubbing sprays, and of rolling prefiltration systems in the exit duct of the fire test cell. Of the countermeasures surveyed, the rolling prefilter system showed the most promise, since it extended HEPA filter lifetimes by factors of two or more.

This paper will concentrate on the effect of control variables; i.e., enclosure air supply, fuel composition and crib porosity, and on the combustion response; i.e., crib burning rate, enclosure temperature rise, oxygen consumption, and CO , CO_2 and total hydrocarbon production. A discussion of the attempts to rationalize smoke aerosol properties, i.e., aerosol mass-and-size distribution on filter plugging phenomena will be included along with results from the effect of countermeasure application on HEPA filter lifetimes.

*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract No. W-7405-Eng-48.

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I. Introduction

An unwanted fire in a space containing radioactive materials can compromise containment measures more dramatically than any threat other than explosion as fire can reduce solids and liquids to aerodynamic aerosols and provide the convective flow to distribute them. The methods by which fire defeats standard containment devices and practices include:

- Overpressurizing the space by generating more gas than can flow through the ventilation.
- Overpressurizing the space by plugging the HEPA filters in exhaust ducts with smoke aerosol.
- Breaching the filter by temperature degradation of the filter media.
- Penetrating the filter with evaporated volatile pyrolysis fractions.

We have attempted to identify through testing some practical and cost-effective countermeasures that will, for a reasonable period, protect HEPA filters from the products of such fires. While a normal HEPA filter lasts from 6 to 24 mo depending on exposure conditions, the duration of exposure prior to HEPA plugging or high-temperature breaching during a severe and smoky fire may be reduced to 5 min. Exposure duration is controlled by the type and geometry of the fuel and by the ventilation patterns of the enclosure. Thus, the device or procedure ultimately used as a countermeasure to protect HEPA systems from potentially high smoke and heat exposure must be capable of increasing the smoke exposure filtration so it lasts through the fire brigade attack and until the source fire is controlled.

The primary purpose of our tests then is to make smoke; this we do in the test cell shown schematically in Fig. 1. We described this test cell and its capabilities in detail in the 14th Conference⁽¹⁾ along with details of some preliminary

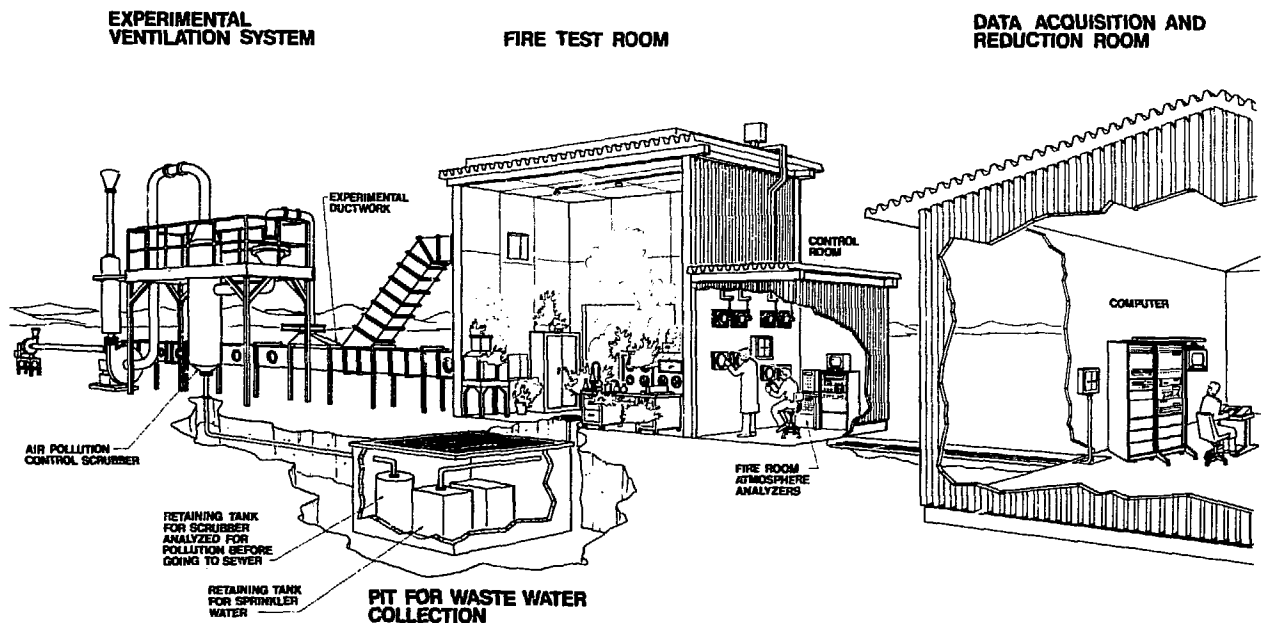
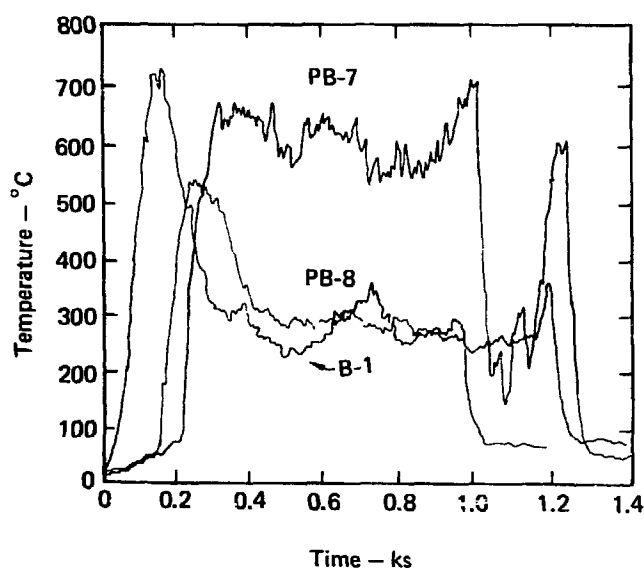


Fig. 1. Full-scale fire test facility.

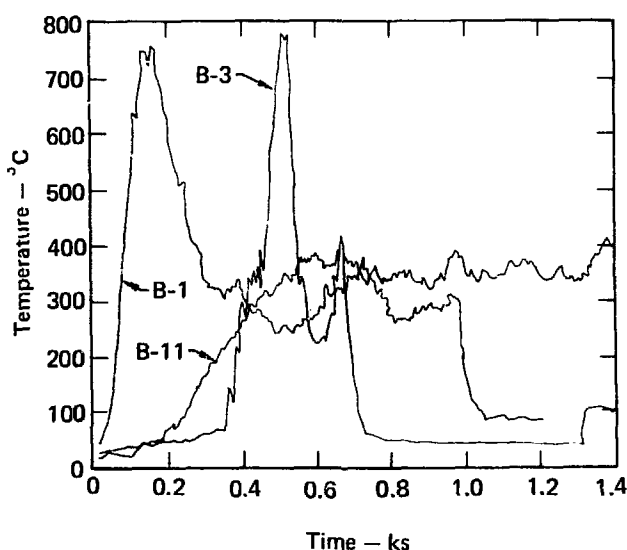
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The effects of crib spacing and ventilation rate on crib burning are shown in Fig. 3. The differences in time to peak burning for PB-8 and B-1 illustrate the effect of crib porosity. However, they exhibit a similar trend due to the effects of ventilation control of the combustion processes; that is, temperature reduction after a high intensity phase was indicative of oxygen starvation in tests PB-8 and B-1. In test PB-7, however, the ventilation rate was sufficient to allow active combustion over the entire test period.

Figure 4 indicates how the characters of different burning polymers compared to that of a fir wood crib. The B-3 curve illustrates the behavior of a fire-retarded PMMA* by its temperature response; although it takes a relatively long time to ignite, once ignited it burns with fierce intensity. The combustion response of polycarbonate is shown by the B-11 labeled curve. Once ignited, polycarbonate burns steadily with moderate heat and smoke release.



B-1 $\Rightarrow$ open crib, 250 l/s
PB-8 $\Rightarrow$ closed crib, 250 l/s
PB-7 $\Rightarrow$ closed crib, 500 l/s



1 $\Rightarrow$ Firwood crib, 250 l/s
B-3 $\Rightarrow$ PMMA - FR crib, 250 l/s
B-11 $\Rightarrow$ Polycarbonate, 259 l/s

Fig. 3. Effects of crib spacing and ventilation rate on crib burning.

Fig. 4. Comparison of the characters of burning polymers to that of a fir wood crib.

Figure 5 illustrates the selected fir and airflow parameters we surveyed during each test. Burn 29 was a supported wood crib (i.e., the standard fuel array used for correlation tests). The displayed data are: plume temperature ($T^{\circ}\text{C}$), oxygen concentration [O_2], burning rate or fuel consumption (m), airflow through the ventilation duct (l/s), and pressure drop across the HEPA filter (ΔP HEPA). When airflow through the HEPA filter reached half the initial flow rate, indicated by the cross on the airflow curve, we considered the filter plugged and terminated the test. We conducted comparative analyses with data taken at the temperature peak and again just prior to significant flow reduction. (In Fig. 5, this would be approximately 400 s). Along with the indicated data, we measured total aerosol concentration and rough aerosol size distribution at the exit to the test cell and just upstream of the HEPA filter. Moreover, we used cryogenic techniques to trap grab samples of aerosol and combustion gas both up and downstream of the filter for chemical analysis.

* Polymethylmethacrylate

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tests in which we installed laboratory furnishings and appliances in the test cell as fuel items. We indicated a major problem with using such items in a quasi-normal array was the nonuniformity of the flame spread and, consequently, of the smoke production. What we needed was a reproducible but severe source of smoke so we could appraise the effects of various countermeasures to increase the smoke exposure lifetimes of HEPA filters.

Our solution was to employ cribs (cross piles of fuel) a fuel array traditionally accepted in fire research. We selected our fuel elements from materials abundant in laboratories where radioactive materials are used or stored. We tested several crib arrangements and fuel element distributions, finally deciding on a series of configurations particular to various fuels that would provide a moderate fuel load (1 to 2 lb/ft²) for the test cell. Figure 2 shows three crib arrays: the top plate shows a natural crib of wood with element dimensions of 1.5 in. on a side and 3.5 ft in length. This crib has a relatively low porosity (i.e., very close element spacing), which reduces the burning rate of the fuel. The second plate shows a crib of wood whose alternate layers are supported on a metal framework. The framework supports plastic fuels that soften and flow during flame exposure, as shown in the third plate of Fig. 2. Since a significant proportion of our fuels are synthetic polymers, we have opted to contain all fuel elements in the same type of support grid. This geometry results in a somewhat higher porosity than the natural crib; hence, the burning characteristics are accelerated.

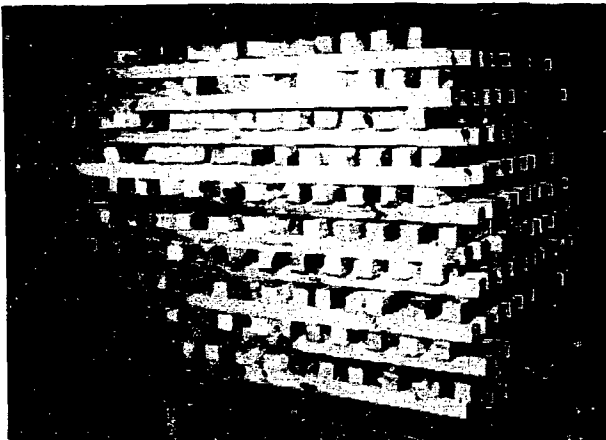


Plate 1

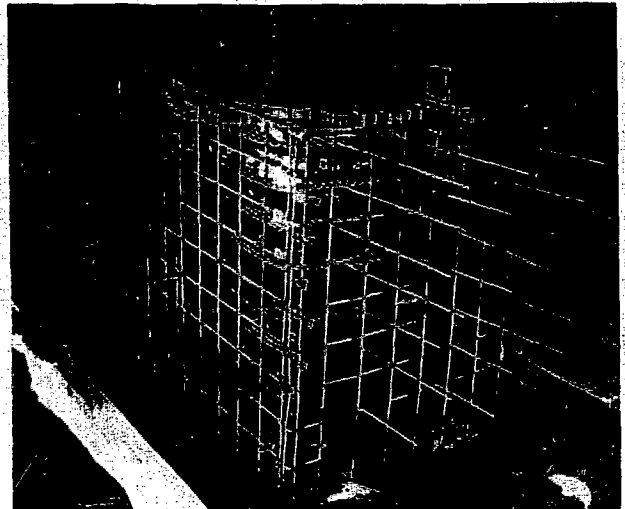


Plate 3

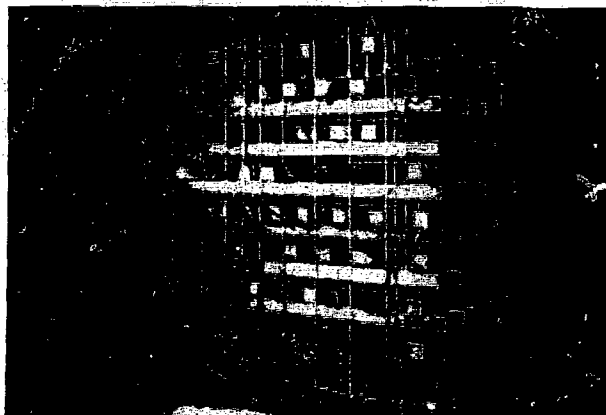


Plate 2

Fig. 2. Crib arrangements for fire tests.

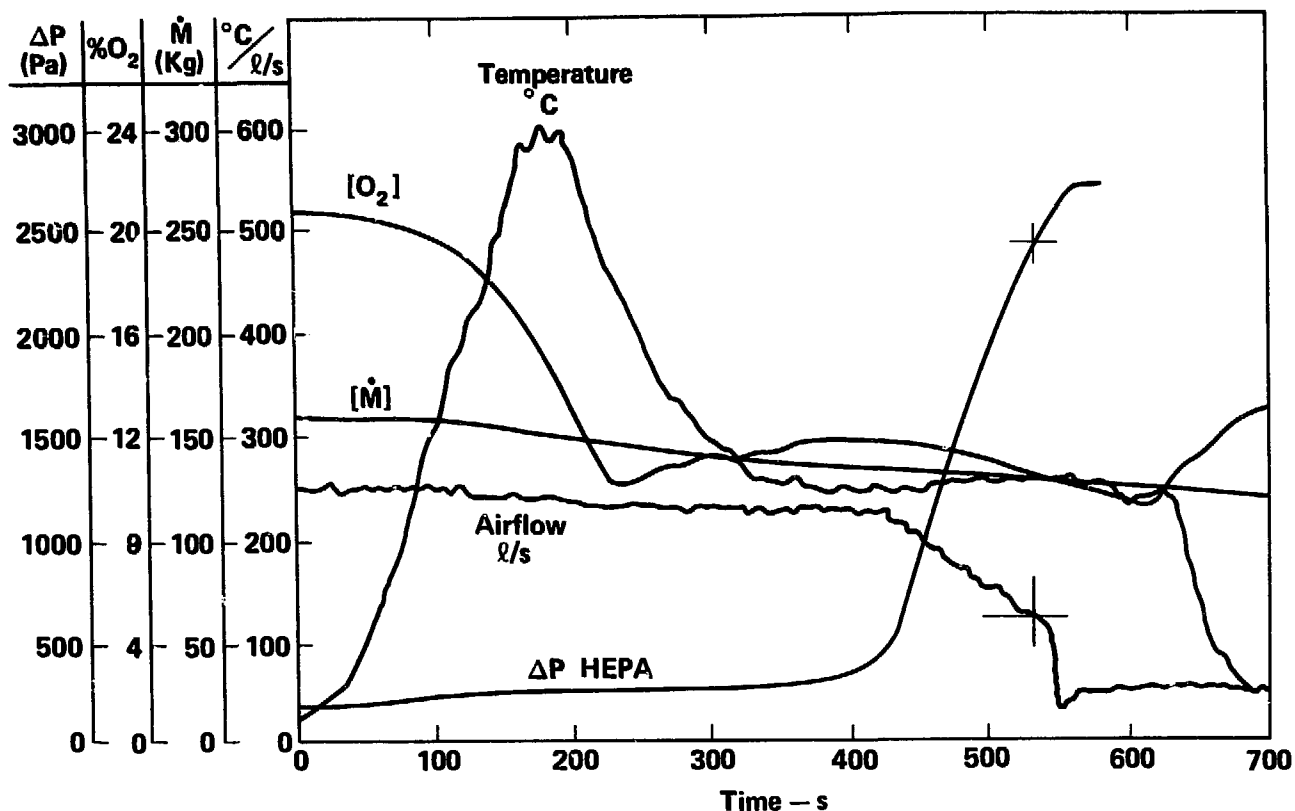


Fig. 5. Selected fire and airflow parameters.

II. Measurements

Because our ignition burner continues to burn throughout the exposure, our crib fires very likely produce more smoke aerosols than would a free-burning crib. For the same reason, the induction period to full burning intensity and, subsequently, to ventilation control is shorter. We are thus able to describe and control the dynamic aspects of our fire source with some degree of success; that is, given the material and a description of the crib parameters, we can approximately predict the combustion response. However, we have been totally unsuccessful in our attempts to characterize smoke aerosols. We can gain some idea of the magnitude of excess pyrolyzate* by comparing the measured weight loss, $\dot{m}$, to the fuel consumption potential as calculated by the oxygen consumption during a stoichiometric combination of fuel and oxygen to products⁽³⁾. Table 1 shows this comparison for some early tests and indicates an order-of-magnitude estimate of the unburned pyrolyzate, which, in almost all cases, is of the same magnitude or of greater magnitude than the burned fractions.

Our attempts to measure the size and mass distribution of smoke aerosols were fraught with complications, mainly because of the high aerosol concentration as indicated in Table 1. Moreover, the chemical and physical interactions in dense aerosols from any source made such measurements almost impossible. Kinetic processes such as agglomeration, evaporation, deposition, gradation, fallout electrostatics, chemical change, and adsorption caused the aerosol to change its character during both sampling and measurement procedures. Regardless of these complications, we took a continuous series of aerosol mass and mass distribution measurements using cascade impactors positioned near the exit to the test cell and at the upstream surface of the HEPA filter. Table 2 summarizes these measurements.

* Partially oxidized pyrolysis fractions that can occur during ventilation-limited combustion processes.

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Table I. Gross estimates of excess pyrolyzate.

Test	Material	Measured fuel mass loss, $\dot{m}_f$ (kg/s)	Calculated fuel consumption from oxygen depletion measurements, $\dot{m}_o$ (kg/s)	Gross excess pyrolyzate, $\dot{m}_f - \dot{m}_o$ (g/s)
PB-7	Fir wood ^a	0.13	0.086	44
B-1	Fir wood	0.12	0.041	89
B-3	PMMA-FR ^b	0.028	0.003	25
B-11	Polycarbonate	0.037	0.019	18
B-15	Dense fiberboard	0.080	0.049	31
B-17	Polyvinyl chloride	0.020	0.008	12
B-18	Fiber-reinforced polyester	0.020	0.020	0
B-20	Composite ^c	0.020	0.016	4
B-21	Fir wood ^d	0.10	0.043	57
B-28	Fir wood ^e	0.1/0.06 ^g	0.038	42 ^h
B-36	Fir wood ^f	0.097/0.058 ^g	0.089	8
B-38	Composite	0.05	0.023	27

^a 500-l/s air supply and low-porosity crib.

^b Polymethylmethacrylate — fire retarded.

^c Crib made with fir wood, PMMA-FR, polycarbonate, fiber-reinforced polyester, and polyvinyl chloride.

^d Low air intake and exhaust duct.

^e High air intake and exhaust duct.

^f 500-l/s air supply.

^g Varies with time for high air intake and exhaust duct tests.

^h Based on average $\dot{m}_f$.

TABLE II. Cascade impactor data

		Impactor at exit of test cell				Impactor at HEPA station			
Test	Material for crib	Conc, Kg/m ³	Gross mass distribution %			Conc, Kg/m ³	Gross mass distribution %		
			d > 3	d > 1	d < 1 ^a		d > 3	d > 1	d < 1 ^a
PB-7	Tight fir wood					0.001	82	2	16
B-2	Loose fir wood					0.005	3	60	37
B-3	PMMA-FR					0.001	3	44	53
B-11	Polycarbonate					0.0007	14	58	28
B-15	Dense fiberboard	0.002	12	71	17	0.002	6	67	27
B-17	Polyvinyl chloride	0.002	10	69	21	0.002	10	63	27
B-18	Fiber-reinforced polyester	0.001	61	21	18	0.002	56	15	29
B-20	Composite	0.006	42	57	1	0.005	25	72	3
B-21	Loose fir wood	0.013	62	32	6	0.009	49	23	28
B-28	Loose fir wood	0.006	68	25	7	0.002	89	u	11

^a d = Aerosol diameter in microns.

^a d = Aerosol diameter in microns.

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TABLE IV. Fir wood crib ventilation and fuel moisture variables

Test	Date	Ventilation Inlet / Exhaust	$\dot{m}$, a kg/s	t_{peak} , b s	t_{plug} , c s	$\frac{t_{peak}}{t_{plug}}$, s	T_{av} , d °C	T_{db} , e °C
B-2	7/76	Low / High	0.12	160	330	2.07	224	100
B-13	4/77	Leak / High	0.07	210	610	2.9	183	105
B-21	7/77	Low / Low	0.10	140	570	4.0	215	90
B-28	11/77	High / High	0.1 0.05	190	540	2.85	172	110
B-30	2/78	High / High	0.077 0.047 0.038	190	No plug	---	-160	120
B-32 (dry)	3/78	High / High	0.12 0.03	160	310	1.9	185	112

a $\dot{m}$ = burning rate.

b t_{peak} = time to peak temperature above crib.

c t_{plug} = time to HEPA plugging.

d T_{av} = average highest test cell temperature.

e T_{db} = dry bulb temperature at HEPA.

cellulosic fuels of high moisture content would burn at a much slower rate than the sensible removal of water from the cellulose would indicate⁽⁴⁾. To test this finding, we kiln dried enough wood for one crib load. The average moisture content of the dried elements was less than 6%. The results are shown in test B-32 where plugging occurred in 310 s. Obviously, control of the moisture content of porous fuels is very important to ensuring reproducibility in tests.

We have tried several countermeasures to keep the HEPA filter from plugging, including rolling prefiltration, increasing the HEPA surface, water scrubbing, stopping the fire with sprinklers, and controlling the materials. Table 5 summarizes our countermeasure experience and condenses the data given in Table 3 to specifically address the relative merits of different countermeasure techniques.

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The weighing protocol for most measurements listed in Table 2 called for holding the filter papers at 60°C for 24 hr before and after measurement. Obviously, we lost all volatile fractions that vaporize at temperatures below 60°C. Even so, the total concentration of solid-phase aerosol appeared to range around 0.002 kg/m³ (2 g/m³). Note, that while we indicate size range, we view these data with reservation. Our total mass measurements of solid-phase aerosol are probably as reliable as anyones.

We knew that by drying the filter after aerosol sampling, we were losing some proportion of the actual in duct aerosol mass. We determined this by sealing the impactors after sampling and by weighing the filters as soon as possible after terminating the test. Then we dried the filters at 60°C and reweighed them. The difference in total mass between the initial and final weighing was a factor of 6 at both the test cell exit and the HEPA exposure surface. Subsequent measurements indicated mass distribution shifts from large ($d > 3\mu$) to small ($d < 1\mu$) particles - an expected trend.

III. HEPA Filter Performance

Table 3 summarizes HEPA filter performance for all tests conducted during this experimental series. Included in the table are fuel consumption ($\dot{m}$), oxygen depletion ($[O_2]$), time to filter plugging (t_{plug}), dry bulb temperature at the filter (T_{DB}), measured total aerosol mass at t_{plug} and calculated excess pyrolyzate. These data show the remarkable ability of HEPA filters to perform well even when exposed to very dense aerosol loads. The shortest time to plug, for an unprotected filter was 320 s, and the measured maximum aerosol mass was 7 gm/m³ (measured from a dry impactor filter array). Since the flow was 250 l/s, the imposed aerosol load was at least 1.75 gm/s and was quite likely much more.

An aspect that modified filter performance was the temperature level at the HEPA station. For dry bulb temperatures of 120°C or higher, the time to plug was significantly longer; in some cases at high temperatures, the filter did not plug. It appeared that some major plugging components of the smoke aerosols remained in gas phase at temperatures greater than 110°C and, consequently, were not adsorbed by the HEPA. This observation was supported by the fact that condensed vapors were observed leaving the exit stack when high-temperature conditions were maintained in the duct and by the chemical determination of hydrocarbon compounds from grab samples taken downstream of the filter.

Table 4 shows that fuel condition and ventilation variables can also affect the HEPA's plugging performance.* The major variable here was supposed to be the ventilation pattern in the test cell, which was equipped with a high and low exit port and two high and low air-inlet ports. [We always use two air inlets (low or high) and one exit port (low or high)]. The first column lists the test number, date, and inlet/exit configuration. The data show that $\dot{m}$ did not vary drastically with the ventilation pattern except for test B-13, where we closed the inlet and found our test cell leaked almost as well as our inlet ducts. [We have subsequently sealed all leaks.]

We also noted that with the Hi/Hi ventilation configuration, the $\dot{m}$ measurement appeared to have a variable character. Most baffling, however, was test B-30; here the filter did not plug. True, the T_{DB} reached 120°C, but we had plugged filters with this condition before. On checking our fuel, we found the wood elements had a 20% average moisture content. Our literature search indicated

* These data are from wood cribs only.

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Table III. Review of test data and HEPA filter response for May 28, 1976 to July 15, 1978.

Test	Fuel	Geometry	Countermeasure	Ventilation rate (f/s)	Ventilation geometry Inlet/Outlet	Rate of measured fuel mass loss, ml (kg/s)	Oxygen depletion (%)	Dry bulb temperature at HEPA, T _{db} (°C)	Time to plug, t _p (kg)	Calculated excess pyrolyzate (g/s)	Measured aerosol mass (g/m ³)
PB-7	Fir wood	Nonporous crib	Free burn	500	Low/High	0.13	15.6	185	0.90	44	1.0
PB-8	Fir wood	Nonporous crib	Free burn	250	Low/High	0.09	14.5	90	0.70	50	12.0
PB-8.2	Fir wood	Nonporous crib	Free burn	250	Low/High	0.10	15.0	100	0.70	59	5.0
B-1	Fir wood	Supported crib	Free burn	250	Low/High	0.13	15.0	100	0.32	89	7.2
B-2	Fir wood	Supported crib	Free burn	250	Low/High	0.12	16.0	100	0.37	76	4.6
B-3	PMMA-FR	Supported crib	Free burn	250	Low/High	0.028	1.6	50	0.40	25	4.1
B-4	PMMA-FR	Supported crib	Free burn	500	Low/High	0.11	7.4	110	1.45	85	1.4
B-5	Diesel oil	3-ft pan	Free burn/scrubbers	250	Low/High	—	—	—	—	—	—
B-6	Fir wood	Supported crib	Scrubber	250	Low/High	0.10	16.0	59	0.37	56	2.2
B-7	Fir wood	Supported crib	Scrubber	250	Low/High	0.11	17.0	31	0.38	63	3.5
B-8	Fir wood	Supported crib	Scrubber	250	Low/High	0.10	17.2	60	0.32	63	1.5
B-9	Fir wood	Supported crib	Fusing sprinkler	250	Low/High	—	12.0	100	No plug	—	—
B-10	PMMA-FR	Supported crib	Fusing sprinkler	250	Low/High	—	2.0	58	0.40	—	—
B-11	Polycarbonate	Supported crib	Free burn	250	Low/High	0.037	12.0	80	No plug	18	—
B-12	Polycarbonate	Supported crib	Fusing sprinkler	250	Low/High	—	4.5	65	0.55	—	—
B-13	Fir wood	Supported crib	Inlet dampers closed	250	Low/High	0.07	16.0	105	0.61	26	—
B-14	Fir wood	Supported crib	Free burn	250	Low/High	0.12	17.5	120	0.67	72	—
B-15	Dense fiberboard	Supported crib	Free burn	250	Low/High	0.08	17.9	110	No plug	31	1.7
B-16	Dense fiberboard	Supported crib	Fusing sprinkler	250	Low/High	—	3.4	75	No plug	—	—
B-17	Polyvinyl chloride	Supported crib	Free burn	250	Low/High	0.02	3.6	60	0.59	12	2.1
B-18	Fiber-reinforced polyester	Supported crib	Free burn	250	Low/High	0.02	12.7	90	0.88	—	1.6
B-19	Fiber-reinforced polyester	Supported crib	Fusing sprinkler	250	Low/High	—	3.5	80	No plug	—	—
B-20	Composite	Supported crib	Free burn	250	Low/High	0.02	8.0	100	0.40	4	6.4
B-21	Fir wood	Supported crib	Free burn	250	Low/Low	0.10	16.8	90	0.57	57	—
B-22	Fir wood	Supported crib	Free burn	250	Low/Low	0.08	18.2	95	0.76	30	—
B-23	Composite	Supported crib	Rolling prefilter	250	Low/High	0.07	12.2	45	No plug	36	13.7
B-24	Composite	Supported crib	Rolling prefilter	250	Low/High	0.07	14.0	35	No plug	32	3.3
B-25	Composite	Supported crib	Rolling prefilter	250	Low/High	0.06	14.0	—	No plug	22	—
B-26	Fir wood	Supported crib	Rolling prefilter	250	Low/High	0.08	19.0	70	No plug	28	16.8
B-27	Fir wood	Supported crib	Prefilter circuit only	250	Low/High	0.09	17.8	80	0.72	41	—
B-28	Fir wood	Supported crib	Free burn	250	High/High	0.1-0.06	13.8	110	0.44	42	27.0
B-29	Fir wood	Supported crib	Free burn	250	High/High	0.1-0.05	11.8	100	0.54	33	10.9
B-30	Fir wood	Supported crib	Free burn	250	High/High	0.077-0.034	13.0	120	No plug	19	4.2
B-31	Fir wood	Supported crib	Free burn	250	High/High	0.06-0.03	13.0	100	No plug	9	13.0
B-32	Kiln-dried fir wood	Supported crib	Free burn	250	High/High	0.12-0.03	14.8	112	0.31	34	3.0
B-33	Polycarbonate	Supported crib	Fusing sprinkler	250	High/High	—	4.5	70	No plug	—	—
B-34	Fir wood	Supported crib	Free burn	500	High/High	0.1	12.2	180	No plug	33	12.4
B-35	Fir wood	Supported crib	Free burn	500	High/High	0.025/0.054/0.035	18.9	180	No plug	0	25.0
B-36	Fir wood	Supported crib	Sonic cooling	500	High/High	0.097-0.058	15.0-11.0	140-75	0.64	8	16.2
B-37	Fir wood	Supported crib	500-f/s filter	250	High/High	0.085-0.42	12.2	105	No plug	30	19.2
B-38	Composite	Supported crib	Free burn	500	High/High	0.05	4.2	103	0.44	27	8.6

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The rolling prefilter appeared* to offer the most promise as an active control since it can be used for a dual purpose; i.e., enhanced filtration** and HEPA fire protection. A passive control strategy might be to simply double the size of the HEPA relative to the design air throughput.

Scrubbing by in duct spray techniques did not increase filter lifetimes appreciably; in fact, there is some indication that the cooling effects of sprays may enhance the condensation of volatile aerosols, thus making plugging more likely. Fire control sprinklers effectively applied to the crib appeared to reduce the intensity of the fire and consequently the smoke production. In only one case did the HEPA plug after sprinkler application. The fuel was polycarbonate and the effect may be propitious.

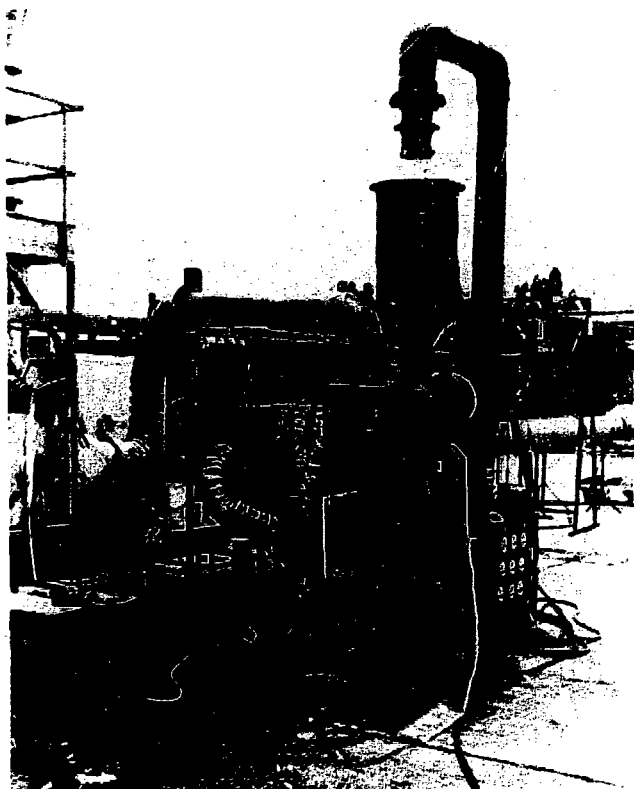


Fig. 6. Rolling prefiltration system installed in the LLL duct system.

IV. Summary and Conclusions

We have exposed HEPA filters to very severe smoke loads from potential fuel materials common in the furnishings and appliances of laboratories containing radioactive materials to identify and test practical filter plugging countermeasures before installing and retrofitting them in DOE facilities. Our tests indicated HEPA filters are inherently capable of handling huge aerosol concentrations

* In August of 1977, LLL rented a pilot model rolling prefiltration system and operator from the Anderson Corporation. The unit is generally used for filtration of air pollution aerosols and is based on high velocity trapping and scrubbing techniques. In our operations, we circumvented the scrubbing section. Figure 6 shows the unit installed in the LLL duct system. The HEPA was downstream from this unit.

** DOE's Enhanced Filtration Project is aimed at increasing the normal operational HEPA lifetime by unique prefiltration procedures.

TABLE V. Effect of various countermeasures on filter performance

Countermeasure	Fir wood		PMMA-FR		Polycarbonate		Dense fiberboard		Fiber-reinforced polyester		Polyvinyl chloride		Composite	
	t_{plug} , s	$\dot{m}$, kg/s	t_{plug} , s	$\dot{m}$, kg/s	t_{plug} , s	$\dot{m}$, kg/s	t_{plug} , s	$\dot{m}$, kg/s	t_{plug} , s	$\dot{m}$, kg/s	t_{plug} , s	$\dot{m}$, kg/s	t_{plug} , s	$\dot{m}$, kg/s
Free burn (enclosure)	320	0.12	400	0.03	No plug	0.04	No plug	0.08	880	0.02	590	0.02	400	0.02
Water scrubber (1-100 l/min)	350	0.11												
Fusing sprinkler	No plug	— ^a	No plug	— ^a	550	— ^a	No plug	— ^a	No plug	— ^a				
Rolling prefilter	No plug	0.08											No plug	0.07
Wet wood	No plug	0.03												
Large HEPA surface	No plug	0.035												
^a For most fires, sprinklers activation occurs before steady-state burning.														

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equal to the maximum rates from a flashed-over* room, however, the filters will ultimately plug. Attempts to control the fire by passive fire control sprinkler systems reduced the smoke load and generally the filter plugging potential of the aerosol. In the absence of a fire control system, prefiltration techniques appeared to hold a promising means of increasing filter endurance under extreme aerosol exposures. The simple expediency of doubling the filter size relative to the design flow rate of the ventilation system also appeared promising as a passive smoke aerosol countermeasure.

So far, none of our filters have been thermally damaged since the heat lost to the duct was sufficient to reduce the temperature of the combustion products to safe levels ($T_{\text{gas}} \leq 180^\circ\text{C}$). However, at gas temperatures greater than 120°C , some volatile components of the combustion products passed through the filter and condensed downstream. We are uncertain if this is a compromise of containment.

We are currently analyzing the chemical components of smoke aerosols responsible for filter plugging. Our gross physical measurements indicate that most fuels tested thermally degrade to specific aerosol mixtures that contain effective filter plugging components. Our efforts in this area will continue at a reduced rate since we intend to concentrate on practical countermeasure techniques unless we can identify a super and universal filter plugging component of all aerosols.

Our future efforts will be directed toward optimizing rolling prefiltration procedures and toward providing scaling guidelines for the inclusion of rolling prefilters in any size containment system.

V. References

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3. W.J. Parker, "An Investigation of the Fire Environment in the ASTM E-84 Tunnel Test," NBS Technical Report, pg 945, (Aug 1977).
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* In this paper we define "flash-over" as the condition when all combustible items in an enclosure are involved in fire.

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DISCUSSION

FREEMAN: I served on an ANSI committee which wrestled with the question, "When is a filter plugged?" Could you give me your rationale for the definition of plugged filter that you define as "When the flow rate is reduced by half"?

ALVARES: We grabbed it out of the air. It appears that when a filter gets to half of the design flowrate it is accelerating to a plugging mode at a very fast rate. It's simply arbitrary.

BURCHSTED: A further comment on this point; earlier results of a program in which filters were totally plugged (i.e., airflow went to zero) showed that time to total plugging was very rapid after airflow dropped to 50% of normal system flow. The value of 50% of flow must be 50% of system flow, not 50% of filter element design flow. A recommended practice for fire/smoke alleviation is to operate filter systems at 50% of nominal filter flow. Plugging, in this case, would be 25% of filter airflow capability.

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AIR CLEANING SYSTEM ISOLATION DURING TORNADO CONDITIONS

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Abstract

The occurrence of a tornado at the site of an operating nuclear power generating station creates unstable atmospheric conditions which can be detrimental to the proper operation of the plant air cleaning system. One means of mitigating or preventing this hazard is isolation of the system from the unstable environment by closure of the system openings during the unstable conditions. For maximum system protection the closure should be an automatic response to the adverse conditions themselves, without reliance on manual or remote sensing or signals.

The interaction of tornado characteristics with velocity/pressure sensitive isolation devices is modeled. Force and time relationships are investigated to evaluate and predict the interaction. Laboratory test apparatus is developed for simulating the flows and pressures induced by the tornado at system openings. Several isolation devices of various sizes are subjected to a range of simulated tornado conditions for observation and evaluation of response time, sensitivity, and dynamic closing forces. The model, apparatus, experimentation, and results are presented.

I. Introduction

It has become commonplace in recent years to protect nuclear power plant air cleaning systems from the effects of tornado induced flows and pressures by means of isolation valves or dampers which close automatically in response to those potentially damaging flows and pressures.

The justification for providing this protection may be based on the requirements of Regulatory Guide 1.117, "Tornado Design Classification", to control the release of radioactive materials and to provide for the safe shutdown of the plant; or on engineering economic analysis of the cost of protection versus the probability of the cost of repair or replacement of system components.

Current practice has been away from "administrative" control of isolation; that is, procedurally controlled manual or remote operation of shutoff devices, and toward the use of valves and dampers which close in direct response to the tornado induced flows or pressures. The effectiveness of such devices depends upon how quickly they respond to the tornado induced conditions, and to what extent this response limits the effect of those tornado induced conditions on the system components.

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It is the purpose of this study to attempt to begin to answer those questions. The laws of fluid mechanics shall be utilized to model and understand the interaction of the various devices with normal and tornado induced system flows and pressures. Tests will be performed to verify the validity of the model selected. Finally, system effects will be evaluated with isolation devices in place and compared to system effects with no isolation from tornado induced conditions.

II. Design Basis Tornado

Regulatory Guide 1.76, "Design Basis Tornado for Nuclear Power Plants" gives the characteristics for the Design Basis Tornado that must be considered by the system designer. For background in the development of this tornado model, the reader is directed to the several references listed. (2) (6) (7)

Those characteristics which have potentially damaging effects on the air cleaning systems are the high rotational and translational winds at the tornado perimeter (624 km/hr, 360 mph) and the very rapid drop in atmospheric pressure within the tornado vortex, (20.7 kPa, 3 psi, in 1-1/2 sec). These changes in ambient conditions are very short lived but take place very rapidly. For this reason, any device which is to provide protection from these conditions must be very responsive.

The possibility of very high winds at the perimeter and uncompensated deficits in the vortex, combined with the possible occurrence at either supply or discharge openings, presents a matrix of system effects which are shown in Table I.

Table I. System Effects

	Supply Openings	Discharge Openings
High Winds	Surges in	Reversal
Vortex Deficit	Reversal	Surges out

III. Isolation Devices

Protection against reversal is provided by the use of appropriately designed check valves or back draft dampers. Normally such devices have some mechanism, such as springs or counterweights, which initiate motion toward closure before actual reversal occurs.

Protection against surges is provided by means of velocity/pressure sensitive devices which allow flows in the normal operating range, but which close when above normal flow rates are encountered. This sensitivity is provided by a mechanism, such as a trip latch,

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spring, or counterweight, which holds the valve or damper open against the forces imposed by normal operation, but which is overpowered by the forces of excessive flow rates or pressure differentials, allowing, or causing the device to close.

Valves and dampers for this application may rely on different mechanics of interaction with the flow stream. Figure 1 shows a device with two separate centrally pivoted, totally unbalanced blades, which rotate in opposite directions to close. This device, identified as "Wing Blade" type in ANSI N509, operates from the transfer of stagnation pressures developed in the apex of the two blades to provide closing moment.

Figure 2 shows the damper in a closed position, isolating the system downstream.

For all types normal flows will cause a closing force which is resisted by the aforementioned mechanisms. However, once the normally expected range of these flows is exceeded, the devices move very rapidly to a closed position, isolating the system from further effect.

While it is quite common for tornado isolation devices to be mounted in ducts, with flow orientation either horizontal or vertical, a more typical installation is that of plenum mounting with missile barrier protection. Such an installation is shown in Figure 3.

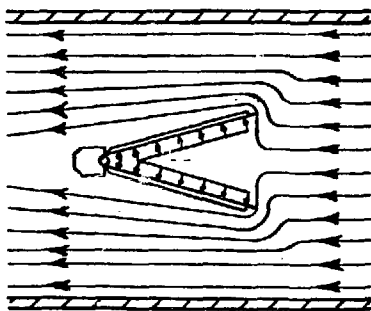


Figure 1 Wing Blade
Damper - open

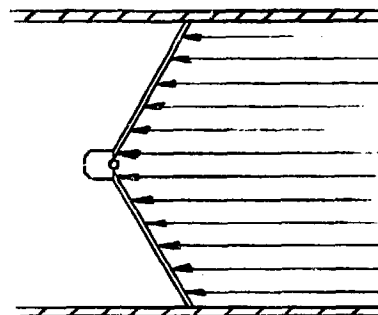


Figure 2 Wing Blade
Damper - closed

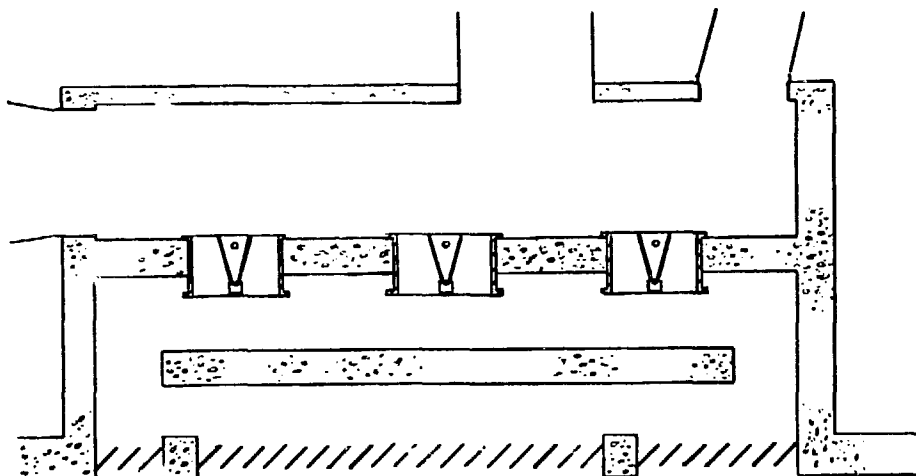


Fig. 3 Typical installation

IV. Analytical Model

A wing blade damper forms an obstruction in a flow path and, as such, can be analyzed from the mechanics of flow about an immersed object. (Figure 4) A pressure differential $P_0 > P_2$ will cause a flow to take place which will impinge on the upstream face of the obstruction, developing a stagnation pressure within the apex of the blades. For the flows and pressures of normal air cleaning system operation this pressure will be equal to the total pressure of the impinging air stream, TP_0 .

As the flow continues around the obstruction, there will be a loss of energy through the combination of eddy formation at the blade edges and friction loss along the downstream, or backblade surface. As the profile of the obstruction increases relative to the flow area (Figure 4b) the loss due to eddy formation will predominate, with most of the loss taking place at the blade edges.

If the blades are free to rotate about a hinge at the apex, which is the case for the wing blade damper, the stagnation pressure acts to move the blades in the direction of flow, while the static pressure distribution on the back side of the blades opposes this motion.

Given these flow induced forces, and the existence of gravity forces on the blades in vertical flow installations, the blades can be made to respond in a controlled manner to changes in the flow conditions by the addition of mechanisms which produce forces of known magnitude and direction, such as the previously mentioned latches, springs, or counterweights.

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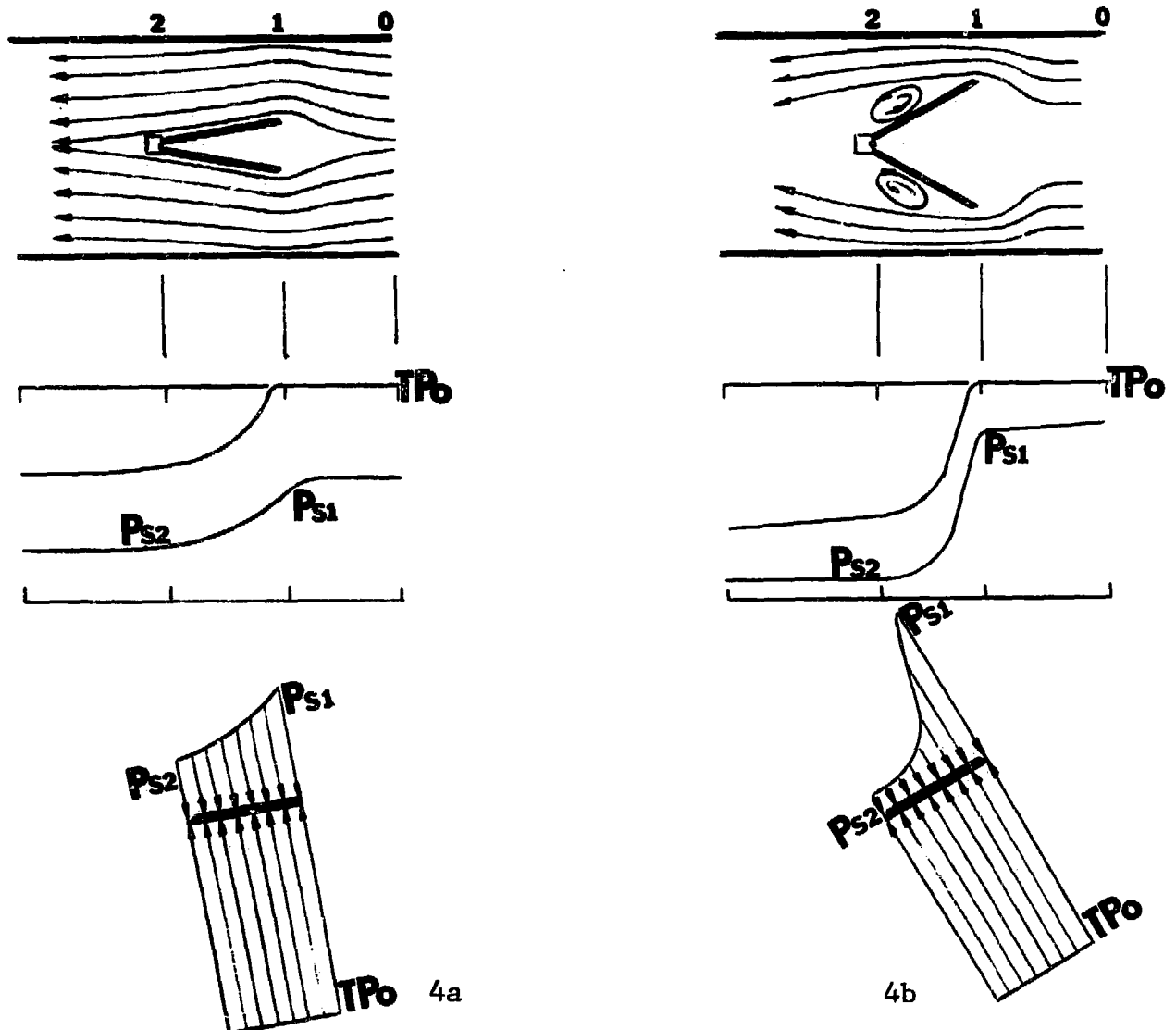


Figure 4. Flow lines, pressure gradients, and free body diagrams - duct flow

For example, if a force were added to Figure 4a equal to $-(TP_0 - \sum P_s)$ the blades would be in equilibrium, allowing flow from $P_0 > P_2$ to take place, as in normal operation. If $P_0 > P_2$ flow were vertical upward, gravity action on the blades would provide this force. As another example, if initial flow were $P_2 > P_0$, with the tornado causing a reversal to $P_2 < P_0$, a force could be added to initiate closing action before reversal occurred.

The resultant of all of these forces causes a moment which is given by:

$$M_R = (TP_0 - \sum_{P_{s1}}^{P_{s2}} P_s) A \bar{X} \pm M_G \pm M_F \quad (1)$$

Where M_G is the moment caused by gravity forces and M_F is the moment caused by the controlling forces F .

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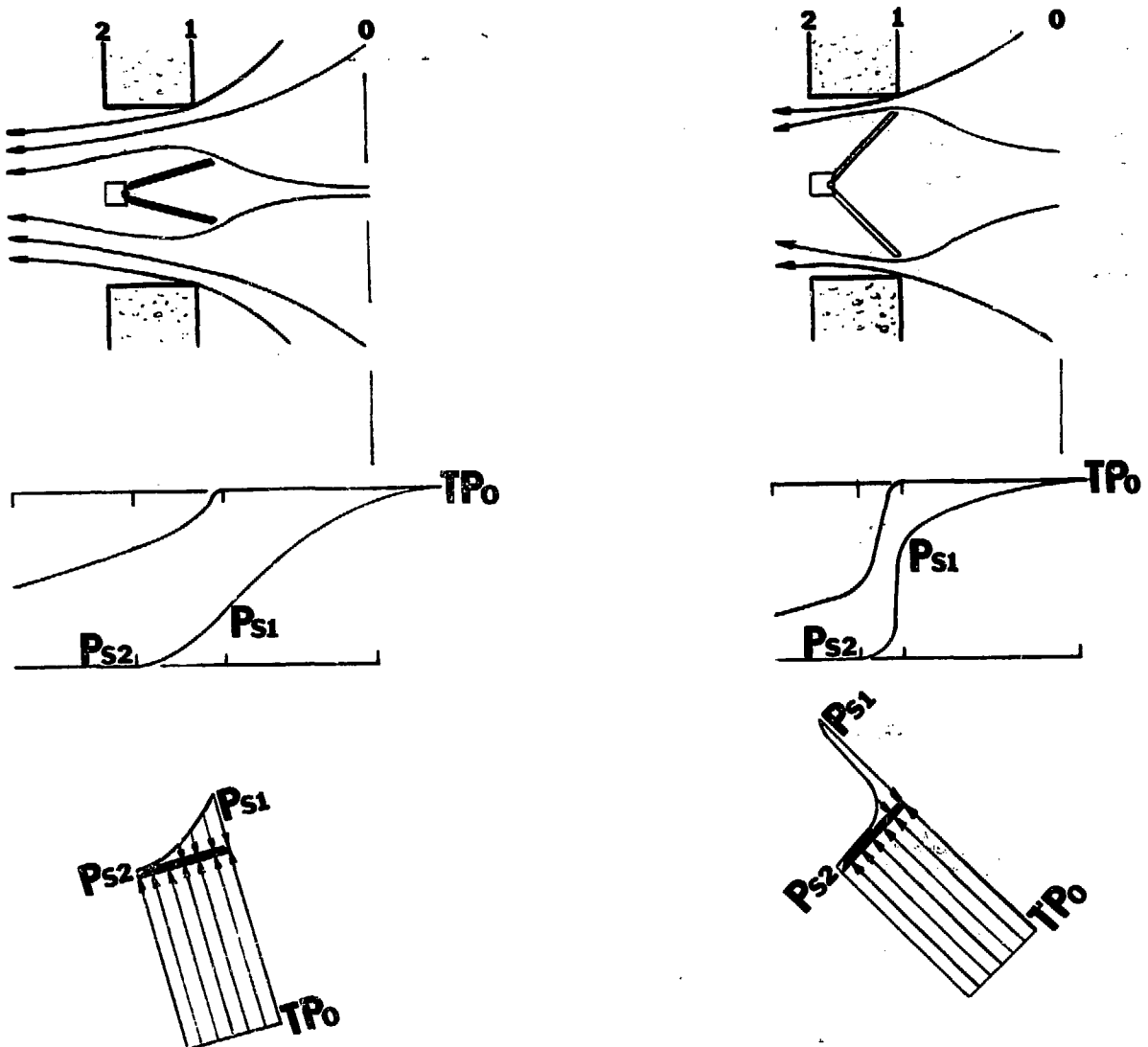


Figure 5. Flow lines, pressure gradients, and free body diagrams - orifice flow

Since A , $\bar{x}$, M_C and M_T are all known for a given application, it remains to determine the flow and pressure transients to which the isolation device must respond to provide the required protection. If an accurate and reliable analysis of the tornado effects on the entire system is available, the degree of protection required and the location of the protective devices will become apparent. Since such an analysis may not be readily obtainable, it may be possible to determine the maximum allowable transients by an evaluation of the limitations of the downstream components, such as filters.

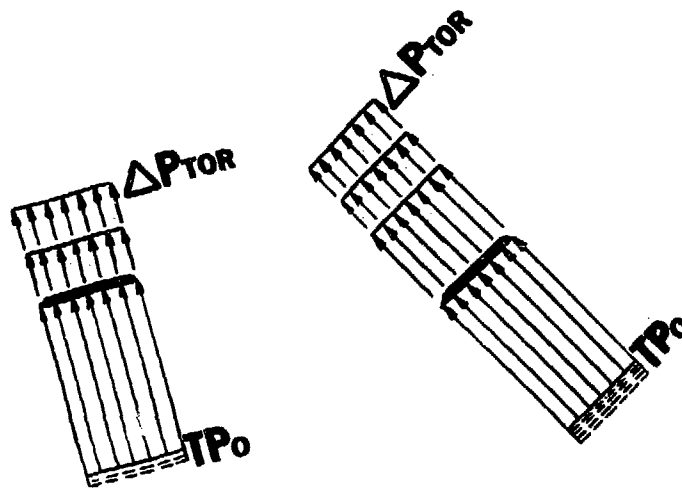


Figure 6. Assumed free body diagrams
orifice flow - during tornado pressure drop

More commonly, however, the decision is made to install the isolation valves and dampers as the first and last components in the system, and to require that they respond directly to the atmospheric conditions of the tornado. While this simplifies the transient solution, it is still necessary to evaluate the time rate of change of the normal system flows and pressures at the inlet and outlet openings.

Fortunately it is very common to have plenums at these openings, and this allows simplifying assumptions to be made. Such a case is essentially that of orifice flow between two large volumes. (Figure 5) For flow through an orifice the major energy loss occurs after the Vena Contracta, and therefore the total energy in the flow stream at (1) is equal to the total (static) pressures at (0) where the velocity is very low. For a wing blade damper located in the orifice, the stagnation pressure in the apex will be equal to the static pressure in the plenum for all normal flow rates. The static pressure distribution $\sum P_s$ will be small relative to TP_0 .

As a tornado induced drop in pressure P_2 occurs, the pressure distribution can be assumed to be that given by the Design Basis Tornado. If the storage capacity of the plenum is sufficient, outflows will have little effect on the static pressure for the short time interval involved and TP_0 can be assumed fixed (Figure 6).

Therefore, if $\sum P_s$ is assumed to be $\Delta P = f(t)$ and TP_0 is assumed constant, with other factors in equation (1) known, the motion of the blade can be defined.

It is this special case of orifice flow from a plenum that will form the basis for the first phase of the testing.

V. Testing

Static Tests

The first phase of testing consisted of utilizing existing laboratory apparatus to verify the static pressure relationships assumed in the model. The apparatus (Figure 7) is essentially that described in AMCA Standard 500-75, "Test Method for Louvers, Dampers, and Shutters."⁽⁸⁾ By varying the position of the control damper to establish "fan" curves and the position of the test damper blades to establish system resistance curves, a family of fan-system curves was created. (Figure 8)

At each point data was collected as shown in Figures 7 and 9. Data collection and adjustment procedures were as given in the AMCA 500 Standard. The force (F , Figure 9) was taken from a force gage connected to the test damper blade. All data was used directly except that the flow was determined by the P_v values at the pitot tube traverse, and the measured closing moment on the blades (M_{meas}) was calculated from a , F , and β .

The test chamber was a cube measuring 3.2 m (10 ft.) on a side, and the test damper was a 1.15 m (42 inch) wing blade damper, equipped with a trip latch which was capable of adjustment for release at varying flow rates. This arrangement was representative of a surge protection device located in a discharge plenum. Increases in flow rates would act to overcome the trip latch and cause closure.

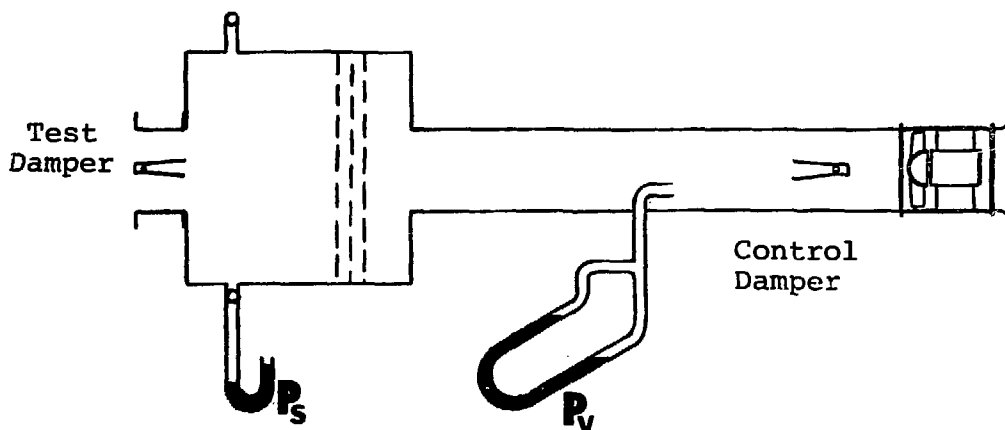


Figure 7 Test Apparatus

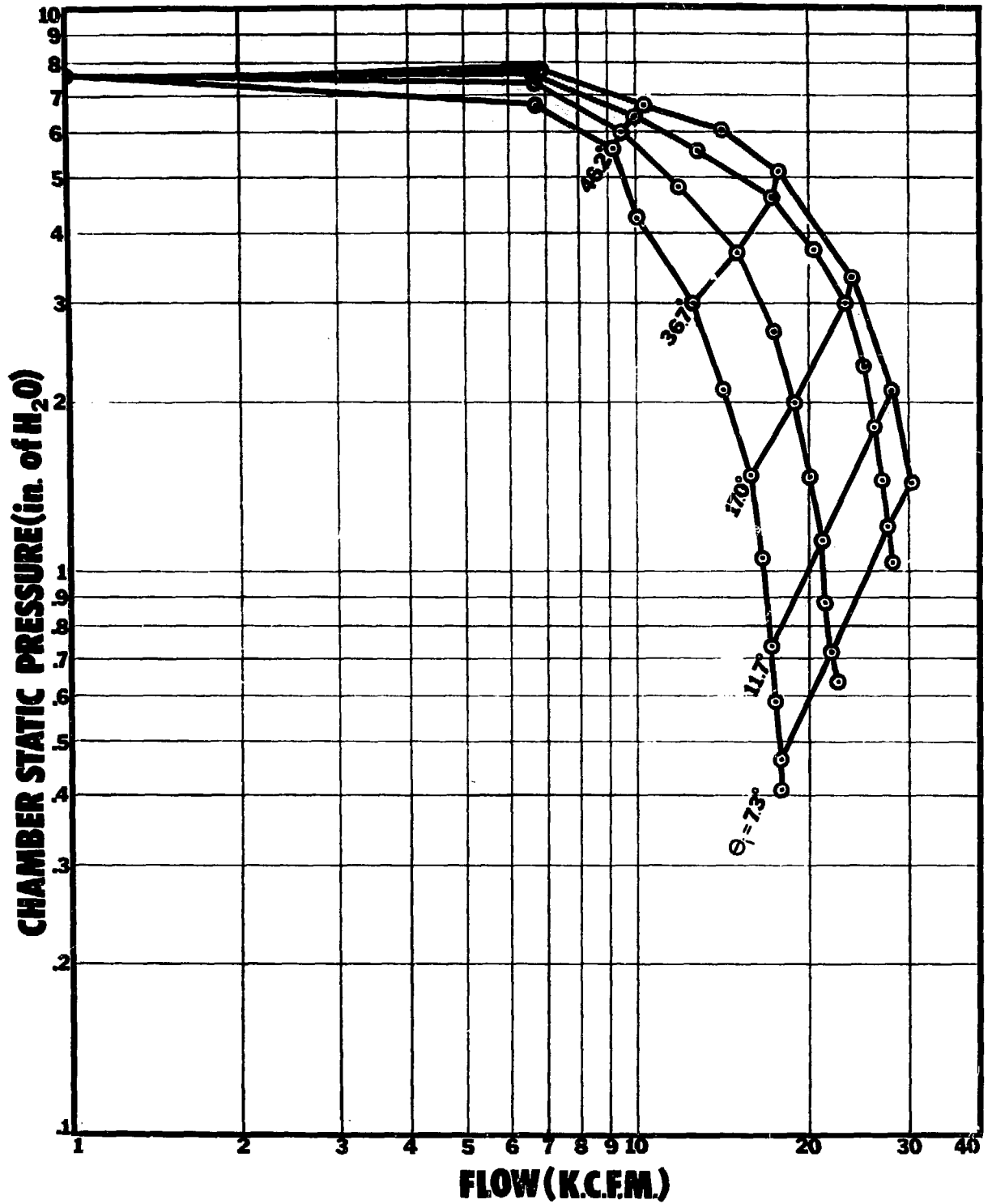


Figure 8 Fan-system resistance curves

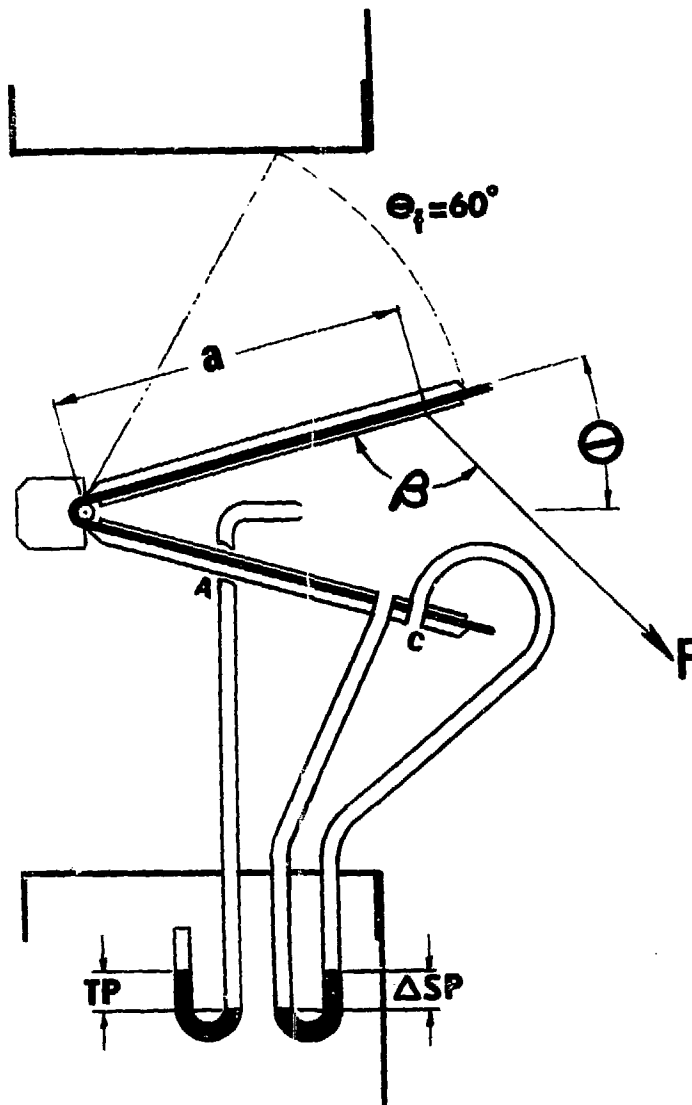


Figure 9 Data Collected

Verification of the assumed model for this arrangement consisted of comparing (1) the static (total) pressure in the chamber (P_s) with the stagnation pressure measured in the apex of the blades (TP) (2) the relationship between the unbalance pressures on either side of the blade at two locations (ΔSP_A , ΔSP_C); and (3) the closing moment calculated from the stagnation pressure (TP) alone acting on the blades (M_{calc}) with the measured moment (M_{meas}).

A comparison of TP and P_s at all data points showed the two values to be equal in all cases, with a maximum deviation of 0.02 inches W.G. The conclusion is that, for the arrangement and flow ranges included, the stagnation pressure in the apex of the blades was equal to the chamber static (total) pressure.

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Comparing ΔSP_A and ΔSP_C gives an indication of the static pressure profile on the downstream surface of the blades. It would be expected that the ratio $\Delta SP_C / \Delta SP_A$ would be small for a more open blade, where some friction drag would occur along the back blade surface, and would increase as the blade angle increased, when separation would occur at the blade edge. These expectations were confirmed by the data (Table II).

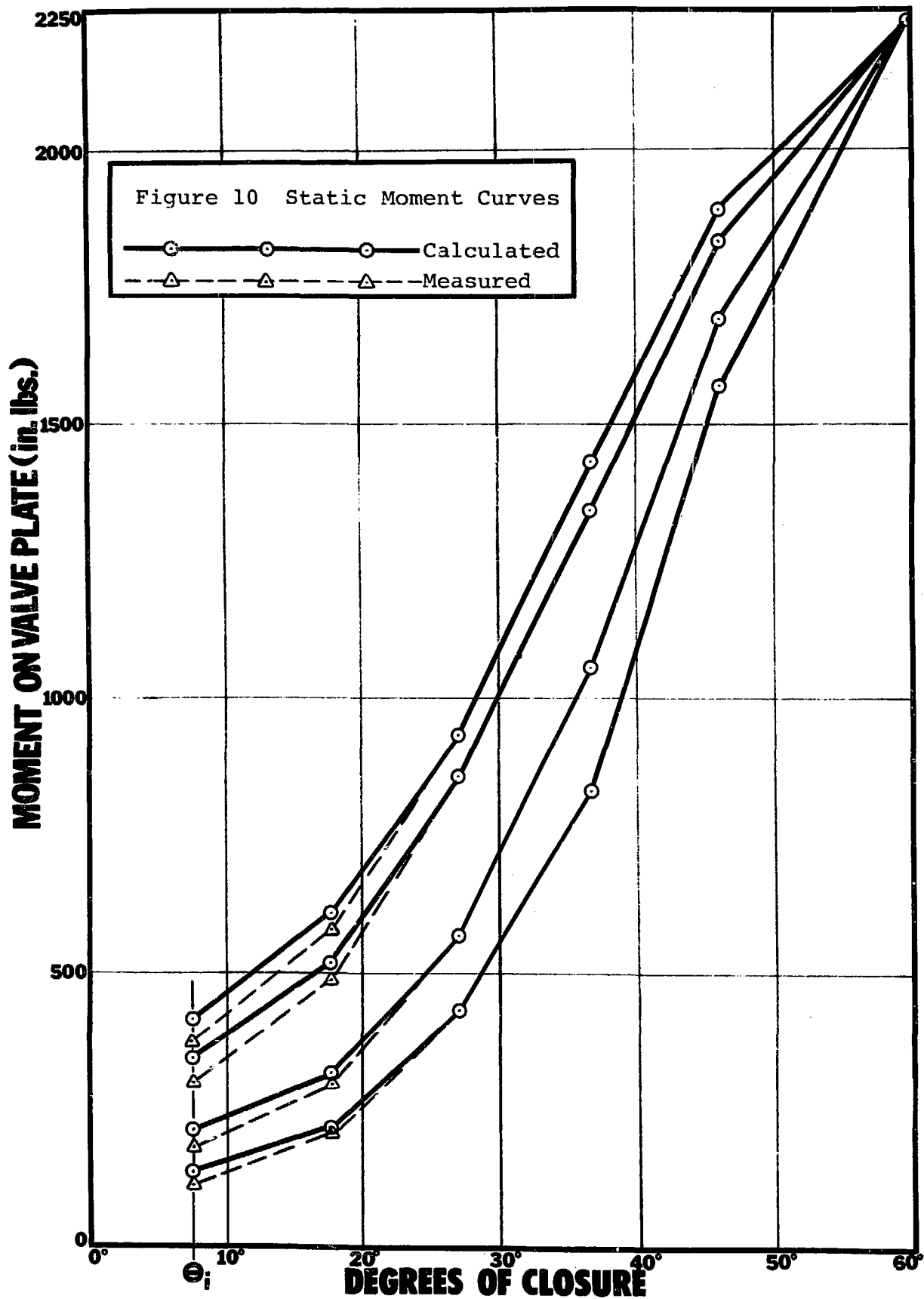
Table II. Pressure profile on back blade surface

Blade Position	$\frac{\Delta SP_C}{\Delta SP_A}$	Maximum Deviation
7.3°	.34	.02" W.G.
11.7°	.46	.01
17.0°	.67	.04
36.7°	1.00	.02
46.2°	1.00	.03

It is interesting to note here that, for a given blade setting, there was no significant change in the back surface profile within the flow ranges of the test series.

The final comparison to be made for the static model is between M_{calc} and M_{meas} . The model assumes for this arrangement, i.e., plenum flow to atmosphere, that the only significant force acting to close the blade is the apex pressure on the inside blade surface. This force would create a moment which is calculated from TP and blade geometry to give M_{calc} . The model acknowledges a static pressure distribution on the back surface but concludes that its effect is insignificant. The difference between M_{calc} and M_{meas} in the testing represents the significance of the back surface pressure distribution, and therefore, the suitability of the model assumption.

The results of the comparison (Table III, Figure 10) seem to substantiate the model assumption. While there is some variation shown at more open blade positions, the difference becomes very small with little blade movement.



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Table III. Blade Moments

Blade Position	$\frac{M_{meas}}{M_{calc}}$ (range)
7.3°	.85 to .93
11.7°	.94 to .99
17.0°	.98 to 1.04
36.7°	.96 to .99
46.2°	not available

Closing Time Tests

The same test apparatus was used to obtain closing times for the test damper when subjected to several different flow conditions. These closing times were then compared to that which would be expected from a quasi-static application of the damper model to those same flow conditions.

The experimental closing times were generated by setting the trip latch to release at a previously defined flow and measuring the travel time from release to closure by means of an electronic clock controlled by switches at the release and closure positions.

By setting the trip latch to release from the position θ_i , at the flow rate previously established for that position on each of the four fan curves used in the static test, the initial conditions of motion are given. The quasi-static assumption is that, as the blade moves toward the closed position, the flow will be governed by the fan curve, and the intersections of the fan-resistance curves will define the pressure relationships at that point.

The test apparatus and procedure give the pressure relationships as a function of position, rather than time, as would be the case for the Design Basis Tornado. However, the $M = f(\theta)$ data (Figure 10) is available to provide the time relationship.

The equation of motion for the blades is that of a mechanism rotating about a fixed point in one plane, and as such, is given by:

$$I_o \ddot{\theta} = M \quad (2)$$

For the quasi-static solution it is assumed that:

$$M = f(\theta) = K\theta + M_i \quad (3)$$

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Where K is the slope of the $M = f(\theta)$ curve and M_i is the moment at the trip point θ_i .

Combining equations (2) and (3) gives:

$$I_O \ddot{\theta} = K \theta + M_i \quad (4)$$

The general solution to this 2nd order differential equation is given by:

$$\theta = C_1 \cosh \left[\frac{K}{I_O} \right]^{\frac{1}{2}} t + C_2 \sinh \left[\frac{K}{I_O} \right]^{\frac{1}{2}} t - \frac{M_i}{K} \quad (5)$$

Therefore, given θ as the angle traveled to closure, K and M_i for the several fan curves (Figure 10), and I_O for the test damper, the predicted closing time, t_{calc} , can be determined.

The results of the comparison of predicted closing times (t_{calc}) and measured closing times (t_{meas}) for the test series (Table IV) indicates that the procedure used gives a reasonably reliable prediction of closing time.

Table IV. Closing Time

Trip Velocity	t_{calc} (seconds)	t_{meas} (seconds)
9.5 m/s (1870 fpm)	.29	.24
		.25
		.24
11.6 m/s (2290 fpm)	.24	.20
		.19
		.20
14.8 m/s (2920 fpm)	.19	.16
		.16
		.16
16.2 m/s (3190 fpm)	.18	.16
		.16
		.16

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VI. Conclusion

The use of automatic isolation devices to protect air cleaning systems from the effects of a tornado requires that these devices respond very quickly to the large, rapid, and short lived changes in atmospheric conditions that occur.

In the absence of a history of experience of plant reaction to tornado effects, the knowledge of behavior of the valves and dampers in response to the tornado induced transient must come from analytical and experimental effort.

Techniques are available for analytical solution of the equations of motion of the devices used, and also for determining the transient conditions within the system. The laboratory simulation of conditions imposed by a real tornado presents a considerable obstacle to experimental work in this area, however, some beginnings are underway.

From the work to date, it can be concluded that very rapid closure times are achievable. As an example, a wing blade damper, similar to the one used in the test, but of heavier construction, when mounted in plenum, discharging horizontally to atmosphere at 9.5 m/s (1870 fpm), and subjected to a 13.8 kPa/sec (2 psi/sec) atmospheric pressure drop, will close in 0.23 seconds, with a final angular velocity of 13.3 radians/sec. During the time of closure the total drop in atmospheric pressure would have been 3.45 kPa (0.5 psi).

This rapid closure response, and resultant reduction in system exposure to the unstable atmospheric conditions, provides the required protection to internal system components.

VII. Effect of Isolation

The predictability of response times has value only if it aids in the evaluation of system effects. There are two possible avenues for this evaluation.

One would be to design and construct a physical scale model of the system to be analyzed and impose a physical simulation of a tornado upon it. Using appropriate similarity and dimensional analysis techniques, the system effects could be evaluated.

Another approach would be to create a mathematical model of the system and impose a tornado simulation in like manner. A means to accomplish this exists in the form of a computer program developed at the Los Alamos Scientific Laboratory. This program, TVENT, has been designed for the specific purpose of modeling HVAC system interaction with events such as tornados. (1) (3) (4) (5)

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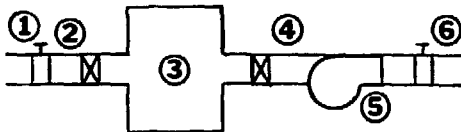
For purposes of illustration, TVENT was used to model a tornado interaction with a very simple system to observe the effects on the system with no isolation devices in place, and then again with isolation occurring.

The model system is shown in Figure 11, and the design basis tornado is shown in Figure 12. It should be pointed out at the outset that dramatic system effects are to be expected because the system is very small, with little natural attenuation, such as missile barriers and duct bends; and with very little system storage, so that small outflows will cause large pressure changes. Also it is to be noted that the tornado model used did not include the high winds surrounding the vortex.

Figures 13, and 14 show the system pressures and flows respectively with the DBT imposed at the system outlet.

For comparison purposes the program was repeated, this time using a controlled closure valve at node 5. The closing time was arbitrarily set at 0.5 seconds, with the resistance-time function as shown in Figure 21. The program instituted valve closure at a point when normal velocity was exceeded by 50%. The system effects for this condition are shown in Figures 15 and 16. Obviously there is a significant reduction in the pressures and flows encountered.

The above process was repeated with the DBT at the inlet with results shown in Figures 17 and 18 for no isolation, and Figures 19 and 20 with a check valve at node 1 to prevent reversal. The valve was instructed to prevent further flow once inlet flow value became negative. However, as the ambient pressure restored, the valve opened to allow positive flow once again. This action explains the very sharp increases around $T = 6$ seconds. It would not be unreasonable to expect such an action in a real system.



☒ Filter
☐ Valve

$Q_1 = 500 \text{ cfm}$
 $\text{Vol. 3} = 10,000 \text{ ft.}^3$

Fig. 11 Model system

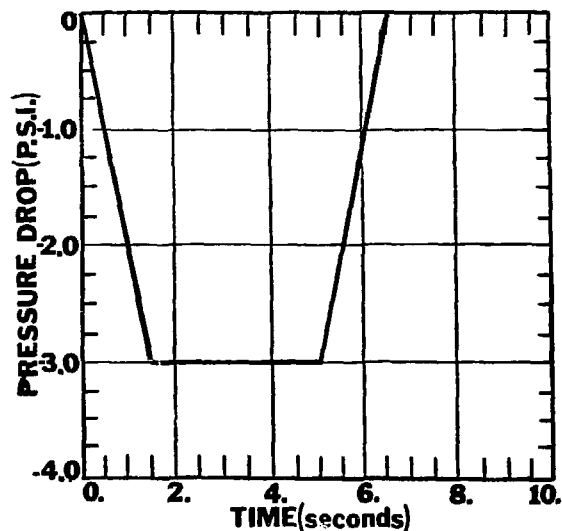


Fig. 12 Pressure Transient

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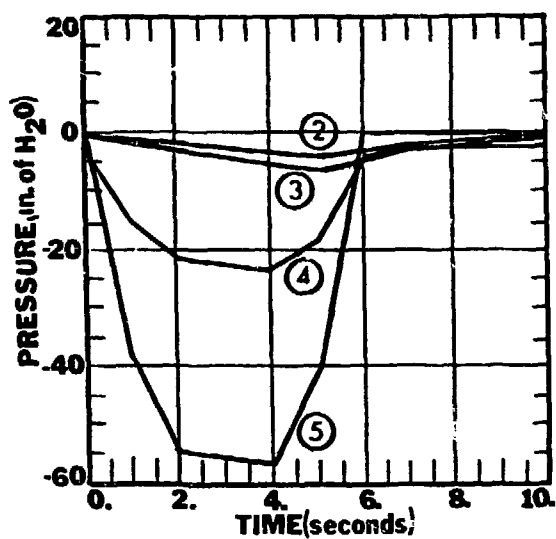


Fig. 13 System pressures - tornado at outlet

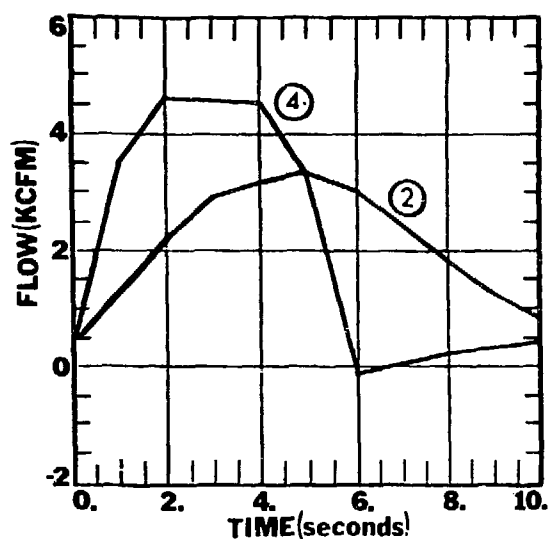


Fig. 14 System flows - tornado at outlet

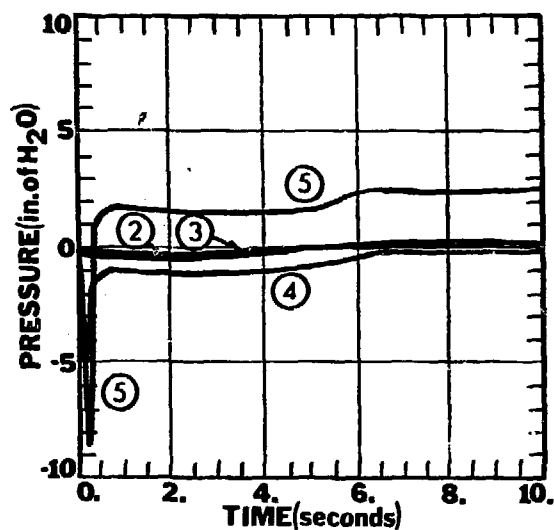


Fig. 15 System pressures - tornado at outlet with isolation valve

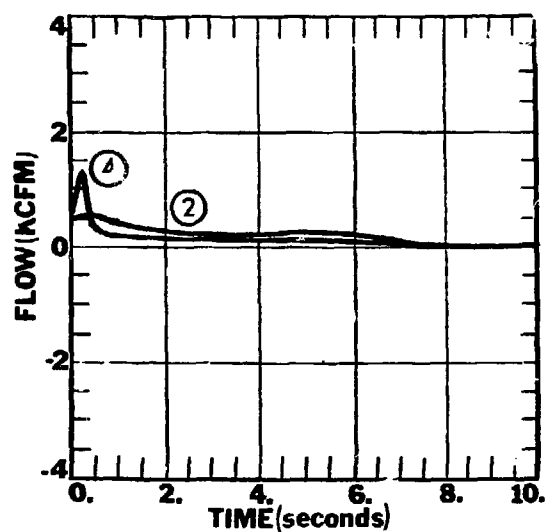


Fig. 16 System flows - tornado at outlet with isolation valve

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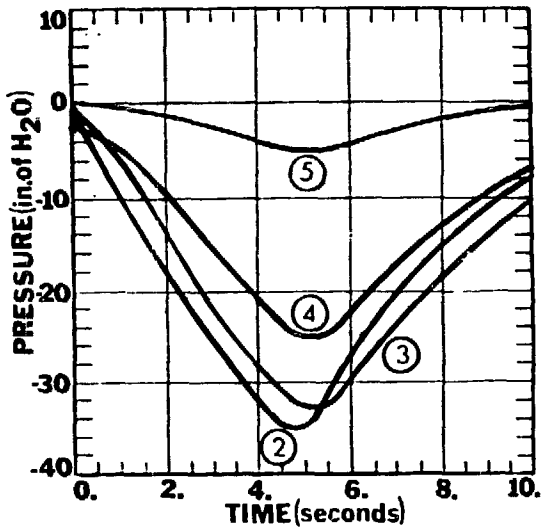


Fig. 17 System pressures - tornado at inlet

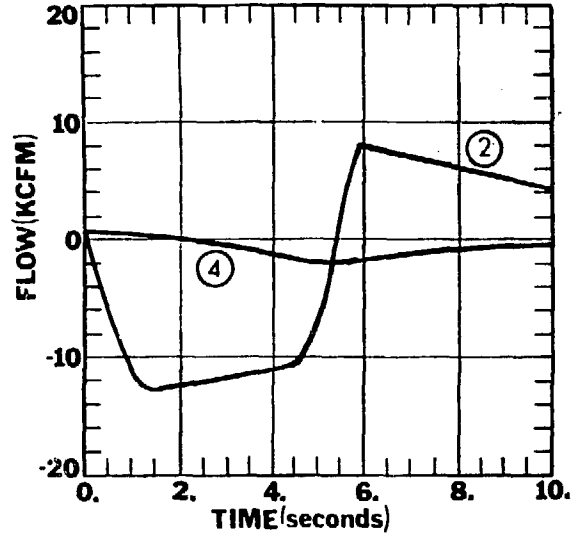


Fig. 18 System flows - tornado at inlet

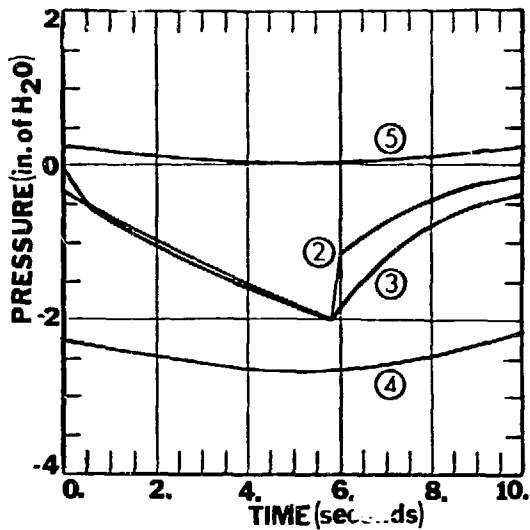


Fig. 19 System pressures - tornado at inlet with isolation valve

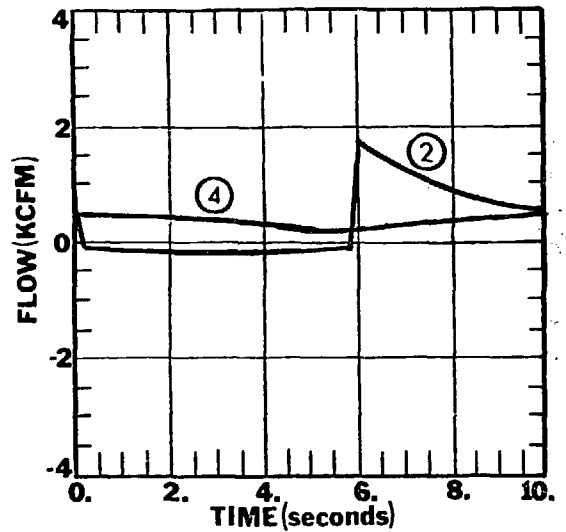


Fig. 20 System flows - tornado at inlet with isolation valve

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Real life systems are enormously more complex than the simple system model chosen, and have some built in attenuation provided by normal system construction. However, this example does illustrate two points. The first is to confirm the intuitive, if qualitative, reasoning that isolation will have positive effect in reducing expected system flows and pressures. Secondly, it provides evidence that a method of quantifying that effect is currently available.

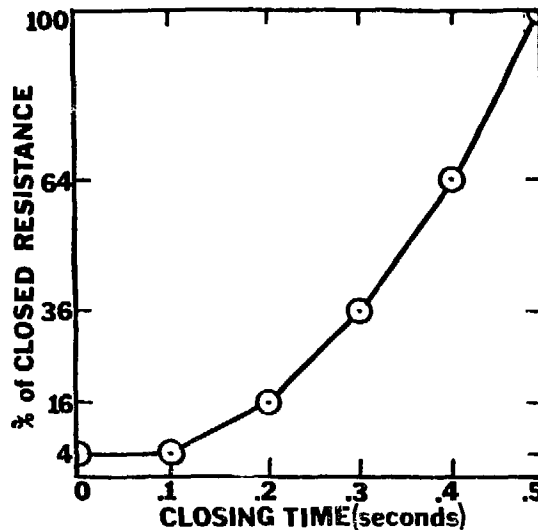


Fig. 21 Resistance curve
for outlet control valve

VIII. Future Effort

The goals of this project as originally stated were to (1) devise a suitable model for the isolation device to allow predictability of responses, (2) design and carry out sufficient testing to verify the validity of the model, and (3) use the predicted response to evaluate the protection provided to real systems.

To this end the following remains to be accomplished:

1. The product model shall be refined to more closely approach real system conditions by the inclusion of dynamic relationships.

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2. Testing shall be carried out under conditions which more closely simulate the Design Basis Tornado. Apparatus for this testing is being developed and is shown schematically in Figure 22. The pressure wave will be simulated by rupturing a disc on the high pressure storage tank, releasing a controlled pulse down the duct to impact with the test valve. Time-pressure readings will be obtained from pressure transducers providing a signal to a time controlled trace. Physical behavior and closing times will be monitored by high speed photography.

3. The LASL TVENT program will be used to analyze and evaluate real systems with and without isolation protection.

4. System effect caused by the rapid closure must be considered in the model, testing, and evaluation. Such effects might include shock wave generation, motor loading of fans and blowers, and duct starvation.

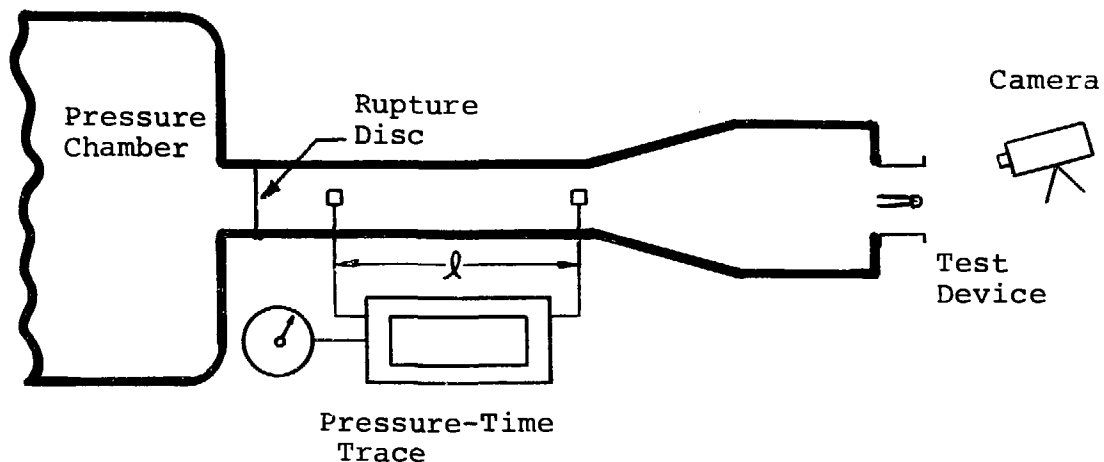


Figure 22 Proposed Test Apparatus

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DISCUSSION

MOELLER: Your map of tornadoes shows that the majority move in a northeasterly direction. Does this have any implications relative to the orientation of air intakes and exhausts in nuclear facilities?

PYSH: It has, but it's never been recognized. I might just say that the Institute for Disaster Research at Texas Tech has done considerable work on tornadoes. In fact, if given the opportunity to attend their symposium on tornadoes, which I think they have every two years, it's worth the adventure provided you can stand West Texas in the summer. They have a lot of meteorological engineering criteria for the tornado model that the NRC uses.

CHEEVER: Could strong ductwork downstream from HEPA filters protect the filters from tornado damage through frictional limitation of flow? Would this make it possible to omit the isolation valve?

PYSH: There are many things that can be done. I put them in a category of passive mechanics. If all you are concerned about is the ductwork, obviously it can be designed for three pounds and it will do the job. But there are filters and fans and other components.

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CHEEVER: What I mean is, would you get enough losses in this ductwork by friction so you would protect the filters that way?

PYSH: I can't answer that question; it depends on the path and it has to be modeled. It's possible that it could be done.

ANDERSON: I might caution you about a navy test I observed one time. We had a guillotine, mushroom-type isolation valve to protect one of our buildings and we conducted a blast test against it. The guillotine worked fine, but the building disappeared.

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EVALUATING NUCLEAR POWER PLANT VENTILATION SYSTEM ADEQUACY IN REDUCING AIRBORNE RADIOACTIVITY EXPOSURE

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Abstract

The problem of analyzing the effectiveness of the ventilation system for reducing airborne radioactivity exposure of personnel in a nuclear power plant is one faced by many nuclear analysis groups in power plant design organizations. This paper presents a rational way of approaching the problems and some simple methods of solving them. In so doing reasonable component leakage values are developed which can be used in solving this and other problems. For chronic leakage the values of 0.1lb/hr for valves, strainers and flanges are derived and 0.5lb/hr for pumps or other active components.

I. Introduction

Increasing attention is being given by the NRC (Nuclear Regulatory Commission) to in-plant radiological exposure and to keeping that exposure as low as reasonably achievable (ALARA) (1). One particular area of increased concern is the mitigation of airborne radioactivity. This is evidenced by the number of items concerned with this exposure source in NRC Regulatory Guide 8.8(1) and the questions concerning it in the latest revision of NRC Regulatory Guide 1.70.3(2). While it is evident that there is a connection between the ventilation in an area and the airborne concentration, it has not been clear exactly what the design relationship was other than increased airflow results in decreased airborne activity. Obviously, there are economic and practical constraints on this latter concept. The question to be answered then is what level of ventilation is necessary and practical and what are the factors affecting the selection?

II. Discussion

A primary source of airborne activity in an area in a nuclear power plant results from the leakage of contaminated liquids. This occurs through small leak paths such as valves, flanges, strainers, pumps and other such "breaks" in the piping and tankage systems. Open tanks or tanks vented into an area are even greater sources of airborne contamination than leaking components.

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In general for an area:

$$\frac{dN}{dt} = -\lambda N + C_s F_i - F_o \frac{N}{V} + \dot{L} C_L P$$

where $\frac{dN}{dt}$ is the time rate of change of the airborne activity, λ is the decay constant for the radionuclide of interest, N is the total number of curies of the airborne radionuclide, C_s is the airborne concentration of the subject radionuclide in the incoming (staged) air, F_i is the incoming airflow rate, F_o is the outgoing air flow rate, V is the free volume of the room enclosure or other area of interest, $\dot{L}$ is the radioactive liquid leak rate into the area, C_L is the subject radionuclide concentration in the leaking liquid and P is the partition factor of the subject radionuclide between the air and the liquid under the conditions extant. Under equilibrium conditions, this reduces to:

$$\frac{dN}{dt} = 0 = -\lambda N + C_s F_i - F_o \frac{N}{V} + \dot{L} C_L P \quad (1)$$

which is simply the familiar rate of input equals output plus accumulation with the added qualifier "minus decay." Re-arranging terms one can solve for the airborne concentration in the area:

$$C_A = \frac{L C_L P + C_s F_i}{(\lambda V + F_o)} \quad (2)$$

where C_A is the airborne concentration of the subject radionuclide equivalent to N/V for the simpler case of non-staged air flow (i.e., uncontaminated input air). For reasonable free volumes and moderate half lives, this equation reduces to:

$$C_A = \frac{L C_L P}{F_o} \quad (3)$$

While this equation is satisfied by any consistent set of units it is normal for the input data to be available with L in lbs/hr, C_L in uCi/ml, P is unitless and F_o in ft³/min. C_A is needed in uCi/cc to compare with MPC values. Hence a conversion constant of $5.34 \times 10^4 \frac{\text{ft}^3\text{-hr}}{\text{lb-min}}$ is necessary. Using the results of this equation,

one can now proceed to judge the adequacy of the ventilation as proposed for a building or alternatively to suggest a ventilation flow rate which will be adequate based on some pre-established criteria.

The decision as to what airborne concentration to allow in an area is a somewhat subjective one. From NRC regulations one might infer that as long as the total exposure does not exceed 40MPC hours per week to an individual the concentration in any area can be many times MPC. A more restrictive interpretation of 10CFR20⁽³⁾, however including the proposed 20.103 would lead one to the conclusion that airborne concentrations should be kept below 25% of MPC unless it is technically impractical to do so. Since it is necessary to post

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an area as an airborne radioactivity area anyway if there is a reasonable chance that someone will be exposed to 25% of the limit, a prudent health physicist would no doubt post an area over 25% of MPC regardless of projected occupancy. Hence, to avoid unnecessary posting and to conform to the strictest interpretation of the code, a maximum of 25% of MPC is recommended as the design goal for airborne radioactivity in restricted areas. In conformance with our own practice with respect to skin dose rates and in the spirit of the ALARA philosophy, a design goal of 10% of MPC for airborne radioactivity in unrestricted areas is recommended. This latter would also make it permissible to tour minors into unrestricted areas of the plant.

III. Derivation of Leakage Terms

Liquid concentrations can be calculated with some measure of success and are necessary information for other radiological calculations during plant design. These parameters are usually known quantities. Likewise, the partition factor can be chosen from available data (such as in WASH-1248)⁽⁴⁾ knowing concentration and temperature of the liquid. One is left, therefore, with liquid leak rate and air flow rate as unknown variables. Since the appropriate air flow rate is the value sought, one is left with the problem of devising a method for arriving at the liquid leakage rate.

Some relevant value of leak rate must be assigned to each area of the plant. Should one pick a constant leakage, i.e., one gallon per minute and apply it to every area? Should the liquid leakage rate be based on the size of pipes that traverse an area? While many systems for assigning leakage or airborne concentrations to an area might be suggested, most of these "distribution formulas" would be open to serious criticism on one basis or another. The calculations would be simple and precise if daily leakage in each area of the plant is known. It is clear, however, that this leakage would vary widely with time and is not available at the design stage.

It was at this point that a more penetrating scrutiny of the sources of leakage in a plant was begun. Obviously, any "breaks" in the piping or tankage are potential sources of chronic leakage. It is the effects of chronic leakage which are to be mitigated. It is impractical to design ventilation to mitigate the consequences of a fountaining valve or a pipe break involving liquids of even moderate concentrations of iodine. The valves, flanges, strainers, pumps, etc., are easily identified as the potential and actual sources of leakage. They represent "breaks" in the otherwise continuous, integral path of piping and tankage. Attaching average leakage values to each component, however, proves to be a difficult and nebulous task. While a few values for such discrete leakage have been proffered in the literature, mainly traceable to B&W (Babcock and Wilcox)⁽⁵⁾ ⁽⁶⁾, no explanation is given for their genesis and, hence, it is impossible to attach a high level of confidence to the numbers. Such information (average leak rates per component) is typically not available given the host of variables involved such as valve types, sizes, materials, manufacturers, operating conditions and quality controls to name a few. It would be fortuitous if the

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range of average leak rates taken over these variables proved to be confined to a narrow, useful band. It would be surprising if individual valves, throughout years of service and repair, remained in such a band. The possibility of finding component average leak rate data has therefore, been rejected and an entirely different tack taken.

To arrive at an appropriate rate for chronic leakage we must determine at what rate a component could leak undetected and unrepaired for the entire life of a nuclear power plant. When a component starts to leak, the liquid may collect on the bottom of the component and form drops which will drip. This phenomenon is essentially insensitive to the host of variables which characterize a valve as they influence only when the leak will start. Conceivably, every component in an area could leak at some low rate which would not be high enough to warrant repair or would escape detection. This is the maximum chronic leakage against which a ventilation system's radiological adequacy should be measured. While plant experienced persons will recognize that much higher leak rates occur routinely, these higher rates would not meet the criteria as chronic leaks, i.e., they would not go undetected very long, would be repaired at the earliest opportunity and would never occur on all components simultaneously without shutdown of part of the plant. Most importantly, leaks of this magnitude cannot be mitigated by practical ventilation design.

It remains to generate such values for chronic leakage. A simple experiment was performed in which the sweep second hand of a watch was observed while verbalizing the word "drip" at regular time intervals starting with one per second and working up to once every fifteen seconds. It seemed apparent that a drop every fifteen seconds would not be noticed by casual observation and that one every second would. Eventually, it was decided that one drop every five seconds might be noticed but would probably not be cause for repair action. Using a conversation factor of twenty drops per cubic centimeter, a chronic leak rate per component of 0.079 lb/hr was calculated. (This checks well, interestingly enough, with the B&W value of 0.0661 lb/hr which works out to 10 drops a minute or one drop every six seconds.) For computational ease as well as additional conservatism, this value was rounded to 0.1 lb/hr or approximately one drop every four seconds.

Pumps tend to leak more before being noticed and, in fact, are designed to leak some water for sealing, lubrication and seal cooling of shaft seals. Based on experience and some literature values (5) (6) it was felt five times as much leakage for pumps was appropriate. A value of 0.5 lb/hr as the chronic leakage of pumps for use in calculating necessary ventilation was, therefore, adopted.

IV. Application of the Technique

Having proposed the chronic leakage values, it remains to show how they are intended to be used. Rigorously, the adequacy of the ventilation in an area could be calculated by counting the exact number of leak paths in the area and multiplying them by the proposed

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values to arrive at the total leakage. This, in fact, was done for one plant to test the technique. As might be expected, the method is extremely tedious, time consuming, and physically exhausting as it requires pouring over P&IDS (piping and instrumentation diagrams) trying to match up the components with the area under study. It requires that the design of the plant be advanced enough to have the P&IDS available. This sometimes means that it is too late to change ventilation design unless the change is critical as the equipment may have already been ordered. Through doing a plant the hard way, it was discovered that a certain maximum number of leak paths were infrequently exceeded. Adding a margin of safety, a value of 0.042 gallon per minute was determined as an upper limit on the chronic leak rate in any area. This value is used for screening in either of two ways: 1) when used with proposed ventilation flow rates to compare the resultant airborne concentration with 25% of MPC, or 2) to compare the calculated air flow rate with practical limits when an airborne concentration of 25% of MPC is used as the starting point of a calculation. Using this screening value, the separately ventilated areas of the plant can be expeditiously checked and the exceptions isolated. Generally, only a few areas will fail the screening tests. These few areas can now be checked with the more rigorous component counting technique to see if they really do not pass. Areas not passing the latter test require additional air flow or may require air flow at a level not practicable. In these cases, a decision must be made to either live with the potential problem with consequent administrative controls and posting or to take measures to affect one of the other variables in the equation.

It should be stressed that the minimization of airborne radioactivity by proper ventilation distribution should be considered as a fine adjustment on such activity. It is capable of minimizing the effects of only a narrow range of concentrations and, hence, liquid leak rates. The primary emphasis should be placed on eliminating leakage.

V. Other Factors to be Considered

While the primary emphasis of this article has been on the relationship of air flow rates to liquid leakage rates, a typical ventilation system review covers other less quantifiable areas. The ventilation system provides one of the prime agents in reducing the spread of airborne contamination. In this respect, a ventilation design should be checked to see that where air is staged (the air from one area also is used to ventilate another area) the flow pattern should be from areas of lower potential contamination to areas of higher potential contamination. The contamination potential of an area is based, of course, on the source strength in the area, i.e., radioactive concentration of the liquid (or gas). To prevent back flow from main ducts into areas or from a higher potential contamination area to a lower one, back flow dampers should be used on the exhaust duct from an area. Areas of potential contamination

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should be maintained at lower pressures with respect to areas of no potential contamination. To maintain the latter, back flow dampers should also be installed on intake ducts into an area such that when a door is open into an area, the air flow is inward. Tanks should always be completely enclosed and the vents from tanks connected directly to the ventilation exhaust duct or as a minimum the vents should have local filters.

VI. Conclusion

No consideration has been given in this article to the industrial hygiene, thermal or comfort factors involved in ventilation design. While an impression may have been given that radiological considerations dominate the design of ventilation systems in a nuclear power plant, the reverse is almost always true. Generally, normal industrial requirements will provide more than enough ventilation to provide radiological adequacy as well. Frequently, the overabundant air flow in some areas based on industrial considerations will allow some re-routing of air flow to areas of radiological concern. It is rarely necessary for additional ventilation capacity to be installed for purely radiological considerations. A radiological review of ventilation design does insure that, given options, the designer will choose those with the least radiological problems. It also allows for the elimination of some potential problems prior to plant start up and the identification of some areas requiring initial attention.

VII. References

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Dep

AIR CLEANING SYSTEMS ANALYSIS
AND
HEPA FILTER RESPONSE TO SIMULATED
TORNADO LOADINGS

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ABSTRACT

A computer code, TVENT, for predicting tornado-induced depressurization in air cleaning systems is described. TVENT easily fits on many computers with input/output formats that are familiar to most analysts and HVAC designers. Our applications of TVENT to several nuclear facilities in Idaho, New York, and New Mexico are described.

Experimental results are described that provide supportive data on the response of individual air cleaning components to simulated tornado loadings. Peak pressure failure results for 610-by-610-mm-by-292-mm (24-by-24-in.-by-11-1/2-in.) standard HEPA filters have been obtained. Structural failure occurred for peak pressures of 9.7 kPa (1.4 psi) to 23.5 kPa (3.4 psi) at pressurization rates from 19.3 kPa/s (2.8 psi/s) to 9.7 kPa/s (1.4 psi/s), respectively. From these data, we derived a relationship between peak failure pressure and pressurization rate. Additional work is planned to evaluate the effects of manufacturer type, peak pressure duration time, number of filter folds, and filter orientation on structural failure.

Flow-resistance data of HEPA filters for use in TVENT are also described. At low flow resistance appears to be mainly caused by a diffusion mechanism, while at high flow the resistance seems to be caused by the mechanism of momentum exchange.

I. Introduction

Safety analysis reviews of proposed HVAC designs for nuclear facilities need to include the effects of tornados on air cleaning systems and the individual components within these systems. We have developed an analytical method to predict tornado-induced pressures and flows within nuclear facilities. This method has taken the form of an easily used, portable computer code called TVENT. We are also developing supportive experimental data of tornado effects on individual air cleaning components. Simulated tornado pulses are imposed on the components to determine their failure points and response at higher-than-normal pressures and flows.

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A description of TVENT and examples of its application to nuclear facility air cleaning systems are described in the following sections. Also reported are experimental data on HEPA filter response to simulated tornado loadings. In addition, flow-resistance data on HEPA filters and integration of these data into TVENT are described.

II. Air Cleaning System Analysis

General

In setting out to develop TVENT, we assumed that to be analyzed, an air cleaning system had to exist either physically or as a finished design on architect-engineering drawings. We also assumed that, for maximum utility, the code must accept input in a form natural to designers of air cleaning systems.

With respect to the first assumption, we found that an air cleaning system does not have to exist physically or as a finished design before it can be analyzed. The computer code can be used in a design mode allowing the analyst to determine tornado transient effects at any time during the design phase. We believe that the code is also useful for determining the effects of minor perturbations (such as loss of a blower) in complex air cleaning systems. We believe our second assumption has produced a more useful code, as TVENT has been used by HVAC engineers that did not have extensive programming experience.

TVENT

TVENT solves steady-state and transient pressure distributions in complex ventilation networks and calculates system flow based on assumptions of

- o One-dimensional, isothermal flow,
- o Lumped parameters (neglecting spatial distribution of components), and
- o Incompressible flow with fluid storage at nodes.⁽¹⁾

Previous studies have shown that for tornado-induced flows inertial effects can be neglected and shock formation will not occur.^(2, 3)

TVENT is written in FORTRAN IV and is designed to be portable; that is, it can be easily transferred from one computer to another with a minimum of change. TVENT has been run on CDC, IBM, UNIVAC, and DEC computers, and requires less than 30K decimal words to run in a nonoverlaid mode on the CDC 7600 computer. The overlaid version of TVENT requires less than 20K decimal words on the same computer.

Input/Output

Input is organized and formatted in a way that is natural to the designer or engineer who prepares data for analysis. Input error

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diagnostic messages are printed to help find errors in data preparation. Checks are made and messages are supplied when possible modeling errors are detected. Portability requirements precluded free format input and film plotting options, which are not found on some computer installations. Output lists and summaries are formatted and organized in a way that is helpful in determining the effects of a transient on system components and the integrity of pressure differentials between containment areas. Line printer plots for the time profiles of selected pressures and flows are also generated.

TVENT Application Examples

The degree of complexity needed in a TVENT model depends (1) on the purpose of the analysis, and (2) on the information available to define the system in its normal operating condition. It makes no sense to model a component that cannot be evaluated. On the other hand, those components (be they blowers, dampers, filters, or rooms) that significantly affect the system's performance must be represented. Certain portions of the model may be shown in much more detail than others depending on the desired refinement in output flows and pressures. Sometimes the effect of one part of the system on another can be crudely, yet accurately, simulated by the minimum number of components that are compatible with the pressure differentials in that area.

We have applied TVENT to a number of DOE nuclear air cleaning facilities to learn about the problems involved in such an application. In all but one case, the greatest difficulty encountered was with evaluating component performance during normal operation. Most of the information needed to define component effects (on the pressure) is not required for the operation of the system and, consequently, never measured in the field. The one exception, Los Alamos Scientific Laboratory's (LASL) new plutonium facility, involved a system that was not yet operational but could be completely defined by the architect-engineer's predictions.

The first application of TVENT was to the Idaho Chemical Processing Plant (ICPP) at Idaho Falls, Idaho.⁽⁴⁾ A recently installed atmospheric protection system (APS) was the subject of our analysis. This is an example of where the flows and pressures in a very specific part of the system are needed, but are not needed in the remaining parts. The TVENT model for this analysis is shown in Fig. 1. It shows the degree of modeling complexity judged to be appropriate for the older portions that are connected to the APS.

The second application of TVENT was to the new plutonium facility at the LASL.⁽⁵⁾ All of the information concerning the air cleaning systems was available from the architect-engineer's calculations and drawings. This proved to be a very worthwhile exercise in the development of TVENT because the application to such a large system tested TVENT's limitations. As a result, we know the code is capable of handling large systems. A small portion of the model used is shown in Fig. 2. This represents half of the first floor containing the laboratories and shops for the recycle and metal fabrication operations. The ducts terminating at the top of the figure lead to the basement that houses the 35 blowers servicing this area. The model of this system contained a total of 168 branches and 110 nodes.

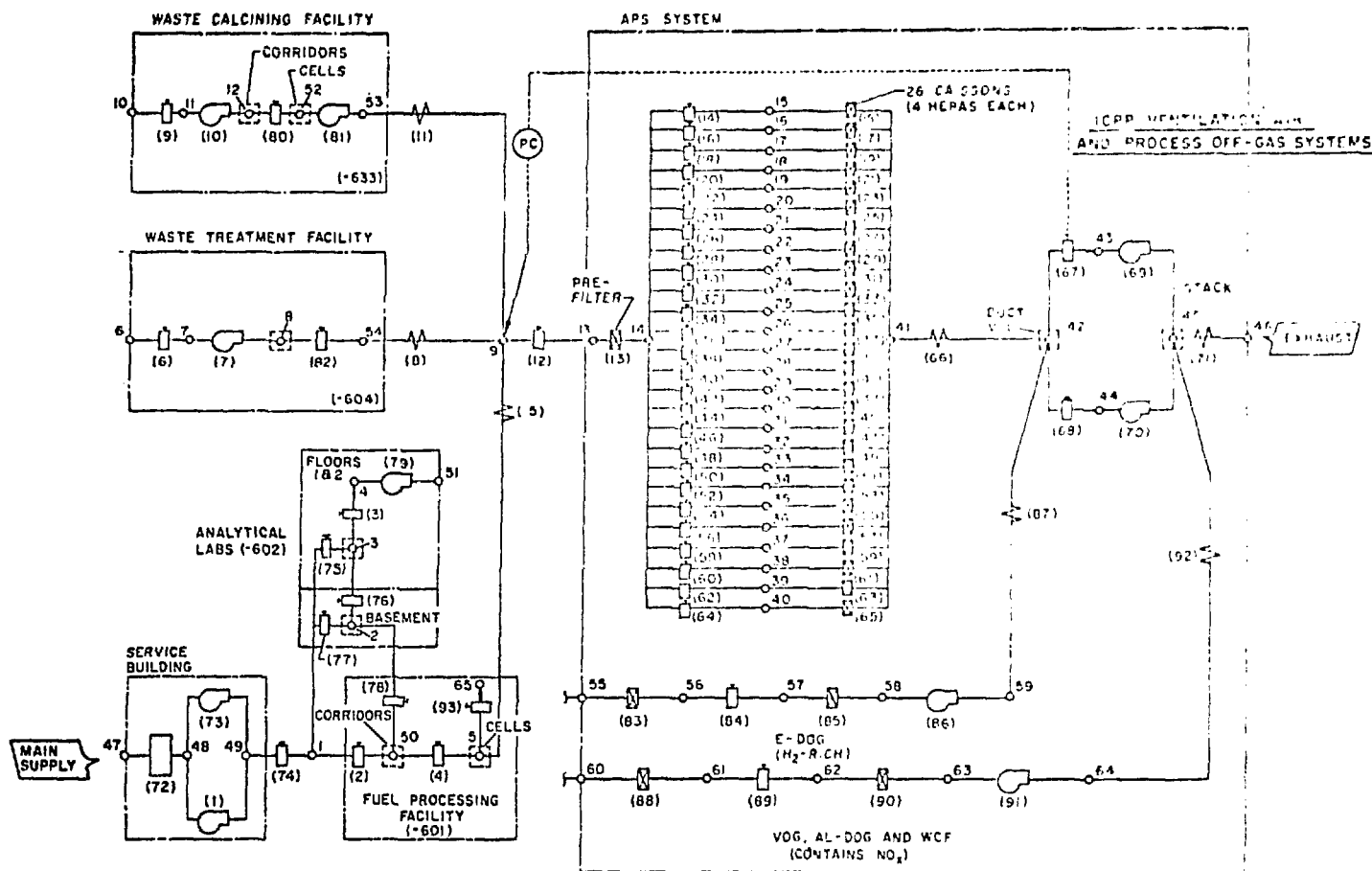


Figure 1 TVENT model of ICPP

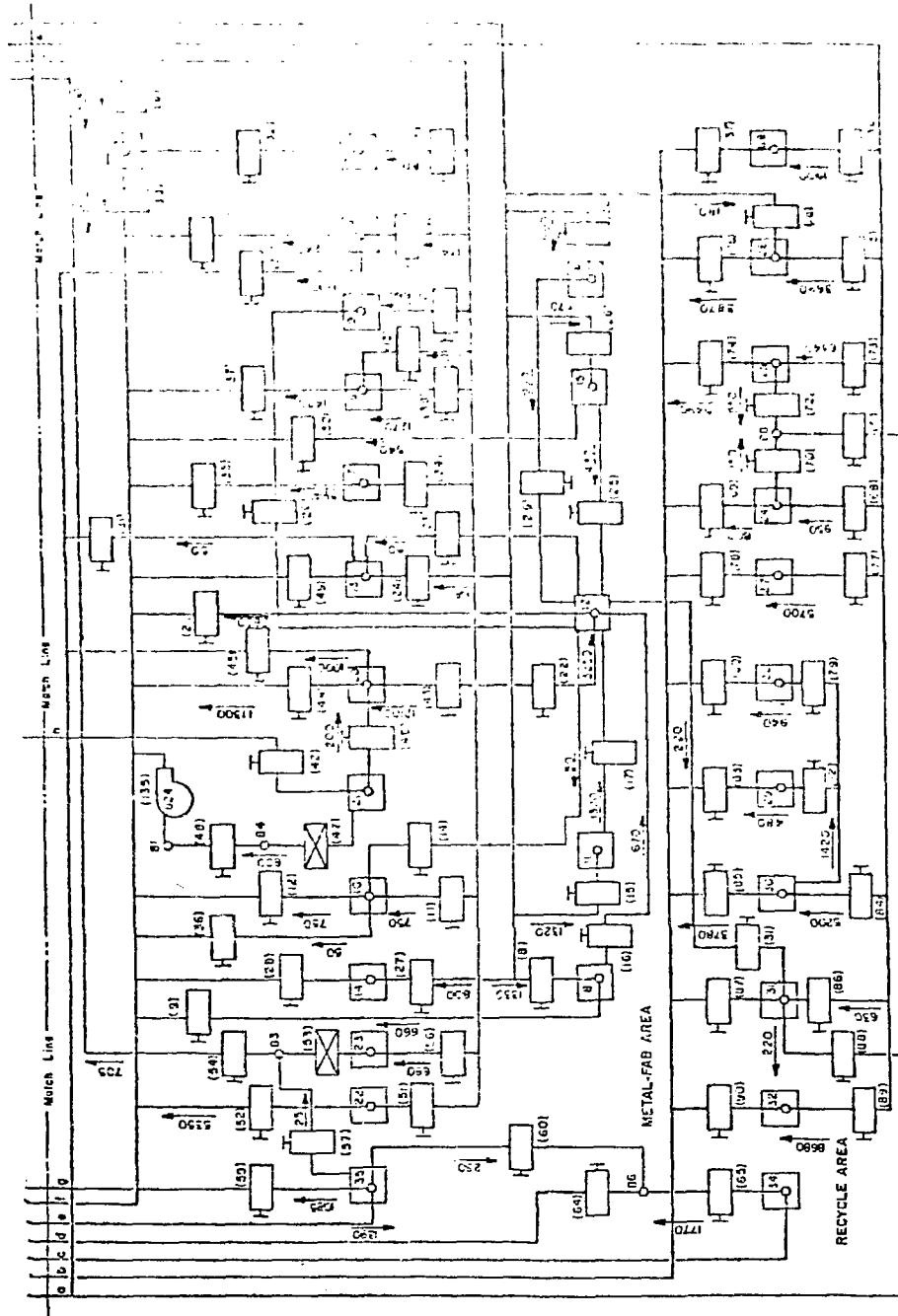


Figure 2 Complex TVENT model showing the LASL plutonium facility

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The third TVENT application was to the head-end ventilation system at the Nuclear Fuel Services facility near West Valley, NY.⁶³ The head-end system serviced the process building housing the major process cells used in reprocessing fuel elements. This facility is no longer operative, but residual contamination in these cells would present a potential danger if it were released to the surrounding environment during a tornado. The first step in studying this problem was to use TVENT to obtain the flows entering or leaving these cells during a hypothetical tornado. Figure 3 shows these flows as well as the flows in the ducts at the supply points for the tornado striking all supply points simultaneously. The duration of the applied tornado was about 10 s and began at time = 10 s.

III. HEPA Filter Response to Simulated Tornado Loadings

General

Our studies of ventilation systems under tornado conditions also involve experimental work. We have fabricated a blowdown apparatus capable of generating transient air flow typical of a severe tornado through full-scale HVAC system components. The usefulness of this facility has been demonstrated by the testing of HEPA filters. We plan to use this facility in future tornado simulation experiments on other components as well.

The blowdown facility is located at New Mexico State University, Las Cruces, New Mexico. The test apparatus consists of an air compressor, two air storage tanks, sequencing valves, a prefilter chamber, and a test section. The sequencing valves control the airflow from the storage tanks through the prefilter chamber, test section, and finally, the test component. A high-speed movie camera was installed at the exit of the apparatus to study HEPA filter response to simulated tornado loadings. A more detailed description of the facility has been published.^{67, 68}

Initial Test Conditions

The purpose of our preliminary tests was to determine the relationship between failure pressure and the pressurization rate for standard 610-by-610-by-292-mm (24-by-24-by-11-1/2-in.) HEPA filters. Experimental determination of peak failure pressure required the sequencing valves to be set to give various pressurization rates. The rate specified in the National Regulatory Commission Guide 1.76 for a Design Basis Region I tornado (the most severe) is 13.8 kPa/s (2 psi/s). Filters were evaluated using pressurization rates at, above, and below this value. The tests were continued until the pressure differential across the filter was above the maximum of 20.7 kPa (3 psi) of the Region I tornado. The high-speed movie camera and all instrumentation recorders were started simultaneously with the sequencing valves. The actual times of filter failure were then found by observing the high-speed movies of the downstream filter face. Also photographed by the camera during the tests were a clock and manometer mounted beside the filters. Pressure differential across the filters and dynamic pressure were measured by electronic pressure transducers and recorded by strip chart recorders. Temperatures in the air storage tanks and the test duct were sensed by thermocouples and also recorded on the strip charts. Humidity was

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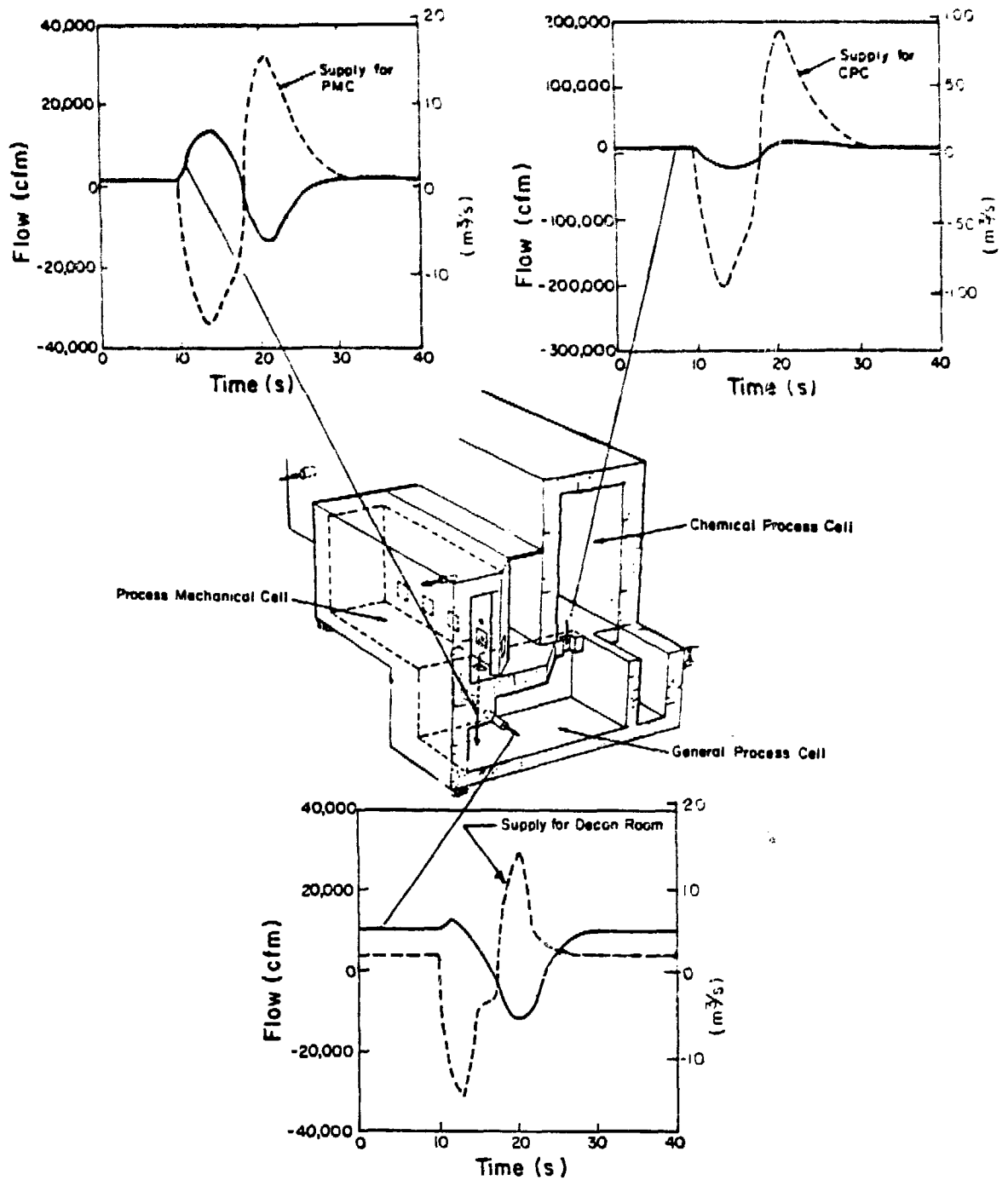


Figure 3 Nuclear Fuel Services cells and resulting transient flows

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measured by a gauge within the prefilter chamber in the low velocity air. This gauge was read and the humidity recorded following each test.

Test Results

The test results are shown in Table I and Fig. 4.⁽⁹⁾ Eleven HEPA filters (from a single manufacturer) with 65 media folds were tested. The data of Table I indicate that structural failure occurred for a peak pressure range from 9.7 kPa (1.4 psi) to 23.5 kPa (3.5 psi) at pressurization rates from 19.3 kPa/s (2.8 psi/s) to 9.7 kPa/s (1.4 psi/s), respectively. Only two HEPA filters withstood the full 20.7 kPa (3 psi) transient pressure pulse. The column listed in Table I under "Break Fold No." indicates where the failure on the downstream filter face occurred. Medium folds were numbered left to right, 1 to 65. Thus in all but three cases, the glued, downstream edge of the filter medium was the initial point of weakness.

The data from Table I are shown plotted in Fig. 4. The solid line of Fig. 4 is a least squares fit to all the data. This fit indicates a trend in the relationship between failure pressure and pressurization rate. That is, the higher the pressurization rate the lower the pressure needed to fail the filters. Notice that the failure pressure at a pressurization rate of 13.8 kPa/s (2.0 psi/s) is

TABLE I. Experimental failure pressure vs. pressurization rate.*

Filter Serial Number	Pressurization Rate kPa (psi/s)	Failure Pressure kPa (psi)	Break Fold No.
41310250	9.0 (1.3)	21.4 (3.1)	1
41310358	9.7 (1.4)	23.5 (3.4)	65
41307976	9.7 (1.4)	13.8 (2.0)	1
41310360	10.4 (1.5)	15.2 (2.2)	65
41310366	11.7 (1.7)	17.9 (2.6)	38
41310859	13.1 (1.9)	17.3 (2.5)	35
41310283	13.1 (1.9)	15.2 (2.2)	65
41310363	13.8 (2.0)	14.5 (2.1)	1
41310268	18.6 (2.7)	16.6 (2.4)	65
41310367	18.6 (2.7)	20.7 (3.0)	28
41310261	19.3 (2.8)	9.7 (1.4)	1

*Note: All filters tested were 610-by 610-by-292 mm (24-by 24-by 11-1/2 in.) each having 65 folds.

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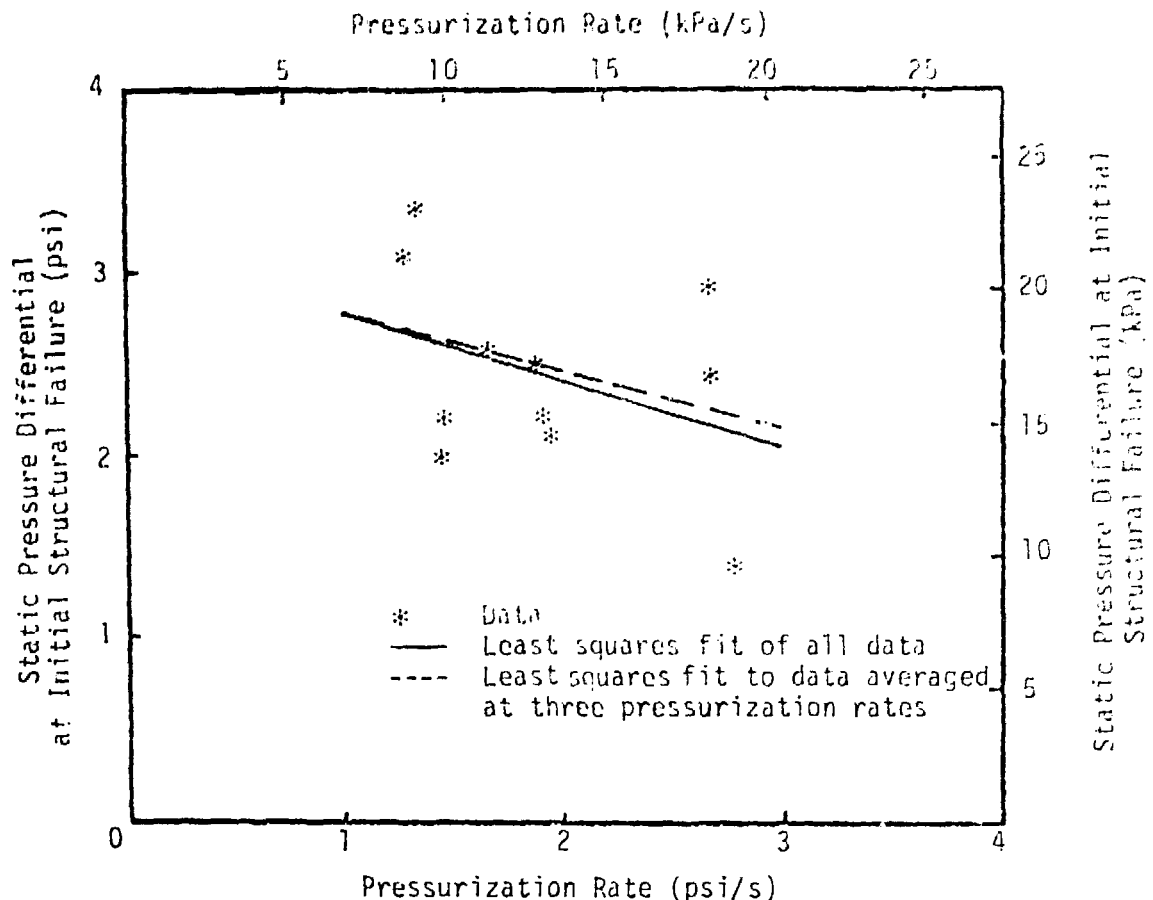


Figure 4 Static pressure differential at the point of initial structural failure as a function of pressurization rate [610- by 610- by 290-mm (24- by 24- by 11-1/2-in.) HEPA filters].

about 16.6 kPa (2.4 psi). Therefore, this type of filter would probably fail if subjected to a Region I tornado pulse. In fact, extrapolation of the least squares curve indicates that all filters of this type are in danger of failure at a 20.7 kPa (3.0 psi) differential pressure for pressurization rates above 3.5 kPa/s (0.5 psi/s).

The tests were run attempting to replicate failure pressure at three different pressurization rates. It proved difficult to repeat pressurization rates exactly. The three rates nominally were 9.7 kPa/s (1.4 psi/s), 13.8 kPa/s (2.0 psi/s) and 19.0 kPa/s (2.8 psi/s). If the data of Table I are treated as if the failure pressures indeed occurred at the nominal pressurization rates and are averaged for each rate, then the dotted line of Fig. 4 results. (The point with a pressurization rate of 11.7 kPa/s (1.7 psi/s) was not used in this analysis.) There is only a slight difference between this method of analyzing the data and taking a least squares fit of all the data.

The small number of filters available for testing did not permit us to obtain completely satisfactory results from a statistical

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standpoint. Therefore, only very general conclusions can be drawn from the tests. First, there appears to be an inverse linear relationship between peak failure pressure and pressurization rate. Second, pressurization rates above 3.5 kPa/s (0.5 psi/s) seem too severe if these HEPA filters are to withstand a 20.7 kPa (3.0 psi) maximum pressure differential that would occur in a Region I tornado.

Flow-Resistance Data

The results of the resistance measurements on 610-by 610-mm (24-by-24-in.) filters appear to agree with earlier results obtained for 203-by 203-mm (8-by-8-in.) filters. These results are shown in Fig. 5. Diffusion and momentum exchange may be used to explain the shape of these resistance curves. At relatively low flow rates, the

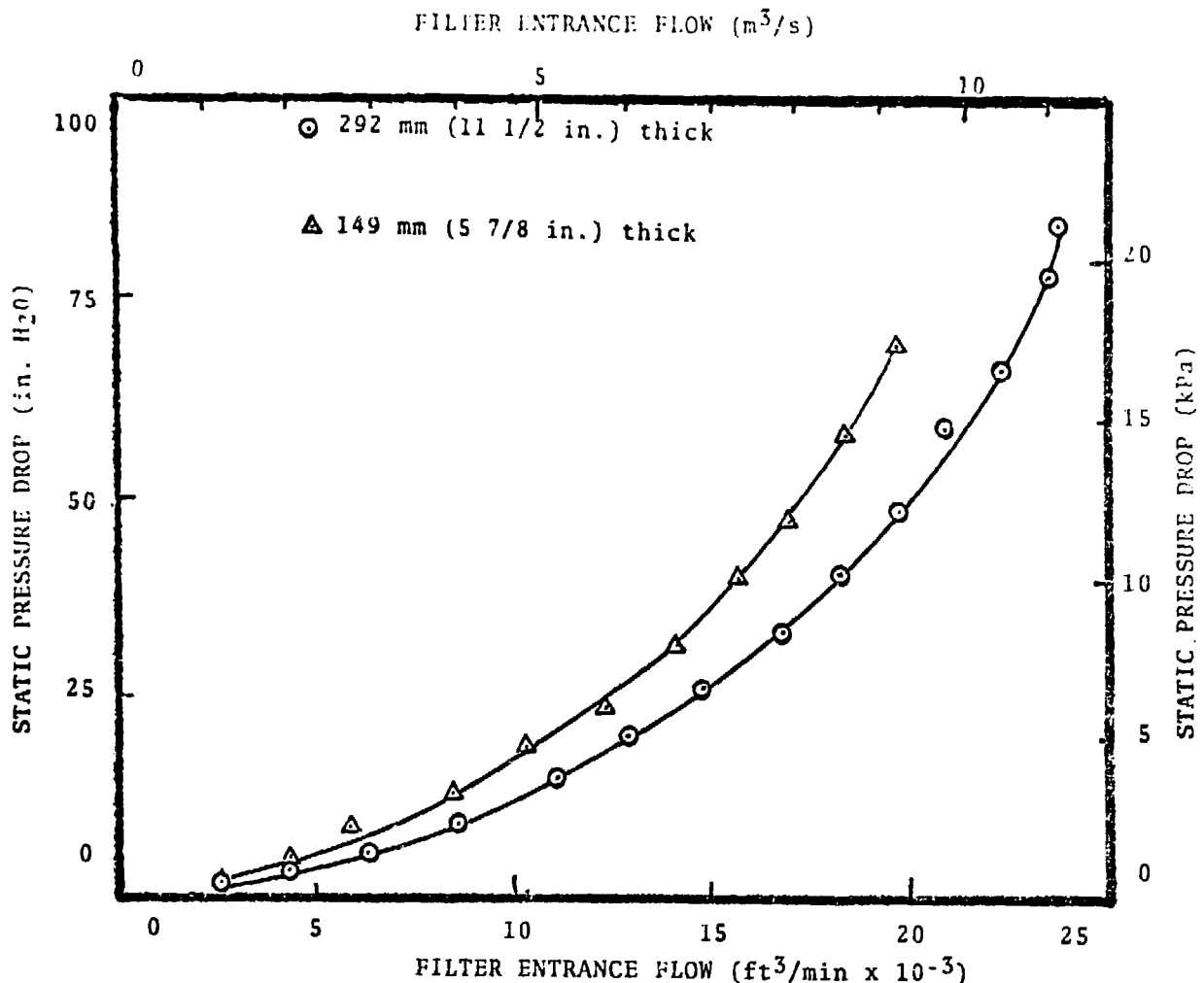


Figure 5 Resistance of 610- by 610-mm (24- by 24-in.) HEPA filters at high flow rates.

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dominate mechanism causing resistance appears to be diffusion, while at higher flow rates, momentum exchange appears to dominate. When momentum exchange dominates, a majority of the flow is probably passing through the folded ends of the fiber mat only, thereby increasing the possibility of inefficient filtration.

IV. Future Work

- o Scale models of air cleaning systems have been constructed. We plan to obtain experimental data from these models and compare them with TVENT results.
- o Analytical development of an explosion computer code that is patterned after TVENT has been undertaken. This computer code will be capable of predicting internally generated pressure time profiles within air cleaning systems.
- o Structural tests will be performed on HEPA filters for simulated tornado transients to evaluate parameters of number of folds, time duration at peak pressure, manufacturers' types, and filter orientation.
- o Efficiency tests of HEPA filters under artificial loading conditions and airstream-entrained aerosol will be performed for simulated tornado transients. Particulate release vs dynamic pressure loading will be determined.
- o Shock tests of HEPA filters will be performed to evaluate both mechanical failure and particulate release. European, standard separatorless, flat pack, and American V-type will be evaluated.

V. Summary

In this report, we have described a computer code called TVENT that can be used by the designer and analyst to predict tornado-induced transients within air cleaning systems. TVENT is easily used, portable, and easily applied to diverse types of air cleaning systems. The computer code with its supporting documentation is available at the Argonne Code Center or through personal communication with the authors.

Structural response of HEPA filters to peak pressure differentials was described. Thus far, structural failure has occurred from 9.7 kPa (1.4 psi) to 23.5 kPa (3.4 psi) with one manufacturer's filters. There apparently is an inverse linear relationship between the peak failure pressure and pressurization rate. Nonlinear flow-resistance data at high flow rates were also obtained. These graphical data can be translated into empirical relationships and incorporated into the computer code TVENT.

Acknowledgements

The authors would like to express their appreciation for the support provided by the Department of Energy, Division of Operational and Environmental Safety, and Division of Waste Management. Support from the Nuclear Regulatory Commission, Division of Safeguards Fuel Cycle and Environmental Research is also greatly appreciated.

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DISCUSSION

MURROW: Is this TVENT code available to anybody? Can we apply to you to run a simulation on a particular system?

GREGORY: Yes, the computer code is available to anyone who wants it. We have a manual that shows people how to use it and the code has been sent to the Argonne Code Center so all documentation and a listing of the computer code is available. We will be glad to help in any way that we can with regard to helping you with modeling. In the past, we have shown the architect engineers how to use it. Typically, it takes about half a day to just take one of their examples and go through it.

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MURROW: Will this code take into account the resistances of the various components (e.g., filters and fans), flowrates, and volumes, etc.?

GREGORY: Yes, it will. There are high-flow-rate, pressure-type loading data that we still need in addition to HEPA filter data. One of the particular things we need relates to the fan response. We are trying to generate that type of data and we will put it into the code.

I must emphasize that much of the data in the experimental facility generates high flow rates. That's what you're going to get with a 3 PSI loading from the tornado. You're not going to get shock waves; it is not a shock loading facility. However, we are building a shock tube where we will be exposing some filters and other components to shock loads from internally generated explosions.

WARNKE: I would like to comment on your paper and the discussions related to tornado induced pressure transients in nuclear HVAC systems. With regard to Allied-General's spent fuel reprocessing plant at Barnwell, we ran an analog simulation of the HVAC system to verify the effects of tornado induced flow on HVAC systems in general and specifically on the final HEPA filter banks. The final HEPA filter bank for the process areas was sized for approximately 72,000 cfm (two 36,000 cfm banks). These were walk-in type filter plenums. The postulated tornado was taken as 0 to -3 PSI ΔP in 3 sec.; -3 PSI ΔP for 2 sec.; -3 PSI to zero PSI in 3 sec. Two cases were considered: (1) tornado at the supply unit, (2) tornado at the stack. The final exhaust filters were considered to have a loading of 5 in.w.g. at the time of a tornado. The tornado-induced flow raised the ΔP across the final HEPA bank to approximately 20 in.w.g. The blowers acted as a choke. With permission of my management I'll send you a copy of the Bechtel study.

CLOSING REMARKS OF SESSION CHAIRMAN:

The papers presented have informed us of the existence of a computer program (TVENT) for predicting steady state and tornado-(or extreme wind-) indicated transient flows and pressures in ventilation systems. For new facilities, many ventilation system alternatives can now be evaluated using the program, and an optimum design selected. The atmospheric parameters of a tornado have been identified and a facility constructed to duplicate these conditions in actual simulations. Preliminary experimental results have demonstrated both qualitative and quantitative tornado damage conditions for operational components.

An experimental facility was described that evaluates countermeasure devices and techniques for control of fires and/or fire related combustion products. Experimental data is now available for engineer/designers to optimize total system engineering parameters for fire protection.

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SESSION IX

AEROSOLS

Tuesday, August 8, 1978

CHAIRMAN: M. Tillery

CHARACTERIZATION OF AIRBORNE PLUTONIUM-BEARING PARTICLES FROM A NUCLEAR REPROCESSING PLANT
S. M. Sanders

AGGLOMERATION CHARACTERISTICS OF FAST REACTOR HCDA AEROSOLS
G. W. Parker, G. E. Creek, A. L. Sutton, Jr.

SODIUM OXIDE/HYDROXIDE AEROSOL PROPERTIES AND BEHAVIOR IN A LARGE VESSEL
R. K. Hilliard, J. D. McCormack,
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ATTENUATION OF PARTICLES THROUGH LEAK PATHS
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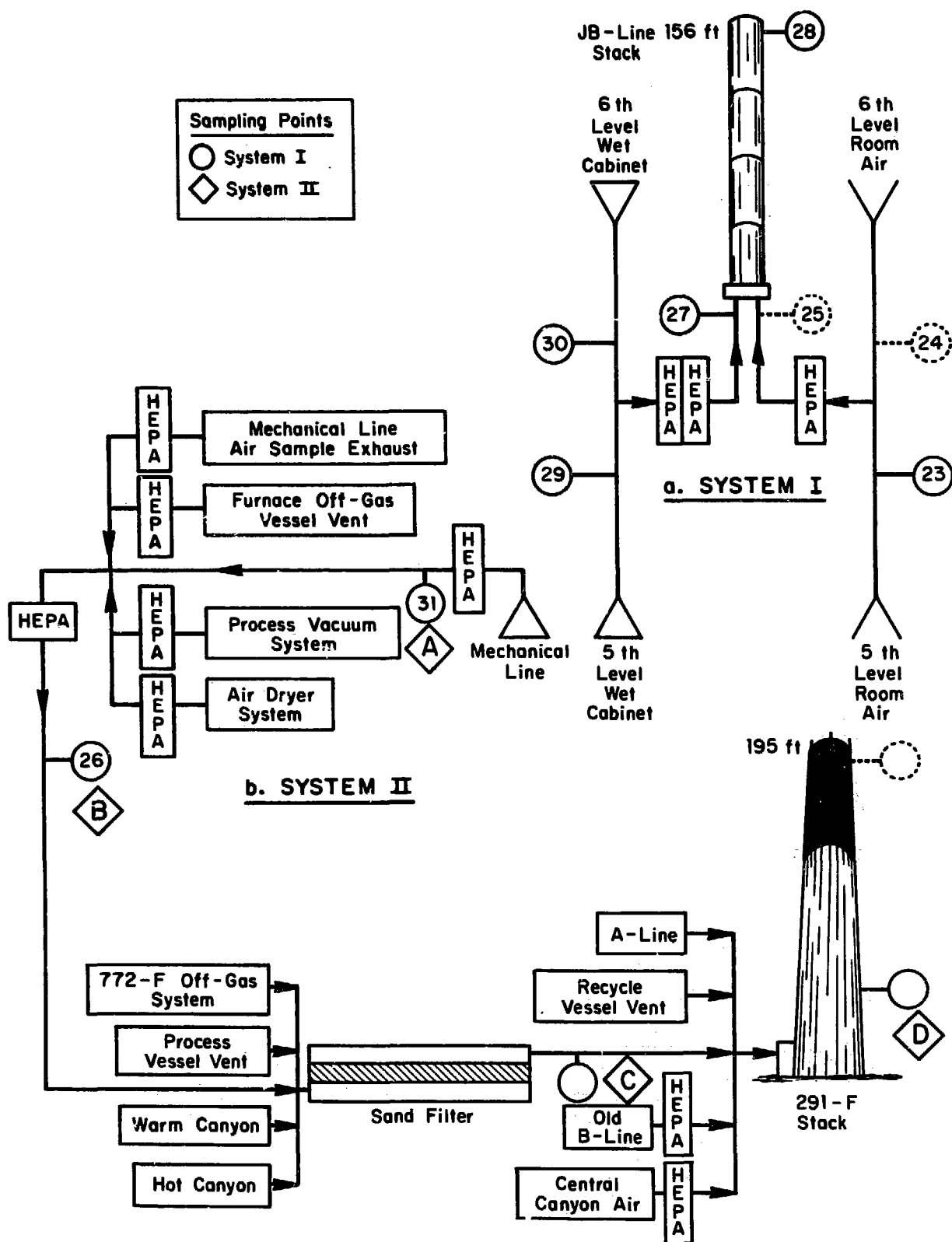


FIGURE 1
EXHAUST SYSTEMS FROM JB-LINE

(Sampling Points 29 and 30, respectively), of filtered air from both locations (Sampling Point 27), and of unfiltered room air from the fifth level (Sampling Point 23), and of air at the 156-foot level of JB-Line stack (Sampling Point 28). In System II, samples were taken of mechanical line air from just beyond the first high efficiency, particulate air (HEPA) filters located in back of the cabinets (Sampling Point A or 31); of the combined air from the mechanical line, air sample exhaust, furnace off-gas vessel vent, process vacuum system, and air dryer system after the second HEPA filter (Sampling Point B or 26); of the air leaving the sand filter which also contained air from the support laboratory off-gas system of Building 772-F, the fuel dissolving and extraction process vessel vent system and Building 221-F canyons containing the process vessels (Sampling Point C); and of air from the 50-foot level in the 291-F stack where air from the sand filter mingles with that from the uranium recovery A-Line and other sources (Sampling Point D). Air was sampled almost continuously at all locations (except at Sampling Point A) during June 1975 for System I and October 1975 for System II. The level of radioactivity at Sampling Point A was so high that samples were collected only during the first two days.

Only 22 particles could be found on filters used at Sampling Point C and D during October 1975. These points were sampled again from the middle of May to the end of September 1977 with the collection of 163 additional particles. Thus, a total of 121 particles were analyzed from System I (16 from sampling point 23; 68 from point 29; and 38 from point 30) and 417 from System II (125 from Sampling Point A; 107 from Point B; 114 from Point C; and 71 from Point D). These figures do not include 20 particles which contained no elements with atomic numbers greater than 9 and were assumed to be organic.

Methods and Materials

Particle Collection

Particles were collected by drawing a fraction of exhaust air through membrane filters. These filters were polycarbonate films 47 mm in diameter with 0.1- μ m diameter pores supported in a polycarbonate aerosol holder.* Air was drawn through the holder by a small diaphragm pump at a rate of four liters per minute to give a face velocity at the filter of 3.8 cm/sec. Since samples were collected from exhaust ducts having varying linear velocities, the sampling technique was anisokinetic. At the above sampling flow rate, the total efficiency for particle collection by the processes of impaction, diffusion, and interception, calculated according to Spurney,³ is 100% for all particles with diameters of 0.001 μ m (the diameter of gas molecules) or larger.

Arrangement of the air sampling system is shown in Figure 2. To determine the fraction of the exhaust sampled, integrated air flow was measured with a dry type test meter** in series with the diaphragm pump. When nitrogen dioxide was present, exhaust gas was passed through two gas drying towers between the filter and the pump. The first tower contained indicating *Drierite*† to remove moisture from the air and save the *Ascarite*†† in the second tower. The self-indicating

* The aerosol holders and membrane filters were produced by Nuclepore Corporation, Pleasanton, California, and obtained from them or Bio-Rad Laboratories, Richmond, California.

** Manufactured by the American Meter Division of Singer.

† Trademark of W. S. Hammond Drierite Company.

†† Trademark of Arthur H. Thomas Company.

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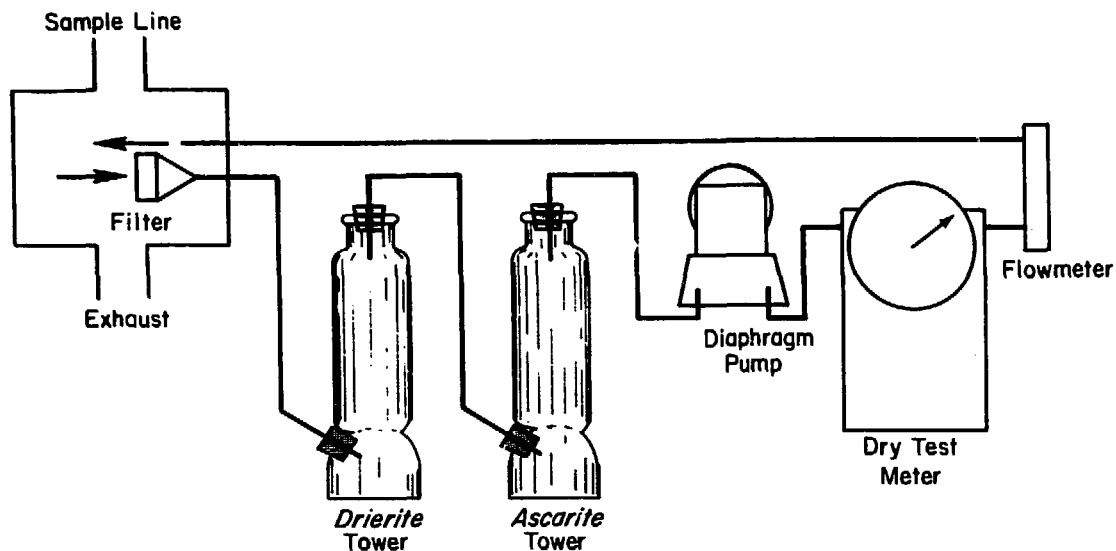


FIGURE 2
ARRANGEMENT OF SAMPLE COLLECTION EQUIPMENT

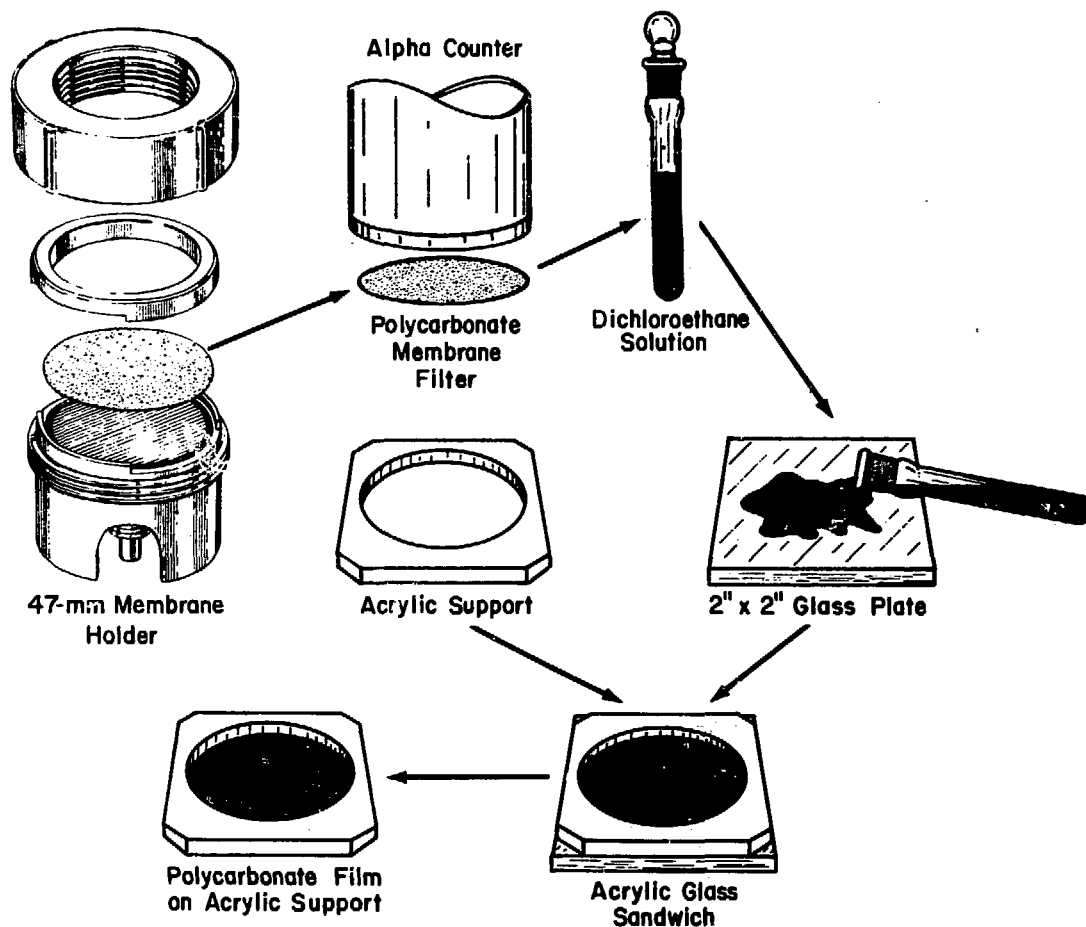


FIGURE 3
PROCEDURE FOR PREPARING POLYCARBONATE FILMS

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Ascarite, in turn, absorbed nitrogen dioxide to protect the pump and the dry test meter. A small flowmeter was mounted on the exhaust side of the dry test meter to give an indication of the instantaneous flow rate through the system. Air from the meter was fed back into the exhaust system to prevent its release to the service area.

Film Preparation

Figure 3 shows the procedure for converting the particle-containing filter membrane to a polycarbonate film. After air was sampled, the radioactivity retained on each filter was measured before it was handled in the laboratory. Each filter was then dissolved in a 40% (v/v) solution of 1,2-dichloroethane in dichloromethane.

The clear polycarbonate solution containing the particles was poured onto a clean, 50-mm (two-inch) square glass plate. One edge of a second 50-mm square glass plate was used to spread the solution evenly over the surface of the first plate. A 50-mm square acrylic support with a 45-mm diameter hole was placed on top of the wet film. The support and plate combinations were placed in covered petri dishes for 16 hours while the films continued to dry.

The glass plates were then removed by dipping the support and plate combinations in distilled water and prying the supports from the glass with tweezers.

Fission Fragment Track Production

The cast polycarbonate film was irradiated in a thermal neutron fluence of about 9×10^{14} neutrons per cm^2 . Films were arranged for irradiation by stacking the supports on top of each other thus sandwiching each film between two supports. Wrapped with each stack were preweighed Type 302 stainless steel disks. The induced radioactivity from 27-day ^{51}Cr in these disks was later measured to determine the thermal neutron fluence to which the particles were exposed.

The packaged stacks were irradiated in a light water-cooled, enriched uranium-fueled standard pile with graphite reflectors.⁴

To make the fission fragment tracks visible with an optical microscope, the polycarbonate film was etched for ten minutes in 6N NaOH at 52 to 55°C.

Alpha Track Production

To identify the fissionable material in each particle, the alpha particle emission rate was measured by coating the polycarbonate film with Kodak Type NTB nuclear-track emulsion (Kodak catalog number 164 4425). Under darkroom lighting, a 4-oz jar of emulsion was immersed in a water bath maintained at 40°C until the emulsion melted. The polycarbonate films were coated with emulsion by holding the supports containing the films vertically by one corner and dipping them into the clear molten emulsion for about one second. The coated films were then maintained at 28°C and about 80% relative humidity until the emulsion cooled and gelled (about 30 minutes).

To determine the alpha particle emission rate for each aerosol particle, the polycarbonate films were stored for one week before being developed. Spun

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aluminum *Desiccators** containing 60 grams of indicating *Drierite* were used to contain the films during this exposure. These were stored in a refrigerator between 4 and 5°C.

At the end of the exposure period, the alpha particle tracks in the emulsion were developed and all substances other than tracks were removed from the emulsion.

The film was prepared for track counting by placing the acrylic support on a 50-mm square, 1.0-mm thick polycarbonate block. Those particles having tracks were located under a microscope using a magnification of 105X. When found, the fission fragment and alpha particle tracks were counted using a magnification of 1000X.

This identification procedure was used to distinguish particle-bound plutonium from uranium. Highly enriched uranium mixtures will give a number of fission fragment tracks comparable to that of the plutonium mixtures. Even if there should be enough uranium to produce fission fragment tracks, mixtures of these isotopes would not produce alpha particle tracks. One femtocurie (fCi) of ^{239}Pu will produce about 22 alpha particles in a week and, when irradiated with a fluence of 8.64×10^{14} thermal neutrons/cm², will produce about 40 fission fragments. In a mixture of low-irradiation plutonium, the number of fission fragments produced will be increased to 53 with between 28 and 33 alpha particles depending on the age of the mixture. Only about half of these particles will produce tracks, yet this radiographic technique is much more sensitive than electron microprobe analysis, which is not sensitive to less than 10 fCi¹.

Particle Isolation

After a particle had been identified and the tracks counted, the particle was excised from the film in a polycarbonate square. At a magnification of 105X, two parallel cuts were made through the emulsion-coated film on either side of the particle using an ultra microlance. The film was then rotated through 90° and two more cuts made forming a square (see Figure 4a). The cut square was then probed in one corner by a tungsten needle, lifted from the film, and placed on a glass microscope slide (see Figures 4b, 4c, and 4d).

The emulsion layers were then removed from the polycarbonate square by placing a cover glass on top of the square. Water was introduced between the cover glass and slide using a glass microbrush. The emulsion was then removed by gently moving the cover glass a few mm from side to side (see Figure 4e); this rolled the swollen emulsion from the surface of the film, but not from the fission fragment tracks themselves. The cover glass was carefully lifted from the glass microscope slide, taking care not to lose the polycarbonate square containing the particle.

Particle Mounting

To mount a particle, the polycarbonate square was placed in a selected grid location on a beryllium sample mounting block** (Figure 4f). These sample mounting blocks were 25 mm in diameter and 13 mm thick and fit the standard electron microprobe sample holders, which grip the sides and provide the necessary electrical contact. The top surface of the block was highly polished and contains a grid

* Trademark of Fisher Scientific Company.

** Walter C. McCrone Associates, Inc. catalog number XIII-403.3.

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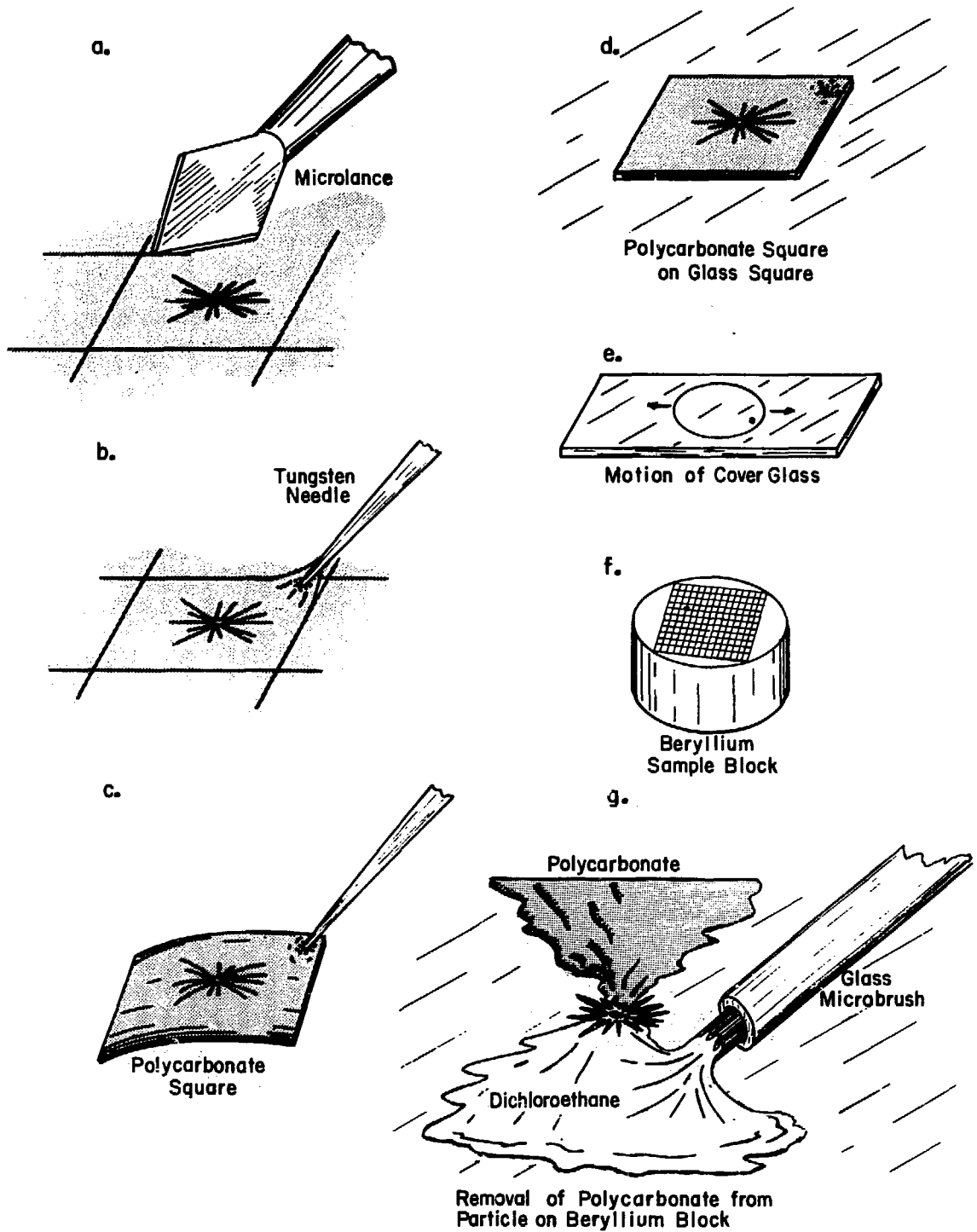


FIGURE 4
PROCEDURE FOR MOUNTING PARTICLE FOR FISSIONABLE MATERIAL IDENTIFICATION

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network of 1-mm squares inscribed on the surface. The squares were numbered in mirror image fashion both vertically and horizontally through the center.

The polycarbonate square was then dissolved and washed back from the particle using dichloroethane from a glass microbrush. Dichloroethane from the brush was dispensed on the beryllium block just in front of the polycarbonate square until the square was engulfed in the solution. The microbrush was then used to push the solution back from the particle. Gelatin replicas of the fission fragment tracks remained with the particles.

A Polaroid picture of each particle was made at a magnification of 556X to identify the particles after the gelatin had been removed.

The gelatin with each particle was oxidized by exposure to an oxygen plasma for three hours in a low-temperature asher.*

To maintain control of particles after the gelatin track replicas were oxidized, each particle was located and photographed again using Polaroid film and a magnification of 556X. An arrow was marked on the film pointing to the particle, so there would be no mistake in what was intended for analysis.

The size of each particle was estimated from these Polaroid pictures taken after oxidation of the completely denuded particles. An average of the smallest and largest dimensions of the photographed particle were measured in mm and divided by the magnification.

Analysis

The plutonium-bearing particles, mounted on beryllium blocks, were analyzed with an electron microprobe. The analyses were made at Arizona State University in Tempe, Arizona, with a Cemeica MS46 electron microprobe equipped with four crystal, wave-length-dispersive spectrometers (take-off angle of 18°) and an energy-dispersive analyzer.

Analytical data included measurements of x-ray intensities and estimates of particle size and shape. These data, along with estimated average densities, were used in the FRAME computer program⁵ as modified for particles work by Armstrong.⁶ These calculations gave the composition of each particle in both elements and oxide weight percents. Oxygen was not measured as such; elements were simply assumed to be present in the oxide form.

Grouping of Data by Enrichment Factors

The results were expressed in terms of "enrichment factors" (dimensionless ratios of elemental concentrations), which enabled the intercomparison of the compositions of plutonium-bearing particles with other atmospheric aerosols and the intracomparison among particles collected from different sampling points. A definition of enrichment factors and an explanation of their development and application in this work is given in the Appendix to this report.

To compare the chemical composition of the particles collected from Systems I and II with each other and with the average for global crustal aerosol, the

* Manufactured by International Plasma Corporation.

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particle analyses were grouped according to the level of the enrichment factors. Four groups were established for each element using the elemental concentration data in Table A-1 of the Appendix. The first group contained those particles which contained no detectable amounts of the element sought. The second group contained detectable amounts with enrichment factors less than one standard deviation below the geometric mean enrichment factor, $\overline{EF}_g/s_g$. The third group contained particles with enrichment factors between the lower and upper limits of one standard deviation from the geometric mean enrichment factor, $\overline{EF}_g/s_g$ and $\overline{EF}_g \cdot s_g$, respectively. The fourth group contained enrichment factors greater than one standard geometric mean enrichment factor, $\overline{EF}_g \cdot s_g$. The third column of Table I gives the percent of the particles analyzed which gave positive analyses for each element. The fourth, fifth, and sixth columns of Table I contain the percent of those having positive analyses which had enrichment factors less than, between, and more than the lower and upper limits of the geometric standard deviation.

To compare the chemical composition of particles collected at the various sample points in System II with each other and global crustal aerosol (Table A-I), this process was repeated and the results are listed in Table II.

Particles having no detectable amounts of an element were not counted with those with enrichment factors less than the lower limit for the geometric standard deviation (s_g) because there can be no zero or negative concentration of enrichment factor values in log-normal frequency distributions. Thus, the size of the three groups are expressed as the percent of the particles giving positive analyses, rather than the percent of the total number of particles.

Particle Evaluation by Size

In this study, particles were selected for analysis on the basis of their being recognizable by their radiating fission fragment tracks under a 160 X dissecting microscope used for their excision from the polycarbonate film. The film was first scanned and the location of the particles marked. A proportion of the particles were then excised without regard for the number fission fragment tracks present. Thus, there was some bias against particles having less than five tracks. The selection of particles for analysis, however, was not biased by physical size. The size of the particles was not measured until after the particles had been mounted and the polycarbonate film containing the tracks dissolved. Since the distribution of the number of tracks per particle appears to be log normal, the size distribution of the analyzed particles is indicative of the size distribution of particles in the aerosol carrying most of the plutonium.

Cumulative frequency plots were constructed for particles from Systems I and II. Particles in each system were first ranked in order of their approximate diameter in μm from the smallest to the largest. A list of the number of particles having successively larger diameters was made. A cumulative total of the number of particles at increasing diameter segments was calculated and then normalized by dividing by the total number of particles from each system. This gave the fraction of the particles having a diameter equal to or smaller than any particular diameter. Table III lists the particle diameters in μm ; and, in Columns 2, 3, 4, 5, and 6, the fraction of the particles having diameters equal to or less than each diameter measured in System I and Sampling Points A, B, C, and D in System II, respectively. These fractions are also plotted on the logarithmic probability graph given in Figure 5.

For comparison, a cumulative frequency plot was also made of the size distribution of particles in natural atmospheric aerosols.

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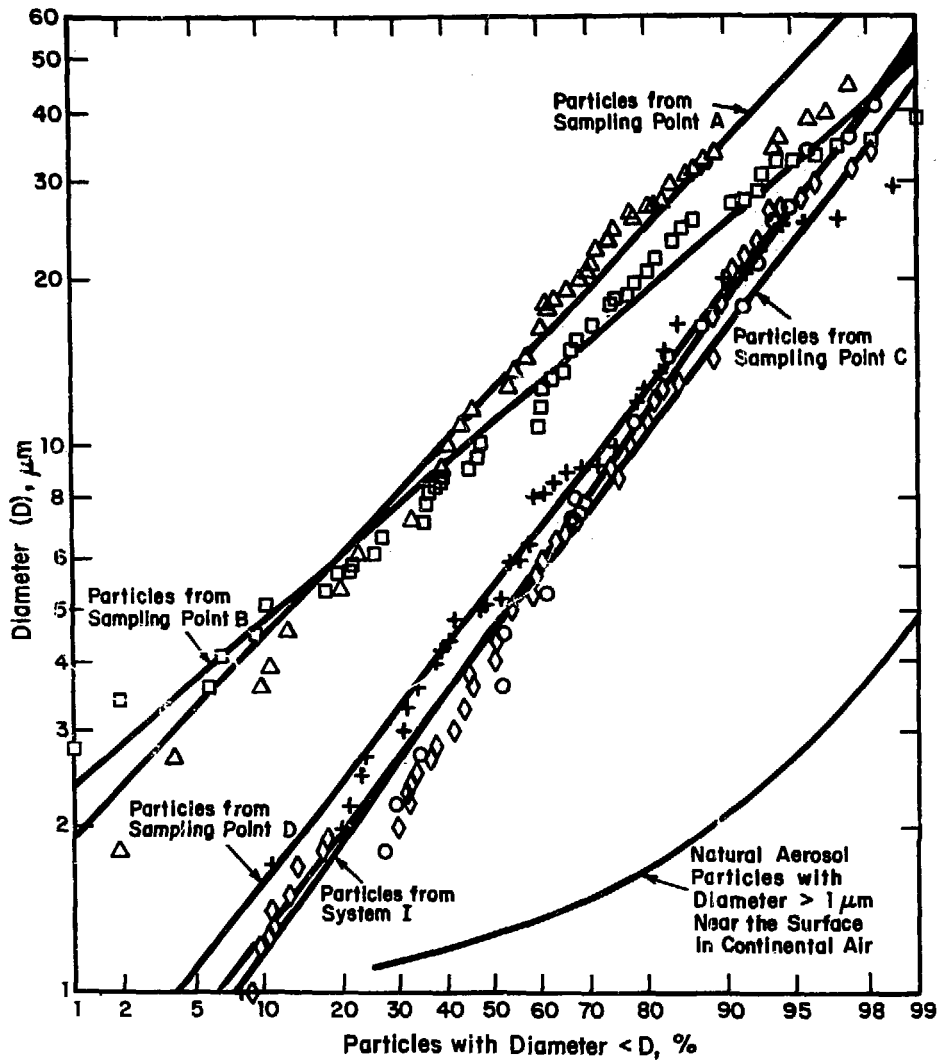


FIGURE 5
SIZE DISTRIBUTION PLOTS FOR NATURAL AND COLLECTED PARTICLES

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Table 1. Comparison of Analyses of Particles From Systems I and II.

Element	System	Positive Analyses, %	% of Positive Analyses ^a		
			Less Than ^b	Within ^c	Greater Than ^d
Si	I	100	47	24	29
	II	99	29	30	41
Al	I	84	0	100	0
	II	88	0	100	0
Fe	I	93	14	35	51
	II	79	36	33	31
Ca	I	70	53	30	17
	II	52	41	40	19
Na	I	70	13	72	15
	II	54	8	81	10
K	I	90	56	30	14
	II	63	35	41	24
Mg	I	51	24	59	17
	II	39	38	52	10
Ti	I	74	20	17	65
	II	31	12	13	76
P	II	1	0	17	83
Mn	I	10	0	0	100
	II	12	4	8	88
Ba	II	0.5	0	0	100
S	I	17	47	47	5
	II	70	28	60	13
Cl	I	34	13	67	21
	II	40	2	82	16
Cr	I	53	0	18	82
	II	29	0	9	91
Ni	I	56	2	25	73
	II	9	0	3	97
Zn	I	64	4	41	55
	II	45	5	52	43
Co	II	1	0	0	100
Sc	II	0.2	0	0	100
Cu	I	36	12	37	51
	II	7	6	29	65
W	I	1	0	0	100
	II	0.5	0	0	100
Cd	II	0.2	0	0	100

a. The percent of the positive analyses less than, within, and greater than one geometric standard deviation of the global geometric mean enrichment factor.

b. $EF < EF_g/s_g$.

c. $EF_g/s_g \leq EF \leq EF_g \cdot s_g$.

d. $EF > EF_g \cdot s_g$.

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Table II. Comparison of Analyses of Particles From Sampling Points A, B, C, and D of System II.

Element	Sampling Point	Positive Analyses, %	% of Positive Analyses ^a		
			Less Than ^b	Within ^c	Greater Than ^d
Si	A	99	35	33	32
	B	98	8	24	69
	C	100	36	31	33
	D	99	40	31	29
Al	A	79	0	100	0
	B	94	0	100	0
	C	89	0	100	0
	D	96	0	100	0
Fe	A	98	31	22	46
	B	100	40	39	21
	C	58	33	41	21
	D	49	46	34	20
Ca	A	56	20	44	36
	B	77	54	38	9
	C	41	45	40	15
	D	27	53	32	16
Na	A	55	18	60	22
	B	90	4	94	2
	C	39	5	82	14
	D	24	6	94	0
K	A	76	20	48	31
	B	73	46	44	10
	C	55	35	32	33
	D	37	54	27	19
Mg	A	63	33	18	9
	B	48	35	55	10
	C	17	47	42	11
	D	21	60	27	13
Ti	A	42	12	6	83
	B	27	10	24	66
	C	30	14	11	74
	D	15	9	18	73
P	A	2	0	50	50
	C	3	0	0	100
	D	1	0	0	100
Mn	A	7	13	0	88
	B	30	3	9	38
	C	5	0	0	100
	D	6	0	25	75
Ba	A	1	0	0	100
	B	1	0	0	100
S	A	58	30	54	17
	B	93	21	69	10
	C	66	24	61	15
	D	61	47	44	9

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Table II., Continued

Element	Sampling Point	Positive Analyses, %	% of Positive Analyses ^a		
			Less Than ^b	Within ^c	Greater Than ^d
Cl	A	43	4	47	49
	B	72	1	99	0
	C	27	0	97	3
	D	10	14	86	0
Cr	A	27	0	6	94
	B	58	0	15	85
	C	13	0	0	100
	D	14	0	0	100
Ni	B	27	0	3	97
	C	4	0	0	100
	D	7	0	0	100
Zn	A	53	14	35	51
	B	88	0	69	31
	C	22	4	24	72
	D	6	0	75	25
Co	B	5	0	0	100
Sc	C	1	0	0	100
Cu	A	22	7	33	59
	B	3	0	0	100
	C	1	0	0	100
W	A	1	0	0	100
	B	1	0	0	100
Cd	D	1	0	0	100

a. The percent of the positive analyses less than, within, and greater than one geometric standard deviation of the global geometric mean enrichment factor.

b. $EF < \overline{EF}_g / s_g$.

c. $\overline{EF}_g / s_g \leq EF \leq EF_g \cdot s_g$.

d. $EF > \overline{EF}_g \cdot s_g$.

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Table III. Comparison of Size Distributions of Particles From Systems I and II With Natural Aerosols^a.

Diameter (D), μm	Fraction With Diameter $\leq D$					Natural Aerosol
	System I	Sampling Point A	Sampling Point B	Sampling Point C	Sampling Point D	
0.4	0.03			0.01	0.01	
0.5	0.04			0.02		
0.9	0.07			0.04	0.06	
1.1				0.09	0.08	0.25
1.2				0.10		0.42
1.4				0.11		0.64
1.7				0.14	0.11	
1.8	0.28	0.02		0.17		0.83
2.2	0.30			0.32	0.21	0.91
2.5				0.34	0.23	
2.7	0.35	0.04	0.01	0.37	0.24	0.949
3.0				0.42	0.31	
3.3				0.43	0.32	
3.6	0.53	0.10	0.06	0.46	0.34	0.979
3.9		0.11				0.983
4.0			0.07	0.51	0.38	
4.4				0.52	0.41	
4.5	0.54	0.13	0.10			0.989
5.0			0.11	0.55	0.48	
5.4	0.62	0.20	0.18	0.60		0.994
5.8			0.20	0.59		
6.1			0.22	0.61	0.56	
6.3		0.23	0.26			0.996
6.7			0.27	0.64	0.58	
7.0				0.67		
7.2	0.67	0.33	0.35	0.68		0.997
7.4				0.68		
7.8			0.36	0.70		
8.0	0.68	0.34	0.37	0.71	0.59	0.998
8.6			0.39		0.63	
9.0	0.75	0.40	0.46	0.74	0.69	0.999
10.0		0.41	0.48	0.78	0.75	
10.8	0.79	0.44	0.60	0.81		
11.7		0.46	0.61	0.82	0.79	
12.6	0.83	0.54	0.62	0.83	0.80	1.000
13.5		0.55	0.66		0.82	
14.4	0.84	0.58		0.89		
14.9			0.67		0.83	
16.2	0.88	0.61	0.71			
17.1		0.62		0.89	0.89	

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Table III., Continued

Diameter (D), μm	Fraction With Diameter $\leq D$					Natural Aerosol
	System I	Sampling Point A	Sampling Point B	Sampling Point C	Sampling Point D	
18.0	0.92	0.64	0.75	0.90		
20.7		0.70	0.81	0.91	0.92	
21.6	0.93	0.71	0.82			
23.4		0.74	0.85	0.930	0.930	
24.3		0.75	0.86			
25.2	0.94	0.78	0.87		0.972	
26.9	0.95	0.81	0.91	0.939		
27.9		0.83	0.92			
28.8			0.93	0.956	0.986	
30.6		0.86	0.935			
31.5		0.87		0.974		
32.4		0.88	0.944			
33.5		0.89	0.963			
34.2	0.959	0.94	0.972	0.982		
35.1				1.000	1.000	
36.0	0.975	0.944	0.981			
39.6		0.968	0.991			
41.4	0.983		1.000			
50.4	0.992					
53.9		0.992				
59.4	1.000					
62.9		1.000				

- a. The percent of the positive analyses less than, within, and greater than one geometric standard deviation of the global geometric mean enrichment factor.

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A very simple function that has been used extensively in atmospheric research to express particle size distribution in both natural and polluted atmospheres is

$$\frac{dN}{dD} = aD^{-b} \quad (1)$$

where N is the number concentration or total number of particles per unit volume having diameters from the lower limit of definition of aerosols up to diameter D in μm . From the relationships

$$dD = D d(\ln D) \quad (2)$$

and

$$\ln D = \ln 10 \cdot \log D \quad (3)$$

the more useful expression

$$\frac{dN}{d(\log D)} = (\ln 10) a D^{-c} \quad (4)$$

is obtained where $c = b - 1$, and $dN/d(\log D)$ is called the number distribution. Junge⁷ found c to be about 3 over the size range $-0.7 < \log D < 1.5$ or $0.2 < D < 32 \mu\text{m}$. Integrating the first equation between D_0 and D ($D_0 < D$) gives

$$N = \left[\frac{aD^{-c}}{c} \right]_{D_0}^{D_0} = \frac{a}{3} \left[\frac{1}{D_0^3} - \frac{1}{D^3} \right] \quad (5)$$

Instead of expressing the distribution as the number of particles per unit volume, it can be expressed as a fraction, F , of the total number of particles or

$$F = \frac{N}{N_T} = 1 - \left[\frac{D_0}{D} \right]^3 \quad (6)$$

where N_T is the total number of particles when $D = \infty$, and $N_T = a/3D_0^3$. To obtain a reasonable distribution, only those particles which could be easily seen with an optical microscope were included. Thus D_0 was assumed to be $1 \mu\text{m}$, and Equation 6 can be expressed as

$$F = 1 - \frac{1}{D^3} \quad (7)$$

The frequency distribution for natural aerosols with particles diameters between $1 \mu\text{m}$ and D , calculated from this expression, is given in Column 7 of Table III and plotted in Figure 5.

To see how closely the distribution of particle diameters resembles a log-normal distribution, the assumption was made that the observed diameters represent a sample of a population having a log-normal distribution. The geometric mean diameter, $\bar{D}_g$, and geometric standard deviation, s_g , were calculated from these data using equations similar to those given earlier for the geometric mean enrichment factor and geometric standard deviation. These values are given in Table IV. Values for the upper 68.27% limit for the diameters were calculated from the product of $\bar{D}_g$ and s_g . The best fit log-normal probability curves were plotted on the

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logarithmic probability graph in Figure 5 by drawing straight lines through coordinates for $\bar{D}_g$ and $\bar{D}_g \cdot s_g$ on the 50.00 and 84.14* cumulative percent abscissae, respectively.

To determine the degree of asymetry, the skewness (SK) of these frequency distributions was calculated using the relationship

$$SK = 3 \left(\frac{\ln \bar{D}_g - \ln d_{med}}{\ln s_g} \right) \quad (8)$$

where D_{med} = the median diameter. A perfect log-normal distribution has a skewness of zero. If a distribution has a higher tail to the right than to the left, it is positively skewed.

Table IV. Distribution of Particle Diameters in Systems I and II.

System	Sample Location	Data Points, N	Geom Mean Diameter, D_g	Geom Std Deviation, S_g	Skewness, SK
I		121	4.64	2.92	0.71
II	A	125	12.27	2.24	0.04
II	B	107	10.82	1.93	0.34
II	C	114	4.48	2.75	0.37
II	D	71	5.43	2.69	0.23

Particle Evaluation by Plutonium Content

Another characteristic studied was the distribution of plutonium among the particles as indicated by the observed number of fission-fragment tracks in the surrounding polycarbonate.

The track distribution among particles from both systems was evaluated in the same way as the particle diameters. The fraction of the particles with the number of tracks equal to or less than a selected number, T, are given for Sampling Points A, B, C, and D in Table V. Figure 6 is a logarithmic probability plot of cumulative percent of particles from each of these sampling points. Figure 7 is a similar plot for particles from four locations in System I. The calculated geometric mean for the number of fission-fragment tracks per particle. The geometric standard deviation, and the skewness for particles from each sampling point are given in Table VI. Best fit log-normal probability curves for each distribution are plotted in Figures 6 and 7. For the comparison of the track distributions for particles from the various sampling points in System I with those from System II, the probability curve for the track distribution for particles from Sampling Point A in System II is plotted with the distributions from System I in Figure 7.

* $50.00 + \frac{68.27}{2}$.

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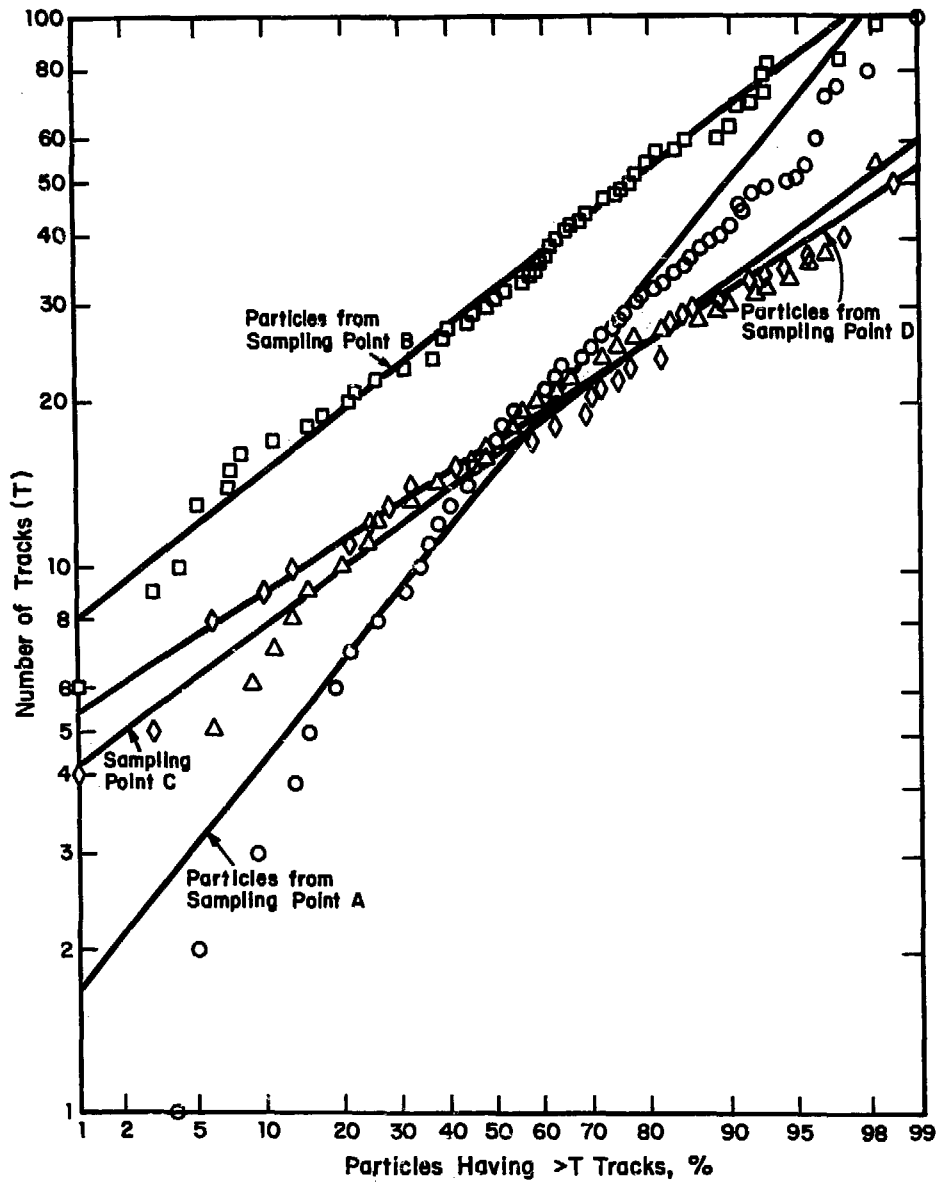


FIGURE 6
DISTRIBUTION OF THE NUMBER OF TRACKS PER PARTICLE FOR
PARTICLES COLLECTED FROM SAMPLING POINT A, B, C, AND D IN SYSTEM II

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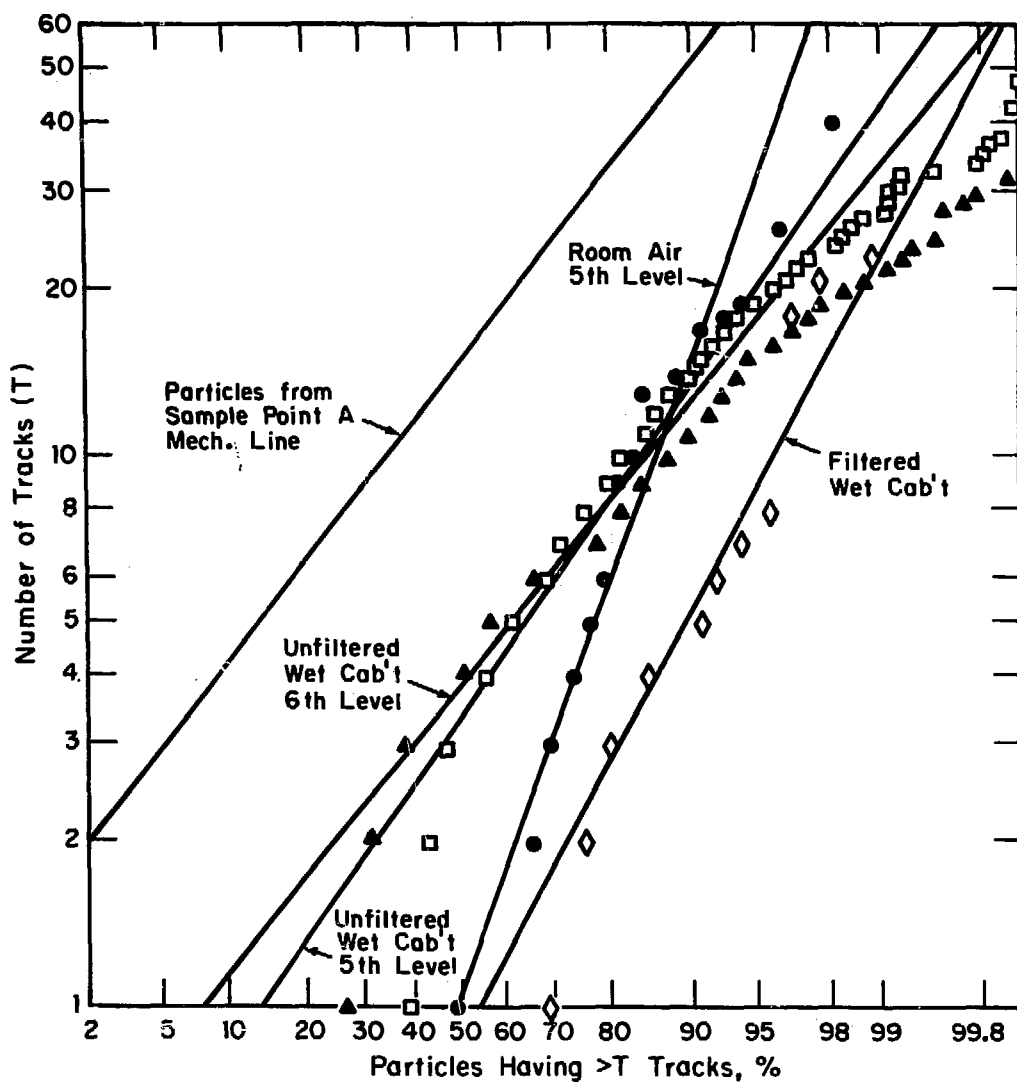


FIGURE 7
DISTRIBUTION OF THE NUMBER OF TRACKS PER PARTICLE
FOR PARTICLES COLLECTED FROM THE MECHANICAL LINE
(SAMPLING POINT A), WET CABINETS, AND ROOM AIR

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Table V. Distribution of Fission Tracks Among Plutonium-Bearing Particles Collected From Sampling Points A, B, C, and D.

Number of Tracks	Fraction With Tracks $\leq T$			
	Sample Point A	Sample Point B	Sample Point C	Sample Point D
1	0.04			
2	0.05			
3	0.09			
4	0.13			0.01
5	0.15		0.06	0.03
6	0.19	0.01	0.09	
7	0.21		0.11	
8	0.26		0.13	0.06
9	0.31	0.03	0.15	0.10
10	0.34	0.04	0.20	0.13
11	0.36		0.24	0.21
12	0.38		0.26	0.25
13	0.40	0.05	0.32	0.28
14	0.44	0.07	0.38	0.32
15	0.45	0.07	0.44	0.42
16	0.47	0.08	0.48	0.46
17	0.50	0.11	0.49	0.58
18	0.51	0.15	0.54	0.63
19	0.54	0.17	0.56	0.69
20	0.59	0.21	0.59	0.70
21	0.60	0.22	0.63	0.72
22	0.63	0.26	0.66	0.75
23	0.64	0.31	0.70	0.77
24	0.68	0.37	0.72	0.82
25	0.70		0.75	
26	0.72	0.39	0.78	
27	0.74	0.40	0.82	
28	0.75	0.44	0.87	0.83
29	0.76	0.45	0.89	0.85
30	0.78	0.48	0.90	0.86
31	0.79	0.50	0.92	0.89
32	0.81	0.52	0.93	
33	0.82	0.56	0.947	0.92
34	0.84	0.58		0.93
35	0.85	0.59		0.944
36	0.86	0.60	0.956	
37		0.61	0.965	0.958
38	0.87	0.62		
39	0.88			
40	0.89	0.63		0.972

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Table V., Continued

Number of Tracks	Fraction With Tracks $\leq T$			
	Sample Point A	Sample Point B	Sample Point C	Sample Point D
41		0.65		
42	0.90	0.66		
43		0.68		
44	0.91	0.69		
46	0.91			
47		0.73		
48	0.92	0.75		
49	0.93	0.76		
50	0.945	0.77		0.986
51	0.950			
52		0.78		
54	0.955		0.991	
55		0.80		
57		0.81		
58		0.84		
59		0.85		
60	0.960	0.89		
63		0.90		
65			1.000	
68		0.91		
70		0.92		
72	0.965			
73		0.93		
75	0.970			
80	0.980	0.93		1.000
82		0.953		
84		0.972		
98		0.981		
100	0.990	0.991		
150	0.995	1.000		
200	1.000			

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Table VI. Distribution of Fission Tracks Among Plutonium-Bearing Particles from Various Sources in Systems I and II.

Source	Data Points, N	Geom Mean of No. of Fission Tracks, $\bar{T}_g$	Geom Std Deviation, s_g	Skewness, SK	Geom Mean Activity Particle, fCi
<u>System I</u>					
Unfiltered 5th Level Wet-Cabinet Air	15,987	3.76	2.56	-0.20	0.09
Unfiltered 6th Level Wet-Cabinet Air	7,042	3.32	2.99	-0.51	0.08
5th Level Room Air ²	53	1.00	8.40	-0.98	0.02
Filtered Wet-Cabinet Air ^a	98	0.87	4.14	-0.29	0.02
<u>System II</u>					
Sampling Point A Air	200	14.74	2.69	-0.43	0.36
Sampling Point B Air	107	32.38	1.78	0.23	0.79
Sampling Point C Air	114	16.50	1.75	-0.22	0.40
Sampling Point D Air	71	17.01	1.65	0.24	0.41

a. Values determined graphically.

Discussion

The most abundant elements in average crustal rock (and soil) are oxygen (46.60%), silicon (27.72%), aluminum (8.13%), iron (5.00%), calcium (3.63), sodium (2.83%), potassium (2.59%), magnesium (2.09), and titanium (0.44%).⁸ With the exception of oxygen, which was not detected by electron microprobe analyses, these elements are also found in most inorganic particles (Tables I and A-I). This supports the idea that most plutonium-bearing particles are airborne crustal material to which minute quantities of plutonium have become attached.

Of particular interest is the quantity of ^{239}Pu contained on these particles. One femtocurie (1 fCi = 10^{-15} Ci) of ^{239}Pu irradiated under the conditions described here should produce 41 fission-fragment tracks. The minimum detection limit for electron microprobe analysis of plutonium is about 0.2 picograms or about 10 fCi of ^{239}Pu ,¹ which is equivalent to 410 fission-fragment tracks. Because of this relatively low sensitivity of electron microprobe analysis, plutonium could be detected by this method in only one of the 558 particles selected for analysis, even though all the particles produced fission-fragment tracks. This single particle was a small, 1- μm -diameter particle, collected from unfiltered wet-cabinet exhaust. It contained 73% PuO_2 by weight (equivalent to 170 fCi of ^{239}Pu) in combination with Fe_2O_3 and mica.

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Of the major crustal elements listed in Table I, silicon and iron were the most ubiquitous being found in most particles. The enrichment factor distribution for these elements, however, does not fall within the log-normal distribution for crustal material. For the enrichment factors of an element to match the log-normal distribution of crustal material in aerosols, there should be about 16% of the enrichment factors of less than one geometric standard deviation, 68% within one geometric standard deviation of the mean, and another 16% above one geometric standard deviation. This lack of conformity may result from the low values for the geometric standard deviations of the enrichment factors for these elements in aerosols.

Only the enrichment factors for sodium and chlorine fall within the log-normal distribution for crustal material. This may be due to the relative high solubility of compounds of these elements and, in the case of chlorine, the high value for the geometric standard deviation.

Particles from System I contain a greater variety of elements than those from System II and thus all but four elements are contained on a higher proportion of particles from System I than from System II. The most striking example was nickel. While 56% of the particles from System I contained nickel, only 9% of those in System II did. The major crustal elements (those in Table A-I comprising 0.4% or more of crustal material) are contained on over half the particles from System I and with the exception of magnesium in particles from sampling Points C and D and titanium are also contained on over half the particles from System II. Some of the minor elements (those comprising 0.1% or less of crustal material) are present in over half the particles, viz, nickel, chromium, and zinc in particles from System I and sulfur, chromium, and zinc in particles from sampling point B of System II. The chromium and nickel may have come from the 304L stainless steel alloy of cabinets and exhaust ducts or the *Hastelloy**-C alloy in the wet cabinets. However, few of the particles contained the proper ratio of chromium-to-nickel found in either alloy. Also, if *Hastelloy*-C contributed the nickel in the particles, some molybdenum should also have been detected.

Of the elements which are present on less than 10% of the particles, all but copper on particles from System II have high enrichment factors. This indicates that the minor constituents of crustal material are not uniformly distributed among particles but are concentrated on a few particles where they represent a major constituent.

The plutonium-bearing particles were larger than natural aerosol particles collected at relatively low altitude (<2.3 km) as seen in Figure 5. Those particles collected from sampling points A and B of System II were larger than those from System I, with geometric mean diameters two or three times those of particles from other locations.

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Particles from all parts of System II also contained on the average more plutonium per particle than those from System I. As seen in Table VI, the geometric mean of the activity in System I, containing only HEPA filters, was a factor of 10 to 100 less than that of System II, which uses a large sand filter before the air is exhausted to the atmosphere. The geometric mean of measured activity per particle released to the atmosphere was 0.4 fCi.

A comparison of the mean diameters of particles collected from different sampling points, given in Table IV, with the mean number of fission fragment tracks for particles from the same location, given in Table VI, indicate a possible relationship between particle size and plutonium content. Correlation coefficients between the cube of the particle diameter and the number of fission fragment tracks from each particle from sampling Points B, C, and D were calculated. These are given in Table VII. These coefficients differ significantly from that expected from a random sample from a population of paired variables having a correlation coefficient of zero. Thus, even though the points on a plot of particle diameter cubed versus number of fission fragment tracks appears scattered, there is a significant correlation between the quantity of plutonium in particles collected from Sampling Points B, C, and D in System II and the particle volume. (Tracks with particles collected at other sampling points, where only ^{239}Pu could be found, were counted but not recorded for each particle. Only where a ratio of alpha particle to fission fragment tracks was needed to distinguish plutonium bearing particles from those having other fissionable materials were the track counts recorded).

Table VII. Correlation and Coefficient of Alienation for the Cube of the Diameter and the Number of Fission Fragment Tracks for Particles From Sampling Points B, C, and D of System II.

<u>Sampling Point</u>	<u>Number of Particles</u>	<u>Correlation Coefficient</u>
B	107	0.69
C	114	0.29
D	71	0.36

Summary

Plutonium-bearing particles collected from a nuclear reprocessing plant exhaust were found to be aggregates of crustal material. High enrichment factors for metals were evidence of the anthropogenic nature of some particles. The radioactivity of the plutonium associated with individual particles was very small (~ 0.4 fCi).

The results of this work indicate the need for further study of the behavior of these plutonium-bearing particles following their release to the atmosphere and subsequent interaction with the environment.

Acknowledgment

The author gratefully acknowledges the assistance of E. F. Holdsworth and J. T. Armstrong of the Chemistry Department of the Arizona State University in Tempe, Arizona, who performed the electron microprobe analyses.

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Appendix: Use of Elemental Enrichment Factors to Express Particle Compositions

Background

Two recent developments in aerosol studies have provided valuable tools for the analysis of particle composition data. The first is the use of ratios of elemental concentrations called "enrichment factors" to compare aerosol compositions. Begun in the early seventies, this technique has gained wide acceptance in the last few years⁹⁻¹³. The second development is the availability of data on the composition of natural aerosols. Rahn⁹ published a compilation of 104 data sets of trace elements in aerosols along with the geometric mean and geometric standard deviation of the enrichment factors for each of the elements. These data sets were from sampling sites ranging from highly industrialized temperature zones to the tropics and poles, and represent all continents except South America, as well as various marine locations. As a framework from which to view much of the order in atmospheric aerosols, Rahn used the concept of aerosol-crust enrichment factors for the elements. This concept has been applied to analyzing data collected in this study to provide for (a) the intercomparison of the compositions of plutonium-bearing particles with atmospheric aerosols compiled by Rahn and (b) the intracomparison among particles collected from different sampling points.

Microprobe Analyses of Particles

To be comparable, results of microprobe analyses must be expressed as elemental ratios. The reason for this is that not all elements which may be present in an aerosol are detected by microprobe analysis. The microprobe used in this study is quantitative only for elements with atomic numbers greater than 10. It is only semi-quantitative for oxygen (the most abundant element in crustal material) as well as other major elements of low atomic number such as hydrogen, fluorine, and carbon. Atmospheric aerosols are known to contain in addition to elements and oxides, carbonaceous material such as sooty carbon and organics and water-soluble ionic material such as sulfate, nitrate, and ammonium ions. Thus elemental weight percents, normalized to 100 based on the elements detected cannot be compared. Even the addition of a hypothetical oxygen concentration, calculated on the supposition that all elements are present as oxides of known valence, will still not account for the organic fraction of particles. However, a ratio of the concentrations of one element to another will normally be relatively unaffected by the concentrations of other elements which may be present and thus can be used for comparisons even when a complete analysis of all the elements in an aerosol or single particle is not available.

Enrichment Factors

A dimensionless ratio of elemental concentrations, called the enrichment factor, has been defined as

$$EF(X) = \frac{(X/Ref)_{\text{aerosol}}}{(X/Ref)_{\text{source}}} \quad (A-1)$$

where EF(X) is the enrichment factor of element X in an aerosol relative to some source material. X/Ref is the ratio of the concentration of element X to the concentration of the reference element, Ref, in both the aerosol and the source material.

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Source Material

Elemental ratios in aerosols or in single particles are normalized by dividing them by ratios of the same elements in a standard source material to obtain the enrichment factors. If a particle is composed of the same material as the source, the enrichment factor will be 1.00 for all elements. If the ratio of an element to the reference element is greater or less than the same ratio in the source material, the enrichment factors will be greater or less than 1.00, and the particle is said to be either enriched or depleted, respectively, in that element.

The most commonly-used crustal source material for continental enrichment-factor calculations is globally-averaged crustal rock. (For marine enrichment-factor calculations, sea salt is used.) Because the composition of plutonium-bearing particles are compared with data reported by Rahn,⁹ the same crustal-rock composition used by him (that reported by Mason)⁸ was selected as the source material composition for this work. Column 2 of Table A-I gives the elemental concentrations in globally-averaged crustal rock for those elements found in plutonium-bearing particles.

Reference Element

Of the various elements which seem to be reliably crust-derived in aerosols, aluminum is generally considered to be the most suitable reference elements. (When sea salt is the source material, the nearly universal choice is sodium.) Aluminum is a major element (81,300 ppm in rock), well-determined by a variety of analytical techniques, and has a minimum of specific pollution sources.

Thus for this work, enrichment factors for element X in most particles were calculated using

$$EF(X) = \frac{(X/Al)_{\text{particle}}}{(X/Al)_{\text{rock}}} \quad (A-2)$$

with aluminum as the reference element and average crustal rock as the source material. However, 18 particles from System I and 48 from System II contained no aluminum. Thus the enrichment factors had to be based on silicon rather than aluminum where

$$EF(X) = \frac{(X/Si)_{\text{particle}}}{(X/Si)_{\text{rock}}} \cdot \frac{(\overline{Si}/Al)_{\text{g aerosol}}}{(Si/Al)_{\text{rock}}} = 0.79 \frac{(X/Si)_{\text{particle}}}{(X/Si)_{\text{rock}}} \quad (A-3)$$

(The second set of ratios is the geometric mean of the global aerosol-crust enrichment factor explained in the next section.)

Using these two relationships, the enrichment factors were calculated from the elemental weight percents obtained for 115 particles in System I and 415 particles in System II. Six small (0.5 to 3.6 μm diameter) iron particles in System I and two particles [$\sim 15 \mu\text{m}$ diameter containing K, Cr, and Fe (1:3:3)] from Sample Point A of System II contained neither aluminum nor silicon and were thus not included in the study.

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TABLE A-I. Elemental Concentrations in Average Crustal Rock and Geometric Mean Enrichment Factors of Various Aerosols

Element	Conc., ppm	Geometric Mean Enrichment Factors					
		Global $\overline{EF}_{g/sg}$	Global $\overline{EF}_g$	Global $\overline{EF}_{g \cdot s_L}$	Remote Marine $\overline{EF}_g$	Remote Conti- nental $\overline{EF}_g$	Urban $\overline{EF}_g$
Si	277,200	0.62	0.79	1.01	0.7	0.7	0.79
Al	81,300	1.00	1.00	1.00	1.0	1.0	1.00
Fe	50,000	1.05	2.06	4.06	2.5	1.5	2.2
Ca	36,000	1.15	2.84	7.04	8.0	1.5	2.9
Na	28,300	0.64	4.44	30.8	$10^2 - 10^3$	0.4	1.81
K	25,900	0.99	1.98	3.98	6.0	1.5	1.63
Mg	20,900	0.64	2.38	8.90	$10^1 - 10^2$	0.7	2.0
Ti	4,400	1.01	1.39	1.92	1.2	1.2	1.63
P	1,050	0.79	2.63	8.71	-	-	2.6
Mn	950	1.45	3.91	10.5	3.0	2	3.2
Ba	425	2.61	5.50	11.6	-	~2	4.8
S	260	228.0	608.0	1620.0	-	-	490.0
Cl	130	100.0	740.0	5470.0	$10^4 - 10^5$	40.0	300.0
Cr	100	2.50	8.11	26.3	20.0	6	6.2
Ni	75	8.74	31.9	116.0	100.0	50	10.8
Zn	70	79.7	257.0	832.0	400.0	80	300
Co	25	0.91	3.52	13.6	4.0	1.5	4.6
Sc	22	0.59	1.17	2.34	0.8	0.8	0.60
Cu	55	34.0	102.0	304.0	150.0	20.0	149
U	1.8	0.92	2.87	8.93	-	-	2.87
W	1.5	4.89	19.1	74.3	-	-	11.0
Cd	0.2	274.0	1920.0	13,400.0	5000.0	2000.0	940.0

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Comparative Aerosol Data

To compare the elemental composition of plutonium-bearing particles with that of atmospheric aerosols, enrichment factors calculated for elements in these particles were grouped according to data supplied by Rahn⁹ for aerosols. In his report, trace element concentrations in aerosols from 104 published and unpublished data sets were used to calculate enrichment factors. From the enrichment factors in each data set, the geometric mean enrichment factor ($\overline{EF}_g$) and geometric standard deviation (s_g) of the logarithmic frequency distributions of enrichment factors were calculated for each element using the following formulae:

$$\overline{EF}_g = \exp \left[\frac{1}{N} \sum_{i=1}^N \ln EF_i \right] \quad (A-4)$$

and

$$s_g = \exp \left\{ \left[\frac{1}{N-1} \sum_{i=1}^N (\ln EF_i - \ln \overline{EF}_g)^2 \right]^{\frac{1}{2}} \right\}$$

where N = the number of data points

EF_i = the enrichment factor of the i th point.

The geometric mean enrichment factors obtained by Rahn⁹ for 19 elements are given in Table A-I for global, remote marine, remote continental, and urban aerosols. The geometric means of the global aerosol enrichment factors include data from all points and may be weighted too heavily toward cities, but they can serve as a useful first approximation to a general aerosol. The urban enrichment factors are geometric means for 29 cities. The enrichment factors for remote continental and remote marine areas were read from the enrichment-factor plots and are therefore somewhat subjective.

To obtain the lower and upper limits for 68.27% of the enrichment factors closest to the geometric mean, values for $\overline{EF}_g/s_g$ and $\overline{EF}_g \cdot s_g$, respectively, were calculated using global values. (When describing concentrations at selected statistical levels remote from a mean, the s_g is a multiplier or divider of the $\overline{EF}_g$, whereas its counterpart Gaussian standard deviation functions as an increment to the arithmetic mean. This is a consequence of the fact that multiplying and dividing values is equivalent to adding and subtracting their logarithms.) The results from these calculations are also given in Table A-I.

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AGGLOMERATION CHARACTERISTICS OF FAST REACTOR HCDA AEROSOLS*

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Abstract

The behavior of vaporized mixed oxide fuel aerosols postulated to result from fast reactor core disruptive accidents is a subject which is intensely evaluated in fast reactor safety analysis, containment design, and site selection licensing procedure.

In this program, surrogate uranium oxide aerosols produced by vapor condensation of super-heated liquid UO_2 have been produced in a variety of ways and have been intercompared with respect to particle morphology, number concentration, deposition behavior, surface area, oxygen content, and diffusion, sedimentation, and agglomeration rates. It has been established that, within a relatively narrow range, the primary particles are nearly identical when examined for initial shape, and size distribution, and effective density. However, primary particles in those aerosols produced by the most rapid heating methods followed by nearly instantaneous cooling are smaller in size and generally spherical, while those produced by slower processes in combination with delayed cooling give rise to a large proportion of crystalline particles which result in more tightly bound agglomerates. Deposition behavior is not significantly affected by this difference.

Stokes diameters, which can be rather accurately calculated from the rate of settling, compare favorably with aerodynamic mean diameters measured with inertial devices. Agglomerate sizes appear to follow the cube root of the maximum initial concentration.

The HAARM-2 version of the HAA-3 aerosol code has been used to obtain detailed time-dependent predicted behavior on a reference closed volume. The interrelations observed suggest that, in a given system, many of the characteristics can be calculated after only a few cases have been experimentally measured.

I. Introduction

A relatively long-standing program in the study of uranium oxide vapor-condensation aerosols has been conducted intermittently at ORNL since 1960, when the initial interest was in the safety analysis of the proposed air-craft nuclear propulsion reactor. This project, although short-lived, served to place some initial values on the partitioning of volatile fission products elements from molten UO_2 .

Continuing through the 1960s as part of the AEC-LWR Nuclear Safety Studies, a long series of fission product release experiments

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were conducted. These included both high-temperature annealing and melting experiments in which the vaporization of UO_2 was relatively minor. The real emphasis on reactor fuel vaporization was initiated by the Division of Reactor Safety Research in support of the larger LMFBR Aerosol Release and Transport (ART) Program. The overall goal of this total program was to provide the analytical techniques and experimental data necessary to assess the transient behavior and the radionuclide release from breeder reactor cores as a result of postulated events of varying severity, up to and including severe hypothetical core disruptive accidents (HCDAs).

II. Experimental Program

The generation of UO_2 and U_3O_8 vapor-condensation aerosols has been investigated by a number of different techniques in order to simulate specific environmental factors and to provide a basis for the evaluation of various separate effects, particularly specific energy input on primary size distribution, and the comparative agglomeration behavior vs aerosol mass concentration from both instantaneous and continuous generating sources. Five different aerosol generation processes have been used in the various experiments:

1. instantaneous UO_2 vaporization by the arc-imaging (solar-type) furnace,
2. instantaneous UO_2 vaporization by capacitor discharge in premelted UO_2 ,
3. continuous U_3O_8 vaporization by the pyrophoric oxidation of molten uranium metal sparged with oxygen,
4. continuous UO_2 vaporization by superheating molten UO_2 in a dc electric arc cold-hearth furnace, and
5. continuous UO_2 vaporization incidental to melting UO_2 in tungsten crucibles heated by RF induction.

Four of these methods are described here in varying details with illustrations of typical results. Only the UO_2 vaporization incidental to UO_2 melting is not discussed further. Details are given elsewhere.⁽¹⁾

Instantaneous Vaporization of UO_2 by the ADL Arc-Imaging Furnace

While this method is not literally "instantaneous," it has the essential characteristics of the process in that only a very small hot zone is produced, similar to the laser method, and the resultant vapor is almost instantly quenched. The sample of UO_2 was placed at the secondary focus of the reimaging mirror (Fig. 1) and was heated to partial melting for 1 to 2 min.

Vaporized uranium oxide particles that collected on Millipore filters during the melting were examined in the electron microscope. The replication method was used to observe particles from specimens melted in air and in helium. It was confirmed by the transmission

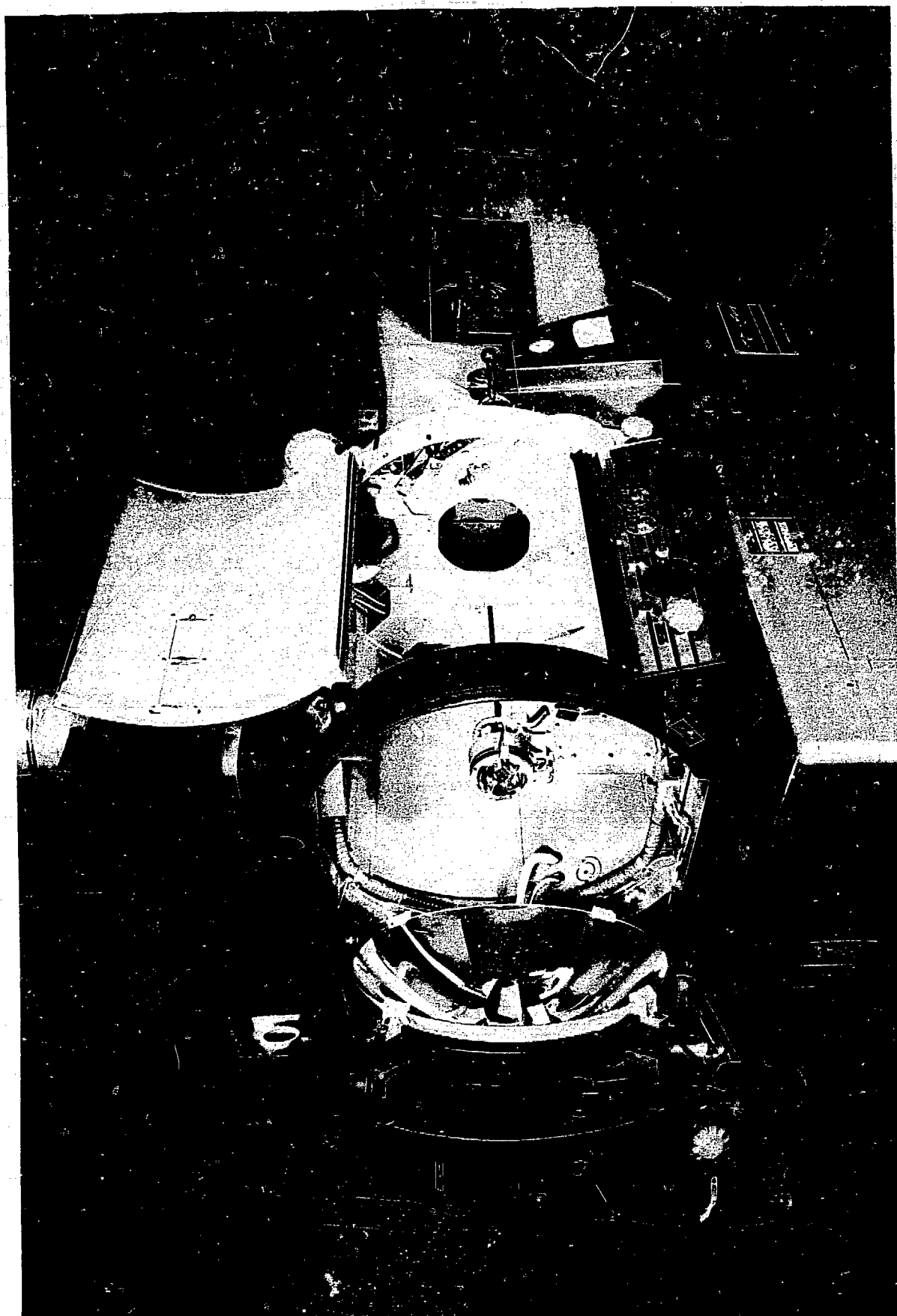


Fig. 1. ADL Carbon Arc-Imaging Furnace

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electron diffraction technique that the oxide collected from melting UO_2 in air is composed of crystalline U_3O_8 (Fig. 2a), while that collected from melting in helium consists of somewhat spherical shapes of UO_2 (Fig. 2b). Ninety-seven percent of the crystalline U_3O_8 particles had a diameter greater than 0.025μ . The average diameter of the U_3O_8 particles was about 0.1μ , while the UO_2 in helium measured between 0.0075 and 0.03μ (averaging 0.01μ). The more rapid cooling that would occur in helium possibly accounts for the smaller spherical shapes. The unusual characteristics of the U_3O_8 particles as seen, completely dispersed, reflect the almost instantaneous collection on a filter before any detectable agglomeration had occurred.

Instantaneous UO_2 Vaporization by High-Energy Capacitor Discharge in Pre-melted UO_2

Development of the capacitor discharge vaporization (CDV) system⁽²⁾ was patterned after the early work at Karlsruhe by Schaikarski and coworkers.⁽³⁾ The approach at ORNL was enhanced by incorporating the suggestions offered by Dr. Schaikarski, which included the direct electric heating of a UO_2 column to nearly complete melting prior to capacitor discharge.

With the CDV technique, ceramic fuel can be put into HCDA-like mean energy states of up to 3000 J/g , which is sufficient to effect prompt disassembly of a fuel pin and flash a significant portion of the superheated liquid fuel to vapor. The efficiency of the energy deposition, which was initially quite low, has been gradually improved by optimizing both the electrical circuitry and the physical configuration of the specimen until most of the power is expended in the vaporization process. The range of time intervals for disassembly can be adjusted to match HCDA theory within about 1 msec .

The aerosol experiments with this method of generation were conducted in a small vessel of about 0.5-m^3 capacity. The installation is shown in Fig. 3.

Some earlier experiments performed at the Arnold Engineering Center's Von Karman Facility were also limited to about the same scale vessel. An example of the rate-of-change data for the CDV-CRI-III is given in Fig. 4.

Continuous U_3O_8 Vaporization by the Pyrophoric Oxidation of Molten Uranium Metal with Oxygen

The initial aerosol generation method used in the Containment Research Installation Vessel (CRI-II) was the rapid oxidation of molten metallic uranium by sparging the melt with gaseous oxygen. The generated aerosol was mainly U_3O_8 , and the yield was limited by the high-temperature attack of the melt on the quartz furnace (Fig. 5). The maximum concentration of U_3O_8 aerosol obtained was little more than 1 g/m^3 , and the method was not considered practical to scale up. Comparative results of these experiments are summarized in Table I.

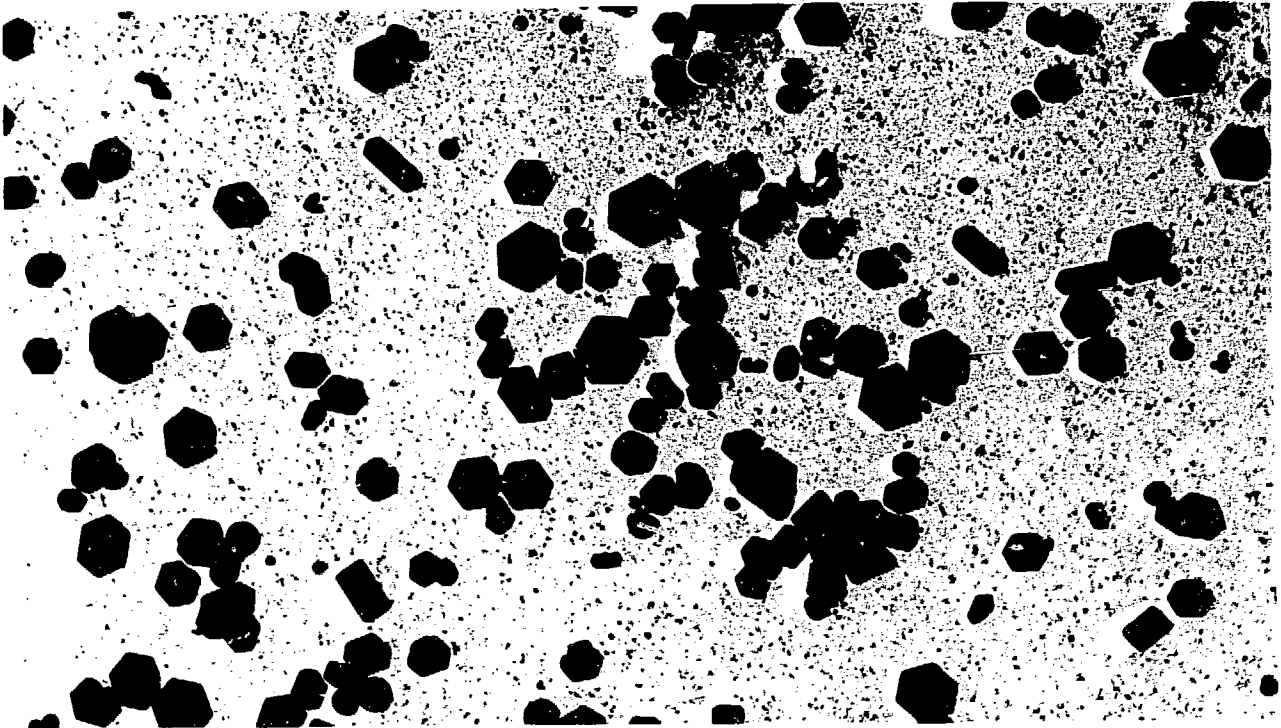


Fig. 2a. Replica of crystalline U_3O_8 particles vaporized from UO_2 melted in air. 40,000X.

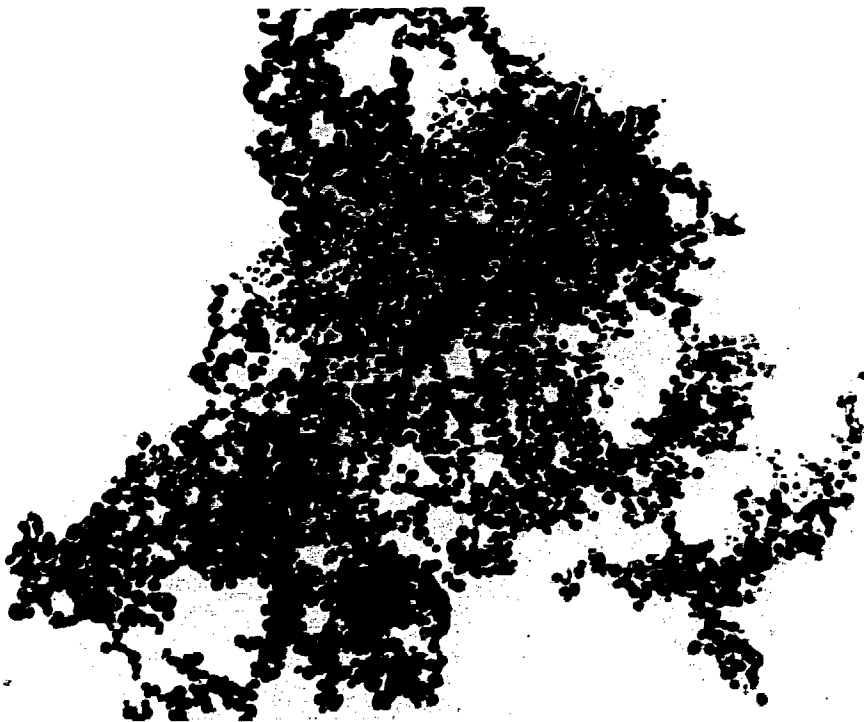


Fig. 2b. Replica of spherical particles of uranium oxide vaporized on melting UO_2 in helium. 69,200X.

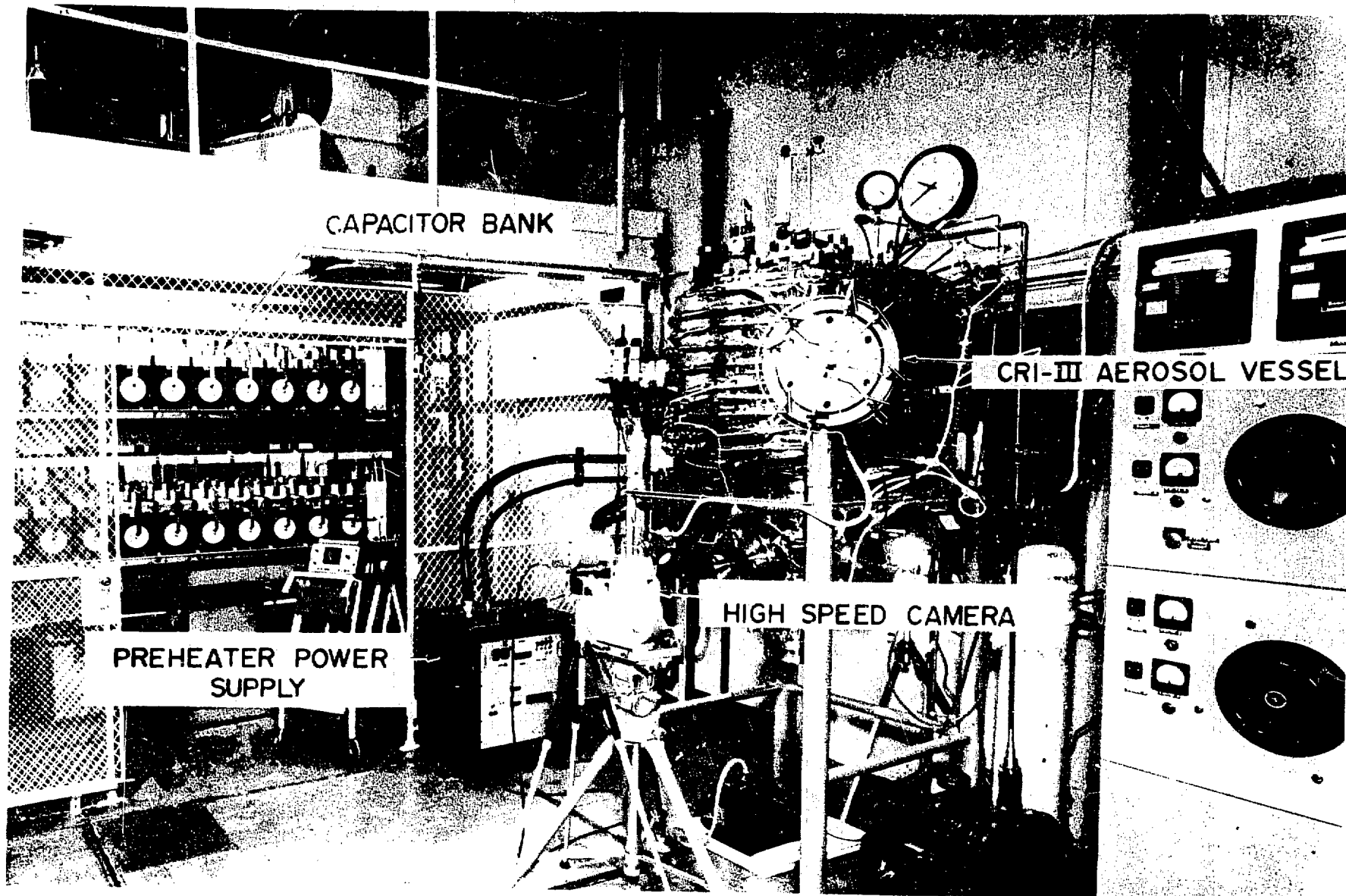


Fig. 3. Overall view of the operational area where ORNL capacitor discharge vaporization experiments are conducted.

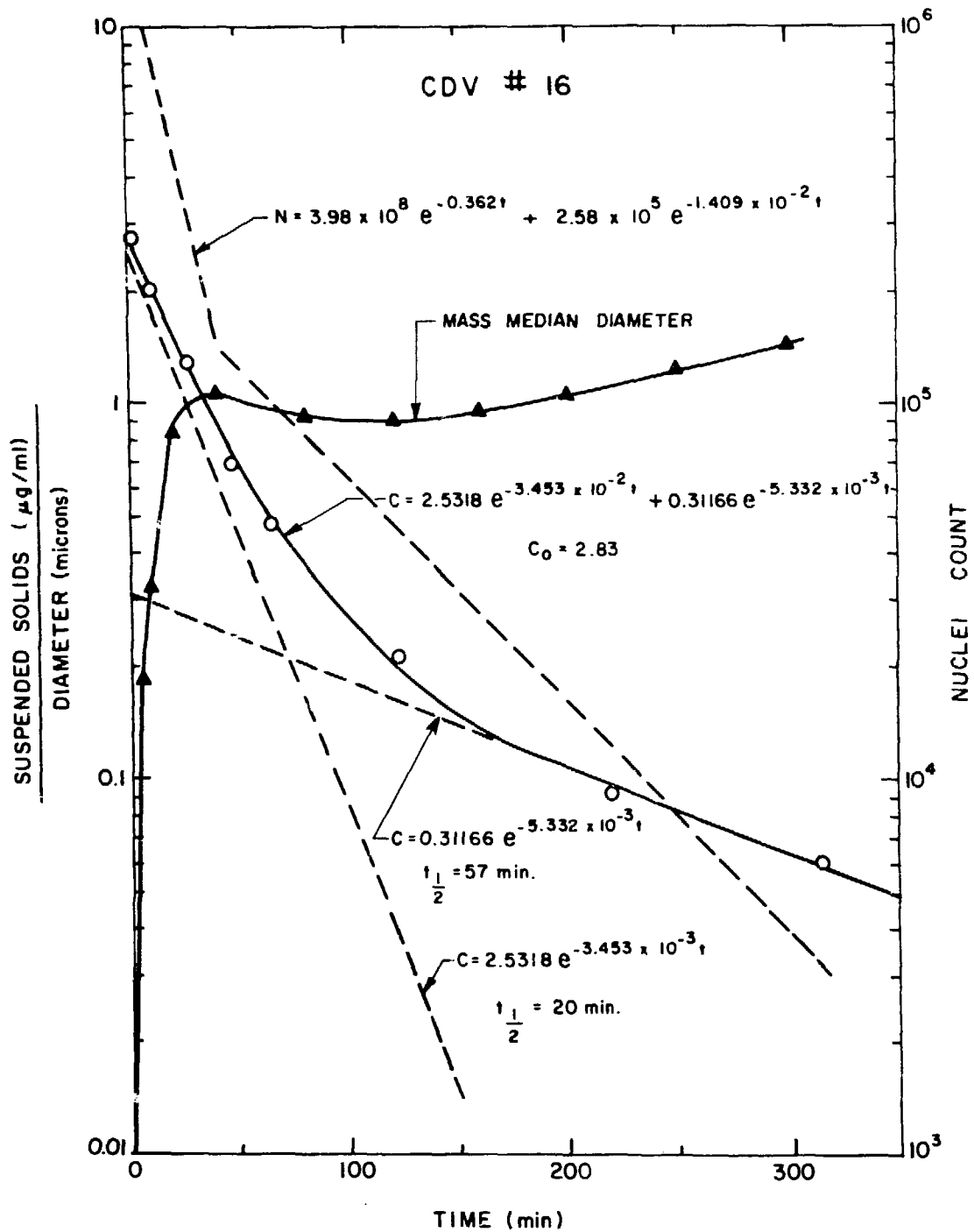


Fig. 4. Histograms for CDV Run 16 showing rate of change in concentration and nuclei count and calculated mass median diameters of agglomerates.

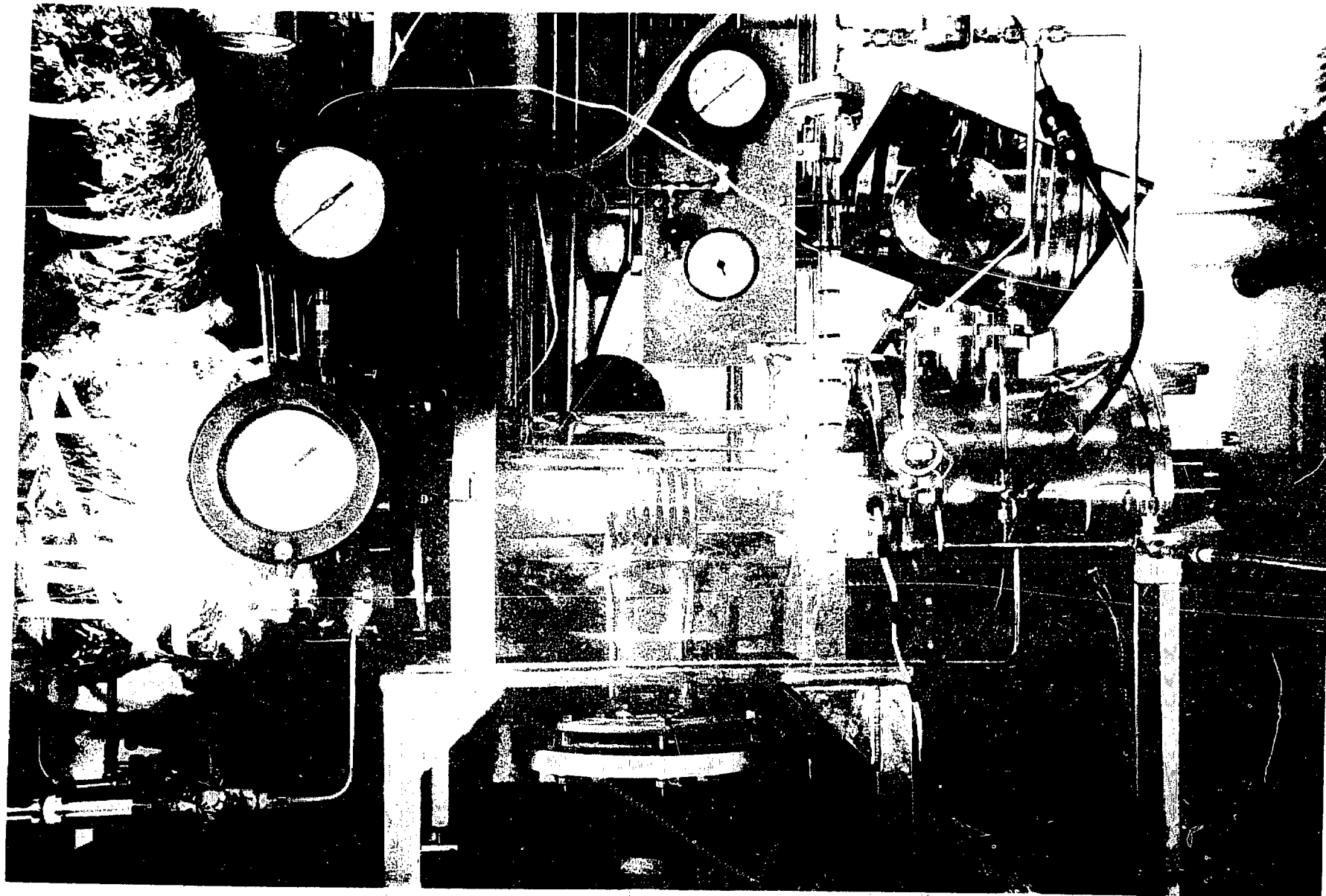


Fig. 5. Furnace for induction heating of metallic uranium.

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The dc Electric Arc Cold-Hearth Vaporization of UO_2

The second method of aerosol generation which was employed in CRI-II was a dc cold-hearth furnace (Figs. 6 and 7) which produces a predominantly UO_2 form of oxide. The actual composition of the aerosol varies somewhat according to the quality of the argon fill gas in the CRI-II vessel. It also varies with the frequency at which new uranium oxide pellets are added to the furnace since the initial melting process releases some surplus oxygen. This method of vaporization could probably be adapted to produce either UO_2 or U_3O_8 by controlling the oxygen content of the carrier gas or by introducing oxygen in the transfer pipe before the vaporized UO_2 cools below the ignition temperature of UO_2 , that is, about 300 to 500°C.

Maximum yields obtained thus far have been about 10 g/m³; however, minor improvements, including increasing the flushing rate of the beehive chamber or inserting a tungsten liner over the copper hearth, should result in a considerable increase in yield. This aerosol source is distinctly not "instantaneous," but can be used as a "continuous" source and made to reach steady-state conditions at relatively high concentrations. A typical example of the deposition behavior and rate-of-concentration change is shown in Fig. 8.

The CRI-II is a 4.52-m³ stainless steel vessel with cylindrical walls, a dished flanged head, and a conical bottom. It was designed, constructed, and operated in the 1960s for use in safety studies of the LWR type in support of the NRTS LOFT program and was reactivated in 1974 in support of the LMFBR-RSR program by NRC. It is housed in a semienclosed, specially shielded, and ventilated area in the ORNL radiochemical hot laboratory. It is presently equipped with two unique fuel aerosol generators designed for attaining high concentrations of UO_2 , U_3O_8 , or Na_2O_x mixed aerosols. One of the generators is the 100-kW dc electric arc cold-hearth furnace (mentioned above) in which molten UO_2 is literally boiled off as vapor. It is capable of vaporizing about 50 g of UO_2 per minute. The other generator (not yet operational) employs a 40-kW dc plasma arc to ignite a mixture of powdered uranium metal and oxygen and is expected to produce even higher concentrations of vapor-condensation oxides.

III. Comparative Sizes of UO_2 and U_3O_8 Primary Particles and Agglomerates

A brief comparison has been made of the primary and agglomerated particle sizes from each of the four separate fuel aerosol behavior projects conducted over the past two years in the Breeder Reactor Transient Release Program. Approximately 41 separate TEM photomicrographs were subjected to individual particle-size determinations for the primary particle evaluation. The average geometric mean primary particle diameter was found to be 0.034 μ for both the uranium-metal pyrophoric oxidation process and the UO_2 arc-furnace process (see Table I for typical examples of each). The particle size of the Arnold Engineering Development Center (AEDC) capacitor-vaporized aerosol averaged 0.025 μ , while that of the ORNL-CDV aerosol averaged only 0.014 μ . These differences are understandable since the first two processes subject the primary particles to a "hot zone" growth

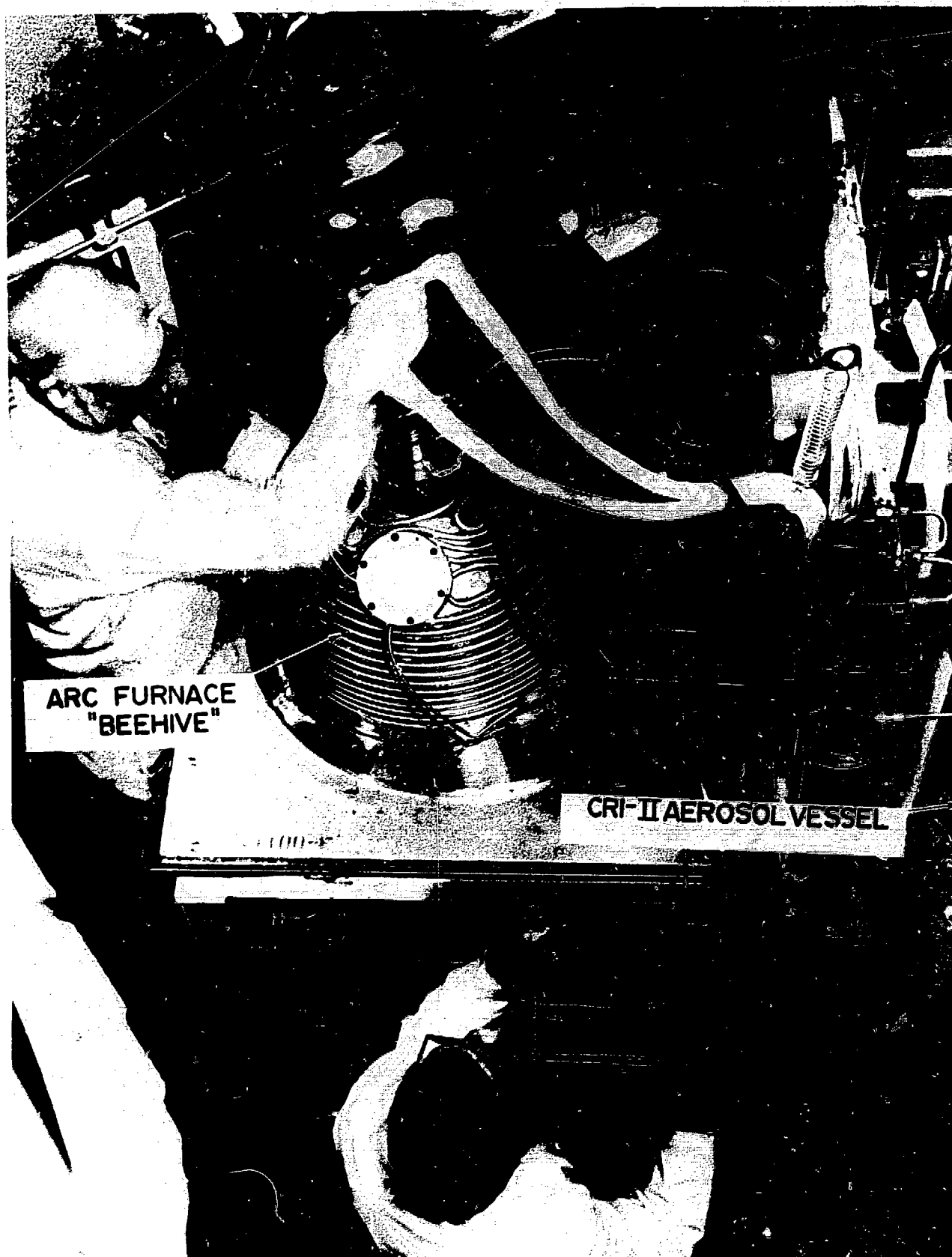


Fig. 6. Electric arc furnace in operation

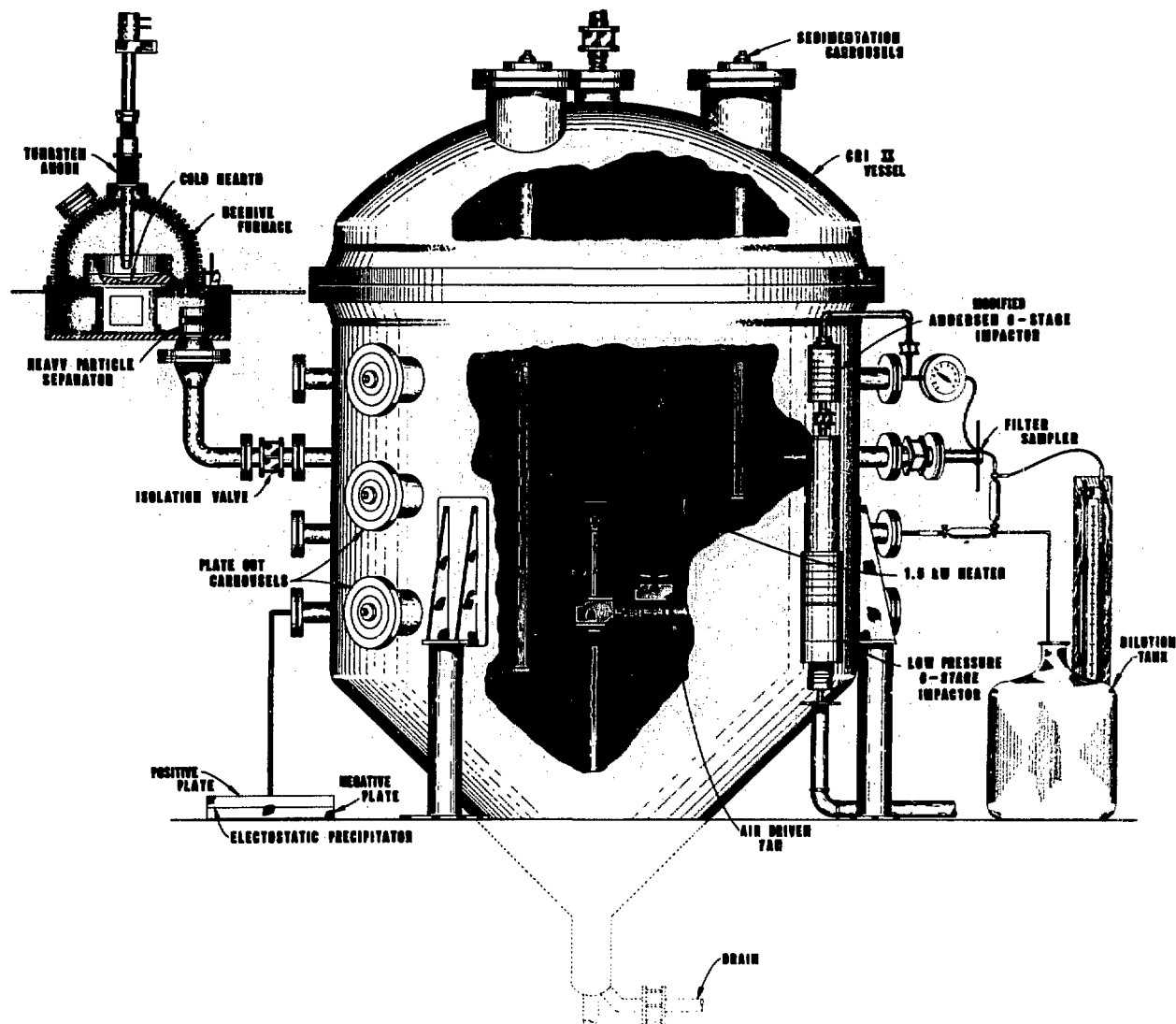


Fig. 7. CRI-II vessel, aerosol sampling equipment and electric Arc Hearth Furnace.

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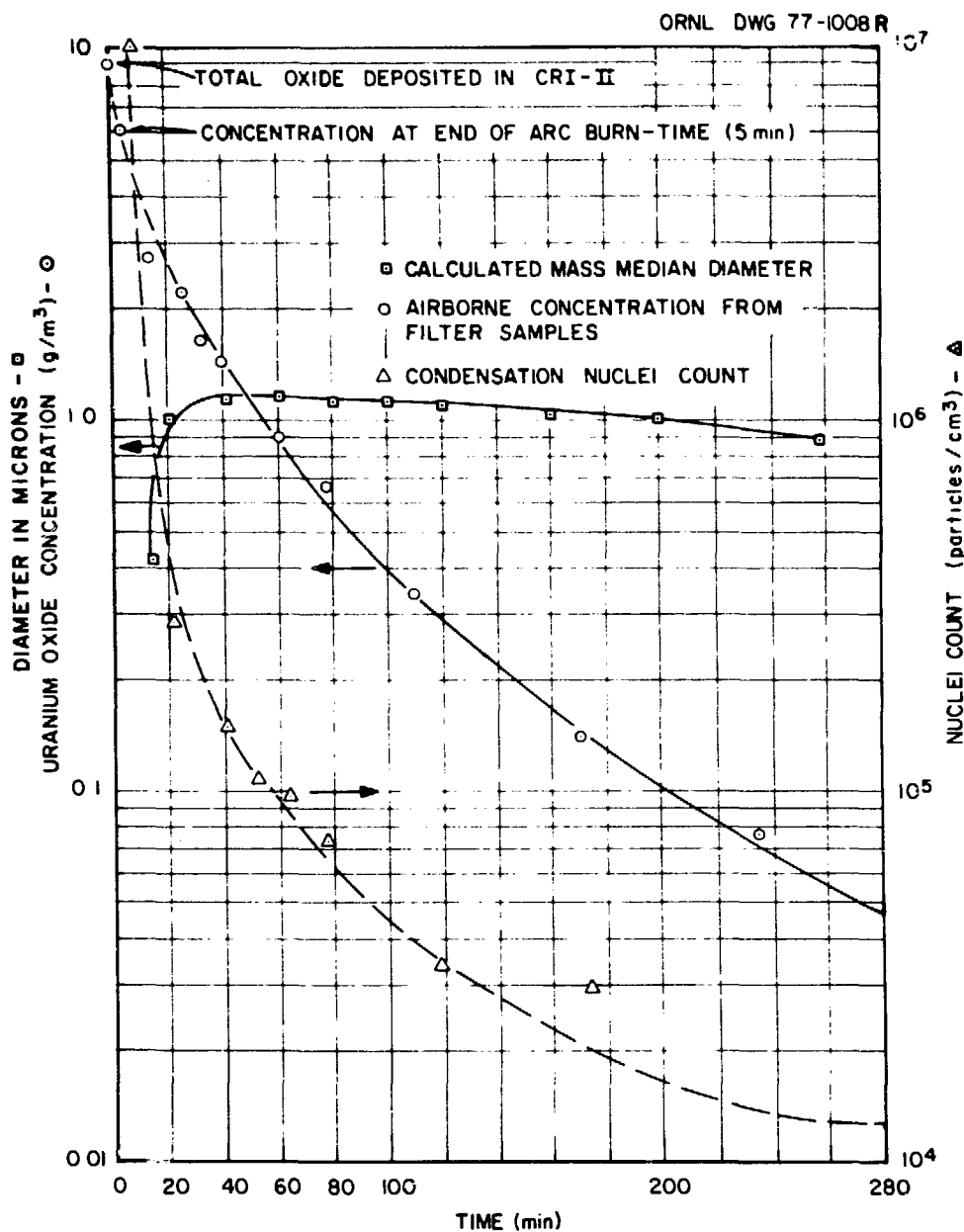


Fig. 8. Histogram of rate of change in airborne UO_2 concentration and condensation nuclei count for Run AF-6.

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process, which is revealed in their high degree of crystallinity (Fig. 9a). The rapidly quenched vapor from the superheated liquid in the capacitor discharge processes should yield a smaller, unenhanced primary particle size (Fig. 9b). The minor difference between the two may be explained by the different voltages and time constants of the electrical pulse.

The differences observed in the primary size distribution are due to the inherent rate of cooling of the vapor. The capacitor-discharge type of aerosol is generated instantaneously compared with the aerosol generated either by the arc furnace or by pyrophoric oxidation. Both of the latter methods have cooling rates several orders of magnitude slower, permitting crystal growth by vapor condensation.

Efforts to determine differences in the behavior of the branched-chain agglomerates of UO_2 and U_3O_8 aerosols have not revealed a clear contrast, although the theoretical densities of these two oxides are 10.9 and 8.3 g/cm³, respectively. Figures 10a and 10b are typical of the arc-furnace and CDV types of UO_2 agglomerates.

IV. Stokes Diameters from Concentration Changes and Impactor Analysis

An additional effort was made to compare the Stokes diameters as calculated from concentration changes with the mass mean diameters (MMDs) determined experimentally by the multistage cascade impactors. These comparisons, with few exceptions, indicate that the calculated maximum Stokes diameters are somewhat greater ($\sim 1.5 \mu$) than the impactor data ($\sim 1.0 \mu$) for most of the arc-furnace (AF) runs. These results are shown in Table II and Fig. 11.

Table II. Comparison of impactor results with
calculated Stokes diameters.

	Andersen impactor data			Calculated	
Run No.	Time of Sample (min)	Mass Median Diameter (μ)	Standard Deviation (σ)	Stokes Diameter (μ)	Maximum Concentration (g/m ³)
		CRI U-pyrophoric oxidation			
10	16-18	0.72	2.04	1.15	1.97
		CRI-II arc-furnace vaporization			
6	27.5-29.4	1.1	2.4	1.62	6.0
		Arnold Engineering Development Center Capacitor discharge			
35S	a	a	a	0.63	0.88
		ORNL-CRI-III capacitor discharge			
7	71	1.46	2.2	1.54	2.04

^aNo impactor samples taken.



Fig. 9a. Photomicrograph of arc furnace UO_2 aerosol agglomerates at 5000X.

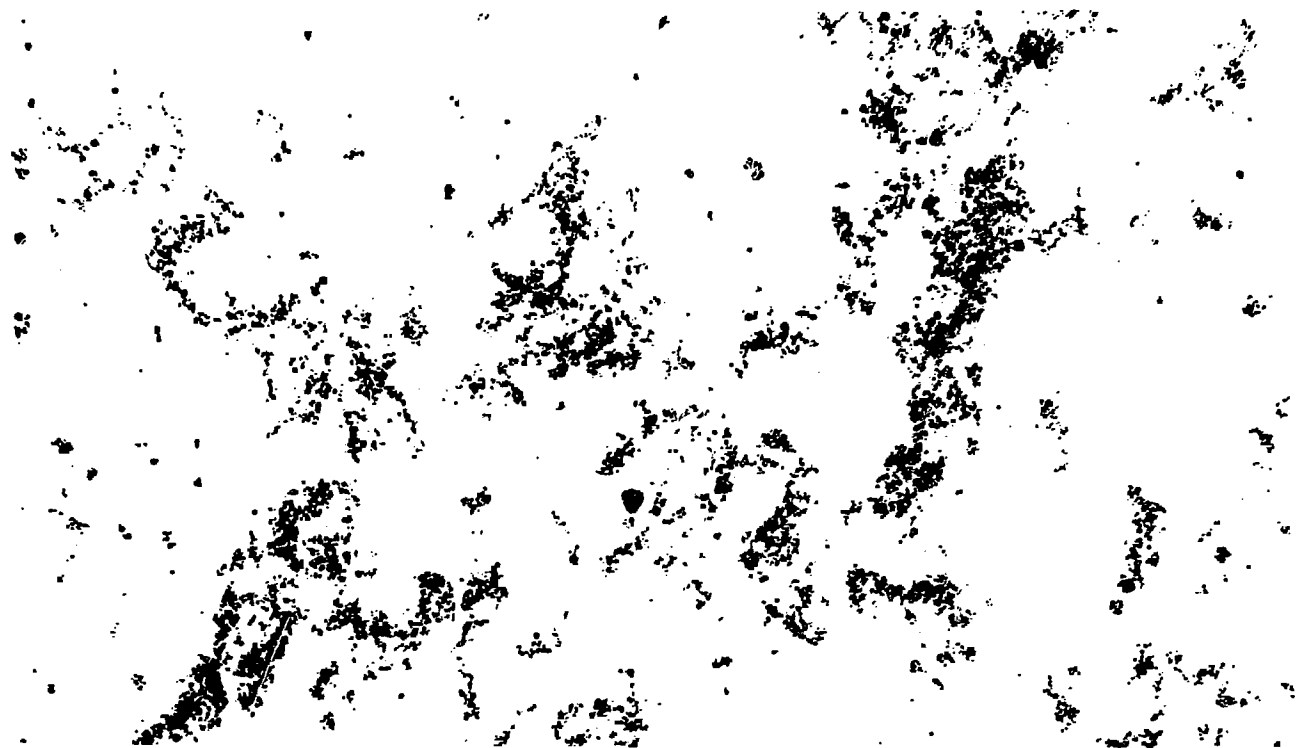


Fig. 9b. Photomicrograph of CDV UO_2 aerosol agglomerates at 5000X showing primary particles slightly smaller than AF particles.

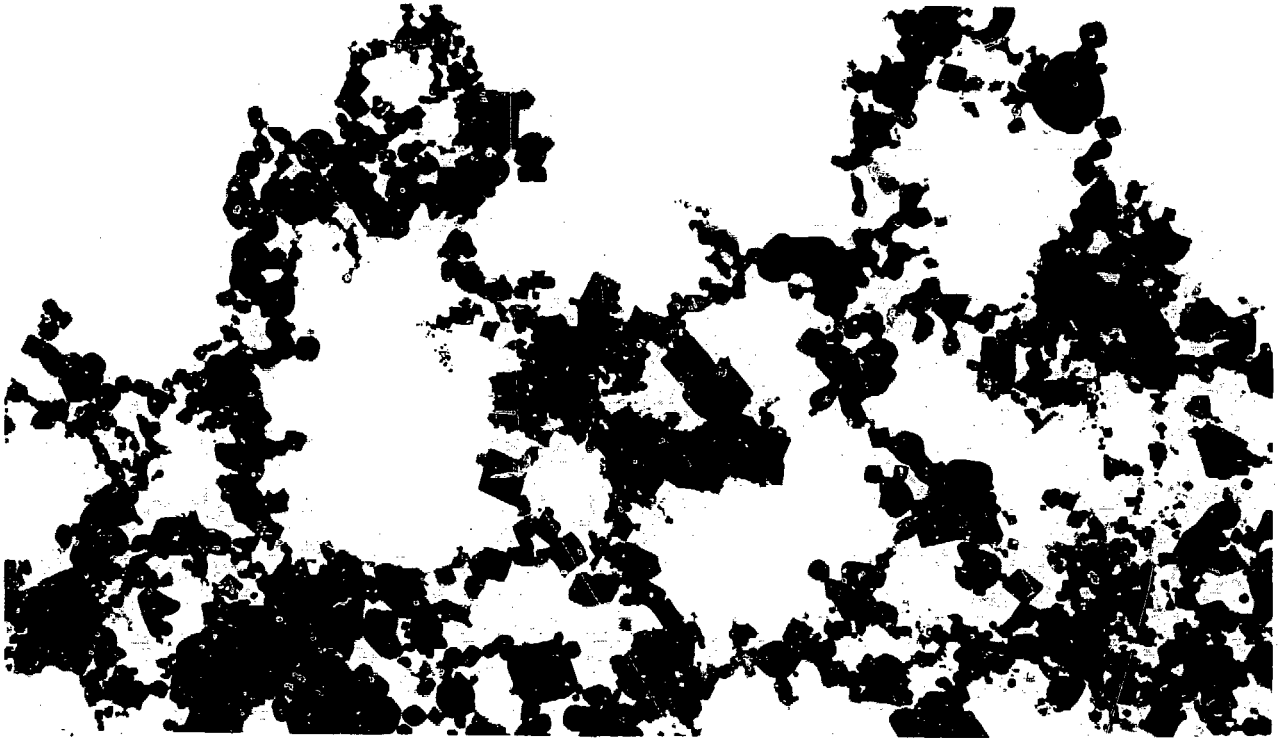


Fig. 10a. Photomicrograph of arc furnace UO_2 aerosol showing high degree of crystallinity of primary particles.

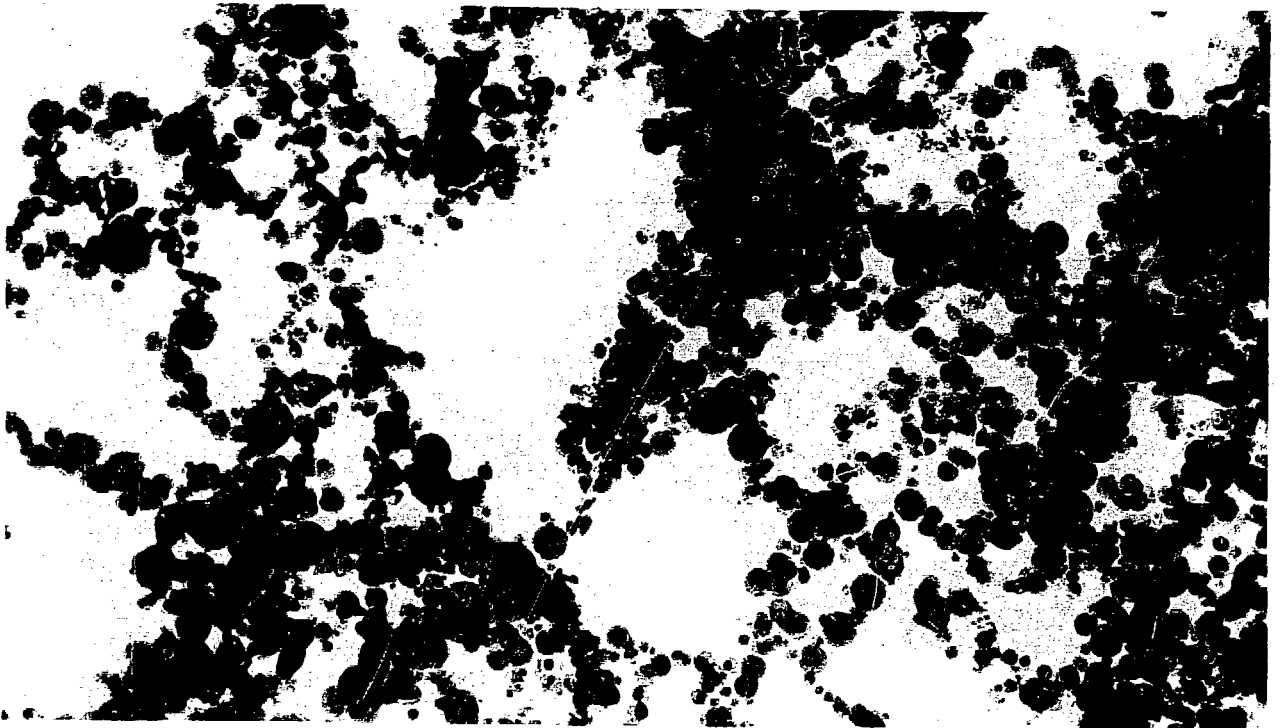


Fig. 10b. Photomicrograph of CDV UO_2 aerosol showing spherical primary particles.

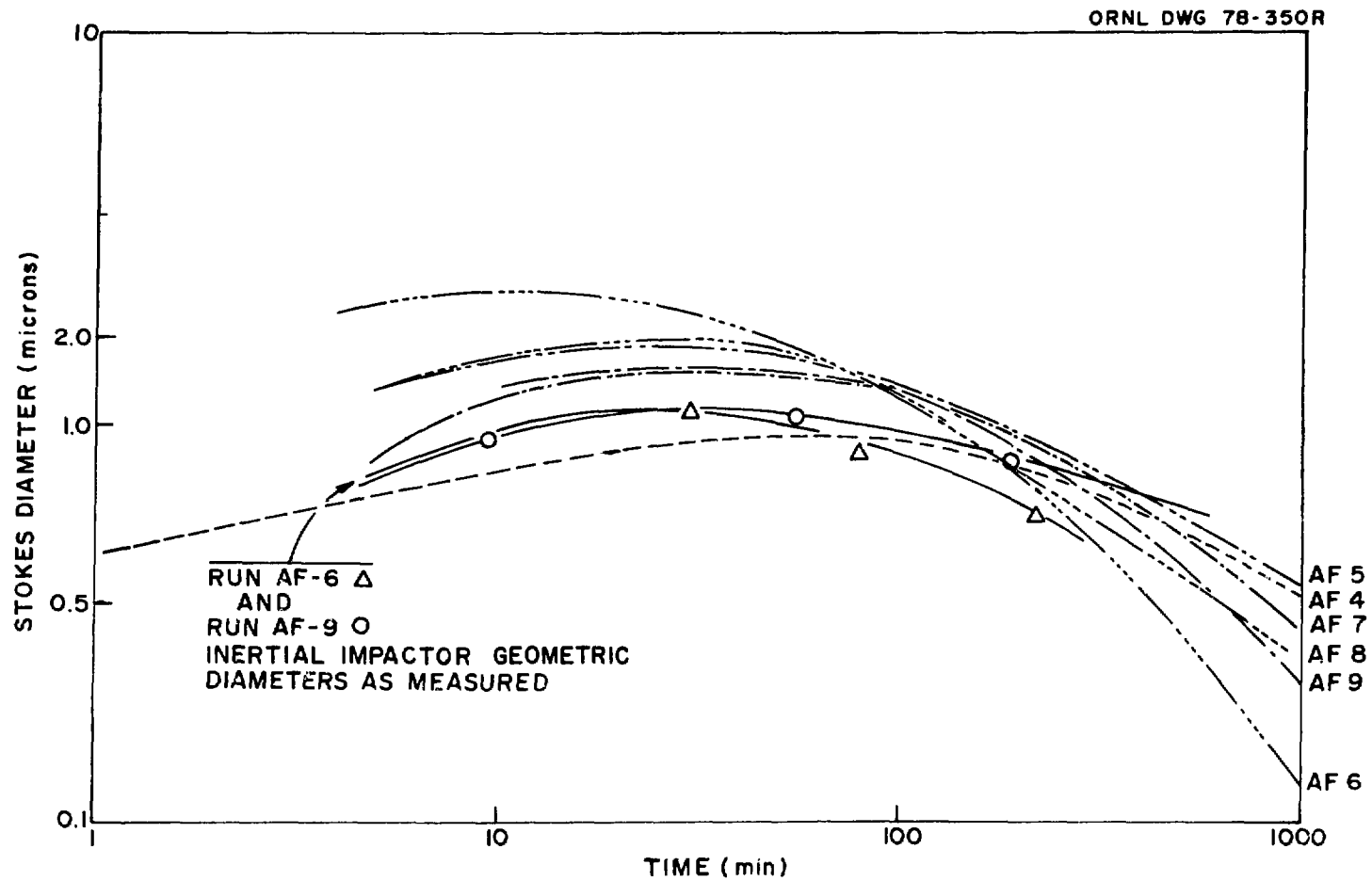


Fig. 11. Stokes diameters computed from airborne UO_2 measurements using plateout corrections derived from deposition sampler data.

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The deposition and plateout samplers expose sample plates to the CRI-II atmosphere for various periods. From these results, the amount of material deposited on the walls and floor of the tank can be estimated by dividing the average amount of plateout or deposition by the total material deposited and by normalizing with respect to the appropriate surface area ratio. Results of these calculations for Run AF-3 are shown in Tables III and IV.

Table III. Results of calculations for Run AF-3.

Exposure Time (min)		Deposition (μg)			Plateout (μg)			Settling Fraction, floor	Plateout Fraction, wall
From	To	Long ^a	Short ^a	Av	High ^a	Low ^a	Av		
0	5	240	820	530	220	200	210	0.419	0.581
5	25	620	470	545	26	19	22.5	0.87	0.126
25	58	400	600	500	18	21	19.5	0.88	0.12
58	92	400	510	455	20	7	13.5	0.906	0.094
92	161	530	320	425	27	5	16	0.883	0.117
161	265	320	290	305	13	27	20	0.813	0.187
265	613	270	130	300	22	5	13.5	0.809	0.191
613	1453	280	190	235	23	27	25	0.728	0.272

^aLong, short, high, and low designate sampler locations at 196, 94, 81, and 157 cm from the tank top, respectively.

The Stokes diameters of particles satisfy the following conditions when airborne concentrations are not adjusted to account for plateout:

$$D_s = \left(\frac{18V\eta}{\rho g} \right)^{1/2} \quad (1)$$

$$C_2/C_1 = e^{-Vt/h}, \text{ or } V = \frac{-h}{t} \ln(C_2/C_1), \quad (2)$$

where

D_s = Stokes diameter (cm),

V = particle velocity (cm/sec),

η = viscosity of air = 1.87×10^{-4} ρ ,

ρ = particle density (g/cm³),

g = gravitational constant (980 cm/sec²),

C_2 = concentration at time t_2 ,

C_1 = concentration at time t_1 ,

t = exposure time in sec ($t_2 - t_1$),

h = tank height (200 cm).

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Combining Eqs. (1) and (2) gives

$$D_s = \sqrt{\frac{-6.3 \times 10^{-5} \ln (C_1/C_2)}{t}}$$

Using the start of an exposure time for t_1 and C_1 and the end of the exposure for t_2 and C_2 , Stokes diameters were computed for Run AF-3 where airborne concentrations have been adjusted for plateout. The results of these calculations are given in Table IV.

Table IV. Stokes diameters for Run AF-3 with corresponding airborne concentrations adjusted for plateout effect.

Exposure time (min)		C_1 ($\mu\text{g}/\text{cm}^3$)	C_2^a ($\mu\text{g}/\text{cm}^3$)	Stokes Diameter (μ)
t_1	t_2			
0	5	1.539	1.498	0.75
5	25	1.44	1.15	1.09
25	58	1.11	0.77	1.07
58	92	0.728	0.506	1.06
92	161	0.483	0.258	0.98
161	265	0.229	0.123	0.80
265	613	0.0978	0.0378	0.54
613	1453	0.0236	0.0078	0.37

^aValues adjusted for plateout.

The concentration of airborne solids (no plateout correction) can be computed from

$$C_i = 1.389^{-0.864} \times 10^{-4} t_i + 0.15^{-0.1818} \times 10^{-5} t_i, \quad (3)$$

which is an empirical fit to the data (t is in seconds, $i = 1, 2$). The plateout effect is accounted for by multiplying the airborne concentration difference [$C_2 - C_1$, as obtained from Eq. (3)] over an exposure time by the settling fraction (Table II) and subtracting this product from C_1 to obtain C_2 , the apparent concentration after the time interval if gravitational settling were solely responsible for the decrease.

Figure 12 is a graph of diameter vs time for Runs AF-1 and AF-3.

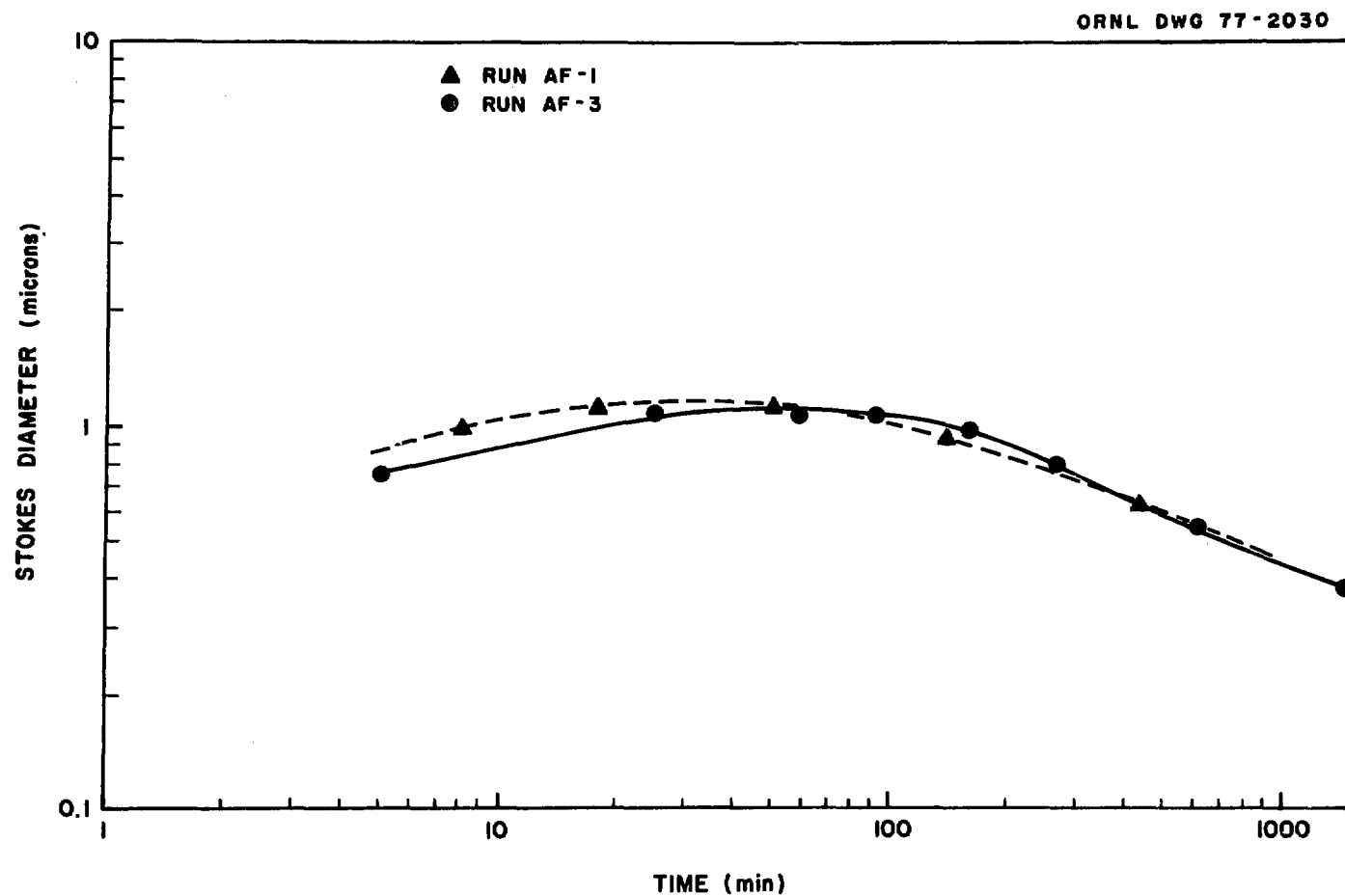


Fig. 12. Stokes diameters for UO_2 agglomerates as calculated from airborne concentration changes due to settling.

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The relative importance of sedimentation, or fallout, compared with plateout is illustrated in Fig. 13 for Run AF-3. The plateout fraction is a nearly instantaneous process accompanying the initial agglomeration step and is probably dominated by Brownian motion of the small primary particles. This is particularly true when the aerosol is settled under room-temperature conditions, as was the case in these two runs. In Run AF-3, the maximum concentration was about 1.4 g/m^3 . The plateout fraction was about 30% of the total, even though Fig. 13 shows that the mass deposited per unit area on the floor was about an order of magnitude higher than that on the walls (because the total wall area is about three times that of the floor).

As a part of the effort to characterize the UO_2 agglomerated chains and clusters and the changes with respect to time, we have installed microscope grids on the fallout carousel between the fallout plates. The results of collecting such particles for Run 8 are illustrated for four time periods in Fig. 14. The photographs appear to confirm a self-preserving size concept during these periods, as does the Stokes calculations.

V. Correlation of Experimental Results with Aerosol Agglomeration Codes

The reference code most widely used in the LMFBR-RSR programs is a Battelle modification of the AI's HAA-3, which is called HAARM-2.⁽⁴⁾ HAARM-2, now being supplemented by HAARM-3, has been used with some success in predicting the relative behavior of aerosols when only one or two parameters are varied.

An example of the use of HAARM-2 is illustrated in Fig. 15, in which the sensitivity of the particle diameter (agglomerate size) is varied in order to find the fraction of material expected to plate out. The total deposition is also derived from the code and compared with the experimental observations for Runs AF-6 and AF-3. Both the match to AF-6 by the $1.0\text{-}\mu$ -diam input and the 16% plateout fraction are in good agreement with actual data.

Another example of the use of HAARM-2 in predicting fuel aerosol behavior is given in Fig. 16, where the expected range in particle diameters as a function of maximum initial concentration is plotted in terms of elapsed time, assuming an instantaneous source. The predicted maximum radius in Curve III converted to geometric mean diameter, and corrected to a theoretical density of 10.9, is 1.05μ . This value agrees closely with the 1.0μ trial diameter and plateout fraction in Fig. 15.

An example of the further correlation with both HAARM-2 and BNL Trap⁽⁵⁾ codes is shown in Fig. 17. This correlation of deposition rates also serves to illustrate the small but distinct differences between UO_2 and U_3O_8 , which are probably due to the density difference expected in the two oxides.

AF 3

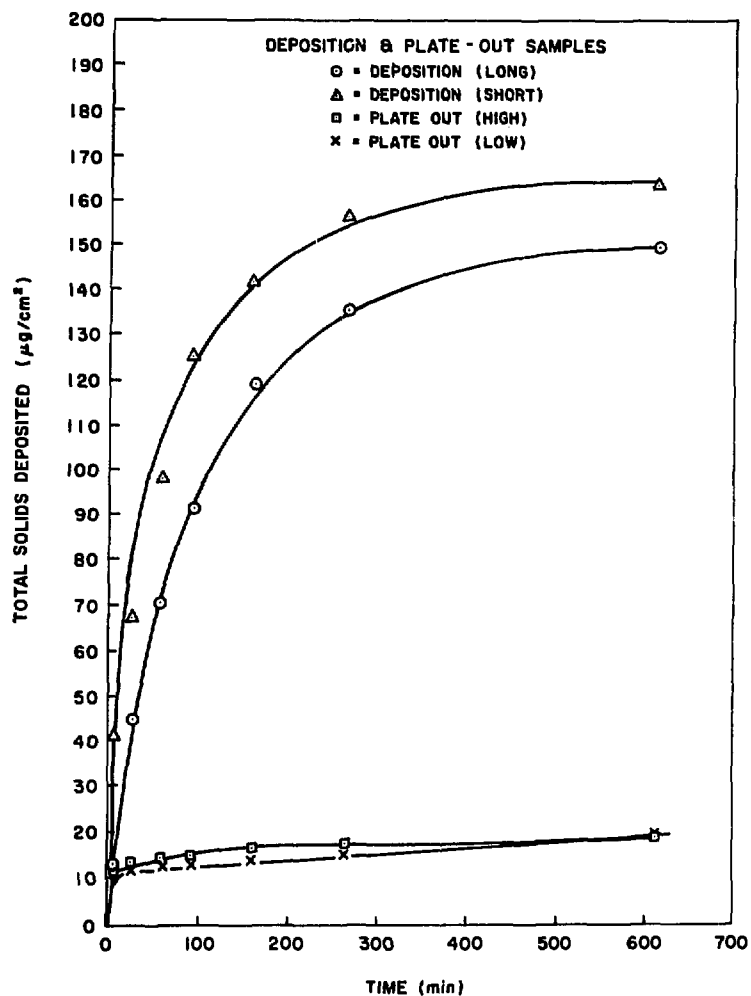


Fig. 13. Accumulated fallout and plateout fractions of UO_2 aerosol, Run AF-3.

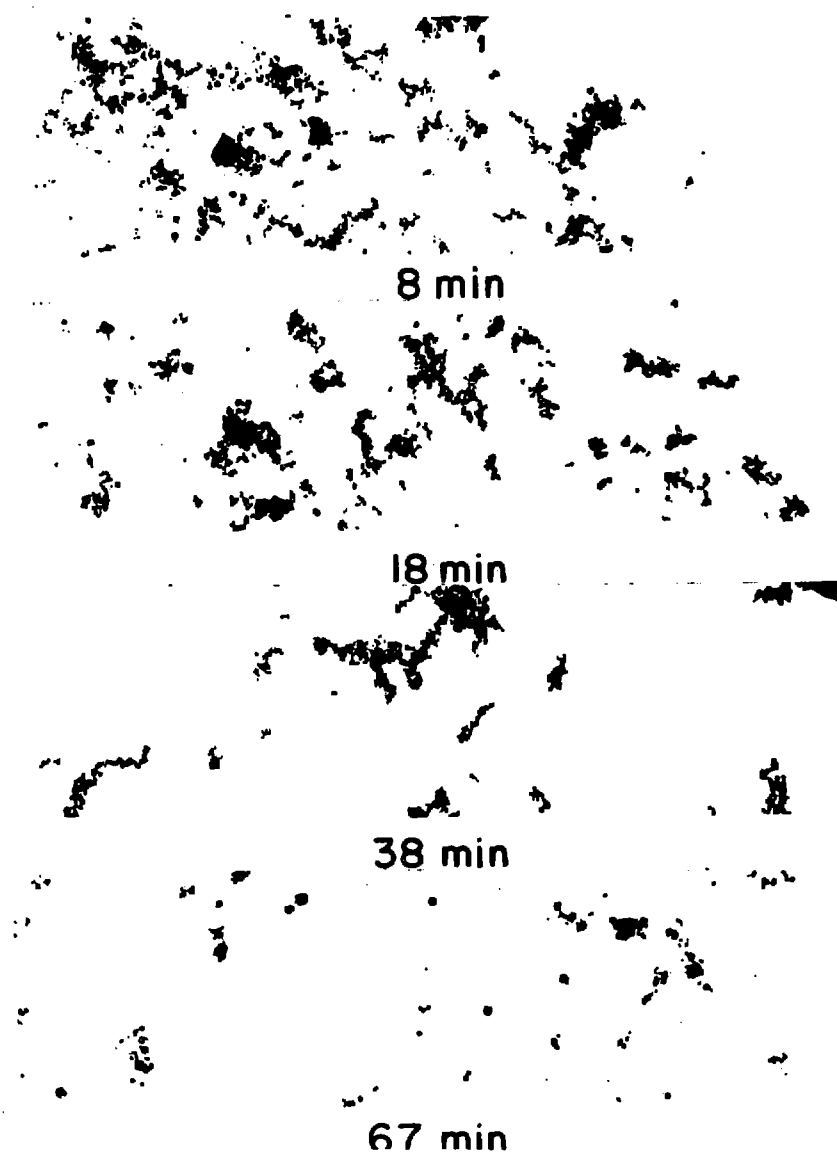


Fig. 14. UO_2 agglomerates settled on microscope grids at different time intervals (5000X).

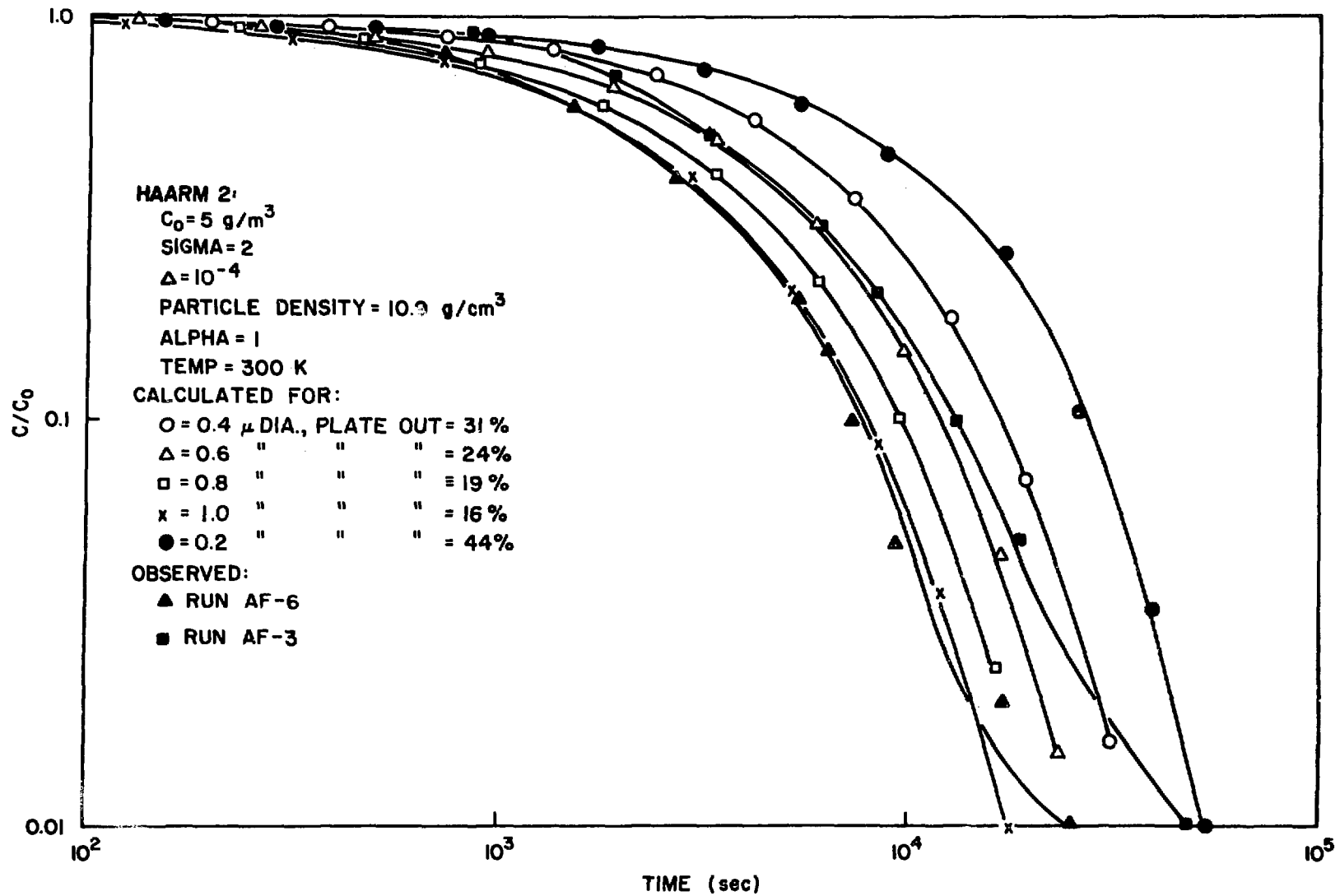


Fig. 15. Correlation of particle diameter with deposition rate as calculated by HAARM-2 and as observed in CRI-II.

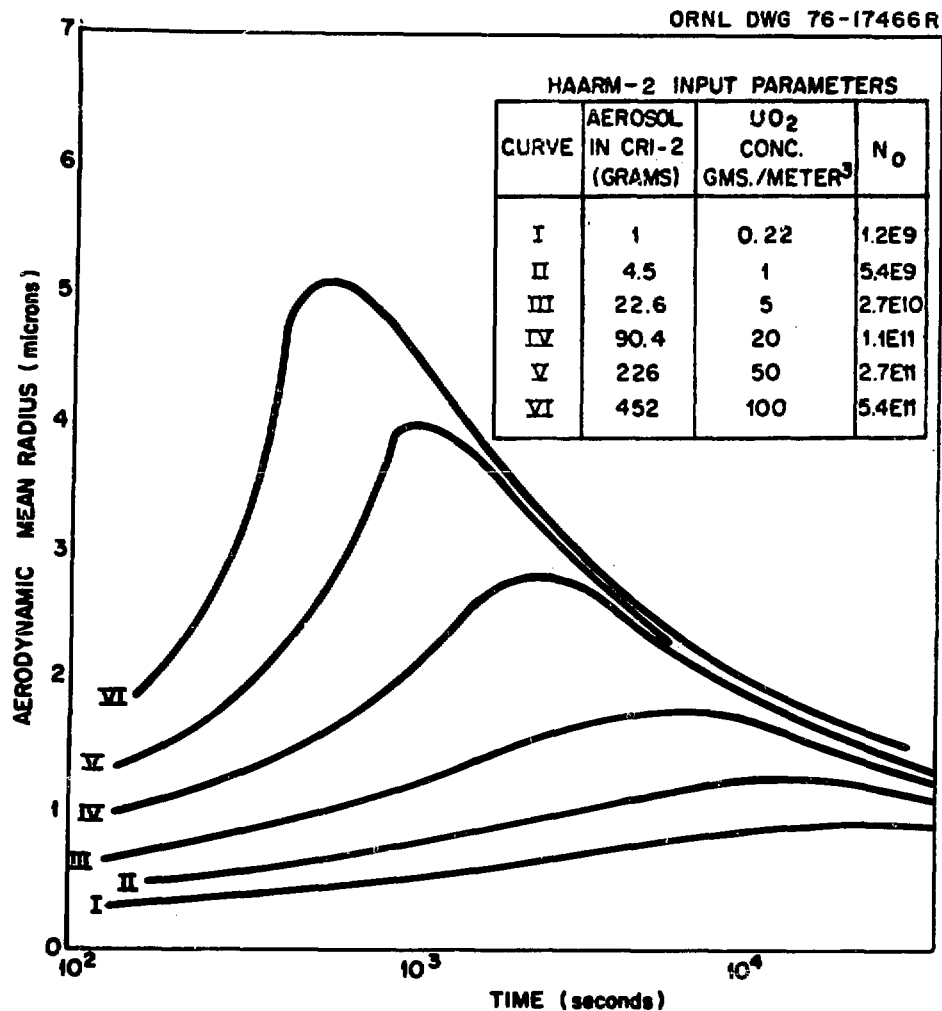


Fig. 16. Predicted time for maximum particle growth vs concentration.

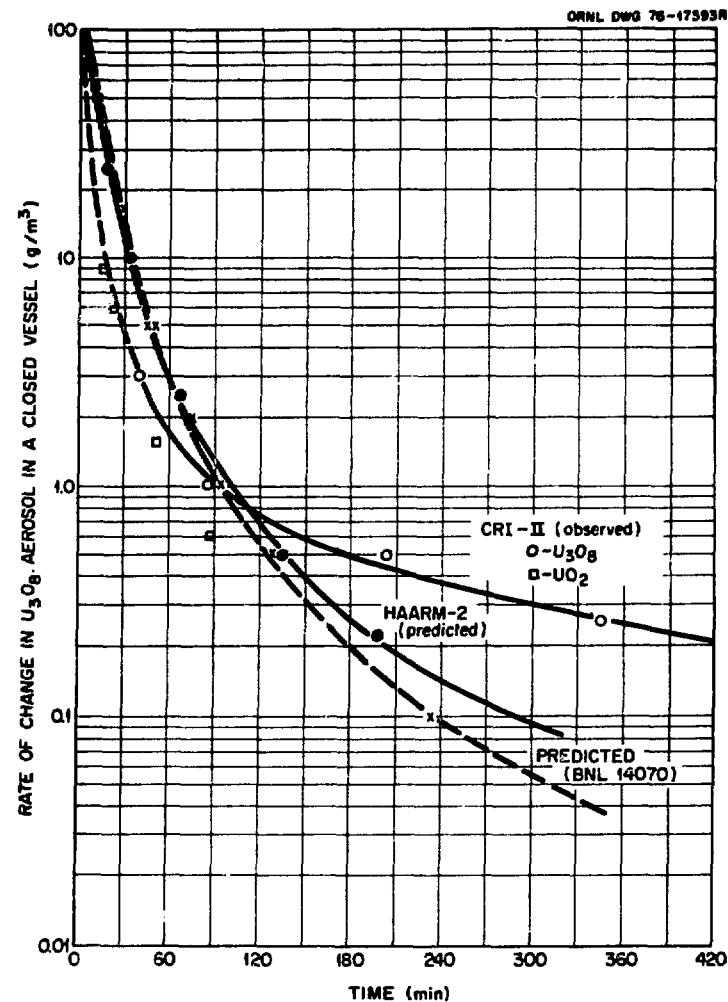


Fig. 17. Deposition rates for airborne concentration change for UO₂ and U₃O₈ in CRI-II as calculated by HAARM-2 and as observed.

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VI. BET Surface Area as a Measure of Primary Particle Size

Whenever a sufficient amount of aerosol has been available (~ 1 g), it has been used to determine the total surface area available for gas adsorption. This area is also a measure of the individual particle surface area and therefore the particle diameter.

In the arc-furnace experiments, where 50 to 100 g of aerosol are generated and deposited both inside the furnace and on the floor of the CRI-II vessel, samples are easily collected and measured for total surface area. The surface area measured between 5 and 7 m²/g for most of the tests. The smaller area is typical of the material plated out in the furnace; the larger area is typical of the settled oxide. The corresponding individual particle diameter calculated for 7 m²/g is 0.08 μ , or about twice that determined by electron microscopy. This difference may be indicative of partially connected primary particles or of the presence of a few large particles that settled on the floor after being ejected from the furnace and thus were never a part of the aerosol. A better technique for collecting the massive aerosol sample by delayed filtration should avoid this contaminant.

VII. Summary and Conclusions

The HAARM and HAA aerosol codes correlate fairly well with our experiments involving single component fuel particles at the limited level of concentrations presently being studied. The codes are useful in demonstrating sensitivity to a selected variable but require careful restriction of the other variables in order to correlate satisfactorily with experimental data. In general, the codes are relatively conservative compared to experimental observations.

The more highly concentrated breeder reactor fuel aerosols that are postulated may be expected to vary considerably in physical characteristics according to the specific hydrodynamic and thermal environment following an HCDA. Extensive variations in the primary particle sizes produced and in the stability of the chains and agglomerates may occur after these aerosols escape into the containment.

Attenuation rates (i.e., rates of change from initial concentration) in a closed vessel initially vary in proportion to the mass concentration for both fuel aerosols alone or fuel and coolant aerosols, since the higher concentrations produce equally heavier agglomerates and more rapid settling rates. Demonstration experiments at the higher levels and more realistic environments will be needed to bound the uncertainties.

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VIII. References

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DISCUSSION

TILLERY: Determination of stoke diameter from the aerodynamic diameter of branched chains, determined by the cascade impactor, requires some assumptions about the slip factor dynamic shape of the branched chains. Did you use the aerodynamic diameter given by the impactor calibration or did you use shape factor values such as those given by Stöber for fibers?

PARKER: We do a straight gravimetric determination of what is on the stage. For the calibration, we simply use the unit density calibration and the bulk density of the material. You just divide one over $\sqrt{\alpha\rho}$ or multiply by $\frac{1}{\sqrt{\alpha\rho}}$.

Not knowing any better you assume α is 1 and that leaves you with $\sqrt{10.9}$, which is 3.3. If you go back and forth from aerodynamic size or unit density size to mass median, you simply use that multiplier.

TILLERY: There is, of course, for the smaller particles a slip factor that has to be put in. Therefore, you have to make an assumption about the slip factor for the branched chains or for the long cylindrical particles if you wish to transpose to a Stokes diameter which is equivalent to a spherical particle of the same density as the particle of interest. I think that would tend to compensate somewhat for the oversize in the Stokes diameter which you get if you fail to include the slip correction factor.

PARKER: I think there are a number of things we could do. We are of the opinion that, at the concentration we are working, we just might "smear" that problem out. But, in all fairness, I would feel much better if we had the centrifuge data to go with it.

SHAW: Have you observed any break-up of aggregates in the first couple of

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stages of the cascade impactor, especially in the large particle size range?

PARKER: Two years ago, we reported at the Aerosol Technology Conference that we had two sets of measurements which were contradictory. The instruments which had sonic jets were giving more particles by almost a factor of 10. For the atmospheric impactor, we presently do not have a way of stating unequivocally that it is not breaking up aggregates. Whenever we get Marvin Tillery's centrifuge, we think we will be able to determine this.

SHAW: Do you use any coating to prevent disaggregation?

PARKER: We use three things: silicone oil coatings, thick filters, and membrane filters. When we vary the amount of material collected on a plate, we get some odd distributions but we can always tell that the unsatisfactory measurements occur because of overloading the plate.

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SODIUM OXIDE/HYDROXIDE AEROSOL PROPERTIES AND BEHAVIOR IN A LARGE VESSEL*

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Abstract

Two large-scale aerosol behavior tests were performed in the Containment Systems Test Facility (CSTF) using sodium pool fires as the aerosol source. The purpose was to characterize the properties and behavior of sodium aerosol particles formed and aged in a large containment vessel. The results will be used as baseline data for comparison with follow-on air cleaning tests. The 20-m high, 850-m³ CSTF containment vessel is approximately half scale of a commercial reactor containment building for parameters which affect agglomeration and gravitational settling. Thus, the test air cleaning systems will be challenged with an aerosol closely representative of that postulated for LMFBR accidents. The results of these tests also offer an excellent opportunity to evaluate computer models which have been developed for the purpose of predicting aerosol behavior during hypothetical accidents in sodium cooled reactor containment spaces.

The two tests differed only in that steam was injected in the second test to simulate the release of water vapor from heated concrete. The aerosol in the first test was a dry mixture of sodium peroxide, sodium hydroxide and sodium carbonate. In the second test it was predominantly wet sodium hydroxide. The addition of water vapor caused more rapid fallout during the aerosol release period, but lower settling rates afterward. The net results was a 25% lower potential for aerosol leakage in the wet test. Detailed analyses of computer model comparisons have not yet been performed, but preliminary HAA-3B code predictions were in good agreement with suspended mass concentration and the ratio of mass settled to plated on walls.

Air cleaning tests will next be performed in the same facility to demonstrate the effectiveness of various types of scrubbers, filters and novel devices for cleaning the very high mass concentration aerosol from the containment atmosphere.

I. Introduction

The aerosol likely to be formed during postulated accidents in sodium cooled reactors has been the subject of considerable study at many laboratories during the past decade⁽¹⁻¹⁰⁾. These studies have encompassed theoretical treatment, computer modeling and experimental measurements. Several areas remain where further information is

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needed. The need for large-scale experiments to clarify the effects of vessel size and shape on airborne concentration has been pointed out⁽²⁾. Present experimental data are limited to vessels 9-m or less in height. Vessel height affects agglomeration time and probably the physical nature of the agglomerate particles. Many postulated accidents involve structures with concrete surfaces that release water vapor when heated⁽¹¹⁾. Water vapor adsorbed by aerosol particles may cause significant chemical and physical changes in the particles.

The CSTF was constructed and operated for the purpose of studying containment and air cleaning systems under light water reactor (LWR) accident conditions⁽¹²⁾. The capability of generating sodium aerosols has been added and an experimental program is under way to develop and demonstrate air cleaning systems under simulated sodium cooled reactor accident conditions⁽¹¹⁾. The CSTF program includes tests with aerosols generated by sodium pool fires and sodium sprays in air and inert atmospheres. In some tests steam and carbon dioxide will be added to simulate off gases from heated concrete. The present paper reports the results of the first two tests which used sodium pool fires in air atmospheres. No engineered air cleaning features were employed. Aerosol depletion was caused solely by passive processes.

II. Experimental Arrangement

Experimental Equipment

A schematic diagram of the experimental arrangement is shown in Figure 1. Table I gives the dimensions of the containment vessel, which was fabricated according to the ASME Section VIII Code (1962) with a design pressure of 0.517 MPa (75 psig) at 160°C (320°F). It is a carbon steel (SA 212-B) vessel with standard dished top and bottom heads. All interior surfaces are coated with a modified phenolic paint. The external surfaces are covered with a 25.4-mm thick layer of fiberglass insulation with a thermal conductivity of 0.0467 W/m°C at 100°C.

Sodium was heated in a 1.9-m³ 304SS vessel to the desired test temperature. At time zero a valve was opened and argon pressure forced the sodium through a 2-in. schedule 40 pipe (52.5-mm ID) into a carbon steel pan centered near the bottom of the containment vessel. The burn pan rested on insulating fire brick and the sides of the pan were insulated. The pan was equipped with a hinged, gasketed lid which permitted termination of the aerosol source on command.

Test Conditions

The two tests, AB1 and AB2, were performed with essentially the same initial conditions, but in the second test steam was injected at a rate equivalent to the release of water vapor from approximately 10-30 m² of hot concrete⁽¹³⁾. Although the aerosol and steam release rates and duration were arbitrary, they are within the range of accident conditions postulated in LMFBR safety analyses.

The initial containment atmosphere was air at normal temperature and humidity. Other test conditions are listed in Table I.

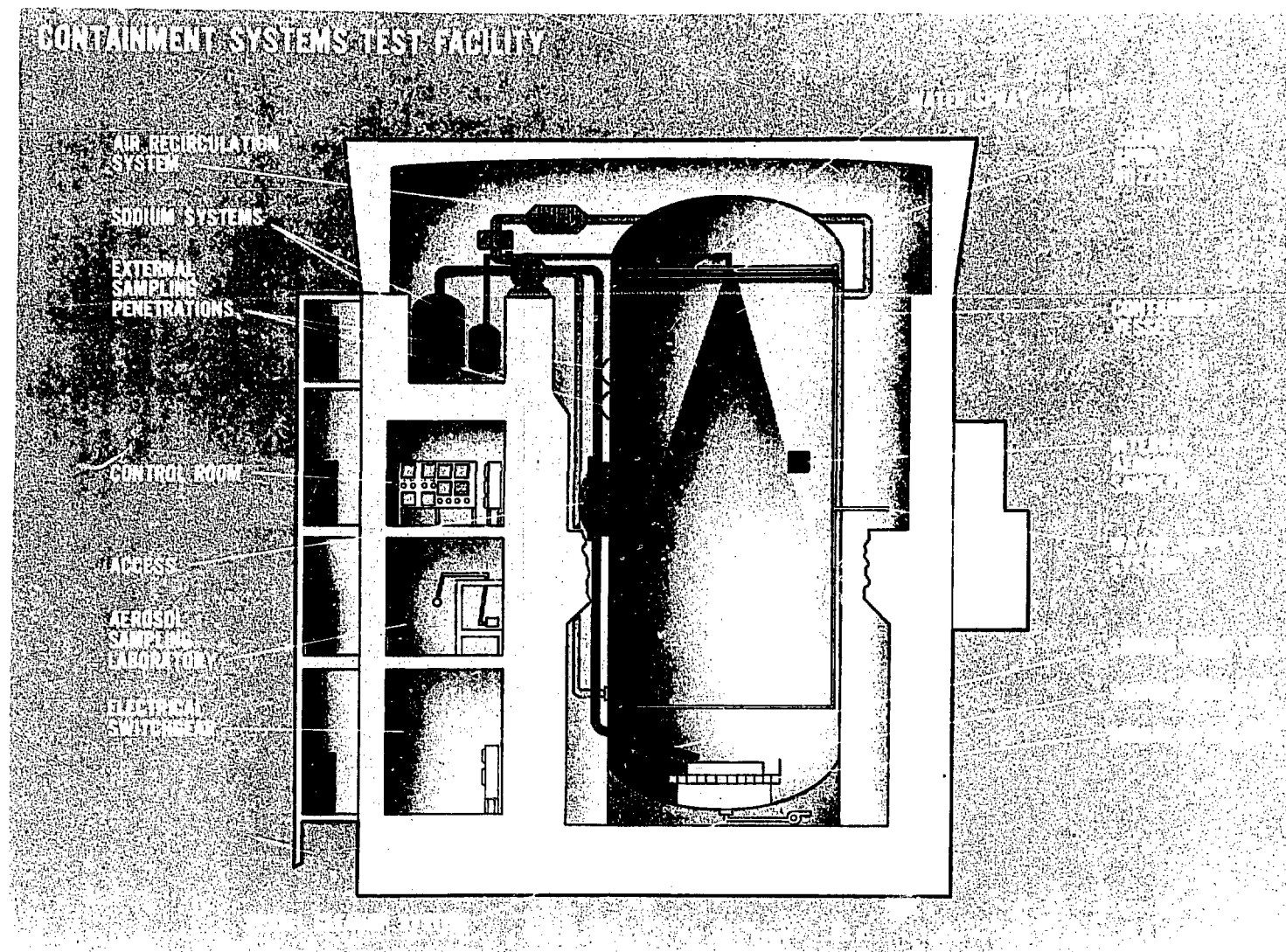


Figure 1. Schematic diagram of the Containment Systems Test Facility.

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Table I. Test conditions for CSTF tests AB1 and AB2.

	<u>AB1</u>	<u>AB2</u>
<u>Containment Vessel</u>		
Diameter, m	7.62	7.62
Overall height, m	20.3	20.3
Volume, m ³	850	850
Total horizontal surface, m ²	88	88
Shell surface, m ²	520	520
Total internal surface, m ²	1000	1000
Vessel steel mass, kg	103,000	103,000
Leakage rate, %/d at 10 psig	2.0	2.6
<u>Sodium Spill</u>		
Mass sodium spilled, kg	410	472
Sodium burn pan surface, m ²	4.4	4.4
Initial sodium temperature, °C	600	600
Sodium fire duration, sec	3600	3600
Total sodium oxidized, kg	154	146
<u>Steam Addition</u>		
Flow started, sec after time zero	-	960
Flow stopped, sec after time zero	-	4560
Flow rate, kg/sec	0	0.019
<u>Initial Containment Atmosphere</u>		
Oxygen, vol %	19.8	20.9
Dew point, °C	10.0	7.6
Temperature, °C	26.5	20.5
Pressure, MPa (absolute)	0.125	0.128
<u>Containment Atmosphere During Test</u>		
Maximum temperature, °C	91	96
Maximum pressure, MPa (absolute)	0.142	0.153
Final oxygen, vol %	14.7	16.7
Final dew point, °C	-40	-1.5
<u>Aerosol Source</u>		
Type	Na pool fire	Na pool fire
Source duration, sec	3600	3600
Average source rate (as sodium), g/s	11.1	10.8
Total aerosol release (as sodium), kg	40.0	38.8
Fraction of oxidized sodium released	0.261	0.266

Test Procedure

The procedure used in each of the tests was to install and calibrate the equipment and instrumentation, dry the vessel to a constant, normal humidity, seal the vessel so that it was essentially leak tight and heat the sodium to 600°C in the auxiliary sodium tank. At a time defined as "time zero", t_0 , a valve was opened and the sodium flowed into the burn pan through the electrically heated delivery line. Figure 2 is a photograph taken through a viewing window near the bottom of the vessel. A dense plume of aerosol particles formed immediately and swirled upward until the aerosol was dispersed throughout the containment volume. The sodium flow lasted 80 seconds and then the sodium supply system was isolated from the containment vessel.



Figure 2. View of sodium spilling into ABl burn pan ten seconds after start of spill.

Temperature measurements were made continuously by 45 thermocouples. Containment atmosphere pressure and gaseous analysis was also continuous. Aerosol characterization was performed by periodic "grab" samples taken through air lock ports at four locations on the containment vessel wall. In addition, clusters of sampling filters with remote operated solenoid valves were located at ten different locations throughout the atmosphere. Besides the suspended mass concentration samples, measurements were made of particle size distribution (by cascade impactors), aerosol chemical composition, particle mobility,

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and instantaneous settling rates (by deposition coupons). Electron microscope grids were exposed and convection current velocity was measured. Temperature, pressure and gas analysis data were recorded on stripchart recorders and on a 100-channel digital data logger.

The experimental aerosol measurements were made with decreasing frequency over a five-day period of time. Then a ventilation system was started to prepare for personnel entry and the internal samples and integral deposition trays were retrieved. Figure 3 is a view of the vessel interior at the time of first entry during AB1. Next, the burn pan with the partially oxidized sodium was removed, weighed, and its contents sampled. A patch of vertical wall surface was decontaminated and the washings analyzed for sodium content. The vessel was then washed by a series of water sprays, beginning with the bottom head, then the vertical walls, then the top head. Finally, steam was injected for several days to clear all crevices by condensation. The volume of each wash was measured and analyzed for sodium concentration to enable a sodium mass balance to be made.

The filter samples and electron microscope grids were protected from exposure to ambient atmosphere by special holders and techniques. The filter papers were weighed to obtain total mass and then analyzed for sodium content by acid titration or flame emission spectroscopy.

III. Experimental Results

Containment Response

The atmosphere within the containment vessel was reasonably well mixed at all times excluding, of course, the region near the surface of the sodium pool and in the plume where mixing occurred. This conclusion was reached by noting that the standard deviation from the mean bulk gas temperature was <5% of the mean and that the suspended mass concentration at ten locations agreed within $\pm 10\%$ during the sodium fire period and within $\pm 40\%$ at long times. The gas composition at two elevations 12 m apart also agreed closely. The oxygen concentration decreased by 5.1% in AB1, 4.2% in test AB2. The containment temperature and pressure are plotted in Figures 4 and 5 for tests AB1 and AB2, respectively.

An attempt was made to measure the convection current velocity near the wall by means of a thermopile-type anemometer inserted near the vessel mid-elevation. A maximum vertical velocity of 1.0 ± 0.3 m/s was measured in both tests at a distance of 25-50 mm from the wall. The highest velocities occurred approximately 20 minutes after the spill in test AB1 and near the end of the sodium fire period during the steam release test AB2. Difficulty was experienced in making this measurement due to deposition of aerosol on the anemometer probe. Very frequent cleaning was required.

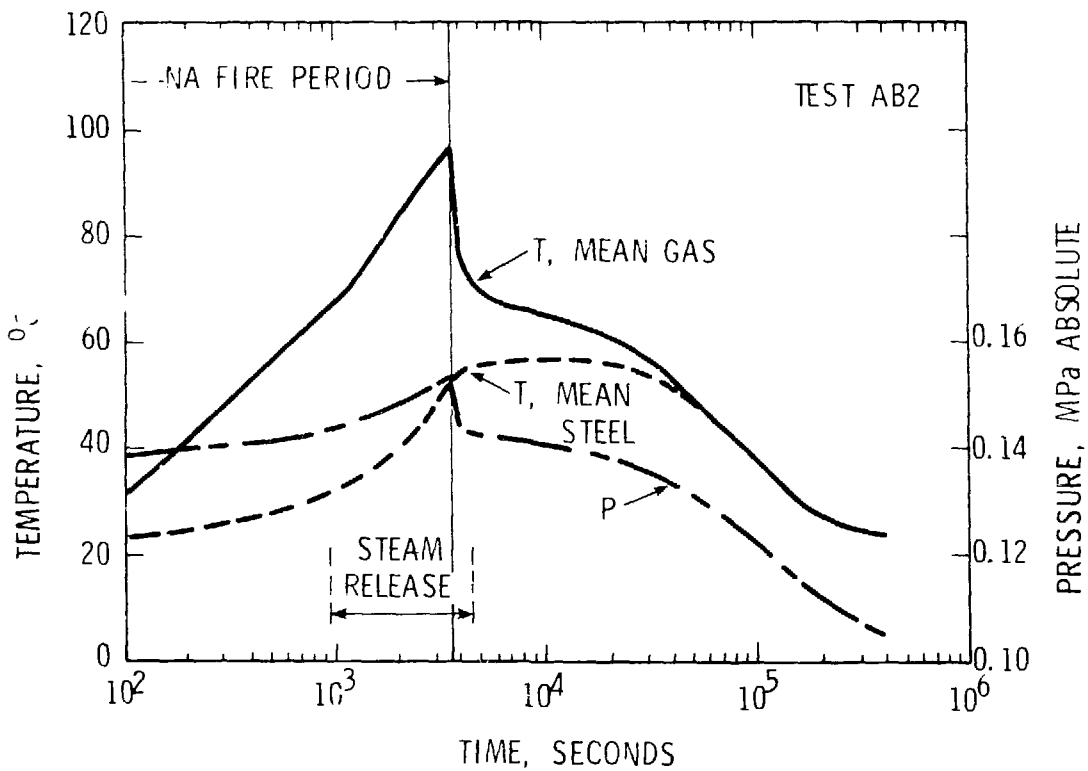
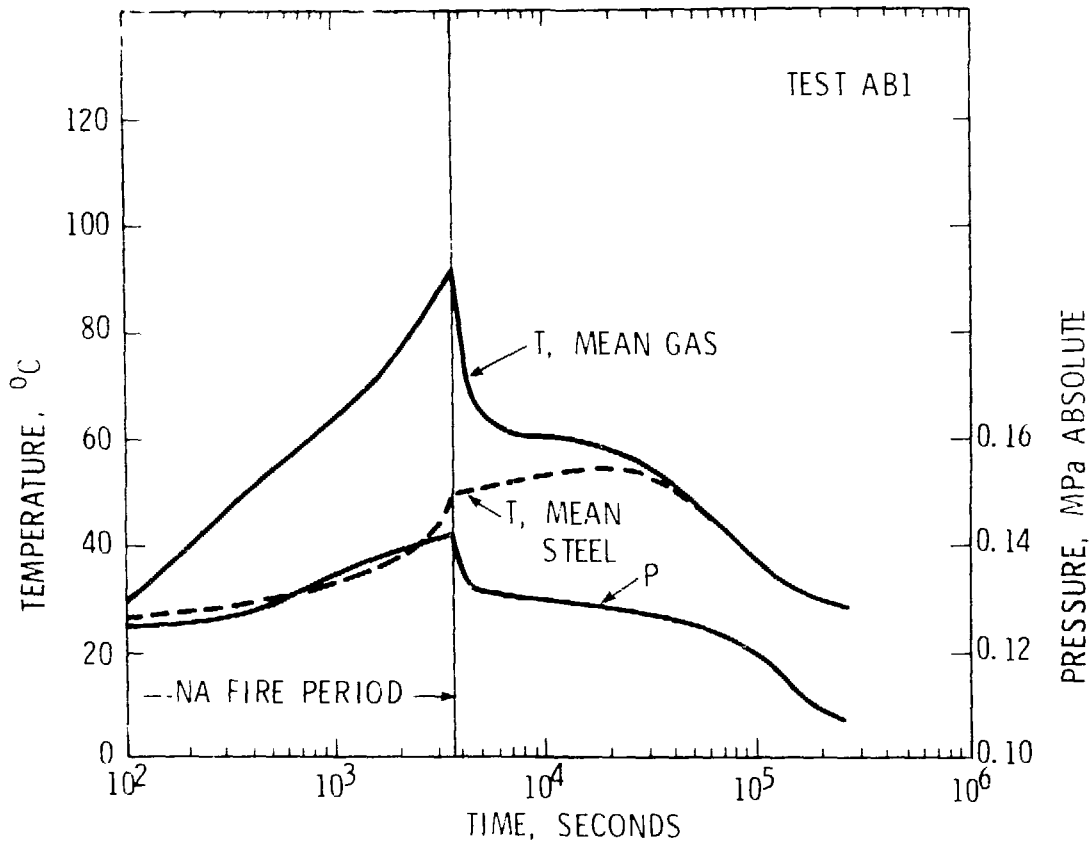
Sodium Fire and Aerosol Generation

The post-test sodium mass balance is given in Table II. Sodium recovered in the water washes accounts for the deposited aerosol mass, since it is reasonably certain that no material escaped from the burn pan except as an aerosol. The sodium oxidation rate was nearly identical for the two tests, averaging 34 kg Na/hr m^2 . Likewise, the



Figure 3. View of vessel through network of cables and wires.

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Table II. Sodium mass balance.

	kg Na	
	Test AB1	Test AB2
<u>Delivered to CV</u>	<u>409</u>	<u>472</u>
<u>Accounted for in CV</u>		
Metallic Na in burn pan	256	326
Oxidized Na in burn pan	117	108
CV water washes	39.8	38.6
Samples	0.2	0.2
Leaked (estimate)	<u>0.02</u>	<u>0.02</u>
Total accounted for ($\pm 15\%$)	413.00	473.00

fractions of oxidized sodium released as an aerosol were nearly the same, averaging 0.26. These observations show that the presence of a source of water vapor in the atmosphere did not affect the sodium oxidation kinetics or the aerosol release fraction significantly. These observations are summarized in Table III.

Table III. Sodium oxidation and aerosol release.

	Test AB1	Test AB2
Mass of sodium reacted, kg	<u>153</u>	<u>146</u>
Average oxidation rate, kg Na/hr m ²	34.8	33.2
Total aerosol mass release, kg Na	40.0	38.8
Average aerosol release rate, kg Na/hr m ²	8.95	8.68
Average aerosol mass source, kg Na/s	0.0111	0.0108
Fraction of oxidized Na released as aerosol	0.261	0.266

Sodium recovery by selective post-test washing of the vessel surfaces showed that approximately 7.3% of the aerosol deposited on vertical surfaces, while 92.7% settled on horizontal surfaces in each test. All horizontal surfaces were effective for collecting aerosol by settling, even surfaces sheltered by overhanging ledges and platforms.

The visibility within the vessel was estimated by observations of visual range made through two viewing windows. The results, shown in Figure 6, are in good agreement with those reported by Reist and Hinds⁽¹⁴⁾ for sodium burning in a 90 m³ cell.

Aerosol Chemical Composition

Samples of the suspended aerosol were taken on Teflon membrane filters and protected from laboratory air until analyzed for chemical composition by a combination of x-ray diffractometry, infrared spectroscopy and wet chemistry. The chemical form of the aerosol in test AB1 is listed in Table IV for several times during the test and for the composite floor deposit. The chemical composition changed continually during the test as reactions occurred with the O₂, H₂O and CO₂ in the atmosphere. Shortly after the sodium spill the aerosol was predominantly NaOH, formed by reaction of sodium oxide aerosol particles with the ambient humidity in the atmosphere. The initial 10°C dew point atmosphere in test AB1 contained 9.0 kg of water vapor,

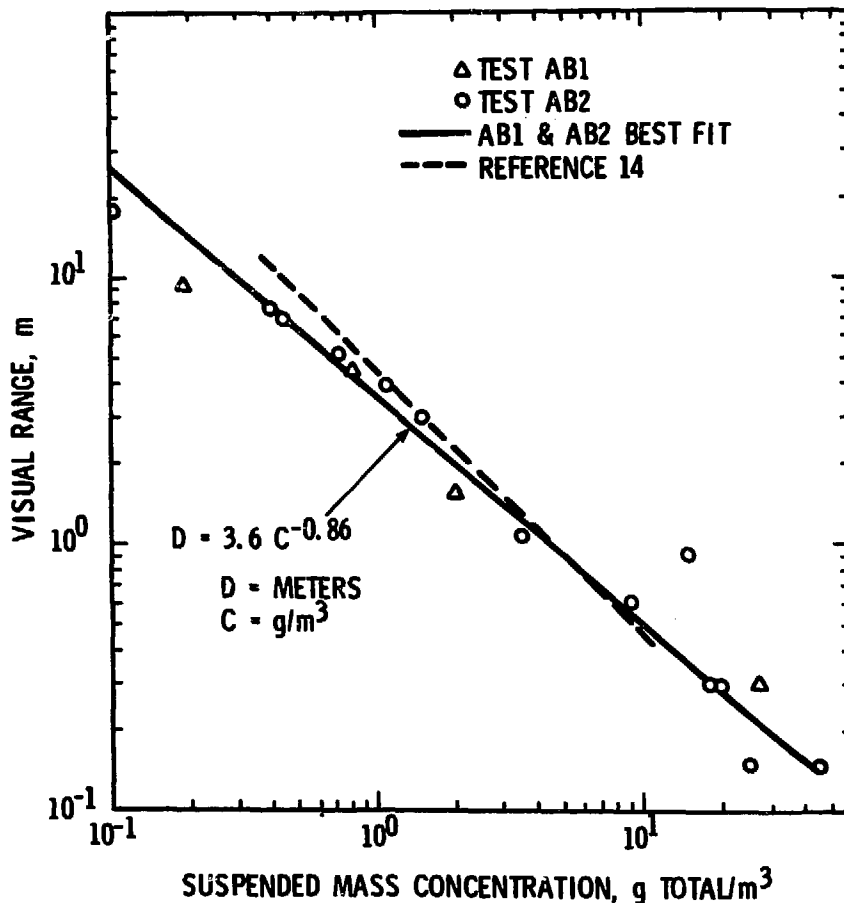


Figure 6. Visual range in a closed vessel during a sodium pool fire.

sufficient to convert half of the released sodium to NaOH. In test AB2 the aerosol was completely converted to NaOH with excess water of hydration and solution. The average composition of the AB2 aerosol was equivalent to a 60 wt% solution of NaOH.

Approximately 75 filter papers taken from four different locations during each test were protected from laboratory air until weighed to obtain total mass. The samples were then analyzed for sodium content and the mass fraction of sodium in the suspended aerosol was calculated and plotted as a function of time in Figure 7. The effect of reactions with the atmosphere is displayed clearly. Analysis for only sodium could lead to significant errors in total mass.

A 2-g sample of the composite settled "dry" aerosol was exposed to the laboratory air (35% relative humidity) and its weight and volume measured as a function of time. Within about 30 hr the weight had doubled and gas bubbles had formed. The color changed from yellow-tan to white. The bulk volume increased by a factor of approximately 15 due to the presence of the gas bubbles. Continued exposure to room air caused the sample to lose weight and form a hard crust of sodium carbonate.

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Table IV. Aerosol chemical analysis - test AB1.

(a)	Mass Fraction				
	Suspended Aerosol ^(b)				Composite ^(c)
	Sample S1 t=16 min	S2 t=46 min	S3 t=190 min	S4 t=610 min	Sample S5 t=5d
Na ₂ O ₂	0.150	0.560	0.430	0.350	0.273
Na ₂ CO ₃	0.029	0.004	0.018	0.061	0.009
NaH	0.0001	0.007	0.0011	0.011	0.0008
NaOH	0.821	0.429	0.479	0.421	0.670
H ₂ O	0.0	0.0	0.072	0.097	0.047
Total	1.0000	1.0000	1.0000	1.0000	1.0000
Total Na	0.573	0.585	0.538	0.519	0.551
Total H	0.0205	0.011	0.020	0.023	0.022
Total C	0.0033	0.0005	0.0021	0.0069	0.0011

(a) Na₂O₂, Na₂CO₃, NaH and Total Na analyzed by wet chemistry and infrared spectroscopy. NaOH and H₂O calculated. Na₂O not detected by x-ray diffraction.

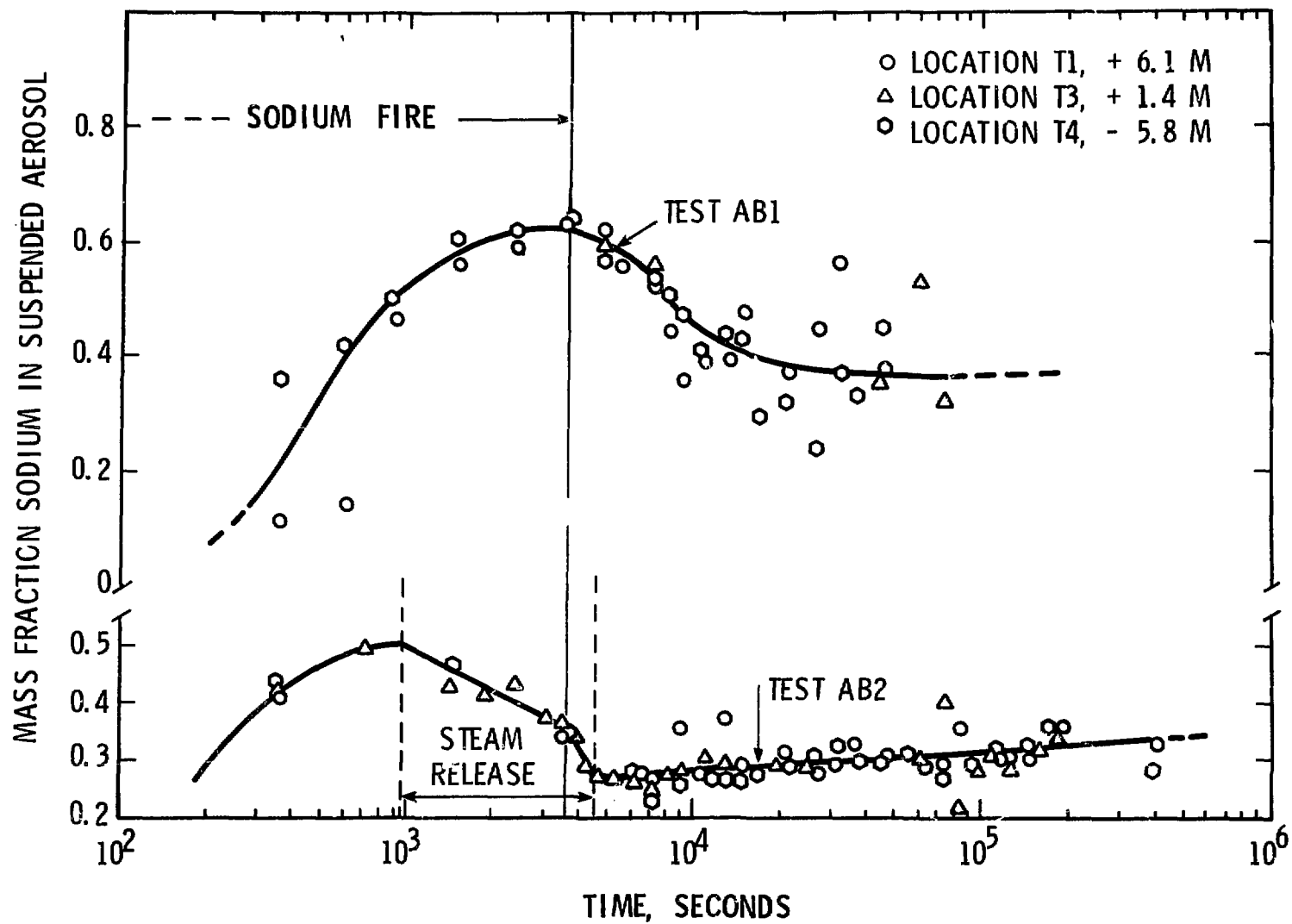
(b) Material collected on membrane filter at stated time.

(c) Floor deposit obtained at end of test.

Suspended Mass Concentration

The concentrations of suspended mass at various times are listed in Tables V and VI for tests AB1 and AB2, respectively. The figures in the tables are mean values of samples taken concurrently at four locations from high, low and central regions in the containment atmosphere. The standard deviation from the mean is also shown. Individual measurements are believed accurate to $\pm 15\%$. The concentrations are reported in two ways—as total mass and as sodium. The data of Tables V and VI are plotted as a function of time in Figures 8 and 9 for total mass. The maximum concentrations of suspended sodium were 15 and 12 g/m³ for tests AB1 and AB2, respectively. The maximum concentrations of total mass were 22 and 34 g/m³.

As soon as the burn pan lid was closed, stopping the aerosol release, the concentrations decreased rapidly with ever increasing half times. The concentration of dry aerosol in test AB1 decreased more rapidly than the wet AB2 aerosol after the source was terminated, but the wet aerosol was removed more rapidly during the source period. The overall result was that the attenuation factor for suspended sodium mass available for leakage was lower in the wet test than in the dry test. This is displayed graphically in Figure 10. The attenuation factor was calculated by integrating numerically the observed concentration over the time period of interest and dividing by the product of the release concentration and time. The attenuation factor is a multiplier in evaluating the mass available for leakage to the environs. Figure 10 shows that the addition of steam was beneficial, with 25% greater attenuation at 2 hr and 15% greater at 5 days. Although these differences are not great, it is important to know that the effect is beneficial rather than harmful because water vapor would be present to a significant extent in many postulated accidents.



HEDL 7806-14.6

Figure 7. Mass fraction of sodium in suspended aerosol.

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Table V. Suspended mass concentration - test AB1.

Time (a) sec	Suspended mass concentration, g/m ³ (c)	
	Sodium	Total Mass
3.6 (2)	5.62 (0) ± 9.0 (-1)	2.7 (1) ± 1.3 (1)
6.0 (2)	1.12 (1) ± 5.0 (0)	2.8 (1) ± 1.4 (1)
9.0 (2)	1.42 (1) ± 8.3 (-1)	2.8 (1) ± 3.5 (0)
1.5 (3)	1.32 (1) ± 2.1 (0)	2.3 (1) ± 4.0 (0)
1.8 (3)	1.30 (1) ± 2.2 (-1)	2.2 (1) ± 1.5 (0)
2.4 (3)	1.60 (1) ± 4.5 (0)	2.6 (1) ± 7.5 (0)
3.1 (3)	1.54 (1) ± 8.4 (-1)	2.5 (1) ± 2.1 (0)
3.6 (3)	1.82 (1) ± 4.4 (0)	2.9 (1) ± 7.3 (0)
4.2 (3)	8.25 (0) ± 2.7 (-1)	1.3 (1) ± 1.0 (0)
4.8 (3)	6.35 (0) ± 1.8 (0)	1.0 (1) ± 2.9 (0)
5.4 (3)	4.30 (0) ± 1.1 (0)	7.4 (0) ± 2.0 (0)
6.3 (3)	2.09 (0) ± 1.3 (-1)	3.7 (0) ± 3.5 (-1)
7.2 (3)	1.80 (0) ± 5.1 (-1)	3.4 (0) ± 1.0 (0)
8.1 (3)	1.02 (0) ± 2.7 (-1)	2.0 (0) ± 5.7 (-1)
9.0 (3)	7.16 (-1) ± 1.4 (-1)	1.5 (0) ± 3.3 (-1)
1.1 (4)	4.54 (-1) ± 5.9 (-2)	1.0 (0) ± 1.7 (-1)
1.3 (4)	3.17 (-1) ± 3.6 (-2)	7.5 (-1) ± 1.5 (-1)
1.5 (4)	2.17 (-1) ± 4.8 (-2)	5.3 (-1) ± 1.4 (-1)
1.6 (4)	1.52 (-1) ± 1.2 (-2)	3.8 (-1) ± 6.4 (-2)
1.75 (4)	1.55 (-1) ± 5.0 (-3)	4.0 (-1) ± 7.3 (-2)
1.93 (4)	8.95 (-2) ± 7.4 (-3)	2.3 (-1) ± 4.5 (-2)
2.09 (4)	9.89 (-2) ± 4.9 (-4)	2.6 (-1) ± 4.8 (-2)
2.60 (4)	5.60 (-2) ± 1.7 (-2)	1.5 (-1) ± 5.3 (-2)
3.20 (4)	2.78 (-2) ± 1.1 (-2)	7.5 (-2) ± 3.3 (-2)
3.80 (4)	2.48 (-2) ± 4.1 (-3)	6.7 (-2) ± 3.5 (-2)
4.60 (4)	1.50 (-2) ± 9.8 (-4)	4.0 (-2) ± 8.0 (-3)
6.09 (4)	7.46 (-3) ± 1.0 (-4)	2.1 (-2) ± 4.1 (-3)
7.38 (4)	5.97 (-3) ± 1.8 (-3)	1.7 (-2) ± 6.1 (-3)
8.70 (4)	2.52 (-3) ± 8.5 (-4)	7.0 (-3) ± 2.7 (-3)
1.03 (5)	2.19 (-3) ± 3.6 (-4)	6.1 (-3) ± 1.6 (-3)
1.08 (5)	1.55 (-3) ± 1.4 (-5)	4.3 (-3) ± 8.4 (-4)
1.19 (5)	1.33 (-3) ± 3.5 (-5)	3.7 (-3) ± 6.5 (-4)
1.27 (5)	1.14 (-3) ± 2.1 (-4)	3.2 (-3) ± 8.5 (-4)
1.35 (5)	1.11 (-3) ± 2.0 (-4)	3.1 (-3) ± 8.2 (-4)
1.49 (5)	7.74 (-4) ± 5.8 (-5)	2.2 (-3) ± 4.6 (-4)
1.82 (5)	5.80 (-4) ± 1.5 (-4)	1.6 (-3) ± 5.2 (-4)
4.17 (5)	6.45 (-5) ± 2.1 (-6)	1.8 (-4) ± 3.3 (-5)
4.27 (5)	1.78 (-2) ± 3.5 (-3)	4.9 (-2) ± 1.4 (-2)
4.42 (5) (b)	1.05 (-2) ± 3.0 (-3)	2.9 (-2) ± 1.0 (-2)

(a) Numbers in parentheses are exponents of base 10; e.g., read 1.05 (-2) as 1.05 x 10⁻².

(b) Ventilation fan started at 4.25 (5) sec.

(c) Mean of 4 sampling locations.

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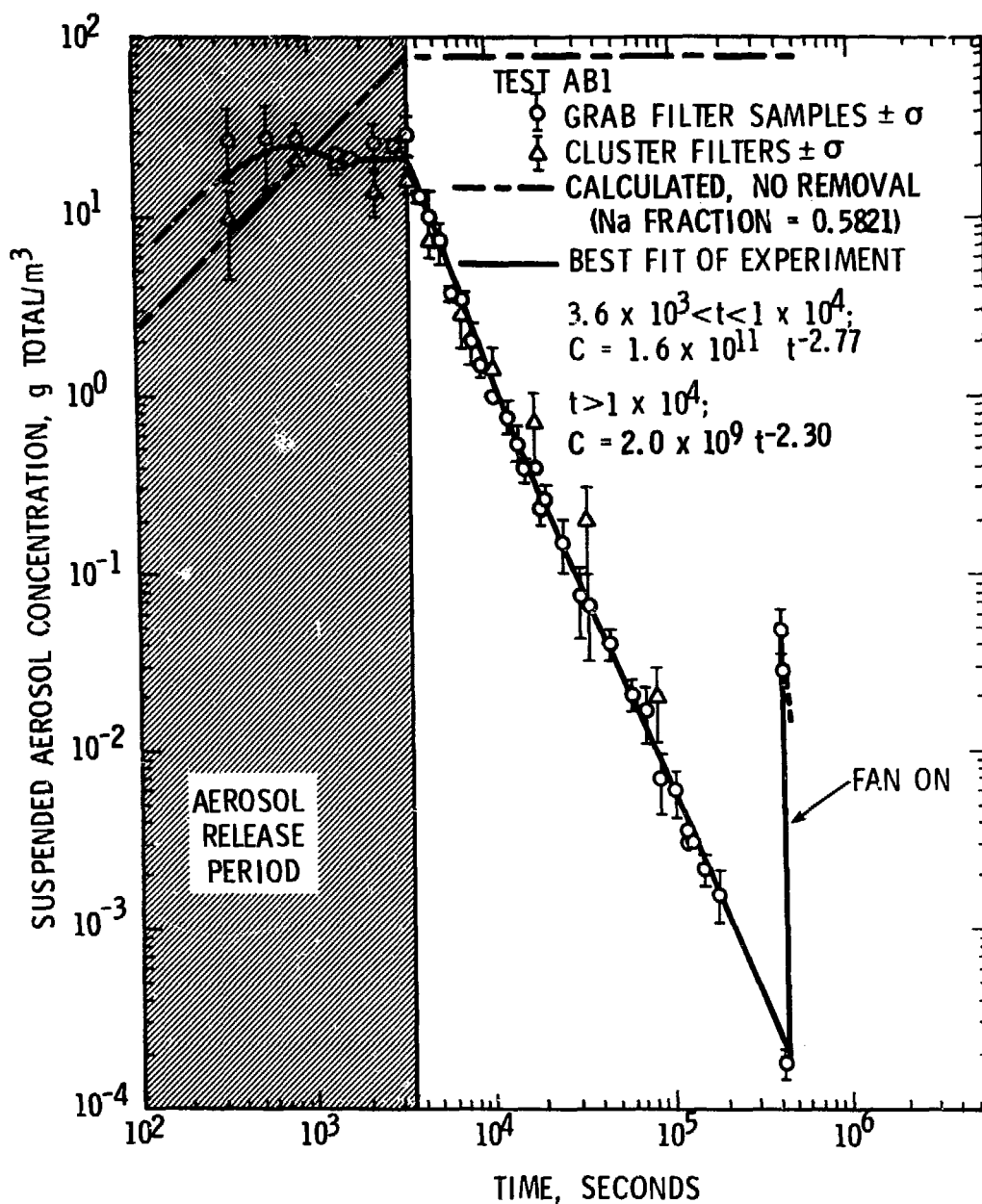
Table VI. Suspended mass concentration - test AB2.

Time ^(a) sec	Suspended mass concentration, g/m ³ (c)			
	Sodium		Total Mass	
3.60 (2)	7.32 (0)	± 4.3 (0)	1.74 (1)	± 1.0 (1)
7.20 (2)	1.09 (1)	± 1.5 (0)	2.22 (1)	± 3.6 (0)
1.02 (3)	7.30 (0)	± 1.5 (0)	1.49 (1)	± 3.3 (0)
1.44 (3)	9.83 (0)	± 1.3 (0)	2.18 (1)	± 3.5 (0)
1.80 (3)	8.97 (0)	± 1.5 (0)	2.22 (1)	± 4.2 (0)
2.40 (3)	1.09 (1)	± 2.9 (0)	2.73 (1)	± 7.8 (0)
3.24 (3)	1.11 (1)	± 1.8 (0)	2.85 (1)	± 5.5 (0)
3.60 (3)	1.26 (1)	± 2.5 (0)	3.60 (1)	± 8.2 (0)
4.32 (3)	9.11 (0)	± 2.5 (0)	3.14 (1)	± 3.4 (0)
4.80 (3)	6.22 (0)	± 1.1 (0)	2.30 (1)	± 4.8 (0)
5.40 (3)	4.29 (0)	± 9.1 (-1)	1.59 (1)	± 3.8 (0)
6.30 (3)	3.26 (0)	± 6.1 (-1)	1.21 (1)	± 2.6 (0)
7.20 (3)	2.24 (0)	± 2.5 (-1)	8.00 (0)	± 1.4 (0)
8.20 (3)	9.92 (0)	± 5.9 (-1)	6.86 (0)	± 2.3 (0)
9.00 (3)	1.61 (0)	± 6.0 (-1)	5.75 (0)	± 2.3 (0)
1.13 (4)	6.58 (-1)	± 2.3 (-1)	2.35 (0)	± 8.9 (-1)
1.29 (4)	3.03 (-1)	± 3.0 (-2)	1.08 (0)	± 1.9 (-1)
1.47 (4)	2.86 (-1)	± 7.2 (-2)	1.02 (0)	± 3.0 (-1)
1.63 (4)	2.53 (-1)	± 9.0 (-3)	9.05 (-1)	± 1.3 (-1)
1.94 (4)	1.88 (-1)	± 2.2 (-2)	6.48 (-1)	± 1.2 (-1)
2.53 (4)	1.20 (-1)	± 1.1 (-2)	4.00 (-1)	± 6.5 (-2)
3.21 (4)	7.07 (-2)	± 2.5 (-3)	2.35 (-1)	± 3.2 (-2)
3.75 (4)	4.90 (-2)	± 5.6 (-3)	1.63 (-1)	± 2.9 (-2)
4.50 (4)	3.28 (-2)	± 3.6 (-3)	1.09 (-1)	± 1.9 (-2)
6.06 (4)	2.05 (-2)	± 2.1 (-3)	6.61 (-2)	± 1.1 (-2)
7.35 (4)	1.57 (-2)	± 1.2 (-3)	5.06 (-2)	± 9.0 (-3)
8.61 (4)	1.08 (-2)	± 9.1 (-4)	3.48 (-2)	± 6.3 (-3)
9.60 (4)	7.63 (-3)	± 2.2 (-3)	2.38 (-2)	± 7.8 (-3)
1.10 (5)	6.05 (-3)	± 5.1 (-4)	1.89 (-2)	± 3.3 (-3)
1.28 (5)	4.95 (-3)	± 1.1 (-3)	1.55 (-2)	± 4.2 (-3)
1.46 (5)	3.17 (-3)	± 2.9 (-4)	1.02 (-2)	± 1.8 (-3)
1.73 (5)	2.15 (-3)	± 3.9 (-4)	6.72 (-3)	± 1.6 (-3)
1.91 (5)	1.50 (-3)	± 1.4 (-4)	4.69 (-3)	± 8.6 (-4)
4.02 (5)	1.72 (-4)	± 1.1 (-5)	5.06 (-4)	± 8.0 (-5)
4.43 (5) ^(b)	<3.0 (-5)		<8.8 (-5)	

(a) Numbers in parentheses are exponents of base 10.

(b) Ventilation fan started at 4.42 (5) sec.

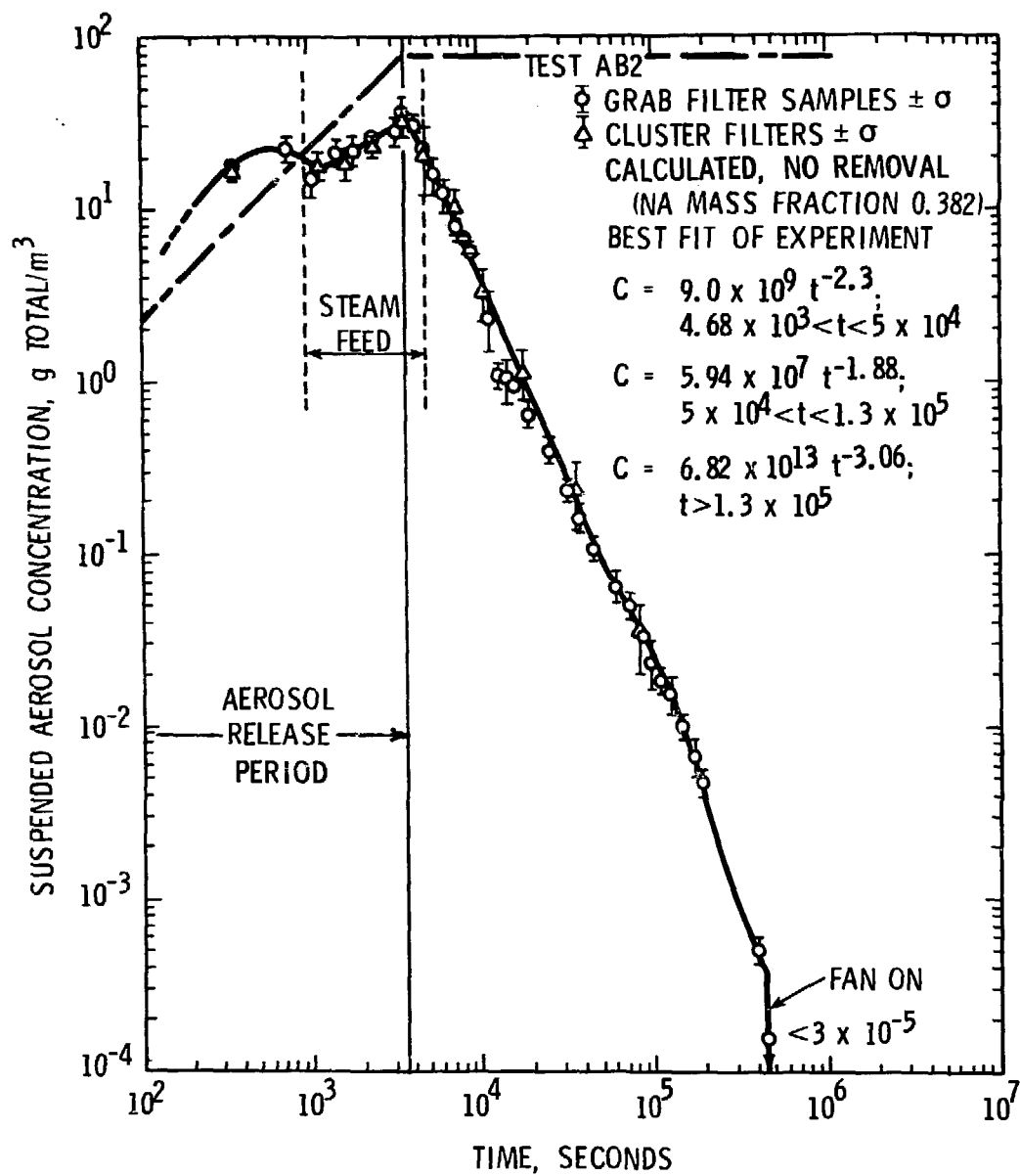
(c) Mean of four sampling locations.



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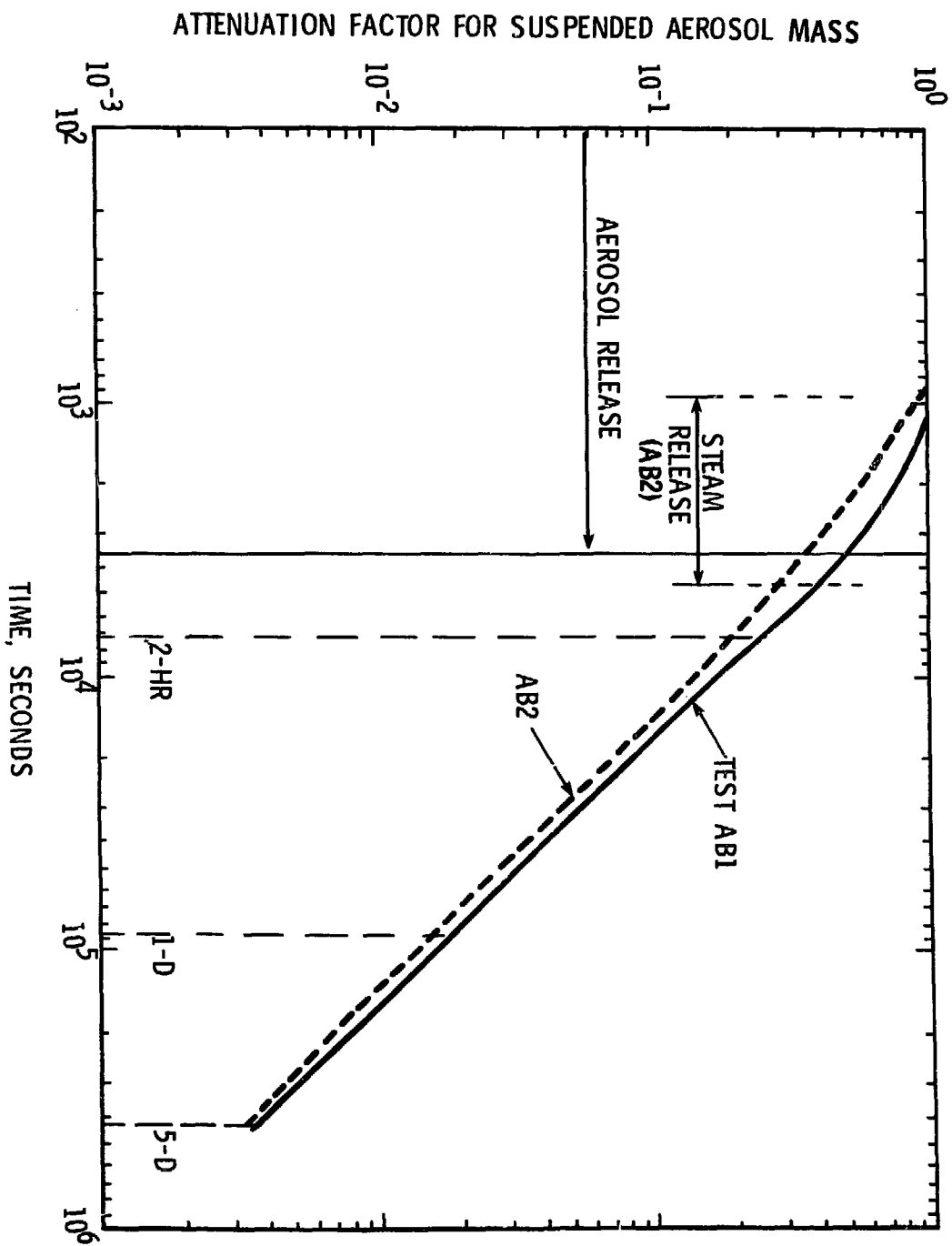
Figure 8. Suspended mass concentration—test AB1.

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HEDL 7806-14.7

Figure 9. Suspended mass concentration—test AB2.



HEDL 7806-14.5

Figure 10. Attenuation factors for suspended mass concentration available for leakage.

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Aerosol Physical Properties

Particle Size Distribution. The aerodynamic size distribution of the suspended aerosol mass was measured by inserting two types of cascade impactors directly into the containment atmosphere in a horizontal position. Although there was no inlet tube, some deposition did occur in the nose sections upstream of the first stages. This material was neglected in analyzing the data. The two types of cascade impactors were an eight-stage multi-circular jet type (Anderson Mark III) and a six-stage multi-rectangular jet type (Sierra Model 226). A typical example of the data plotted on log-normal paper as the stage cut-off diameter vs. the mass fraction penetrating that stage is shown in Figure 11. If the particle distribution is log-normal, a straight line should be obtained and the two major parameters of particle size distribution can be obtained: the aerodynamic mass median diameter, AMMD, and the geometric standard deviation, σ_g . Most of the 44 cascade impactor samples taken in these tests gave good approximations to a straight line on log-normal paper, though a few showed significant deviations from it.

Samples were taken simultaneously from high and low containment locations. No consistent difference was found for the two locations, which supports the conclusion made earlier that the gas space was well mixed. The results for the upper and lower samples were averaged and plotted as a function of time in Figures 12 and 13 for the AMMD and σ_g , respectively. The rectangular jet impactor gave a significantly larger median diameter than the circular jet impactor, especially at early times when the particles were larger. The reason for this discrepancy has not been resolved. Possible, as suggested by Allen and Briant⁽¹⁵⁾, this is caused by interception of very large particles of lower than average density on the first few stages, but it is not known why a rectangular geometry would accentuate this effect.

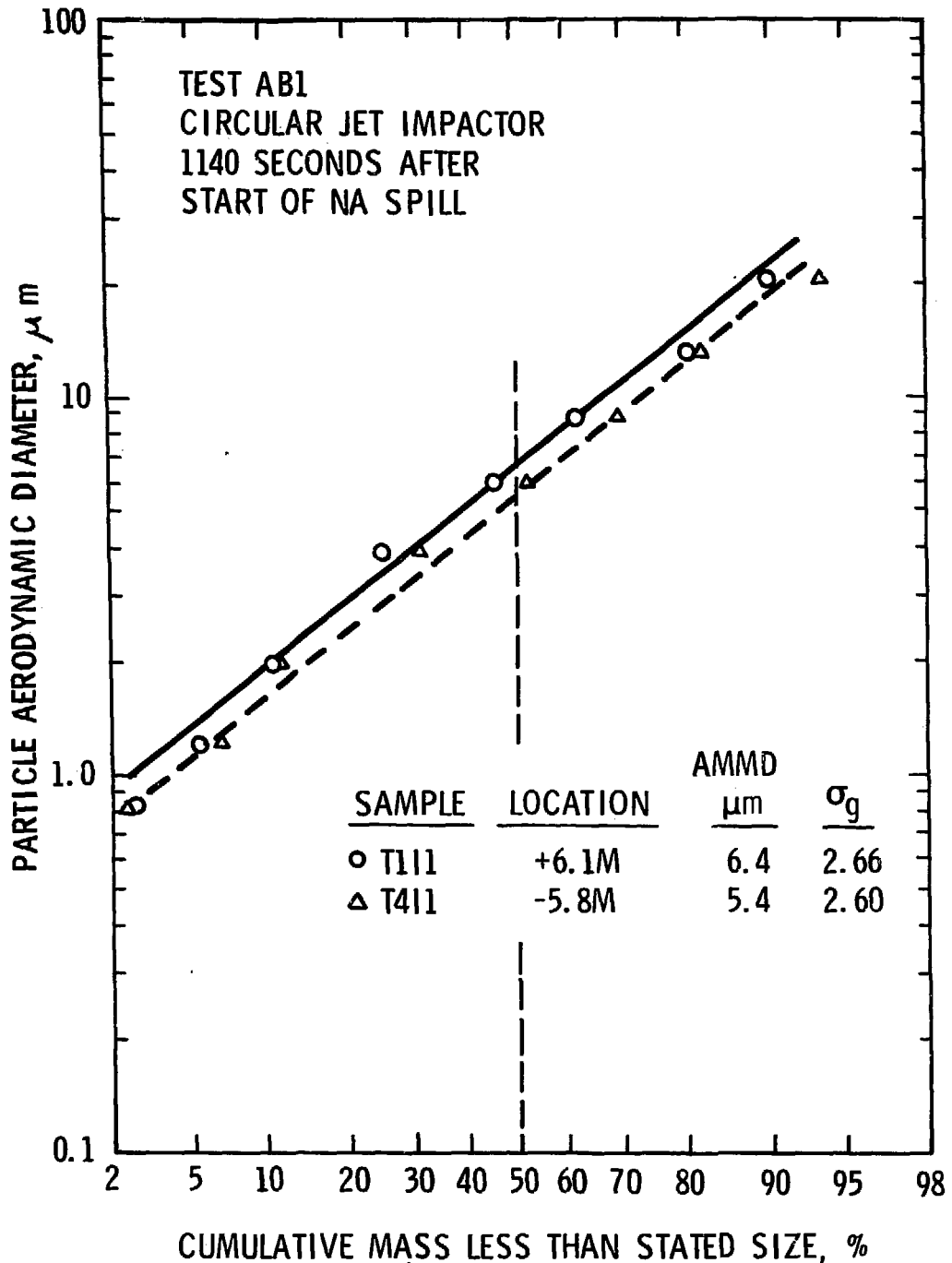
A comparison of the cascade impactor results with the settling diameter, d_s , calculated from the rate of depletion of airborne concentration is given in Figures 14 and 15. If the distribution is log-normal, the aerodynamic settling diameter can be calculated from the cascade impactor by equation (1).

$$d_s = \text{AMMD} \exp (\ln^2 \sigma_g) \quad (1)$$

The aerodynamic settling diameters were also calculated from the airborne concentration curves (Figures 8 and 9) by differentiating the curves and equating with the Stokes settling removal term. This neglects wall plateout, which is reasonable in view of the fact that 93% of the mass settled and only 7% plated. Thus,

$$\frac{dC}{dt} = - \frac{u_t A_s C}{V} \quad (2)$$

where C = time dependent suspended mass concentration
 u_t = terminal settling velocity
 A_s = horizontal surface
 V = gas volume.



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Figure 11. Typical cascade impactor data plot on log-normal paper.

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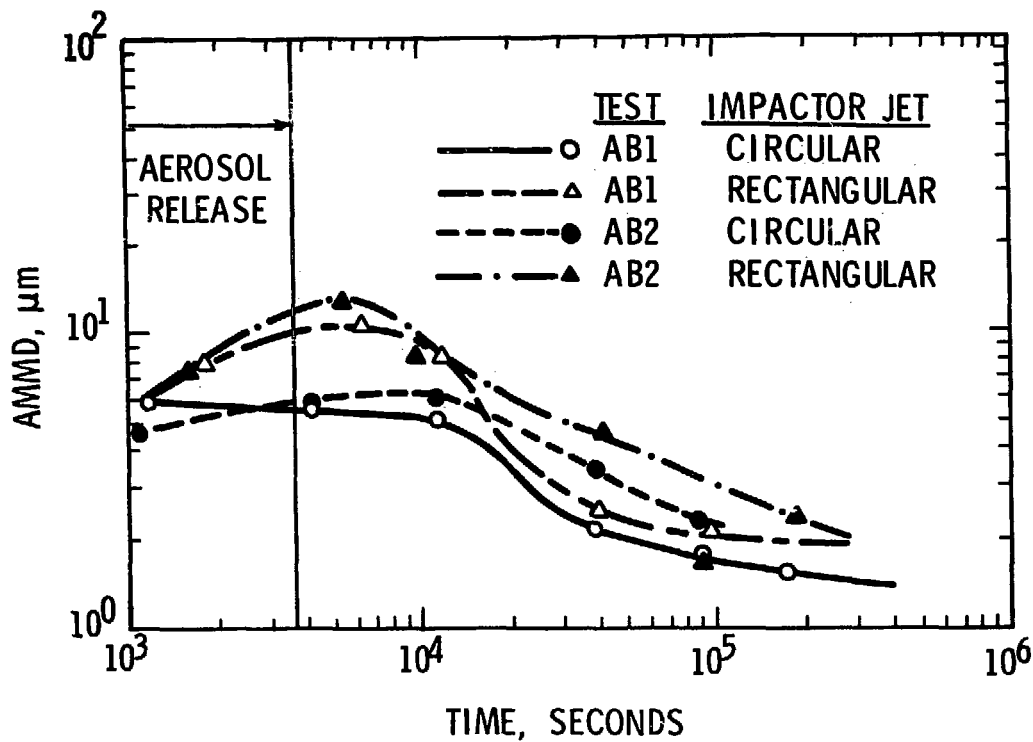


Figure 12. Aerodynamic mass median diameter of suspended particles.

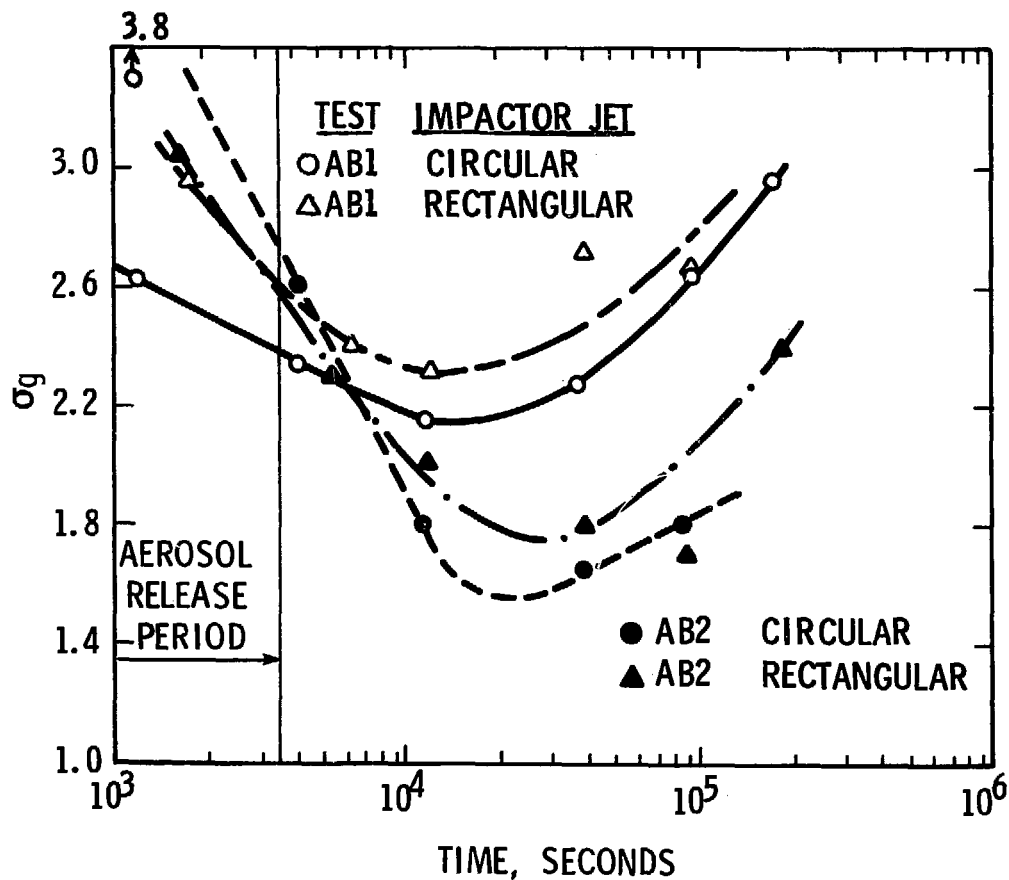


Figure 13. Standard geometric deviation of suspended particle size.

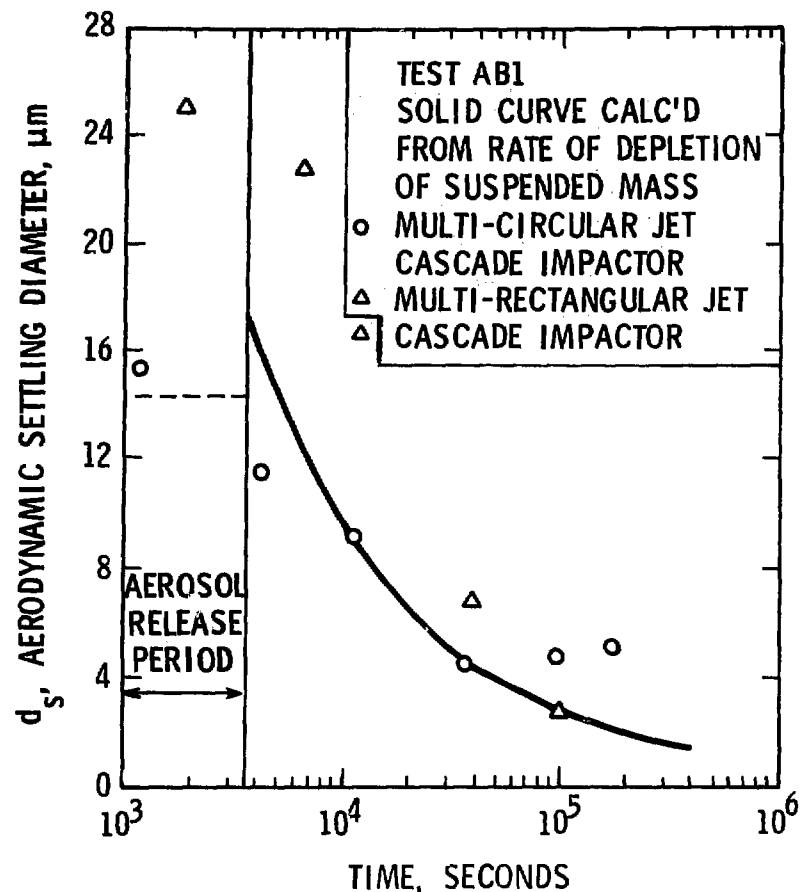


Figure 14. Comparison of aerodynamic settling diameters from cascade impactor and airborne depletion data—test AB1.

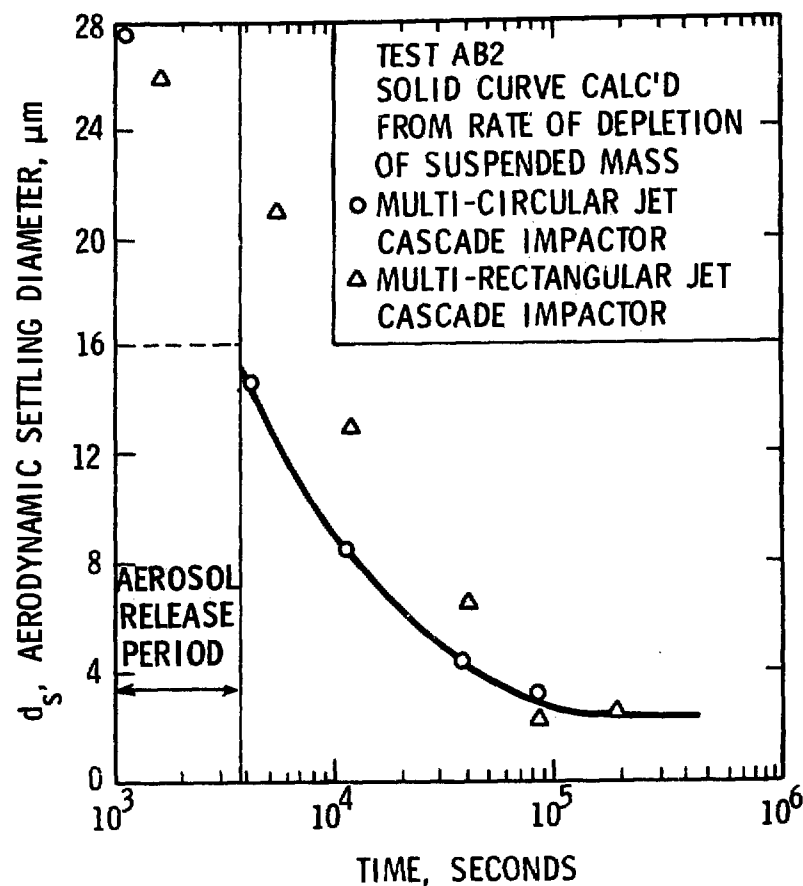


Figure 15. Comparison of aerodynamic settling diameter from cascade impactor and airborne depletion data—test AB2.

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The settling velocity, u_t , was calculated by equation (2) and the settling diameter calculated from Stoke's law:

$$d_s = \left(\frac{18 \mu_f u_t}{g \rho_p} \right) \quad (3)$$

where μ_f = fluid viscosity
 g = acceleration due to gravity
 ρ_p = effective particle density.

In using equation (3), the particles were assumed to be spherical with unit density in order to compare with the similar assumption used for the cascade impactor data. Figures 14 and 15 show that fairly good agreement was obtained with the circular jet impactor.

Particle Density. The bulk density of the composite deposited aerosol materials recovered from the floor after test AB1 was determined by weight and volume measurements to be 0.33 ± 0.03 g/cm³. This agrees well with the value of 0.32 reported by First and Yusa (19). After tapping 100 times in a graduated cylinder, the bulk density increased to 0.60 ± 0.10 g/cm³. The material density of the AB1 material was not determined, but a similar sample with the same nominal composition (test AB3) was found to have a material density of 2.3 ± 0.3 g/cm³ by immersion in acetone. The handbook value, based on the analysis of sample S5 in Table IV is 2.30 g/cm³.

No density measurements were made of the aerosol material in test AB2, but it was believed to have been liquid during most of the test. At the end of the test 13.4 liters of 65 wt% NaOH solution was drained from the vessel. However, portions of the vessel surface had a deposit of solid material. The agglomerated aerosol particles probably contained some void space due to oxygen gas emitted by the reaction of Na₂O₂ and water and to the insolubility of the small amounts of sodium carbonate which formed. No direct measurements were made of the particle density.

Electron Microscope. Electron microscope grids were exposed briefly to the containment atmosphere at various times. They were protected from exposure to ambient air by withdrawing the holder into an o-ring sealed protector before removing it from the containment vessel. Figure 16 shows some typical photo micrographs of samples taken at four times during test AB1. The indices shown in the figure are set at 10 μ m. The projected diameter of the primary particles ranged from 0.1 to 0.5 μ m. The settled agglomerates were irregular shapes of approximately 1-5 μ m diameter at 8 minutes. They had grown to 1-30 μ m diameter spheres by the end of the sodium fire. The larger particles appeared to contain voids, but this observation is suspect because the vacuum in the transmission electron microscope may have altered the geometry of the particles. Shadowing techniques were not used, but stereoscopic examination showed the particles to be approximate spheres.

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**AEROSOL PARTICLES AT 8 MINUTES DURING
TEST ABI**



Figure 16a. Time 8 min.

**AEROSOL PARTICLES AT 56 MINUTES DURING
TEST ABI**

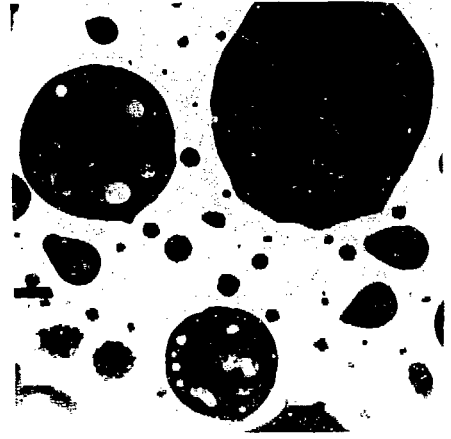


Figure 16b. Time 56 min.

**AEROSOL PARTICLES AT 190 MINUTES DURING
TEST ABI**

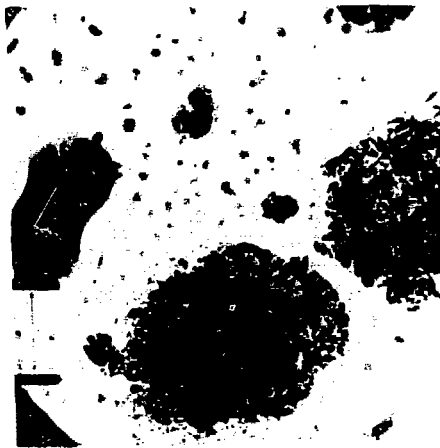


Figure 16c. Time 190 min.

AEROSOL PARTICLES AT 25 HR DURING TEST ABI

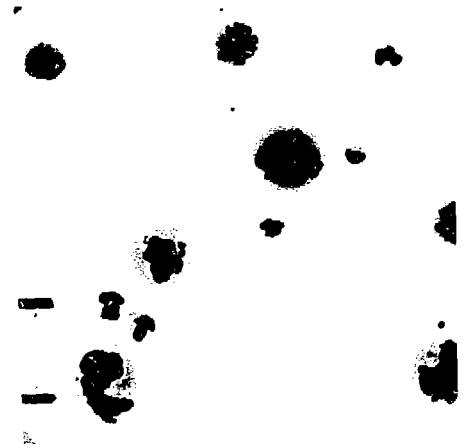


Figure 16d. Time 1500 min.

Figure 16. Electron photomicrographs of gravity settled particles at various times in test ABI.

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Electric Charge

Particle charge distributions were measured by the use of a parallel plate charge analyzer of the type described by Gillespie and Langstroth⁽¹⁶⁾. For both tests the sample flow was started 30 minutes after the sodium spill and lasted for 10 minutes. Filtered sheath air flow at 11 cm³/sec was recirculated by a small diaphragm pump and a net sample flow of 2 cm³/sec was bled through the analyzer and its backup filter. The apparatus was operated at 10,000 V for AB1 and 4000 V for test AB2. Any particle with a mobility greater than 7.7×10^{-6} and 1.9×10^{-5} cm²/volt sec, respectively, would be collected on one of the plates. Table VII gives the distribution.

Table VII. Particle charge distribution.

	<u>AB1</u>	<u>AB2</u>
Mass fraction on positive plate	0.46	0.07
Mass fraction on negative plate	0.45	0.07
Mass fraction on backup filter	0.09	0.86
Limiting mobility, cm ² /v-sec	7.7×10^{-6}	1.9×10^{-5}
Equivalent charge on 7- μ m dia. particle	7	18
Equivalent charge on 1- μ m dia. particle	1	3

The data of Table VII indicate that the "dry" particles in test AB1 had a higher charge than the wet aerosol, but that the net charge was close to zero in both tests.

IV. Discussion and Conclusions

The results of these tests support the following conclusions and generalized observations. The sodium burned at an average rate of 34 kg Na/hr m², releasing 8.8 kg Na/hr m² as aerosol. The presence of a steam source in the atmosphere had no significant effect on the mass release rate. The mean atmosphere temperatures increased by 64.5 and 75.5°C for the dry and wet tests, respectively. The additional heat generated in test AB2 was due to the condensation on and reaction of steam with the aerosol particles.

It is hypothesized that the aerosol particles were formed by reaction of sodium vapor with oxygen within a few centimeters of the sodium pool surface, rapid nucleation to form a high concentration of primary particles in the high temperature plume, and rapid agglomeration to form aggregate particles during the plume rise and dispersal throughout the vessel. Essentially uniform suspended mass concentration was accomplished very quickly, but the few seconds in the high concentration plume caused the particles to grow to diameter of the order of one micrometer by the time they were dispersed.

The aerosol was formed as Na₂O₂ particles in the oxygen-rich atmosphere. As these particles were dispersed throughout the vessel atmosphere, they reacted very rapidly with the water vapor present in the normal air test. Sufficient water vapor was present to convert approximately half of the total released aerosol to NaOH. Within a

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few minutes the aerosol was in the form of sodium hydroxide solution particles. As more Na_2O_2 particles were generated, they agglomerated with the sodium hydroxide particles. Some of the water of dilution in the sodium hydroxide solution particles diffused to the co-agglomerate Na_2O_2 to react to NaOH and release oxygen gas. The released gas caused a volume increase, with corresponding reduction in the effective density of the particle. Some reaction with the CO_2 occurred, but the supply of CO_2 was limited. In the test where additional water vapor was added as steam, sufficient water was available to convert all the aerosol to NaOH with excess water of solution. The overall result in both tests was an aerosol that varied with time in chemical composition, density and shape.

Cascade impactor samples showed that the size distribution of the suspended particles was approximately log-normal. Samples taken later than 24 hr indicated some departure from log-normal. Approximately 7.3% of the released mass was recovered from the vertical walls, while 92.7% settled on horizontal surfaces. All horizontal surfaces collected aerosol by sedimentation, although deposition trays exposed for the entire test put 0.4 m below an overhanging ledge received only ~70% as much as unshadowed trays. This shows that aerosol models should consider all horizontal surfaces, not just the vessel cross section.

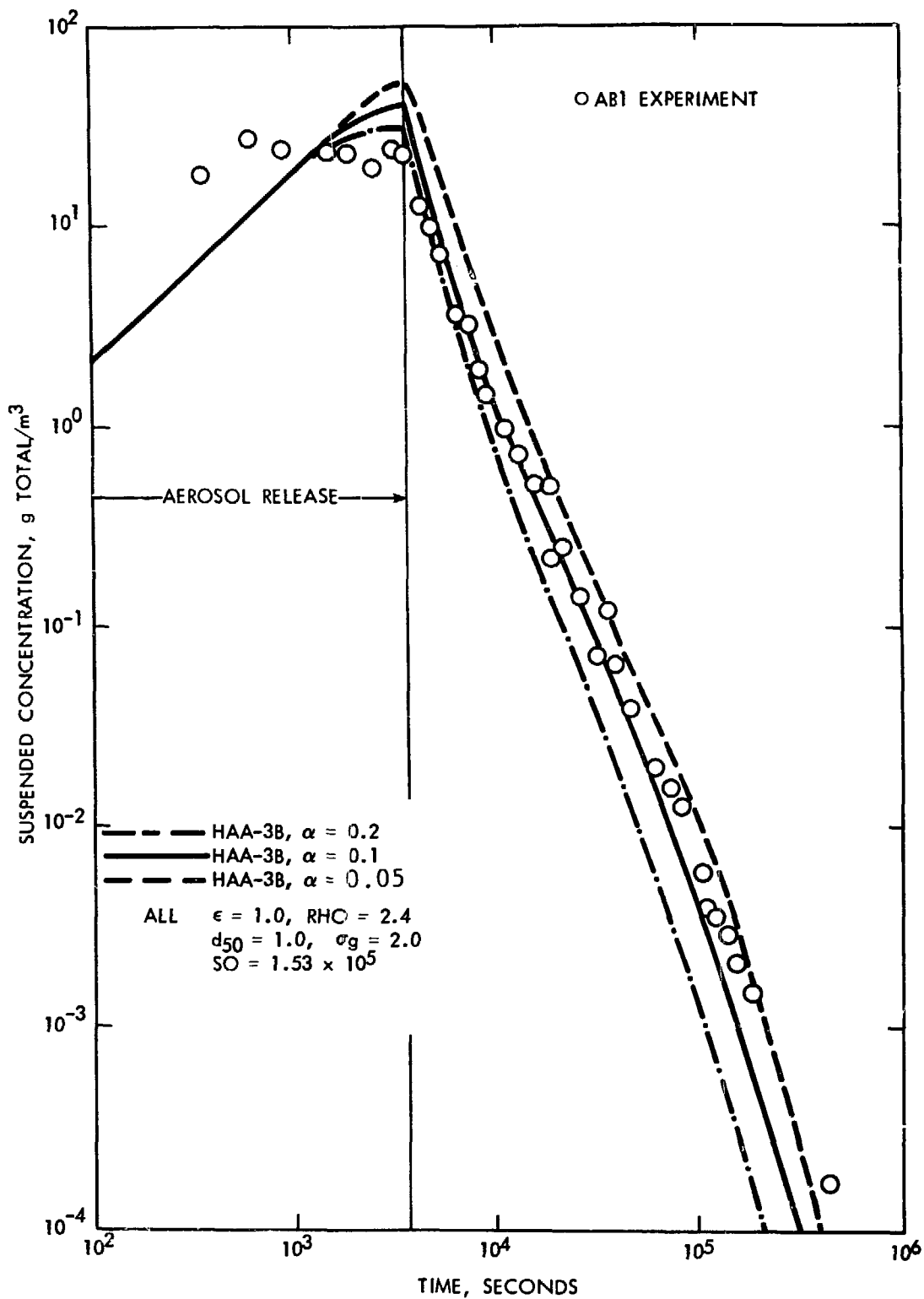
No attempt was made in this paper to evaluate specific aerosol behavior models and computer codes. However, it is of interest to note that pre-test predictions of the suspended mass concentration using the HAA-3B⁽¹⁷⁾ and HAARM⁽¹⁸⁾ codes agreed reasonably well with the experiment. One problem encountered in the pre-test predictions was the need for an accurate method to forecast the aerosol mass release rate. Even when conditions are well known, as in the present tests, there was a large uncertainty in the source rate. The HAA-3B code was rerun post-test, using the average experimentally determined source rate. The HAA-3B predictions for suspended mass concentration are compared with experimental results in Figure 17. Good agreement is shown except at the very early times, where the code underpredicts because it does not account for condensation of water vapor. The HAA-3B code predicted that 6.2% of the total release would be plated on walls and 93.8% settled to horizontal surfaces. This compares well with 7.3% and 92.7%, respectively, for the experiments. A more complete evaluation of aerosol codes is underway at HEDL and elsewhere.

It is concluded that the results of these large-scale tests in the CSTF will contribute importantly to increased confidence in the ability to understand and predict nuclear aerosol behavior during accidents in sodium cooled facilities.

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HEDL 7806-331.1

Figure 17. Comparison of HAA-3B computer code predictions with experiment.

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DISCUSSION

SCHIKARSKI: I congratulate you on your fine experiments. I want to make two remarks. In the presence of enough moisture and CO_2 in the air, the reaction to sodium carbonates is fast. This, I guess, agrees with your findings. It is a favourable effect because Na_2CO_3 is not a chemically aggressive material. The change of composition of particles in the vessel affects many parameters assumed to be constant in the codes (HAARM-2, PARADISEKO IIb, etc.). I think these codes should be improved in that respect.

HILLIARD: Thank you, Dr. Schikarski. I agree that the conversion to carbonate is very rapid when you have water vapor present. I think Jerry McCormack is going to speak on Thursday about some filter loading tests in which we injected carbon dioxide and steam and found it produced sodium carbonate very quickly, just a few minutes. But, in a sealed containment vessel that has a limited supply of carbon dioxide, i.e., no added carbon dioxide, we found an excess of sodium hydroxide. All the carbon dioxide was used up and there was no more available. Therefore, the chemical composition depends on the size of the sodium hydroxide release relative to the vessel size. Your point regarding new input to the codes is certainly relevant. I think that the density and shape of the particles is important. These will be different for dry carbonate compared to moist sodium hydroxide.

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ATTENUATION OF PARTICLES THROUGH LEAK PATHS

by

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Abstract

Particulates of sodium oxide were passed through narrow leak passages. The particulates impacted on the walls and built up plugs which eventually stopped the flow. Conditions under which plugging occurs were determined. Material which did pass through the leaks was increased in size because of agglomeration on the wall followed by detachment. The data shows that radioactive particulates escaping through leaks following a hypothetical FBR accident will tend to plug the leaks in both the reactor vessel and the containment building. Also, the particulates will generally increase in size so as to be nonrespirable.

I. INTRODUCTION

Gas borne particulates both tend to plug small passages in leak paths and also increase in size following impaction and subsequent detachment from leak-path walls. This behavior mitigates the consequences of hypothetical LMFBR Core Disassembly Accidents (CDA) by reducing the leakage both through reactor head seals and through containment building walls.

A series of tests has been made to assess the proportion of CDA particles in the cover gas escaping through assumed displaced head seals and to assess the quantity and size of the particles which could escape into the containment building.

In a second series of tests, gas containing sodium combustion product aerosols was allowed to leak through capillary-type leak paths to determine the fraction of respirable size particulates which were transmitted. With current building leak rate measurement technology, the limit of leak detection sensitivity is usually assumed to be approximately 0.1 vol % leakage per day at a pressure differential of 0.68 atmosphere for a steel reactor containment building. Assuming the total containment leak is accommodated by a single hole, the size of the leak can be represented by a hole 1 to 2 mm in diameter.

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II. PLUGGING OF HEAD SEAL LEAKAGE PATHS (TEST SERIES 1)

Following a CDA in a LMFBR, the accompanying sodium slug motion would stretch the head tiedown bolts and part of the aerosol mixture* in the cover gas could escape into the reactor containment building through broken seals. Generally, these seals would consist of a sodium filled seal, followed by a series of inflatable elastomer seals. To simulate these seals on a small scale, a multiple bend leak path was constructed, see Figure 1. This leak consisted of a series of small brass chambers having the approximate configuration of the passages in the head seal. Small diameter pipe connected the chambers together.

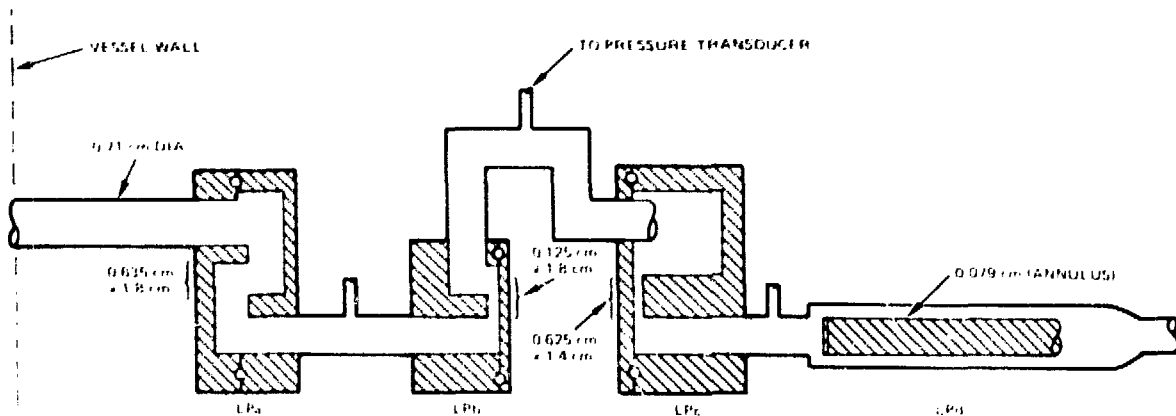
An airborne sodium oxide concentration of 200 to 700 g/m³ was produced in the high-temperature concentration aerosol (HTCA) Test Apparatus to characterize the plugging of a head seal leak. A schematic of the apparatus is shown in Figure 2. The apparatus consisted of a 427 liter vessel, two reservoirs (one for 530°C sodium - one for atomizing gas), sodium spray nozzles, and oxygen flooding nozzles. All the sodium was expelled in a few seconds and subsequently produced a very concentrated, high-temperature aerosol of sodium oxide carried by a mixture of nitrogen and oxygen. This aerosol was used to simulate the material presumed to escape from the reactor cover gas following a CDA - a mixture of core debris, sodium droplets, and gases.

In Test 1a, preliminary data on leak plugging in the head seal leak path was obtained by connecting the multiple bend leak path directly to the HTCA vessel. A spray of 100 g of 5 μ m drops of sodium was injected into the HTCA vessel in approximately 4 seconds producing an aerosol concentration of approximately 234 g/m³. The HTCA gas and airborne particles were allowed to vent directly through the multiple bend leak path to the outside atmosphere.

The maximum gas flow rate in the leak path was 8.5 l/sec. This occurred at approximately 1.5 seconds.

*The aerosol concentration in the cover gas following a CDA has been estimated to be ~ 4000 g/m³.

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Figure 1. Multiple bend leak path.

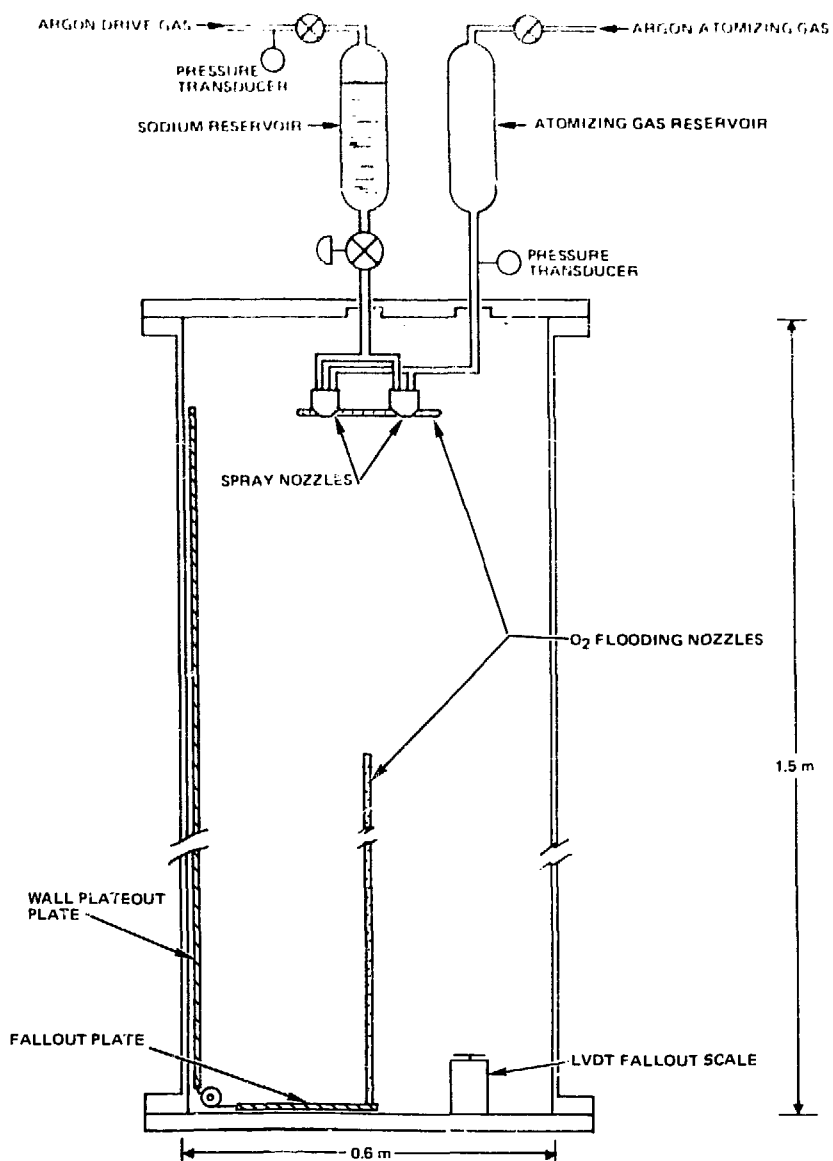


Figure 2.
Schematic of HTCA
Test Apparatus.

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The leak path plugged in 6 seconds. Post-test analysis of the data indicated that the plug could withstand 2.8 atmosphere differential pressure and that approximately 7 g of sodium, in the form of sodium oxide, was trapped in the leak path assembly. The mass of sodium found in each section of the assembly is shown in Table 1.

TABLE 1

Sodium Oxide Aerosol Collected in Multiple Bend Leak Path
(Test 1a)

Leak Path Section	Type of Leak Path	Mass of Sodium in Leak Path (g)	Percentage of Mass
S-1 (LPa)	Na Seal	6.37	91.15
S-2 (LPb)	Margin Seal No. 1	0.57	8.16
S-3 (LPc)	Dynamic Seal	0.027	0.39
S-4 (LPd)	Margin Seal No. 2	0.018	0.26
Exit	--	~0.003	~0.04

The size of the 0.04% of the sodium oxide particles which exited the leak path assembly was measured by an eight-stage Anderson impactor. Approximately 95% of the mass collected in the impactor was $>6.8 \mu\text{m}$. The median size of the particles before passing into the leak path was $2.5 \mu\text{m}$ (AED 50).

In another test, 1b, the leak path was exposed to a particle concentration of 30 g/m^3 at a pressure differential of 0.068 atmosphere. The plugging time for this set of conditions was less than 30 seconds.

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III. PLUGGING OF CONTAINMENT SHELL LEAKS (TEST SERIES 2)

In Test Series 2, sodium combustion product aerosols were allowed to pass through capillary leak paths representing flaws in a steel containment vessel to determine the attenuation of the respirable size aerosols.

These aerosol leak tests were conducted in a 427 liter vessel with flat plates on each end of the cylindrical vessel. The test vessel and simplified schematic of the injection system are shown in Figure 3. The injection system consisted of a heated pot containing sodium, an air inlet line, solenoid valve, and a pressure switch. The concentration in the vessel was regulated by a manual valve located between the solenoid valve and burn pot. A pressure switch sensed the vessel pressure and operated a solenoid valve to maintain a constant pressure in the vessel. During each test run, both total volume of gas which passed through the leak path and gas flow rate were recorded.

For the majority of the tests, the aerosol was allowed to leak through smooth capillaries 1 to 2 mm diameter by 4 cm in length. An example of the sampling methods are shown in Figures 4a and 4b. The wet test meter was modified to measure the gas flow rate as well as accumulated flow. The dry air manifold was used to determine the plug strength at the end of the test. In some tests, the air manifold was not used and the capillary entrance was mounted flush with the inner surface of the vessel wall.

Typical results for a given set of conditions in which plugging occurs is presented in Figure 5. For this series of tests, the aerosol concentration and humidity were maintained constant. The data indicates a direct relationship between pressure applied across the capillary and the mass required to plug it. In addition, measurements of the plug mass and the mass escaping have shown that the major fraction of the entering aerosol mass is the plug mass. Further tests have shown that a higher initial pressure applied across the capillary before plugging usually requires a greater pressure to unplug.

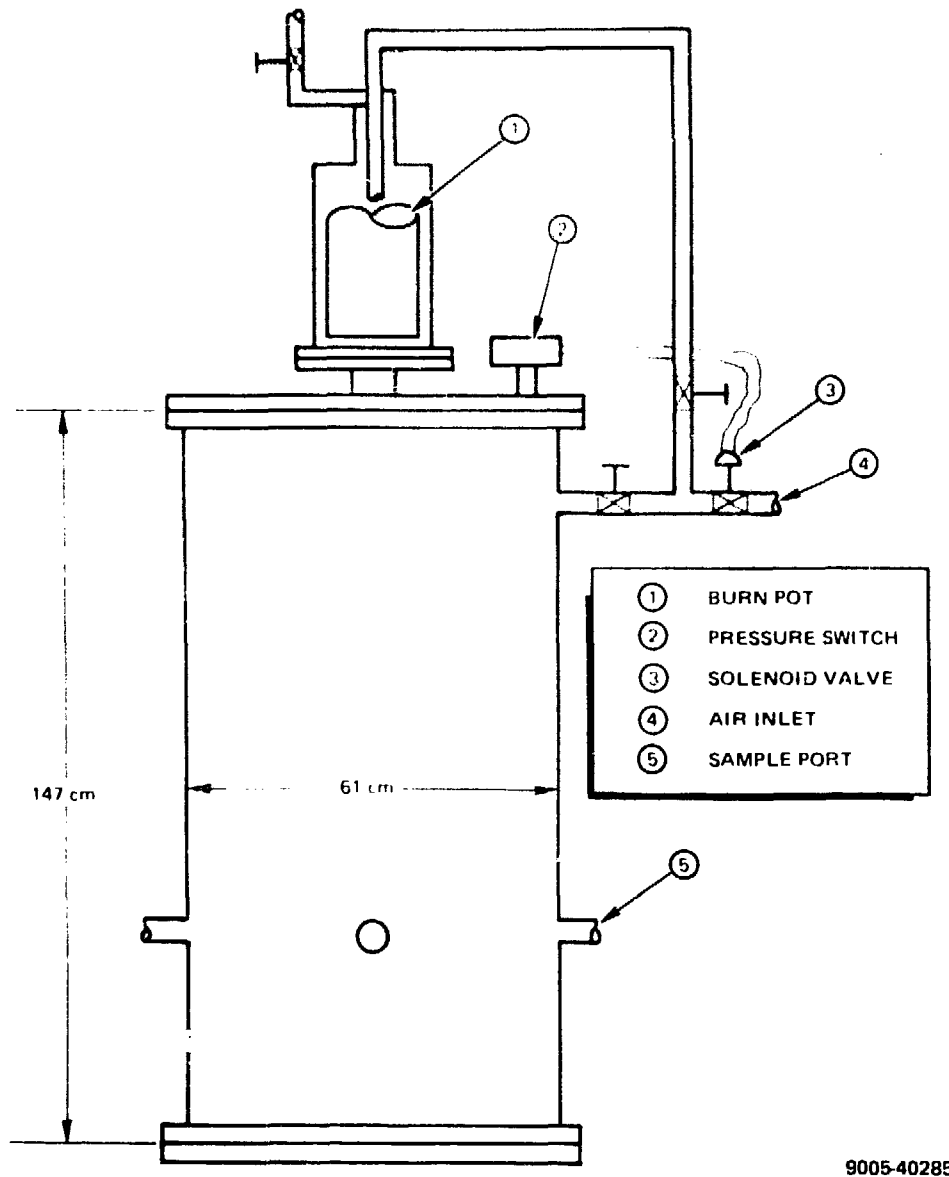
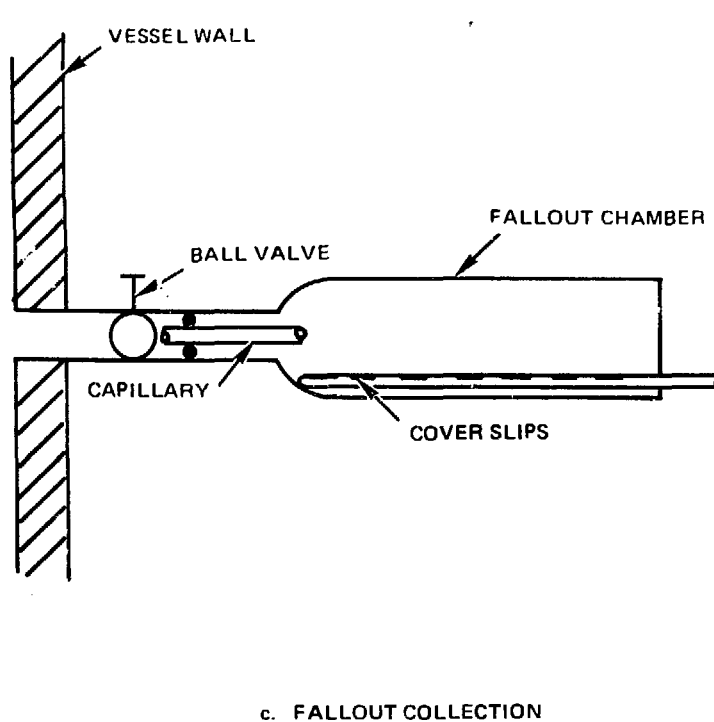
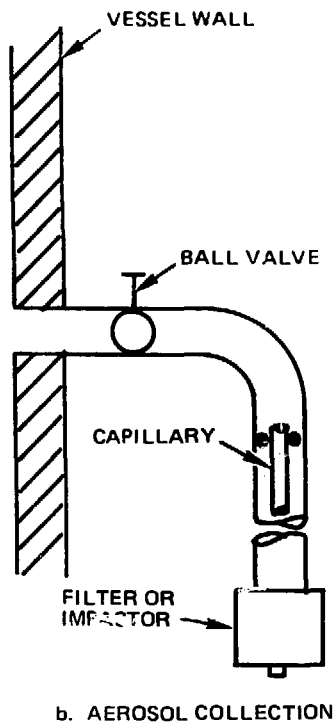
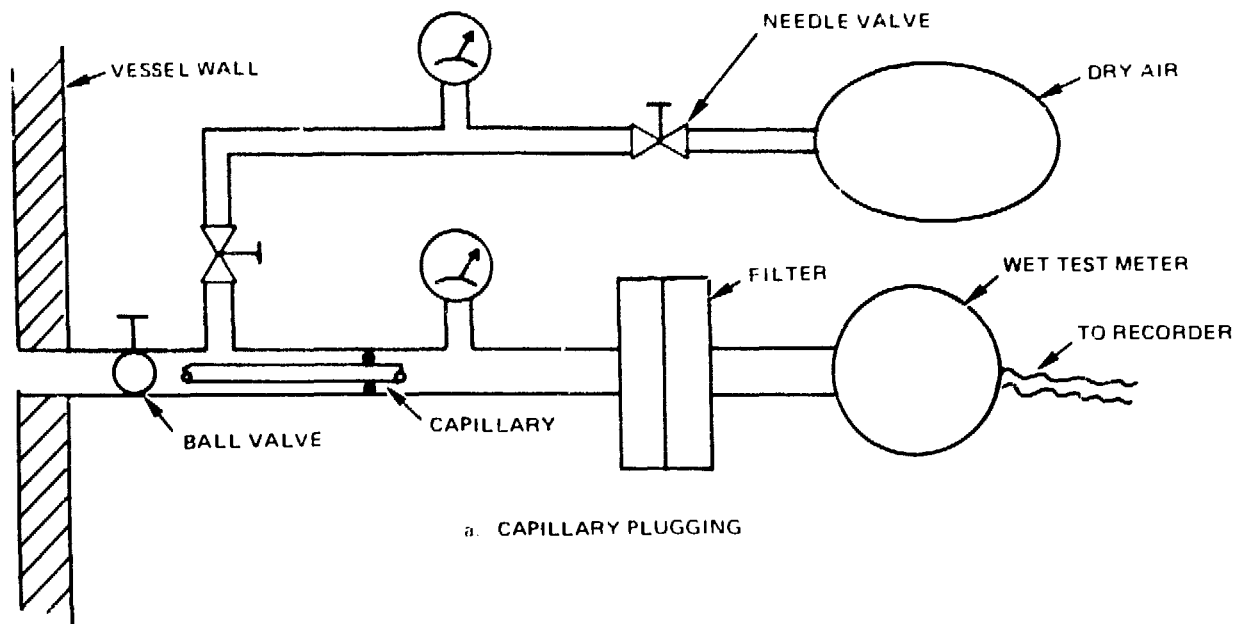


Figure 3. Test vessel.

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9005-40193

Figure 4. Sampling methods.

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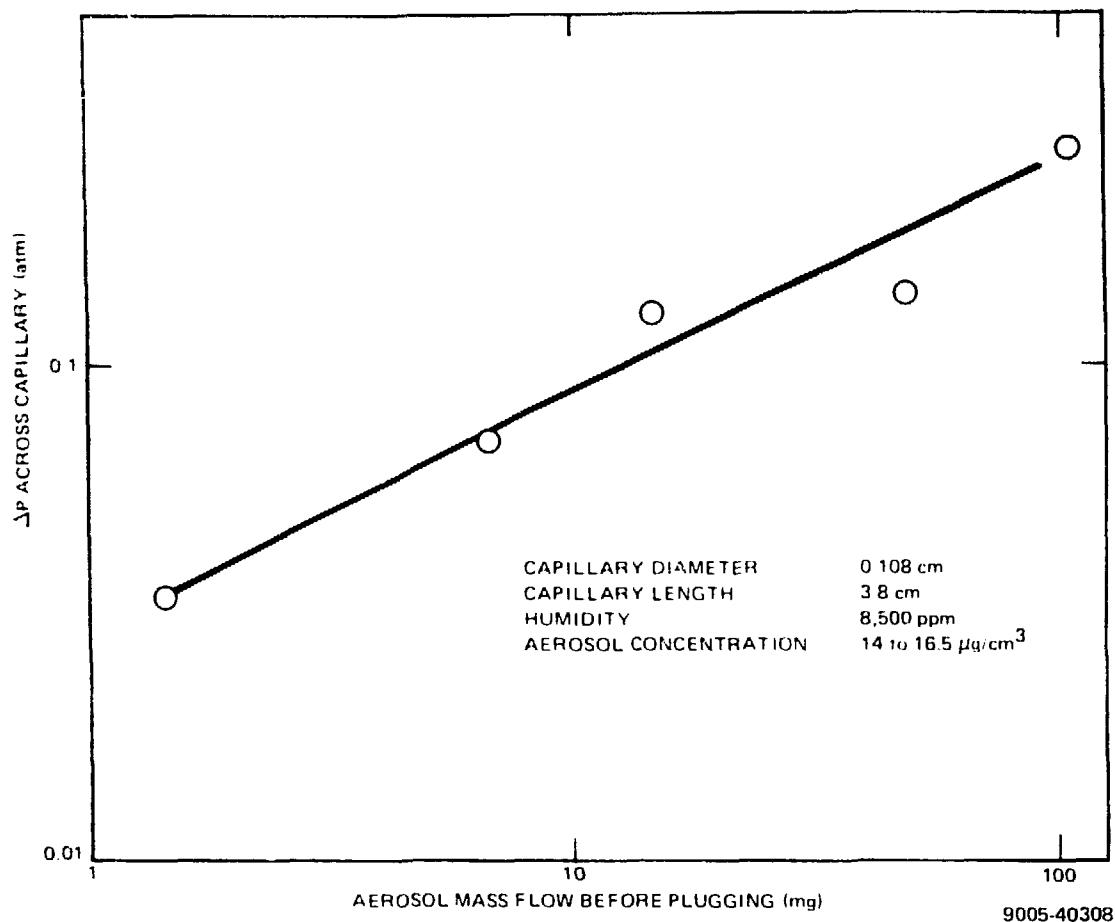


Figure 5. Pressure across capillary vs plugging mass.

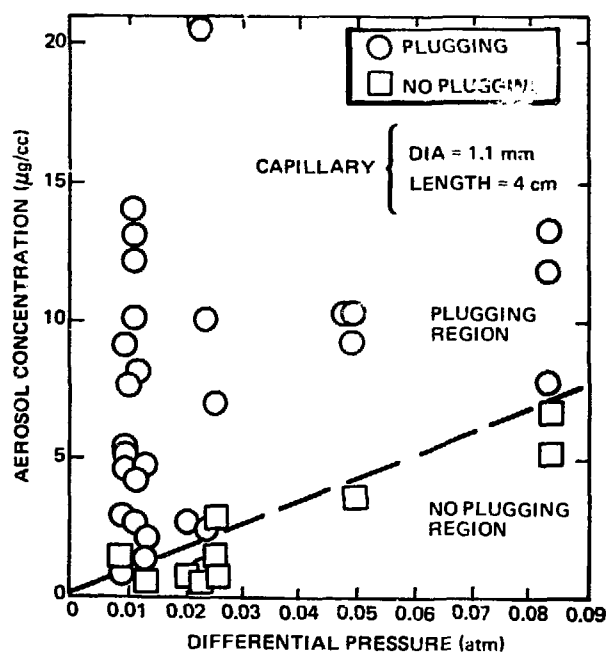


Figure 6. Conditions for Plugging.

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After grouping several tests together as in Figure 6, the data indicates that given a certain pressure differential, there is an aerosol concentration above which plugging occurs and below which no plugging occurs. The line of separation depends primarily on the diameter of the leak path although humidity also plays an important part.

In one set of experiments, tests were performed to document by photography the particulate buildup or deposition during the plugging sequence of capillaries. These tests were performed in a sodium oxide aerosol environment which varied from 0.5 to 3.5 g/m³ in concentration and approximately 1.8 to 2.5 μ m in size (AED 50). The pressure in the vessel was maintained at 0.068 atmosphere with the dew point ranging from -10°C to 5°C (~3500 ppm).

Figure 7 shows a photograph of entrance plugging where the particulates do not follow the air stream and impact at the entrance. When such a buildup of oxide occurs the plug acts as a filter even though some gas leakage through the capillary still exists.

A plug formed inside the capillary is shown in Figure 8. For this type of leakage, deposition occurs directly on the walls of the capillary.

Figure 9 is a photograph of a capillary exit in which plug creep occurred. As can be seen, a considerable mass of oxide may accumulate before plugging terminates the gas flow.

To illustrate the events that take place when a plug does not form, a strike plate was placed approximately 10 cm from the capillary exit. The results as shown in Figure 10, indicate that large agglomerated particulates were formed in the capillary and then expelled. These particulates are shown to be considerably larger than respirable particulates (i.e., >> μ m). Before passing through the median size was about 2.8 μ m (AED).

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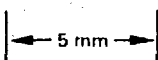
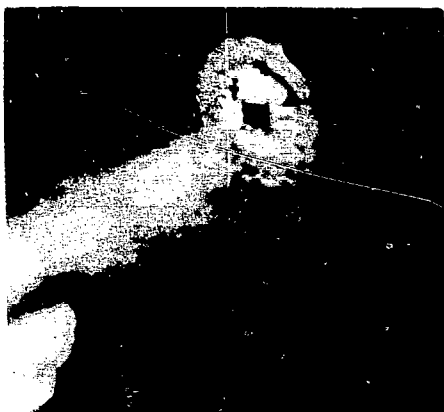


Figure 7. Entrance plugging.

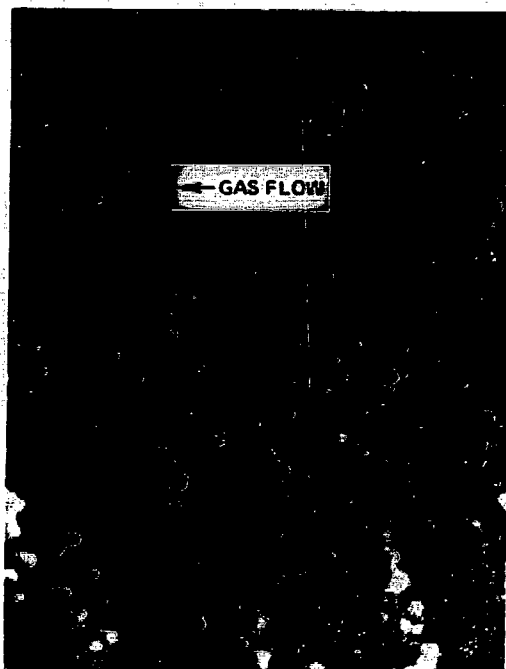
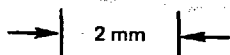


Figure 8. Inside wall of longitudinal section of capillary following plugging.



9005-40270



Figure 10. Impact zone exit
from plate impact.



Figure 9. Impact zone exit
from plate impact.

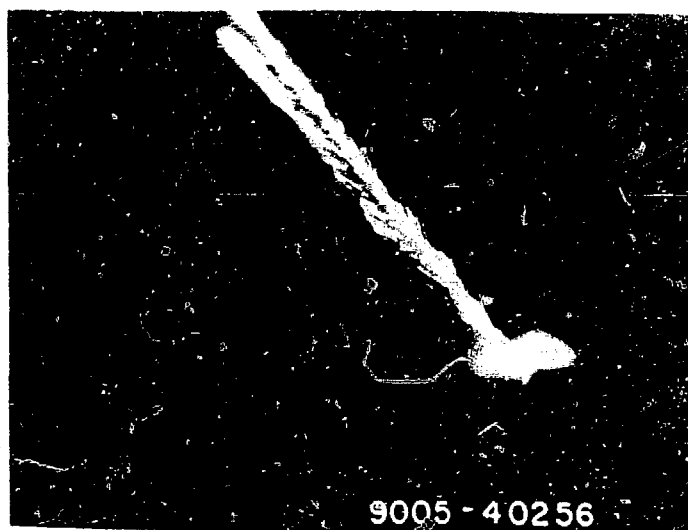


Figure 11. Dendritic column of
sodium oxide formed on
impact plate.

44

1. 2. 3. 4. 5. 6. 7. 8. 9. 10. 11. 12. 13. 14. 15. 16. 17. 18. 19. 20. 21. 22. 23. 24. 25. 26. 27. 28. 29. 30. 31. 32. 33. 34. 35. 36. 37. 38. 39. 40. 41. 42. 43. 44. 45. 46. 47. 48. 49. 50. 51. 52. 53. 54. 55. 56. 57. 58. 59. 60. 61. 62. 63. 64. 65. 66. 67. 68. 69. 70. 71. 72. 73. 74. 75. 76. 77. 78. 79. 80. 81. 82. 83. 84. 85. 86. 87. 88. 89. 90. 91. 92. 93. 94. 95. 96. 97. 98. 99. 100. 101. 102. 103. 104. 105. 106. 107. 108. 109. 110. 111. 112. 113. 114. 115. 116. 117. 118. 119. 120. 121. 122. 123. 124. 125. 126. 127. 128. 129. 130. 131. 132. 133. 134. 135. 136. 137. 138. 139. 140. 141. 142. 143. 144. 145. 146. 147. 148. 149. 150. 151. 152. 153. 154. 155. 156. 157. 158. 159. 160. 161. 162. 163. 164. 165. 166. 167. 168. 169. 170. 171. 172. 173. 174. 175. 176. 177. 178. 179. 180. 181. 182. 183. 184. 185. 186. 187. 188. 189. 190. 191. 192. 193. 194. 195. 196. 197. 198. 199. 200. 201. 202. 203. 204. 205. 206. 207. 208. 209. 210. 211. 212. 213. 214. 215. 216. 217. 218. 219. 220. 221. 222. 223. 224. 225. 226. 227. 228. 229. 230. 231. 232. 233. 234. 235. 236. 237. 238. 239. 240. 241. 242. 243. 244. 245. 246. 247. 248. 249. 250. 251. 252. 253. 254. 255. 256. 257. 258. 259. 260. 261. 262. 263. 264. 265. 266. 267. 268. 269. 270. 271. 272. 273. 274. 275. 276. 277. 278. 279. 280. 281. 282. 283. 284. 285. 286. 287. 288. 289. 290. 291. 292. 293. 294. 295. 296. 297. 298. 299. 300. 301. 302. 303. 304. 305. 306. 307. 308. 309. 310. 311. 312. 313. 314. 315. 316. 317. 318. 319. 320. 321. 322. 323. 324. 325. 326. 327. 328. 329. 330. 331. 332. 333. 334. 335. 336. 337. 338. 339. 340. 341. 342. 343. 344. 345. 346. 347. 348. 349. 350. 351. 352. 353. 354. 355. 356. 357. 358. 359. 360. 361. 362. 363. 364. 365. 366. 367. 368. 369. 370. 371. 372. 373. 374. 375. 376. 377. 378. 379. 380. 381. 382. 383. 384. 385. 386. 387. 388. 389. 390. 391. 392. 393. 394. 395. 396. 397. 398. 399. 400. 401. 402. 403. 404. 405. 406. 407. 408. 409. 410. 411. 412. 413. 414. 415. 416. 417. 418. 419. 420. 421. 422. 423. 424. 425. 426. 427. 428. 429. 430. 431. 432. 433. 434. 435. 436. 437. 438. 439. 440. 441. 442. 443. 444. 445. 446. 447. 448. 449. 450. 451. 452. 453. 454. 455. 456. 457. 458. 459. 460. 461. 462. 463. 464. 465. 466. 467. 468. 469. 470. 471. 472. 473. 474. 475. 476. 477. 478. 479. 480. 481. 482. 483. 484. 485. 486. 487. 488. 489. 490. 491. 492. 493. 494. 495. 496. 497. 498. 499. 500. 501. 502. 503. 504. 505. 506. 507. 508. 509. 510. 511. 512. 513. 514. 515. 516. 517. 518. 519. 520. 521. 522. 523. 524. 525. 526. 527. 528. 529. 530. 531. 532. 533. 534. 535. 536. 537. 538. 539. 540. 541. 542. 543. 544. 545. 546. 547. 548. 549. 550. 551. 552. 553. 554. 555. 556. 557. 558. 559. 560. 561. 562. 563. 564. 565. 566. 567. 568. 569. 570. 571. 572. 573. 574. 575. 576. 577. 578. 579. 580. 581. 582. 583. 584. 585. 586. 587. 588. 589. 590. 591. 592. 593. 594. 595. 596. 597. 598. 599. 600. 601. 602. 603. 604. 605. 606. 607. 608. 609. 610. 611. 612. 613. 614. 615. 616. 617. 618. 619. 620. 621. 622. 623. 624. 625. 626. 627. 628. 629. 630. 631. 632. 633. 634. 635. 636. 637. 638. 639. 640. 641. 642. 643. 644. 645. 646. 647. 648. 649. 650. 651. 652. 653. 654. 655. 656. 657. 658. 659. 660. 661. 662. 663. 664. 665. 666. 667. 668. 669. 670. 671. 672. 673. 674. 675. 676. 677. 678. 679. 680. 681. 682. 683. 684. 685. 686. 687. 688. 689. 690. 691. 692. 693. 694. 695. 696. 697. 698. 699. 700. 701. 702. 703. 704. 705. 706. 707. 708. 709. 710. 711. 712. 713. 714. 715. 716. 717. 718. 719. 720. 721. 722. 723. 724. 725. 726. 727. 728. 729. 730. 731. 732. 733. 734. 735. 736. 737. 738. 739. 740. 741. 742. 743. 744. 745. 746. 747. 748. 749. 750. 751. 752. 753. 754. 755. 756. 757. 758. 759. 760. 761. 762. 763. 764. 765. 766. 767. 768. 769. 770. 771. 772. 773. 774. 775. 776. 777. 778. 779. 780. 781. 782. 783. 784. 785. 786. 787. 788. 789. 790. 791. 792. 793. 794. 795. 796. 797. 798. 799. 800. 801. 802. 803. 804. 805. 806. 807. 808. 809. 810. 811. 812. 813. 814. 815. 816. 817. 818. 819. 820. 821. 822. 823. 824. 825. 826. 827. 828. 829. 830. 831. 832. 833. 834. 835. 836. 837. 838. 839. 840. 84

NUMBER 200 000

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DISCUSSION

SCHIKARSKI: Have you found any dependence of particle composition on the penetration or plugging behavior?

NELSON: There's a lot of dependence on the composition as well as moisture. If you have a lot of moisture in the reactor vessel you have very wet sodium hydride and essentially, you have a liquid going through the capillary rather than a dry aerosol. If you have a smooth capillary such as the ones we used, it's difficult to plug them with liquid, because it just keeps on going through. However, if you have a smooth capillary and an aerosol which is extremely dry, why you have a material which does not readily plug within the capillary but forms fluffy deposits on the side. It cannot stick to the side and keeps popping out. Composition does make a lot of difference.

JORDAN: Do you measure temperature distribution in the test capillary? Do you measure particle size entering and leaving the capillary?

NELSON: We assume the leak path is in the outer containment and for this location, temperature will not be much of a problem. Normally, we measure particle size entering and leaving the capillary.

JORDAN: Do you attempt to identify this behavior with filtration theory?

NELSON: Normally, the particle size that we are working with when we expose the capillaries is somewhere between 1 and 4.5 micrometers. We have used extremely low concentrations so that the size would not increase by agglomeration. We seldom get sizes less than one micrometer.

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SESSION X

TEST METHODS

Wednesday, August 9, 1978

CHAIRMEN: J. L. Kovach, J. W. Jacox

- THE NEW WASTE CALCINATING FACILITY PROCESS OFF-GAS FILTRATION SYSTEM AND REMOTE
IN-PLACE DOP TEST
H. H. Loo, R. R. Smith, G. E. Bingham,
T. T. Allan, D. E. Wilcox
- EVALUATION OF THE HEPA FILTER IN-PLACE TEST METHOD IN A CORROSIVE OFF-GAS ENVIRON-
MENT
L. P. Murphy, M. A. Wong, R. C. Girton
- IN SITU TESTING OF TANDEM HEPA FILTER INSTALLATIONS WITH A LASER SINGLE PARTICLE
SPECTROMETER SYSTEM
B. G. Schuster, D. J. Osetek
- MEASUREMENTS WITH SELF-IDENTIFYING AEROSOLS
T. Kyle, B. G. Schuster
- A METHOD FOR COMPARING AIR VELOCITIES BETWEEN BULK CARBON AND ASSOCIATED TEST
CANISTERS
H. C. Parish
- PERFORMANCE EVALUATION OF AN 18,000 CFM FILL-IN-PLACE ADSORBER UNIT
R. D. Porco
- EXPERIMENTAL DETERMINATION OF FISSION GAS ADSORPTION COEFFICIENTS
R. Lovell, D. W. Underhill
- A RADIOIODINE DETECTOR BASED ON LASER INDUCED FLUORESCENCE
A. P. Baronavski, J. R. McDonald
- AN INORGANIC RADIOIODINE ADSORBER WITH LOW NOBLE GAS RETENTION
C. Distenfeld, J. Klemish

Dup

THE NEW WASTE CALCINING FACILITY PROCESS OFF-GAS
FILTRATION SYSTEM AND REMOTE IN-PLACE DOP TEST

H. H. Loo, R. R. Smith, G. E. Bingham
Allied Chemical Corporation
Idaho Chemical Programs - Operations Office
Idaho Falls, Idaho

T. T. Allan, D. E. Wilcox
Flanders Filters, Inc.
Pacific and Western States Office
Burlingame, California

Abstract

The crabtrap, one component of a remotely-operable, experimental, in-place DOP test system for process off-gas HEPA filters being developed at Flanders Filters was tested with good results. The crabtrap is a device which is positioned downstream of a HEPA filter during in-place DOP testing to thoroughly mix and collect the air and DOP passing through the filter so the efficiency of the filter and the integrity of the supporting framework can be determined from outside the ventilating system. Concurrently, a test program was also initiated at the Remote Maintenance Development Facility located in the Idaho National Engineering Laboratory. The objective of the effort was to verify that a remotely operable and maintainable (by a single-arm manipulator) off-gas filtration system could be designed for use in the New Waste Calcining Facility being built at the Idaho Chemical Processing Plant. The capabilities demonstrated in the program included the remote replacement of filters, filter housing doors, seals, the filter support and elevating units, and remotely testing the filters using DOP aerosol within a space-limited plenum.

I. Introduction

The in-place dioctyl phthalate (DOP) test required by government regulations to determine the efficiency of High Efficiency Particulate Air (HEPA) filters is frequently incompatible with many other components in a ventilating system. Research and development has been performed at Flanders Filters, Inc. on various in-place test devices that could be incorporated in a filter housing in efforts to (a) reduce the space required to perform a successful in-place DOP test, (b) be able to accomplish the test (inject the DOP, operate the test equipment, etc.) from outside the plenum or filter housing, (c) independently test each filter in a multiple filter installation for efficiency, and (d) reduce the amount of DOP to be generated at a given time by testing the filters one at a time. Hopefully, the results of this program will have special significance in larger filter banks, installations where two or more filters in series must be tested, and in systems where the maintenance of the filters must be performed from a remote location.

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The specific objectives of the program were directed towards (a) devising a method of achieving uniform dispersion of the DOP aerosol in the airstream immediately ahead of each HEPA filter and its supporting framework without the industry recommended seven to ten duct diameters and without using bypass ducting, and (b) providing a downstream device that would mix, collect and permit sampling for filter efficiency a short distance from the filter.

In early tests, dispersion was attained by electrically polarizing the DOP particles as the DOP was introduced into the airstream immediately ahead of the HEPA filter. A sheet metal partition was installed upstream of and between contiguous filters to isolate the DOP inlet and the dispersion apparatus which were installed ahead of each filter. However, because of potential maintenance problems and the fire hazard associated with the electro-static method, this method of dispersing DOP was abandoned and an effort began to develop a mechanical solution. However, capability of isolating filters to permit individual testing has been retained.

A device called a crabtrap (because of its unusual shape and the way that early versions opened and closed) has been used from the beginning of the program to sample the airstream a short distance downstream of a filter. One crabtrap device is installed downstream of each filter so that individual filters can be tested. Readings taken at the crabtrap have been found comparable to samples taken ten or more duct diameters downstream of the filter.

II. Description

Crabtrap

The crabtrap is shaped like a funnel, except that the "spout" (sample box) has been recessed into the body of the funnel, and the trap is square rather than round (Fig. 1 and 2) giving it the appearance of a pyramid with its top cut off. The base of the pyramid is 24 in. by 24 in. to correspond in size to the face area of the filters commonly used in larger ventilating systems. At the top where the sample box is recessed, the opening is 6 in. by 6 in. About a third of the way up from the base, the pyramid is broken on three sides and hinged on the fourth to permit it to be opened after testing. When in the test position, air leaving a filter is forced through the crabtrap, with a great deal of turbulence and mixing due to the sharply sloping interior walls of the pyramid and the sidewalls of the sample box. Air that has been diverted through the crabtrap is then sampled for an efficiency reading at a point 13 in. beyond the box. When the test is completed, the crabtrap is swung to the side leaving a 17 in. by 17 in. opening for the passage of air.

When more than one filter in parallel is being tested, all crabtraps are closed simultaneously so that the air through the system remains balanced.

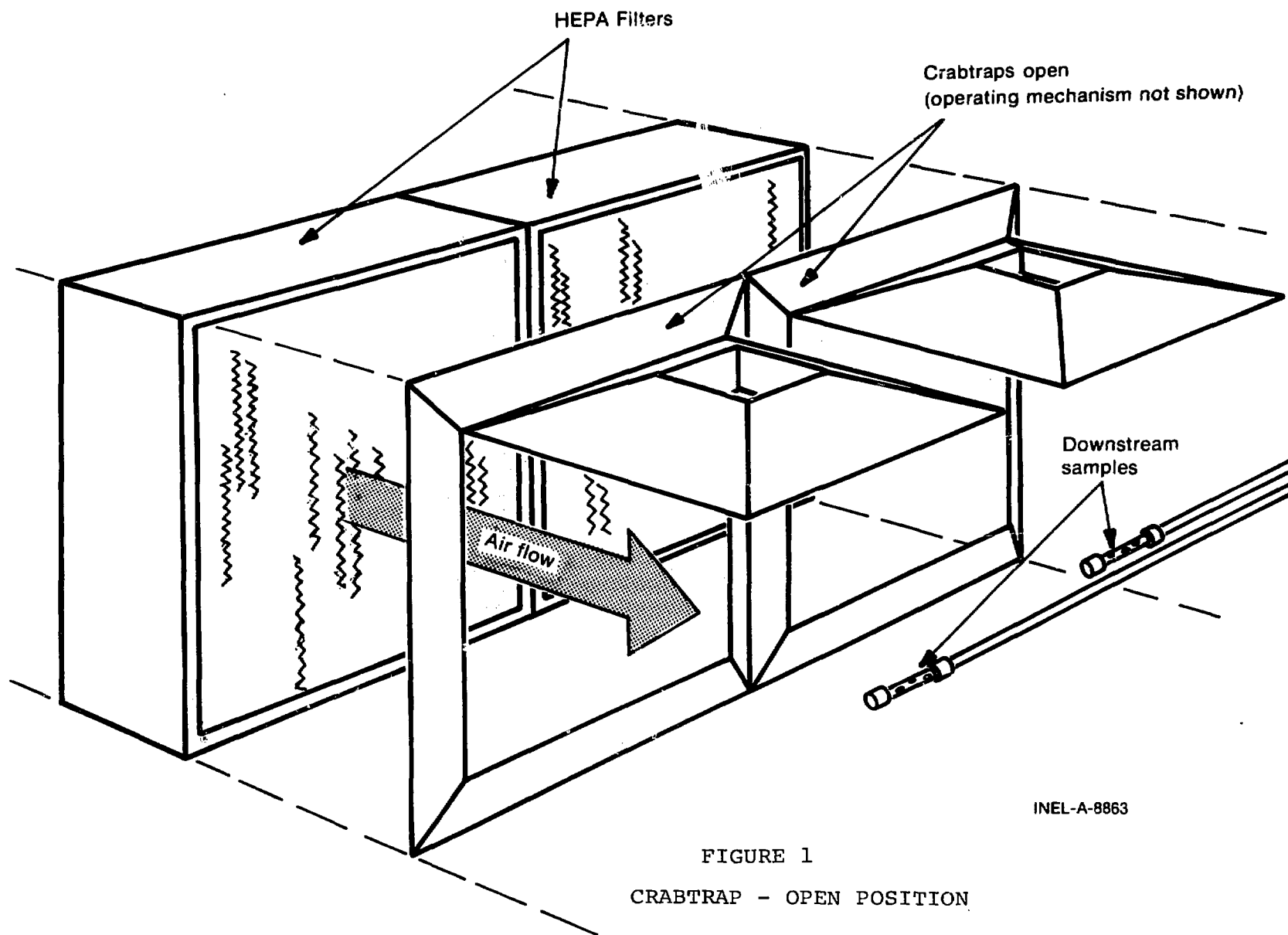


FIGURE 1
CRABTRAP - OPEN POSITION

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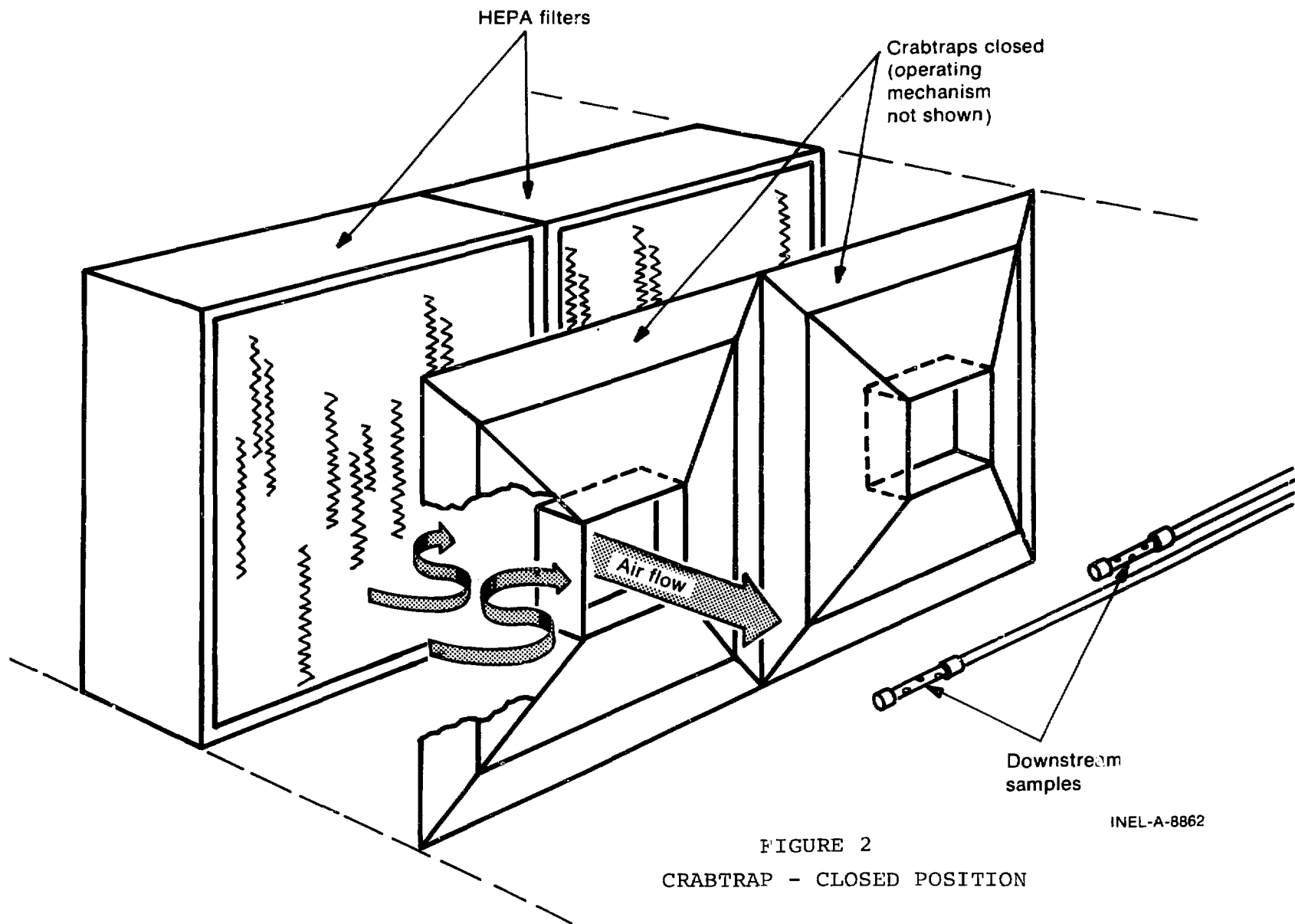


FIGURE 2
CRABTRAP - CLOSED POSITION

INEL-A-8862

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III. The New Waste Calcining Facility

Concurrent with Flanders Filters' remote DOP testing program, the New Waste Calcining Facility (NWCF) Project initiated a series of remote development programs. These programs were designed to confirm the remote replacement philosophy developed for high maintenance equipment within the NWCF. The specific objective of each program was to verify that all the remote operations designed for use in the NWCF could be accomplished. The remote design philosophy will contribute significantly to the control of problems such as lengthy facility downtime, costly decontamination efforts, and high personnel radiation exposures as compared with a nuclear facility with direct handling and replacement concepts.

As part of the remote development program, an off-gas filtration system incorporating the remote replacement philosophy was designed for use in the NWCF. To demonstrate the operability of the design, a full scale section of the remotely operable and maintainable filtration system was fabricated and tested at the Remote Maintenance Development Facility (RMDF) located at the Idaho National Engineering Laboratory (INEL). The design of this filtration system included positive positioning and sealing of the filters, remote filter replacement, remote in-place DOP filter testing, and remote replacement of selected filter plenum components which may require maintenance. To accomplish project objectives, the operability and compatibility of the filter system and the remote handling equipment was also developed and proven along with all the specific remote operating requirements.

As developed and designed, the NWCF process off-gas filtration system will contain four parallel filter plenums, each containing (in series) a prefilter plus two HEPA filters. The prefilter and HEPA filters will be 95% and 99.95% efficient, respectively, on 0.7 micron mean diameter DOP aerosols. Any three of the filter plenums are capable of accepting the total NWCF off-gas flow, thus permitting isolation of any single plenum for maintenance, filter replacement, or other remote operations. Overall, the system is designed to provide an off-gas decontamination factor for particulates of 8×10^4 . Figure 3 shows the final design of the NWCF off-gas filtration system.

IV. Remote Testing

To simulate actual NWCF operating conditions, all remote handling operations in this test program were performed using a wall-mounted electro-mechanical, single-arm manipulator behind a simulated shielded viewing window. At the completion of the remote test program, the following remote operations had been successfully demonstrated with the aid of an impact wrench, special filter cover and handling tools:

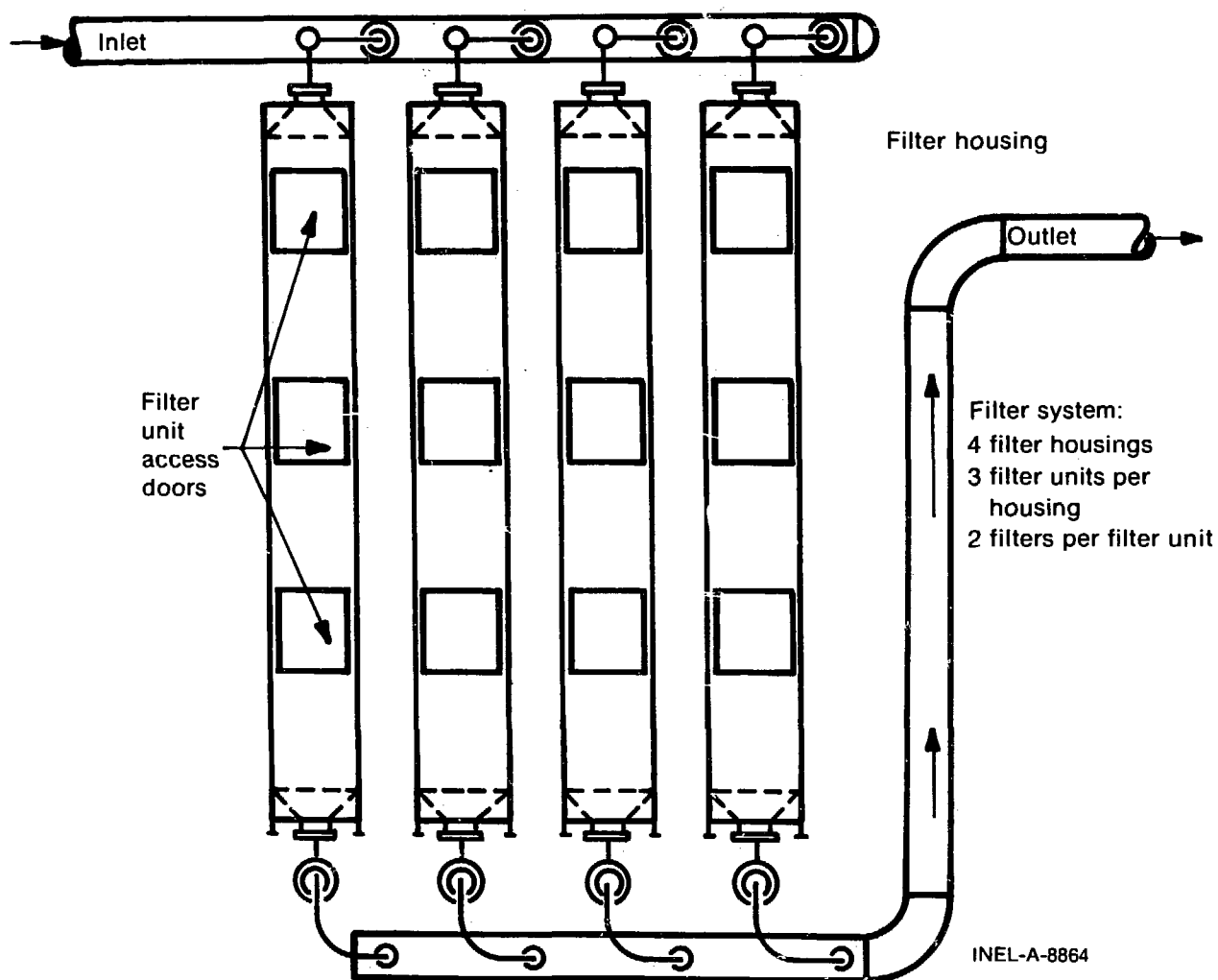


FIGURE 3
NWCF FILTER SYSTEM

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1. disengaging the filter housing door bolts;
2. lowering and aligning the filter housing door;
3. operating the filter elevator mechanism for lowering or raising (sealing) the filters;
4. removing and replacing the filters;
5. transporting the filters from the filter cell;
6. removing the filter housing door for seal replacement;
7. removing the elevator assembly for repair.

Figures 4 through 9 show the auxiliary handling tools used and the remote operations verified in the test program.

V. DOP Testing

The NWCF design criteria requires that after replacement, each filter be tested in-place using 0.7 micron mean DOP aerosol, thus verifying that the filter will meet federal regulations. Unless such a test is conducted, the remote filter replacement procedure does not prove that the filter is in position and sealed, or that the filter has remained undamaged during replacement.

Due to the limited distance available between each filter unit (approximately 4 ft) in the NWCF filter plenums, conventional in-place DOP test methods (requiring approximately 10 duct diameters upstream for DOP injection and 15 duct diameters downstream prior to sampling) could not be utilized. Thus, another test method had to be developed to verify the integrity of the newly replaced filters. After several design iterations, a non-conventional DOP test system was finalized and tested. Two major problems, good upstream DOP distribution and good representative downstream samples, were finally solved through the use of a conical disperser, a crabtrap, and a multiple port sample line. With the aid of these special devices, the final DOP test system gives results comparable with conventional test methods.

The basic in-place DOP test set-up is illustrated in Figure 10. The conical disperser, crabtrap, and multiple port sample line are shown in Figures 11 and 12. In the test program, DOP aerosol was injected with approximately 200 scfm of clean air through the 5 in. DOP injection line and dispersed by the conical disperser upstream of the filter. DOP samples were collected upstream (10 in. in front of the filter face) at 12 different points on the same plane to determine the disperser's distribution characteristics. Samples were also collected downstream of the crabtrap and compared with samples collected by conventional methods.

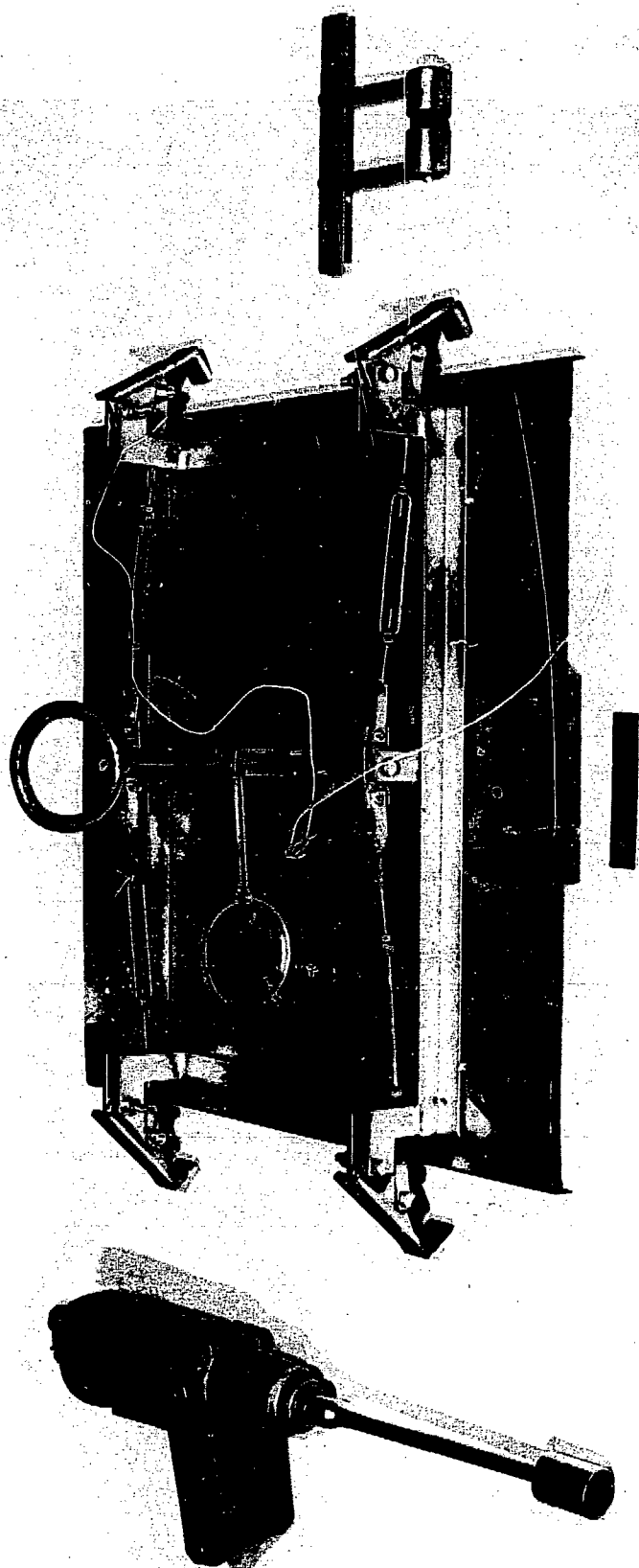


FIGURE 4
AUXILIARY TOOLS

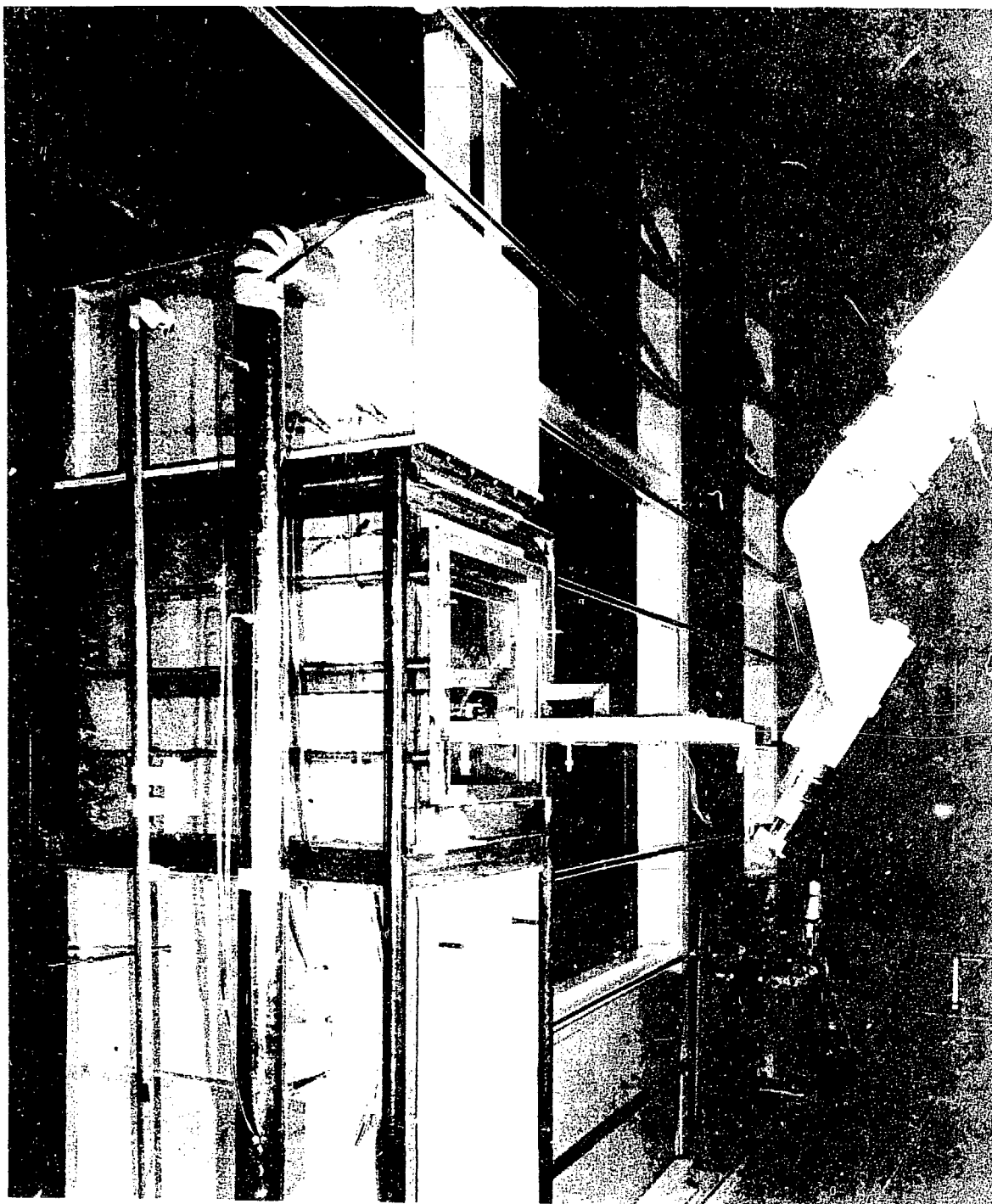
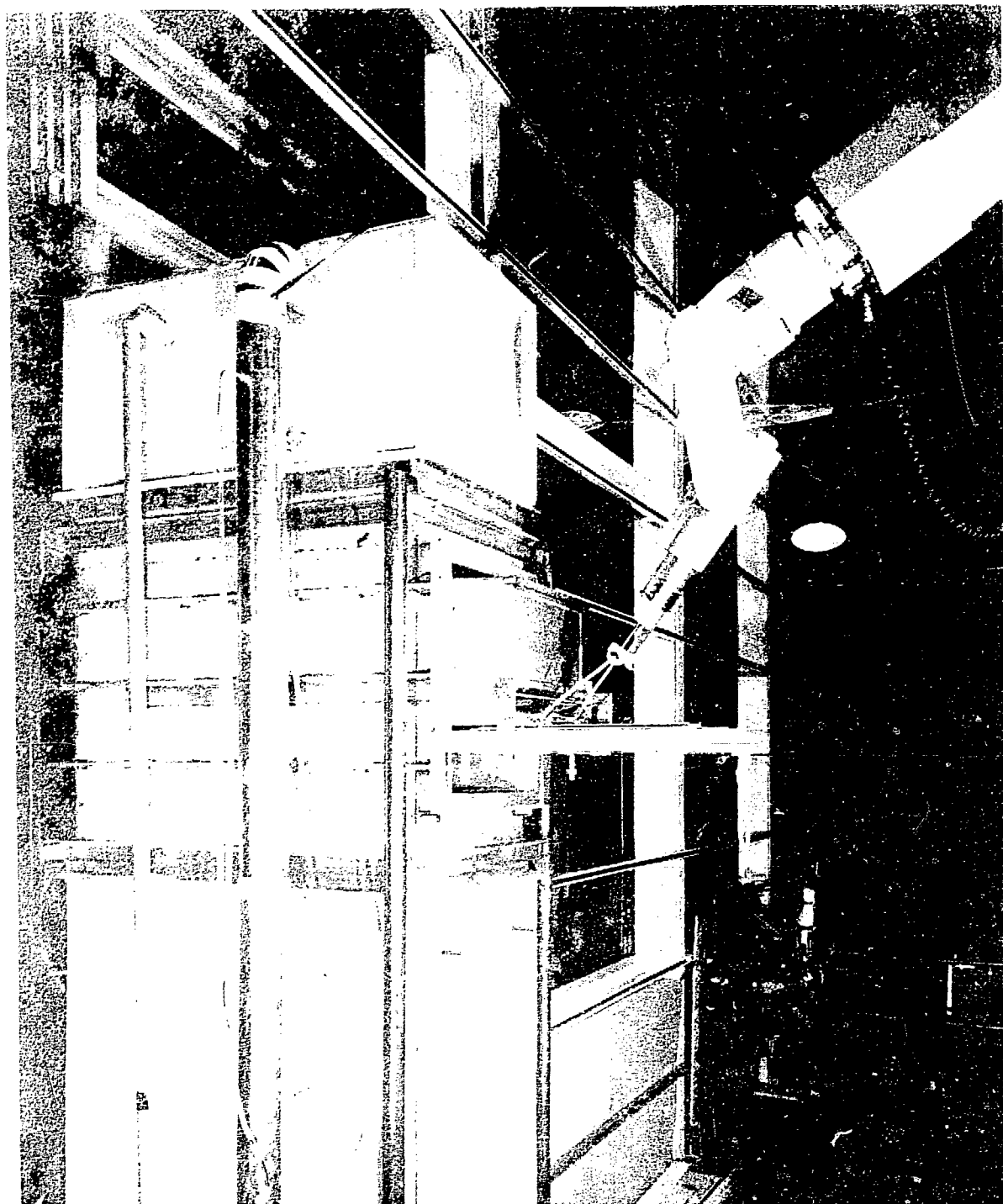


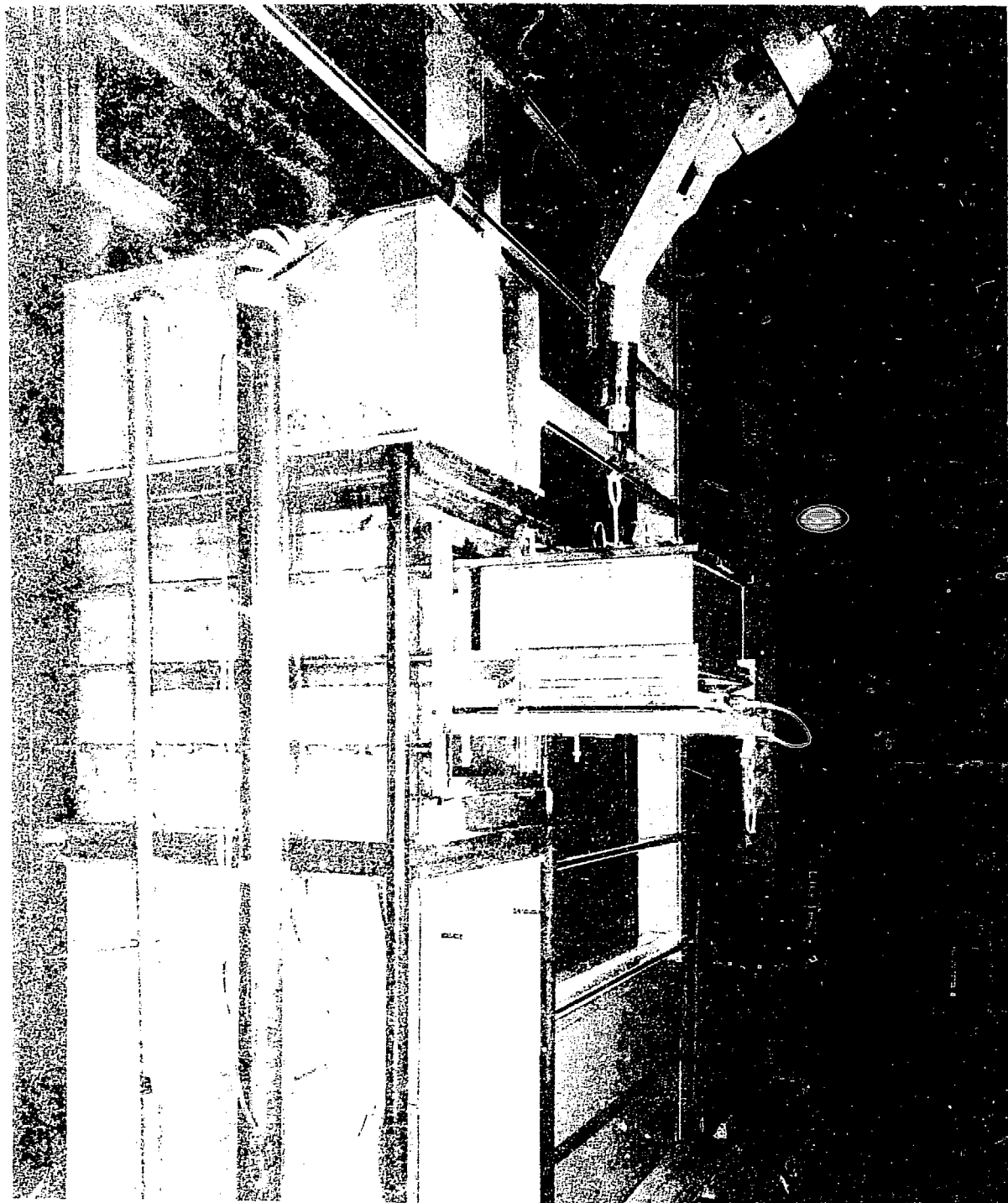
FIGURE 1
LOADING AND ALIGNING FILTER ELEMENTS

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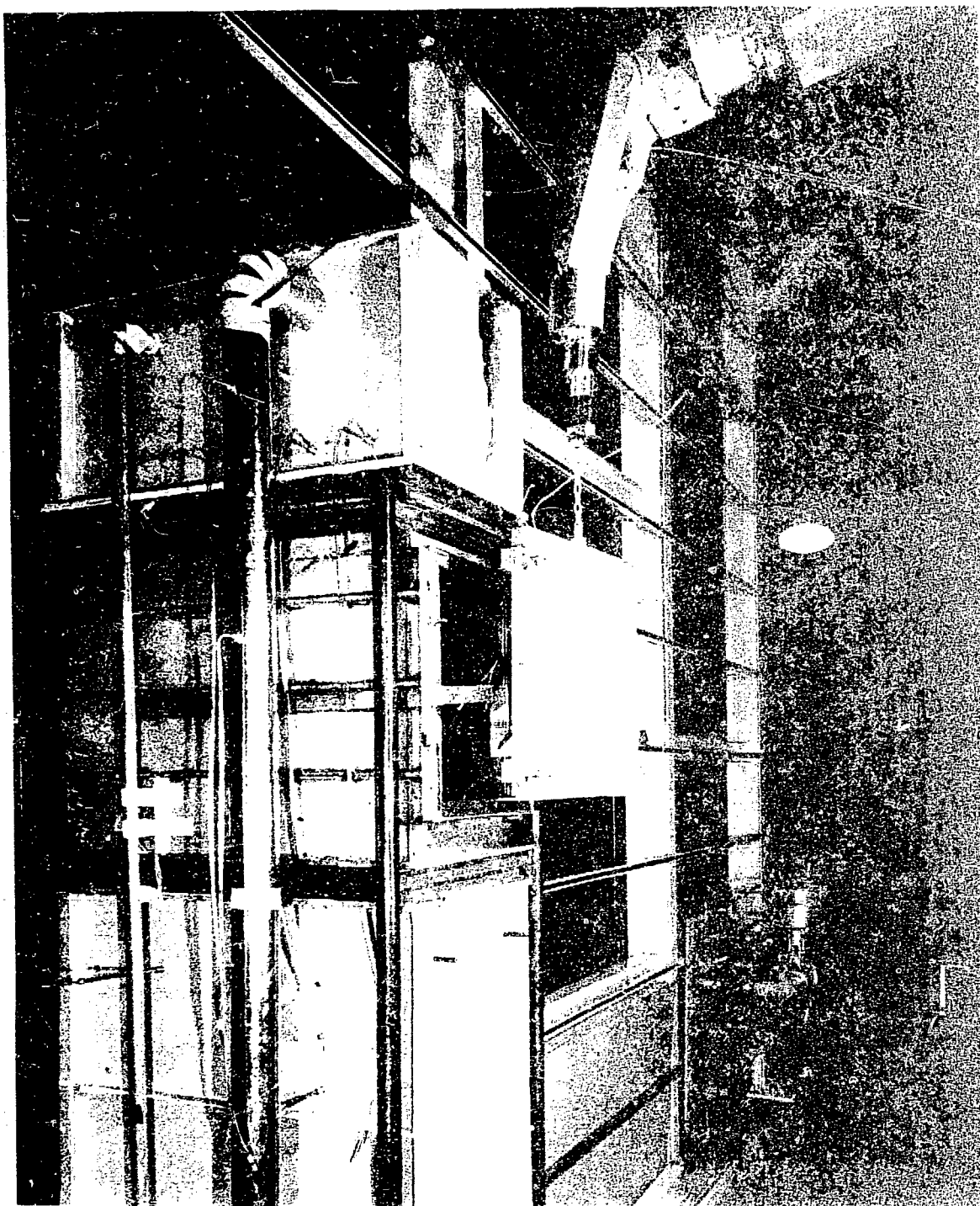
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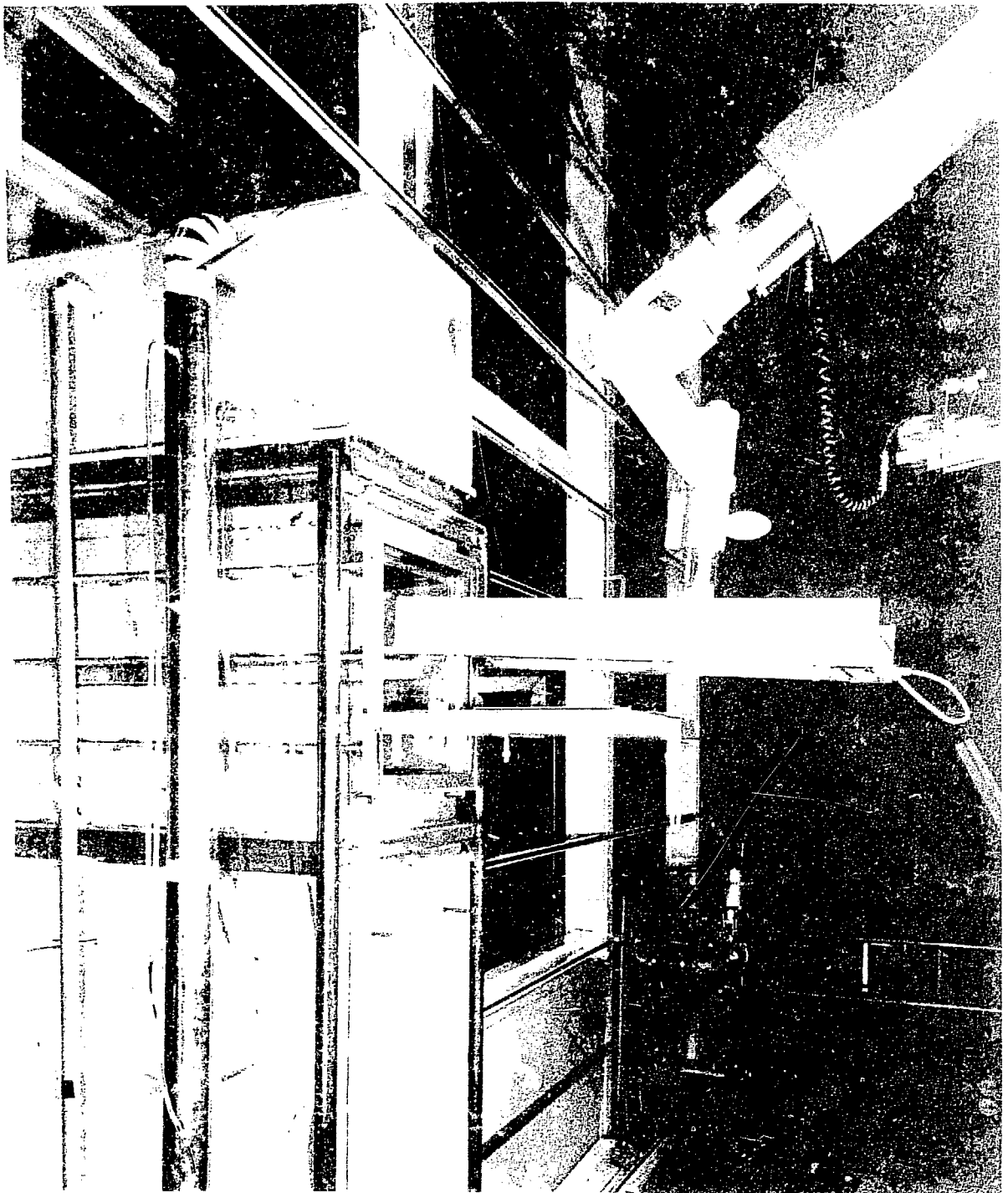
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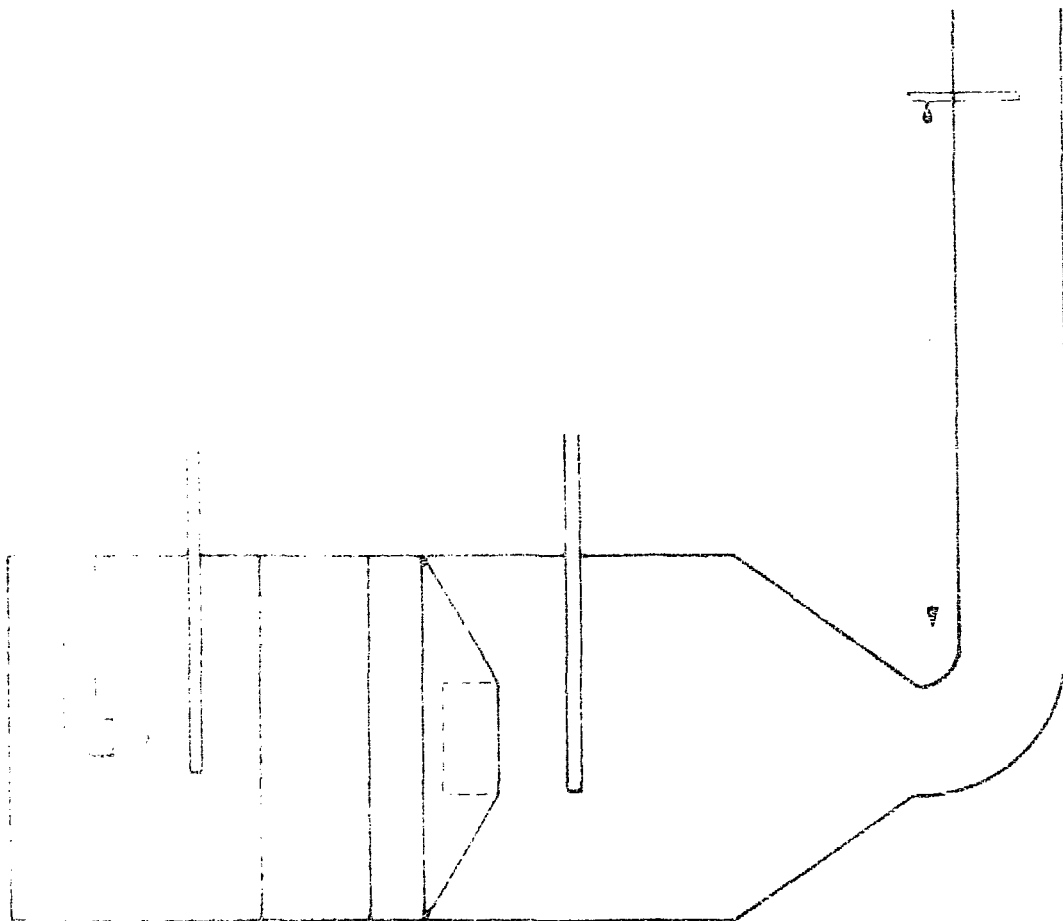
1981 DOE NUCLEAR AIR CLEANING CONFERENCE

1981 DOE NUCLEAR AIR CLEANING CONFERENCE





1961-1962 FARM AND LEANING CONFERENCE



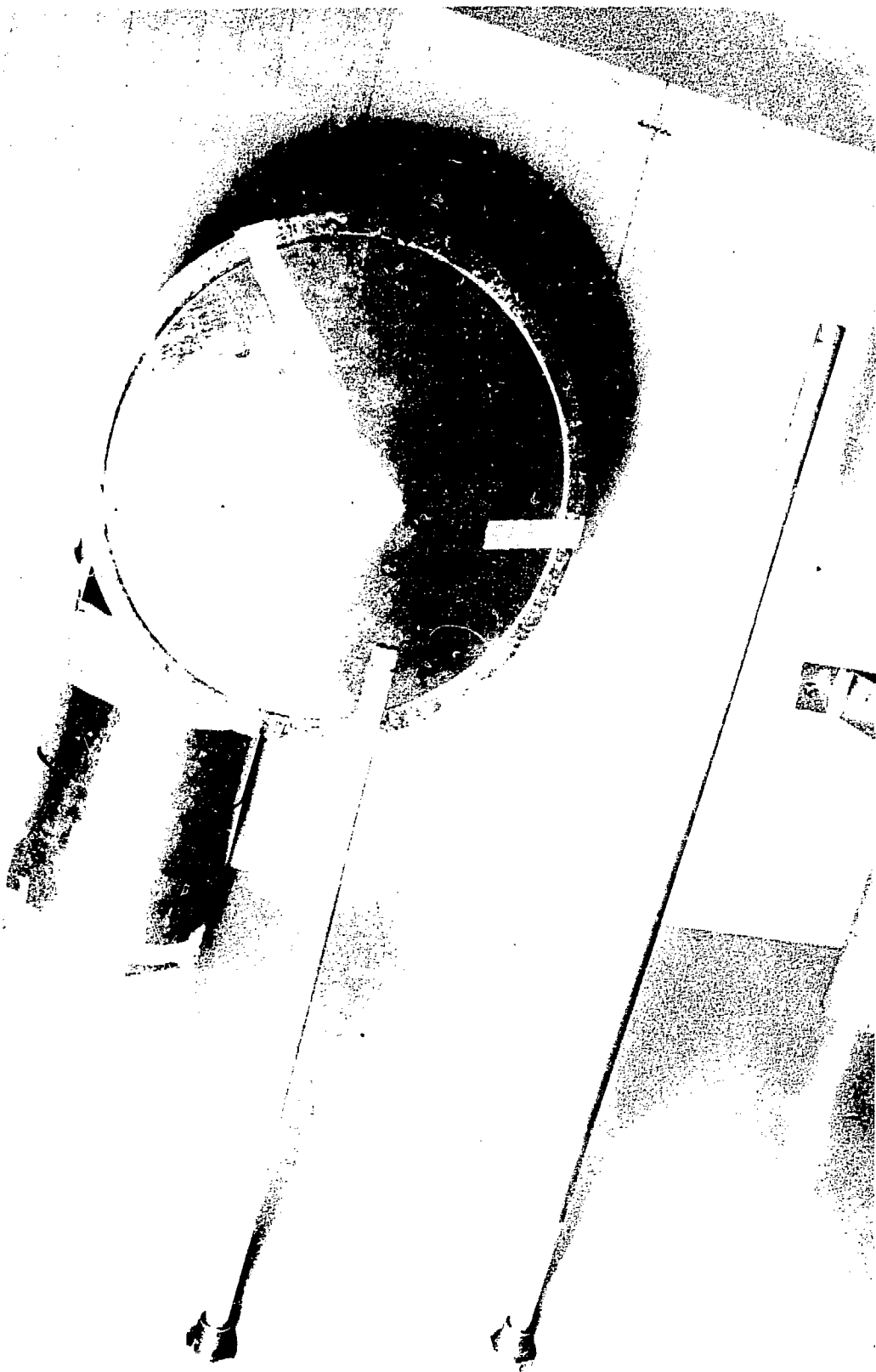


FIGURE 11
CONICAL DISPERSER

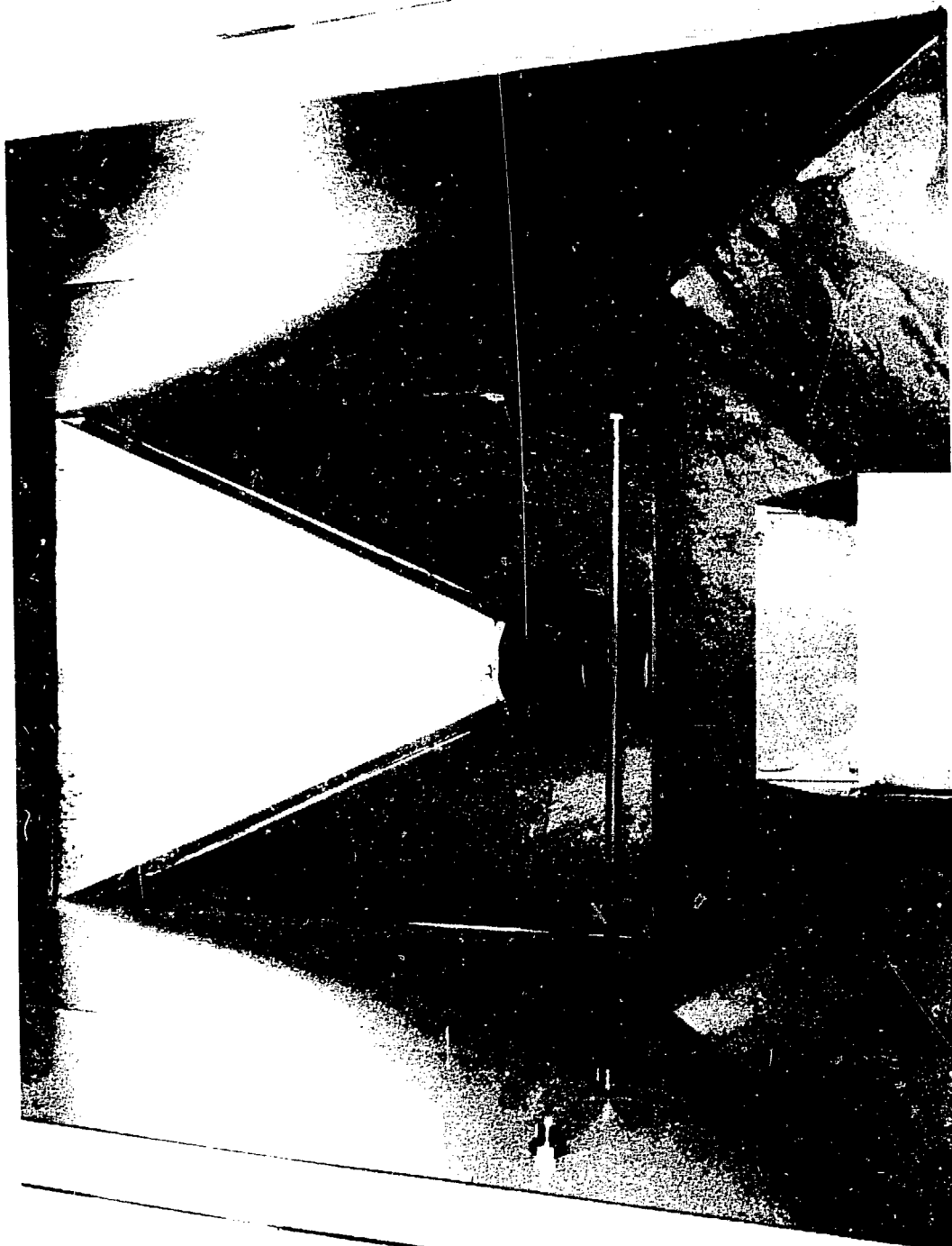


FIGURE 12
DOWNSTREAM SAMPLE LINE AND CRABTRAP-PARTIALLY OPEN

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The sample data collected in this test program were in close agreement with sample data collected by conventional DOP test methods. In addition, typical sample readings collected upstream of the filter were within 1% of one another. The DOP sample collected downstream of the crabtrap was within 0.02% of the sample collected at the conventional DOP sample line which was more than 15 duct diameters downstream of the filter. These results indicate that the conical disperser has an almost perfect DOP distribution characteristic and that the crabtrap and multiple port sample probe will determine filter efficiency to within 0.02% of conventional sample methods.

Based on these preliminary test results, the final design of the in-place DOP test system to be used in the NWCF will essentially be identical to the test arrangement. However, the filter plenum manufacturer will be required to perform a final acceptance test to prove that the DOP test system to be used in the process is workable. Preliminary results, on a similar test set-up, (submitted by the plenum manufacturer) is given in Table I. The plenum manufacturer's set-up is identical to the one tested at the RMDF, other than the crabtrap was turned around.

Table I. In-place DOP test results

| <u>Filters
Tested</u> | <u>Tested Air
Flow (acfm)</u> | <u>DOP
Upstream</u> | <u>Downstream
Samples
(crabtrap)</u> | <u>Downstream
Samples
15 duct Ø</u> |
|---------------------------|-----------------------------------|-------------------------|--|---|
| 1A | 193 | 100% | .015 | .015 |
| 1B | 223 | | .008 | .015 |
| 2A | 207 | | .012 | .012 |
| 2B | 204 | | .012 | .018 |
| 3A | 209 | | .070 | .058 |
| 3B | 209 | | .030 | .039 |
| 4A | 200 | | .017 | .019 |
| 4B | 193 | | .022 | .026 |
| 5A | 199 | | .025 | .029 |
| 5B | 186 | | .070 | .038 |
| 6A | 185 | | .022 | .024 |
| 6B | 219 | | .024 | .024 |
| 7A | 203 | | .140 | .130 |
| 7B | 212 | | .120 | .065 |
| 8A | 217 | | .065 | .065 |
| 8B | 206 | | .080 | .039 |
| 9A | 220 | | .055 | .055 |
| 9B | 221 | | .120 | .055 |
| 10A | 188 | | .027 | .028 |
| 10B | 195 | | .140 | .110 |

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VI. Summary

The test program conducted at the INEL's RMDF has proven to be extremely valuable in insuring a workable system prior to finalizing an equipment design. This program conducted at the RMDF has demonstrated that remote filter replacement, and filter housing preventive maintenance, and remote in-place DOP testing can be accomplished within a compact (space-limited) filter plenum. Development is continuing on other mechanical methods of introducing and dispersing the DOP ahead of the filters. One promising possibility of attaining dispersion is turning the crabtrap around so that the top of the pyramid points upstream and the sample box is recessed towards the filter. The DOP, introduced upstream of the reversed crabtrap, is forced to mix with the air being diverted through the sample box and is dispersed as it leaves the sample box prior to entering the filter. Further study is necessary in this area. The reversed crabtrap may also be used for sampling.

Presently, space is at a premium because of ever-increasing costs, yet ventilation systems are required to have an ever-increasing number of components in their make-up such as prefilters, entrainment separators, carbon adsorbers and HEPA's in series. Compact filter trains are less costly to build and easier to maintain, but are not always compatible with the requirements for an acceptable in-place DOP test. Also, at present, "hands on" maintenance of filters is under careful scrutiny for safety reasons. For the above reasons, remote testing of filters is certain to have increasing importance in years to come. Finally, the development of a workable in-place test requiring a minimum of space wherein individual filters can be tested is more stringent than challenging an entire filter bank. That is, when an entire bank of filters is challenged with DOP where only one filter might be leaking, the leak is diluted by the great volume of clean air passing through the rest of the filters (dilution effect), and the portion of the leaking filter that does not leak, thus perchance masking the leak. Testing filters individually will limit this effect.

DISCUSSION

FREEMAN: What costs are we looking at for this automated feature?

LOO: The total cost for fabricating the four filter housings to be used in the New Waste Calcinating Facility is \$435,000. However, I would like to point out that these filter housings are designed and fabricated in accordance with the ASME code, Section VIII, Div. 1 to a design pressure of 10.0 psig positive and 8.5 psig vacuum, thus increasing the cost somewhat.

FREEMAN: What is the air volume?

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LOO: Each filter housing contains two 24 x 24 x 11-inch filters. If you use the separatorless-type filters, there will be about 3,000 cfm of air through each filter housing.

T. T. ALLAN: I would like to comment further on Mr. Sam Freeman's question regarding the cost of the four housings because the response, \$435,000, was not adequately qualified. The cost of all the internal components of the assembly (twelve sets in all) to operate, test and maintain the 24 HEPA filters and prefilters was 20% of the total cost and that included several years of development cost. The housings themselves, or vessels as they should have been called, are each 20 ft. high, 4 ft. deep, and 2 ft. wide and were designed to withstand operating pressures of ± 7 psi. They are to be constructed of 5/16 in. 304L stainless steel contained in reinforcing angles with 2 in. of insulation and a 304L stainless skin. The vessels were 80% of the cost.

If similar, remotely operable units were to be constructed of heavy gauge stainless without the operating pressures that were required for these units, they would cost 15-20% of the figure given by Henry Loo.

Finally, it should be pointed out that in the existing calciner hot cell, as in similar hot cells in other facilities, the procedure is to dispose of the entire housing or vessel with each filter change at a cost of several thousand dollars per housing. The ability to change just the filter remotely will be a considerable savings in maintenance costs on the NWCF.

WARNKE: Since these filters are remotely-operable and remotely maintained, have you done any time and motion studies with regard to remote change-out and in-place DOP testing? I would be interested in the time required for filter change-out, assuming the filter is already inside the cell and prepared for changeout.

LOO: Assuming the filter is completely prepared for change-out, the operation takes about 20 minutes per filter.

WARNKE: Also, how do you transfer the filter into the cell and what time frame are we looking at for this operation?

LOO: We have designed a parking area separation between the filter housing and the outside building corridor. We transfer the unused filters into this buffer zone and then into the hot filter area. Since the facility is not yet constructed, we have no exact idea of the time required.

WARNKE: What time frame are you looking at for DOP testing each filter bank? Again, assuming all DOP generating and testing equipment is ready to go and all that is required is the actual testing?

LOO: Assuming all equipment is ready, the DOP testing would take about 15 minutes per filter.

VAN BRUNT: Can you explain the much higher penetration of filter 7B and 9B? They appear to be much worse than any of the others.

LOO: The tests were conducted on two consecutive days, with tests 1-5 on the first, and 6-10 on the second. The much greater deviation noted on the second day is probably attributable to some inadvertent change in the set-up.

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W up

EVALUATION OF THE HEPA FILTER IN-PLACE TEST* METHOD IN A CORROSIVE OFF-GAS ENVIRONMENT

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Abstract

Experiments were performed to determine if the combined effects of temperature, humidity and oxides of nitrogen (NO_x) hinder the in-place testing of high-efficiency particulate air (HEPA) filters used for cleaning the off-gas from a nuclear waste solidification facility. The laboratory system that was designed to simulate the process off-gas contained two HEPA filters in series with sample ports before each filter and after the filter bank. The system also included a reaction bomb for partial conversion of NO to NO_2 . Instrumentation measured stream flow, humidity, NO_x concentration, and temperature. Comparison measurements of the DOP concentrations were made by a forward light-scattering photometer and a single particle intra-cavity laser particle spectrometer. Experimental conditions could be varied, but maximum system capabilities were 95% relative humidity, 90°C , and 10,000 ppm of NO_x .

A 2^3 factorial experimental design was used for the test program. This design determined the main effects of each factor plus the interactions of the factors in combination.

The results indicated that water vapor and NO_x interfere with the conventional photometer measurements. Suggested modifications that include a unique sample dryer are described to correct the interferences. The laser particle spectrometer appears to be an acceptable instrument for measurements under adverse off-gas conditions.

I. Introduction

A nuclear waste solidification facility similar to the Waste Calcination Facility (WCF) at the Idaho Chemical Processing Plant (ICPP) produces an off-gas stream that has characteristics unique to the nuclear industry. The calcination process, the conversion of radioactive liquid wastes to solids, emits gases that are cleaned by cyclones, a scrubber, silica gel adsorbers, and HEPA filters (Figure 1). At the filters, the stream has a temperature of 90°C , a relative humidity of 95%, and an oxides of nitrogen concentration of 10,000 ppm. The filters are changed-out when radiation levels exceed a

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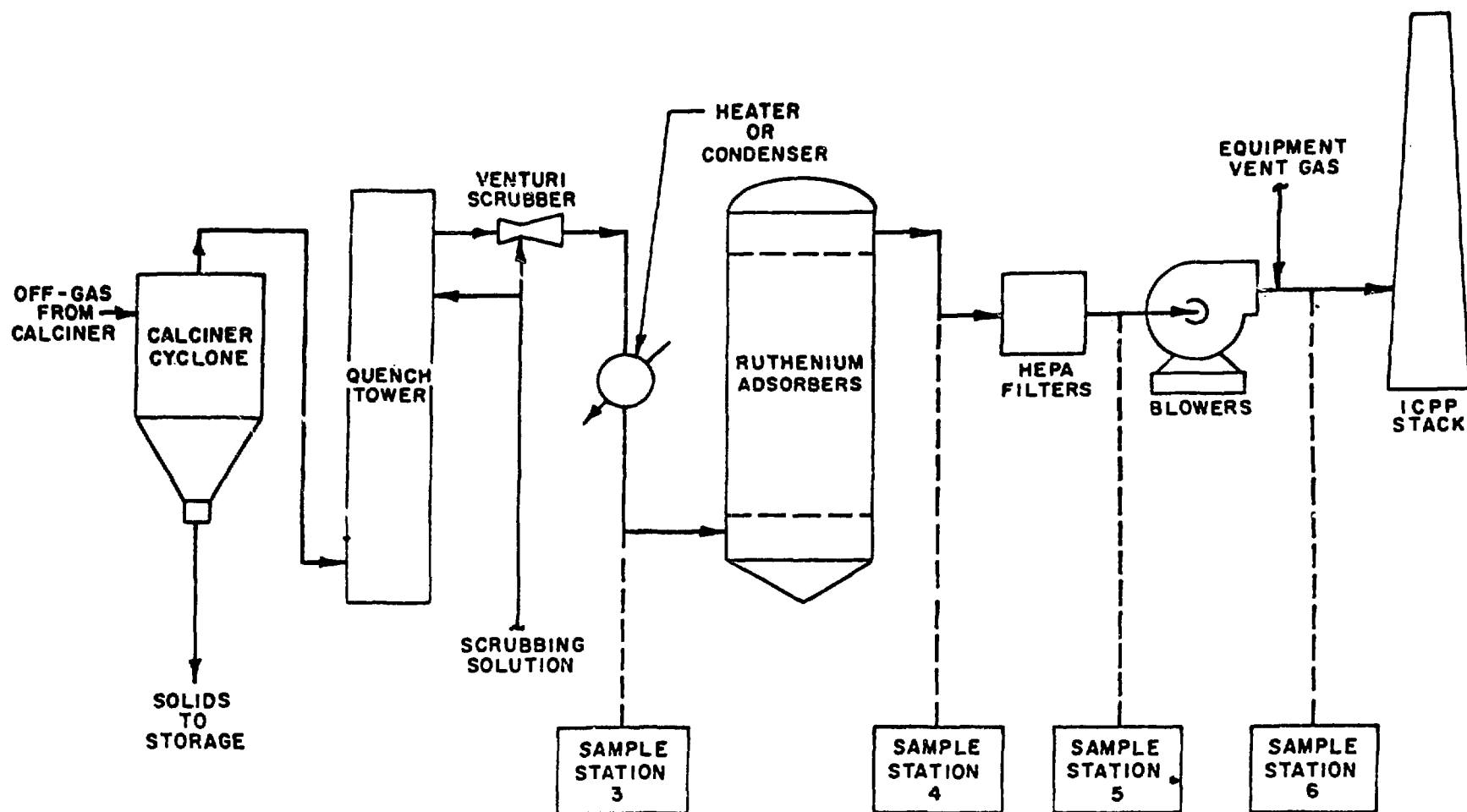


Figure 1. WCF OFF-GAS DECONTAMINATION SYSTEM

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safe operating limit, when pressure drop is high, or when a filter breach is indicated. A breach can be detected by the continuous monitoring of radiation in the off-gas stream after the filters or by a "fail-test" of the DOP in-place test method.¹ However, the DOP test method has provided erratic and unreliable results and the need for filter changeouts is usually determined by the monitor.

The HEPA system in the WCF consists of three parallel banks of filters; each bank has two HEPAs in series. Each filter bank can be tested separately by closing the air flow valves. A sample port does not exist between the filters.

The objective of this HEPA filter testing program is to evaluate the combined effects of temperature, humidity, and oxides of nitrogen on the standard DOP test method, and to determine if the laser is an acceptable instrument for measuring filter efficiencies in series in this type of environment. The conventional forward light-scattering photometer (Air Techniques Model TDA-2DN with .01% scale) and a laser particle spectrometer (Particle Measuring Systems Model ASAS-300) are used as the aerosol measuring instruments. Because of the highly radioactive environment of the WCF off-gas, a laboratory experimental system was constructed to simulate the off-gas conditions excluding radioactivity.

Each of the three parallel HEPA filter banks in the WCF off-gas system can be opened and closed for testing of individual banks. Since the filters in each bank cannot be tested separately, possible test errors can occur due to agglomeration and evaporation between filters. Light-scattering photometers have an effective dynamic range of about 10^4 and, therefore, are not acceptable instruments for a two-stage system.² In the simulated system, the photometer and laser spectrometer are used as comparison instruments for testing of the single HEPA filter and the laser is used for testing the two HEPA filters in tandem. The laser is capable of measuring protection factors of 10^8 , and it has the additional advantage of measuring various particle sizes in the 0.15 to 3.0 μm range. Protection factors as a function of particle size can be obtained.

The work that is reported in this paper is a part of the total program to evaluate in-place testing of HEPA filters in a corrosive environment. Work to date has involved the DOP test method. Future work will include comparison tests with the NaCl and uranine methods.

II. Experimental Procedure

The DOP studies were conducted with two 20-cm x 20-cm x 15-cm (8-in. x 8-in. x 5-7/8-in.) HEPA filters in tandem. Measurements of the DOP concentrations are taken upstream of both filters, between the filters, and after the final filter.

The apparatus shown in Figure 2 was used to conduct all testing. The system flow of 25 scfm was provided by a lubricated rotary vane vacuum pump. With a pressure drop of 5 inches of water, as in our

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system, this pump has a flow capacity of 44 scfm. The inlet air is filtered through a 30-cm x 30-cm x 15-cm (12-in. x 12-in. x 5-7/8-in.) HEPA filter. The flow is measured after the inlet filter with a linear mass flow transducer.

Up to 10,000 parts per million (ppm) concentration of NO_x can be obtained downstream of the flow transducer by mixing measured amounts of NO and O_2 in a 7L reaction bomb. The gases are supplied from cylinders and their flow into the reaction bomb is controlled by mass flow controllers. The NO_x concentration is measured by a chemiluminescent analyzer. A blender is used to dilute the NO_x to a range within the measurement capabilities of the analyzer. Measurements of both NO and NO_2 can be obtained with the analyzer. A 50% NO - 50% NO_2 mixture was maintained during the tests.

Steam is injected into the system to furnish the moisture requirements of 50 to 95% relative humidity. The steam is supplied by an electric boiler. The moisture content can be controlled by boiler pressure and flow control valves. Moisture measurements are made by a dew point hygrometer. The operating temperature can be controlled to 0.5°C accuracy at any temperature between 66 and 93°C. The temperature is monitored by thermocouples at selected points along the system. Dioctylphthalate (DOP) is added downstream from the steam. The average DOP aerosol concentration was $102 \mu\text{g/L} \pm 5 \mu\text{g}$. Concentrations were determined by collection of the aerosol on a filter and by the photometer. A by-pass line in the system allows the filters to be avoided during startup and preliminary system checkouts. Another linear mass flow transducer in the by-pass line measures total flow. The system contains four sampling points. They are located before the by-pass, before Filter 1, and after Filter 2. All four points are connected to the light-scattering photometer or laser particle analyzer by a heat-traced and insulated line.

When conducting tests at dew points above 60°C, a drying tube was used to lower the sample dew point without removing aerosol. A sample from any point can be directed through the drying tube. The device operates by a process called permeation distillation.³ In this procedure, a wet gas flows continuously into a bundle of tubes made of silicone polymer which is permeable to water vapor. Continuous drying is accomplished when the water vapor diffuses through the membrane and is swept by the dry gas flow in the reverse direction. The driving force of the process is the difference in relative humidities of the two streams. The dryer is designed in a shell and tube configuration, as shown in Figure 3. The wet sample passes through the tube side of the dryer and exits at the end of the bundle. The purge gas enters the unit on the shell side of the feed gas outlet and flows countercurrent to the wet feed gas. Drying efficiency and capacity are based on the dryer geometry (length and diameter) and the ratio of the purge-to-feed gas. The optimum purge-to-feed ratio is 1.5 to 1.

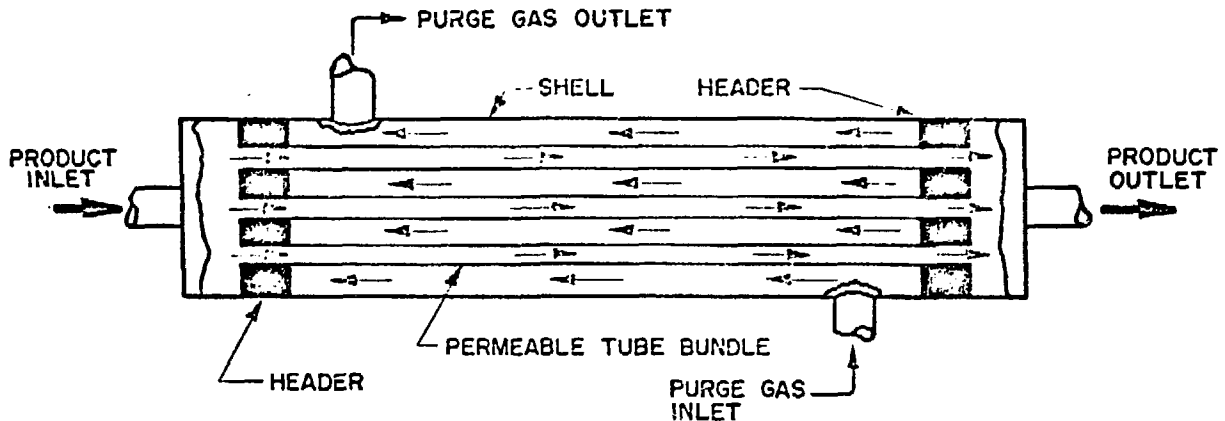


Figure 3. PERMEATION DISTILLATION DRYER

The HEPA test filters are sealed to the frame by a rubber base adhesive. These filters and the WCF process off-gas filters have identical specifications. Pressure drop is measured across each filter. After the test filters, the water is removed from the system by a condenser and a cryogenic probe. The dry stream then passes through the pump and is exhausted to the building ventilation system.

III. Experimental Results and Discussion

Baseline Measurements with Photometer and Laser

Preliminary tests were performed to establish a baseline for photometer and laser measurements of DOP in dry air. The tests were performed at a flow of 25 scfm and temperatures of 66 and 93°C. Each test period was two minutes, and 100 µg/L of DOP challenged the filters.

The results of the baseline tests are shown in Table I. These results were used as correction factors for all test measurements. Of particular interest is the great sensitivity of the laser measurements.

Photometer and Laser Measurements in Corrosive Environment

The effects of humidity, nitrogen oxides (NO_x) and temperature on the standard DOP test method were examined using a 2³ factorial design. The design consists of eight tests, with three experimental variables (NO_x, temperature, relative humidity) that may be replicated as many times as desired. The important feature of this design is that it incorporates the extreme points of the factor space. The ranges of the variables were 500 to 10,000 ppm NO_x, 66 to 93°C, and 50 to 95% relative humidity.

The first DOP test was performed at 500 ppm NO_x and 50% relative humidity. The photometer gave a poor response. Additional tests were performed using room air (16% RH) and 10,000 ppm NO_x. The results are shown in Table II.

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Table I. Baseline measurement with photometer and laser

| Test Conditions | | | % Penetration | | | |
|---------------------|---------------------|----------------|---------------|-------------------------|----------|------------|
| Relative Humidity % | NO _x ppm | Temperature °C | Photometer | | Laser | |
| | | | Filter 1 | Filter 1&2 ^a | Filter 1 | Filter 1&2 |
| *16 | 0 | 66 | .002 | -- | .0001 | .0001 |
| 16 | 0 | 66 | .003 | -- | .0004 | .0002 |
| *16 | 0 | 66 | .002 | -- | .0004 | .0002 |
| 16 | 0 | 93 | .002 | -- | .0002 | .0002 |
| *16 | 0 | 93 | .002 | -- | .0002 | .0001 |

* Test performed using drying tube.

^a Sensitivity of the photometer was not great enough to measure penetration of second filter in a series.

Test Conditions: flow, 25 scfm; challenge DOP concentration, ~100 µg/L; test period, 2 min.; temperature, 66 and 93°C.

Table II. Water vapor and NO_x effects on measuring instrumentation

| Test Conditions | | % Apparent Penetration | |
|---------------------|-----------------------------------|----------------------------------|-------|
| Relative Humidity % | NO _x Concentration ppm | | |
| | | Photometer | Laser |
| 50 | 500 | 0-40% on 100% scale ^a | .0005 |
| 16 | 500 | Negative Response ^b | .0002 |
| 50 | 0 | 0-40% on 100% scale | .0004 |
| 16 | 10,000 | Negative Response | .0002 |
| 50 | 10,000 | 0-40% on 100% scale | .0001 |

^a The readings on the photometer were erratic varying from 0-40%; no readings were obtainable on the more sensitive scales of .01, .1, and 1%.

^b The photometer reading was below zero on the .01 and .1% scales.

Note: All tests were conducted at 66°C and measurement made downstream of the first filter.

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This difference in the performance of the photometer and the laser in humid atmospheres may be attributed to the difference in sampling areas of the two instruments as shown in Figure 4. In a forward light-scattering photometer, the sample is drawn through a solenoid valve and into the scattering chamber where the measurements are made. The stream exits the scattering chamber and is exhausted through a vacuum pump. The volume of the sample chamber of the photometer can cause a gas stream with a high dew point to be cooled followed by condensation. By contrast, the sampling area of the laser is extremely small. The sample is pulled through two small jets. This makes condensation an unlikely occurrence in the laser.

The response of the photometer when measuring a gas stream that contains NO_x appears to be caused by light absorption by NO_2 . The standard DOP test method requires that "clean filtered air" be drawn through the scattering chamber while adjusting the stray light so that the photometer meter reads zero. After an adjustment is made in this manner and the gas stream containing NO_x is sampled, the light is not scattered forward but is absorbed by the NO_2 and the result is the negative response of the photometer's meter. The effect was eliminated by making the stray light adjustment while sampling a stream that contains NO_x . No NO_x interference was noticed with the laser.

The problem of condensation occurring in the photometer sample chamber was solved by installing the drying tube to remove water vapor. A comparison of data collected with the photometer and the laser is shown in Table III. The data indicate that the photometer performance improved greatly after the compensation for the NO_x and the removal of water vapor. However, the laser's sensitivity is still superior.

An analysis of the penetration data based on a two-level factorial design and the 95% confidence interval for an effect indicate:

Photometer

- (1) H_2O effect = $15 \times 10^{-3}\% \pm 2 \times 10^{-3}\%$; adding water to the stream enhances the penetration of test filters.
- (2) NO_x effect = $1 \times 10^{-3}\% \pm 2 \times 10^{-3}\%$; the effect of NO_x cannot be established within the confidence limits.
- (3) Temperature = $-4 \times 10^{-3}\% \pm 2 \times 10^{-3}\%$; increasing the temperature lowers the penetrations.
- (4) $\text{H}_2\text{O} \times \text{NO}_x$ interaction = $-4 \times 10^{-3}\% \pm 2 \times 10^{-3}\%$; the interaction between H_2O and NO_x lowers penetration.
- (5) $\text{H}_2\text{O} \times \text{temperature}$ interaction = $3 \times 10^{-3}\% \pm 2 \times 10^{-3}\%$; the interaction between H_2O and temperature is very small as compared to experimental error, and may be questioned, but it also lowers penetration.

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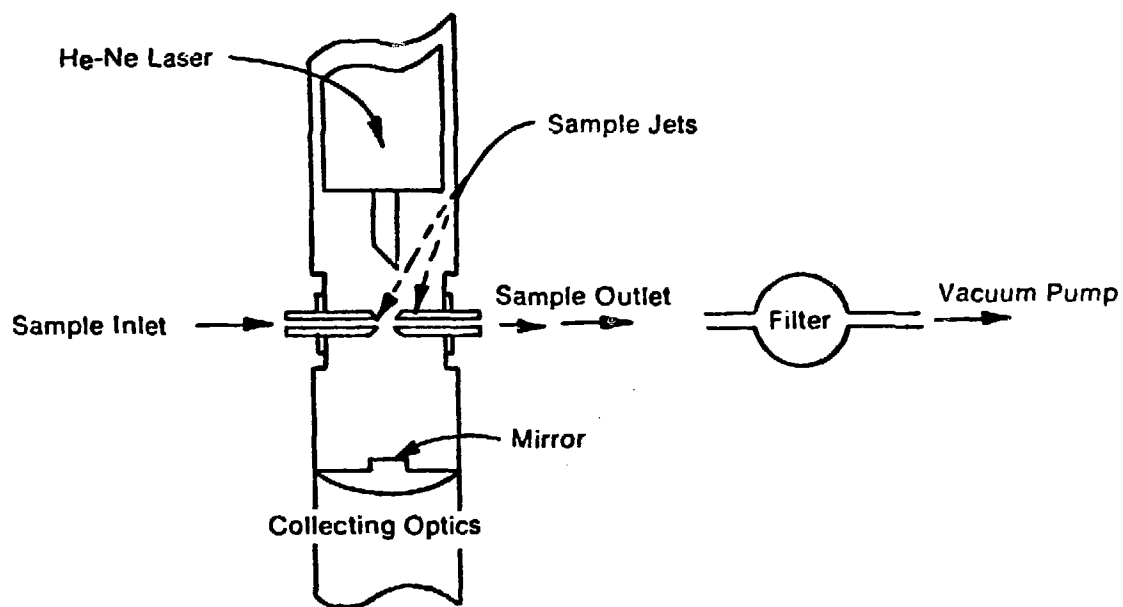
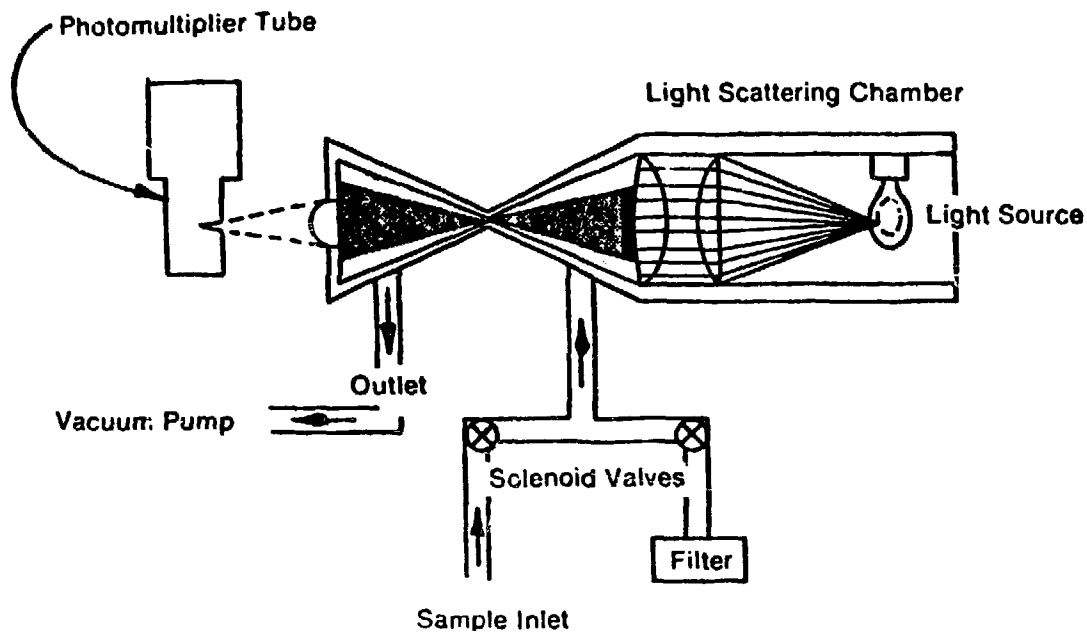


Figure 4. PHOTOMETER AND LASER SAMPLING SCHEMES

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Table III. Measured HEPA filter efficiencies in corrosive environment

| Test Conditions | | | % Penetration | | | |
|---------------------|---------------------|----------------|---------------|------------|----------|------------|
| Relative Humidity % | NO _x ppm | Temperature °C | Photometer | | Laser | |
| | | | Filter 1 | Filter 1&2 | Filter 1 | Filter 1&2 |
| 50 | 550 | 66 | .010 | -- | .0021 | .0007 |
| 95 | 500 | 66 | .030 | -- | .0025 | .0016 |
| 50 | 10,000 | 66 | .014 | -- | .0021 | .0018 |
| 95 | 10,000 | 66 | .030 | -- | .0048 | .0007 |
| 50 | 500 | 93 | .008 | -- | .0270 | .0027 |
| 95 | 500 | 93 | .025 | -- | .0420 | .0021 |
| 50 | 10,000 | 93 | .014 | -- | .0220 | .0065 |
| 95 | 10,000 | 93 | .020 | -- | .0310 | .0058 |

Eight-test experimental design; design points were duplicated using both instruments for measurements.

Test period, 2 min.; DOP concentration, ~100 µg/L (photometer), 10⁶ counts (laser); the experimental uncertainty for laser measurements was ±.0008%.

Sensitivity of the photometer was not great enough to measure penetration of second filter in a series.

- (6) NO_x x temperature and H₂O x NO_x x temperature = $1 \times 10^{-3}\%$ ± $2 \times 10^{-3}\%$, and $2 \times 10^{-3}\%$ ± $2 \times 10^{-3}\%$, respectively; the interactions between NO_x and temperature, and H₂O and NO_x and temperature are insignificant.

Laser

- (1) H₂O effect = $6.0 \times 10^{-4}\%$ ± $9.6 \times 10^{-4}\%$; adding water vapor has no effect on DOP penetration of the filters.
- (2) NO_x effect = $7.5 \times 10^{-4}\%$ ± $9.6 \times 10^{-4}\%$; the presence of NO_x in the test stream does not affect laser measurement.
- (3) Temperature effect = $-2.6 \times 10^{-3}\%$ ± $9.6 \times 10^{-4}\%$; rises in temperature cause decreased filter penetration.
- (4) H₂O x NO_x = $3.0 \times 10^{-4}\%$ ± $9.6 \times 10^{-4}\%$; the interaction between H₂O and NO_x is insignificant.
- (5) H₂O x temperature = $-5 \times 10^{-4}\%$ ± $9.6 \times 10^{-4}\%$; the interaction between H₂O and temperature is also insignificant.
- (6) NO_x and temperature = $8.5 \times 10^{-4}\%$ ± $9 \times 10^{-4}\%$; the interaction between NO_x and temperature cannot be established within the confidence limits.

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- (7) $H_2O \times NO_x \times \text{temperature} = 3.0 \times 10^{-4\%} \pm 9.6 \times 10^{-4\%}$; the interaction of all three variables is insignificant.

Computation of factor effect showed that water vapor was the only main effect when using the photometer as the measuring instrument. No main effect was indicated during use of the laser.

IV. Conclusion

Accurate results cannot be obtained when using the conventional forward light-scattering photometer in a corrosive off-gas environment to measure DOP concentrations by the ANSI N-101.1 method. The test data indicate interferences in the method due to water vapor and NO_x . However, by modification of the method to include a unique sample drying process before the sample enters the photometer, the water vapor interference can be eliminated. The NO_x effect on the photometer reading can also be corrected by a stray light adjustment. Temperatures used in these tests do not affect the conventional photometer.

The laser spectrometer appears to be a very capable instrument for filter test work. Its high sensitivity allows for the measurement of penetration factors in a corrosive off-gas environment when using two HEPA filters in series. It is not affected by water vapor, NO_x , or temperature under the described test conditions.

V. References

1. ANSI N-101.1-1972, "Efficiency testing of air-cleaning systems containing devices for removal of particles," New York (1972).
2. B. G. Schuster, D. J. Osetek, "The use of a single particle intracavity laser particle spectrometer for measurements of HEPA filters and filter systems," Proceedings of the 14th ERDA Air Cleaning Conference, CONF 760822.
3. Perma Pure Products, Inc., "Continuous drying of process sample streams," Manufacturer's Literature, Oceanport, NJ (1973).

DISCUSSION

STEINBERG: Did you take the scattering chamber apart after the test to see if there was any water in it?

MURPHY: Yes, we did. We ruined the photometer and had to buy another one.

STEINBERG: But, was there any water in the scattering chamber?

MURPHY: I assume that it had dried by the time we figured out what was going on and took it apart. We could see a white residue on the inside of the scattering chamber from dried water.

STEINBERG: We've run tests under high humidity conditions although not with

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NO_x. Even when the scattering chamber was wet inside we have had pretty good results. Do you think it was the NO_x that might have been giving you the trouble?

MURPHY: No, I'm pretty sure it was condensation.

KOVACH: Have you made any runs with only high humidity and no NO_x?

MURPHY: Yes, with our standard room air (16% RH) and 500 ppm NO_x we got a negative response. When we increased water vapor to 50% RH and NO_x to 500 ppm or 10,000 ppm, the response of the photometer ranged from 0 to 40%, with no stability in the readings. This is shown in Table II of the paper.

STEINBERG: Usually, that sort of photometer response is indicative of something inside the scattering chamber that gives you extra stray light. This, in turn, gives you an erratic response. But, my curiosity is directed towards the water vapor because we have run under humidity conditions much higher than 50% with fairly good results. So, maybe you should try it again sometime.

MURPHY: OK, but I think that when the temperature drops below the dewpoint of your stream, you are going to get condensation inside the photometer. If you keep above the dewpoint, which we did not, you won't get condensation. Because our extreme condition was going to be 95% relative humidity, we decided to go to a drying tube. The dewpoint at 95% humidity is 90°C. I talked with several people who said that we shouldn't heat the stream up much more than 80°C to make measurements with the photometer and with the laser. That's why we decided to dry the stream before we made our measurements.

STEINBERG: The next time you try it, you might also try putting heating tape around the scattering chamber.

MURPHY: That's what I'm saying. The reason we didn't do that is because of the extreme conditions of these tests. They require that our dewpoint be at 90°C and we didn't want to heat the chamber up over 80°C. That's why we decided to use the drying tube; it would enable us to make measurements at 50 and 95% relative humidity without modifications to the system.

STEINBERG: What is the significance of the laser results? Were they able to tell you something about the condition of the HEPA filters, i.e., whether they were good or bad? Whether you had any leaks or not?

MURPHY: It told us that an increase in temperature caused increased penetration of the filters. That was the result of my tests with the laser.

ANDERSON: The data in Table III indicate that the second filter in a tandem filter combination is not as effective as the initial filter. I would suspect that the laser detection system is the explanation for the difference in the reported penetration results.

MURPHY: That's right.

ANDERSON: If both filters are in tandem, the number found should be anywhere from the sixth to the ninth decimal place to represent the combined efficiency.

MURPHY: I've had discussions with my section chief about this and he said the same thing. I'm not sure I understand what he's saying or what you're saying, but the way I took this measurement was probably contrary to what he recommended. This measurement is based on the upstream challenge aerosol concentration and the

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downstream aerosol concentration after the second filter. The numbers shown are what I got.

ANDERSON: By your data, you're indicating that the second filter was not effective inasmuch as it did not take very much more of the material out than one filter, alone. Take the case in Table III where you show .0021 for the first filter and .0007 for the second. If you assume that the effluent from the first filter is the input to the second, you divide 7 by 21 to find the second filter is only 30% effective.

MURPHY: That's exactly what it came out to be. We went through Dr. Schuster's paper on testing HEPA filters in series, and these are the results I got from my experimentation.

RIVERS: I believe you should indicate whether the percent penetrations were for individual filters or for two in series. Also, was the same pair of filters used for all experiments?

MURPHY: The same pair of filters was not used in all tests. I had trouble controlling the atmospheres in all the tests, but the design was sufficiently random that changing the filters didn't affect the test results.

IN SITU TESTING OF TANDEM HEPA FILTER INSTALLATIONS
WITH A LASER SINGLE PARTICLE SPECTROMETER SYSTEM*

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Abstract

System components and methodology for in place testing of tandem HEPA filters are described. The system consists of a very high volume thermal DOP generator for producing sufficient challenge aerosol, a single particle intra-cavity laser particle spectrometer for high sensitivity detection, a forward light scattering photometer to measure challenge aerosol concentration, and a diluter to interface the spectrometer and photometer measurements. Results of the measurements on 11 tandem HEPA installations are presented along with problem areas and recommendations for future work.

I. Introduction

In order to implement the testing of tandem HEPA filter installations, a series of feasibility studies was initially performed on a small laboratory scale system. Using the concepts of measurement and digital counting obtained by implementing an intra-cavity single particle laser spectrometer rather than the conventional instantaneous analogue read-out of a forward light scattering photometer, tandem filter protection factors (PF) of 10^9 were readily measured. Challenge aerosols composed of solids (NaCl) and liquid (DOP) were employed. The size distributions of these two aerosols were essentially identical, as were the individual filter PF's, as a function of particle size. These measurements displayed a minimum PF at $0.19 \mu\text{m}$ with increasing protection on either side of this minimum. The fundamental difference in the use of liquid versus solid aerosols was in the rapid increase of pressure drop, i.e., filter plugging, that occurred with solid particulates, hence resulting in a semi-destructive test. The similarity of other results, however, suggests that DOP testing is a valid technique for exploring the critical behavior of filters.⁽¹⁾ The data also implies that since the size characteristics of a challenge aerosol are changed during the filter interaction, that the light scattering photometer PF's are really only a ratio of light scattering signals rather than a ratio of particle counts, masses, or areas. This follows from the very complex dependence on radius, wavelength, index of refraction, and concentration of the light scattering processes occurring within the photometer. In addition, the product of the PF's measured individually on a tandem system is likely to over estimate the system PF. This is a consequence of the first filter removing the most easily collected particles, hence effectively providing a

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challenge to the second filter of mostly difficult to collect particles. Only if the challenge aerosol were absolutely monodisperse would there be no measurement differences between the two detectors in the above situation, providing that the particle concentrations were kept low enough to avoid multiple scattering in the photometer and excessive coincidence errors in the spectrometer.

One additional advantage in the use of the spectrometer is its ability to integrate, i.e., store the particle count per size channel, over any required time period. This, coupled with the complete lack of instrument noise (a common feature of digital devices) enhances the detectivity for low particle concentration. In other words, it is not just an instantaneous rate, but the rate integrated over time that is measured. For extremely low concentrations, the measurement is facilitated by increasing the counting time.

In order to implement this technique for field test use, several new pieces of equipment had to be designed and built. Chief among these was a high volume thermal DOP generator⁽²⁾ for producing the very large challenge aerosol concentrations required for tandem HEPA filter installation testing. This generator output is equivalent to 10 of the smaller commercially available units. A diluter for diluting the very high upstream challenge aerosol concentrations so that they are amenable to single particle measurement was also constructed.

The need for tandem HEPA filter testing was dictated by the desire to more accurately characterize system performance. Current test methods are limited to testing separate stages and may overestimate system performance. In addition to decreasing system downtime and worker exposure during testing, considerable dollar and space savings may be effected by designing tandem filter ventilation systems so they need not undergo stage-by-stage testing.

II. Test Apparatus

For tandem HEPA testing, protection factors of 10^7 to 10^8 are encountered, as opposed to 10^3 to 10^4 for single filter testing. It is obvious that much greater concentrations of challenge aerosol are required for the tandem test. Even though the detectivity of the laser spectrometer is considerably enhanced over that of the photometer, excessive integration times are undesirable. Additionally, the presence of downstream background aerosol, due usually to ductwork leaks, can mask the presence of very small concentrations of test aerosol. Typically, a challenge aerosol of 10 to 100 times the concentration normally used for single filter testing is required. The laboratory system of a six stem Laskin generator and associated compressed air supply, when considered in the multiplicity required for an industrial application, is far too cumbersome. In several tests, commercially manufactured thermal DOP generators were used, but their individually variable outputs and operating periods made multiple units awkward to work with. The DOP reservoirs in these devices were enlarged, which increased the operation time, but the combined output from the several generators did not behave in a temporally constant fashion and the tending of two to four separate generators proved unacceptable.

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So that these difficulties could be overcome, an effort was directed at constructing a thermal DOP generator which would have an output several times that of the individual commercial generators and using the same basic principles of operation. The successful completion and splendid performance of this system provided confidence in the further scaling to a system providing ten times the output, i.e., 100 gms of DOP and 34 liters of CO₂, per minute, when operated at 30 psi.

The primary aerosol detector is a LAS-X single particle intracavity laser spectrometer specifically designed for this measurement. The instrument measures particles in a size range of 0.1 μm to 3.1 μm in 0.1 μm increments. The counts per size channel are stored and automatically printed out along with appropriate house keeping data such as time, date, and sample time. Graphical displays can be obtained with the use of an auxiliary oscilloscope or x-y recorder. Sample flow rate is 15 cm³/s, considerably greater than previous designs of this instrument.

The advantages of such an instrument are manifold. The ability to measure a size spectrum obviates the problems associated with trying to generate a monodisperse aerosol and, in fact, characterizes a filter system's efficiency as a function of size. The digital technology associated with single particle counting insures against instrumental background. The counting feature enables a measurement to continue until a statistically significant sample of N counts has been registered, the percentage error then being proportional to $N^{-1/2}$. This is all in contradistinction to the instantaneous photometer type of readout which derives from a gross scattering of light from a population of particles, and is insensitive to single particles.

In the measurement of very high concentrations of upstream challenge aerosol, precautions must be taken to insure that any optical measuring system is not operated in a range for which it was not designed. In the case of the particle spectrometer, this limit is set by a maximum count rate, or electronically, by the frequency bandwidth of the instrument. For a photometer, the limitation is imposed by the onset of multiple scattering. This typically occurs when the optical depth reaches a value of 0.1 or greater. For the range of aerosol sizes normally encountered in filter test measurements, this corresponds to a concentration of order 10⁷/cm³, which is reasonably below the concentrations used for the tandem testing procedure described in this paper. However, the upstream concentration will saturate the single particle detector.

In order to make use of the high saturation level of the photometer upstream and the great sensitivity of the single particle detector downstream, a diluter section is used in the measurement. This consists basically of a small wind tunnel into which HEPA filtered room air is drawn. A probe iso-kinetically samples the upstream challenge aerosol, introducing it into the diluter, where it is mixed with the much larger volume of clean air. Flow rate and pressure drop are controlled by varying blower power and throttling the air flow. Appropriate instrumentation is included to measure

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flow rate and pressure differentials. The dilution ratio, which is typically in the 10^{-3} range, can be computed and/or measured. Since the size distribution of the diluted sample should be unchanged from the original population, the photometer is used to determine this value. Measurement of the diluted sample with the spectrometer will then provide the upstream count rate, which is proportional to the challenge concentration.

The diluter also serves a second, very important purpose in that it is responsible for bridging three of the seven or eight orders of magnitude associated with the measurement of the protection factor. Although the spectrometer dynamic range is essentially unlimited, decreasing the challenge concentration to a point where it is directly measurable would result in enormously long downstream measurement times.

The photometer used for these measurements may be any one of the conventional forward scattering types now used for conventional HEPA filter testing. It should be of the linear read-out variety and be gain switchable over at least three noise-free orders of magnitude.

The two major systems of DOP generator and diluter are mounted on two-wheel hand carts. The two detection instruments are hand portable. The complete system is shown in Figure 1.

III. Procedure

The methodology associated with a tandem HEPA filter system test is not, with the exception of the dilution procedure, much different from conventional filter tests, but because of high protection factors usually involved, systems with flow rates greater than 20,000 CFM should usually not be attempted. The DOP generator is first emplaced as far upstream as is practicable. Appropriate iso-kinetic probes are installed and, if possible, centered immediately upstream and downstream of the tandem plenum. The upstream probe will usually consist of a probe pair so that the flows for both the diluter and photometer may be matched. The diluter has internal probes matched to the photometer and spectrometer flows. The generation of DOP is then initiated and monitored with the upstream sampling photometer until a steady state of generation obtains, at which time the photometer is switched to the diluter and a measurement made. Simultaneously, a diluter measurement is made with the spectrometer. Because of the still high concentration in the diluter, a spectrometer measurement for one minute will be quite sufficient. Over 10,000 counts will be registered in this interval, assuring an accuracy of better than 1%. The upstream concentration is now proportional to the product of the upstream photometer reading and the diluter spectrometer count rate divided by the diluter photometer reading. The downstream reading is made directly with the spectrometer. Ten minutes are typically required to obtain a count of 100, which will provide 10% accuracy.

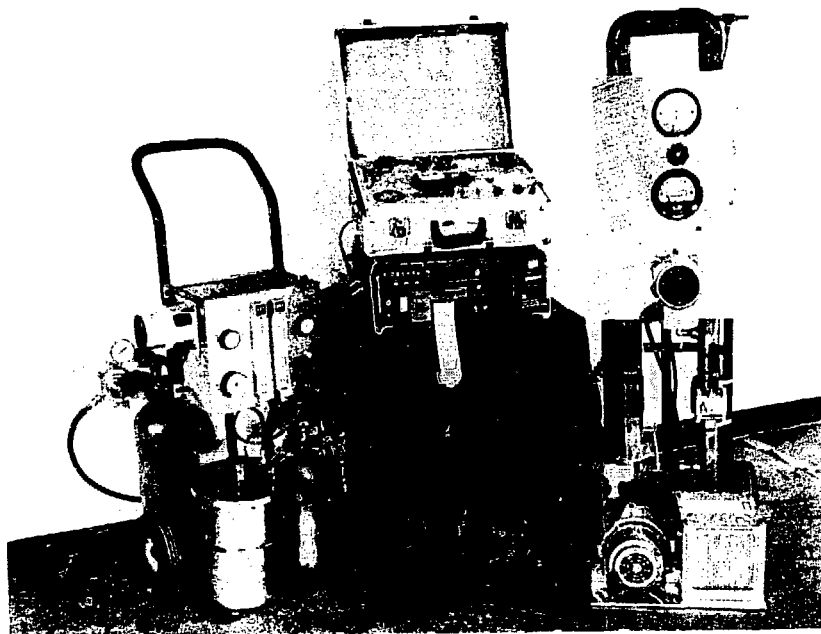


FIG. 1
FILTER TEST APPARATUS - GENERATOR, DILUTER, DETECTORS

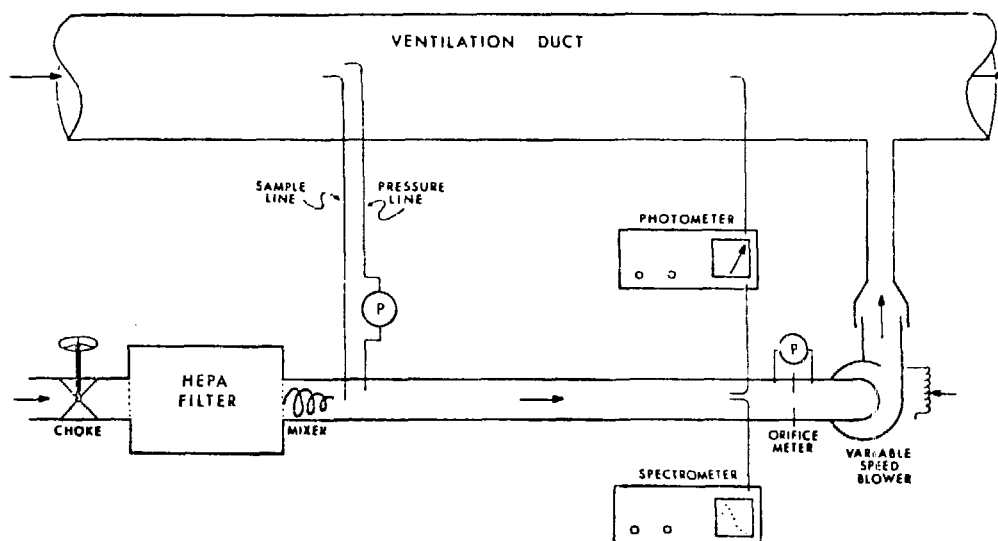


FIG. 2
TEST PROCEDURE SCHEMATIC

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In a great many instances, a downstream background count will be present, due usually from in-leakage of ambient air into the downstream ducting, which is at a negative pressure with respect to the ambient air. In this case, background measurements should be made (with the aerosol generator turned off) before and after the system is challenged, so that they may be subtracted from the actual test measurement. Upstream background usually poses no problem because it is overwhelmed by the enormous challenge concentration. In some instances, the downstream background may be so high that it masks the test measurement. In this case, a lower limit on the system performance can still be obtained using the noncorrected downstream count. A schematic of the measurement procedure is shown in Figure 2.

IV. Results

Table I tabulates the results of nine systems measured during the acceptance testing phase at the new LASL Plutonium facility. These measurements were accomplished with an earlier model low flow rate spectrometer designed primarily for laboratory use. Conventional photometer data, for the acceptance tests, were acquired for individual banks. As anticipated, the tandem protection factors derived from these latter measurements usually overestimated the system performance by about a factor of two.

The data shown in Table II were acquired with the LAS-X spectrometer. Because of operational constraints associated with the test schedule for LASL filter systems, only these installations were able to be tested up to this point, but they are of particular interest. System 1 shows a typical downstream background count rate which is small enough so that it can be successfully subtracted from the test count rate to produce a reasonably accurate test. System 2 displays a very different sort of behavior in that the background count rate increases after the system has been challenged and in fact is greater than the count rate taken during the challenge. It appears that there is an interaction between the DOP and the material collected on the filter which creates the change. Simultaneously, there is an increase of pressure drop of 25 mm across the filter. This particular system has been measured several times with similar results. Laboratory tests on clean filters do exhibit an increase in P.F. immediately after and during the challenge, but no background due to DOP was ever observed. The P.F. for this system was computed by using the maximum downstream value observed, 1.13, and calling this the lower limit or minimum P.F. for the system.

V. Conclusions and Recommendations

The system that has been described can, of course, also be used for measuring single filter bank installations. One such system of 160,000 CFM flow rate was, in fact, successfully measured; a task virtually impossible with conventional test methods. The system has performed reliably and is readily portable. One problem area that has already been discussed is that of excessive downstream background. This situation is now under study using the

TABLE I
TA-55 FILTER TESTS

| <u>System</u> | <u>Tandem Test
Protection Factor</u> |
|---------------|--|
| C | 7.2 ± 1.3
10^7 |
| F | 2.7 ± 0.3
10^7 |
| A | 1.3 ± 0.2
10^8 |
| G | 1.8 ± 0.4
10^7 |
| B | 9.8 ± 2.8
10^7 |
| D | 5.6 ± 1.6
10^7 |
| E | 5.5 ± 1.3
10^7 |
| I | 4.2 ± 0.4
10^5 |
| H | 6.2 ± 0.7
10^6 |

TABLE II
CMR FILTER TESTS

| | <u>System</u> | |
|--------------------------|----------------------------|-----------------------------|
| | <u>CMR 1</u> | <u>CMR 2</u> |
| Flow Rate | 17500 CFM | 16400 CFM |
| Upstream
Count Rate | 8.450×10^7 | 6.184×10^7 |
| Downstream
Count Rate | | |
| Before Test | $.30 \pm .10$ | $.29 \pm .10$ |
| After Test | $.30 \pm .12$ | $.13 \pm .20$ |
| During Test | $2.20 \pm .27$ | $.30 \pm .10$ |
| Downstream
Net | $1.90 \pm .29$ | $1.13 \pm .20$ |
| P.F. | $(4.4 \pm .7) \times 10^7$ | $(5.5 \pm 1.0) \times 10^7$ |

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concept of a self-identifying challenge aerosol that can be distinguished from background particulates. The phenomenon of changed efficiency due to the interaction of the challenge aerosol medium and the filter is one which should be thoroughly studied in order to avoid falacious high efficiency measurements on older, partially loaded filters.

VI. Acknowledgements

The authors wish to express their gratitude to John Ortiz and Elmer Garcia for their aid in construction of the DOP generator and for participation in the field test program; and to Marvin Tillery for his helpful consultation.

References

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DISCUSSION

DUGGAN: Do you know what percent of in-leakage there was downstream of the total design flow of the system?

SCHUSTER: I think it's very small, usually from pinhole leaks or welding leaks. From the point of view of contamination, one doesn't worry too much about it because the air is always going into the duct rather than coming out of it and it is also downstream of the filters. But it does mess up the measurements simply because one cannot distinguish between the challenge aerosol that has been introduced and whatever else leaks in by any method that relies on conventional electromagnetic scattering.

DUGGAN: Have you made an effort to pinpoint those leaks and reduce them to decrease your background?

SCHUSTER: We decorated lots of ducting with an awful lot of yellow tape.

CHEEVER: How long did the elevated background persist downstream from the HEPA filters after DOP testing the no. 2 system? Did it come back to the original background?

SCHUSTER: We monitored one system for about 36 hours and it took many hours to come back down. That was an unusual case. We think we were making smoke in that system. We don't know what was on the filter, but we suggested that when it was pulled that it ought to be analyzed chemically to determine what was on it.

MORROW: Can you explain the problem of anomalies with NO_x as expressed by the previous speaker?

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SCHUSTER: I think I'm going to refer that to Tom Kyle because he's more of a spectroscopist than I and knows more about the adsorption of NO_x in the visible range.

MURROW: One more question, is your laser spectrometer available commercially?

SCHUSTER: Yes, it is.

MCDONALD: Can you tell us the cost of the laser instrumentation relative to the cost of the forward scattering photometer system?

SCHUSTER: It's approximately two to two and a half times as much. The systems that we have are somewhat different because we usually provided specifications that make them essentially custom instruments. I think if other people would specify the same type of instrument, the price would go down. Ours have usually been prototype instruments.

WILHELM: Was there a large temperature effect due to the operation of the aerosol generators?

SCHUSTER: Typically, there isn't. As far as I know, nobody tampered with the blowers so, presumably, the flow was constant.

WILHELM: I am not asking about the flow, only the temperature, because you had a huge generator in front. Was there no increase in temperature during testing?

SCHUSTER: Yes, I believe that there was a slight increase in the temperature. After all, we were boiling off DOP and it is hot. But, by the time it got to the filters I doubt if there was very much of a temperature rise. I believe it was less than a degree centigrade.

STEINBERG: What was your upstream concentration of DOP?

SCHUSTER: The number I remember is 10^8 or 10^9 particles per liter. I can't give it to you in terms of mass per unit volume.

STEINBERG: I wonder if, with your generator, you were putting so much DOP on the filters that it was coming through and that was why you were getting penetration?

SCHUSTER: When we did the original tests, with clean filters, in the laboratory, we looked for that phenomenon. We put a lot of DOP on the filters, and turned off the generators, and then looked to see if anything was coming off. It wasn't.

STEINBERG: Did you notice any wetting by DOP on the upstream side of the filter?

SCHUSTER: In the laboratory, when we took the filters apart, we could see wetting, but we could not see any particles coming off downstream.

STEINBERG: How about in your test outside the laboratory; was there any wetting?

SCHUSTER: We did not go into the plenum to look at the filters.

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STEINBERG: Usually, wetting is indicative of more DOP than you need to run a test.

SCHUSTER: Just remember that we were looking at two filters in tandem. In the laboratory we deliberately overloaded them to see if DOP would come off. I showed an anomalous example deliberately to indicate what might happen with a dirty filter, not a clean one.

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Dup

MEASUREMENTS WITH SELF-IDENTIFYING AEROSOLS*

by

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ABSTRACT

The use of modern single particle detection of aerosols has made it possible to test multiple HEPA filter systems. Problems have been encountered in such tests with the background aerosol on the downstream side of the filters. Since the ducts are maintained at a negative pressure, ambient aerosols leak into the ducts and provide concentrations of particles which are comparable to the number of particles of the test aerosol passing through the filters. The background count creates an upper limit to the protection factor which can be measured.

This limitation can be overcome by the use of an identifiable aerosol. The most direct method of identifying an aerosol is to use fluorescent particles, which has been done in the past. In the present case, the fluorescence of the individual particle is used, rather than dissolving the particles in solution to develop the fluorescence. All the advantages of single particle detection are retained, such as orders of magnitude greater sensitivity, digital data techniques, and freedom from instrumentation drift. Possible new measurements include backfeed in air distribution systems, and the re-entrainment of particles from surfaces within air cleaning systems.

Two systems based upon fluorescent solid or liquid particles are being developed. Both involve single particle detection and excitation by a portable helium-cadmium laser.

The advantage of the solid particles system is the ability to capture the particles on a filter. The filter can then be dissolved and the number of particles counted and sized, since the particles do not dissolve. This also permits the use of greater sample flow rates than is possible with a real time system.

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I. Introduction

The testing procedures for HEPA filters involves introducing an aerosol upstream from the filter, then measuring the concentration of the aerosol previous to and after passing through the filter. In the past, the aerosol concentration was usually measured by means of the light scattered from a volume of air large enough to contain many particles. More modern equipment makes it possible to both count and determine the size of individual particles.

The capability of dealing with individual particles relieves some previous restrictions required to ensure valid testing. The ability to determine the size of the individual particles allows the test aerosol to have a wide range of sizes. By counting particles within a narrow size interval and neglecting all other sizes, the filtration efficiency can be described for the particle size range of interest. Although not required, considerations of efficient generation indicate that a reasonably narrow size distribution from the aerosol generator is desirable.

The most important improvement that single particle detection allows is in sensitivity. Use of light scattering from multiple particles requires a minimum concentration of particles per unit volume to provide a measurement. In the case of single particle detection, there is no lower limit of aerosol particle concentration that can be detected. The practical limit is in terms of time available to await the arrival of a particle in the measuring volume.

The increased sensitivity means that a single test can be used to determine the protection factor (PF) for multiple stages of HEPA filters. This has been tried and found to work. However, an unexpected problem was encountered that sometimes caused the technique to be unusable. The problem arises because the duct in which the filter is located is at a lower pressure than the surrounding room, and air from the room can leak into the duct. The aerosol particles in the room air are then counted because they cannot be distinguished from the aerosol particles introduced upstream from the filter. If the aerosol particles from the room air were only a small percentage of the total particles being counted, there would be no problem, but when the PF is high, the opposite case can occur. That is, only a small change in the number of particles is noted when the aerosol generator is turned on or off, because most of the generated particles are removed by the filter and leaked-in air is the primary source of particles.

The solution to the problem of aerosol particles leaking into the ducting and confusing the filter test is to generate an aerosol that is identifiable and to only count those particles that are identified as coming from the aerosol generator. All particles scatter light to some degree, and so some technique other than light scattering is required.

II. The Use of Fluorescent Particles

There is a method by which the particles can be identified and yet maintain the advantages of light scattering measurements. It is the use of fluorescent particles. When these particles are illuminated with light of one wavelength, they emit light at a longer wavelength. Thus, they can be distinguished because the emitted light is of a different wavelength than the illuminating light. The fluorescent nature of the particles causes them to emit light when illuminated by light of the proper wavelength, and this provides a signal akin to scattered light, but distinguishable from it by the shifted wavelength. Therefore, they can be measured by much the same techniques as light-scattering measurements.

In operation, one wavelength of light will illuminate the particle, and a longer wavelength will be emitted, and this longer wavelength is detected. This requires illumination by something other than white light. Otherwise, scattered and emitted light could not be distinguished, eliminating the unique identification of aerosol particles from the generator. Very small particles must be detected requiring a very intense source of illumination. These requirements are best satisfied by a laser, especially the need for high intensity light.

There are many naturally fluorescent materials, but few of these are highly fluorescent. Rather than search for naturally fluorescent materials from which an aerosol can be generated, it is usually more practical to determine the substance for the aerosol, then locate a fluorescent dye that can be used to make it highly fluorescent.

A dye for DOP is a desirable approach as it would allow all the DOP particle technology to be used, and thus reduce the complexity of the undertaking. The DOP technology includes answers to questions of the toxicity of the materials used for the testing of filters and the safety problems when introducing the test aerosol into the air cleaning system. These problems are more severe than it might appear, as can be illustrated by an example. The physical properties of glycerol made it attractive as an aerosol material, and an additive for making it fluorescent is known. However, this approach had to be dropped when it was pointed out that the nitric acid vapor in some air cleaning systems could react with the glycerol to form nitroglycerin.

A survey of 18 manufacturers and distributors of fluorescent materials resulted in no suggestions as to additives that would cause DOP to become fluorescent. Therefore, an experimental approach was used, during which 113 different dyes were added to DOP as a test of solubility. Those having a reasonable solubility were also analyzed in a fluorometer to determine the wavelength of maximum absorption of radiation and the emission spectra. The result was six materials that appear to satisfy the present requirements.

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III. Particle Generation

Most fluorescent dyes would decompose at the temperatures encountered in thermal DOP generators. The use of a thermal generator is desirable because of the high output this type of generator provides. A method was sought to provide for dispersal of DOP in an aerosol with particle diameters of a few tenths of a micron, but with total throughput of up to a few liters per hour. It is not usually possible to generate droplets this small with a nebulizer or spray system, but at least one system has been found that satisfies the need for a high volume generator of submicron particles of DOP other than a thermal generator.

The unit is called a sonic atomizer and is commercially distributed by the Sonic Development Corporation, primarily as a spray for breaking fuel oil into very small droplets to promote better burning of the oil. The unit operates by using compressed air to create a standing sonic shock wave when the air flows through a convergent-divergent nozzle into a resonator chamber at supersonic velocity. This can be better visualized by reference to Fig. 1. As the air is reflected from the resonator, it creates a standing shock wave between the cavity and the body of the nozzle. Liquids pumped into the shock wave are vigorously sheared into fine droplets.

Tests were carried out on one of these units using DOP as the liquid and a compressed air pressure of 40 psi. Droplets having a mass median diameter of 0.35 microns were produced. The range of droplet sizes was too great to be considered monodisperse, but not unduly wide. It should be recalled that the particles are to be sized by the instrument so a monodisperse source of droplets is not required. The volume output of DOP in the form of the aerosol droplets was not determined, but on the basis of the manufacturer's specifications for water instead of DOP, a volume flow of 1 or 2 l/h seems reasonable.

The detection and sizing of the fluorescently dyed DOP can be carried out in much the same way as the presently used laser single particle measuring system. Two modifications are needed, but these are such that a new instrument would have to be purchased. The primary modification is the use of a helium-cadmium laser instead of a helium neon. The He-Cd laser provides a blue light output rather than red, which is required to induce the fluorescence in the dyes that are soluble in DOP. The second modification is the insertion of filters in front of the detector, which pass light of the wavelength where fluorescence occurs, yet blocks light at the wavelength of the laser. The blocking of the laser wavelength is necessary to avoid signals due to light scattering by aerosol particles that may or may not be fluorescent.

IV. Solid and Liquid Particles

It was originally felt that the most desirable system of identifiable particles would involve the use of DOP with fluorescent dyes. The advantage of using DOP was that it is accepted as producing no hazard by being introduced into an air cleaning system.

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Some of this advantage is lost when dyes are introduced along with the DOP, because many, if not most, of them are suspected of having undesirable effects on health. A more significant advantage of DOP is the small but finite vapor pressure. This permits large quantities of DOP to be introduced upstream from the HEPA filter, so that a large burden is captured by the filter, with no long-term degradation of the filter, because the DOP will evaporate away rather than remain on the filter. Contrast this to the use of a solid nonevaporating particle. The large number of particles that must be introduced to allow a test of a filter system with a large protection factor could provide a permanent loading of the filter that would be a significant fraction of the total burden which the filter is capable of accepting. The implicit assumption in this reasoning is that the testing is the same for the solid as for the DOP aerosol.

Let a filter system have PF of P , which means that if N_1 particles are introduced before the filter, a number N_1/P will pass through the filter system. Let the flow of the air through the filter system be F_1 unit volumes per unit time, and let the sampling system extract air at a flow rate F_2 . The number of particles collected by the sampling system will be N_2 and is given by:

$$N_2 = \frac{N_1}{P} \frac{F_2}{F_1} . \quad (1)$$

If there is a background of particles that confuses the measurement, there is an advantage in supplying the N_1 particles in a very short time, but if the particles are to be identifiable, as in the present case, the rate of introduction is unimportant except for the time that the test requires. The number of particles that must be counted for the test is determined by statistical considerations and can be taken to be of the order of a thousand. The values of P and F_1 are determined by the system under test. The determining factor for the number of particles that must be introduced is F_2 alone. The larger F_2 is, the smaller the quantity of aerosol that must be generated in order to test the system.

The limitation on F_2 is the time that must be allowed to detect and size each particle. Most of the time there is no particle within the sensitive volume of the instrument, but when one does enter, it must remain in the volume for a minimum length of time to allow the signal-to-noise ratio to be large enough to provide a valid measurement. With the single particle laser spectrometer, the flow rates cannot exceed a few milliliters per second.

It is the limited flow rate that the measuring system requires that makes solid particles rather than DOP particles of interest. The DOP particles are liquid and must be measured in the air itself, as they would be absorbed or altered if collected on a filter, but solid particles do not have this shortcoming. By using solid particles, a large flow rate can be used for the sampling system, which will capture the particles on a membrane filter. The filter can then be dissolved in a proper solvent, and the liquid used to dissolve it will then contain the undissolved aerosol particles

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that were on the filter. Perhaps hundreds of liters of air were put through the filter to collect the particles, but they are now contained in a few milliliters of the liquid. The liquid can be put through the measuring system at a low flow rate and the individual particles sized and counted.

An example will illustrate the power of the collection of solid particles on a membrane filter in testing. Assume a standard (24-in. square) HEPA filter system is to be tested. The flow rate F_1 , is 2.8×10^4 l/min and the PF is 10^8 . The sample will be collected on a 47-mm filter using a flow rate, F_2 , of 25 l/min and a total of 10^3 particles must be collected for a statistically valid test. Putting these values into (1) gives the result that a total of 4×10^{13} particles must be released upstream from the filter in order for the test to be statistically valid. If these particles are taken to have a specific gravity of 2, then only 1.14 g of material are required for the test. With this overall quantity of material, there is no significant problem with loading the filter.

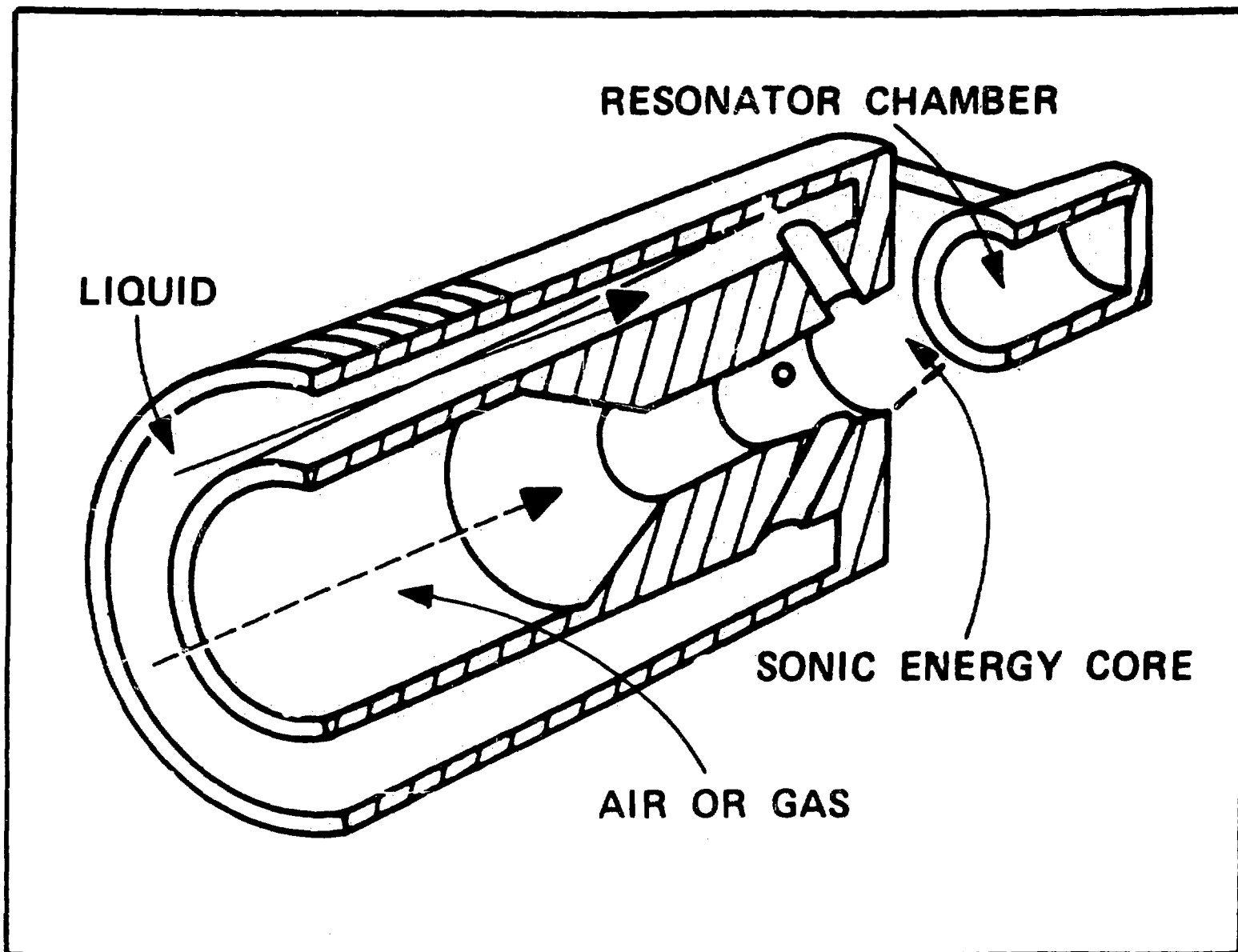
It is instructive to contrast the limitations of different approaches to filter testing. When filter systems are tested with light-scattering photometers, the particles must exceed a certain concentration, say 100 particles/cm³, before they can be measured with an acceptable accuracy. In the case of multiple HEPA filters, the concentration of aerosol particles getting through the filter is too low for a measurement, and so the unit cannot be used for multiple HEPA filter tests. The single particle measuring systems have the sensitivity for measuring systems with high PFs, but leaks of room air into the system often contribute more particles than the aerosol generator, which prevents a valid test. This points out the need for an identifiable particle to use in the testing.

It might seem that an identifiable bulk material could serve the same purpose. For example, particles of sodium fluorescein could be generated, fed through the air cleaning system, and then captured on a membrane filter. The filter could then be dissolved along with the aerosol particles and the bulk fluorescence of the solution measured. In such a case, a concentration of the fluorescent material of one part in 10^8 would be needed. A simple calculation shows that in the case of multiple HEPA filters, the first filter would become overloaded before the required quantity of material was collected.

A single particle detection system using fluorescent particles can provide the desired measurement using either liquid or solid particles. There are advantages to either of these types of particles, but the solid particle approach is the one being followed at this time.

V. Fluorescent Plastic Particles

The solid aerosol particles being used have a number of desirable properties. The material is manufactured by Day-Glo Color Corporation for use as a fluorescent ink in the printing industry, and as a result of the large-scale production is reasonably priced.



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The particles are composed of a thermoplastic material with a fluorescent dye included within the plastic. This type of material is quite inert, and thus should retain the dye within it so that no hazard would be expected from breathing air containing the particles.

The particles are approximately spherical in shape and seem to have a smooth surface. Tests on the size spectra have been carried out. These tests give the mass median diameter of the particles in the range of 0.25 to 0.35 microns. The difference in the values determined is due to different experimental arrangements and different methods of measurements. The size spectrum is not wide, being a lognormal distribution with a dispersion of 1.35.

The operational use of the identifiable particle measuring system with solid fluorescent particles to test filter systems will proceed in somewhat the following manner. The particle dispersing unit will be an atomizer that disperses liquid droplets containing the solid particles. The liquid evaporates leaving only the fluorescent particle.

Air samples will be pulled immediately before and somewhat downstream from the filter system. Air from the system is simultaneously pulled through each of two membrane filters during the time the particles are being dispersed. These samples may be either returned to a central location, or they can be dissolved in a few milliliters of liquid and the particles counted immediately. That is, the system that sizes and counts the particles will be portable and capable of providing the test result in a few minutes; but if it is thought to be a better procedure, the filters can be brought back to a quality controlled situation for the quantification of the test results.

A system such as the one described here should be capable of providing more accurate tests of filter systems since it can test multiple stages of filters in a single test. The testing of multiple stages of filters in a single test should also provide economic savings in the cost of testing the filters.

DISCUSSION

McDONALD: We wish to point out that we have at NRL identified suitable fluorescent tags for DOP to use with a fluorescence detection. A 100 watt mercury lamp and 90° collection optics with photon counting gives a suitable dynamic range for single filter challenge. Toxicity and upstream filter wetting should not pose a problem. The dynamic range of the challenge technique, about 10^5 , is straightforwardly extendable for challenge of tandem filters. This is more of a comment than a question.

KYLE: If your light scattering photometer has to have a minimum concentration of 10 particles per cubic centimeter downstream, you must have 10^9 particles per cc upstream if you require a protection factor of 10^8 .

McDONALD: You can read in an analog mode if you want to. However, you can just as easily count photons and count as long as you wish. This is much the same way you do it by particle counting.

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KYLE: I am concerned that when you wet the filter, you get an invalid test. I don't doubt you can read the signal; whether you've done a valid test is the question.

McDONALD: I see no reason why it should require wetting if you have a sufficient dynamic range to work with.

KYLE: Like 10^9 particles per cubic centimeter?

McDONALD: We are talking about developing a technique for testing one filter with 10^5 or 10^6 particles per cc. I don't know what this system will give in terms of discrimination against other particles.

KYLE: The need is to get by without upstream wetting. I have been looking at the numbers and it's doubtful that you get a valid test when you go to very high upstream concentrations.

SHAW: I have two questions. One, it seems to me that by using a sonic jet particle generator for the nebulization of fluorescent particles you make a lot of agglomerates. Have you checked to make certain you have monodisperse particles?

KYLE: You have to dilute down pretty thin in the solution.

SHAW: Because your concentrations are high, i.e., 10^9 , you must have a lot of agglomerates.

KYLE: I don't recall using 10^9 particles. There isn't a minimum concentration we must reach upstream. Because we are counting individual particles, we don't need very high concentrations. We plan to select the dilution on the basis of (a) the particle sizes that can be detected with a single particle spectrometer and (b) avoidance of agglomerates.

SHAW: All I'm saying is that it should be checked to make sure what the amount is. Otherwise, by just measuring all the collected particles in the solution, you may filter large numbers of agglomerates and think you are doing a very good job.

KYLE: We will have to make sure they are not agglomerated. I certainly agree with you.

VAN BRUNT: When you dissolve the filter, aren't you worried about scattering and ghost effects on liquid fluorescence? Light measurements of particulate-liquid mixtures are extremely difficult unless you have taken the above mentioned effects into account.

KYLE: I have not taken them into account. I have looked in a cursory fashion and have not found any problems, but it is possible that problems will occur.

MUSEN: Did you check to see how the dye degraded the DOP?

KYLE: No, we did not.

**A METHOD FOR COMPARING AIR VELOCITIES
BETWEEN BULK CARBON AND ASSOCIATED
TEST CANISTERS**

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Abstract

An important design element to insure that a test canister will adequately represent the condition of the bulk adsorbent is to provide for a uniform face velocity between the bulk adsorbent and test canister.

This paper describes a suggested test method for comparing the air velocity through bulk carbon with that through associated test canisters. The rationale behind the test are discussed as are some advantages believed to be offered by this test method. The test method was applied to a full scale adsorber unit at CVI and the test and its results are described.

While this procedure has been developed specifically for activated carbon adsorbers, it is believed to be adaptable to others such as silver zeolite, although the use of a different test gas may be required. Also, the possible extension of this test method to more general applications is suggested.

Introduction

It is generally known that the impregnated carbons traditionally used in HVAC systems for I_2 and CH_3I removal are subject to weathering, a phenomenon that causes a progressive decrease in the adsorber performance. Test canisters subjected to service conditions comparable to those for the bulk carbon are periodically removed, laboratory tested and the performance level of the bulk carbon is extrapolated. It is apparent that this approach will provide meaningful results only if the carbon in the test canister is weathered to the same degree as the bulk carbon. This condition will not be met unless the flow through the test canister is representative of the flow through the bulk carbon. Primarily, this requires that the face velocity for the test canister be essentially the same as that for the bulk carbon.

If the test canister face velocity is much less than that for the bulk carbon, weathering of the bulk carbon will be more advanced than that of the test canister and the test results will over estimate the performance of the bulk carbon. On the other hand, if the face velocity for the test canister is too much greater than that for the bulk carbon, a severe economic penalty may result since the bulk carbon may be replaced pre-maturely.

Several factors may influence the velocity ratio between the test canister and the bulk carbon including the following:

- a) Variation in average particle size
- b) Variation in packing density (particularly probable if the method of filling is different)
- c) Difference in static pressure drop across the test canister compared to that across the bulk carbon
- d) Difference in bed depth between the test canister and the bulk carbon.

Some experimental and analytical data regarding the effect of the first two factors above are included in Reference 1 and 2. The effects of the last two factors are apparent. In view of the above, it was considered desirable that some means be established for measuring the relative face velocity (velocity ratio) between the bulk carbon and that in the test canister.

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This paper describes a suggested test method that is considered to offer the following advantages:

- It provides a unique capability for measuring velocity ratios at several discrete, local points as opposed to average values over larger areas.
- It offers a means of direct measurement of the relative velocities without in anyway disturbing the system or the parameters being measured while the adsorber bed and test canister are operated simultaneously in the normal fashion.
- It does not rely on measuring the low face velocities associated with this type of system.
- It can be conducted utilizing equipment already required for filter testing.

Rationale Behind Test Method

The test method described herein uses a physical adsorption process to directly compare the velocities through the bulk carbon and the test canisters. For a physical adsorption process, the standard holdup equation relates the adsorbate breakthrough time to various system parameters. These parameters relate the delay time, τ , to the adsorbent mass, m , the dynamic adsorption coefficient, k , and the volumetric flow rate, V , as shown in Equation 1 below (see References 3 or 4 for one development of the delay equation).

$$\tau = \frac{mk}{V} \quad \text{Equation 1}$$

where

τ = the mean breakthrough time (min)
 m = the charcoal mass (g)
 k = the dynamic adsorption coefficient (cm³/g)
 V = the actual volumetric flow rate (cm³/min)

Equation 1 is generally valid for physical adsorption processes operating in the linear range of adsorption. By applying Equation 1 to both the bulk adsorbent and test canister and forming the ratio of the holdup times, a useful expression (see Equation 3A) is obtained from which the velocity ratios can be determined from known or measureable parameters. First, the mass "m" is expressed as density times volume.

$$m = \rho AL \quad \text{Equation 2}$$

Where ρ = charcoal density (g/cm³)
 A = adsorbent cross section (cm²)
 L = depth of adsorbent (cm)

Define v = superficial face velocity (cm/min)
 $v = V/A$

Subscript b — parameters relating to the bulk adsorbent

Subscript c — parameters relating to the canister

Substituting into Equation 1 and dividing the holdup time for the test canister by that for the bulk carbon

$$\frac{\tau_c}{\tau_b} = \frac{\rho_c L_c k_c v_b}{\rho_b L_b k_b v_c} \quad \text{Equation 3}$$

It is expected that the test described herein would be performed on newly installed carbon and that the carbon used in the test canisters would be nominally the same as that used for the bulk carbon so that $k_c = k_b$.

Therefore, using $k_c = k_b$ and rearranging

$$\frac{v_c}{v_b} = \frac{\rho_c L_c \tau_b}{\rho_b L_b \tau_c} \quad \text{Equation 3A}$$

Equation 3A provides the desired expression for velocity ratios as a function of known or measurable system parameters and was used for the analysis of the experimental results.

Equation 3A is a result of assuming that $k_c = k_b$. The effects of unaccounted for variations in the dynamic adsorption coefficients due to weathering are considered in the following. In a similar manner, the effects of unaccounted for variations in packing density were also examined.

Effect on Variations in "k" Values

If the test were to be performed on a system that has been weathered, it is believed that the test results would still be meaningful and that the net effect of weathering on the test results would be to make the test more sensitive to a poor flow distribution.

This can be illustrated by an example using a hypothetical system in which the face velocity through the bulk adsorber is somewhat higher than the test canister face velocity. It is assumed that the system is operated for a period of time prior to the test being performed so that the bulk carbon and the test canister are both weathered to some degree, which might result in a decrease in both k_c and k_b . However, since $v_b > v_c$, the bulk carbon would be weathered to a greater degree than the test canister carbon. It follows that when $v_b > v_c$ then $k_c \geq k_b$ and therefore from Equation 3 both act in the same direction to increase the breakthrough time ratio.

From the above, it is concluded that any foreseeable variation in the dynamic adsorption coefficient would tend to make the test results conservative by indicating a higher variation in velocity than is actually present. Furthermore, even this concern can be eliminated by testing the system when the adsorbent is installed. This is usually the appropriate time to conduct flow tests in any case.

Sensitivity of Results to Density Ratio

It is considered highly important that the packing density be uniform throughout the bulk carbon and consistent with the packing density in the test canister. However, it would be unrealistic to assume that this would always be the case.

The average packing density for both the bulk carbon and the test canister can be determined if the weight of adsorbent and the volume of the adsorbers are known. However, the accuracy of the estimated densities could be questioned. It is probably more instructive to consider the consequence of undetermined variations in the packing densities.

The most important consideration is in the limits of the variations. Granular carbon has been found to demonstrate a variation in packing density, for a given sample, of approximately $\pm 10\%$, depending on how well it is packed. However, given a $\pm 10\%$ variation in packing density, the resultant variation in velocity would be expected to be the order of 300% from minimum to maximum velocity, other conditions being the same (see Reference 2). Therefore, there is a cause-effect relationship between density ratio and velocity ratio but the relative magnitudes make the density ratio a second order effect.

One other consideration is relevant: the direction of variation in packing density ratio and velocity ratio is such that both act to influence the holdup time ratio in the same direction. For example, if $p_c > p_b$ then, other conditions being equal, $v_b > v_c$ due to a higher flow resistance in the canister. From Equation 3, it follows that the two ratios supplement each other to increase the holdup time ratio τ_c/τ_b . Since τ_c/τ_b is measured and used in Equation 3A to estimate the velocity ratio, the test would be slightly more sensitive to variations in velocity due to non-uniform packing (considered to be the major cause of non-uniform flow).

Test Equipment

The test method incorporates a pulse injection technique in which the fluorocarbon refrigerant R-12 was injected upstream of the filter. R-12 was used in this test for convenience because it could be detected using equipment utilized for the standard in-place leak test for carbon adsorbers. Other adsorbates could undoubtedly be used. The mean breakthrough time for the bulk carbon and the canister were measured by sampling the air downstream of each component individually and recording concentration versus time.

The bulk adsorber/test canister physical configuration and general flow pattern are illustrated in the plan view in Figure 1. The adsorber/canister system was a full scale HECA® filter and CANSET® canister manufactured by CVI.

The test apparatus is illustrated schematically in Figure 2 with a photograph of the inlet air duct, filter housing and gas chromatograph equipment included as Figure 3. The HECA® unit was designed for 42.5 m³/min flow. However, all testing was done with laboratory fans providing an aggregate flow of 38.2 m³/min.

A Valco Model 1200 Halocarbon Monitor gas chromatograph with a Varian A-5 strip chart recorder were used to monitor the R-12 breakthrough downstream of both the bulk adsorbent and the test canister.

Air flow rates were measured with a pitot tube located upstream of the filter unit as illustrated in Figure 2. R-12 was injected upstream of the fan as also illustrated in Figure 2.

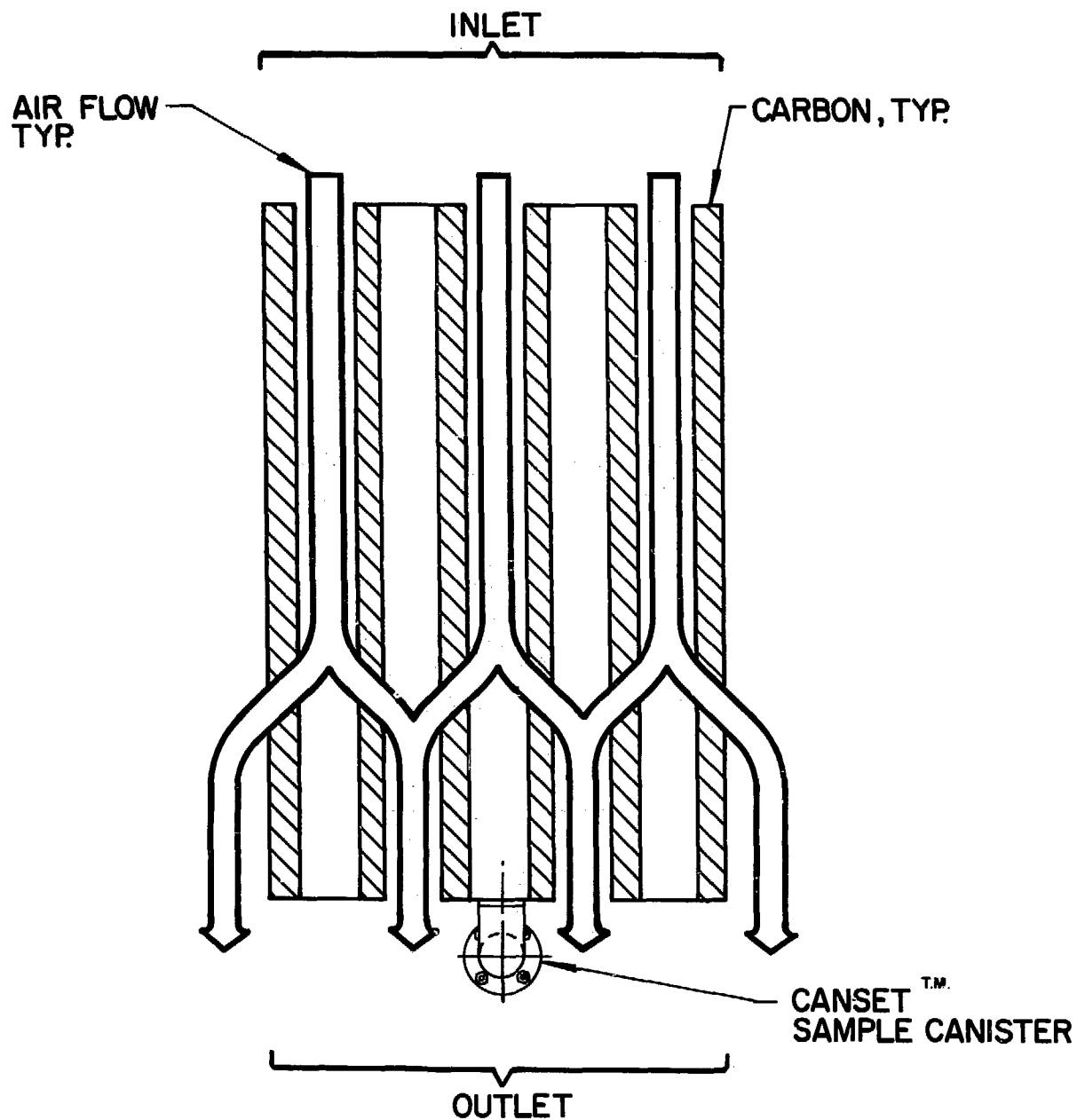


FIGURE 1. CROSS SECTION OF HECA[®] ASSEMBLY AS USED IN THE BREAKTHROUGH TEST

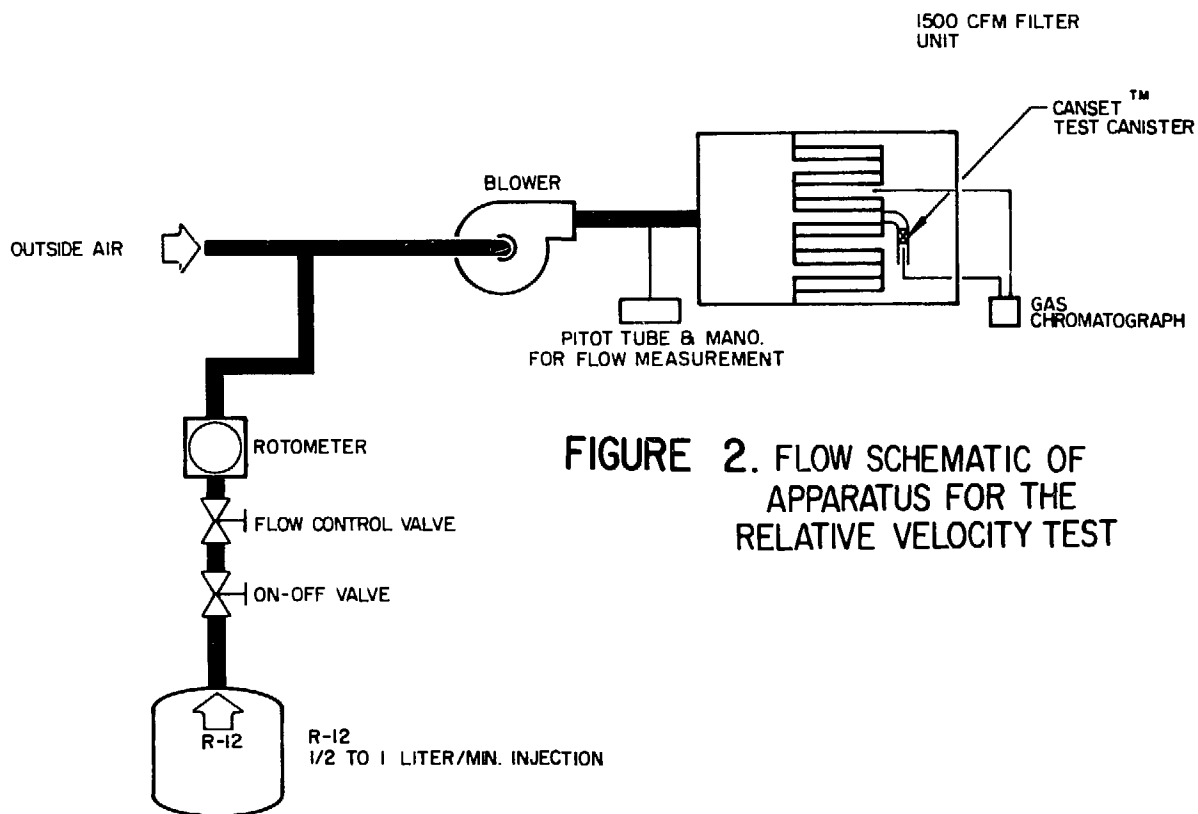


FIGURE 2. FLOW SCHEMATIC OF APPARATUS FOR THE RELATIVE VELOCITY TEST

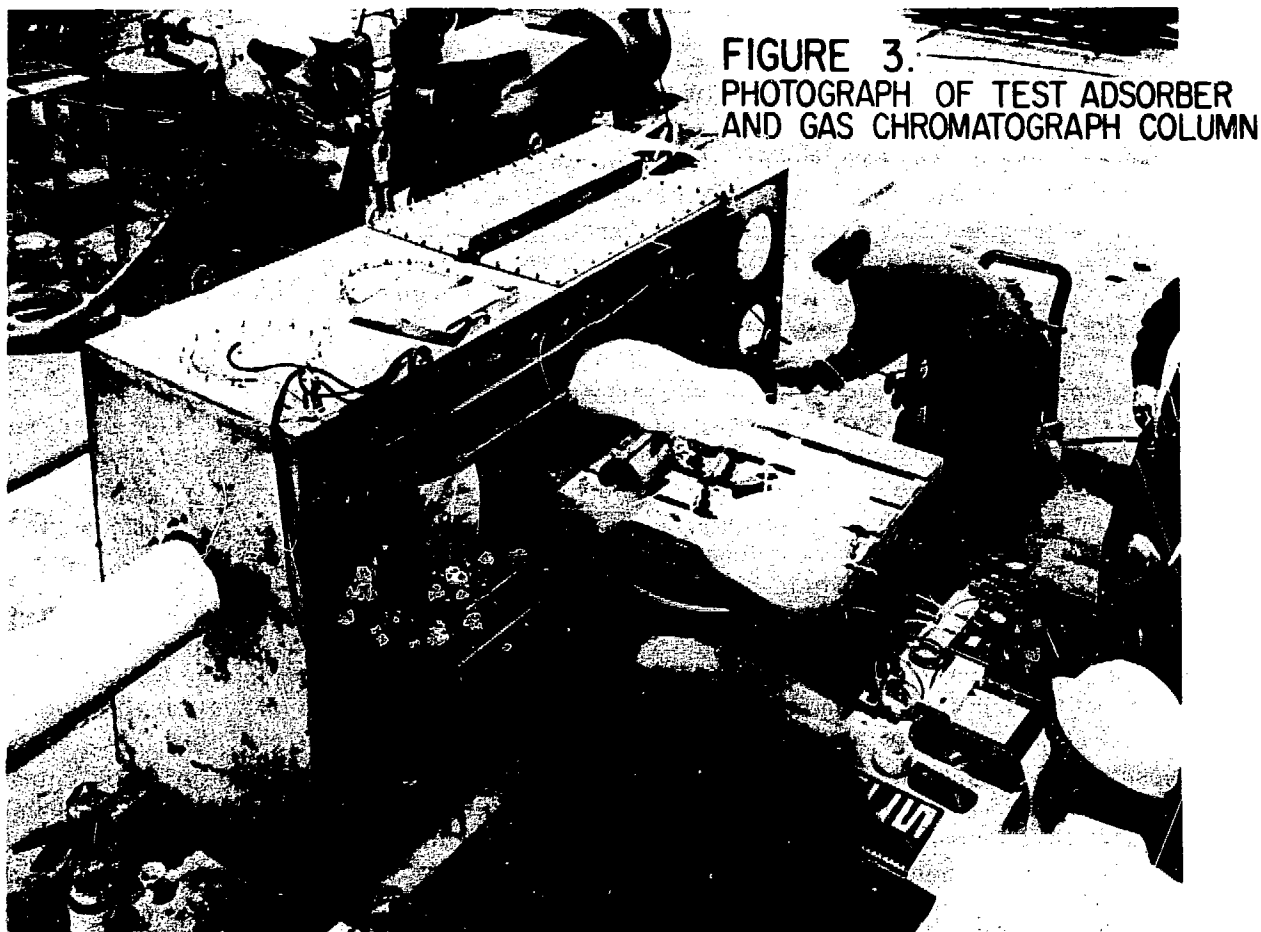


FIGURE 3. PHOTOGRAPH OF TEST ADSORBER AND GAS CHROMATOGRAPH COLUMN

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Test Procedure

Prior to the first test, the bulk adsorber beds and the test canister were filled with carbon using a gravity packing method.

In subsequent testing, the system was purged and the effluent examined for R-12 periodically until it contained no detectable R-12. When the system was clean, the R-12 bottle was prepared for injection by adjusting a throttling valve to produce the desired flow rate of R-12. The R-12 was discharged to the atmosphere in such a manner that it would not be drawn back into the test system in subsequent runs. The R-12 flow rate was then terminated by means of an isolation valve until the beginning of the test.

A velocity traverse was performed for the inlet duct and the centerline velocity coefficient was determined. The G. C. was set up to draw a sample of gas and analyze it once every two minutes. A manual switching valve was provided so that the sample could be alternately drawn from downstream of the bulk carbon and downstream of the test canister. By using a single instrument to monitor the effluent from both the bulk carbon and the test canister, any instrument error would be common to both and the effect of any error would therefore be minimized. The characteristic breakthrough behavior of R-12 under test conditions resulted in pulses sufficiently broad that sampling each stream once every four minutes provided a very adequate description of the effluent pulse. These pulses generally took from 2 to 4 hours to elute, depending primarily on air temperature.

The G. C. was started approximately one hour prior to the test in order to allow the G.C. to stabilize. After the G. C. was stabilized the fans were started and the average duct velocity determined using the pitot tube to measure the centerline velocity and applying the centerline coefficient determined previously. The volumetric flow rate was calculated from the average duct velocity times the duct cross sectional area.

With the air flow established, the R-12 isolation valve is opened to initiate the R-12 flow, the strip chart recorder started and the start time indicated. As indicated previously, the sample lines were manually switched to provide alternate readings from downstream of the test canister and from downstream of the bulk carbon. The test was generally continued until most of the R-12 had been eluted.

Results and Discussion

The results of the R-12 breakthrough tests are illustrated in their most basic form in Figures 5, 6, 7 and 8 which show the G. C. output as a function of time for both the bulk carbon and the test canister effluents. The G. C. output is shown as peak height in percent of full scale.

Data from Test Number 2 are plotted in Figure 5. Generally, data in Test Number 2 were taken alternately from downstream of the bulk adsorber and the test canister. However, a few data points were taken in succession from the same sample line to confirm that switching from one sample line to the other did not introduce any unexpected effects on the results.

This was done, for example, at 154 minutes into the run when two measurements were taken in succession downstream of the canister followed by measurements taken in succession downstream of the bulk carbon bed. This procedure was repeated again at 200 and 222 minutes into the run. Whether or not the sample lines were switched made no observable difference in the results as evidenced by the continuity of the traces when consecutive readings were made.

From Figure 5 it can be seen that the elution curves from the bulk carbon and the test canister are very similar in shape and nearly simultaneous, indicating that the flow through each is very comparable. These results can be quantitatively evaluated from the mean breakthrough times from each. The mean breakthrough times were evaluated by graphically integrating the area under the curves as plotted in Figure 5. The resultant mean breakthrough times are also shown in Figure 5. For Test Number 2, the mean breakthrough time for the test canister preceded that for the bulk carbon by 3.6%, indicating the velocity through the test canister is higher than that through the bulk carbon. The analysis of the data is presented later. Results, including adjustment for path lengths, are tabulated in Table 1.

The results of Test Number 4 are illustrated in Figure 6. As in Test Number 2, the test canister was in normal operation with no artificial flow impediment. The mean breakthrough times were evaluated as for Test Number 2, from which it was determined that the mean breakthrough time for the test canister preceded that for the bulk carbon by 8.9% (see Table 1 for complete results).

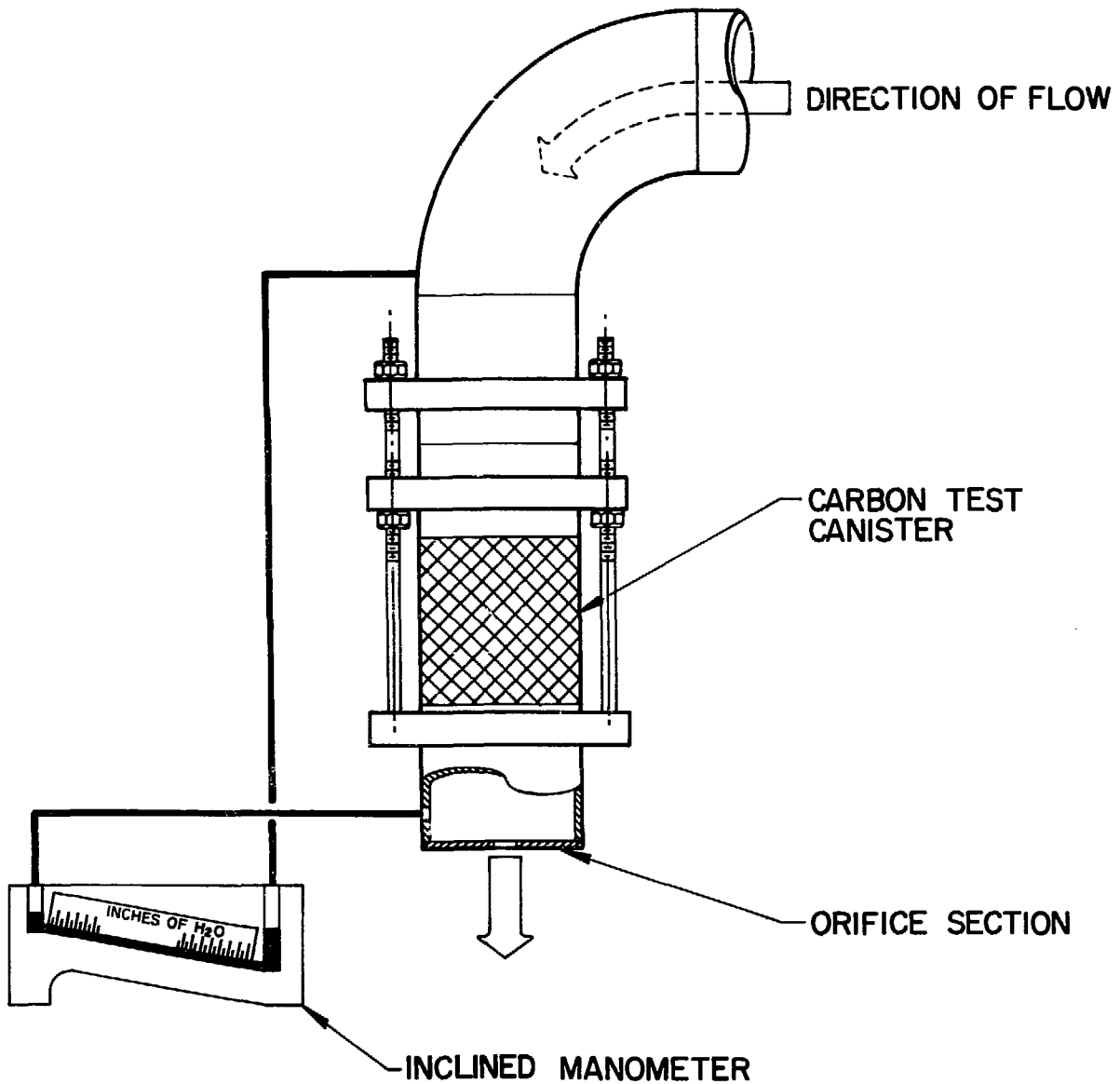


FIGURE 4. DIFFERENTIAL PRESSURE MEASUREMENT FOR CARBON TEST CANISTER

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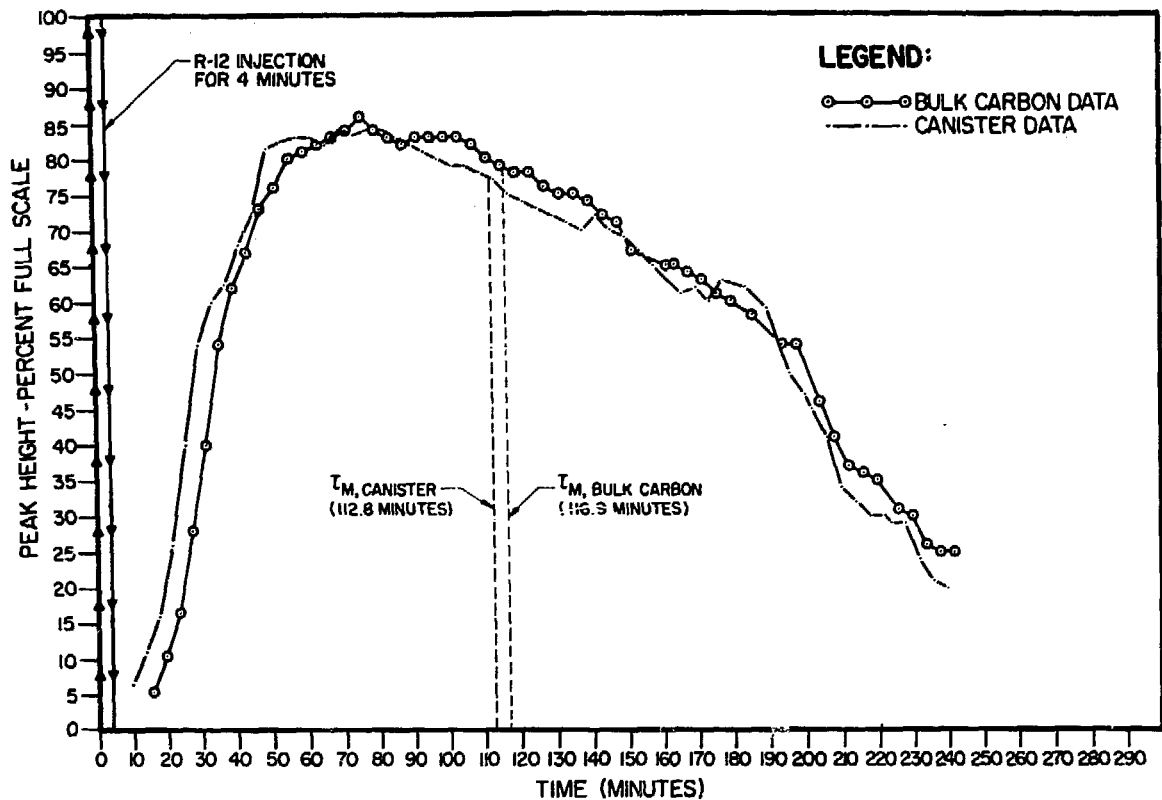


FIGURE 5. BREAKTHROUGH DATA FOR TEST NUMBER 2

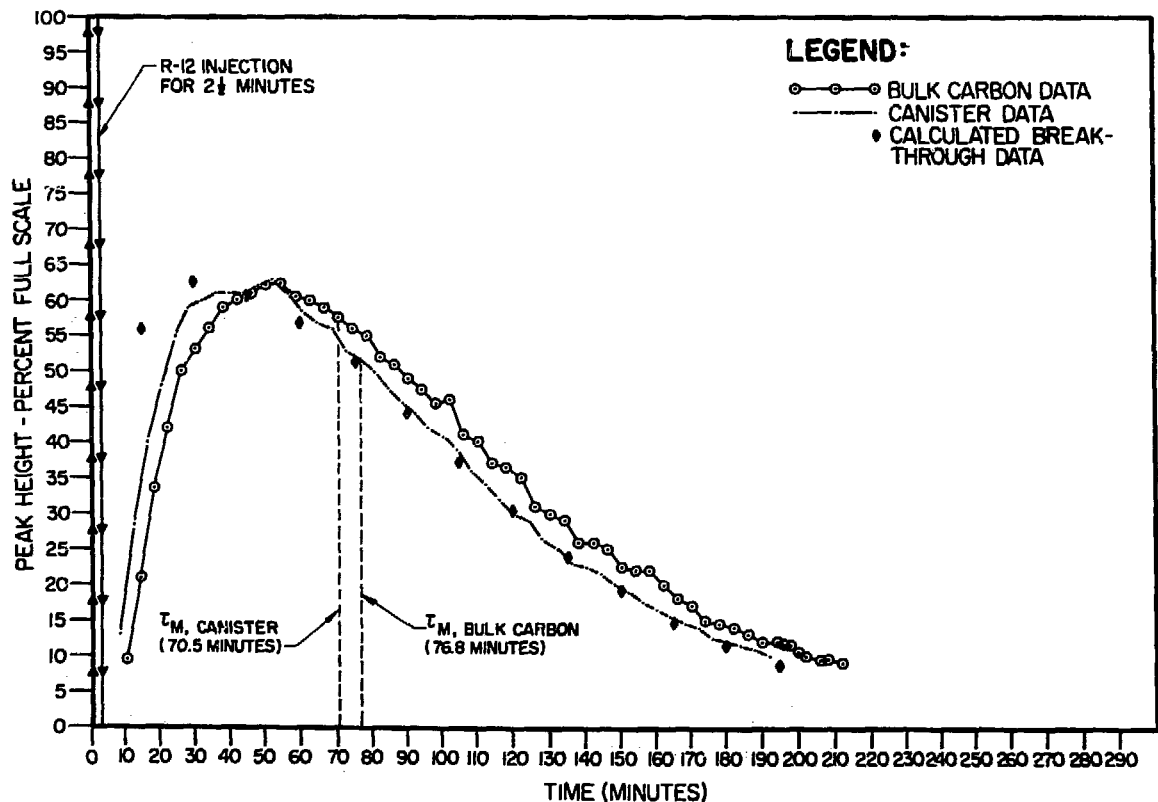


FIGURE 6. BREAKTHROUGH DATA FOR TEST NUMBER 4

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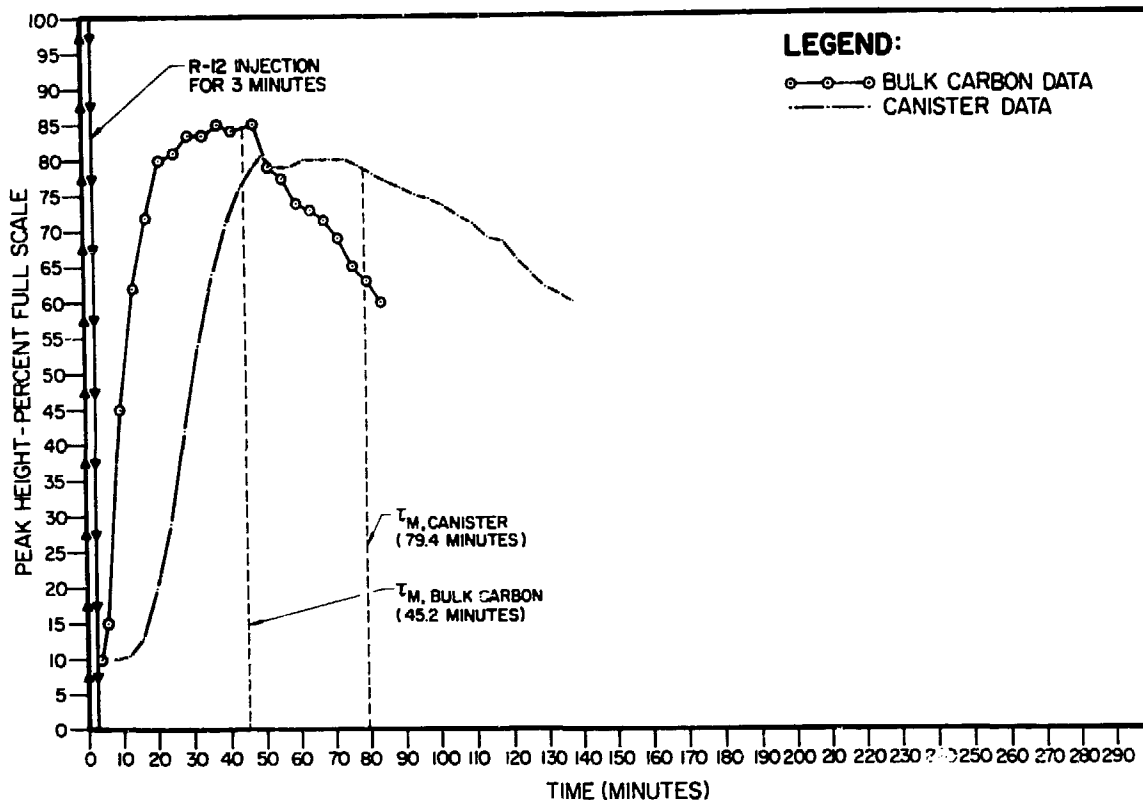


FIGURE 7. BREKTHROUGH DATA FOR TEST NUMBER 5

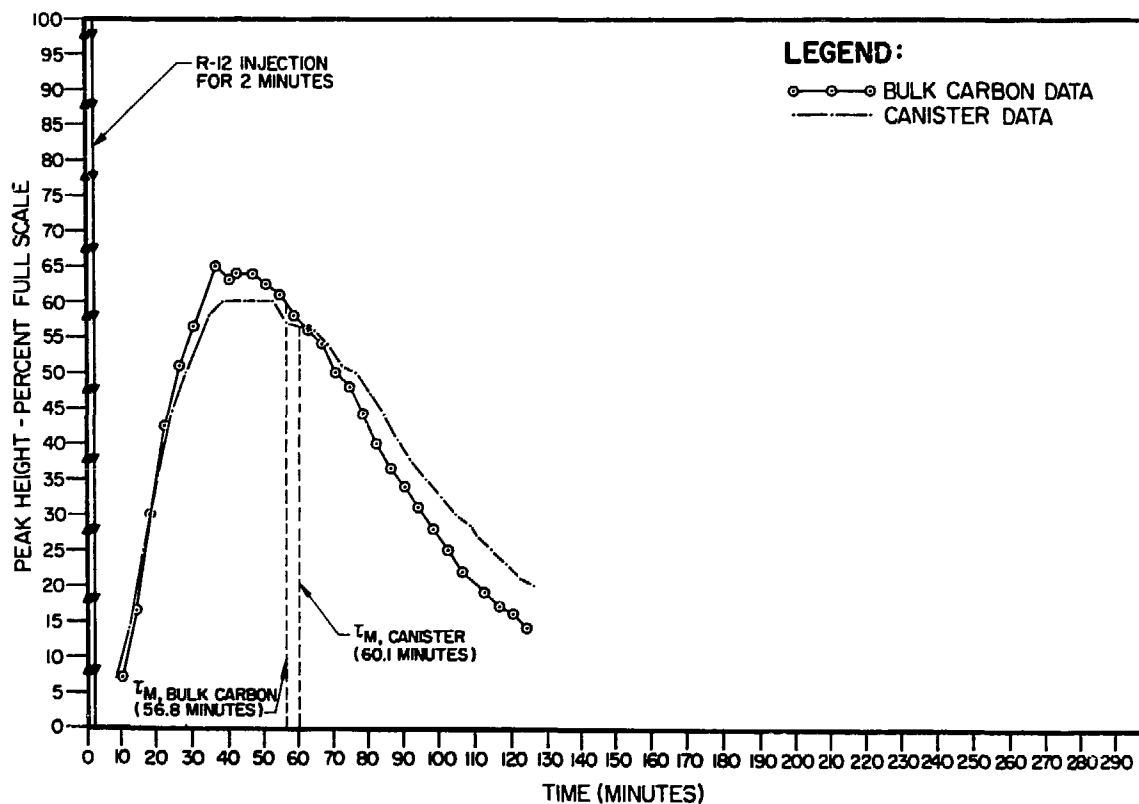


FIGURE 8. BREAKTHROUGH DATA FOR TEST NUMBER 7

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The elution curves from Test Numbers 2 and 4 obviously do not exhibit the symmetrical pulse customarily observed for linear, physical adsorption systems. However, this was not too surprising because of the extremely small bed depth (nominally 2 inches). Nevertheless, it was decided to calculate the shape of the breakthrough curve, assuming a linear, physical adsorption process, and to compare the results with the observed breakthrough curve.

The calculations were performed using a physical adsorption model presented in Reference 5. The dynamic adsorption coefficient and height of transfer unit (HTU) were estimated from the test canister data of Test Number 4 to be $32,490 \frac{\text{cm}^3}{\text{g}}$ and 1.65 cm., respectively. These empirically determined parameters were used to calculate the breakthrough curve. Therefore, the calculated breakthrough curve was forced to reproduce the mean breakthrough time and one point on increasing and one point on the decreasing curve. However, as seen in Figure 6, the shape of the entire calculated breakthrough curve agrees well with the experimental data, thereby increasing confidence in the assumption that the adsorption can be characterized as a linear, physical adsorption process.

To further evaluate the test method, the flow through the test canister was artificially decreased to see whether the breakthrough curve for the bulk adsorber preceded that for the test canister, as predicted by Equation 3A. In Test Number 5, the flow through the test canister was partially blocked by taping off its discharge nozzle and progressively enlarging an opening in the tape until the test canister flow was approximately $\frac{1}{2}$ the normal flow. The relative flow was estimated by monitoring the pressure drop across the canister carbon bed only (see Figure 4) as the downstream flow resistance was developed. The flow was considered to vary as the ΔP to the 1.22 power (from carbon vendor data). The pressure differentials for the bulk carbon and the test canister carbon were not compared since it was considered that this could lead to substantial error. Instead, the pressure drop through the test canister carbon was measured both before and after adding the flow resistance, while the bulk carbon flow was maintained essentially constant. The velocity through the bulk carbon and the test canister are assumed to be approximately the same prior to throttling the test canister flow. Therefore, the ratio of the test canister velocity, after throttling, to that before throttling would be approximately the same as the ratio of the test canister velocity to bulk carbon velocity, after throttling. The results of the test, as can be seen in Figure 7, indicate that the bulk carbon eluted R-12 much more rapidly than the test canister, as it should. It will be noted that the test data were taken for a period of about 140 minutes and that neither bulk adsorber nor the test canister assembly had completely eluted the R-12 during this time interval. However, the data taken clearly demonstrates that the mean breakthrough time is closely related to the air velocity. The best estimates of the breakthrough times, shown in Figure 7, indicate the mean breakthrough time for the test canister to be 56.9% that of the bulk carbon (see Table 1 for complete results). Using the test canister carbon as a flow meter as described above, the canister velocity was estimated to be 51.8% that of the bulk carbon. This was considered to be satisfactory agreement, especially considering the approximate nature of the flow indication from the pressure differentials (considered to be probably less accurate than the relative breakthrough method).

Data from Test Number 7 are presented in Figure 8. This was another normal run in the sense that flow through the test canister was not artificially decreased as was done in Test Number 5. Again, the R-12 was eluted in very nearly the same time from both the bulk adsorber and the test canister. However, the mean breakthrough time for the canister lags that for the bulk carbon by about 5.5%.

Results for Test Numbers 1, 3 and 6 were not reported. Test Number 1 was a shakedown test to determine the appropriate quantity of R-12 to be injected, the approximate breakthrough time, etc. Test Numbers 3 and 6 were aborted due to an inadvertent change in total blower flow rate during the course of the test and due to an artificially induced flow imbalance, respectively.

Summary of Results

As indicated previously, the mean breakthrough times were evaluated by graphically integrating the area under the breakthrough curves. The resultant mean breakthrough times are shown in Figures 5 thru 8. The ratio of the mean breakthrough times are also included in Table 1. The density ratio, $\frac{\rho_c}{\rho_v}$, was taken to be unity. The ratio of the test canister bed depth to the bulk carbon bed depth was determined to be approximately 0.97. The mean breakthrough time ratios and the bed depth ratio were used in Equation 3A to calculate the relative velocities presented in Table 1.

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From Table 1, the canister velocity was greater than the bulk carbon velocity in two out of three tests (not including Test Number 5). The maximum indicated difference was 8.3%.

The locations of the sample points downstream of the bulk carbon were not the same for all tests. The relatively minor variations in the measured velocity ratios probably reflect several factors as; 1) both the bulk adsorber and test canister were drained and refilled at least once during the test program, 2) possible variations in bulk carbon velocities as the sample point was moved from one point to another and 3) the reproducibility of the test (the latter was not directly determined).

Conclusions

The results and observations of this effort indicate that the test method can be used, as intended, to measure the ratio of the superficial face velocity for the test canister to the superficial face velocity for the bulk carbon. This velocity ratio may then provide valuable information to establish the relative effectiveness of the test canister.

A basis of the test was the assumption of a linear, physical adsorption system. The shape of the breakthrough curve, as calculated from an analytical model, closely reproduced the shape of the observed breakthrough curve. This increased confidence that the system satisfied the above assumption.

The test method was checked directly by artificially decreasing the superficial face velocity through the test canister to approximately 50% of full flow by means of an external flow resistance. The results (velocity ratios) from the breakthrough test method agreed with results obtained from measured pressure drops to within 3.4%.

Although it was beyond the scope of this effort to demonstrate, it is believed that this test could be extended to measure the flow uniformity through the bulk carbon. By placing two or more pickups at various locations on the outlet screen, this test method would offer a unique means for determining relative velocities at a number of discrete points, on a local basis, rather than less meaningful average velocities over a much larger area.

The above tests demonstrated the flow through the test canister, CANSET[®], to be representative of the flow through the HECA[®] bulk carbon.

Acknowledgements

Our appreciation is expressed to Fred Leckie who provided and operated the G. C. and associated equipment. Fred's expertise in this area greatly facilitated the testing.

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| Test
Number | Ratio of mean
breakthrough
times
$\frac{\tau_b^{(1)}}{\tau_c}$ | Ratio of
adsorber
depths
$\frac{L_c}{L_b}$ | Ratio of
flow
velocities
$\frac{v_c}{v_b} = \frac{L_c}{L_b} \times \frac{\tau_b}{\tau_c}$ | Velocity
ratio from
canister ΔP
$\frac{v_c}{v_b}$ |
|------------------|---|---|--|--|
| 2 | 1.036 | 0.97 | 1.005 | — |
| 4 | 1.089 | 0.97 | 1.057 | — |
| 5 ⁽²⁾ | 0.569 | 0.97 | 0.552 | 0.518 |
| 7 | 0.945 | 0.97 | 0.917 | — |

(1) Subscript b refers to bulk carbon, c refers to canister.

(2) Introduced artificial flow resistance downstream of canister and estimated velocity ratio both from R-12 breakthrough and from ΔP across canister.

TABLE 1 Summary of Breakthrough Tests

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DISCUSSION

BELLAMY: Why is there a difference of 3% in the bed depths between the canister and the bulk bed?

PARISH: The bulk carbon was designed to be approximately 3% deeper to provide slightly more flow resistance through the bulk carbon relative to the flow canister as a conservative measure. The tests referred to were conducted in 1975. In recent years, we've gone to a little deeper bed relative to the canister as this helps to ensure that the flow resistance to the test canister, everything else being the same, will be less and therefore that there will be a slightly greater flow through the test canister.

BELLAMY: And your newer designs will have more conservatism?

PARISH: Yes.

BELLAMY: How much more?

PARISH: I'd say, on the average, an additional one or two percent.

DEMPSEY: Is the purpose of all this to be able to go back to the lab with the samples and analyze them for iodine sorption capability?

PARISH: Yes.

DEMPSEY: Our friends from other countries who have been attending these meetings for the last ten to twenty years have been telling us repeatedly that we should make iodine tests in the field because the moisture content of the carbon makes a drastic difference in the results, and we don't know what the moisture content is. It seems like such an easy step to go from what you're doing to making the iodine test in the field, perhaps on a limited fraction of the bed with a micromicrocurie of radioiodine, if you'd like, so that we could get some real field data. Is anyone considering that?

PARISH: I don't believe I am the proper one to answer that question. I would point out that although there is an interest in testing in the field in the manner you suggest, you still would want to assure yourself of an appropriate velocity distribution between various points in the adsorber bed. Perhaps you would not need a test canister if you were testing in the field. Nevertheless, I think you would still want to assure yourself that you had a proper flow distribution through the bed, at least within the $\pm 20\%$ called for in Regulatory Guide 1.52. To that extent, this test, or some other type of flow distribution test, would still be needed.

DEMPSEY: Am I correct in assuming that the proportion of flow through the canister to that through the whole bed is one to a thousand?

PARISH: Something on that order, yes.

DEMPSEY: I am suggesting that by introducing the radioactivity into a very limited area of the bed, knowing exactly what the relative flow rate is, we could use extremely small amounts of radioiodine: one thousandth or a hundred thousandth less than what would have to be used for testing the entire bed.

PARISH: You mean, if you were injecting only upstream of the test canister?

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DEMPSEY: Of that exact area. Perhaps there's an idea here that we could get the Nuclear Regulatory Commission to consider with us.

PARISH: I think that's an interesting concept; I hadn't heard it before.

RIVERS: You were speaking of ASTM D2804 which is in the process of establishing test conditions for the canisters after they are taken back to the laboratory as well as before they are installed. It is essential that we first have a baseline measurement of the carbon that is installed in the filter system by sampling and evaluating it in the same type of canister. Then, after service, the test canisters can be brought back to the laboratory and put through at least one of the test procedures that the original carbon batch was subjected to. At the moment, the feeling is that the appropriate test procedure should be one that couldn't possibly be done in the field because it is to be conducted at 80°C and 95% RH. Of course, that condition doesn't necessarily exist in the field system. Therefore, we have a problem in deciding what is to be the most desirable set of test conditions in the field or laboratory.

KOVACH: I have some questions and comments. I think just taking the mean breakthrough time is not really an accurate way of deciding how uniformly flow is distributed between the main bed and the test bed. Differences in the tail can be very important and the entire breakthrough curve is needed to show whether you have uniformity of flow or not. Although you didn't stress it, your curves match quite reasonably on most of the tests. I think that whoever is using this test procedure may be well warned to evaluate the exact differences when they develop the full breakthrough curve and to evaluate the entire region on a statistical basis. I don't think we always develop enough data to say that two samples are really identical. If we start using different canister bed depths to counteract potential differences in the adsorber when we run through the test conditions, we may be getting ultra-conservative and this can be just as bad from an economic standpoint when considering the frequency of changing out adsorber beds.

PARISH: I believe, when there is adequate flow distribution, that the mean breakthrough time is sufficient to establish the flow distribution and that is what it is that we are looking for. I will agree that differences in the tail of the curve or, for that matter, in the initial rise reflect changes in the height of a transfer unit which might be related to particle size or some other factor which has to do with the mass transfer in the adsorbent. But I stick to the claim that if you match the mean breakthrough times, based on the conditions that are outlined in the report, that you can establish whether or not you have uniform velocity through the adsorber. Other information can be obtained by examining the entire breakthrough curve.

KOVACH: The only comment I have is that Edgewood Arsenal uses the R-12 method to evaluate adsorber life. Identical mass-produced adsorbers are tested routinely with R-12 and measured for pressure drop. Their results show very limited, or no direct, correlation between pressure drop and R-12 values, both of which should relate to air velocity through the bed. Therefore, neither pressure drop nor R-12 mean number relates to flow rate through the bed. Do you have any comments on this observation?

PARISH: The reason we did not use ΔP measurement as the primary indication of flow distribution is because there are too many other variables such as packing density and particle size which affect the flow resistance and therefore ΔP . Thus I would not be surprised at the lack of correlation you noted.

PERFORMANCE EVALUATION

OF AN

18,000 CFM FILL-IN-PLACE
ADSORBER UNIT

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Abstract

Under a commercial contract MSA was required to assemble and factory test a complete 18,000 CFM air filtration train before shipment to the plant site. The filter train tested was approximately 14 ft. wide by 9 ft. high by 31 ft. long and weighed an estimated 33,500 pounds. It contained four stages of filtration — moisture separators (prefilters are substituted for this test), upstream and downstream HEPA filters and a Type III (Fill-In-Place) Sorber. Filter train design followed the guidelines set forth by ANSI N509-1976 and the tests were conducted under factory controlled conditions per ANSI N510-1975. (1), (2)

In addition to the ANSI N510 tests, an extensive program was conducted to determine "slot velocities", bed packing density, bed flow distribution, and effect of filling methods on particle size and sorber performance for the Type III High Efficiency Sorber.

The purpose of the testing was to demonstrate that after filling, the Type III sorber will function as designed for contaminant removal and thereby qualifying the fill method and Type III sorber design. Additional criteria were to develop and establish suitable testing procedures which can be implemented to demonstrate functional performance on any Type III sorber unit.

I. Introduction

In commercial power reactors, filtration units are used in various air-decontamination systems which prevent release of fission products to the atmosphere and/or cleanup for "in-house" environments. With heavy emphasis from the public, NRC* and other agencies the trend is to insure that these components are reliable and most of all that the units meet their design performance.

The purpose of this paper is twofold. The first is to demonstrate that a Type III sorber unit can meet all the applicable tests as outlined in ANSI N510 when the tests are conducted at the factory. The second purpose is to develop some tests which are not currently available to hopefully insure that the Type III sorber

*Nuclear Regulatory Commission.

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is reliable. It is erroneous to assume that these units when designed to current specifications and installed accordingly are adequate because manufacturing techniques, installation procedures, method of loading, size of unit and other factors can significantly affect the performance.

It should be realized that for most filtration systems the desired efficiency is 99.95% or higher. It appears paradoxical that when one reviews the requirements of HEPA filtration vs. gaseous filtration that the requirements put forth on HEPA filtration performance far exceed any performance tests conducted on the Type III sorber and still we see some significant HEPA filter rejection rates by ERDA* Q.A. stations. Furthermore, almost any person who deals with sorbent testing and bed filling techniques is aware of pitfalls in this area. It is not unusual to note significant flow variation and/or flow resistance because of bed packing density which, of course, would result in poor overall performance of the unit.

II. Test Facilities

A schematic of the test facility is shown in Figure 1. The air enters the filtration plenum through a rectangular inlet located in the top of the plenum approximately 13 feet by 1-1/3 foot. The air then travels through a mixing plenum approximately 14 feet wide by 8 feet high by 3 feet deep and continues through the prefilter, heater and HEPA filter sections of the plenum. The air then enters the Type III sorber and after-HEPA filter section and exits at the filtration plenum through an outlet duct approximately 13 feet by 1-1/3 foot at the top of the plenum. The duct transitions to a 45 inch diameter section where the static pressure taps are located. The 45 inch diameter duct is connected to a 40 horsepower centrifugal fan equipped with a flow control damper. Figure 2 depicts the test facility.

III. ANSI N510 Testing

Procedures

All testing was performed in accordance with procedures outlined in the ANSI N510 standard. Since these procedures are standard throughout the industry, they are not detailed herein.

Air Flow Capacity Test

An air flow capacity test was run in accordance with Section 8.3.1 of ANSI N510. The pitot tube traverse method was used to generate the necessary data for establishing the centerline velocity pressure vs. volume flow rate curve shown in Figure 3.** The manual damper was adjusted to give a centerline velocity pressure of .20, i.e. 18,000 CFM.

As an additional check to substantiate flow, the pressure drop of each HEPA filter in the upstream HEPA filter bank was determined after the unit test was completed. The resistance of

*Energy Research & Development Administration.

**ANSI N510, Para. 11.4.

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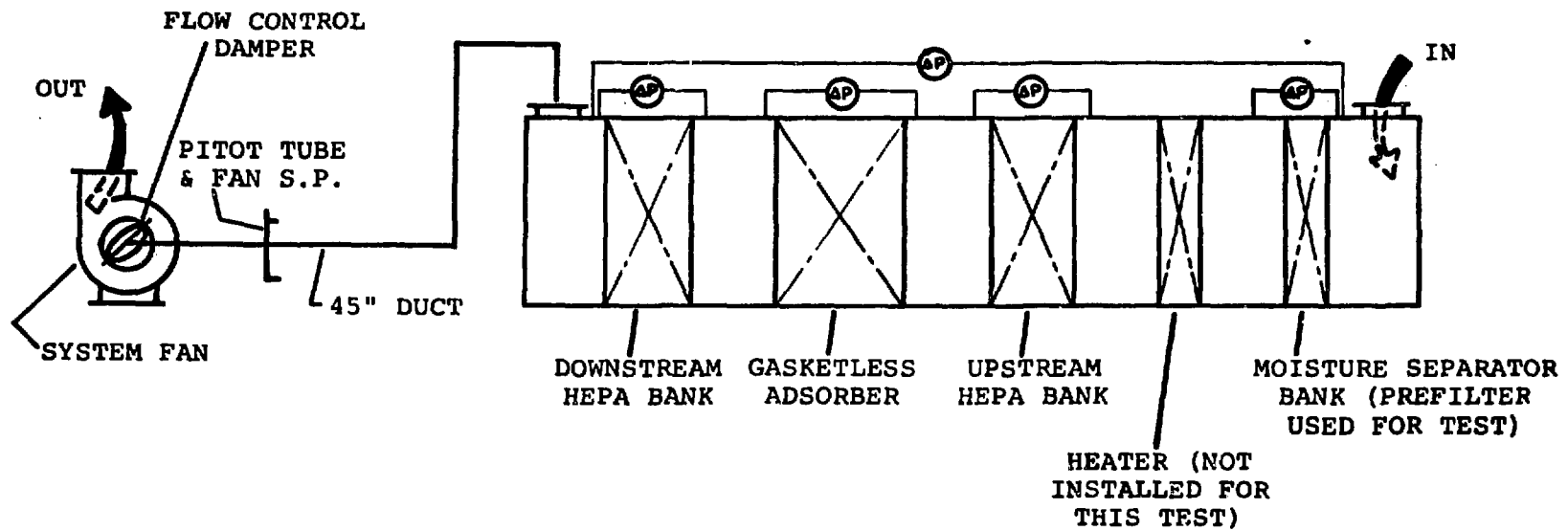
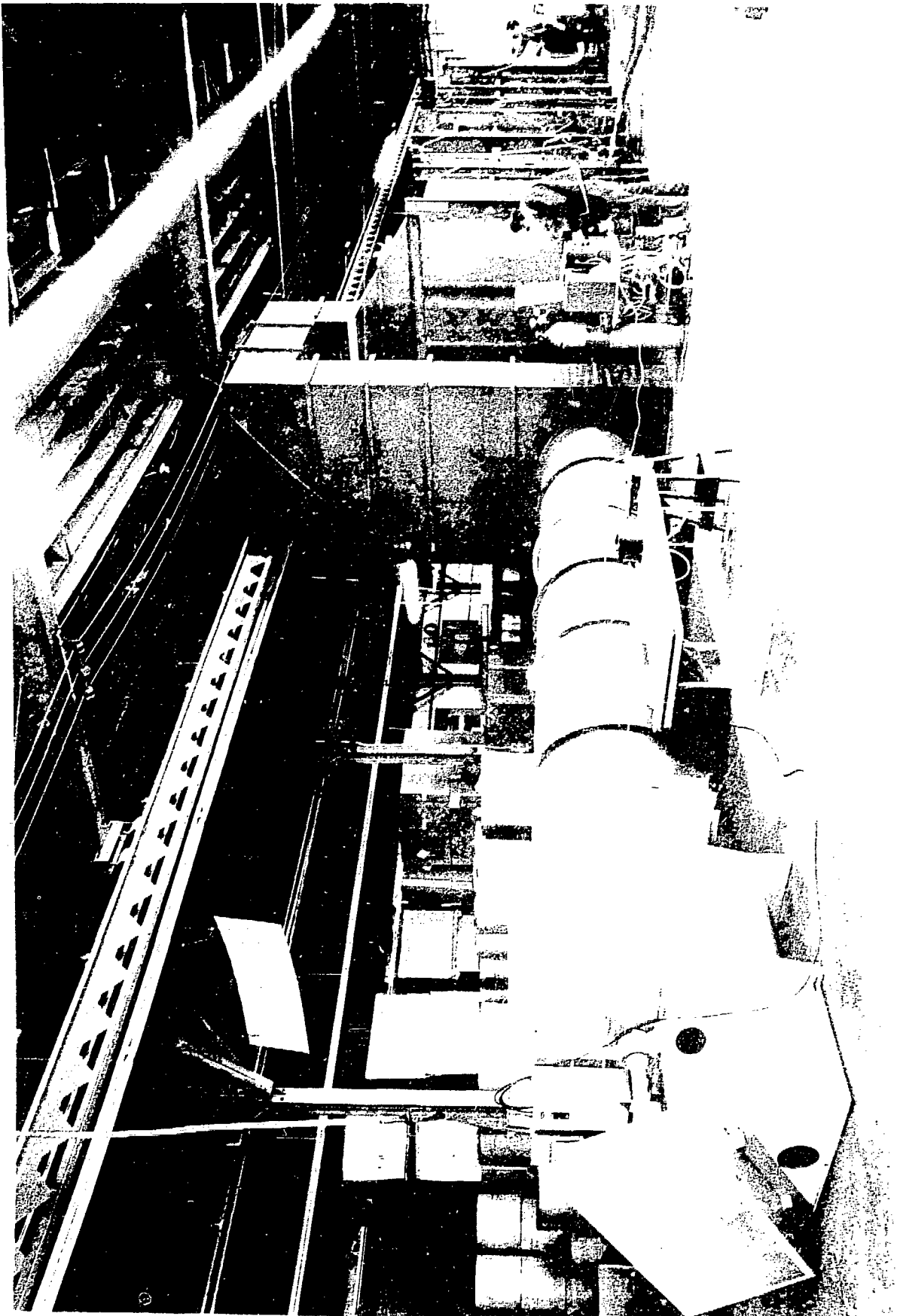


Figure 1. System flow schematic.



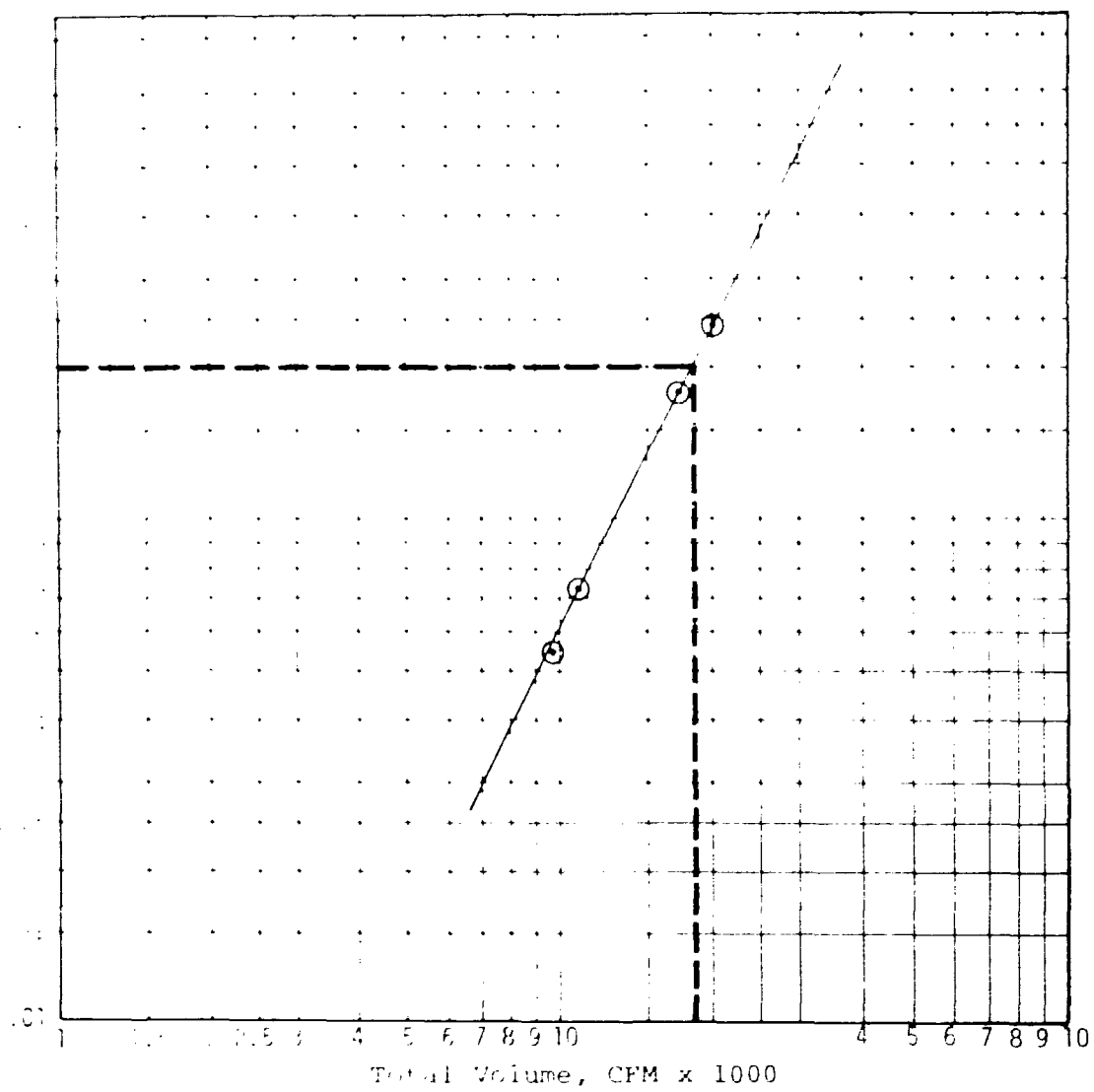


Figure 1. Volume flow calibration curve.

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each filter was obtained on the production tester* (DOP machine) for each filter at 1000 CFM and averaged, see Table I. The average resistance was found to be 0.84 in. W.G. for 18 filters (18 filters x 1000 CFM/filter = 18,000 CFM). The average bank resistance obtained during unit testing was 0.83 in. W.G., a variance of 1.2% which indicated the volume flow as measured was correct.

Table I Upstream HEPA resistance —
production tester at 1000 CFM.
Resistance, inches W.G.

| | | | | | |
|-----|-----|-----|-----|-----|-----|
| .88 | .82 | .84 | .83 | .82 | .83 |
| .87 | .84 | .81 | .85 | .90 | .84 |
| .82 | .78 | .88 | .77 | .86 | .86 |

Note: Each block represents filter location in the bank.

Average = 0.84 in. W.G.

Air Flow Distribution

An air flow distribution test was performed across the upstream HEPA filter bank in accordance with ANSI N510, Paragraph 8.3.2. The data collected during this test is shown on Table II.

Table II Air flow distribution velocity.

| Velocity, FPM | | | | | |
|---------------|-----|-----|-----|-----|-----|
| 280 | 280 | 285 | 295 | 280 | 280 |
| 280 | 275 | 285 | 295 | 280 | 280 |
| 275 | 265 | 285 | 295 | 280 | 275 |

Note: Above readings corrected for anemometer calibration. Each block represents filter location in the bank.

Average velocity = 281 FPM
Allowable range - 225 to 337

*Penetrometer, Filter Testing, DOP, Mil-Q107-Doc. No. 136-300-175A, U.S. Army, Edgewood Arsenal, Maryland. (3)

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The velocities were taken upstream of the face at the center of each HEPA filter with an anemometer. The average velocity was calculated using the equation:

$$\bar{V} = \frac{\sum_i^n V_i}{n} * \quad (1)$$

where $\bar{V}$ = average air flow velocity through the housing

V_i = individual anemometer readings

n = number of anemometer readings

The average velocity is 281 FPM. The allowable range of all readings per ANSI N510 is $\pm 20\%$ or 225 to 337 FPM. The actual readings are within $\pm 6\%$ of the average velocity.

An additional check of volume flow was performed by taking the superficial flow area of each HEPA filter times the number of HEPA filters times the average velocity.

$$C = A \times N \times \bar{V} \quad (2)$$

where C = volume flow, ft.³/min.

A = face area of a HEPA filter = 3.52 ft.²

N = number of HEPA filters = 18

$\bar{V}$ = average velocity, ft./min.

$$C = 3.52 \times 18 \times 281$$

$$C = 17,804 \text{ CFM}$$

This volume flow is within 1.1% of actual.

Sorber Residence Time Calculation

The Type III sorber residence time was calculated from the equation:

$$T = \frac{nNt(A-b)**}{28.8 Q_s} \quad (3)$$

where T = residence time, sec.

n = number of cells in system = 1 for Type III sorber

N = number of sorbent beds per cell = 18

t = thickness of adsorbent beds = 4 in.

A = gross area of all sorbent-bed screens of one cell = 159,840 in.²

* Per ANSI N510-1975, Para. 8.3.2.

**Per ANSI N510-1975, Para. 8.3.3.

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b = area of baffle, margin, and blank area
of all sorbent-bed screens of one
cell = 29,736 in.²

Q = volumetric flow rate of system, from
Para. 8.3.1 step 4 = 18,000 CFM

s = number of screens of one cell = 36

$$T = \frac{1 \times 18 \times 4 (159,840 - 29,736)}{28.8 (18,000) (36)}$$

$$T = 0.5 \text{ sec.}$$

Air Aerosol Mixing Uniformity Test

The air aerosol mixing uniformity test was performed in accordance with Para. 9 of ANSI N510. Aerosol is introduced into the air stream at a previously selected injection point. Aerosol concentration readings are taken across a plane parallel to, and a short distance upstream of the HEPA filter bank; the uniformity of these readings establishes the acceptability of the injection port location. Results are given in Table III. The results show uniform mixing within $\pm 10\%$.

Table III Aerosol uniformity.

| Penetrometer Meter Reading | | | | | |
|----------------------------|-----|-----|-----|-----|-----|
| .65 | .60 | .65 | .60 | .65 | .65 |
| .55 | .55 | .60 | .60 | .60 | .55 |
| .55 | .55 | .55 | .60 | .55 | .60 |

Note: Readings taken 6" in front of up-stream HEPA. Each block represents the filter location in the bank.

Average = .592

Acceptable range - .533 to .651

In-Place Leak Tests of the Sorber

In-place leak test of the sorber stage will be described later in Section VII.

IV. Bed Pressure Drop Characteristics

Before filling the Type III sorber with carbon, differential pressure readings (ΔP_1) were taken at various flows to determine the "blank" pressure drop characteristics of the bed. These values are listed in Table IV. After filling the sorber, the tests were repeated and the total pressure drop (ΔP_2) of the sorber (unit + 4 inch depth of carbon) was recorded at the same flow rates as noted in Table IV. From these values, bed resistance was determined and

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correlated with the Reynolds Number through the bed. The correlation between sorber resistance (ΔP_3) and the Reynolds Number from Figure 4 is:

$$\Delta P_3 = .025 (R_e)^{1.209} \quad (4)$$

where ΔP_3 = differential pressure, inches H₂O
 R_e = Reynolds Number

Table IV Sorber resistance data.

| Flow
CFM | ΔP_1
Empty
In. H ₂ O | ΔP_2
Actual
In. H ₂ O | Bed Resistance
$\Delta P_3 = \Delta P_2 - \Delta P_1$
In. H ₂ O | R_e |
|-------------|---|--|--|-------|
| 21,800 | 0.13 | 1.29 | 1.16 | 24.61 |
| 19,300 | 0.11 | 1.15 | 1.04 | 21.79 |
| 18,000 | 0.10 | 1.13 | 1.03 | 20.32 |
| 17,000 | 0.10 | 0.995 | 0.90 | 19.19 |
| 16,200 | 0.095 | 0.93 | 0.83 | 18.29 |

The above test results showed close agreement ($\pm 5\%$) with the pressure drop measured in the laboratory on a 3" dia. bed.

V. Effects of Filling Method on Particle Size and Sorbent Performance

To insure the functional performance of the Type III sorber, it is imperative that the method used to fill the gasketless sorber does not change the particle size distribution or bulk density of the charcoal beyond the acceptable range given in ANSI N509. As a result, any fill method used must be qualified.

MSA qualified two methods of filling. The methods were:

- 1) Sifter Fill Technique*- Method A
- 2) Pneumatic Fill with Vibration - Method B

The Sifter Fill Technique* combines pneumatic conveying of the sorbent from the shipping container to the fill hopper and sifter fill from the hopper to the Type III sorber (see Figure 5, Method A). The Pneumatic (low pressure) Fill Method (see Figure 5, Method B) conveys the sorbent from the shipping container to the

*Sifter Fill Technique, "A Noiseless Method for Filling Packaging Containers", W. E. Gross, Army Chemical Center, Maryland. (4)

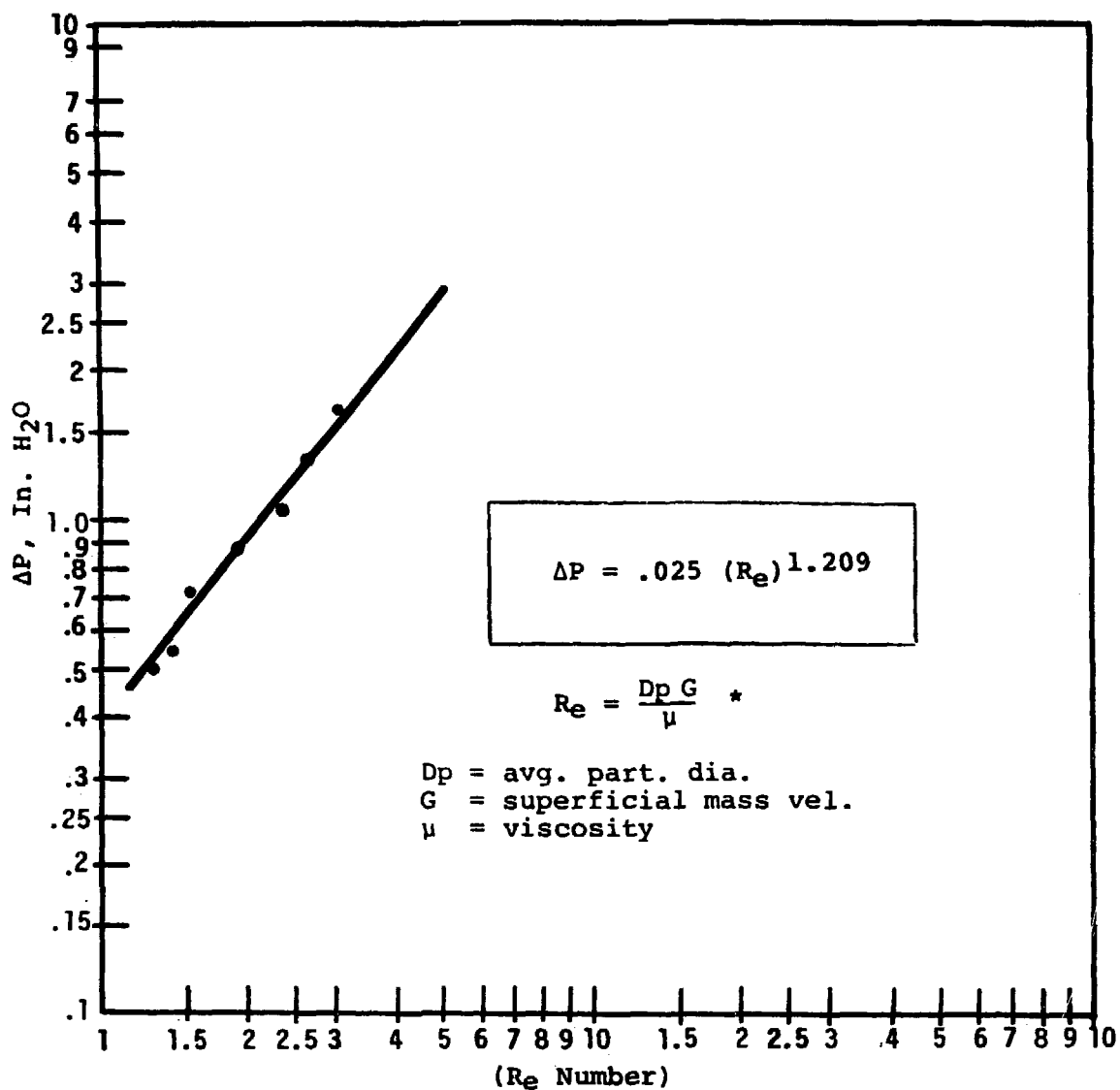


Figure 4. Sorber resistance vs. Reynolds number.

*Perry, R. H., Chemical Engineer's Handbook, 4th Edition, p. 5-50, Equation 5-164. (12)

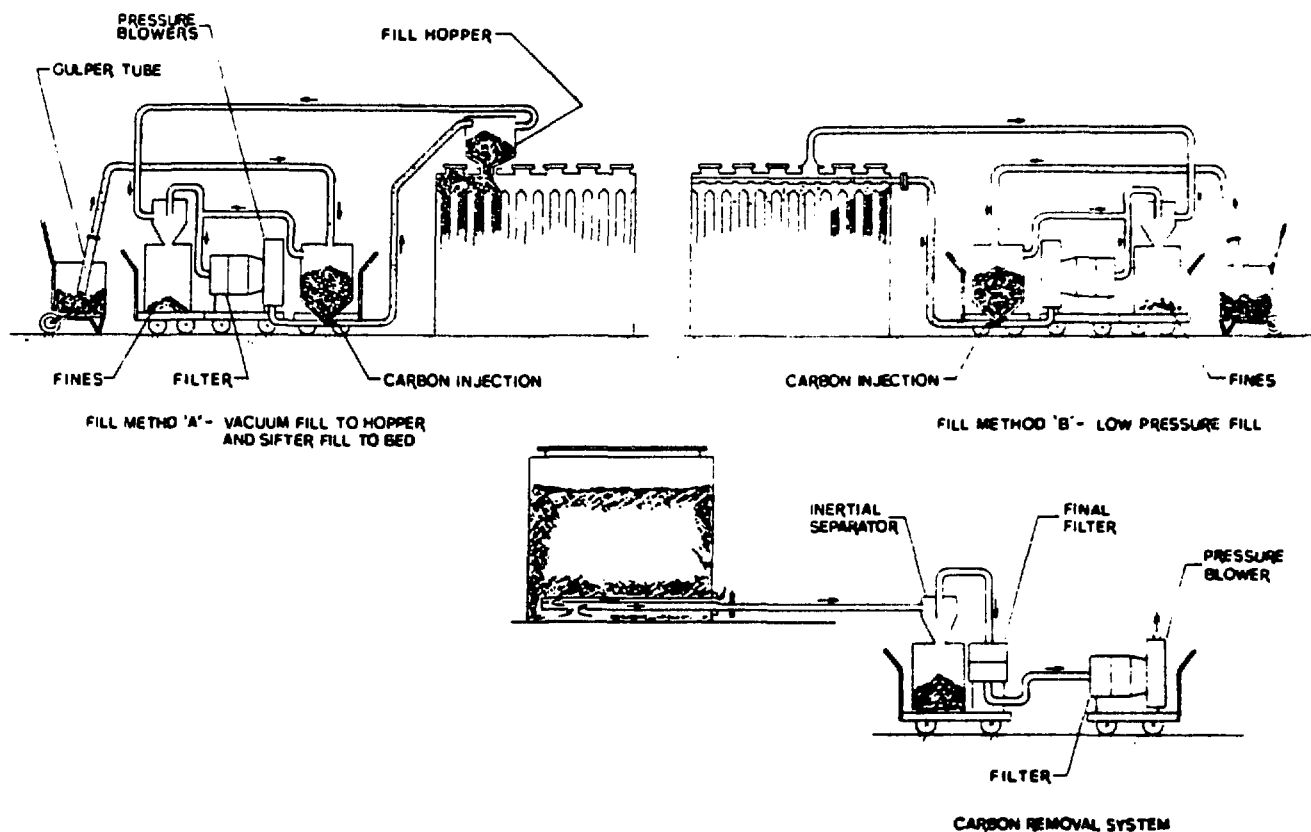


Figure 5. Gasketless adsorber fill and removal methods.

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sorber unit. Vibration must be used with this method to insure uniform packing density. It should be noted that both of the above methods were achieved through research and development. Conventional conveying systems or simple dumping of sorbent into the Type III unit were proven unsatisfactory on early tests.

The 18,000 CFM unit was filled using Method A. During the filling stage, samples of charcoal were taken to determine what changes occurred in particle size during transfer. Samples were taken from the shipping drum and at the fill hopper prior to dropping the charcoal into the sorber.

Table V shows the particle size distribution on the charcoal sampled from the shipping drum and the system fill hopper. Average values in the drum and in the fill hopper can be listed as follows.

Table V Particle size distribution, sifter fill.

| | <u>Mesh Size **</u> | <u>Drum</u> | <u>Hopper</u> | <u>Allowable*</u> |
|-------------------------|---------------------|-------------|---------------|-------------------|
| Through 6, retained on | 8 | 1.5% | 1.2% | 5% |
| Through 8, retained on | 12 | 45.4% | 42.4% | 40-60% |
| Through 12, retained on | 16 | 51.5% | 53.2% | 40-60% |
| Through 16, retained on | 18 | 1.6% | 3.2% | 5% max. |

The above data shows that this method does not effect the particle size distribution of the carbon.

Fill Method B was qualified using the same methods. During the filling stage, samples of charcoal were taken to see if any changes occurred in particle size distribution during transfer. Samples were taken from each shipping drum and from the bed after each drum was transferred.

Table VI shows the particle size distribution of the charcoal sampled from the shipping drum and the adsorber bed. Average values were:

Table VI Particle size distribution, pneumatic fill with vibration.

| | <u>Mesh Size **</u> | <u>Drum</u> | <u>Sorber</u> | <u>Allowable*</u> |
|-------------------------|---------------------|-------------|---------------|-------------------|
| Through 6, retained on | 8 | 1.0% | 1.3% | 5% |
| Through 8, retained on | 12 | 41.9% | 42.8% | 40-60% |
| Through 12, retained on | 16 | 53.8% | 53.4% | 40-60% |
| Through 16, retained on | 18 | 3.3% | 2.5% | 5% max. |

*Allowable values per Reg. Guide 1.52, July 1976. (5)

**Mesh size distribution tests were conducted per ASTM D2862-70. (6)

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There was no significant change in the particle size distribution of the charcoal resulting from it's transfer from the shipping drum into the sorber.

Bed Packing Density

Bed packing density was determined by calculating the volume of the adsorber bed and measuring the weight of charcoal used to fill the bed.

$$\text{Bed density} = \frac{\text{wt. of sorbent}}{\text{volume of bed}} \quad (5)$$

For Fill Method A and B, the calculated bed packing density was 31.52 lbs./ft.³ or 0.50 gms./ml. The average apparent density of the sorbent used was 31.92 lbs./ft.³ or 0.510 gms./ml. for Method A. The apparent density of the adsorbent used was 35.57 lbs./ft.³ or 0.569 gms./ml. for Method B.

The minimum packing density required by ANSI N510 is 0.38 gms./ml.

Apparent density tests were conducted per ASTM D2854-70. (7)

VI. Velocity Measurements

Velocity Profile Across the Sorber Slots

Procedure. The sorber air slot velocities were determined by taking velocity measurements 10 inches downstream of each slot. Ten inch long rectangular ducts, having the same cross-sectional area of the air slots were attached to the exit slots of the sorber unit, as depicted in Figure 6. A hot wire anemometer probe was placed at seven locations in each slot as indicated in Figure 6. The probe tip was located at the center line of each slot. Readings were recorded for each of the seven locations of each slot.

Results. The results of the test are summarized in Table VII.

The acceptable range is considered $\pm 20\%$ of the average velocity of 1145 FPM or 916 to 1374 FPM. Ninety-five (95) percent of the readings were within this range.

Theoretical average slot velocity is 867 FPM. This is calculated as follows:

$$SV = \frac{C}{SA} \quad (6)$$

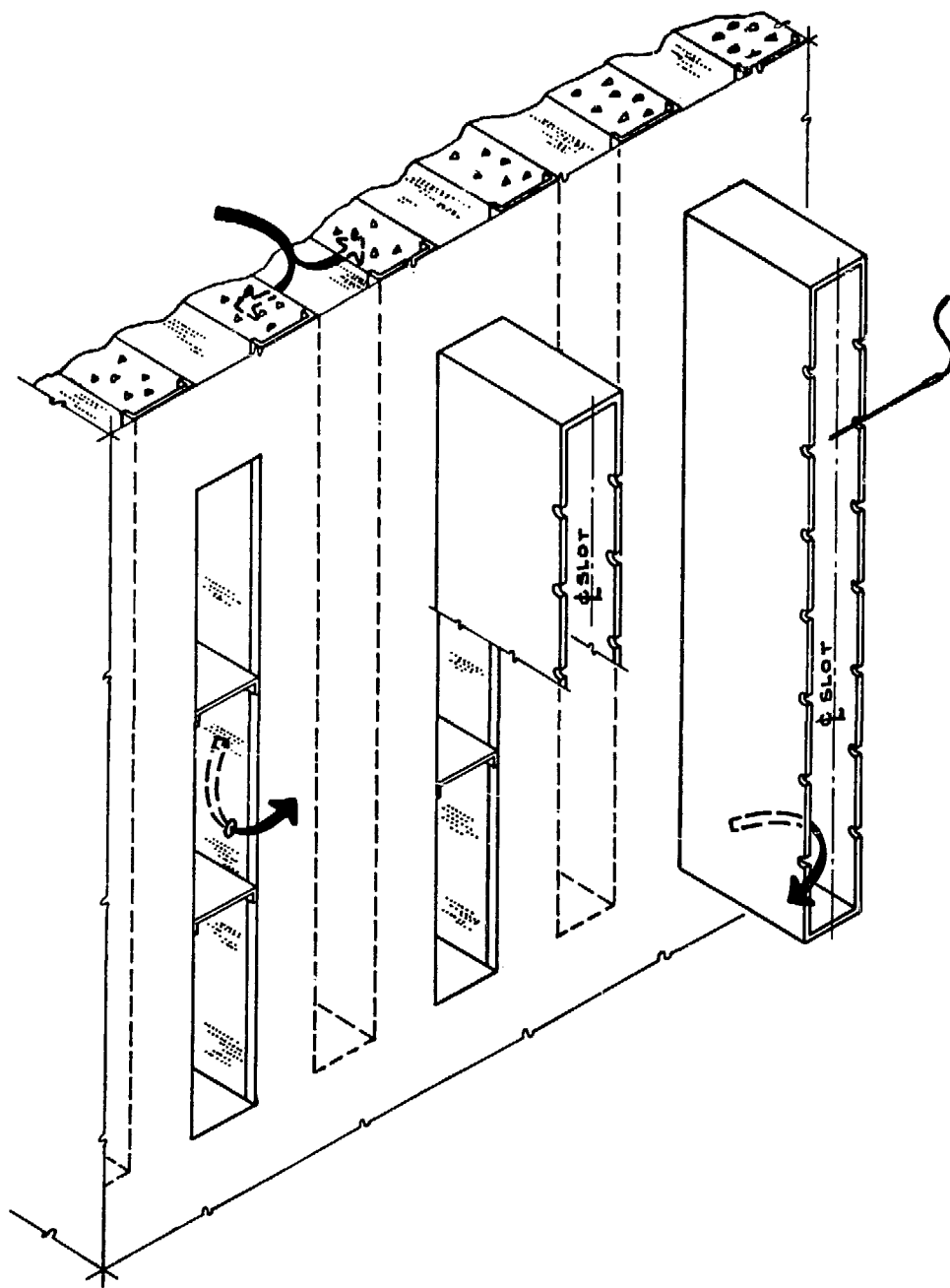


Figure 6. Sorber velocity profile.

Table VII Slot Velocities, fpm - center line velocities vs. probe location.

| Location | Slot 1 | Slot 2 | Slot 3 | Slot 4 | Slot 5 | Slot 6 | Slot 7 | Slot 8 | Slot 9 |
|----------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| 1 | 1300 | 1325 | 1280 | 1290 | 1230 | 1250 | 1380 | 1370 | 1175 |
| 2 | 1050 | 1100 | 1000 | 1060 | 1025 | 1110 | 1140 | 1100 | 1100 |
| 3 | 1040 | 1050 | 1090 | 1125 | 1010 | 1140 | 1170 | 1160 | 1070 |
| 4 | 1040 | 1060 | 1040 | 1400 | 1100 | 1140 | 1125 | 1200 | 1060 |
| 5 | 1050 | 1060 | 1100 | 1400 | 1090 | 1200 | 1360 | 1060 | 1050 |
| 6 | 940 | 1125 | 1150 | 1210 | 1040 | 1075 | 1360 | 950 | 940 |
| 7 | 960 | 1130 | 1025 | 1210 | 980 | 1070 | 1360 | 960 | 925 |

Note: Readings are corrected for temperature and pressure.

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where SV = slot velocity, FPM
C = volumetric flow, CFM = 18,000 CFM
SA = total slot area, ft.²

Total slot area is calculated by:

$$S.A. = \text{No. Slots} \times \text{Slot Height} \times \text{Slot Width} \quad (7)$$

where No. Slots = 9
Height Slot = 78.1 in.
Width Slot = 4.254 in.

$$S.A. = \frac{9 \times 78.1 \times 4.254}{144} = 20.76 \text{ ft.}^2$$

$$\text{Slot Velocity} = \frac{18,000}{20.76} = 867.05 \text{ FPM}$$

The reason for the disparity between the average slot velocity reading of 1145 FPM and the theoretical slot velocity of 867 FPM is most likely a combination of testing technique, bed packing and that the flow is in the transition zone between laminar and turbulent flow.

Velocity Through Test Canisters

Since the sorbent test canisters are a vital part of the Type III sorber unit, the velocity across each test canister was checked.

Procedure. A 15 inch long tube of the same diameter as the test canister was installed on the outlet of the test canister. A hot wire anemometer probe was placed at the open end of the tube with the probe tip located at the center of the tube. With the flow rate through the sorber at 18,000 CFM, readings were recorded.

Results. The average velocity through the test canisters was 39.9 FPM. The allowable range of velocities through the test canisters is 32 to 48 FPM. The actual range was 37.5 to 43 FPM. The test canister design was therefore determined satisfactory.

VII. Sorber Efficiency Tests

In-Place Leak Tests

The Type III sorber was leak tested with Freon-11 at a concentration of 20 PPM at an air flow rate of 18,000 CFM in accordance

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with ANSI N510, Section 12. The efficiencies determined exceeded the acceptance criteria. Efficiency is determined by the equation:

$$\text{Eff.} = 1 - \frac{R_{DS}}{500 R_{US}} * \quad (8)$$

where R_{DS} = Recorder units downstream
 R_{US} = Recorder units upstream

| | <u>Time</u> | <u>Reading</u> |
|----------|-------------|----------------|
| R_{DS} | 60 sec. | 2 |
| R_{US} | 150 sec. | 36 |

$$\text{Eff.} = 1 - \frac{2}{500 \times 36} = .9999 \text{ or } 99.99\%$$

Therefore the Type III sorber unit meets the leak test requirement of ANSI N510.

Freon Breakthrough Test with Time

The primary purpose of this test was to check the Freon adsorption on the charcoal beds throughout the adsorber. To accomplish this the Type III unit was challenged with 20 PPM of Freon-11 at rated flow (18,000 CFM). Gas samples were withdrawn at various predetermined points with time directly downstream of each slot. The efficiencies were measured and calculated per the method outlined in DP-1082. Figure 7 depicts the data collected in this test.

The curve shows the adsorption history for the various carbon beds at different stations until breakthrough occurred. This is the point where the exit concentration begins to rise rapidly with time. It should be noted that the breakthrough uniformity for the various slots and not the time the bed lasts is the important factor of this test. That is, all slots as monitored show approximately the same concentration gradient with time. This is indicative of a uniformly packed bed.

Quantitative Determination of Freon-11 (Trichlorofluoromethane) Sorbed on Charcoal

During removal of the charcoal from the bed, fifty-four (54) samples were taken to determine the amount of Freon sorbed on the charcoal.

The quantitative determination of Freon-11 sorbed on carbon was accomplished by a procedure utilizing solvent desorption and gas chromatography. The procedure was adapted from the method used for the determination of organic vapors in industrial atmospheres. (9), (10)

*Per Savannah River Report DP-1082. (8)

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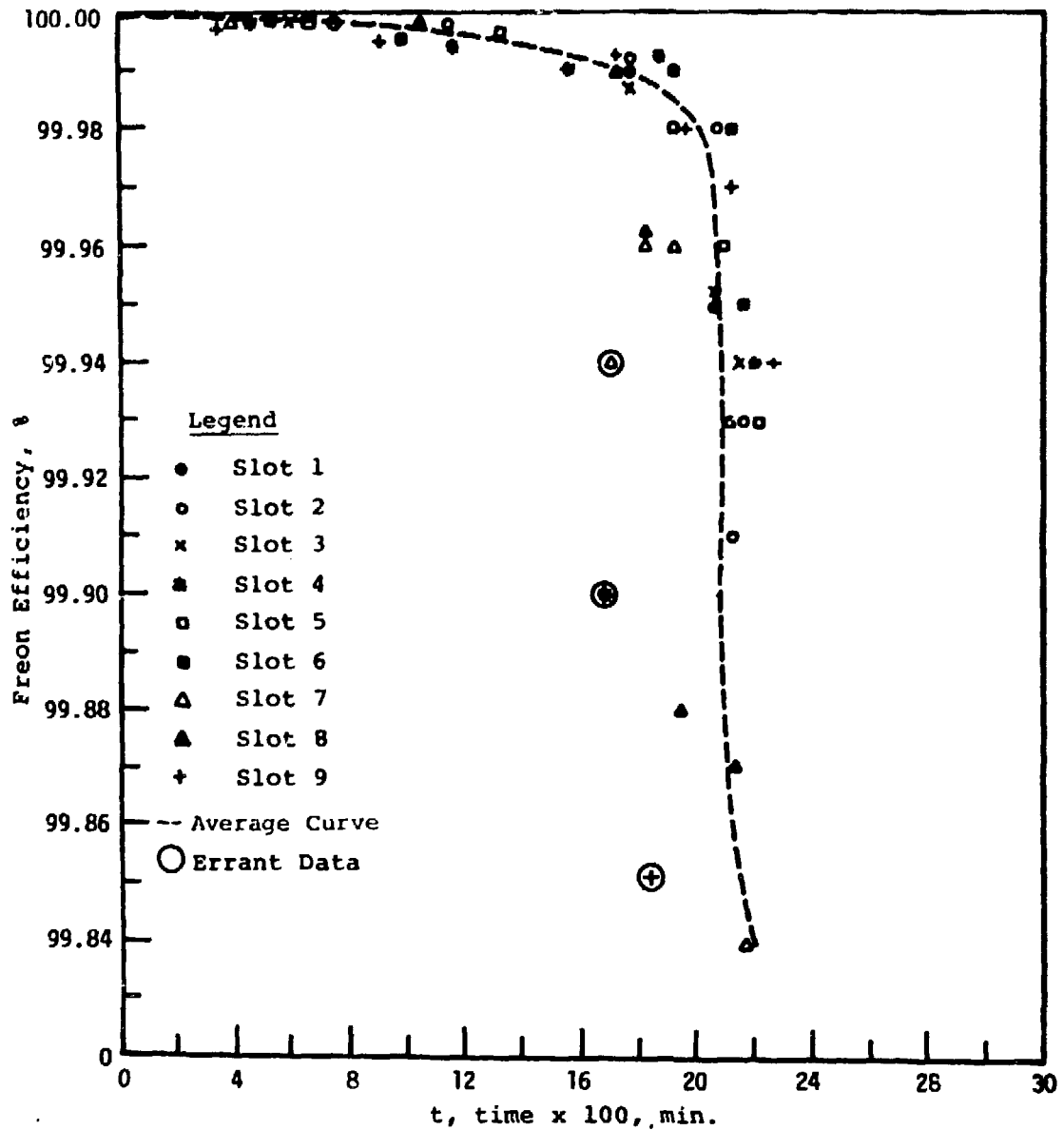


Figure 7. Freon breakthrough curve.

| | Bed Number | | | | | | | | | | | | | | | | | |
|--------|------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 |
| Top | 58.8 | 51.3 | 51.2 | 51.4 | 67.1 | 54.1 | 68.9 | 40.3 | 57.3 | 48.3 | 68.3 | 44.6 | 72.7 | 51.8 | 47.1 | 56.0 | 75.6 | 61.5 |
| Middle | 55.3 | 50.4 | 79.0 | 69.0 | 74.5 | 51.6 | 66.7 | 48.1 | 44.7 | 67.9 | 43.8 | 63.6 | 70.9 | 82.4 | 68.8 | 51.4 | 53.1 | 57.9 |
| Bottom | 42.3 | 43.2 | 53.2 | 48.4 | 61.0 | 45.5 | 53.5 | 78.5 | 44.5 | 75.3 | 86.9 | 75.2 | 55.6 | 58.9 | 59.8 | 44.4 | 54.4 | 59.4 |

Average = 58.7

high 86.9 + 48%

low 40.3 - 31%

Expected error $\pm 35\%$

$\pm 20\%$ Air Dist.

$\pm 10\%$ Freon Mixing

$\pm 5\%$ Testing Error

35%

Expected Range

38 $\longleftrightarrow$ 79

95% of Readings in Expected Range

Table VIII Freon-11 adsorbed on carbon, mg/g.

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Results. Ninety-five percent of the samples were within the expected range of $\pm 35\%$. The results are shown in Table VIII. The average amount of sorption was 58.7 mg. Freon-11/gram of charcoal.

VIII. Inherent Problems with the Type III Sorber

The basic problem with the Type III sorber unit is that there is no test method currently available to assure functional performance in the field. The conventional Type I and Type II cells per AACC Standard* CS-8T are filled and tested at the factory prior to installation and again after installation.⁽¹¹⁾ Furthermore, these sorber cells have undergone extensive testing and each manufacturer's cell must meet the strict requirements put forth in AACC Std. CS-8T. Therefore, there is a strong need for a standard method for qualification testing of a Type III unit. Furthermore, field personnel should be trained and certified in this area.

Another potential problem is field repair. The most common repair necessary would be to the perforated portion of the sorber bed. In addition to accidental tearing or cutting of the screen, a not uncommon occurrence is accidental release of the water spray system. If the wet charcoal is allowed to remain in the sorber unit for a few days, corrosion damage could result to the perforated area due to the corrosive nature of the charcoal. It is very difficult to effect screen repair in the field.

IX. Conclusions

A deep bed Type III sorber unit is shown to meet current applicable test standard ANSI N510 when tested at the factory under controlled test conditions. Furthermore, test methods were developed to demonstrate flow distribution through the bed. The results of this program indicated that the Type III sorber unit when filled and tested by factory trained personnel can exhibit reasonable flow distribution through the bed; however, extreme care and extensive testing is necessary to prove or verify the efficacy of an on-line unit.

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DISCUSSION

KOVACH: I'm always glad to see the test procedures developed in the last few years being used and found applicable to our newer systems.

HALLIGAN: What is the MSA recommendation for the first step to deal with the problem of an initial field test failure on your carbon bank? The choice is either to remove the carbon (to eliminate possible voids) or to perform a shroud test. Both require a substantial amount of effort. Have you considered the approach you would recommend?

PORCO: We have considered it. One would want to check quality assurance documents to begin with and, also, to check that the units were properly filled. You can try a shroud method but I would have a tendency to withdraw the charcoal, recheck it, and then follow the recommended fill technique once again.

HALLIGAN: Both are fairly difficult and require a lot of effort in the field. It is a decision that people who perform the work have to make. Shroud tests on the designs are very difficult. You didn't mention how you would test the HEPA filter banks. Did you give any thought to test ports and DOP distribution upstream of the last HEPA filter bank?

PORCO: The unit I discussed was designed to ANSI 509. Test ports and injection ports are all there for testing per ANSI 510.

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HALLICAN: I am glad it has been worked out because achieving satisfactory DOP distribution on the downstream side, in accordance with ANSI 510, is difficult. Systems that big, employing HEPA filter-carbon-HEPA filter, pose some serious testing problems in the field.

PORCO: These units are tested every day and I know ours pass. I can't speak for everybody.

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EXPERIMENTAL DETERMINATION OF FISSION GAS ADSORPTION COEFFICIENTS*

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Abstract

Large charcoal beds have been used for a number of years for the holdup and decay of radioactive isotopes of krypton and xenon. Reliable design of these beds depends on an accurate knowledge of the adsorption coefficient of krypton and xenon on the adsorbents used in these beds. It is somewhat surprising that there is no standard procedure of determining the adsorption coefficient for krypton and xenon. This paper gives fundamental information needed to establish a standardized reproducible test procedure.

Particular points examined in this paper include:

- 1) The breakthrough curves commonly used to analyze dynamic adsorption data can lead to serious systematic errors.
- 2) The adsorption coefficient, if calculated from the arithmetic holding time, is independent of geometric factors such as the shape of the adsorption bed and the irregular shape of the adsorbent.

I. Introduction

Precise, reproducible measurements are crucial to the nuclear industry. It is ironic that activated charcoal, one of the most useful materials in the nuclear industry, is also one of the most difficult materials to characterize. For example, what does it mean when we say that a certain sample of activated charcoal has a surface area of 1,500 square meters per gram? There are many assumptions behind the values stated for surface area and the consumer in the nuclear industry may not be aware of all of these assumptions. Is the user of this charcoal aware that the value for the surface area will depend on:

- 1) the selection of the vapor used to obtain an isotherm?
- 2) the temperature at which the isotherm is run?
- 3) the range of pressures over which the data is considered to be useful?
- 4) the choice of an equation to fit the data?
- 5) the area assumed to be taken up by a single adsorbed molecule?

The assumptions made in this measurement are important to the reported

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value. To illustrate this point a specific incident can be cited. Several years ago a study at the Harvard Air Cleaning Laboratory involved testing charcoals from various sources as adsorbants for noble gases. One of the parameters that we wished to determine was the relationship between the surface area and the ability of the charcoals to adsorb noble gases. It turned out that the surface areas, as reported by the manufacturers, were obtained by a variety of methods and that as the adsorbed molecule for these determinations we had results from nitrogen, propane, and iodine adsorption. Are these data equivalent? Probably not, but we did not realize it at that time.

Since this experience it has been useful to us to try to characterize the types of measurements that can be made on charcoal according to the reproducibility of these tests. Table I below shows how we have tried to rank the various types of analyses.

TABLE I. Precision of Selected Tests

| Measurement | Degree of Precision | Reason |
|-----------------------------|---------------------|--|
| Temperature)
Pressure) | High | These are thermodynamic properties which can be measured at equilibrium. |
| Bulk Density | Moderately Good | Depends on force by which charcoal is packed. Also influenced by adsorbed air, which can be about 1% of measured value. |
| Surface Area | Fair | Assumptions as noted above. |
| Removal of
Methyl Iodine | Poor | Depends on rate of chemical reactions. Kinetic data are always difficult to reproduce even under the best of conditions. |

The purpose of this paper is to evaluate the underlining accuracy of dynamic procedures for determining the adsorption coefficients of charcoals for noble gases. As will be seen, a strong argument can be made that the measurement of the adsorption coefficient, like the measurement of surface area, depends on many assumptions, some of which may be valid and others not. However, the conclusion in this paper will be that a dynamic adsorption coefficient can be determined precisely and given an exact physical meaning. It is our hope that this paper will contribute to the development of standardized procedures for the measurement of noble gas adsorption coefficients.

II. Use of Breakthrough Curves

Dynamic adsorption data are usually obtained by injecting a pulse of noble gas into a gas stream passing through a charcoal bed and then measuring the effluent noble gas concentration. The dynamic

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adsorption coefficient can then be determined from these data. The usual procedure for the analysis of these data has been to fit the data to a two-parameter curve in which one of these parameters is the adsorption coefficient. In this manner, a "best" value for the adsorption coefficient can be found. As shown in Table II, there are a number of theoretical curves that have been used for this purpose.

Table II. Theoretical Curves Available

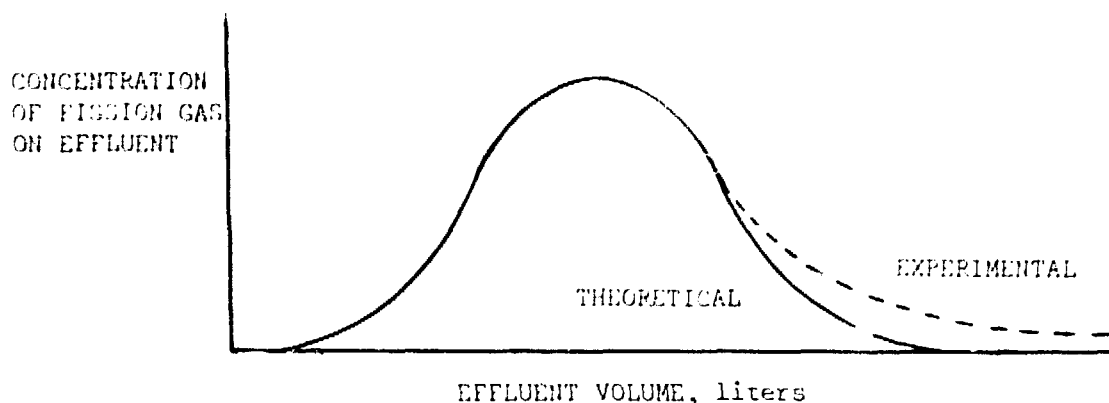
| Author of Equation | Second Adjustable Parameter | Comments |
|-------------------------|--|--|
| Browning ⁽¹⁾ | Number of subdivisions in bed in which mixing occurs | Doubtful that such subdivisions (called theoretical Chambers by Browning) exist. |
| Burnette ⁽²⁾ | Mass transfer rate across air film surrounding each particle | In general, air film does not control mass transfer in adsorption |
| Madey ⁽³⁾ | Gas phase diffusion | Valid only at low carrier gas velocities |
| Rosen ⁽⁴⁾ | Diffusion in adsorbent | Assumes adsorbent consists of internally homogeneous spheres of equal diameter |

The basic difficulty is that these equations, although they can in themselves appear complex, represent fairly simple models of fission gas behavior, and each can account individually for only one mechanism of mass transfer. Therefore, these equations are not strictly applicable to normally operating fission gas adsorption beds, in which a number of mass transfer mechanisms are important. There is no guarantee that the use of any one of these equations will be appropriate and lead to results of a specified accuracy. In fact, it has been our experience that in general, fission gas breakthrough curves exhibit a tailing which is greater than that predicted by any one of the above equations. The reason for the presence of this excessive tailing, we have believed, is the presence in the charcoal adsorbent of pores or pore areas, where the diffusion of fission gases is highly restricted. Only a small fraction of the input fission gas may reach such areas, but once this occurs these gases find it equally difficult to diffuse out of these areas. Hence, as the sketch below shows, generally, the breakthrough curve has an excessive tailing. Until recently we were not certain how to handle this effect in calculating the adsorption coefficient.

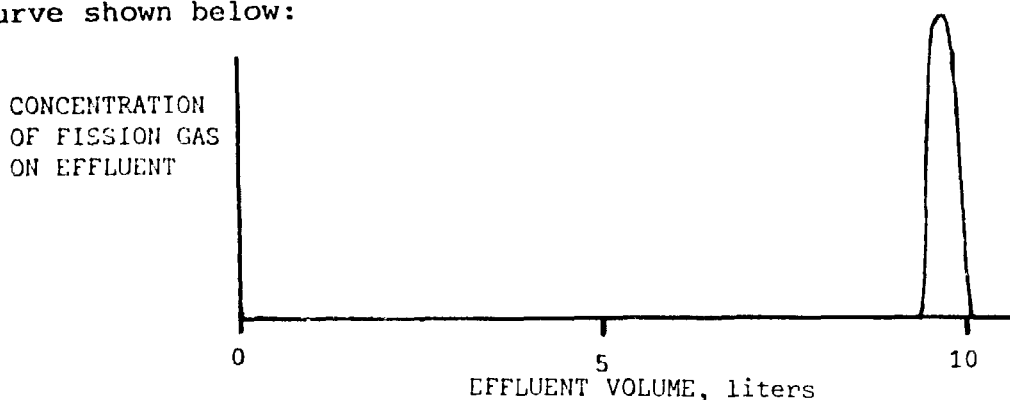
III. Direct Calculation of the Adsorption Coefficient

It is our belief that the adsorption coefficient is best calculated directly from the experimental data without use of a theoretical

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breakthrough curve. To show in a simplified manner how this is done, suppose that we inject a pulse into a bed and obtain the breakthrough curve shown below:



It is apparent from this curve that the bed retarded the release of the input fission gas over a period of time in which 10 liters passed through the beds. The adsorption coefficient is then calculated as

$$k = \bar{V}/m, \quad (1)$$

where $\bar{V}$ = the retention volume,

and m = the mass of charcoal.

In any real bed the effluent output appears as a skewed Gaussian curve, and the arithmetic average retention volume can be calculated as

$$\bar{V} = \int_0^{\infty} c V dV / \int_0^{\infty} c dV \quad (2)$$

The use of the arithmetic average retention volume to calculate the adsorption coefficient has the following advantages:

1. The result is consistent with all the breakthrough curves cited above. For example, we used a computer to generate the breakthrough curve predicted by the Madey equation, and then applied this method of analysis to that curve. In this test of this procedure the actual error was less than one part in a thousand, and the error could have been further reduced by increasing the precision of the

integration procedure used for this analysis. On the other hand, if we take the same breakthrough curve and try to fit it by a least squares procedure to the curve predicted by one of the other theories, e.g., Browning's theoretical chamber model, a finite error will be found that cannot be reduced by taking more data points. In other words, the procedure of taking the arithmetic average is such a general procedure that it can be used to analyze data from each of the popular models for breakthrough curves, even though these models themselves cannot be used interchangeably between themselves.

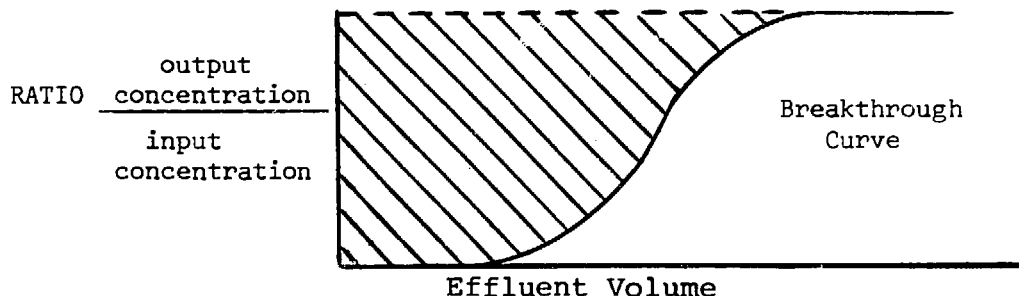
2. This procedure is sufficiently simple as to be carried out using a desk calculator. In contrast, some of the breakthrough curves mentioned above involve Bessel functions, and least squares fitting to these curves requires a computer.

3. As long as the effect of the pressure drop can be ignored, the results obtained from this type of analysis are independent of either the shape of the bed or the flow velocity of the carrier gas. If corrections must be made for the effect of pressure drop, they are extremely difficult to incorporate into any of the breakthrough curves cited above.

4. The "tail" of the breakthrough curve is included in the analysis by the arithmetic mean holdup time. In contrast, other methods of analysis are not able to account for the tailing, which results, we think, from ill-defined pore restrictions in the charcoal granules.

5. The results have a precise physical meaning: The adsorption coefficient obtained in this manner is identical to the static adsorption coefficient obtained if the charcoal is allowed to come into equilibrium with a constant concentration of fission gas.

The proof of this latter assertion is as follows: If a bed is exposed to a constant concentration of fission gas, as it approaches equilibrium saturation with the fission gas, the breakthrough curve will have the following shape:



The hatched area above the breakthrough curve is proportional to the amount of material removed by the bed from the air stream and therefore can be used to determine the quantity of material adsorbed by the bed at equilibrium saturation. If certain rather general assumptions are made, such as a constant carrier gas velocity, constant diffusion coefficients, and a linear isotherm (all these factors were assumed in the derivations of the breakthrough curves which were cited in the previous section), then the hatched area will be identical with

the arithmetic average holdup volume determined from a pulse input.

In order to make this paper as readable as possible, we have left out as much mathematics as possible. This includes the mathematics involved in proving the above statement. However, the more adventuresome of the readers may be interested in a paper which appeared in Nature last year.⁽⁵⁾ The central theorem, as stated in this paper, is that for simple flow systems "the mean residence time of material is independent of internal characteristics and always equal to the ratio of total holdup of material to its overall flow rate through the system." It is also stated that this theory has been "central to the development of a model independent theory of chromatography which supersedes previous treatment, based on specific models of the chromatographic process by obviating the restrictive assumptions on which these are founded." It is our belief that the best procedure for determining fission gas adsorption coefficients is based on the same powerful mathematical technique.

IV. Summary

The use of the mean retention volume to calculate the adsorption coefficient is both a simple method of analysis and probably the most precise procedure currently available.

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DISCUSSION

WOOD: The pulse technique is advantageous only with a relatively inefficient bed. With sorbent beds highly efficient for a test adsorbate (such as charcoal and iodine) no pulse would come through unless the challenge pulse overloads the bed.

UNDERHILL: For the study of noble gas adsorption, it is possible to use pulses of ^{85}Kr or ^{133}Xe . These radionuclides can be detected at very low concentrations. The equations given here are too simple to account for removal by chemical reactions. However, this can be done and the appropriate equations were given at a previous Air Cleaning Conference.

WOOD: These are inert gases. I'm thinking in terms of radioiodine.

UNDERHILL: I mentioned radioactive krypton and xenon. I didn't include iodine because iodine reacts chemically in the bed. The technique described here would not apply to radioiodine unless you had a form that wasn't reactive, and that's not in the cards.

WOOD: A second question. Did you say that the top, or the median, of the pulse curve was the only important thing?

UNDERHILL: No, I didn't. What I'm really trying to say is that the entire breakthrough curve is important.

DEITZ: Does not a pulse technique that you advocate introduce both adsorption and desorption coefficients displaced in the time interval of the pulse?

KOVACH: In reference to Dr. Deitz's question, in all dynamic adsorption processes, whether pulsed or continuous, both adsorption and desorption rates influence the process.

UNDERHILL: Kovach's explanation is absolutely correct. This has been looked at. If you have a very constricted pore system, very few atoms will diffuse in. Most of the adsorbate may simply bypass the constricted pore area and produce tremendous tailing. This results in slow adsorption which can't be detected when looking at only one part of the curve. But the long tail represents the small amount that did get in. I am trying to use a classical mathematical proof to point out that if the pores are wide open, you can diffuse in and diffuse out very rapidly and the K value determined from this type of analysis is identical. It doesn't make any difference how constricted the pore area is; you will get the same K value. It is independent of the diffusion coefficient. That is really remarkable.

LEVINS: While I agree, in principle, with the methods you advocate, practical problems can arise in some circumstances using the method of moments when applied to pulse data. These arise because integrations must be taken to infinity. In the presence of severe tailing or base-line drift, a practical decision must be taken as to when to terminate the integration. Some work with which I am familiar found that experiments were apparently non-reproducible when processed by the method of moments because the tail made a significant, but not experimentally reproducible, contribution to the total area. When, however, the curves were fitted in the region near the peak, which is known with greater precision, the agreement was excellent.

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UNDERHILL: We had no difficulty applying the technique to the adsorption of krypton. With a higher concentration of radon you may have been able to analyze accurately the tailing section of your breakthrough curves. We found with krypton that if too little is injected, the tail becomes difficult to analyze.

PARISH: It seems to me that this tailing effect may be a practical consideration in regeneration. Have you taken a look at that?

UNDERHILL: Spreading mechanisms, including those that cause tailing, are central to calculations of regeneration efficiency. There have been a number of theoretical calculations of the effect of tailing on regeneration, but to my knowledge, the actual measurements of this effect in regard to fission gas adsorption and desorption are not sufficiently accurate to permit accurate application of these theories.

PARISH: I would just like to clarify a point. This analysis would not apply to delay beds but only to batch type systems?

UNDERHILL: To regeneration systems, where you are heating and cooling.

JONAS: The methods of Rosen, Masamune and Smith, and Schneider and Smith, using statistical moments, are based on the inlet gas concentration being on the linear portion of the isotherm. However, we are sometimes concerned with challenge concentrations of nerve gas which are on the plateau of the isotherm. For these cases, the Wheeler type kinetic equation holds very well since it can pinpoint the time at which a very small concentration exits the bed. The generic type of sigmoidal curves obtained from complete breakthrough may not be able to accomplish this time pinpointing as well as the kinetic breakthrough equations.

UNDERHILL: Let me add one additional comment about the appropriateness of this type of analysis to the particular problem that it was assigned to. When you're dealing with offgases from a nuclear reactor, it can be shown that the releases are in the range of parts per million. There are, already, part per million concentrations in the air. Therefore, there will be isotopic exchange and the assumption of linearity works beautifully. There is no question about it for this particular case. For the case that you're dealing with, it sounds like the type of work that a mathematician would love to do. It's a very interesting problem.

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A RADIOIODINE DETECTOR BASED ON LASER INDUCED FLUORESCENCE

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Abstract

A technique is described to measure airborne concentrations of $^{129}\text{I}_2$ in nuclear power generation and reprocessing plants. Due to its low specific activity, long half-life, and low energy decay products, radio-counting techniques for ^{129}I are not practical. A laser fluorescence technique based on coincidences between a ^3He - ^{22}Ne laser and $^{129}\text{I}_2$ absorption lines is used and is shown to be sensitive to near 10^{-12} g/cm³ of $^{129}\text{I}_2$ in a scrubbed system; at 1 atm of argon and 1 atm air the sensitivities are reduced by factors of 86 and 131.7 respectively. Possible interferences, measurements under flowing conditions, and ultimate sensitivities are discussed. An evaluation of the technique and extension to a portable, real time monitoring instrument for use in the field is given.

I. Introduction

Needs and Applications for Radioiodine Monitoring

Of the six major iodine isotopes produced in nuclear reactors, ^{129}I represents only about 1% of the main fission product iodine isotope yield.¹ Although this isotope has the lowest specific activity and the lowest energy decay products, after a few days of cooling outside the reactor the major iodine isotope present is ^{129}I , because the next longest-lived isotope ^{131}I has a decay half-life of 8.0 days. In view of the very long ^{129}I half-life of about 17 million years, new attention has been directed to its cumulative effects.^{2,3} In spent fuel processing ^{129}I will have to be recovered and stored indefinitely because of its longevity. Because of the low specific activity, low energy decay products, and because ^{129}I is a minor iodine isotopic constituent in an active core, its detection by counting techniques is often slow and impractical.

Activated charcoal is the most widely used material employed for iodine removal, and it is also used as a means for decay storage. Because organic compounds of iodine are adsorbed less well than I_2 on activated charcoal substrates, in some applications charcoals are used which have been impregnated with substances which react chemically with I_2 and compounds of iodine or which undergo an isotopic exchange.¹ Among examples currently used are charcoals impregnated with KI (or other inorganic iodine compounds) in which the stable isotope of the impregnant iodine can undergo an exchange (or reaction) with the radioiodine. Note that such an exchange can release a stable iodine into the effluent.¹ The efficiency of charcoal filters is strongly dependent upon their environmental history,

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(particularly extended exposure to very high relative humidity)⁴ and the desorption rate of iodine from the filters is strongly dependent upon temperature.⁵ It appears that there exists a need to monitor and measure quantitatively in real time, iodine concentrations in several areas:

- (a) as a continuous monitor to observe breakthrough behind critical filter facilities;
- (b) as a testing and trouble shooting field measuring tool to test the integrity of in place filters;
- (c) as a monitor of fairly high ^{129}I ambient concentrations in spent fuel reprocessing; and
- (d) as a monitor to observe accidental large releases of radioiodine in the spent fuel reprocessing cycle.

Proposed Research

For the reasons outlined above, NRL in cooperation with DOE (Oak Ridge Operations Office) has contracted to design and laboratory test an instrument for the detection of low concentrations of $^{129}\text{I}_2$ in ambient atmospheres. The proposed instrument is to be based upon a laser induced fluorescence technique. In the design and evaluation of the proposed instrument the following characteristics were to be stressed:

- (a) the specified final instrument must be easily portable and capable of use under field conditions;
- (b) the instrument should be specific for ^{129}I ;
- (c) the specified instrument should be capable of making real time measures - preferably on flowing gas samples;
- (d) the final field hardened instrument should be capable of continuous operation over extended periods of operation; and
- (e) the instrument should be as sensitive as possible consistent with an economic appeal which would allow widespread use in the industry of the field instrument.

II. Experimental

Approach to Experimental Problem

A laser induced fluorescence technique was chosen for application to monitoring of I_2 concentrations. In this technique a laser photon of appropriate frequency impinging upon the sample is selectively absorbed by the iodine molecule placing it in a metastable excited electronic state. The excited iodine molecule can relax by collisional processes back to the ground state in which case it will not be detected, or it may relax by reemitting a photon (of longer wavelength) by a fluorescence mechanism. This fluorescence emission may be detected by a photoelectric detector. The resulting

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fluorescence signal, after appropriate corrections are made for competing collisional relaxation processes, collector efficiency, detector response characteristics, and other emissive interferences, can be used to determine the I_2 concentrations in a gas sample. Several problems which must be considered in the application of this technique to quantitative measurements are enumerated below:

(a) a laser excitation frequency must be chosen which corresponds to an optical resonance of the specific molecule to be detected;

(b) excitation frequencies must be selected which do not correspond to resonances of other species in the sample (i.e., other I_2 isotopes);

(c) evaluation must be made of all non-fluorescing deactivation pathways which the selected molecule may undergo;

(d) possible fluorescing interferences from other species must be accounted for; and

(e) allowance must be made for all possible molecular forms in which the atom to be detected may occur and appropriate corrections be made.

Spectroscopic Considerations

The I_2 molecule has a very characteristic absorption in the visible region of the spectrum. The major absorption features and the electronic transition assignment have been known for many years^{6,7} for the red band system. The transition is a spin forbidden process $\tilde{B} (^3\Pi_{out}) \leftarrow \tilde{X} (^1\Sigma_g^+)$ which is made relatively allowed by the heavy atom nature of the atoms involved. The visible band fluorescence of this system was first critically evaluated over 70 years ago.⁸ Scores of studies of both absorption and emission have been carried out in the intervening years. Recently the absorption lines in this transition have been chosen to use as a new secondary wavelength standard by workers at the National Bureau of Standards. For this application it has been necessary to catalogue very precisely many of the absorption features of this spectrum. Hanes and Dahlstrom,⁹ Wallard¹⁰ and Hanes *et al.*,¹¹ have frequency stabilized a $^3\text{He}-^{20}\text{Ne}$ laser to hyperfine components in the absorption spectrum of $^{127}\text{I}_2$ at 633 nm and Knox and Pao¹² have frequency stabilized a $^3\text{He}-^{20}\text{Ne}$ laser to hyperfine components in the $^{129}\text{I}_2$ spectrum near 633 nm. More recently Schweitzer,¹³ *et al.* have carried out similar work using a $^3\text{He}-^{22}\text{Ne}$ laser monitoring resonances of $^{129}\text{I}_2$. It was shown in the latter work that the gain curve of the $^3\text{He}-^{22}\text{Ne}$ laser overlaps ~20 hyperfine components in the $^{129}\text{I}_2$ spectrum, but does not have any interferences from absorption features of the more common isotope $^{127}\text{I}_2$.

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For the above reasons the $^3\text{He}-^{22}\text{Ne}$ laser was chosen as the preferred excitation source for the laser induced fluorescence experiments on I_2 .

Equipment Design

$^{129}\text{I}_2$ and ^{22}Ne Spectral Characteristics

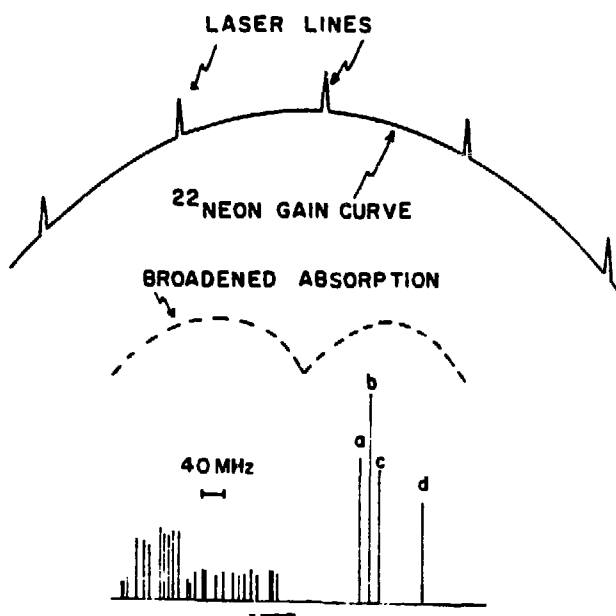


Fig. 1 — Spectroscopic characteristics of the $^{129}\text{I}_2$ spectrum and the $^3\text{He}-^{22}\text{Ne}$ laser. The line spectrum at the bottom is the hyperfine absorption spectrum of $^{129}\text{I}_2$ measured by Lamb Dip spectroscopy from Ref. 13. The dashed curve above this represents the Doppler and pressure broadened absorption of $^{129}\text{I}_2$ characteristic of ambient temperature and pressure conditions. The solid curve at the top schematically represents the $^3\text{He}-^{22}\text{Ne}$ laser gain curve while the spikes on this curve schematically represent five oscillating modes characteristic of the laser employed in this work.

In Figure 1 a portion of the spectrum of $^{129}\text{I}_2$ in the region of the ^{22}Ne gain curve is shown. These spectral lines are shown as measured by Lamb Dip spectroscopy and are not pressure or Doppler broadened. The wavelength of the B feature is $6329.90\text{\AA}$. The B feature is separated from the A feature by $\sim 0.003\text{\AA}$. In experiments such as those conducted in this work the Doppler and pressure broadening of the features shown in Figure 1 will merge all these hyperfine components into an apparent continuum absorption as represented schematically by the dashed curve immediately above the stick spectrum. A schematic of the gain curve of ^{22}Ne laser is shown immediately above the dashed curve as a solid line. A ^{22}Ne laser oscillating between two cavity mirrors can lase upon a

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selected frequency or frequencies anywhere on this gain curve. The actual operating characteristics (bandwidth, number of cavity modes operating and mode spacing) of an individual He-Ne laser will depend upon gas mix, discharge current and cavity length of the laser employed.

Specifications of ^3He - ^{22}Ne Laser

The laser purchased for this is a basic Jodon Model HN-7 configured for TEM₀₀ operation at 6328Å. A special fill of ^{22}Ne was specified for the laser. The power output of the laser upon arrival was 16.0 mwatt, a value which has held constant over several months of operation. The spectral characteristics of the laser were measured with a Coherent Optics Model 380 Interferometer. We find that there are five to six cavity modes oscillating at all times. The oscillation of these cavity modes with their characteristic 260 MHz cavity spacing is indicated schematically superimposed upon the laser gain curve shown in Figure 1. Since the Doppler width of the I_2 resonances is ~200 MHz, any frequency jitter in the ^{22}Ne laser frequencies is not reflected in the absorption efficiency of the pseudo continuous absorption of $^{129}\text{I}_2$ across the whole ^{22}Ne laser gain curve.

Fluorescence Cells and Photomultiplier Detection

The fluorescence cells used for static measurements are constructed of quartz and are 1 inch cubes. The scattered laser light is rejected by combinations of corning glass longwavelength pass sharp cut filters and specially constructed dielectric blocking filters. The 6328Å laser light is rejected, and all fluorescence of $\lambda \sim 6600\text{Å}$ is detected. The detector is an RCA 31000A 2" photocathode extended S-20 response photomultiplier. A block diagram of the breadboard detector is shown in Figure 2.

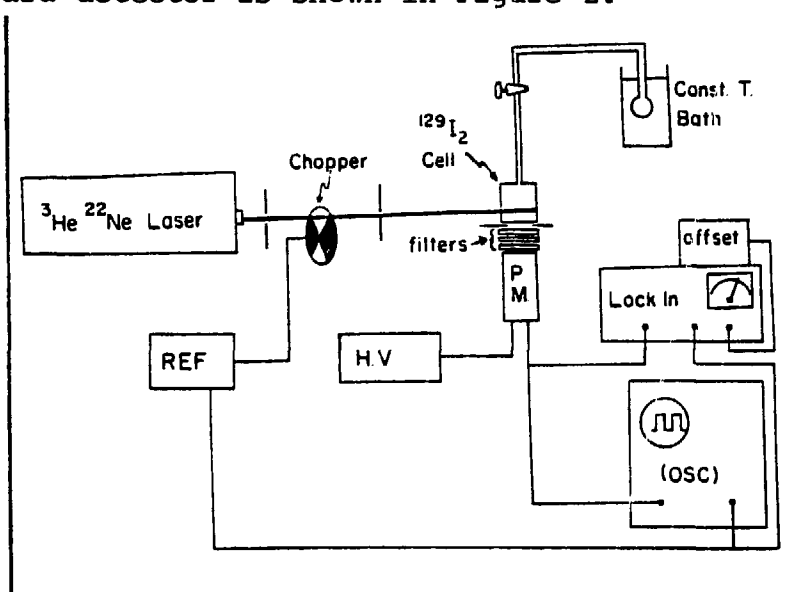


Fig. 2 — Schematic representation of the breadboard detector employed in this work

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$^{129}\text{I}_2$ Samples and Pressure Control

In order to generate a set of working curves, it is necessary to make static measurements on known pressures of $^{129}\text{I}_2$. The $^{129}\text{I}_2$ samples were obtained commercially from ICN, Inc. in sealed ampules each containing 3 mg of $^{129}\text{I}_2$. The samples were introduced into the fluorescence cells on a vacuum line and then sealed off. The $^{129}\text{I}_2$ pressure is controlled in the static cells by means of constant temperature slush baths. I_2 vapor pressure curves as a function of temperature were taken from the International Critical Tables. Various constant temperature slush baths were employed to control the temperature between -55 and 0°C. In addition, various measured temperature baths were produced with ice-NaCl- H_2O mixtures. A Table of convenient slush baths with corresponding temperatures and I_2 pressures is shown in Table I.

Table I

| <u>Compounds</u> | <u>Slush Temperature</u> | <u>I_2 Pressure (mtorr)</u> |
|-----------------------------------|--------------------------|---|
| H_2O (ice) | 0°C | 31 |
| H_2O (ice) + NaCl | 0°, -14°C | 31 - 7.0 |
| CCl_4 | -22.5°C | 2.6 |
| Chlorobenzene | -47°C (-45.2°C) | 0.13 |
| Chloroform (CHCl_3) | -53°C | .035 |
| N_2 (liquid) | -196°C | 0 |

Fluorescence Signal Detection and Processing

Because of the ease of operation and availability of equipment, it was decided to measure fluorescence signals by means of lock-in amplifier detection. It should be possible to obtain equivalent results by photon counting techniques. The lock-in amplifier employed is a PAR Model HR-8 used with the appropriate chopper and chopper reference. Because of small, but finite, scattered laser light signals, we have found it convenient to employ a PAR Model 123 A.C. zero offset to null out the scattered laser light signal when measuring low level signals. Using this nulling device, one can easily zero out scattered laser light down to < 10 volts full scale thus extending the sensitivity by ~ a factor of a hundred.

The photomultiplier signal is also monitored by an oscilloscope which is convenient for monitoring the signal level at relatively high I_2 pressures for purposes of monitoring photomultiplier dark current levels and for aligning the laser and fluorescence cells, etc.

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Notes on Handling $^{129}\text{I}_2$ in Static Cells

During the course of this work we have noted a deterioration of fluorescence signals from our $^{129}\text{I}_2$ reference cells on several occasions. These problems were tracked down to a H_2O contamination problem. I_2 reference samples which have been exposed to a trace contamination of H_2O have much different vapor pressure characteristics than when completely dry. The introduction of one atmosphere of moist laboratory air is sufficient to form a surface layer coating over the I_2 samples which is visible to the eye. Resublimation of these samples does not allow one to recover the samples effectively. In all cases, contaminated samples of I_2 give significantly lower equilibrium vapor pressure of I_2 than do the pure samples. We have been able to introduce dry air into the fluorescence cells and subsequently reclaim the I_2 reference samples if one uses clean vacuum techniques and carefully handles the samples. I_2 samples for reference cell purposes are best prepared under dry high vacuum techniques and should be permanently sealed off.

III. RESULTS

Static Measurement

Single Component Gases

Using the detector design shown in Figure 2, $^{129}\text{I}_2$ fluorescence signals were measured from two different fluorescence cells for two separate samples of $^{129}\text{I}_2$ over many repetitions at various temperatures in cells containing only $^{129}\text{I}_2$. Apparent pressure equilibrium is obtained after ten minutes with a new temperature slush bath. Before and after each series of measurements the sample is frozen out at 77°K to check the background correction factor. The upper curve in Figure 3 shows a plot of fluorescence signal intensity as a function of $^{129}\text{I}_2$ pressure (and concentration). Measurements have been made over iodine concentrations between 5×10^{-7} and $4 \times 10^{-10} \text{ g/cm}^3$. A good straight line signal response is obtained over this range. For the lower concentration measurements the ability to measure signals is not limited by fluorescence signal, but by the uncertainty in the known vapor pressure curves of I_2 as a function of temperature. The practical limits of sensitivity for fluorescence measurements are indicated by the dotted line extension of this curve. Since the signals follow a linear response curve, it is possible to measure in evacuated systems I_2 concentrations to near one picogram per cm^3 . The working curve so obtained is in no way dependent upon a need to independently measure the absolute I_2 concentration by vapor pressure curves or other techniques below the 10^{-10} g/cm^3 level.

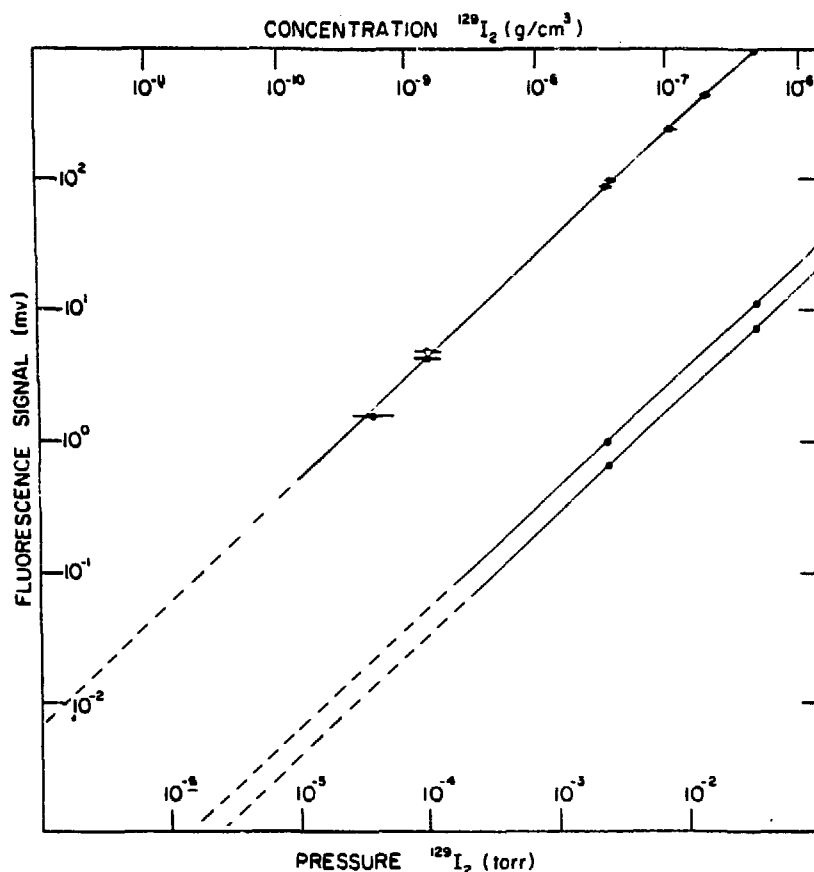


Fig. 3 — Working curves for $^{129}\text{I}_2$ fluorescence intensity as a function of pressure or concentration. The upper curve is measured for $^{129}\text{I}_2$ in evacuated static cells. The horizontal error bars represent the uncertainty in the I_2 pressure resulting from uncertainty in the absolute temperature-vapor pressure measurements for I_2 . The vertical error bars are smaller than the size of the dots in the figure, i.e. $\sim \pm 10 \mu\text{volts}$. The center and lower curves are fluorescence signal response for $^{129}\text{I}_2$ in a static cell in the presence of 1 atm of argon and 1 atm of air respectively.

Multicomponent Gas Mixtures

Working fluorescence signal curves were also obtained for $^{129}\text{I}_2$ in the presence of a rare gas buffer and in the presence of air. Since bimolecular quenching of I_2 fluorescence is a well documented process, the loss of fluorescence signal in the presence of other gases is predictable.¹⁴ However, since the quenching process is dependent upon the pressure, the collision partner, and the excitation wavelength, independent measurements are required for this new experimental setup. As it is anticipated that measurements will be made with this detection system only under either evacuated conditions or in the presence of one atmosphere of air or some other buffer gas, multicomponent gas mixtures were measured only at one

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atmosphere total pressure. The middle curve in Figure 3 shows a plot of I_2 fluorescence signal as a function of I_2 concentration in the presence of one atmosphere of Argon while the lower curve is in the presence of one atmosphere of air. In the presence of Argon the fluorescence signals are decreased by a factor of 86.0 while in air the signals are decreased by a factor of 131.7 over the respective values under evacuated conditions. The quenching cross sections for He, Ar, N_2 , O_2 and CO_2 from Capelle and Broida¹⁴ are given in Table II for excitation at 6234Å. The relative fluorescence efficiencies observed in this work are in essential agreement with what is expected based upon the relative quenching efficiencies of the gases in Table II. It is apparent that there is little advantage to be gained from scrubbing I_2 from the air and rediluting it in an Argon carrier gas. On the other hand, the data in Table II indicate that one might gain a factor of ten to twenty in increased sensitivity if one were to make measurements in one atmosphere of He gas as opposed to air.

Table II

| <u>Quenching Gas</u> | <u>Cross Sections</u>
(in 10^{-16} cm^2) |
|----------------------|--|
| He | 0.44 $\pm$ 0.26 |
| Ar | 5.62 $\pm$ 0.24 |
| N_2 | 5.0 $\pm$ 0.6 |
| O_2 | 7.8 $\pm$ 0.3 |
| CO_2 | 22.1 $\pm$ 0.9 |

With the equipment as presently set up, it is feasible to measure $^{129}I_2$ concentrations at atmospheric pressure (air) to concentration levels at or below 10^{-10} g/cm^3 . As this limit of sensitivity appears to be in the range required for applications of the proposed instrument, the specified equipment design will be built around measurements to be made in ambient air. This will simplify the design of the final instrument and will avoid the requirement for scrubbing I_2 and rediluting it into inert carrier gases. It will also avoid the possible pitfalls resulting from the tendency of I_2 to plate out and hang up on otherwise clean surfaces.

Measurements Under Flowing Conditions

At the present time we have not extended our measurements to flowing cells. To make laboratory measurements under these conditions will require:

- (a) A much larger inventory of radioactive $^{129}I_2$ than used in the static experiments.

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- (b) glassware, gas metering, and flow measuring equipment;
- (c) construction of flowing gas cells; and
- (d) an independent means of measuring the $^{129}\text{I}_2$ effluent from our experiments.

Such measurements are to be desired for several reasons:

- (a) the measurements will demonstrate that the technique can be transferred from measurements under static conditions to a performance level which will be required in the field;
- (b) an independent crosscheck can be made between this technique and a wet chemical analysis technique currently used in our laboratories;
- (c) the proposed monitor can be evaluated against radiocounting analysis; and
- (d) laboratory procedures need to be developed for use in evaluating prototype fluorescence detection instruments to be acquired.

IV. EVALUATION OF THE LASER FLUORESCENCE TECHNIQUE

Applications and Limits of Sensitivity

Laboratory measurements with the breadboard detector clearly demonstrated that an $^{129}\text{I}_2$ monitor based upon the laser fluorescence technique will be a sensitive, practical and relatively inexpensive field monitoring instrument for $^{129}\text{I}_2$ measurements down to or below the one hundred picogram/cm³ level. Such an instrument will be capable of real time instantaneous measurements or capable of operating in an integrating mode to give accumulation loads over a period of time. The components of the prototype instruments should be capable of continuous operation over many thousands of hours for use as a monitoring device. The field and prototype models will be essentially self-contained and readily portable for use in the field checking the integrity of power plant charcoal filters. The dynamic concentration range of the prototype and field instruments should be in excess of 10^5 and if required and specified will be capable of measurements over a range of 10^8 , i.e., up to $\sim 10^{-2}$ g/cm³ of $^{129}\text{I}_2$.

Interferences and Isotopic Scrambling

Measurements have been made with the breadboard detector for sensitivity using samples of $^{127}\text{I}_2$. The signals measured indicate that $^{127}\text{I}_2$ is detected with a sensitivity of less than 1% that of $^{129}\text{I}_2$. It is unclear at this point whether fluorescence signals from $^{127}\text{I}_2$ result from excitation by the plasma discharge light in the He-Ne laser or whether there exist very weak unmeasured $^{127}\text{I}_2$

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resonances in the region of the laser gain curve. These determinations can be made if the need warrants it.

If a very large excess of $^{127}\text{I}_2$ over $^{129}\text{I}_2$ is present in the effluent gas to be measured, a significant portion of the ^{129}I present will be tied up in the mixed isomer molecule $^{127}\text{I} - ^{129}\text{I}$. Our detection sensitivity for this molecule and other mixed isomer molecules is undetermined.

We are presently setting up the experimental apparatus to determine the effect of such isotopic scrambling. The tests will be conducted with known concentrations of the mixed isotopic species, and the importance of this scrambling will be assessed.

We are also concerned with interferences from other gaseous products of the fuel reprocessing cycle. At the present time, the only apparent interferent may be NO_2 . NO_2 is known to absorb over the entire visible region and fluoresces strongly. However, if NO_2 fluorescence is determined to be a strong interferent, several steps may be taken to minimize its effect on $^{129}\text{I}_2$ measurements:

- (a) the fluorescence may be blocked by a suitable combination of bandpass filters;
- (b) the NO_2 may be scrubbed before $^{129}\text{I}_2$ measurements are taken;
- (c) at pressures of ~ 1 atmosphere the quenching of NO_2 fluorescence results in a reduction of the fluorescence quantum yield by $\sim 10^5$ and so may pose no problem.

In any case, we are currently investigating the effect of NO_2 on the measurement of $^{129}\text{I}_2$ concentration.

It has been pointed out that a portion of the iodine present, particularly downstream from charcoal filter beds, is in the form of alkyl iodides (CH_3I , $\text{C}_2\text{H}_5\text{I}$, etc.). The laser fluorescence technique developed herein is completely insensitive to ^{129}I in any organic form. Work underway at NRL¹⁵ on a catalytic pyrolyzer has shown the capability of conversion of organic iodides to molecular iodide. If such a device is required, its inclusion immediately prior to the gas intake of the laser fluorescence detector can be provided for. This instrument should also serve to strip I_2 adsorbed on the surface of any particulate matter in gas stream to be detected.

Specification of Prototype Instrument

At this point laboratory studies have made it possible to specify many of the desirable design features which should go into the construction of the prototypes for $^{129}\text{I}_2$ field monitors. A contract has been let for the procurement of a prototype instrument.

TENTATIVE DETECTOR DESIGN

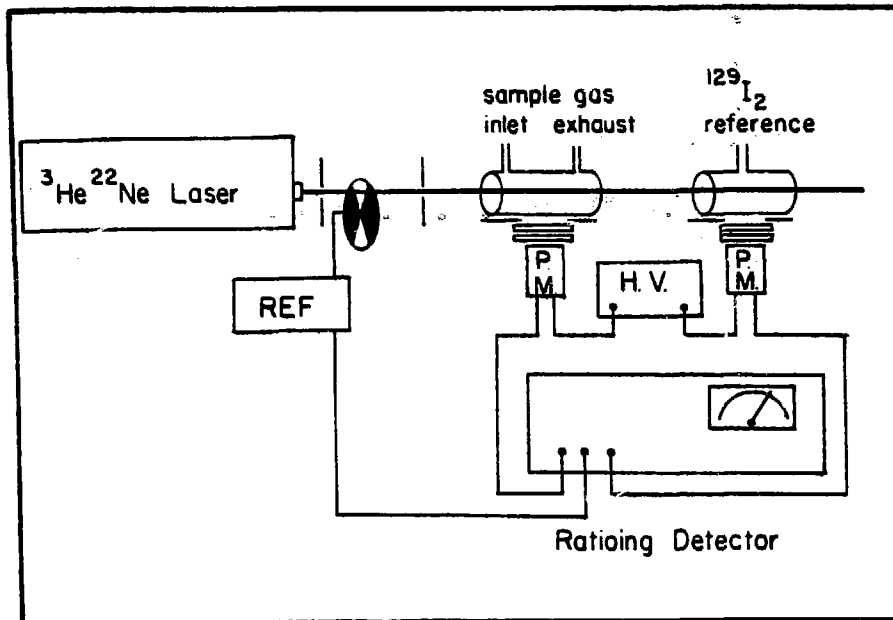


Fig. 4 — Schematic diagram of a proposed prototype iodine monitoring device.
 Explanations and possible changes are noted in the text.

A schematic block diagram of the proposed prototype detector is shown in Figure 4. The prototype instrument contains a reference sample of $^{129}\text{I}_2$ which will be used to ratio against the unknown samples to be measured. In this way, small variations in the laser power or alignment of components will not effect the accuracy of measurements. The fluorescence is monitored by lock-in detection, and the mechanical chopper has been replaced by a "Tuning Fork" chopper due to space limitations. The final design instrument contains microprocessing circuitry to allow automatic sensitivity changes to be made and to supply digital readout and integrated sample loadings. The prototype instrument also allows for some internal reference measurements (i.e., laser power, absolute reference signal intensity, background signal nulling, etc.) to aid in field adjustment and trouble shooting. The prototype system is of a ruggedized and modular design which will allow for ease of portability and operation. The only external connection which is required is a standard 110V ac power outlet.

Other Design Options

It should be noted that several options are feasible for incorporation into a laser fluorescence iodine monitor. The present studies have been built around an assumed need to specifically measure $^{129}\text{I}_2$. By changing the specifications of the excitation laser it is possible to build a detector which will detect essentially the sum of all isotopic I_2 species. Such a detector would not be

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specific for $^{129}\text{I}_2$, but if used in conjunction with the detector specified in this work, it would be possible to detect both $^{129}\text{I}_2$ and total I_2 .

If a requirement becomes apparent to measure much lower concentrations of $^{129}\text{I}_2$ than is feasible with the present detector, it should be possible to extend the limits of sensitivity of the proposed instrument by using I_2 scrubbing techniques for larger samples of air followed by measurement of $^{129}\text{I}_2$ in a carrier flow of helium. It would likely be possible to extend the sensitivity limits by several factors of 10 for the proposed instrument.

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DISCUSSION

DUGGAN: Two questions: can you give us an idea of the approximate cost of this instrument, and also what specific applications do you see for it?

MCDONALD: With respect to cost, the components for the instrument are under \$3,000 off the shelf. The prototypes cost about \$40,000. We have a guarantee from the supplier to make more for under \$20,000. I don't know what they will finally cost if they are produced in bulk. Initially, it will be tested in the field stack to measure effluent wastes.

PARKER: There's a possibility this instrument could be recommended for use in reprocessing offgas streams for measurement of iodine. Have you taken a look at all the possible constituents that might be in dissolver offgas that could interfere? I am specifically thinking of chlorine compounds.

MCDONALD: I have talked to many people about what substances might be present in offgas effluents. The only molecule I can identify that absorbs at 6328 Angstroms is NO₂. There are no chlorides or other halogens that are close to that absorption region. Molecular bromine might be present, but it will be several orders of magnitude below iodine.

HULL: This is a comment, rather than a question. This is a very elegant paper; I am very impressed with what you have done. But it seems to me, as the

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previous questioner has suggested, that if the device described has any application, it is to reprocessing facilities. Your paper mentions power reactors, for which, it seems to me, the cost would be all out of proportion to the value of the measurements. It seems to me that the driving force for this, as well as for the removal of ^{129}I from offgas streams, which was described yesterday, is EPA's release standard in 40CFR190. I suggest that even if one is a devout believer in the linear hypothesis, that the cost per health effect averted from the application of this standard is preposterous, especially when the cost of the development and application of your instrument is added to the costs EPA identified in their cost-benefit analysis. I feel that we should try to keep in mind the reasonableness of what we are trying to accomplish, as well as the challenge to our technical abilities.

McDONALD: The use of detectors such as I have described by the nuclear power industry involves economic, political, and emotional decisions in which I am not, and do not wish to be, involved. Our task was to develop this detector because DOE believed that it was in their best interests to determine its feasibility. The modest detector which we have designed is likely not nearly sensitive enough to measure ^{129}I concentrations at power facilities at the present time. I do feel, however, that DOE and other facilities are correct in being far-sighted enough to invest in R & D for instrumentation not required by current effluent standards but which may be vitally needed in 5-10 years. The development time for such diagnostics strongly mitigates against their development after the fact.

GIBSON: In the figure with the schematic diagram, you show the laser beam first absorbed by the sample cell and then passing on to the reference cell. Could you explain why this is done instead of having the beam split for the sample and reference cells?

McDONALD: Any conceivable $^{129}\text{I}_2$ concentration in the sample cell will absorb only a negligible fraction of the HeNe laser light. More important, if there were a high concentration of noniodine absorbing species such as NO_2 , or scatters, the light subsequently impinging on the reference cell will be reduced in intensity as it was in the sample cell, thus giving a ratio signal which is representative of the light available to each cell.

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AN INORGANIC RADIOIODINE ADSORBER WITH LOW NOBLE GAS RETENTION*

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Abstract

A silver loaded silica gel was developed as an adsorber for air monitoring subsequent to a release from containment power reactor accident. Requirements of high efficiency for known radioiodine species under wide ambient conditions of humidity and temperature and low noble gas adsorption efficiency are satisfied by the material.

Silver loadings from 2 to 24% by adsorber weight have been tested against organic radioiodine, hypiodous acid, elemental radioiodine, and noble fission gases. Relative humidity was varied between 5 and 99%, and stay times of .11, .073, and .055 seconds were used.

Silver loading requirements depend on sampling duration and relative humidity. Environmental monitoring, to define safe living areas, requires about 25 ft³ of air be sampled and analyzed by the proposed system of an air mover, an adsorber, and a civil defense readout instrument. A silica gel adsorber can be used with a 4% silver loading for an efficiency of better than 93% with a .11 second stay time, and for all ambient conditions tested. Similar tests using 4% silver loaded 13X molecular sieve or about 60% silver zeolite yielded lower efficiencies.

Xenon adsorption was $<5 \times 10^{-3}\%$ at 55°C with no post-release flushing. This value was about 1/20 of the value for charcoal under the same conditions.

A silver loading apparatus was developed to easily process ~5 Kg of silica gel per day. The method involves prehumidification, boiling under high vacuum, oven drying, sieving, and finally packaging.

I. Introduction

Radioactivity releases from power reactor containments under accident conditions are expected to be extensive and will spread over a large geographic area. Concentrations of pertinent radioisotopes can be predicted mathematically, but with an uncertainty that will increase with distance from the reactor. Inhalation exposure will depend significantly on atmospheric stability and vertical cloud dispersion. Evacuation of personnel will be governed by thyroid dose commitments and whole body exposures. The lack of reliable air concentration predictions and the need to define evacuation boundaries combine to require air concentration measurements be made in the environment.

* Research performed at Brookhaven National Laboratory under contract with the U.S. Nuclear Regulatory Commission.

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The probability of a reactor accident was estimated⁽¹⁾ to range from 1×10^{-4} to 5×10^{-7} per reactor year. The less probable, and more damaging, accidents would have U.S. probability of about 2×10^{-3} per hundred power reactors life times of 20 years assumed per reactor. The small aggregate accident probability and large geographic area potentially involved lead to a desirable solution of a significant number of inexpensive air sampling apparatus which can be stored indefinitely.

Costly measurement methods using gamma analysis can be avoided by developing a sample specifically for iodine, thereby permitting any beta or gamma detector to be used for measurement. Particulate fission products include dozens of noniodine radionuclides. Use of a prefilter⁽²⁾ before the adsorber bed separates the activity into gaseous and particulate fractions, and allows a determination of gaseous radioiodine.

Adsorption of fission gases relative to iodine can be reduced by using an appropriate inorganic adsorber. Several commercial inorganic adsorbers were tested, but were too expensive or inefficient for the organic or hypoiodous acid forms of iodine. A silver impregnated silica gel adsorber was developed that had over 90% efficiency for collection of radioiodine for sampling times of several minutes. The material provided corresponding Xenon efficiencies of less than .04% at temperatures above 7°C.

The properties of the silver loaded silica gel adsorber were used in conjunction with vacuum cleaner air movers and Civil Defense GM survey meters to make possible economical air sampling and assessment following release from containment accidents.

Adsorber Loading Method

Solutions of silver nitrate were used to impregnate molecular sieve and silica gel. Since cost is an important factor, silver loadings of less than 8% by weight were extensively studied. Loadings of >20% by weight were easily accomplished with about the same loading efficiency.

The loading method⁽³⁾ started with prehumidification of factory dry silica gel⁽⁴⁾ material. Keller⁽⁵⁾ found that dry zeolite pore structures would fracture with addition of liquid water. Prehumidification with moist air allowed the heat of adsorption to be controlled to prevent fracturing.

Kilogram quantities of dry silica gel sieved to >30 mesh were prehumidified by 4 liters per minute of lab air bubbled through distilled water maintained at 60°C. The process required about 18 hours, overnight, for complete saturation. The wet gel was slurried with an aqueous solution of the silver salt, and boiled under vacuum for about 2.5 hours. After boiling, the material was transferred to drying trays and heated in an air-circulated oven at 105°C for 24 hours. The dried adsorber was resieved and the >30 mesh fraction was stored in clean gallon sized metal paint cans ready for use.

Loading efficiency was studied⁽⁶⁾ by measuring the quantity of silver on the absorber surface. This was accomplished by tumbling the silica gel pellets for 4 to 76 hours to allow self abrasion to remove the surface layer. Three fractions, which proved to be time independent, were developed by sieving, using 30, and 80 mesh screens. Nearly all of the material was retained on the 30 mesh screen, and broken pellets, corners and sharp edges were trapped on the 80 mesh screen. Particles smaller than 80 mesh were assumed to represent the silica gel pellet surface and constituted less than 0.2 by weight of the total. Analysis for silver

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indicated that about .2 to .4% of the silver was on the <80 mesh fraction for 4 to 24% silver loaded silica gel stock. Clearly the pore loading efficiency was better than 99.6% since some part of the <80 mesh fraction consisted of pellet fragments.

A water leaching experiment⁽⁶⁾ was conducted with prehumidified 4% silver loaded silica gel. A silver recovery efficiency of $88\% \pm 3$ indicated that the impregnated silver was in the nitrate form since the oxides and nearly all silver salts are not very soluble. Further, the silver nitrate appeared to be weakly held by the silica gel.

CH₃I Generator

A fission iodine generator was constructed⁽⁷⁾ using <3 micron thick ²³⁵U alloy foil positioned around a 10^8 neutron/sec AmBe source. Approximately 3m² of foil, containing 16.2 gm of ²³⁵U, was assembled in a 12 cm wide strip. The foil was positioned on a continuous strip of .15 cm thick by 12 cm wide open mesh polyethylene-polypropylene and wound on a form around the neutron source. The resulting 22 cm spool was placed in an aluminum pressure tank and all voids, except in the mesh, were filled with paraffin. The assembly provided an average thermal neutron fluence of about $7 \times 10^4 \text{ cm}^{-2} \text{ sec}^{-1}$ along the outer end of the spool.

The pressure chamber, filled with argon gas, 180 psig, containing .3% CH₄, was used for iodine exchange reactions. Approximately 5.4% of the ¹³⁵I theoretical yield was measured, corresponding to a CH₃¹³⁵I activity of 0.16 microcuries. A method outlined by Keller⁽⁸⁾ was used to test and confirm the chemical species. The species differentiator indicated the discharged mixture consisted of 95 to 99% organic iodine and 1 to 5% HOI. Since both oxygen and water are absent in the generator, the HOI indication was probably due to HI.

The generating method used had the advantage of also producing all of the fission gases; so that tests were made with noble gases as well as organic radioiodine.

HOI Generator

The HOI generator was patterned after Kabet^(9,10,11). Carrier free ¹³¹I was added to a $3 \times 10^{-5} \text{ M KIO}_3$ water solution. The pH was adjusted to 2.0 with H₂SO₄, and the mix was placed in a fritted bubbler. Helium gas was used to strip HOI from the solution. Demisting was accomplished with a Gelman glass fiber filter. A second helium stream of about the same flow rate, .04 CFH, was used to dry the combined stream prior to passing through a stack of six 40 mesh copper screens described by Barry⁽¹²⁾. The screens were routinely pickled in HI, washed in distilled water, and air dried immediately before use.

The Keller species differentiator indicated that about 95% of the iodine was adsorbed on the p-iodaphenol impregnated alumina adsorber. The remaining 5% was trapped on the TEDA impregnated charcoal backup filters.

I₂ Generator

Convenience and economy of multiple use prompted the conversion of the easily generated and controlled HOI into I₂. This required the reduction of I⁺ by oxidation of Cr₂O₃ at 510°C.

The HOI - He generator stream was passed through a 1 cm diameter quartz tube mounted in a tube furnace. Glass wool was used to center the C₂O₃ in the furnace. Nichrome heating wire was wound around the discharge end of the quartz tube and the teflon tubing used to transfer the I₂ to the air stream leading to the test adsorber

Table I. CH₃I Adsorption Tests

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp,
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|------------|-------------------|--------------|---------------------------|-------------|--------------------|----------------|----------------------|------------------|------------|
| Silica Gel | 0 | 6x16 | 45 | 41 | - | 23 | .11 | - | .01 |
| Silica Gel | 0 | | 84 | 40 | - | 25 | .11 | - | .003 |
| Silica Gel | 2% Ag | 6x16 | 17 | 66 | - | 23 | .11 | - | .99+ |
| Silica Gel | 2% Ag | | 80 | 41 | - | 26.5 | .11 | - | .75 |
| Silica Gel | 3.5% Ag | 6x16 | 17 | 66 | - | 23 | .11 | - | .99+ |
| Silica Gel | 4% Ag | 6x16 | 6 | 47 | - | 8 | .08 | - | .99+ |
| Silica Gel | 4% Ag | | 7 | 49 | - | 8 | .05 | - | .98 |
| | 4% Ag | | 36 | 44 | - | 8 | .08 | - | .93 |
| | 4% Ag | | 35 | 45 | - | 8 | .05 | - | .89 |
| | 4% Ag | | 67 | 42 | - | 8 | .11 | - | .98 |
| | 4% Ag | | 67 | 42 | - | 8 | .08 | - | .94 |
| | 4% Ag | | 86 | 40 | - | 13 | .11 | 2 | .79 |
| | 4% Ag | | 88 | 40 | - | 24 | .11 | - | .94 |
| | 4% Ag | | 90 | 40 | - | 24 | .11 | - | .85 |
| | 4% Ag | | 97 | 42 | - | 8 | .11 | - | .93 |
| | 4% Ag | | 96 | 42 | - | 8 | .05 | - | .92 |

(cont'd)

Table I. continued

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp.
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|------------|-------------------|--------------|---------------------------|-------------|--------------------|----------------|----------------------|------------------|------------|
| Silica Gel | 4% Ag | | 98.5 | 34 | - | 8 | .05 | - | .81 |
| Silica Gel | 6% Ag | 6x16 | 98 | 34 | - | 8 | .05 | - | .96 |
| Silica Gel | 8% Ag | 6x16 | 1 | 110 | 2 | 22 | .11 | - | .99+ |
| | 8% Ag | | 8 | 28 | - | 8 | .05 | - | .99+ |
| | 8% Ag | | 17 | 71 | - | 27 | .11 | - | .99+ |
| | 8% Ag | | 22 | 35 | - | 25 | .11 | - | .99+ |
| | 8% Ag | | 36 | 48 | - | 8 | .11 | - | .99+ |
| | 8% Ag | | 36 | 48 | - | 8 | .08 | - | .98 |
| | 8% Ag | | 35 | 48 | - | 8 | .05 | - | .97 |
| | 8% Ag | | 72 | 45 | - | 8 | .08 | - | .96 |
| | 8% Ag | | 62 | 48 | - | 8 | .05 | - | .93 |
| | 8% Ag | | 85 | 37 | - | 23 | .11 | - | .97 |
| | 8% Ag | | 85 | 38 | - | 13 | .11 | 2 | .94 |
| | 8% Ag | | 92 | 37 | 40 | 24 | .11 | - | .37 |
| | 8% Ag | | 97.5 | 40 | - | 8 | .11 | - | .97 |
| | 8% Ag | 6x16 | 96 | 39 | - | 8 | .08 | - | .98 |

(cont'd)

Table I. continued

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp.
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|-------------------|-------------------|--------------|---------------------------|-------------|--------------------|----------------|----------------------|------------------|------------|
| Zeolite | ~60% Ag | 2 0x40 | 98 | 34 | - | 8 | .05 | - | .81 |
| Zeolite | ~60% Ag | | 98 | 34 | ~40 | 27 | .08 | - | .82 |
| Zeolite | ~60% Ag | | 98 | 35 | ~40 | 28 | .034 | - | .48 |
| 13X Mole
Sieve | 4% Ag | 8x12 | 99 | 32 | - | 8 | .05 | - | .06 |
| Char. G215 | (R-51)AO | | 88 | 36 | - | 9 | .04 | - | .75 |
| Char. G618 | TEDA | 18x30 | 91 | 32 | ~ 1 | 8 | .113 | 11@110°C | .95 |
| | TEDA | | 95 | 33 | ~ 1 | 8 | .106 | 11@110°C | .96 |
| | TEDA | | 75 | 35 | ~ 1 | 6 | .06 | 11@110°C | .87 |
| Char. W-337 | None | 8x16 | 67 | 45 | 24 | 8 | .44 | 8 | .99 |
| | None | | 95 | 33 | 120 | 8 | .44 | 10 | .97 |
| | None | | 96 | 34 | 70 | 8 | .05 | 10 | .26 |
| Char. BC845 | TEDA | 14x30 | 97 | 33 | - | 8 | .05 | - | .99+ |
| Char. BC540 | KI ₃ | 14x30 | 97 | 40 | - | 8 | .05 | - | .88 |
| MSA #2133 | Char. | Cloth | 44 | 35 | - | 8 | .8CFM | - | ~.01 |

Table II. HO^{131}I Adsorption Tests

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp.
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|------------|-------------------|--------------|---------------------------|-------------|--------------------|----------------|----------------------|------------------|------------|
| Silica Gel | 0 | 6x16 | <10 | 31 | - | 28 | .11 | - | .82 |
| | 0 | | 85 | 42 | - | 26 | .11 | - | .51 |
| | 0 | | 88 | 40 | - | 26 | .11 | - | .31 |
| Silica Gel | 4% Ag | 6x16 | <10 | 33 | - | 26 | .11 | - | .93 |
| | 3.7% Ag | 6x16 | 13 | 41 | - | 8 | .11 | - | .99+ |
| | 3.7% Ag | | 12 | 43 | - | 8 | .08 | - | .99+ |
| | 3.7% Ag | | 13 | 43 | - | 8 | .05 | - | .99 |
| | 4.0% Ag | | 35 | 35 | - | 8 | .05 | - | .99+ |
| | 4.0% Ag | | 45 | 36 | - | 8 | .11 | - | .97 |
| | 4.0% Ag | | 65 | 40 | - | 8 | .05 | - | .97 |
| | 4.0% Ag | | 75 | 41 | - | 8 | .11 | - | .99+ |
| | 4.0% Ag | | 85 | 40 | - | 26 | .11 | - | .71 |
| | 4.0% Ag | | 86 | 42 | - | 28 | .11 | - | .81 |
| | 4.0% Ag | | 95 | 41 | - | 30 | .11 | - | .76 |
| | 4.0% Ag | | 97 | 42 | - | 8 | .11 | - | .99+ |
| | 4.0% Ag | | 97 | 42 | - | 8 | .08 | - | .98 |
| | 4.0% Ag | | 97 | 43 | - | 8 | .05 | - | .74 |

(cont'd)

Table II. continued

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp.
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|-------------------|-------------------|--------------|---------------------------|-------------|--------------------|----------------|----------------------|------------------|------------|
| Silica Gel | 4.0% Ag | | ~100 | 40 | - | 16 | .11 | 14 | .63 |
| Silica Gel | 8.0% Ag | 6x16 | 5 | 51 | - | 8 | .08 | - | .99 |
| | 8.0% Ag | | 5 | 52 | - | 8 | .05 | - | .98 |
| | 8.0% Ag | | 40 | 37 | - | 8 | .11 | - | .99+ |
| | 8.0% Ag | | 50 | 38 | - | 8 | .05 | - | .79 |
| | 8.0% Ag | | 63 | 40 | - | 8 | .11 | - | .99+ |
| | 8.0% Ag | | 68 | 37 | - | 8 | .05 | - | .98 |
| | 8.0% Ag | | 86 | 37 | - | 28 | .11 | - | .71 |
| | 8.0% Ag | | 96 | 40 | - | 8 | .11 | - | .99+ |
| | 8.0% Ag | | 96 | 40 | - | 8 | .08 | - | .99 |
| | 8.0% Ag | | 98 | 39 | - | 8 | .05 | - | .92 |
| Zeolite | ~60% Ag | ~20x40 | ~100 | 39 | - | 30 | .11 | - | .26 |
| 13X Mole
Sieve | 4% Ag | | 75 | 40 | - | 26 | .11 | - | .18 |
| Char. W-337 | 0 | 8x16 | 71 | 45 | 15 | 8 | .44 | 8 | .99+ |
| | 0 | | 96 | 39 | 15 | 8 | .44 | 8 | .99+ |
| | 0 | | 98 | 35 | 40 | 8 | .05 | 10 | .66 |

(cont'd)

Table II. continued

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp.
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|--------------|-------------------|--------------|---------------------------|-------------|--------------------|----------------|----------------------|------------------|------------|
| BC-845 | TEDA | 14x30 | 7 | 52 | - | 8 | .05 | - | .99 |
| | TEDA | | 96 | 42 | - | 8 | .11 | - | .99+ |
| | TEDA | | 95 | 43 | - | 8 | .08 | - | .99+ |
| | TEDA | | 95 | 42 | - | 8 | .05 | - | .99 |
| Char. BS-540 | KI ₃ | 14x30 | 9 | 49 | - | 8 | .05 | - | .99 |
| | KI ₃ | | 97 | 40 | - | 8 | .11 | - | .99+ |
| | KI ₃ | | 97 | 40 | - | 8 | .08 | - | .99+ |
| | KI ₃ | | 96 | 40 | - | 8 | .05 | - | .99+ |
| MSA-2133 | Char. | Cloth | ~100 | 38 | - | 15 | 3.8CFM | - | <.01 |

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chamber. A minimum transfer line temperature of 100°C was maintained with an average temperature of >160°C.

A modified Keller species differentiator was used to analyze the I_2 discharge stream. The elemental iodine fraction was measured with a bank of 6 Barfy (12) screens replacing the standard cadmium iodide loaded chromasorb. Results indicated that 18% of the iodine was on the Gelman glass filter, followed by 73% on the screens, 2% on the HOI adsorber, and finally by 7% on the TEDA impregnated charcoal. The 18% trapped by the glass filter was probably due to adsorption rather than particulate filtration. In any event, at least 91% of the total was elemental iodine.

Results

Attempts were made to summarize the results graphically. However, the number of parameters were too extensive requiring a tabulation of important variables. Some of the parameters may require specification. Pre-run, run and flush times refers to the number of minutes that the temperature and humidity controlled air passes through the trial adsorber. Test CH_3I , HOI, I_2 , or Xe were released during the run stage. Flow conditions were set by the stay time defined as the quotient of adsorber bed volume and volume flow rate.

In general the data follow increasing silver impregnation from 0 to 8%, decreasing stay time, and increasing relative humidity. Certain other inorganic adsorbers results are next with charcoal and a cloth filter material completing the list.

Adsorber criteria (7) dominated the testing scheme. In general the adsorber material had to be highly efficient, better than 90%, for any known species of iodine under all expected conditions of temperature and humidity. Sampling time was changed in mid-experiment from a maximum of 15 minutes to 5; so that two sampling times appear in the data. Finally the adsorber must be at least 2 orders of magnitude less efficient for noble gases than iodine.

Table I summarizes the CH_3I results. The unimpregnated silica gel is inefficient for CH_3I adsorption. For high humidity and a stay time of 0.11 seconds, 4% silver loaded silica gel had an adsorption efficiency of 93% for an 8 minutes sampling time. Increasing the silver content increased the efficiency; but was not considered to be cost effective.

The commercial silver zeolite had a surprisingly low efficiency considering the >60% silver loading and small particle size.

Charcoal impregnated with TEDA is more efficient than 4% Ag - silica gel. However, high noble gas adsorption efficiency of several percent, Table IV, precluded its use.

HOI results are presented on Table II. It is interesting to note that HOH and HOI have one polar end in common. Hydrophilic adsorbers, such as silica gel, should be efficient for HOI, and this was confirmed with silica gel. Increasing silver impregnation tends to increase adsorption efficiency, but again 4% Ag - silica gel is adequate.

Silver zeolite had a poor efficiency of 26%. Addition of a 30 minute equilibration run under test conditions of temperature, humidity, and flow rate apparently saturated the zeolite with water thereby reducing efficiency. The same effect was expected and noticed during the longer runs with silica gel.

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The charcoal HOI results were similar to the organic iodine test. All of the charcoals were highly efficient. Impregnation doesn't appear to be as important for HOI as for CH_3I .

As expected the elemental iodine results, Table III, are uniformly good. The highly reactive atomic and molecular iodine is attracted by any surface cooler than the sublimation temperature such as the unimpregnated silica gel.

The tests reflecting the Xenon results were more involved. A mixture of noble gases and organic iodines from the fission generator was split into two flow rate monitored and controlled streams. Room temperature, silver zeolite filters were used in each leg to quantitatively trap the iodines. One of the streams was sent to an air duct leading to the test chamber, and the other to two sequential charcoal adsorbers maintained at dry ice temperatures.

Ge(Li) analysis of the 250 keV ^{135}Xe line indicated that all of the cold leg Xenon, >99%, was trapped on the zeolite and first cold charcoal adsorber. Analysis of the radioiodine activities in both legs provided reliable flow fractions between the two legs to supplement flow meter results.

Xenon results, Table IV, indicate that silica gel is about $.09 \div .0004 \sim 200$ less efficient than TEDA loaded charcoal at about 10°C . The charcoal results are comparable to earlier work shown in Figure 1. The two BC 845 charcoal results fall slightly below the results obtained independently by Seigworth⁽¹³⁾ and Collins⁽¹⁴⁾.

(2) Ratios of Xe to I counting rates expected for perfect adsorbers were calculated for the nine pressurized water and boiling water reactor accidents postulated in WASH 1400⁽¹⁾. Selection of 4% Ag - silica gel would allow clear iodine detection in all but the BWR 5 case. At 7°C ambient air temperature, the iodine concentration would be overestimated by a factor of 2.7 for 2 hours total decay and 7.5 for 24 hours. Since the average temperature is $>7^\circ\text{C}$ and the adsorber bed temperature is normally appreciable due to water adsorption the overestimation will be largely reduced. For typical conditions of 20°C and 50% RH the air leaving the bed was measured to 20°C higher than the entering air. This would reduce the overestimate by a factor of 3 to 5. Further the magnitude of the radioactive release for the BWR 5 case is the smallest of the cases considered, and would result in a total iodine release of only about 2 curies. This would lead to a very small iodine exposure problem.

The ideal adsorber for radioiodine monitoring subsequent to a release from containment accident should have the following properties:

- a. The adsorber should be highly efficient for all species of iodine expected in the environment.
- b. Noble gas adsorption efficiency must be at least two orders of magnitude smaller than the iodine.
- c. Long term storage should not degrade the adsorber.
- d. The adsorber must be economical as hundreds of kilograms are required.

All of criteria are satisfied by 4% silver loaded silica gel in a system requiring a sampling time of several minutes and a .11 stay time.

Table III. $^{131}\text{I}_2$ Adsorption Tests

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Temp
°C | Pre-Run
Minutes | Run
Minutes | Stay Time
Seconds | Flush
Minutes | Efficiency |
|------------|-------------------|--------------|---------------------------|------------|--------------------|----------------|----------------------|------------------|------------|
| Silica Gel | 0 | 6x16 | 97 | 37 | 0 | 8 | .05 | - | .92 |
| Silica Gel | 4% Ag | 6x16 | 8 | 46 | - | 8 | .06 | - | .99+ |
| | | | 37 | 36 | - | 8 | .06 | - | .99+ |
| | | | 66 | 36 | - | 8 | .06 | - | .99+ |
| | | | 95 | 34 | - | 8 | .06 | - | .99+ |
| | | | 98 | 36 | - | 8 | .05 | - | .98 |
| Silica Gel | 8% Ag | 6x16 | 98 | 35 | 30 | 10 | .05 | - | .94 |

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Table IV. ^{135}Xe Adsorption Tests

| Material | Impregnant
W/O | Mesh
U.S. | Relative
Humidity
% | Inlet Temp.
°C | Outlet Temp.
°C | Run Time
Minutes | Stay Time
Seconds | Efficiency |
|-------------|-------------------|--------------|---------------------------|-------------------|--------------------|---------------------|----------------------|--------------------|
| Silica Gel | None | 6x16 | 45 | 39 | 45 | 23 | .11 | 3×10^{-5} |
| Silica Gel | 4% Ag | 6x16 | 75 | 39 | 59 | 25 | .11 | 5×10^{-5} |
| Silica Gel | 0 | 6x16 | 84 | 40 | 61 | 25 | .11 | 4×10^{-5} |
| Silica Gel | 4% Ag | 6x16 | 34 | 7 | 12 | 10.5 | .11 | 4×10^{-4} |
| Char. G618 | TEDA | 18x30 | | 125 | 119 | 18 | .11 | 1×10^{-3} |
| Char. BC845 | TEDA | 14x30 | 34 | 12 | 12 | 10.5 | .006 | 5×10^{-3} |
| | | | | | | Normal to | .11 | 9×10^{-2} |

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Table V. Xenon to Iodine Shielded DCV 700 Response
for Various WASH 1400 Containment Releases

| | Xe/I | |
|-------|------------------------|-----------------|
| | ----- Decay Time ----- | |
| | <u>2 hours</u> | <u>24 hours</u> |
| PWR 1 | .04 | .10 |
| 2 | .04 | .10 |
| 3 | .11 | .30 |
| 4 | .18 | .51 |
| 5 | .25 | .71 |
| 6 | 2.97 | 8.38 |
| 7 | 4.17 | 11.70 |
| 8 | .53 | 1.50 |
| 9 | .78 | 2.20 |
| BWR 1 | .07 | .19 |
| 2 | .03 | .09 |
| 3 | .26 | .74 |
| 4 | 11.10 | 31.20 |
| 5 | 6700.00 | 18900.00 |

Xe includes 133, 135, 138

I includes 131, 132, 133, 134, 135

$$Q_m = \frac{\mu \text{Ci/g CHARCOAL}}{\mu \text{Ci/cc CARRIER GAS}}$$

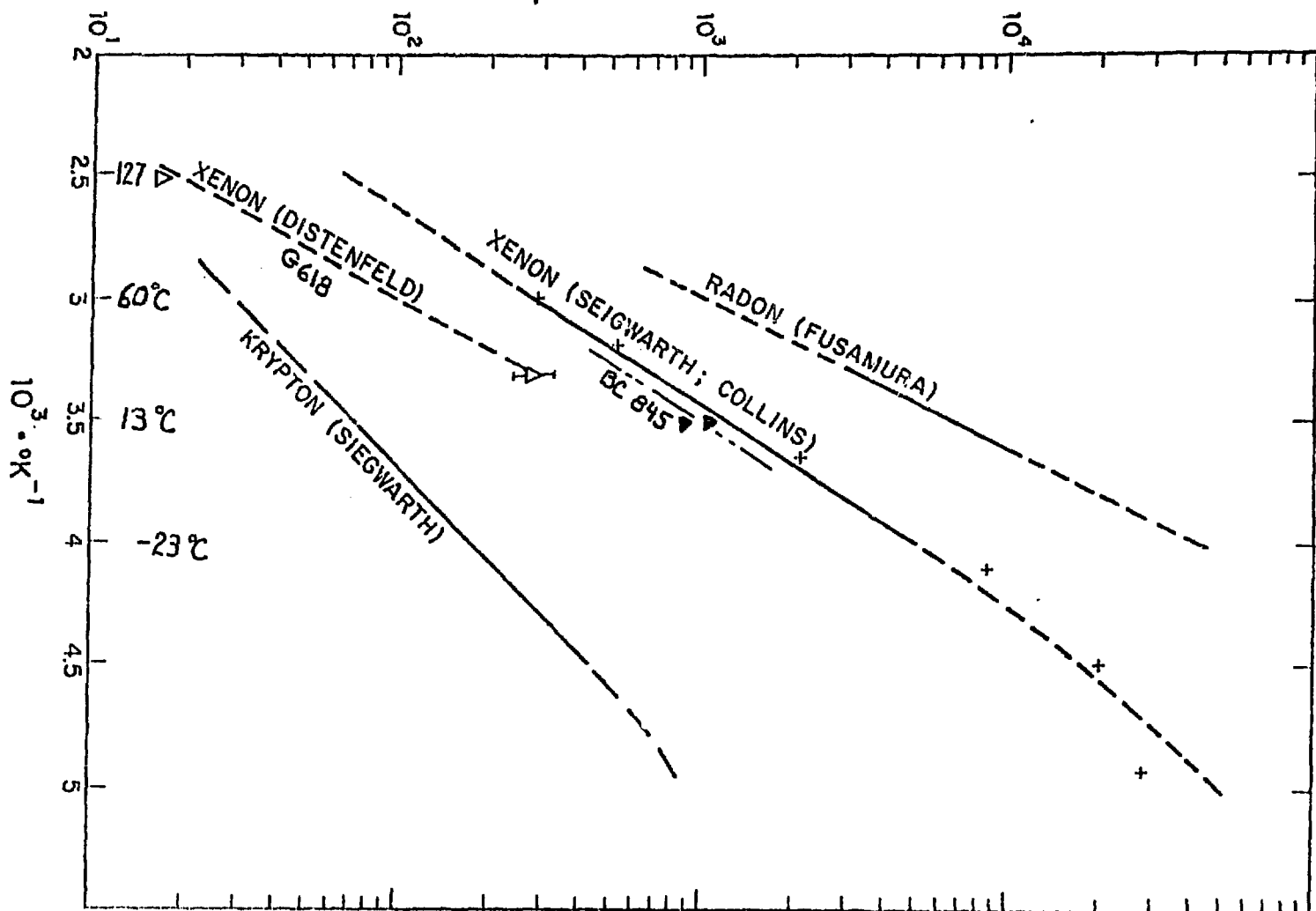


Figure 1. Temperature dependence on noble gas adsorption on charcoal.

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DISCUSSION

DEUBER: In many of your experiments you had no preconditioning. Do you expect reasonable results without preconditioning?

DISTENFELD: The adsorber is used in the field without preconditioning of any sort. In fact, the adsorber must be stored away from moisture prior to use. This is done so that the laboratory tests reflect field conditions and are not conservative.

DEUBER: Can you really expect a reasonable retention efficiency if you do no preconditioning?

DISTENFELD: Yes, I have demonstrated that in my paper.

DEUBER: What will be the sampling time for measurements in the field?

DISTENFELD: The collecting cycle will be five minutes and all tests were run at eight minutes, so that the adsorber was still unsaturated, with respect to water, at the end of the collection cycle. Therefore, I expect better results in the field than in the laboratory, i.e., 95 to 99% efficiency for all species of iodine.

DEUBER: Other iodine species than those examined in your tests, such as organic iodides, might occur in the case of an accident. These might be retained to a lesser extent by your material. Shouldn't it, therefore, be more reasonable to sample with impregnated carbon and flush off the rare gases if necessary? We have made measurements with other species than those mentioned by you, and when we find good retention with charcoal, the retention is always very poor with inorganic adsorbers impregnated with silver nitrate.

DISTENFELD: We considered doing that very thing; in fact, the original work was a heating operation while sampling. Alternately, a heating operation afterwards in clean air. But, a containment loss accident will cause contaminated areas that may extend for several tens of kilometers. A flushing operation would have to be done at high temperature, 110°C or more, for 10 to 15 minutes, and in uncontaminated air according to studies reported in BNL 23112 and BNL 21541. Because of these restrictions, and the power needed for hot flushing, we developed the silver loaded silica-gel as the alternative and better solution. I understand that higher polymers or higher organics are even more efficiently absorbed.

DEUBER: Just to give you one example, we made measurements with iodine benzene and we found that retention with inorganic absorbers was very poor compared to charcoal.

DISTENFELD: I am not arguing that our material has a better absorption efficiency. I am saying that, under the conditions of the test, the absorption efficiency was fully adequate, i.e., in excess of 90%.

HULL: Dr. Deuber's paper yesterday, on species identification in nuclear power plants and your paper today indicate that there is some question about the presence of hypoiodous acid and, particularly how much hypoiodous acid there might be in an accident situation. If there are any questions about the ability of charcoal systems to retain HOI, even if impregnated, as compared to silica gel, perhaps we should be looking at correctly employed strategies--particularly for incident situation--if a large amount of HOI is anticipated in them. At Brookhaven we have a system for the production of ^{124}I and ^{126}I by the irradiation of CsCl targets by protons underwater in a tank, in which there was a recent minor incident producing an air release of the ^{124}I and ^{126}I . This was completely un-

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detected by conventional (unimpregnated) charcoal samples both in the stack and in the field. However, silica gel samples for tritium showed unusual counts in the high energy region in liquid scintillation counting. When run on a Ge-Li detector, the ^{124}I and ^{126}I were readily apparent. This is a form of iodine that was completely undetected by the conventional charcoal systems for detecting iodine and there was so little of it that it was called undetectable but it was quite obvious on the silica gel system. Seriously, it is hypiodous acid and, if this is so, we had better start taking a look at what we expect from this situation, particularly in an incident.

KOVACH: Before we go further, I would like to mention that I certainly would not call an unimpregnated carbon sampler a "conventional system." I don't think any power reactor would have anything to do with an unimpregnated charcoal sampler for iodine.

HULL: I was talking about using them for field sampling, particularly if you are not looking for organic compounds in the field.

DISTENFELD: Impregnated charcoals should always be used because of much better weathering resistance and efficiency for other than I_2 iodine species. Stay time should be maintained at a reasonable level: approximately ≥ 0.1 seconds.

DEUBER: After five years of operating iodine monitors in German reactor stations with silicic acid impregnated silver (published in 1974 in the 13th Air Cleaning Conference), we found that there are still problems. Figures for xenon retention and a description of the instrument used are given in this reference. We found that one of the problems is that in between the granules of the sorption material there is a gas volume containing rare gas. You can't avoid it. The only way to get rid of it is to flush it with fresh air. This is important if you measure the offgas of boiling water reactors, as we do continuously, because sometimes the level of rare gas is so much higher than the level of iodine. We tested the instrument and found it can be flushed out in five minutes with fresh air from pressurized tanks. But this is not the only problem. Cesium and rubidium, the daughter products of xenon and krypton, pass through the aerosol filter in the form of gas and stick partly to the sorption material. We are not through with that problem, at the moment. I only wanted to point out that the measurement of very small amounts of iodine in the environment, at the level of rare gases, is still a problem.

DISTENFELD: The retention data that I showed did not include flushing. The efficiency cited was for a non-flush system.

WILHELM: Radioactive particles of the daughter products Rb and Cs from Kr and Xe may influence the measurements, as we learned from iodine monitoring in the exhaust air of a BWR. They may be generated after the air has passed the aerosol filter. Those measurements were performed with AC 6120, a silver nitrate impregnated silic acid, which is regularly used as an iodine sorption material in the exhaust air of German LWRs for monitoring with a continuously working iodine monitor.

SIPP: We also measure iodine in the environment with cartridges to determine iodine species. It started four years ago when we had leaky fuel. Sampling showed about a 33% split between methyl iodide, hypiodous acid, and elemental iodide. So, we've had no problems with halogens or inert gases in environmental monitoring. It looks like such sampling would be useful during an accident situation where there would be a larger amount of xenon, and I think it's very worthwhile.

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CLOSING REMARKS OF SESSION CHAIRMAN:

KOVACH:

I always feel at these conferences that I should remind people that there were other air cleaning conferences before this one and that it would be very helpful if they would all go through the Proceedings that have been published from the Second Air Cleaning Conference on. We can learn a lot of things from what has been done before. Inevitably, by attending the Conferences, you see a certain cycling. Some of the problems come up and some of the same solutions also come up. So, I think it would be very beneficial for all of us if we would go back and see what was done in the early days. Some of that work isn't so bad.

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SESSION XI

LUNCHEON MEETING

Wednesday, August 9, 1978

CHAIRMAN: D. W. Moeller

SOME PERSPECTIVES ON RADIATION PROTECTION Dr. Warren K. Sinclair

OPENING REMARKS OF SESSION CHAIRMAN:

The year, 1978, marks the 50th anniversary of the founding of the National Council on Radiation Protection and Measurements. Today we are honored to have with us as our luncheon speaker, the President of the Council, Dr. Warren K. Sinclair.

Dr. Sinclair is a native of New Zealand who came to the United States about twenty-five years ago and has been a citizen for the past twenty years. He received his bachelors degree and masters degree in physics from the University of New Zealand. Thereafter, he completed the doctorate at the University of London where he studied under Professor W. V. Mayneord, a pioneer in radiation protection.

During the early years of his career, Dr. Sinclair showed an interest in the academic profession and has been closely associated with it ever since. From 1945-47, he served as Lecturer in Physics and Radiological Physics, University of New Zealand; from 1947-54, he was Assistant Physicist, Royal Cancer Hospital, and Lecturer, University of London; and from 1954-60, he was Head, Physics Department, M.D. Anderson Hospital and Professor of Physics, University of Texas. Subsequently, Dr. Sinclair became Professor of Biophysics at the University of California in Berkeley, and in 1970, he became Director, Division of Biological and Medical Research, Argonne National Laboratory. In 1974, he was appointed Associate Laboratory Director, a position which he continues to hold today. In addition to these many duties, Dr. Sinclair also serves as Professor of Radiation Biology, Department of Radiology, University of Chicago and as President of the Radiation Research Society. He is also a past president of the American Association of Physicists in Medicine and currently serves as a member of the International Commission on Radiation Units and Measurements, the International Commission on Radiological Protection, and the U. S. Scientific Delegation, UNSCEAR. He was elected to the Presidency of the NCRP in 1977. In short, he is a very busy man and it is a delight to have him with us today. Dr. Sinclair will speak to us on the subject of "Some Perspectives on Radiation Protection."

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SOME PERSPECTIVES ON RADIATION PROTECTION

Warren K. Sinclair, President
National Council on Radiation Protection and Measurements

I am pleased to have this opportunity to address you on the subject of protection from ionizing radiation and especially to describe to you some of the work of the National Council on Radiation Protection and Measurements (NCRP) of which I am the President, and of the International Commission on Radiological Protection (ICRP) of which I am a recently elected member.

Both of these bodies have almost exactly half a century of history - and indeed the ICRP celebrated its 50th anniversary in Stockholm recently. It was in Stockholm in 1928 that the Second International Congress of Radiology established the International Commission on Radiological Protection, and Lauriston Taylor, of the USA, was one of its founding members. He also organized, that same year, the Advisory Committee on X-ray and Radium Protection which became the forerunner of first, the National Committee on Radiation Protection, and in 1964, the National Council on Radiation Protection and Measurements chartered by the Congress of the United States as a non-profit corporation "...to collect, analyze, develop, and disseminate in the public interest, information and recommendations about (a) protection against radiation and (b) radiation measurements, quantities, and units, particularly those concerned with radiation protection."

The Council has a membership of some 75 scientists, physicians and engineers, elected for six-year terms (from the nominations of its members and its more than 30 collaborating organizations) drawn from the fields of radiation physics, biology, nuclear medicine, radiology, public health, nuclear engineering, environmental science, biophysics, genetics, microwave and ultrasound studies, dentistry, veterinary medicine, epidemiology and many other branches of science. In addition, its scientific committees and study groups include more than 300 additional scientists selected for their expert knowledge in the given subject area of the committees' work.

NCRP reports are produced by establishing, in a specific subject area, a scientific committee or study group of six to ten persons who draft a report which is then submitted to the entire Council for review. All substantive comments from the Council, and sometimes from other additional reviewers, are accounted for before the report is published as a report of the NCRP. The preparation and reviewing process takes time and NCRP reports reach the press often, regrettably, only slowly; but the process is one of careful deliberation and the detailed review process has certainly been responsible for the maintenance of high quality in the technical accuracy of the reports and in the quality of their guidance and recommendations. All work by Council and committee members is voluntary, which also contributes to the length of the process but ensures an independence of viewpoint and judgment generally not possible by other means. The true role of the NCRP is to provide this deliberate, expert opinion as free as possible of special interests or groups, and keeping always the public interest and the

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national good, as perceived by this expert body, in mind. The scope of the NCRP program is as broad as the impact of radiation on our lives and environment. The NCRP therefore seeks to render useful information and advice about all matters pertaining to radiations and protection against them.

The NCRP has a total of about 60 scientific committees and study groups of which more than 35 are currently active. The NCRP presently has two important committees dealing with non-ionizing radiation, but for purposes of discussion here, we shall limit our attention to committees concerned with ionizing radiation only.

For purposes of explanation and clarification, the work of these committees and groups can be divided into three principal categories:

1. Scientific Committees and Study Groups concerned with basic radiation protection criteria:
 - SC-1 Basic Radiation Protection Criteria
 - SC-40 Biological Aspects of Radiation Protection Criteria
 - SC-57 Internal Emitter Standards
 - Study Group on Human Exposure Experience
 - Study Group on Comparative Risk
2. Scientific Committees concerned with the assessment of dose to members of the general public:
 - SC-28 Radiation Exposure from Consumer Products
 - SC-41 Radiation Resulting from Nuclear Power Generation
 - SC-43 Natural Background Radiation
 - SC-44 Radiation Associated with Medical Examinations
 - SC-45 Radiation Received by Radiation Employees
 - SC-48 Apportionment of Radiation Exposure
3. Scientific Committees concerned with matters of guidance. For example:
 - SC-46 Operational Radiation Safety
 - SC-51 Radiation Protection in Pediatric Radiology
 - SC-15 Radiation Protection in Teaching Institutions
 - SC-16 Dental X-ray Protection
 - SC-17 Radiation Protection in Veterinary MedicineOr Scientific Committees concerned with information on selected topics of importance. For example:
 - SC-38 Waste Disposal-Krypton-85 in the Atmosphere
 - SC-36 Tritium Measurement Techniques
 - SC-35 Environmental Radiation Measurements
 - SC-24 Radionuclide and Labeled Organic Compounds Incorporated in Genetic Material

By way of explanation of the NCRP program, I will make just a few comments on each of these three, starting with the third category and working back.

A task group of SC-38 on Waste Disposal, in 1975, produced NCRP Report No. 44, Krypton-85 in the Atmosphere - Accumulation, Biological Significance and Control Technology¹, which may be of some particular interest to air quality engineering experts.

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The report describes the properties of ^{85}Kr , and makes estimates of future power requirements and ^{85}Kr releases to the year 2000; it describes the fate of ^{85}Kr , the dosimetry, the projected exposure of the population from ^{85}Kr , the biological significance of the absorbed dose due to ^{85}Kr , and the status of control technology procedures. The final discussion and summary indicates that, depending on nuclear power assumptions in the year 2000, which may now be modified due to changes in policy on reprocessing, the atmospheric inventory of ^{85}Kr and the doses to skin from diffused ^{85}Kr ($T_{1/2}$ 10.7 yr., β , 0.51 MeV), and the whole body dose will rise from 1970 to the year 2000, as shown in the table:

| | 1970 | 2000 |
|------------------------------------|-----------------------------|-------------------------|
| Inventory of $^{85}\text{Krypton}$ | 55 megacuries | 3600-6200
megacuries |
| Dose to Skin | 0.02 mrem/yr. | 2-3 mrem/yr. |
| Dose to Whole Body | 2×10^{-4} mrem/yr. | 0.02 mrem/yr. |

While the dose to whole body is negligible, the dose to the skin may become a factor to be considered. The report indicates that various control technologies are available which could remove ^{85}Kr from waste gases, including fluorocarbon extraction, precipitation as a clathrate, selective membranes or cryogenic distillation or absorption.

Another report of potential interest to this audience is NCRP Report No. 50, Environmental Radiation Measurements². I would like to draw your attention to the major headings in the Table of Contents as follows:

- Introduction
- Natural and Manmade Environmental Radioactivity and Radiation Fields
- Requirements for Measurement and Surveillance Programs
- In Situ Radiation Measurements
- Collection and Preparation of Samples for Laboratory Analysis
- Laboratory Measurements
- Conclusions: Trends and Problem Areas.

Among the second category of committees, i.e. those addressing population exposure, two have already produced reports; one, Report No. 45, Natural Background in the United States³, is a definitive report on this subject for the USA.

This report evaluates the contributions to human exposure of external cosmic radiation and cosmic ray-induced radioactivity, external terrestrial radiation, inhaled radioactivity, and internally deposited radionuclides. It concludes that the average exposure to an individual in the USA is as shown below:

| | |
|--------------------------|----------------|
| Average for the U.S. | ~ 80 mrem/year |
| Maximum (Denver) | ~125 mrem/year |
| Minimum (Atlantic Coast) | ~ 65 mrem/year |

These numbers are often rounded off to $\sim 100 \pm 25$ mrem/year.

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The report also deals with contributions to the dose from fallout and from air travel. Some of you might be interested in the latter because many of you probably travel quite a lot. I have added a few small calculations to the material in Report No. 45 in the slide that follows:

Dose from Air Travel (NCRP Report No. 45) *
Mid-latitude flights ~ 5 hours = 2.5 mrem
High latitude (polar) flights ~ 10 hours = 10 mrem.

Frequent Traveler at Mid-Latitudes
100,000 miles per year at 500 mph = 200 hours
(or about 2% of total available time!)
Dose = $\frac{200}{5} \times 2.5$ mrem = 100 mrem (or a doubling of natural background.)

Pilot and Crew
100 hours per month maximum - (Actual 60-65 hours)
 $\frac{100}{5} \times 2.5 \times 12 = 600$ mrem/year
(Note: This is well below occupational levels, 5000 mrem/year.)

Contribution of Air Travel to Population Dose
(NCRP Report No. 45)
 ~ 1 mrem/year

This shows that a very frequent traveler may double his exposure to natural background and that pilots and crew may get substantial exposure if they fly up to the maximum amount. These exposures are, however, considerably below those currently allowed for occupational workers.

Another very recent report in this category is NCRP Report No. 56, Radiation Exposure from Consumer Products and Miscellaneous Sources (1977)⁴. This report considers thirty or more sources of exposure to segments of the public from consumer products. These include electronic products such as television receivers, x-ray devices such as airport inspection systems, sources such as incorporated radionuclides in building materials, or (more bizarre) uranium in dental porcelain, or sources such as luminous materials in watches, clocks or instrument dials. A few of the estimated average exposures to the population from representative sources are as follows:

| Source | Tissue | No. of People Exposed | Average Annual Population Dose |
|----------------------------|------------|-----------------------|--------------------------------|
| Television Receivers | Gonads | 100,000,000 | 0.5 mrem |
| Airport Inspection Systems | Gonads | 10,000,000 | 1 μ rem |
| Luminous Wristwatches | | | |
| Radium | Gonads | 10,000,000 | 150 μ rem |
| Tritium | Whole body | 16,000,000 | 50 μ rem |
| Combustible Fuels (Coal) | Lungs | 50,000,000 | 0.5 - 1 mrem |
| Building Materials | Whole body | 100,000,000 | 3.5 mrem |

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The overall exposure to the public from all such sources is less than 5 mrem/year, and most of this (70%) comes from building materials, primarily U, Th and ⁴⁰K in masonry (as opposed to frame construction). Note, this includes office exposure for many people as well as private housing (100,000,000 people receiving 7 mrem/yr is equivalent to a dose of 3.5 mrem/yr to the entire US population of 200,000,000).

At this point I thought it might also be of interest to you to know about some important sources of exposure and the average doses to which the average member of the public is estimated to be exposed.

Estimated "Average" Annual Exposure to Members of U.S. Population

| | | |
|----|--------------------------|--------------|
| A. | Natural Background | 100 mrem |
| B. | Air Travel | 1 mrem |
| C. | Radioactive Fallout | |
| | Whole Body | ~0.5 mrem |
| | Bone | 2 mrem |
| | (Alaskan Eskimos (max.)) | 50 mrem) |
| D. | Occupational | ~0.8 mrem |
| E. | Consumer Products | < 5 mrem |
| F. | Nuclear Power | < 1 mrem |
| | Waste Management | < 0.001 mrem |
| G. | Medical | 55-75 mrem |

The two kinds of committees I have discussed so far, namely those concerned with guidance or information, and doses to the public, are of interest and considerable value. However, the most important function of the NCRP (and the ICRP) is contained in my first grouping; namely the work of committees concerned with Basic Radiation Protection Criteria. In the NCRP, in the past, a single committee established the basic radiation protection criteria for external exposure, and the basic report here is that known as NBS Handbook 59 (NCRP Report No. 17) published in 1954⁵, after an extended period of examination and close cooperation with the ICRP. It set levels of permissible occupational exposure (weekly 0.3 rem= 15 rem/yr) which, in 1957, were effectively lowered when age pro-rata was introduced. The new basic occupational level then became 5(N-18) rem/yr, where N is the age in years. These occupational levels have been widely adopted by government agencies in rule-making, and by industry and many others for guidance.

These levels were essentially repeated in the NCRP Report No.39 Basic Radiation Protection Criteria, issued in 1971⁶. At that time, the NCRP, following the philosophy of absence of harm, saw no reason to alter the levels. These are the levels the NCRP recommends at the present time and are summarized as follows:

| | | |
|--------------|--|--------------------|
| Emergency | Life Saving (>age 45) | ‡ 100 rems |
| | Non-life Saving | ‡ 25 rems |
| Occupational | Whole Body (N=age in years) | ‡ 5 (N-18) rems |
| | Extremities higher | |
| Public | Individual | ‡ 0.5 rem in 1 yr. |
| | Average | ‡ 0.17 rem av./yr. |
| | (i.e. 170 mrem or ~ 2 x natural background). | |

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Nevertheless, the process of examining levels and their suitability both for external radiation and for internal emitters released in the air and water is a continuing one. The primary responsibility for this rests with NCRP's SC-1, but in 1971 a new committee was set up, SC-40, on Biological Aspects of Radiation Protection Criteria, to aid Committee 1 in the evaluation and analysis of important radiobiological information which would enable better extrapolation of data obtained at high doses to the low doses of protection level interest. This Committee has been examining the important matter of dose-rate effects and their influence on the dose response curve at low doses. It will also examine the important matters of the relative effectiveness of high-LET radiation at low levels. These examinations will result in estimates of risk being supplied to Committee 1 for radiation exposure circumstances relevant to radiation protection.

At the same time a new Study Group on Human Exposure Experience will be examining all sources of human exposure taking into account not only the excellent surveys of Japanese data from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)⁷, but considering, also, the validity of recent studies such as those reported by Mancuso et al.⁸, exposures at the Portsmouth naval shipyard, military exposures during bomb tests, and other sources of potential value in establishing levels at which harm exists. Many other sources in the medical field such as the induction of thyroid tumors in persons given x-ray therapy to the head and neck region may also yield important information. Medical studies tend to receive less publicity than studies on workers, but they may yield more information ultimately.

The NCRP also has a Study Group on Comparative Risk which seeks to examine the hazards from other agents to which the public and occupational workers may be exposed, and to place radiation hazards in context with them. In all of these cases, while the principle of "as low as practicable (ALAP)," or "as low as reasonably achievable (ALARA)" is the basic operating tenet, when limits above which exposure is unacceptable must be defined, they should be based on similar considerations no matter what the exact source of the hazard may be, if the effects on the individual and/or the population are comparable. The basis should, perhaps, be acceptable risk, and I might point out that the NCRP has another group examining this question specifically in relation to waste disposal procedures.

SC-57 on Internal Emitter Standards is examining the maximum permissible levels of radionuclides in air and water, and is studying the metabolism of nuclides introduced to the body by these routes and, therefore, causing exposures in workers and others at levels controlled by the recommendations of SC-1. SC-1 and SC-57 work closely on these matters and are heavily engaged on their programs at the present time and I expect both committees to come out with highly significant reports within the next two years.

Now let me turn briefly to the ICRP. The ICRP comprises a Chairman and 12 members of the main commission elected as individuals for four-year terms. Currently these members are drawn from nine countries, and many other countries are represented among the

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individuals serving on the four main committees of the ICRP and on their task groups. The ICRP program also includes the study of the fundamental principles upon which radiation protection measures are based as well as general guidance on matters of radiation protection. More specific guidance tends to be left appropriately to the national groups of each country. Also, the United National Scientific Committee on the Effects of Atomic Radiation⁷ provides much of the radiobiological assessment, population exposure information, and worldwide impact of sources that ICRP can use, and therefore does not need to generate independently. In recent years, ICRP has been very active in developing the basic principles of radiation protection and in 1977 produced ICRP Publication 26⁹. In this connection, it is interesting to note that the ICRP initiated publication of its Annals in 1977, and the reports of the ICRP are now readily available by subscription to these Annals.

The principal features of ICRP Publication 26 are as follows:

The report

- o discusses risk in detail
 - total stochastic risk, 10^{-2} Sv^{-1} *
 - total genetic risk, $4 \times 10^{-3} \text{ Sv}^{-1}$
- o limits occupational exposures for adults irradiated whole body to 50 mSv/year (5 rems/year), i.e. somatic risk has a maximum value of 5×10^{-4} /year
- o does not allow for age proration, i.e. the occupational limit is 5 rems/year even if exposure has been much lower in previous years
- o permits higher exposure of individual organs when body is irradiated non-uniformly (see below)
- o suggests that exposure of individual members of the public be limited to 5 mSv, in which case the average exposure should not exceed 0.5 mSv, corresponding to a risk of 10^{-6} /year, which is deemed to be acceptable when compared with other risks
- o promotes principles of optimization of protection procedures to keep exposures well below these limits (i.e. ALARA).
- o emphasizes that the upper limit (5 rem/year) is rarely received by workers, that 10-15% of this level is an average, and this average exposure of radiation workers is comparable with the hazards of other safe industries.

*1 Sievert (Sv) = 100 rems.

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The weighting factors for non-uniform radiation are as explained in the following two paragraphs quoted from ICRP Publication 26:

- (104) For stochastic effects the Commission's recommended dose limitation is based on the principle that the risk should be equal whether the whole body is irradiated uniformly or whether there is non-uniform irradiation. This condition will be met if

$$\sum_T w_T H_T \leq H_{wb,L}$$

where, w_T is a weighting factor representing the proportion of the stochastic risk resulting from tissue (T) to the total risk, when the whole body is irradiated uniformly, H_T is the annual dose equivalent in tissue (T), $H_{wb,L}$ is the recommended annual dose-equivalent limit for uniform irradiation of the whole body, namely 50 mSv (5 rem).

- (105) The values of w_T recommended by the Commission are shown below:

| Tissue | w_T |
|-----------------|-------|
| Gonads | 0.25 |
| Breast | 0.15 |
| Red bone marrow | 0.12 |
| Lung | 0.12 |
| Thyroid | 0.03 |
| Bone surfaces | 0.03 |
| Remainder | 0.30 |

The value of w_T for the remaining tissues requires further clarification. For the reasons stated in paragraphs 58 and 59 the Commission recommends that a value of $w_T = 0.06$ is applicable to each of the five organs or tissues of the remainder receiving the highest dose equivalents, and that the exposure of all other remaining tissues can be neglected. (When the gastro-intestinal tract is irradiated, the stomach, small intestine, upper large intestine and lower large intestine are treated as four separate organs.)

The ICRP has also been revising the permissible levels for internal emitters and a report of its Committee 2 on the first 22 elements, some 200 radionuclides considered to be of primary importance, will go to press this fall. It is not yet available for general distribution, but its approach is to develop dose equivalent limits for internal exposure which are equated with those of ICRP 26 for external exposure. These will define a Committed Dose Equivalent, H_{50} , the dose caused by the intake of a nuclide over 50 years, which must not deliver an annual dose greater than 5 rem/year (weighted for organs if necessary). Secondary levels which will conform to these dose limits are expressed as Annual Limits of Intake (ALI's) either by ingestion

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or inhalation. In the case of inhalation, a Derived Air Concentration is defined such that the ALI is divided by the volume of air inhaled by Reference Man in a working year. Total ingestion includes liquids and solids and is controlled by the ALI for ingestion. No separate value for water will be given. This new approach by ICRP in the internal emitter area is yet to be tested in practical application and working experience on the part of health physicists is needed.

Another area in which the ICRP has made a significant contribution, recently, is in the issuance of its Publication 27, Problems Involved in Developing an Index of Harm¹⁰. This report is not intended to be a definitive report but rather tries to develop a method for equating risks from very different sources. For example, the industrial accidents or injuries. This effort is well worth further study and refinement. One general conclusion to be derived from this report is that radiation exposure at the average level of occupational workers (10-15% of the upper limits themselves) is comparable with the hazards of "safe" industries. If exposures are actually at the upper limit however, the risks, estimated on the basis of the linear hypothesis, are more comparable with the relatively dangerous industries like construction and mining.

In view of the trend toward greater and greater safety in all fields leading presumably to further reductions in accident rates, and as the approaches to comparative risks from different sources of hazard become better appreciated, it will be increasingly necessary to re-examine the philosophy of radiation protection. Hopefully, in time, more definitive quantitative information on the risks at low levels may become available, possibly demonstrating the extent to which the assumed linear hypothesis over-estimates the hazard. An important aspect of this is that we must keep analyzing actual data from all sources of radiation exposure of human beings as it accumulates. Positive information will enable future estimates of risk to be more accurate than they are now and thus recommendations for limits for occupational workers and for the public can be put on a steadily sounder foundation.

* More recent estimates involving a more extensive direct survey (R.W. Wallace and G.A. Sondhaus, *Aviat. Space and Environ. Med.* 49 (4): 610-613, 1978) indicate appreciably lower values for the average exposures due to air travel.

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SESSION XII

NEW AIR CLEANING TECHNOLOGY FROM EUROPE

Wednesday, August 9, 1978

CHAIRMAN: D. W. Moeller

NEW AIR CLEANING TECHNOLOGY IN THE UNITED KINGDOM

R. G. Dorman

SOME DEVELOPMENTS IN NUCLEAR AIR CLEANING IN FRANCE

P. Sigli

GAS CLEANING RESEARCH AND ENGINEERING IN BELGIUM

W. R. A. Goossens, L. H. Baetslé,
A. Bruggeman, G. Collard

DEVELOPMENTS IN NUCLEAR AIR CLEANING IN GERMANY

J. G. Wilhelm

IAEA/NEA ACTIVITIES IN THE FIELD OF GASEOUS WASTES

J. Dubé, Y. Zabaluev

OPENING REMARKS OF SESSION CHAIRMAN:

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

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

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

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NEW AIR CLEANING TECHNOLOGY IN THE UNITED KINGDOM

R. G. Dorman
Chemical Defense Establishment
United Kingdom

PORTON GAB
LONDON

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

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

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

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

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

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

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

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

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

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

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

DISCUSSION

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

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

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

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

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

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

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SOME DEVELOPMENTS IN NUCLEAR AIR CLEANING IN FRANCE

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YONENAS - BOY - 1975, 1976, 1977

I. IODINE TESTING

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

I.1. Modular unit testing

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

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

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

In order to obtain these performances this installation comprises:

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

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

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

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

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

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

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

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

I.2. Iodine in situ testing

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

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

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

II. IODINE TRAPPING IN A REPROCESSING PLANT

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

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

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

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

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

III. KRYPTON RECOVERY (3)

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

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

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

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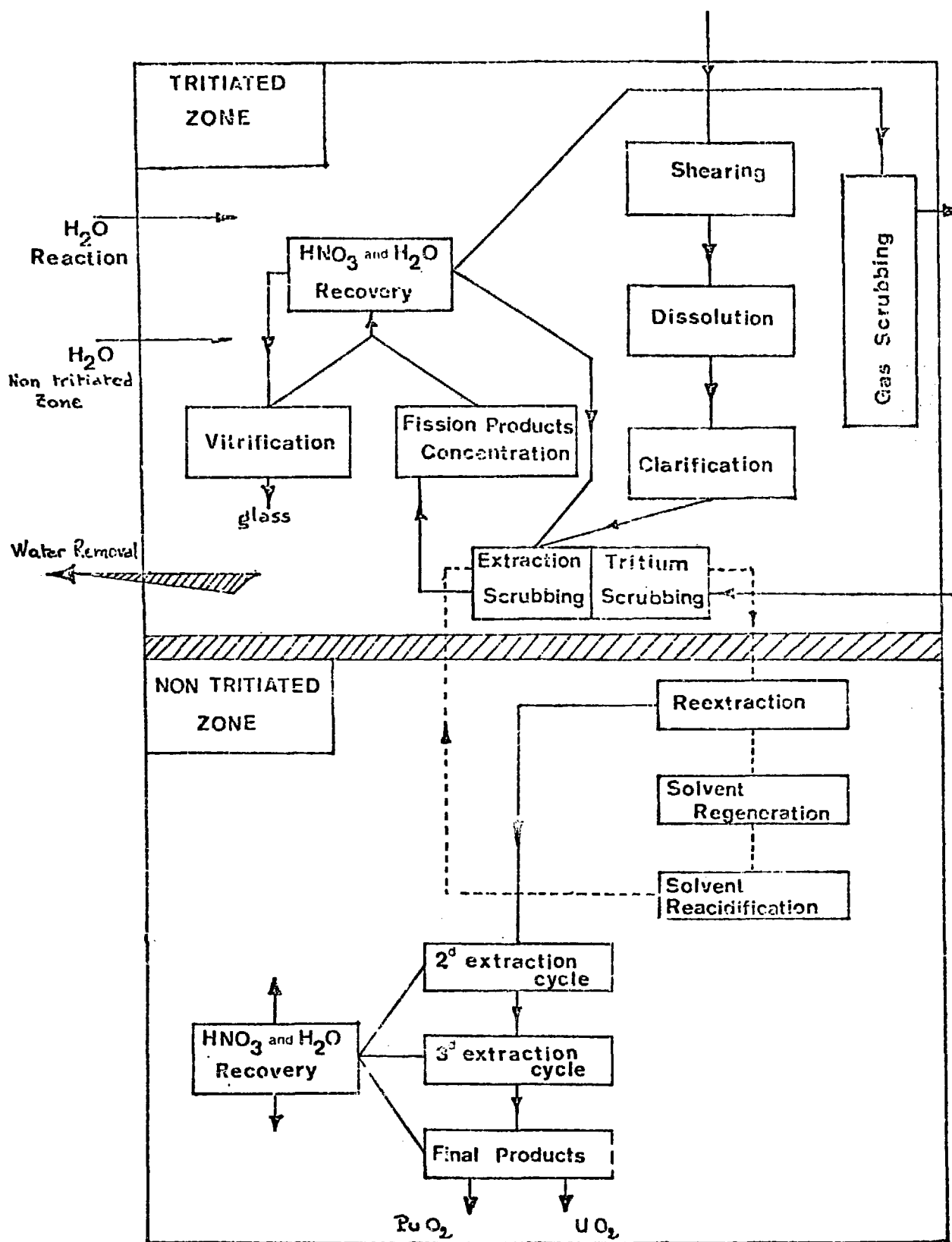


Fig. 1 - TRILEX process

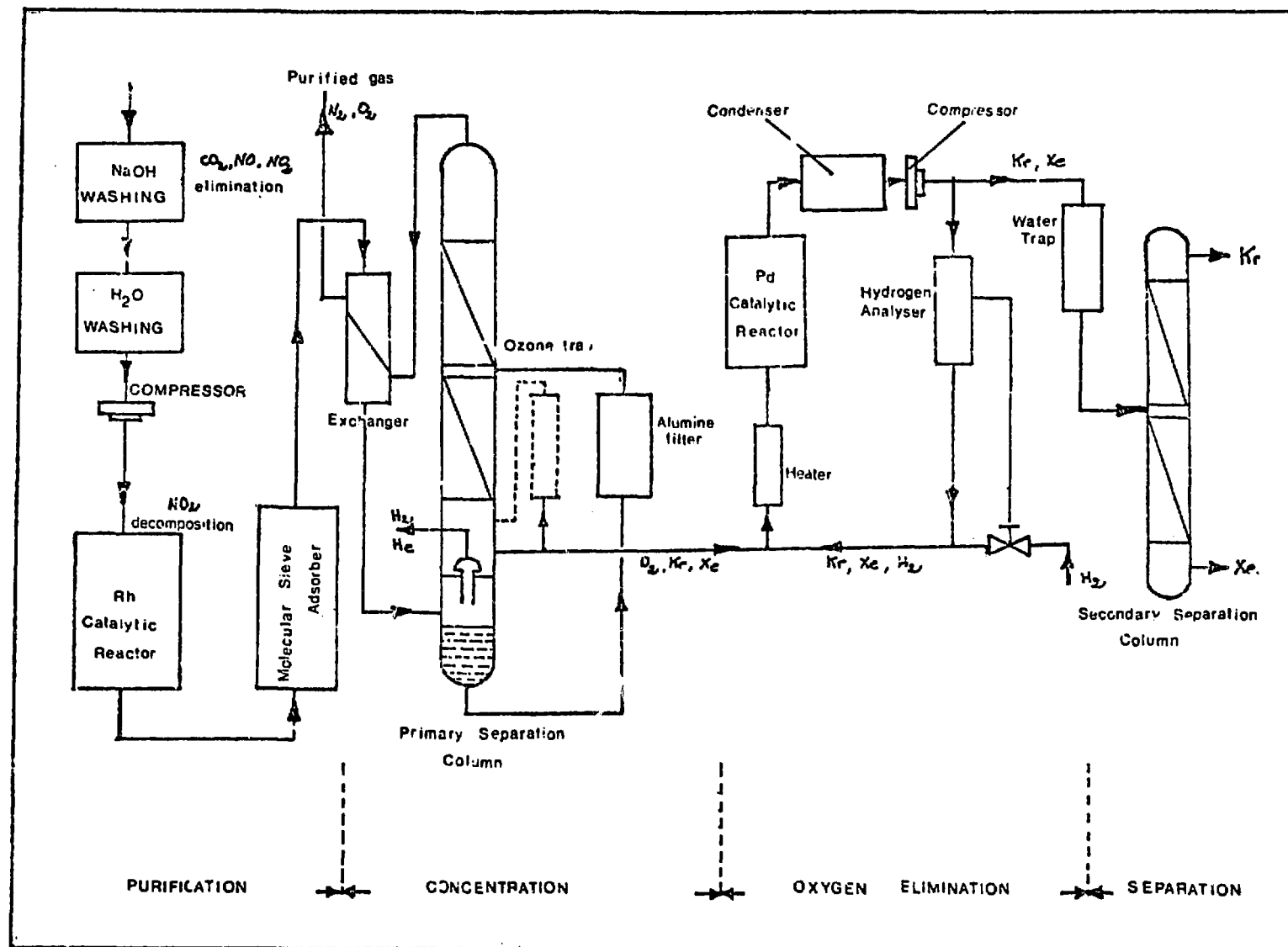


Fig. 2 - Krypton Recovery flow-sheet

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IV. TRILEX PROCESS (4)

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

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

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

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

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

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

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

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DISCUSSION

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

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

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

SIGLI: Principally isotopic exchange (water-tritiated water) or electrolysis followed by H₂-HT separation.

BROWN: You mean electrolysis and then a catalytic exchange?

SIGLI: Yes.

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

SIGLI: Yes, it is.

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

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

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

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

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

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

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GAS CLEANING RESEARCH AND ENGINEERING IN BELGIUM

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I. Introduction

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

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

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

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

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

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

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

III. The Pilot Delay-Bed

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

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IV. The Laboratory Set-Up for Iodine Sorbents

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

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

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

V. The Integrated Gas Purification Test Loop

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

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

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

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

VI. Test Rigs for the Removal of Particulates

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

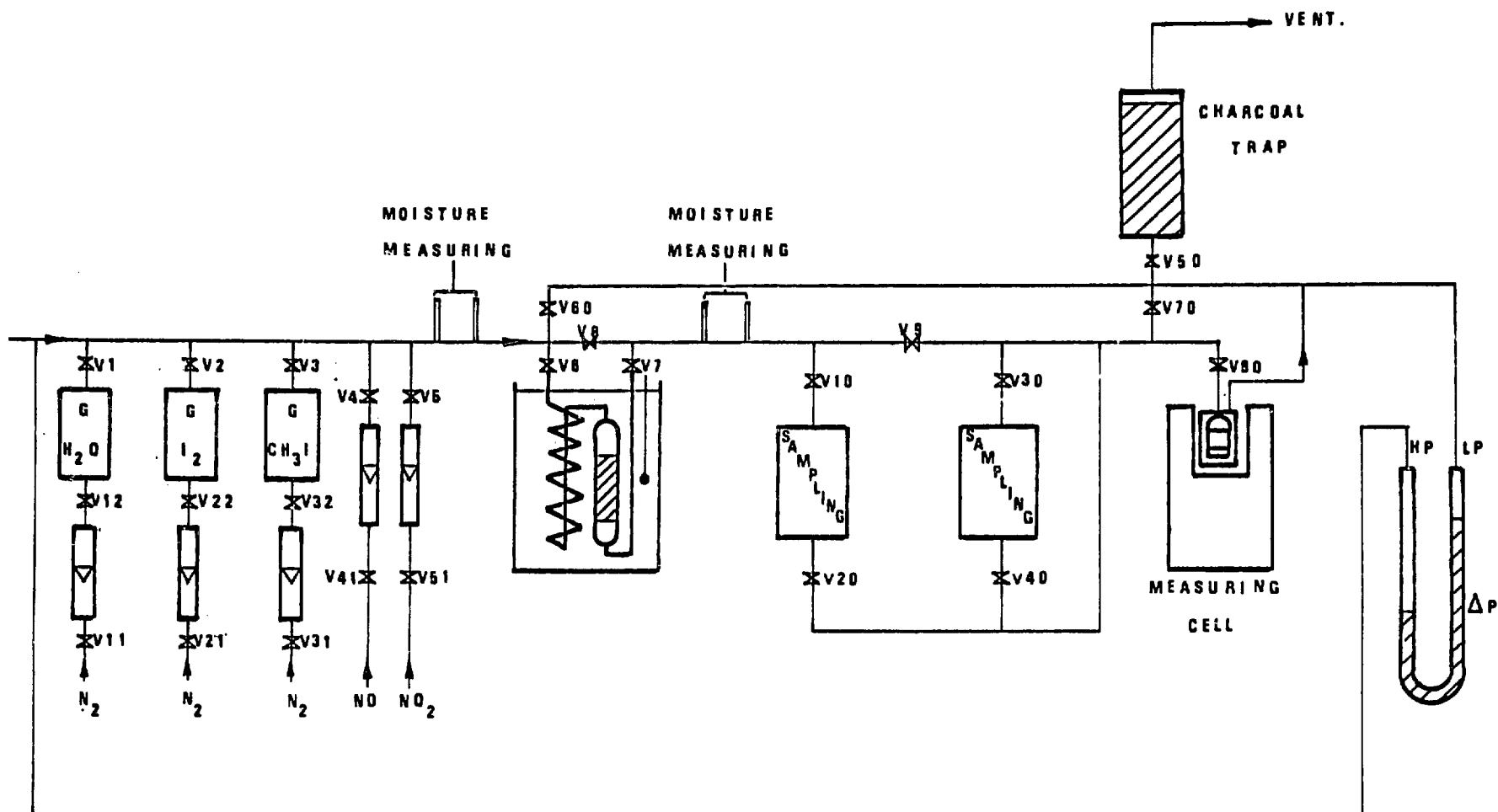


Fig. 1 : SCHEME OF $1 \text{ m}^3 \text{ h}^{-1}$ LOOP

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

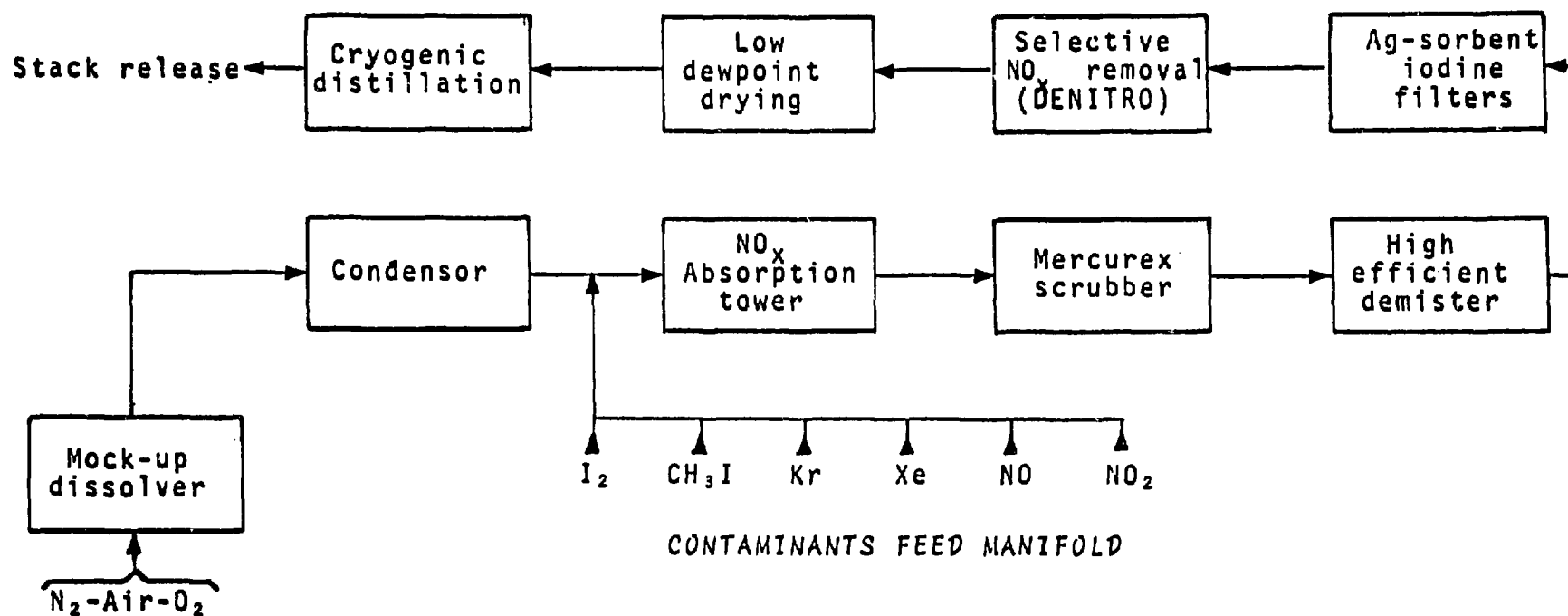
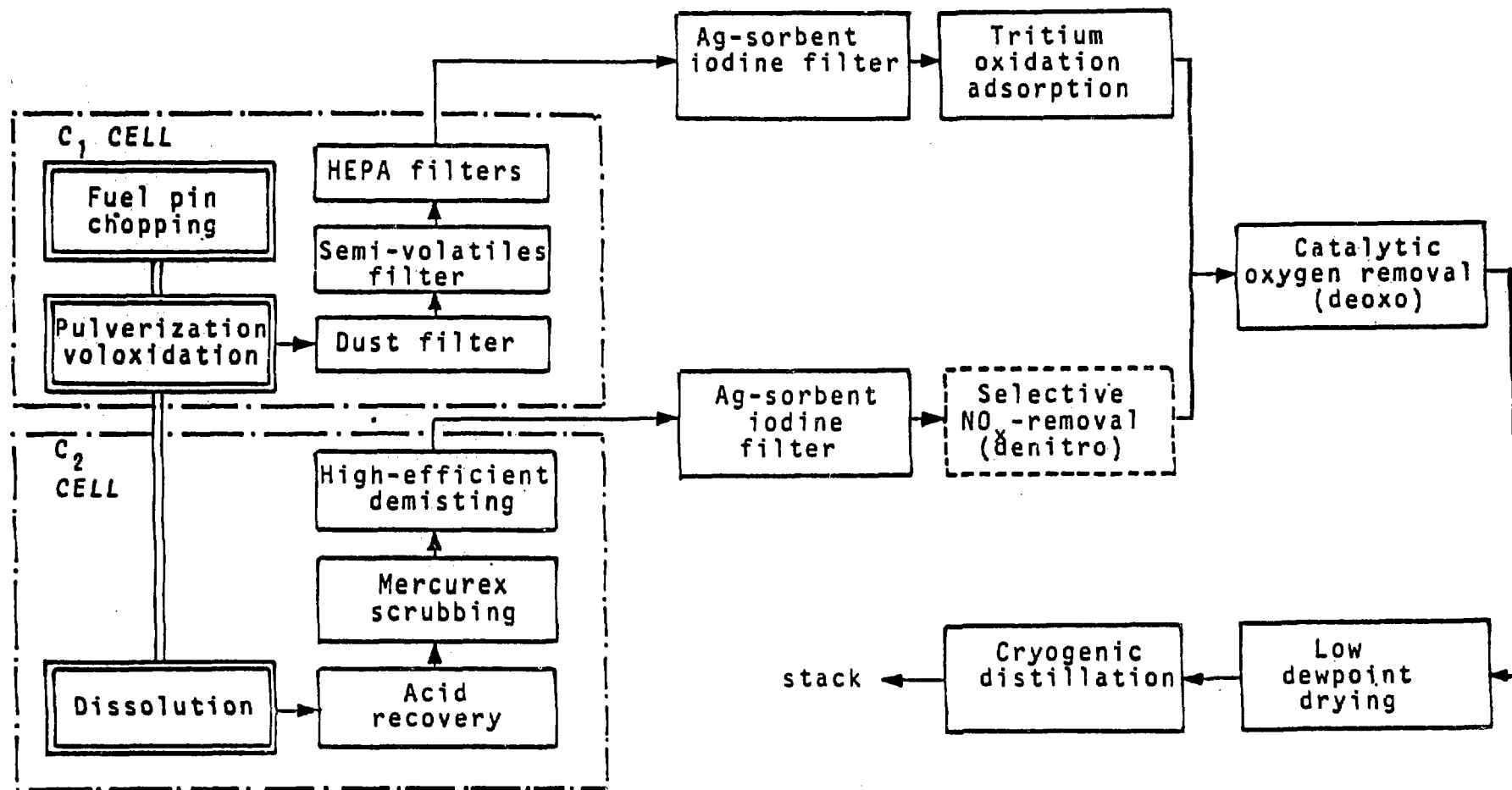


Fig. 3 : BASIC OFF-GAS PURIFICATION SCHEME (HERMES)



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

VII. The Experimental Program on Tritium

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

VIII. Conclusion

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

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DISCUSSION

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

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

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

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

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Developments in Nuclear Air Cleaning in Germany

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I. New Filter Designs for Nuclear Power Plants

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

I.1 HEPA-Filters

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

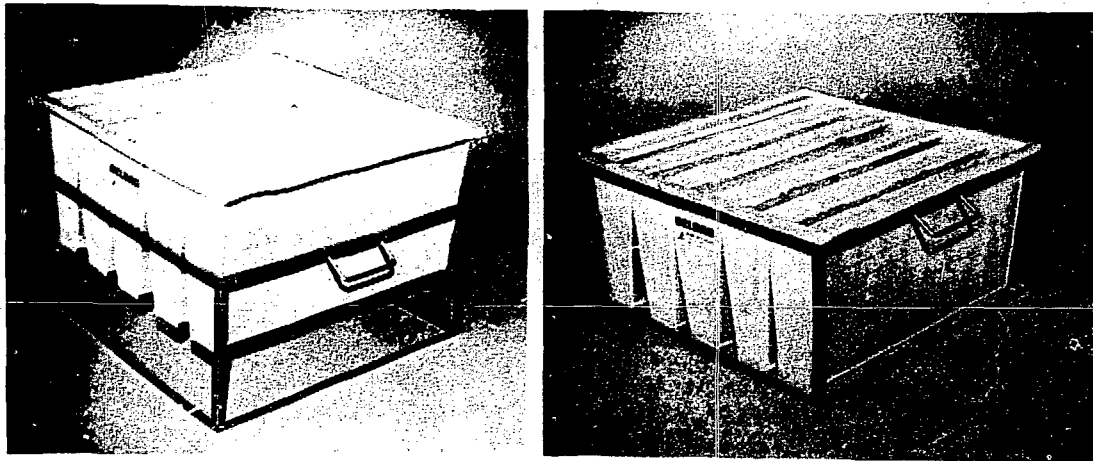


Fig. 1

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

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

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

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

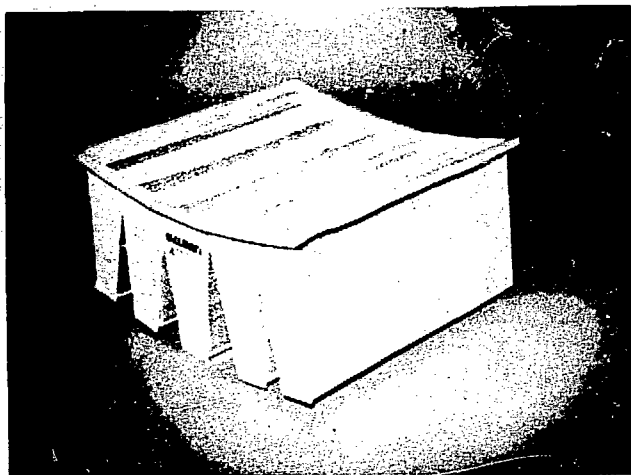


Fig. 2

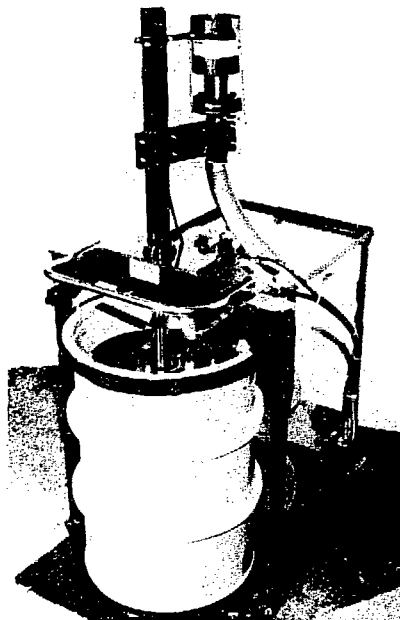


Fig. 3

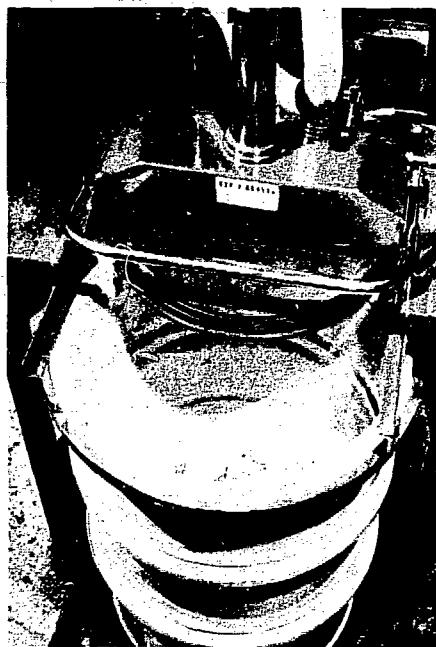


Fig. 4



Fig. 5

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Another recent development is a filter element which consists of a standard 200 l waste drum with integrated HEPA filters. The drum at the same time acts as a filter housing. The special design of the filter elements and the sealing relative to the wall of the drum at the diagonally opposed edges splits the gas stream into unfiltered and filtered gas streams within the drum. The maximum volume flow through such a HEPA filter may be up to 5000 m³/h. The filter is connected directly to the ventilation ducts. When replaced, the drum with the integrated filters is disengaged and sealed. This work can easily be performed by remote handling equipment.

I.2 Iodine Sorption Filters

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

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

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

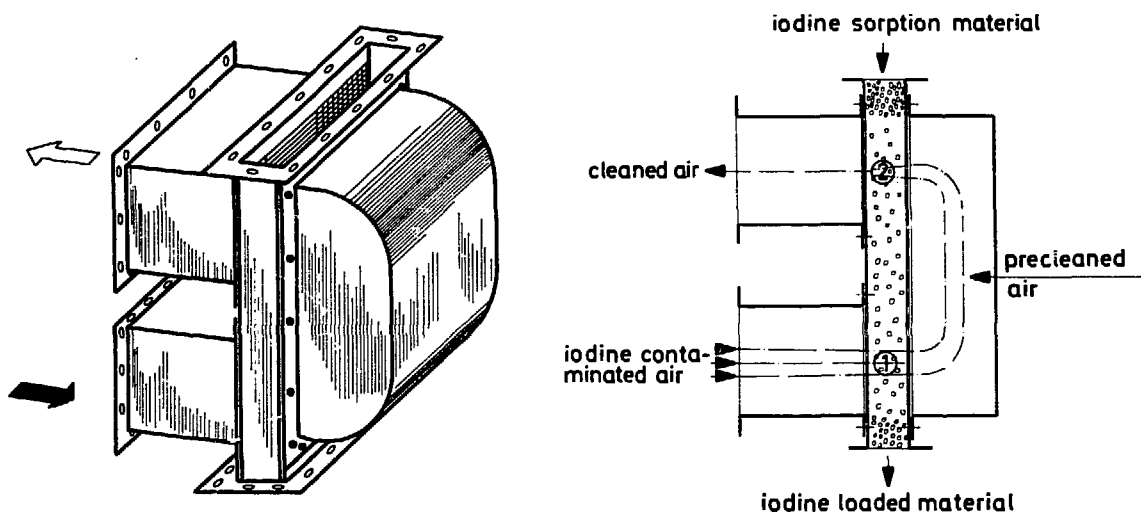


Fig. 4 Multiway iodine sorption filter

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

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

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

II. Testing Iodine Sorption Filters

In the Federal Republic of Germany, iodine sorption filters installed to control the consequences of accidents must be examined for their removal efficiencies prior to startup of the respective nuclear power plant and at regular intervals. These checkups are increasingly becoming the rule also for iodine sorption filters used for exhaust air cleanup in normal operation of a nuclear power plant. As a rule, the examination includes an in-place test and testing of activated carbon samples in the laboratory at specific time intervals. Both the in-place test and the laboratory examination have so far been carried out with radioactively labeled methyl iodine as the test medium. Now that iodine species samplers (2) have been developed which allow a clear distinction to be made between elemental iodine and organically bound iodine (methyl iodide, as a rule) it is possible, provided that there is a sufficient concentration of ^{131}I in the filter intake air, to measure iodine sorption filters used in normal operation of a nuclear power station without additional feeding of a radioactive test agent directly by way of their removal behavior relative to these iodine components. For an activity concentration of 10^{-12} Ci/m³ in the filter intake air a decontamination factor of 1000 can be proved by means of a solid state detector for sample counting. The measured results obtained are always much more precise than measurements of activated carbon samples in the laboratory, because the true loading condition of the activated carbon with pollutants during the measuring period remains unchanged; there is no need for conditioning of the activated carbon in respect of the uptake of water vapor prior to determining the removal efficiency.

III. Off gas Treatment in Reprocessing Plants

III.1 Reprocessing Plants for LWR Fuel

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

- Iodine vaporisation from the feed solution and from the recovered acid (HET test rig, flow 50 m³/h at STP, cold and tracer trial operations).

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- Separation of large and fine droplets, removal of the bulk quantity of iodine accumulated in the solid AC 6120 iodine sorption material and removal of particulates in HEPA filters (PASSAT test rig (3) with 1 : 1 components, flow 250 m³/h at STP, cold and tracer trial operation, tests of remote handling systems).
- Removal of krypton (4) including the separation of O₂ and residual NO_x by catalytic reduction with H₂, adsorptive retention of H₂O, CO₂, NH₃ etc., and cryogenic distillation in two columns, first the N₂-Kr-Xe three-component mixture, then the Kr-Xe two-component system (cold pilot plants for gas flows of 50 m³/h at STP called ADAMO and KRETA).

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

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

III.2 Reprocessing of HTR Fuel

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

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

one cyclone for particles with diameters $\geq 5 \mu\text{m}$.

one rotational flow centrifugal separator for particles $\geq 1.8 \mu\text{m}$.

Improvements are to achieve removal down to particle diameters of approx. 0.6 μm .

- the removal of aerosols, I and Kr from the burner off gas (AKUT II test facility with a flow of 10 m³/h at STP). Aerosols are removed in an electrostatic precipitator with downstream HEPA filters; tritium, which is present as tritiated water, is adsorbed onto molecular sieves.

CO is transformed into CO₂ by oxidation with oxygen on a palladium catalyst.

The off gas consisting of CO₂, 0.25 % O₂ and the fission noble gases, is compressed and liquefied. Kr is separated from the bulk CO₂ by distillation. The distillation column is designed for a decontamination factor of 1000 and a product of 0.1 % Kr in CO₂.

- removal of the Xe and Kr noble gases from the dissolver off gas. Two cryogenic separation units have been developed:

- (1) the KRYOSEP I facility (flow 5 m³/h at STP) equipped with demister, HEPA filters, iodine filters, O₂ and NO_x removal by catalytic reaction with H₂, and removal of the water by means of molecular sieves. In a subsequent cryogenic separation unit Xe is frozen out and separated from residual Kr by distillation. The liquefied N₂-Ar-Kr mixture is distilled and the Kr-rich fraction is removed and filled in pressurized cylinders;
- (2) the KRYOSEP II facility (flow 1 m³/h at STP) for the removal of Kr and Xe from the off gas of a continuous dissolver with a closed loop helium purge gas cycle. In this facility Xe and Kr are frozen out separately after the same preliminary treatment as described under (1) above. Xe is cleaned of the residual Kr in a batch distillation column.

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References

- (1) J. Furrer, R. Kaempffer, J.G. Wilhelm, Ageing and poisoning of iodine filters in nuclear power plants, Kerntechnik Vol. 18, No. 7 (1976).
- (2) H. Deuber, J.G. Wilhelm, Determination of the Physico-Chemical ^{131}I Species in the Exhausts and Stack Effluent of PWR Power Plants, paper 6-7 of this conference.
- (3) J. Furrer, J.G. Wilhelm, K. Jannakos, Aerosol and Iodine Removal System for the Dissolver Off Gas in a Large Fuel Reprocessing Plant, paper 6-2 of this conference.
- (4) R.v. Ammon, H.G. Burkhardt, E. Hutter and G. Neffe, Development of a Cryogenic Krypton-Separation System for the Off Gas of Reprocessing Plants, paper 7-2 of this conference.
- (5) H. Barnert-Wiemer, M. Heidendael, E. Merz, H. Vygen and G. Schröder, paper 6-1 of this conference.

DISCUSSION

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

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

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

BURCHSTED: And the area of the medium?

HAYN: 8 sq. meters.

BURCHSTED: Is the medium fiberglass?

HAYN: Yes.

BURCHSTED: What is its thickness?

HAYN: The thickness is 0.015 in.

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

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

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

HAYN: At least 10 in. w.g.

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

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

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

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

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

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

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

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

BROWN: Do you have any plans for tritium recovery?

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

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

WILHELM: This is under investigation.

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

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

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

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

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

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

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

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IAEA/NEA ACTIVITIES IN THE FIELD OF GASEOUS WASTES

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Introduction

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

1. Objective

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

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

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

2. Scope

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

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

3. Requirements

System designs are determined by three factors:

- (a) Government regulations and guidelines;
- (b) Process control needs;
- (c) Effluent stream characteristics.

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

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

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

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

4. Description of Present Systems

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

- (a) Representative sampling rarely occurs due to non-homogeneous gas streams, non-isokinetic sampling, and line losses due to plate-out, adsorption, impaction, temperature changes, proper location of probes, etc.
- (b) Monitoring of low-energy beta emitters are difficult to detect when high energy radiation from other radionuclides is present.
- (c) Real-time monitoring is necessary in order to allow operating personnel to minimise abnormal releases of radioactive effluents. No facility surveyed has continuous I-129, H-3, Ru-106 or C-14 monitors. Continuous particulate monitors are generally gross monitors.
- (d) Alarm monitors are not able to detect an increase of "x" counts per minute immediately after installation of a new sample collector, but this same "x" counts per minute could trigger the alarm monitors just before removal by the collector.
- (e) The reliability of monitoring data is difficult to evaluate due to the numerous possible interferences in the complex gas streams. Calibration of monitoring equipment with pure radionuclides only adds to the difficulties as it does not take into account interferences from other components of the off-gas stream. Monitoring reliability under abnormal conditions is also not fully understood.
- (f) The reliability and sensitivity of Kr-85 monitors in some complex and corrosive environments is unknown.
- (g) The problem in particulate monitoring is essentially that of representative sampling. In addition, sampling filters have varying efficiencies, depending on particle size, gas stream conditions, flow rates, etc.

5. "State-of-the-Art"

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

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

- (a) The results of the enquiry are complete enough to permit an approximation of the "state-of-the-art" monitoring of off-gases for different radioisotopes as well as those isotopes that are currently measured at different types of facilities.
- (b) The different monitoring techniques cannot be compared without making specific value judgements on sensitivity, range or specificity of the method and these criteria may not be applicable to a specific type of installation.

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

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

- (a) Design of sampling systems should be made before construction of facilities.
- (b) Use of multiple isokinetic samples probes.
- (c) To avoid condensation problems, use of heated sample lines, collectors and adsorbers, and filters.

6. Advanced Monitoring Methods

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

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

7. Research and Development Recommendations

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

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

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

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TABLE I

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

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

TABLE II

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

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

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TABLE III

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

| Sampling | Measurement |
|--|---|
| 1. Flow-thru flask, chamber or vessel. | 1. Gamma spectrometry with sodium iodine or Ge-Li detectors with single and multi-channel analysers and rate meters.

2. Proportional counters and Geiger Muller tubes.

3. Plastic scintillator.

4. Ionization chamber. |

TABLE IV

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

| Sampling | Measurement |
|---|--|
| 1. Adsorbtion of iodine on silver zeolite, charcoal, silver impregnated material. | 1. Liquid scintillation counting.

2. Neutron activation analysis.

3. X-ray spectrometry. |

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TABLE V

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

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

TABLE VI

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

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

* for Sr-90

** for Ru-106

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TABLE VII

Advanced Monitoring Methods for Different Radioisotopes

| Isotope | Monitoring methods |
|-----------------------|---|
| Tritium | <ol style="list-style-type: none"> 1) Differential monitor 2) Condensation followed by liquid scintillation counting 3) Water or ethylene glycol bubbler and large surface area plastic scintillator (continuous) 4) Possible laser monitoring techniques |
| Carbon-14 | <ol style="list-style-type: none"> 1) Possible laser monitoring techniques 2) Adsorption in a liquid media followed by liquid scintillation counting |
| Krypton-85 | <ol style="list-style-type: none"> 1) Silicon surface barrier detector after removal of interfering radioisotopes 2) Laser monitoring possible if ultraviolet lasers become available |
| Iodine-129 | <ol style="list-style-type: none"> 1) Laser induced fluorescence 2) Intracavity absorption laser spectroscopy 3) Molecular beam quadrupole mass spectrometry |
| Iodine-131 | |
| Particulates | <ol style="list-style-type: none"> 1) Surface ionization mass spectroscopy 2) Annular impactor for alpha particulates 3) "AIRTRACE" laser techniques 4) Moving filter tape monitors for transuranic particulates |
| "Ramp alarm monitors" | |

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

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

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

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

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

8. Conclusions

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

- (a) Three types of monitor are actually needed - inventory monitors, alarm monitors, and process control monitors.
- (b) The particular nuclides that should be monitored, and the methods for monitoring them, should be mentioned in the regulations.
- (c) Specific removal devices should be continuously monitored at the clean-up device for process control and alarm purposes.
- (d) Inventory monitoring at present is most reliably accomplished with fixed collectors which are removed periodically and brought to a laboratory for analysis.

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CLOSING REMARKS OF SESSION CHAIRMAN:

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

In a general sense, the papers in this session addressed three areas of technological development. First was the matter of the increasing emphasis on air cleaning systems that can be operated for longer periods of time. In this regard, the use of prefilters to protect the primary air cleaning units ~~was~~ emphasized. Also stressed in these programs was the use of components that can be maintained with a minimum of occupational exposure and which can be disposed of more readily. Second was the effort to develop facilities for improved testing of air cleaning systems under a wider range of parameters (temperature, pressure, humidity). This includes the application of a range of test aerosols, and evaluations of removal efficiencies for specific radionuclides in specific chemical forms at concentrations normally anticipated in operating facilities. Third was the increasing attention being directed to the long range need for the removal and retention of selected radionuclides such as ^{14}C , ^{131}I , ^3H , and ^{85}Kr . This last effort is being addressed primarily to air cleaning problems associated with spent fuel chemical processing plants. This is an area in which scientists in the European countries are moving forward rapidly and I believe it is clear to all that they are ahead of similar research and technological developments within the U.S.

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

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SESSION XIII

OPEN-END

Wednesday, August 9, 1978

CHAIRMAN: M. W. First

USE OF INSTALLED TEST CANNISTERS FOR REPRESENTATIVE SAMPLING OF VENTILATION SYSTEMS
CHARCOAL ABSORBERS C. A. Moore

A REMOTE SAMPLING SYSTEM FOR HEPA (1500 CFM) IN-PLACE FILTERS WITH A FLUORIDE
(CHANNEL) SEAL SYSTEM L. G. Musen

RADIOACTIVE WASTE MANAGEMENT: THE NEED FOR MULTIPLE BARRIERS
J. Sokol, M. Cooper

CSL ENGINEERING DESIGNER HANDBOOK A. Shacter

CPP EXPERIMENTAL HEPA FILTER MEDIA W. D. Hanson

OPENING REMARKS OF SESSION CHAIRMAN:

The Open-End Session will be devoted to brief technical notes, to short presentations of new work that is in progress but not yet far enough along to justify a full formal report, to descriptions of puzzling observations that the audience will have an opportunity to respond to from their own experiences, and to statements of a policy or other nature that would not easily fit into any other part of the program.

The first speaker on the program is J. P. Olivier from the Organization for Economic Cooperation and Development. A formal presentation of his remarks is contained in the last paper of Session 12, entitled "New Air Cleaning Technology from Europe." Regrettably, the authors of this paper were unable to attend the Conference. Mr. Olivier has some items he would like to bring to our attention on sampling and measurement techniques for gaseous wastes and the testing of HEPA filters.

OLIVIER: I come from the Nuclear Energy Agency of OECD which is an inter-governmental organization. We are interested in and have been active in nuclear safety and waste management. Our interest in air cleaning technology started only two years ago. We are working to prepare a study on effluent releases from the nuclear fuel cycle and trying, on the basis of new ICRP Publication 26, to look at all the impacts in such a way that we can determine cost-benefit trade-offs for retention of radionuclides such as krypton-85, tritium, carbon-14, and iodine-129.

FIRST: Thank you for bringing this information to our attention. I am sure we will all be interested in reading the details in the paper by Dubé and Zabaluev. Your suggestion of an international agency to do testing is an interesting one. I don't think it would be valuable to repeat what is being done in each of the several countries, but I can think of at least a couple of areas where an international body could have an important impact. For example, they could pos-

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sibly resolve the several different test methods used for HEPA filters. We in the U.S. have the DOP method. U.K. has the sodium chloride method. France has the dye method. All of these are familiar to you. It would be useful if an international agency would study all these methods and tell us how they relate to one another. Another area for international action would be in HEPA filter design. We are trying to decide whether the European small pleat design is better or worse than the U.S. and U.K. wide pleat design. Certainly this is an important area for standardization.

Our next speaker is Mr. Charles Moore who is with the Florida Power and Light Co. He would like to tell us a bit about the use of installed test canisters for representative sampling of ventilation systems employing charcoal adsorbers.

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USE OF INSTALLED TEST CANNISTERS FOR REPRESENTATIVE SAMPLING OF VENTILATION SYSTEMS CHARCOAL ABSORBERS

Charles A. Moore
Chemistry Supervisor
St. Lucie Nuclear Plant
Florida Power and Light Company

INTRODUCTION

In Nuclear Power Plants, activated charcoal is routinely installed in the Emergency Safeguards Ventilation Systems for reduction of iodine releases under postulated post-accident clean-up conditions. Since charcoal is subject to several types of degradation, representative sampling methods must be utilized in order to perform analyses to ensure these systems are maintained at a proper level of iodine removal efficiency.

A common method of sampling currently in use is to install small test cannisters containing charcoal from batches used to fill the main absorber beds. These are usually placed so that the samples are exposed to the same general environmental conditions as the absorbers, i.e., temperature, humidity, etc.

CONCERN

While current standards recommend that the flow conditions of the test cannisters be the same as the main absorbers, limited testing has been performed to verify such conditions do exist in field installations.

CRITERIA

- 1) Typical Technical Specifications issued on ESF Ventilation Systems in the last few years require that--a laboratory analysis of a carbon sample from at least one test cannister--demonstrates a removal efficiency of 90% for radioactive methyl iodide when the sample is tested in accordance with ANSI N 510-1975---etc.
- 2) Regulatory Guide 1.52, Rev. 2 (March 1978) "Design, Testing, and Maintenance Criteria for Post Accident Engineered Safety Feature Atmosphere Clean-Up System Air Filtration and Absorber Units of Light Water Cooled Nuclear Power Plants" states in Section 6, paragraph b that---The efficiency of the Activated Carbon Absorbers Section should be determined by laboratory testing of representative samples of the activated carbon exposed simultaneously to the same service conditions as the absorber section.
- 3) ANSI N 509-1976 "Nuclear Power Plant Air Cleaning Units and Components" Appendix "A" Paragraph A.2.1 states that--The superficial face velocity of any test cannister shall be shown by calculation or direct measurement to be within ± 20 per cent of the superficial face velocity of the absorber bed.

LOCATION OF TESTING

The St. Lucie Nuclear Plant is an 853 MWE, Combustion Engineering Pressurized Water Reactor Plant of the Florida Power and Light Company System, located on the Atlantic Coast of Florida approximately 100 miles north of Miami. The plant was

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licensed in March, 1976 and commenced power operation in April, 1976.

The ventilation charcoal filtration devices were supplied by Mine Safety Appliance Company of Evans City, Pennsylvania. The plant systems were designed and installed by Ebasco Services, Inc., New York, New York, the Architect-Engineer on the St. Lucie Project.

CHARCOAL SYSTEM GENERAL DESCRIPTION

The St. Lucie Plant has four (4) major emergency safeguards ventilation systems containing activated charcoal which are addressed in the Plant Technical Specifications:

- 1) ECCS Area Exhaust System--Two (2) trains--30,000 CFM each
- 2) Shield Building Ventilation System--Two (2) trains--6,000 CFM each
- 3) Control Room Emergency Recirculation System--One (1) train--2,000 CFM
- 4) Fuel Handling Building Exhaust System--One (1) train--9,800 CFM

These systems contain the Mine Safety Appliances (MSA) tray-type absorber beds. The charcoal beds are two (2) inches in depth and each tray has a surface area of approximately eight (8) square feet. The beds are designed for a flow of approximately 333 CFM per bed, which gives a face velocity of approximately 42 feet per minute. St. Lucie Plant Technical Specifications allow a flow variation of +10 per cent from design.

The Test Cannisters have two (2) inches charcoal depth also and are 2½ inches in diameter. The cannisters are mounted between absorber racks with ¼ inch, Schedule 40 pipe, which has a total length of 33½ inches. The cannisters are located on the upstream side of the absorber racks.

TEST PROGRAM

During the Spring, 1978 refueling outage of the St. Lucie Plant, while conducting the routine Ventilation System surveillance, a test was performed to determine if the flow conditions of the test cannisters were representative of the actual flow conditions of the Main Absorber Trays.

The Shield Building Ventilation System operating at design flow of 6000 CFM, was selected as the test system. Measurements were taken with an Alnor Type 8500 Thermo-Anemometer outside the trays with flow through to the center and on the screen face of the carbon test cannisters. Measurements were repeated several times on different trays and different cannisters.

RESULTS

Measurements on the trays and cannisters showed a nominal face velocity of 45 feet per minute. (MSA in house testing has shown similar results.)

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CONCLUSION

Both the cannisters and carbon trays have a bed depth of two (2) inches and measurements in each case showed a nominal face velocity of 45 feet per minute; therefore, the cannisters in addition to environmental conditions are also representative of flow conditions at the St. Lucie Plant.

BENEFITS

Cannisters provide a convenient means of sampling charcoal without disturbing the main absorber installations. Alternate methods of acquiring such samples involve disassembling expensive trays and in some cases would involve additional in-place testing.

PERSONNEL PARTICIPATING IN THE TEST PROGRAM

- 1) Mr. Fred D. Leckie, Nuclear Containment Systems, Contractor for Ventilation Systems Testing at St. Lucie Plant.
- 2) Mr. Charles A. Moore, Chemistry Supervisor, St. Lucie Plant, Florida Power and Light Company.
- 3) Mr. Robert J. Frechette, Senior Plant Technician, Chemistry Department, St. Lucie Plant, Florida Power and Light Company.

ACKNOWLEDGEMENTS

- 1) Mr. Roger Zavodoski, Nuclear Regulatory Commission, Region II, Atlanta, Georgia and
- 2) Mr. Ron Bellamy, Nuclear Regulatory Commission, Washington, D.C. for providing incentive for this effort.

DISCUSSION

FIRST: After some of the other papers we've heard at this meeting, this seems like a very simple way to do it. Do other people complicate the problem?

HULL: Did you find comparable results for filters in steady and routine use? I assume the loadings are quite different for charcoals that are not on stream all the time.

MOORE: The systems that we have are all stand-by systems with the exception of the fuel ventilating system which is operated in conjunction with fuel movement for a period of time. We have just done measurements on our shield building ventilation system; we haven't done a complete set of measurements on all of the systems.

HULL: At 6,000 cfm, I would guess that the shield building is a recirculating clean-up unit within the building, isn't it?

MOORE: It exhausts out of the stack.

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Kup

A REMOTE SAMPLING SYSTEM FOR HEPA (1500 cfm) IN-PLACE FILTERS WITH A FLUID (CHANNEL) SEAL SYSTEM*

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"ABSTRACT"

High efficiency particulate aerosol (HEPA) filters are used extensively for air cleaning systems at the Idaho National Engineering Laboratory (INEL). These filters are occasionally located in inaccessible areas, in high radiation or toxic areas, or where design does not permit correct sampling distances. This paper describes certain advantages in using a specially designed multihole sampling probe in conjunction with a separatorless, fluid (channel) seal, filter and crab trap system. All work utilizing this system is conducted from outside the area, thus reducing personnel exposure and testing time, and providing accurate sampling. This system has enabled the INEL to meet the requirements for testing filter systems that otherwise could not have been tested.

1.0

Introduction

High efficiency particulate aerosol (HEPA) filters are used extensively for air cleaning systems at the Idaho National Engineering Laboratory (INEL). The Department of Energy (DOE) and the American National Standard (ANSI) Institute standard N-101.1 and N-510 require evaluation prior to placing a HEPA filter system in service and at regular intervals thereafter. These filters are occasionally difficult to evaluate because they can be located in banks, stages, series, inaccessible locations, and high level radiation or toxic areas (Figures 1,2,3,4,). This paper describes both the current method, and the remote sampling system for HEPA filters located in areas where accessibility is difficult.

2.0

Methodology

"CURRENT IN-PLACE TEST METHOD"

The current in-place method for evaluating HEPA air cleaning filter systems consist of introducing a challenge aerosol agent of dioctyl-phthalate (DOP), 0.3 micron partical size, in the air stream at 10 duct diameters upstream from the filter face. This distance ensures a thorough mixing of DOP in air. The sample ports are 5 to 7 duct diameters upstream, and 7 to 10 duct diameters downstream from the filter face.

* Work supported by the Idaho National Engineering Laboratory operated under the auspices of the U. S. Department of Energy, Idaho Operations Office, under contract No. EY-76-C-07-1570.

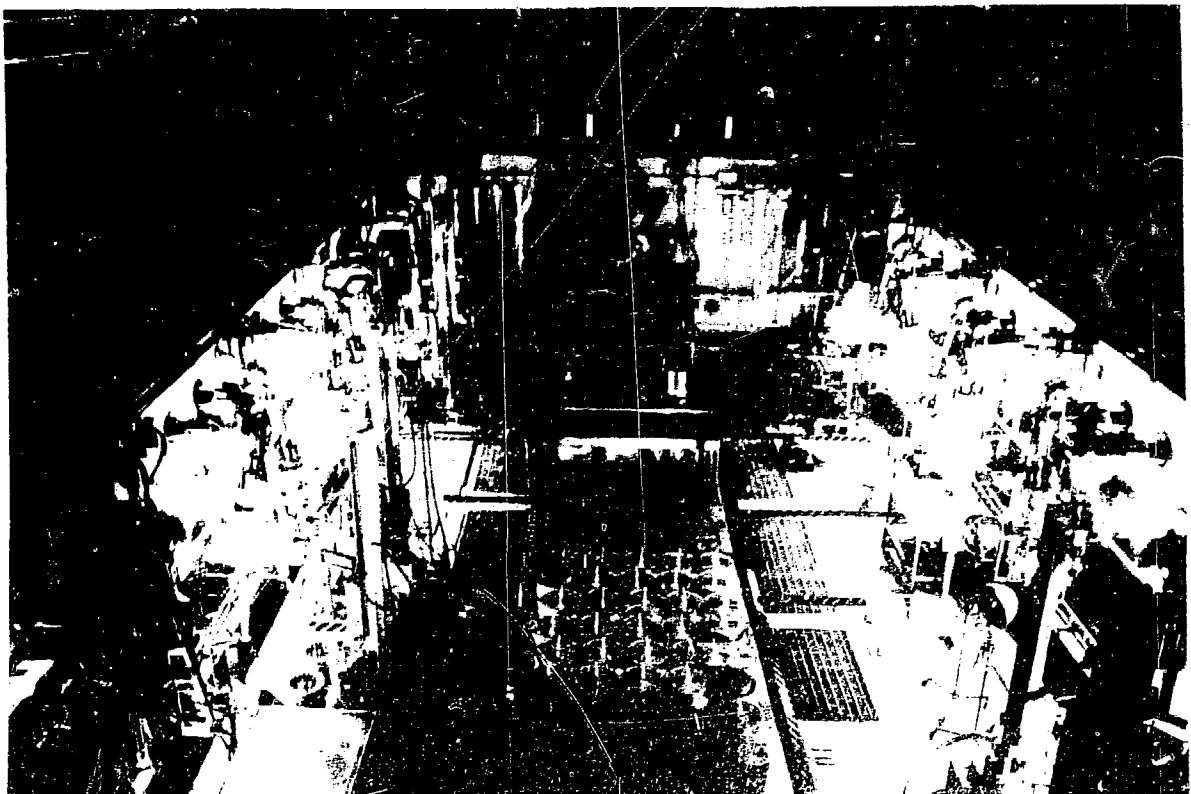
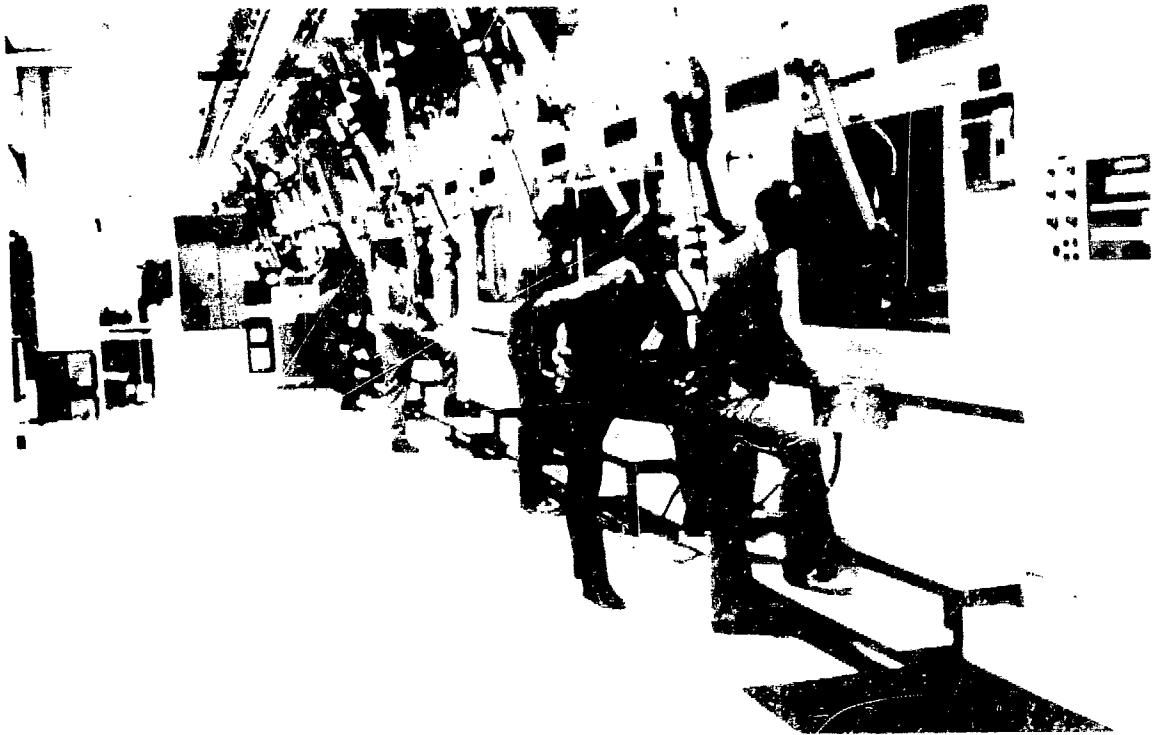


Fig. 2 Oxygen deficient or toxic area.

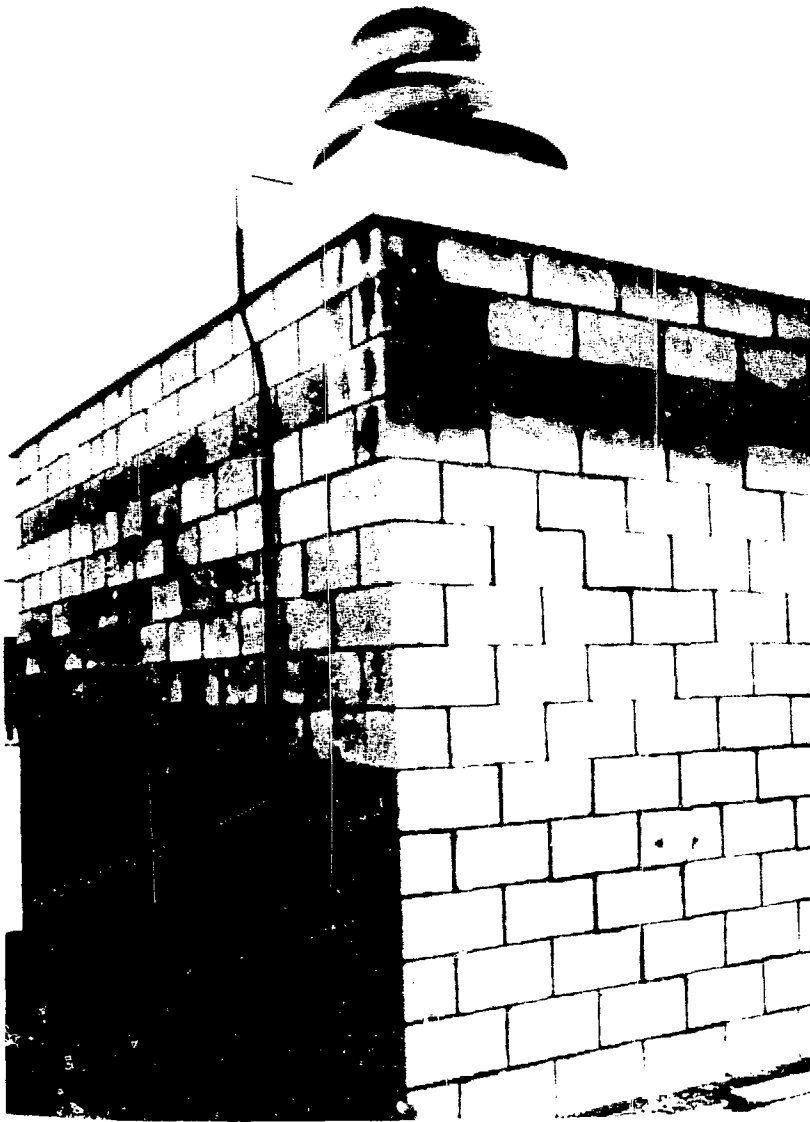


Fig. 3 Kerote area.

A probe connected to a light scattering photometer is then inserted into the sample ports to determine the DOP concentration upstream and downstream of the filter face. The above method requires personnel to be in the immediate vicinity of the filter face during testing.

The accuracy of the test is reduced whenever the filter system is located in inaccessible areas, high radiation areas, or where design does not permit correct sampling distances. Some of the problems encountered are:

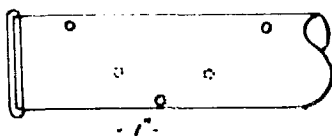
- (1) Personnel are exposed to excessive toxic material or radiation during testing of filters.
- (2) A challenge agent of DOP aerosol cannot be introduced at the proper distance from the filter face. This will result in an uneven distribution of DOP across the filter face.
- (3) DOP samples cannot be collected at the proper distance downwind of the HEPA filter.
- (4) Testing time is increased.



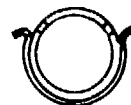
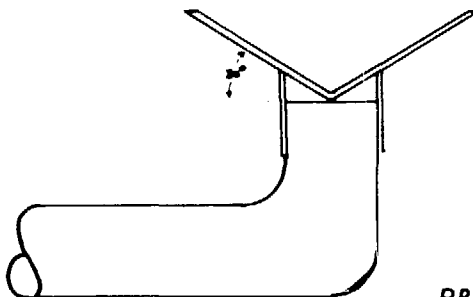
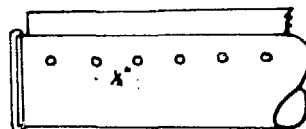
Fig. 4 Air circulation area, bank of filters.

D.O.P. INJECTORS

PROBE W/ HOLES
VARIANCE IN DUST $\pm 11\%$



PROBE W/ BAFFLE
VARIANCE IN DUST $\pm 7\%$



PROBE W/ INVERTED CAP
VARIANCE IN DUST $\pm 3\%$

Fig. 5 DOP injectors.

1. INJECT DOP WITH INVERTED CAP,
2. USE CRAB TRAP,
3. USE SPECIAL PROBE.

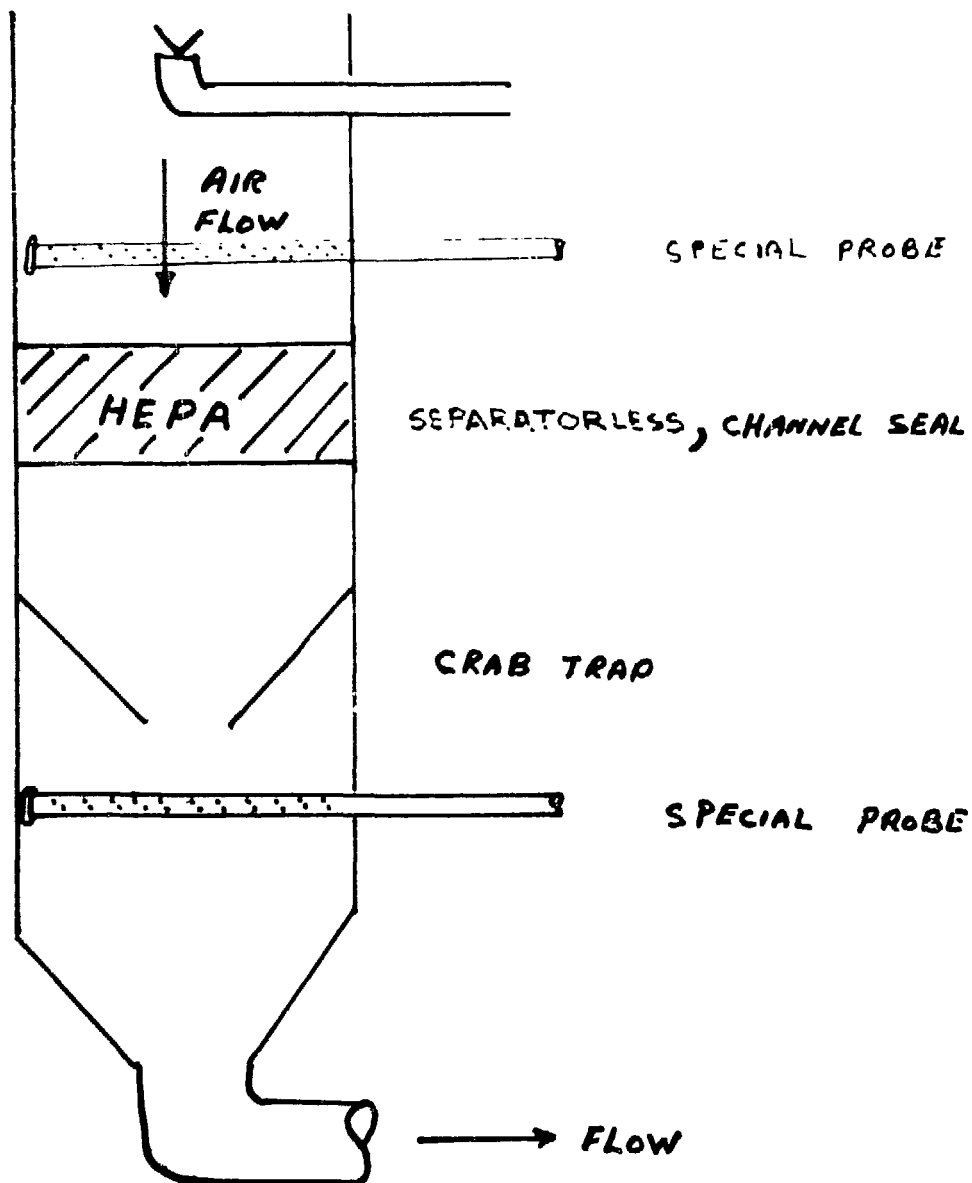


Fig. 6 Sample method for filter in series or small sample distance.

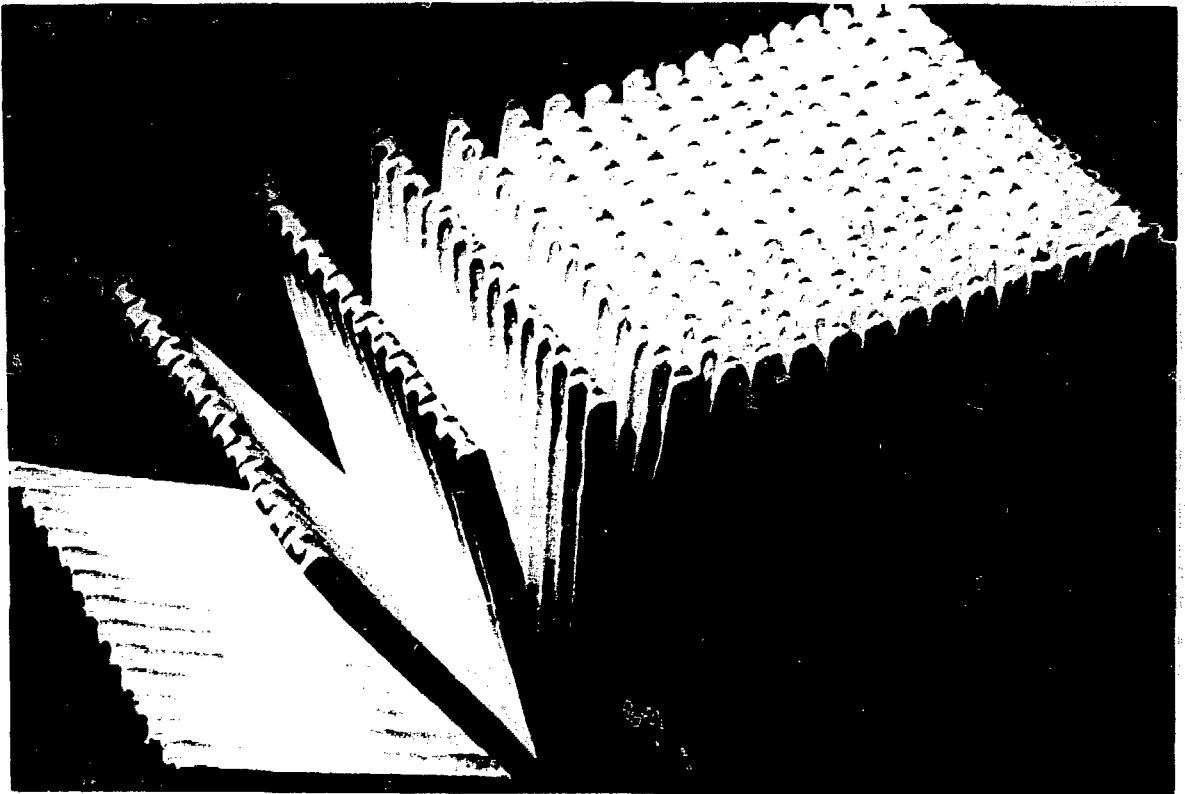
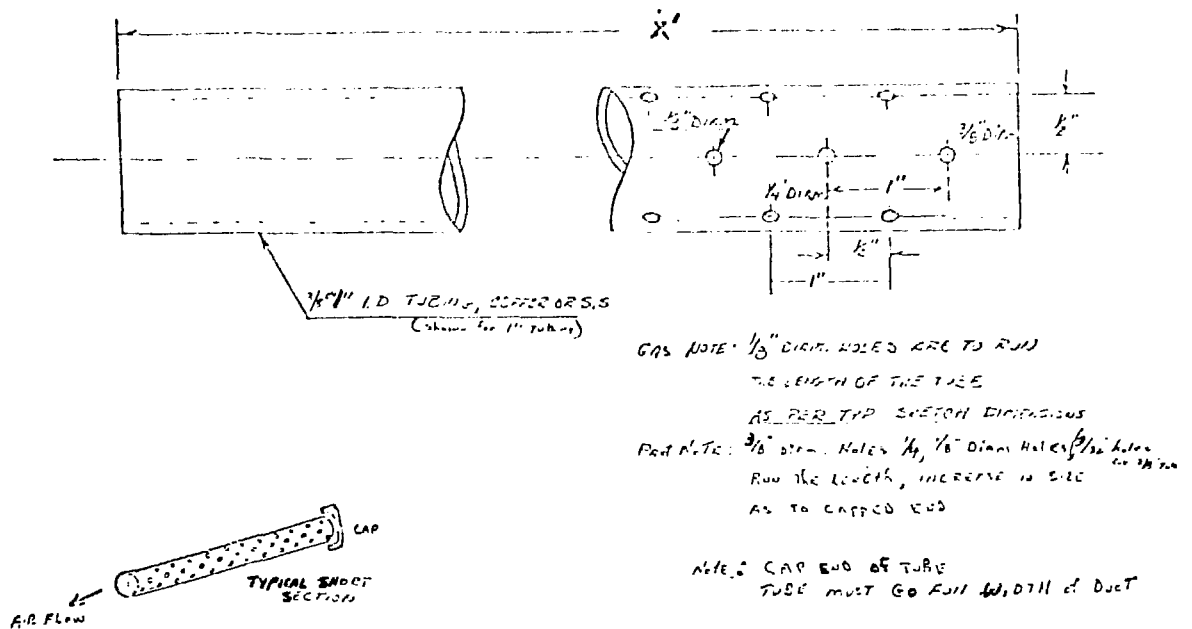


Fig. 7 Separatorless filter.



PROBE FOR HEPA EXHAUST DUCT

Fig. 8 Sample probe with multiholes.

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"REMOTE TEST METHOD"

The remote test method equipment consists of the following:

- (a) DOP injector probe (Figure 5).
- (b) Crab trap (Figure 6).
- (c) Separatorless filter with fluid "channel" seal system (Figure 7).
- (d) DOP multihole sample probe (Figure 8).

DOP is injected into the system by using the inverted cap injector probe (Figure 5). A "100 " DOP concentration is obtained upstream of the filter face. Downstream from the injector probe is a nuclear grade separatorless HEPA filter. A fluid "channel" seal system is used with these filters. This reduces gasket seal problems on installation by about 80 per cent. This type of filter system is used almost exclusively at the INEL for remote sampling.

Located downstream from the filter system is a "crab trap" designed by Flanders Filters, Inc. It is designed to ensure a turbulent mixture of DOP in air, and can be moved out of the way for normal air flow operations. The "crab trap" is located downstream from the filter at a distance equal to the width of the filter.

The multihole sample probe is located downstream from the "crab trap" $1\frac{1}{2}$ times the width of the "crab trap" opening. The initial test utilized a variety of sample probes. These probes were isokinetic, straight pipe, bent pipe, pipe with holes on the side, and a "U" pipe. The probe determined to be the best overall was the multihole probe (Table I).

Tubing with an inside diameter of 0.375 inches is connected from the permanently installed multihole duct sample probe to a remotely located light scattering photometer (Figure 9). The DOP injection line is also connected from the injector probe to the DOP generator. The DOP generator is located in the same area as the light scattering photometer (Figure 9). All lines should be kept as short as practical to reduce lag time, to increase efficiency, and reduce the possibility of DOP plating out on the lines.

3.0

Discussion

HEPA filters are occasionally located in inaccessible areas, high radiation areas, or where design does not permit correct sampling distances. This remote test method was developed to reduce possible employee exposures, improve testing efficiencies, and reduce testing time. The remote sampling system, in one instance, has enabled testing of 26 individual filters in a hot cell, by one person, in less than one hour with no measurable personnel exposure.

"ADVANTAGES OF THE SYSTEM"

- (a) The DOP filter test is conducted from outside the immediate area, thus reducing personnel risk.
- (b) The DOP sample injection can be used to test one or more units either

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individually or as a system.

- (c) The DOP sample injection is evenly distributed over the filter face.
- (d) An accurate DOP test can be obtained when filters are in close space arrangements of a bank.
- (e) The remote system reduces personnel exposures.
- (f) The remote system reduces personhours required for the test and enables one person to perform an accurate test.
- (g) The separatorless filters have a 2 to 3 times longer life expectancy with virtually no sealant problems.

"SUGGESTED TEST PROCEDURES"

Installation

- (1) Install a dispersion cap in the center of the duct and install a fall-away "crab trap"
- (2) Install a channel system knife edge and a separatorless channel (fluid) system HEPA filter
- (3) Install a circular drilled multihole sample probe in the correct location across the entire duct as indicated by a traverse.

Procedure

- (1) Check out and set up for operation the photometer and the generator (ensure proper warmup of the generator)
- (2) Blow a small amount of gas or air through the sample tube to ensure a relatively clean line
- (3) Connect the aerosol generator to the injection pipe
- (4) Connect the photometer to the upstream sample tube
- (5) Swing "crab trap" into the sample position
- (6) Obtain the upstream sample
- (7) Move the photometer sample tube to the downstream sample tube and obtain a sample
- (8) Disconnect the photometer and swing "crab trap" into the stored (manual) position
- (9) Record and evaluate the sample data

Conclusion

The remote test system has proven very successful when HEPA filters are located in inaccessible areas, high radiation or toxic areas, or where design does not permit correct sampling distances. This system has enabled the INEL to meet the requirements for testing HEPA filter systems that otherwise could not have been tested

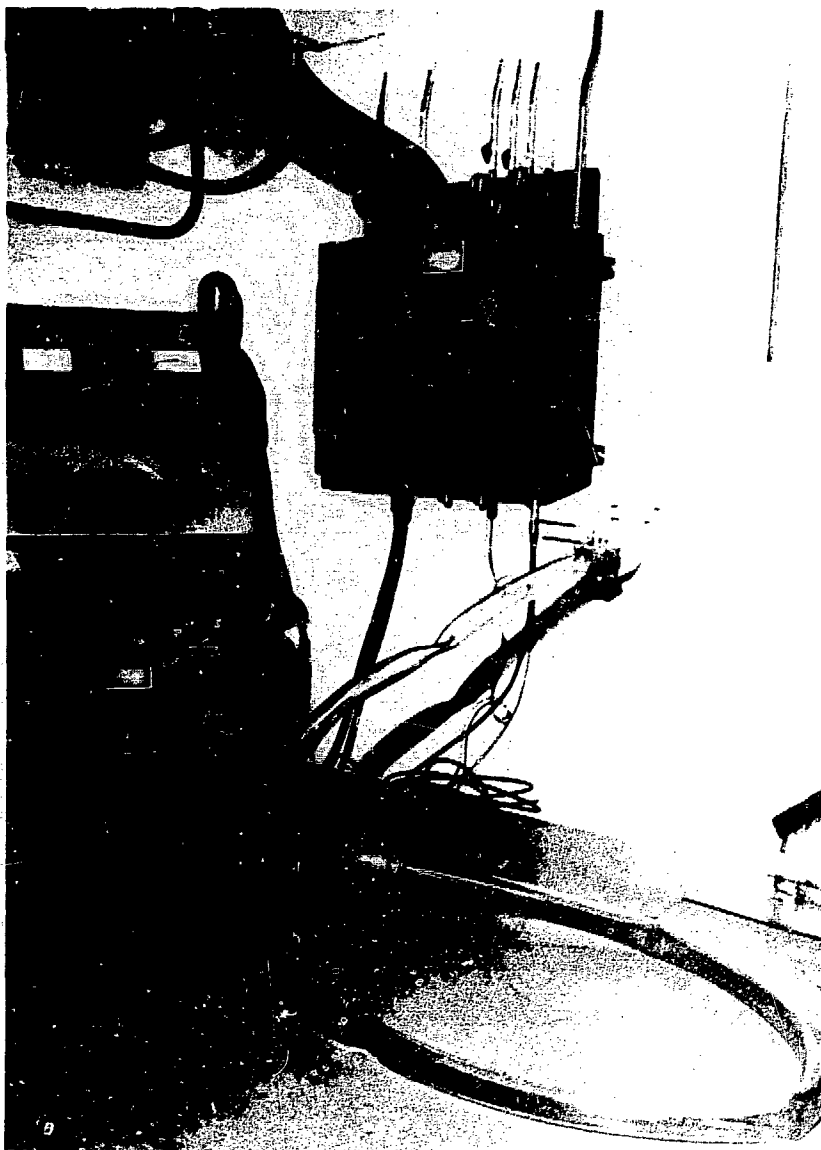


Fig. 9 Remote sample by one person.

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DETECTOR TDA-2D

(flow rate 25 and 50 f/m)

| Test Number | Upstream Concentration | Type of Probe
(downstream of filter) | | | | | |
|-------------|------------------------|---|----------------------------|-------------------------|-------------------------|------------------------|---------------|
| | | Probe with Holes
(3/8 in.) | Straight Pipe
(3/8 in.) | Isokinetic
(1/8 in.) | Isokinetic
(1/4 in.) | Bent Pipe
(3/8 in.) | "U" Bent Pipe |
| 1 | 100 | 0.013 | 0.011 | 0.011 | 0.011 | 0.012 | 0.015 |
| 2 | 100 | 0.013 | 0.011 | 0.012 | 0.012 | 0.013 | 0.016 |
| 3 | 100 | 0.014 | 0.011 | 0.012 | 0.012 | 0.013 | 0.015 |
| 4 | 100 | 0.013 | 0.011 | 0.012 | 0.012 | 0.002 | 0.016 |
| 5 | 100 | 0.014 | 0.012 | 0.012 | 0.012 | 0.013 | 0.017 |
| 6 | 100 | 0.013 | 0.012 | 0.012 | 0.012 | 0.012 | 0.016 |
| 7[a] | 100 | 3.1 | 2.6 | 2.8 | 2.8 | 2.7 | 4.0 |
| 8[a] | 100 | 3.4 | 2.8 | 2.7 | 2.8 | 2.4 | 3.2 |
| 9[a] | 50 | 3.2 | 2.7 | 2.8 | 2.8 | 2.5 | 3.9 |
| 10[a] | 25 | 3.2 | 3.1 | 2.9 | 3.0 | 2.8 | 4.0 |
| 11[a] | 100 | 3.3 | 2.6 | 2.8 | 3.0 | 3.0 | 6.4 |
| 12[a] | 100 | 3.2 | 2.5 | 2.7 | 2.6 | 2.3 | 4.0 |

[a] Hole in filter unit. The ratio was best on the special probe and compared very well to isokinetic.

Table I Sample Probe Test Comparison

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DISCUSSION

FIRST: We have heard other papers in this Conference on testing. This is a little different variation. Mr. Musen, could you tell me why you thought it was worthwhile to do your studies to see if isokinetic or nonisokinetic sampling had an effect when the particle size is well below one micrometer?

MUSEN: The main reason is that we wanted to look at all the different types of sample probes. We considered isokinetic sampling to be the state of the art.

FIRST: I would be very skeptical of a multiple port probe if you did have particles that were larger than one micrometer because then inertial effects would be present. Even though the holes were distributed uniformly over your probe, there is no guarantee that the particles would be uniformly distributed over the duct into which the probe was placed so you might get some false results for this reason.

MUSEN: This was considered but since the filter would not otherwise be capable of being tested, it was better to use it than not to test the filters at all. A loss of only $\pm 4-6\%$ in efficiency resulted.

FIRST: Dr. Sokol's subject matter is a review of the current technical status of high level waste disposal with emphasis on the recommended use of multiple barriers.

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RADIOACTIVE WASTE MANAGEMENT THE NEED FOR MULTIPLE BARRIERS

J. Sokol and M. Cooper
Westinghouse Advanced Power Systems Divisions
Madison, Pennsylvania

Abstract

Isolation criteria for the disposal of high-level radioactive waste are critically examined. The results indicate that the essential period for isolation of high-level waste is about 1,000 years. Multiple barriers, such as a solidified, leach-resistant waste form (i.e. fixation in glass), a corrosion-resistant outer container, and a stable geological formation, which limit the transport of radioactivity into the human food-chain are recommended. The multiplicity of barriers allows for the unlikely event of failure in one or two of the barriers while still providing adequate isolation of the waste.

I. Introduction

The essential period for the containment of high-level waste is about 1,000 years (Figure 1). This is based on a comparison of the hazard using the federally allowed Maximum Permissible Concentrations (MPCs) in water, of the fission product and actinides in the waste produced from the reprocessing of a ton of nuclear fuel to the hazard of the naturally occurring, radioactive uranium ore which was mined to produce this ton of fuel.^(1,2) The MPCs take into account the biological effects produced by individual radionuclides and the use of MPCs in water reflect the fact that flowing water is the most likely mechanism for transporting the radionuclides in the water to the human food chain.

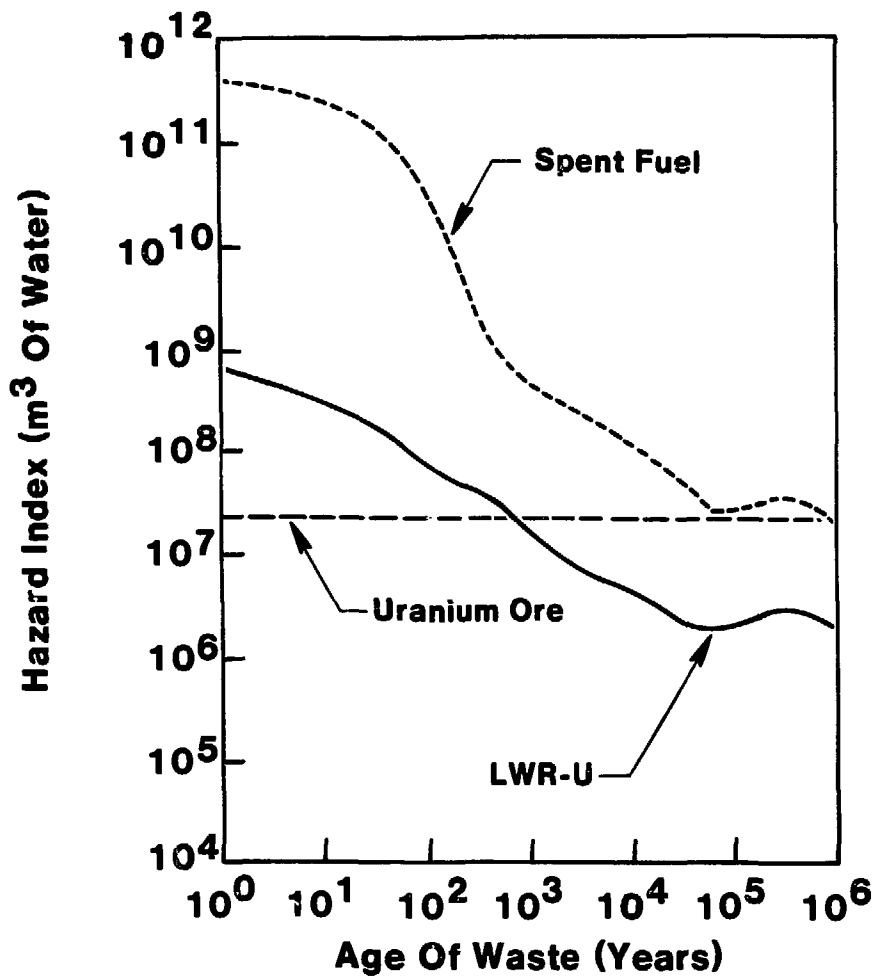
Since the first 1,000 years are the essential period for containment of the waste and considering the advances which have been made on waste management technology, a waste management system which includes an engineered barrier, as well as geological barriers, has the best chance of meeting the technical and public acceptance requirements for a waste management system. Having a multiplicity of barriers allows for the imperfect knowledge which will exist about the integrity of any one of these barriers. Such an approach for the disposal of high-level radioactive waste would involve consideration of the solid form of the waste, the engineered outer container, and the geological medium for disposal.

II. Solid Waste Forms

For all practical geological disposal modes, the leach rate of the ultimate product is an important factor in determining potential hazards from buried waste. Borosilicate glass waste forms have been produced in the U.S. with initial leach rates of 10^{-6} g/cm²/day. Battelle Laboratories has estimated that the leach time for such a 1-ft. dia. x 10-ft. long glass monolith is approximately 150,000 years.

Similar borosilicate glass forms have been produced in France with initial leach rates in the range of 10^{-7} to 10^{-8} g/cm²/day. Nepheline syenite glass waste forms from Canada show a leach rate of 4×10^{-11} g/cm²/day in ground water after remaining in situ for 18 years.⁽³⁾

SUMMARY OF HAZARD-INDEX COMPONENTS



Reference: NUREG 0216,1977

Figure 1

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III. Engineered Containment

Historically, the U.S. Program has not emphasized the concept of a well-engineered outer container for high-level waste. For example, in bedded salt the stainless steel canister was assumed to be corroded by the salt in a few years.⁽⁴⁾ One of the objections to Project Salt Vault in Lyons, Kansas was the lack of retrievability for the waste.⁽⁵⁾ In a waste disposal system developed in Sweden, a containment system, which uses a layer of lead 100 mm thick surrounded by 6 mm of titanium, has been devised to provide 500 to 1000 years of retrievability.⁽⁶⁾ To add to the integrity of their high-level waste disposal system, the Swedish plan includes a packing of bentonite and quartz-sand around the canisters which decreases the flow rate of the water by a factor of 200 or more and therefore reduces the release rates of radionuclides in the unlikely event that water enters the repository.

IV. Geological Disposal Mediums

For two decades, disposal in bedded salt has been the leading high-level waste disposal medium in the U.S. The advantages of bedded salt are its high thermal conductivity, resistance to radiation damage, and the ability to flow plastically under moderate pressure and "heal" breaks in the formation.⁽⁷⁾

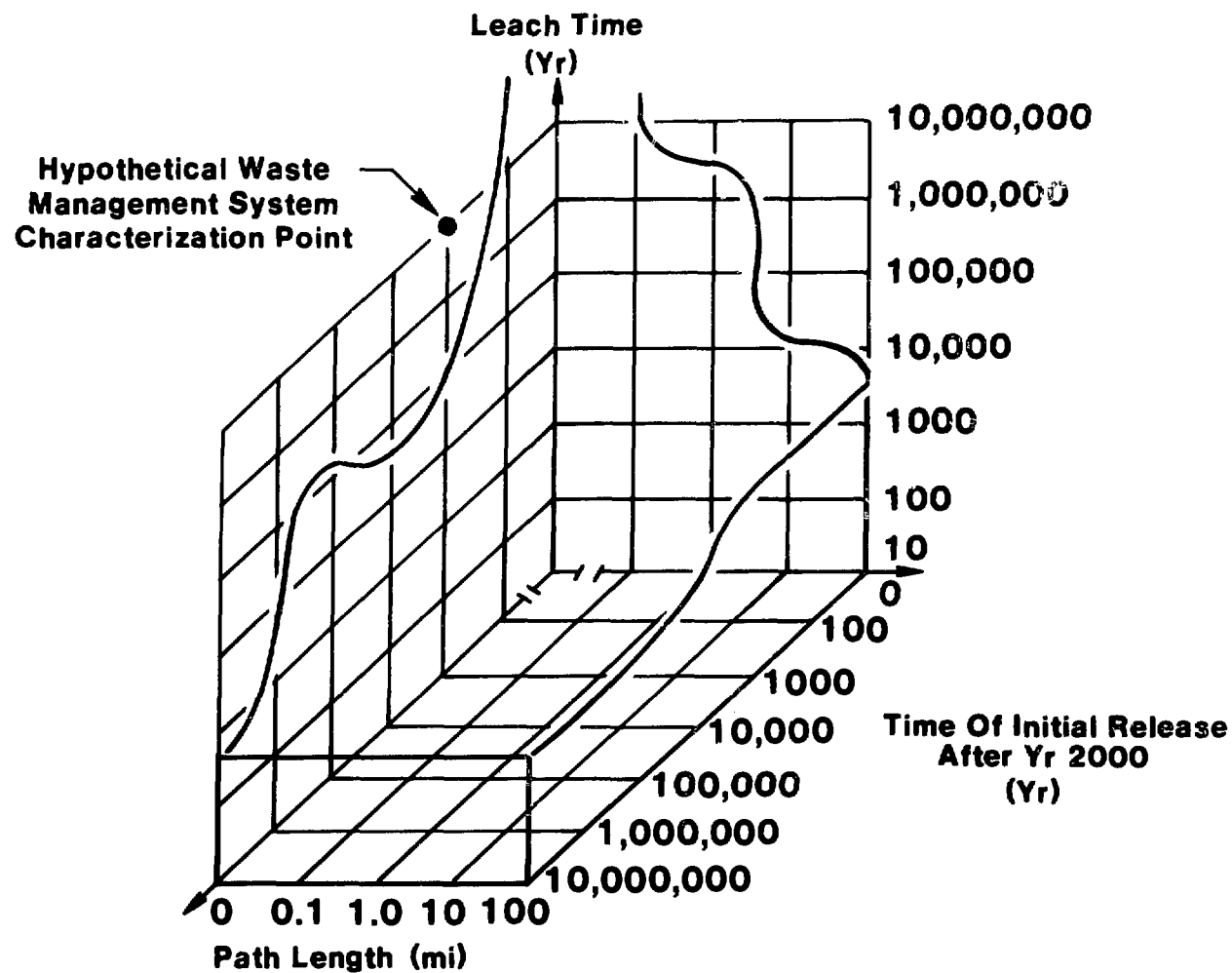
The crystalline rocks, such as the granites and basalts, under investigation in both the U.S. and Sweden, lack some of salt's desirable thermal and mechanical properties but represent a less corrosive environment. Also, the public image of this type of material is better ("hard as a rock"). Since the number of disposal sites in bedded salt which meet all the requirements for disposal of radioactive waste may be limited, other geological mediums are under investigation in the U.S. and overseas which appear to also offer the potential as an acceptable medium for geological disposal of radioactive waste.⁽⁸⁾

V. Hazard Assessment

A realistic assessment of the waste disposal hazards must include considerations of the leach-resistance of the waste form, the engineered containment, and the absorption of the waste in the surrounding geological medium. Recent studies in both the U.S. and Sweden have considered the effects of these various barriers.

Studies by Battelle Pacific-Northwest Laboratory considered all the solidified high-level waste produced by commercial nuclear power plant operations by the year 2000 placed in a repository in typical western U.S. desert soil. These calculations considered variations in the leach-resistance of the solidified waste form, the time before containment is breached, and the path length between the repository and a body of water at the surface. The results were in terms of a radiation dose to an individual who obtained all his food and water from water which has been contaminated.⁽³⁾ The results of these studies (Figure 2) indicated that using realistic assumptions, the maximum dose to this individual would be a fraction of the normal dose due to background radiation. In fact, using this analysis and assuming a well engineered container (1000 years of containment) and a high leach-resistance glass (10^{-8} g/cm²/day), the high-level waste would not pose a serious environmental hazard even if buried in near surface storage in a dry area.

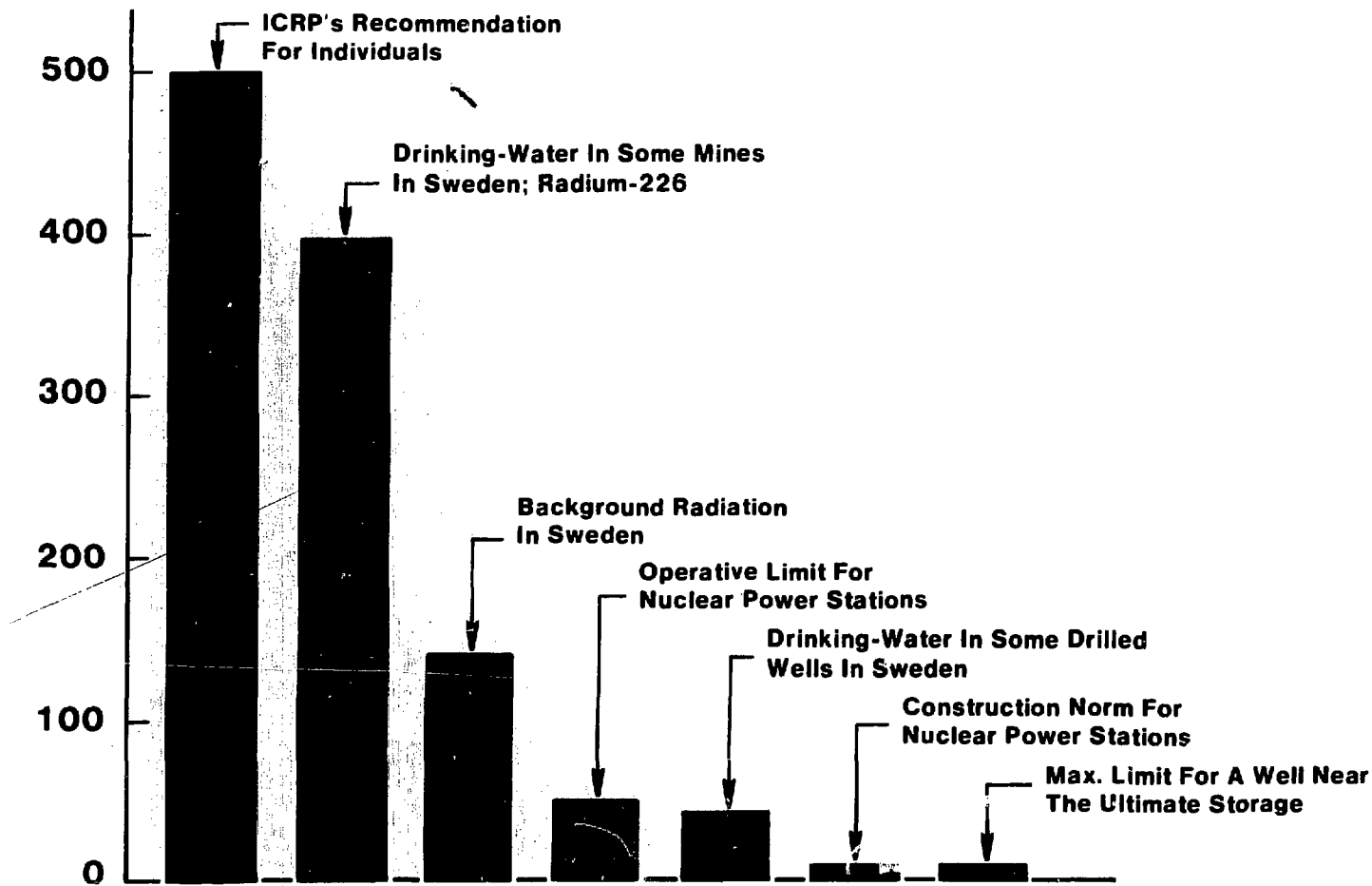
WASTE MANAGEMENT CONTROL SURFACE FOR INCREMENTAL BACKGROUND DOSE



Reference: BNWL-1927, 1975

Figure 2

Radiation Dose mrem/year



Reference: KBS, Report No. 1, 1978

Figure 3

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A similar study done in Sweden (Figure 3) for solidified high-level waste disposed in granite showed that the maximum dose to an individual would not be more than 13 mrem (about 10% of background radiation) and is below the dose some people in Sweden receive from naturally occurring minerals in drinking water (about 40 mrems).⁽⁹⁾ Of course, if the engineered barrier were still intact, then no radioactivity would be released and a small defect in the engineered barrier, such as a crack or hole, would only allow a small fraction of the radioactivity (the 13 mrem) to reach the critical group.

By combining a series of engineered barriers, which can be qualified by accelerated life testing, and geological barriers, a disposal system can be developed using current technology which can meet the current political/public acceptance, as well as technical requirements for the safe disposal of high-level waste.

DISCUSSION

HULL: From the health physics standpoint, there is a lot of radioactivity in nature, but nobody worries about it because it is dispersed. Since the long run problem of high-level wastes is not so much the amount as the concentration, doesn't this make deep ocean sediment disposal, that the British are looking at, attractive? If the containers leak from there, the effluent would be widely dispersed. Also, it would be far removed from manmade intrusions. Nobody is going to sink into it unless they're looking for oil at 10,000 or 20,000 foot depths some time in the future.

SOKOL: There has been some work done on this by Nuclear Safety Associates. You're talking about human intrusion which is apart from general public health. If you say that after 1,000 years the container is going to go and the material is going to spread out somewhat, it turns out that it doesn't have to spread out very much before you are down to the same concentrations of uranium ore you started with. In other words, you would only have to go 30 feet to the side of these 1 x 10 ft. canisters before you would be down to concentrations similar to the uranium ores people are finding in places like Canada and early parts of the Grant's Belt mineral district. Therefore, it requires a system of container design that optimizes for containment, which is important for the general public, and isolation, which is related to the fear that some idiot will dig 3,000 feet down into salt or granite for something that he can find easier elsewhere. One of the advantages of granite is you can mine it deeper and this may make it easier to optimize for containment and isolation from human intrusion. But you can do the same if you adopt more than a single barrier.

HULL: Maybe we have to do this in order to obtain public acceptance of nuclear power, but I'm a health physicist and a member of the American Public Health Association and I keep thinking, "If only we could transfer this money that we're spending for all this redundancy plus redundancy into real public health issues, we could really improve the public health." I think we ought to keep saying to the public that it could be much better spent. This will create a perception of sensible options on risk prevention.

SOKOL: I will reiterate what I said about containment buildings. Early European reactors did not have large concrete and steel containment structures. They now have to have them. Your point is well taken, but we have to deal with the political realities of our times.

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DUGGAN: Would you comment on the cost of using a three containment system for disposal of nuclear wastes? It seems that it is political to some extent; that we are trying to make the public feel more safe about the way we plan to dispose of our wastes. Yet, at the same time, we can't make disposal of the waste more expensive than producing the power in the first place. How do you balance cost-effectiveness against gains in assurance of containment?

SOKOL: Most of the figures that I have seen from the Department of Energy show that very elaborate schemes, such as separating out parts of the waste and putting it in outer space shift power cost 5% at the most. When you consider that waste disposal represents a small fraction of the total fuel cycle cost, you have tremendous freedom in designing the system. The Swedes have said that economics--at least initially--is not a factor. We want a system that will be safe without a lot of dickering over cost-effectiveness. You may find a system 15, 20, or 30 years from now that lets you eliminate or reduce some of your dependence on one of the barriers, but right now you're talking about being penny-wise in terms of saving money on one of these barriers and pound foolish because you won't have the generating stations that you need and you won't have the lowest price form of generating system that you need. People have talked about a tenth of a mil per kilowatt hour for some of these disposal schemes, and there is tremendous leeway in that economic area, so it's silly to get hung up on that part of it; at least at this stage of the game. I have my own reasons for not being keen on disposal in outer space, but that's another topic.

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CSL ENGINEERING DESIGN HANDBOOK

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Abstract

An engineering design handbook recently published describes a high efficiency air cleaning filtration system. This system, modified from the basic nuclear concept, was designed to meet the criteria for a chemical demilitarization facility. Topics covered in the book include both basic and detailed filtration and ventilation design considerations.

Presentation

I am here today to discuss a recently released Chemical Systems Laboratory document entitled "Engineering Design Handbook for Air Cleaning for Chemical Demilitarization." (Figure 1).

Why prepare an Engineering Design Handbook? The answer is simple. There is a lack of published information available on this subject and the information is relevant to both government and industry.

In late 1973, the Chemical Systems Laboratory located at Aberdeen Proving Ground, Md., was assigned the task of designing an air cleaning system for a facility located at the Tooele Army Depot, Utah. This facility, called Chemical Agent Munitions Disposal System (CAMDS), (Figure 2), was to dispose of obsolete chemical munitions containing toxic agents. To meet the stringent requirements imposed by Public Law and other regulations relating to safeguarding the health and safety of workers and the environmental quality of air discharged to the atmosphere, a high efficiency air cleaning system was required. By high efficiency air cleaning system, I do not mean a general industrial or air pollution control system, but a system equivalent to a nuclear air cleaning system, capable of handling the extremely hazardous material to be contained.

During the initial design effort, an extensive search for published literature on filtration systems was conducted. The typical chemical filtration system at that time was the chemical-biological-radiological (CBR) unit which consisted of a roughing filter, a HEPA filter and a specially made wood encased adsorber. When this design was compared to the basic nuclear concept, the nuclear concept had more potential advantages. The information that was incorporated into the various designs came mainly from ORNL-NSIC-65 (the predecessor of the current nuclear air cleaning handbook) and "Industrial Ventilation" (a manual prepared by the American Conference of Government Industrial Hygienists).

Over the next four years the CAMDS system was completed and successfully tested. Since this system was judged to be a significant improvement over previous air cleaning systems built by the US Army, it was desirable that the experience gained as a result of the effort be suitably documented to provide baseline data for designers of future systems of this type. Thus, this handbook was prepared.

The information contained in the handbook is shown in Figures 3 - 5. Figure

AD

CONTRACTOR REPORT ARCSL-CR-78022

**ENGINEERING DESIGN HANDBOOK
FOR
AIR CLEANING FOR CHEMICAL
DEMILITARIZATION**

by

**Marvin E. Siegel (AAI Corp.)
Arthur Shacter (CSL/APG)**

**AAI Corporation
P.O. Box 6767
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Project No. 655



**US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
Chemical Systems Laboratory
Aberdeen Proving Ground, Maryland 21010**

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FIGURE 1

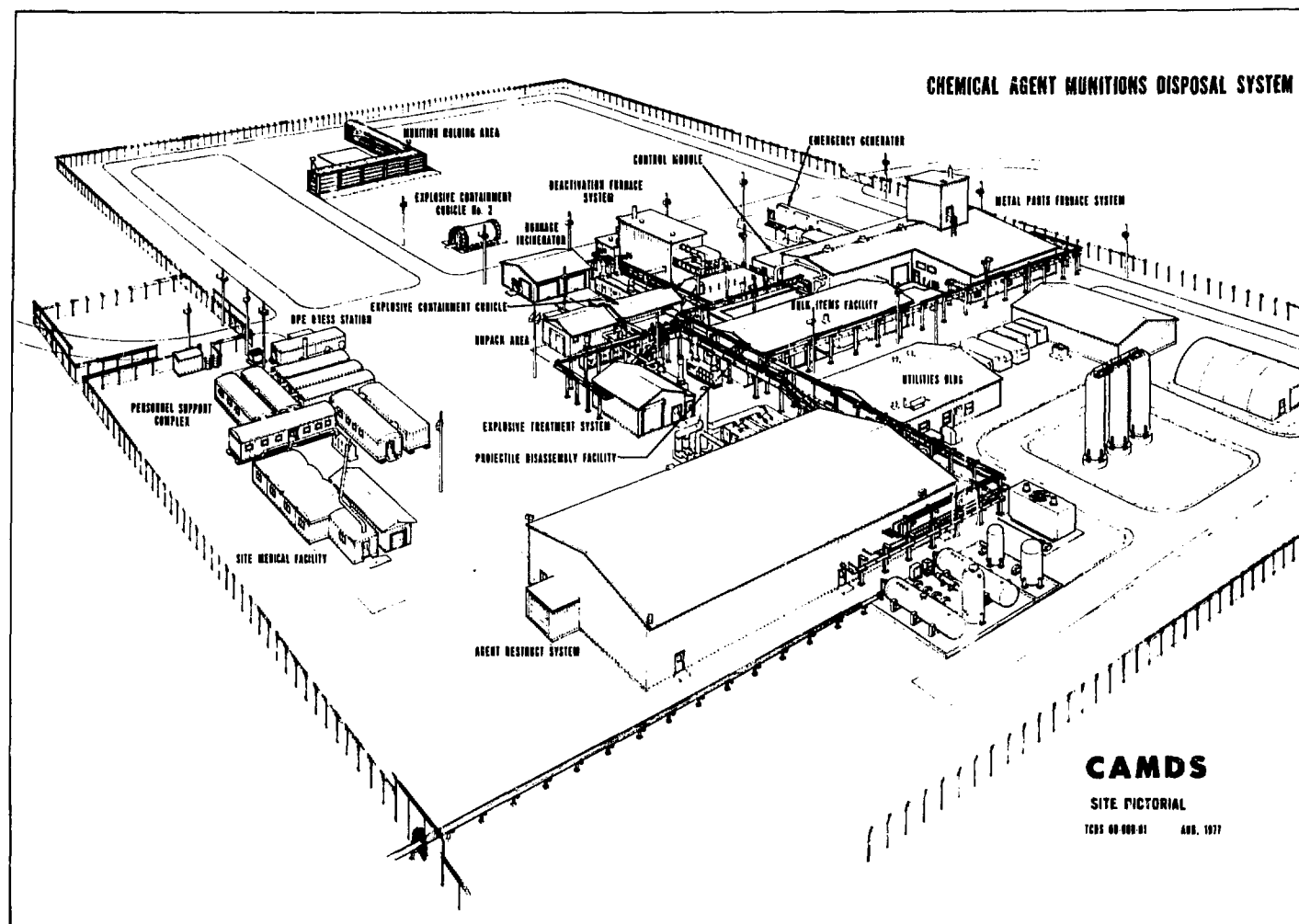


FIGURE 2

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TABLE OF CONTENTS

- 1. INTRODUCTION**
- 2. REQUIREMENTS FOR AIR CLEANING FILTRATION SYSTEMS**
- 3. BASIC DESIGN CONSIDERATIONS FOR EXHAUST SYSTEM**
- 4. DETAILED DESIGN CONSIDERATIONS**
- 5. OPERATIONAL CONSIDERATIONS**
- 6. BASIC MAINTENANCE AND STORAGE CONSIDERATIONS**
- 7. TESTING**
- 8. APPLICATION OF AIR CLEANING CONCEPTS TO DATS**
- 9. LITERATURE CITED**
- 10. GLOSSARY OF TERMS**

APPENDIXES

- A. PROPOSED MAXIMUM ALLOWABLE LIMITS FOR EMISSIONS OF GB, VX, AND MUSTARD FOR CHEMICAL DEMILITARIZATION**
- B. SPECIFICATION FOR CAMDS AIR CLEANING SYSTEM**
- C. DETAILED ADSORBER BED CONSIDERATIONS**
- D. GENERAL CHARACTERISTICS OF ACTIVATED CARBON**

FIGURE 3: HANDBOOK CHAPTER HEADINGS AND APPENDIXES

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3. BASIC DESIGN CONSIDERATIONS FOR EXHAUST SYSTEM

3.1 AGENT VAPOR REMOVAL METHODS

3.2 CRITERIA FOR DETERMINING VENTILATION RATES

3.3 FILTER CAPACITY

3.4 SAFETY

3.5 OPERATION AND DESCRIPTION OF KEY COMPONENTS

3.6 EXTERNAL COMPONENTS

3.7 EXTREME ENVIRONMENTAL CONDITIONS

FIGURE 4: CHAPTER 3 - SUBHEADINGS

4. DETAILED DESIGN CONSIDERATIONS

4.1 FILTRATION SYSTEM

4.1.1 CONFIGURATION

4.1.2 FILTER HOUSING DETAILS

4.1.3 FILTERS AND ADSORBERS

4.2 VENTILATION SYSTEM DESIGN CRITERIA

4.2.1 HOODS

4.2.2 DUCT DESIGN

4.2.3 AIRLOCK DESIGN

4.2.4 FAN SELECTION

4.2.5 DAMPERS

4.2.6 AUTOMATIC CONTROLS

4.2.7 MAKEUP AIR

4.2.8 HEAT CONTROL

4.2.9 REDUNDANCY AND SAFETY FEATURES

FIGURE 5: CHAPTER 4 SUBHEADINGS

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3 lists the 10 chapter headings and the appendixes. Note Appendixes C and D. These were written by Dr. Leonard Jonas of the Research Division of Chemical Systems Laboratory. Appendix C is a mathematical description on how the adsorptive process works and Appendix D explains how various factors effect the adsorptive capability of carbon.

The major portion of the design information is contained in Chapters 3 and 4 as shown in Figures 4 and 5.

These next figures are typical of the information contained in this handbook. Figure 6 is an isometric of a walk-in type filter housing. The handbook discusses in detail, three different types of filter housing designs: The walk-in, the bag-in, bag-out, and the walk-in with stationary absorber. Five separate banks of cells (prefilter, HEPA, adsorber, adsorber and HEPA) can be seen in the filter housing. All of the CAMDS filter housings have this same bank arrangement. The handbook also explains the function of each bank, discusses mounting frame configurations, clamping techniques for the cells, instrumentation, testing, etc.

Figure 7 shows one of the bag-in-bag-out filter housings as it is installed at the CAMDS site. You will note that this filter housing is located outdoors. The notes in the picture are pointing out two butterfly dampers in the ductwork and an inter-connecting duct to another filter, which enables a partial parallel redundancy to be achieved between the filter housing shown in the foreground and one in the background.

Figure 8 shows the overall CAMDS filter system layout. Each solid black rectangle represents one filter housing and each dark black line represents the ductwork. There are 10 separate filter units. The filter units vary in capacity from 333 CFM to 30,000 CFM. (Filter housings #9 and #10 with a capacity of 15,000 CFMs each are in parallel. You will note that most of the air cleaning systems are connected to one and in some cases, two other air cleaning systems. This provides a partial parallel redundancy without requiring additional filter housings. The handbook discusses in both general and specific terms, series and parallel redundancy, sizing of filter unit, exhaust stack requirement, access requirements, power requirements, etc.

As you may have noted from these last three slides, the CAMDS system is an extrapolation of the basic nuclear air cleaning concept, modified to meet the specific requirements of ventilating a chemical demilitarization facility, and the filter housings are similar to some nuclear units.

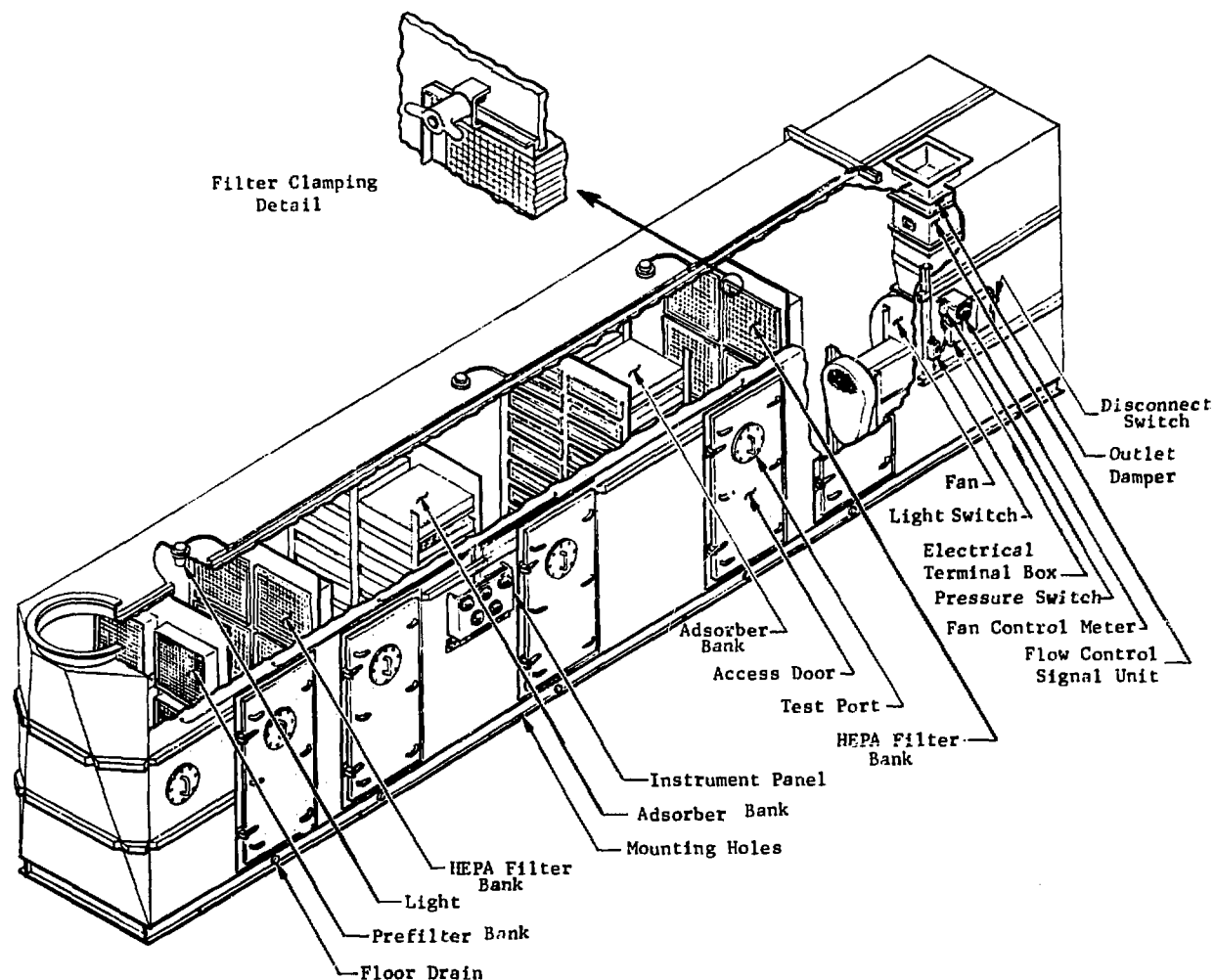
The major differences between this system and the typical nuclear system are:

(1) Adsorbent - The CAMDS adsorber cells contain an unimpregnated activated carbon whereas the nuclear field uses an iodine impregnated carbon.

(2) Location - All CAMDS filter housings are located outdoors whereas most nuclear units are located indoors.

(3) Adsorber Bank Arrangement Requirement - CAMDS has two series - redundant adsorber banks (2" bed depth) with agent monitoring equipment between, whereas this is not a requirement in the nuclear field.

(4) Maximum temperature of (110°F) and maximum pressure requirements of 24" H₂O are lower than for nuclear systems.



Type II Filter System (6,000 Cfm), Walk-In
(Courtesy Of CTI-Nuclear, Inc.)

FIGURE 6

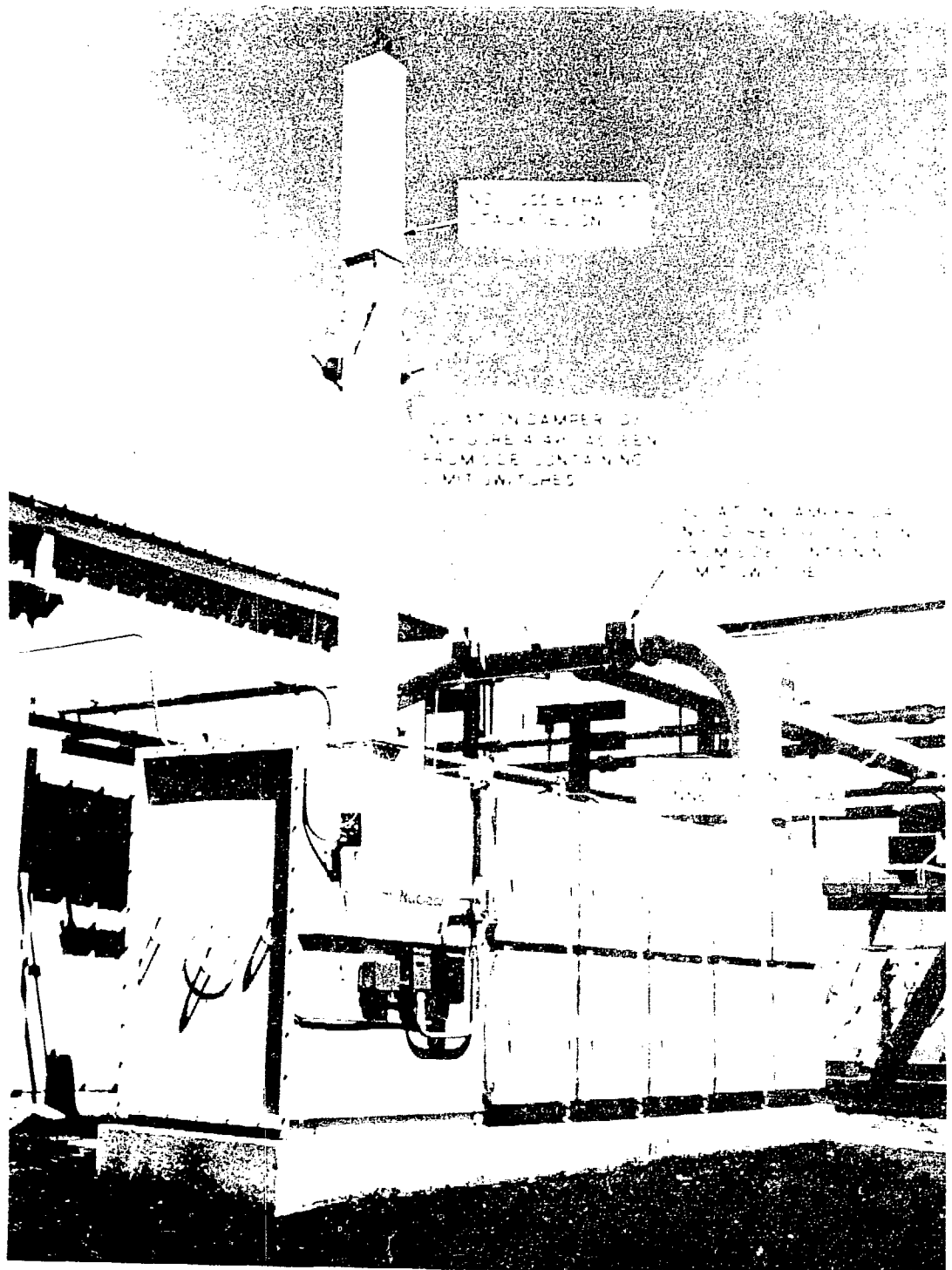


Figure 7 Type 1 Filter System (2,000 CFM) for Explosive Treatment System At CAMDS Showing Isolation Dampers And Interconnecting Network

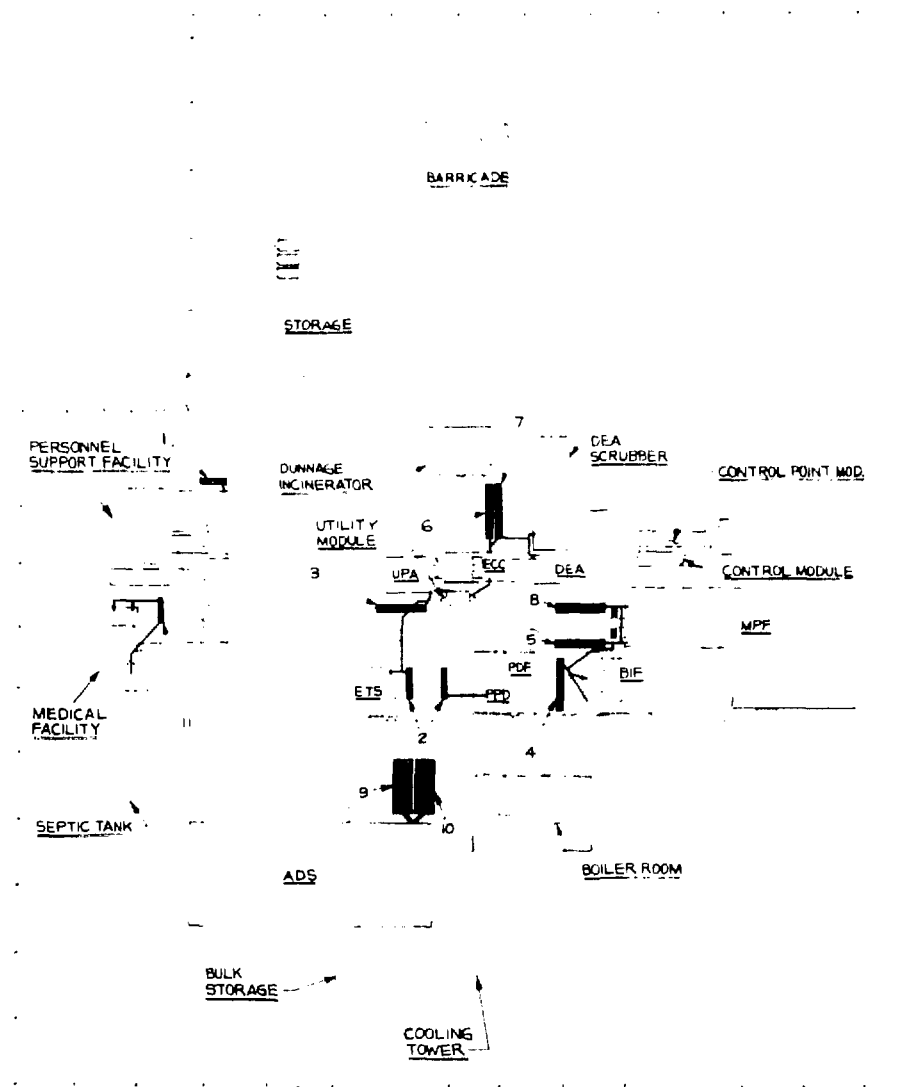


Figure 8 CAMDS Filter System Layout

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(5) No sprinkler system requirement for CAMDS.

As more stringent environmental and personnel safety requirements are continually being imposed, there may be a corresponding demand for high efficiency air cleaning systems in certain industrial situations. This handbook has been prepared primarily for those designing air-cleaning systems in the chemical demilitarization operations as the nuclear air cleaning handbook was prepared for those in the nuclear field. As Chemical Systems Laboratory was able to extrapolate extensive information from the nuclear air cleaning handbook, personnel working in various air cleaning areas of the chemical or industrial field may be able to obtain information from this handbook which will make their task easier and their end product more technically sound.

This handbook has been assigned a #ADA-056-389 by Defense Document Center and is currently being processed through normal channels. When processing is complete, this handbook will be available from the National Technical Information Service at 5285 Port Royal Road, Springfield, VA 22161, area code 703-557-4650.

DISCUSSION

DENNISON: Would you comment on your instrumentation regarding the efficiency of your untreated charcoal beds in dealing with heavy hydrocarbons or other hydrocarbons for that matter?

SHACTER: The instrumentation to be used for the CAMDS high efficiency air cleaning systems will consist of two bubbler systems and an M8 Alarm. One bubbler system will sample between the two adsorber banks and the other bubbler system will sample at the exhaust stack. The M8 Alarm will sample between the two adsorber banks with provisions to have the sample source switched to the exhaust stack should a breakthrough of the first adsorber bank occur. The chemical analysis of the bubbler solutions will achieve sensitivities of 3×10^{-5} mg/m³, 3×10^{-6} mg/m³, and 3×10^{-3} mg/m³ for the three toxic agents to be contained. The M8 Alarm is an electrochemical detector with an immediate response, but only a 102 mg/m³ sensitivity. The only suggestion I could offer would be to use a gas chromatograph in series with a detector selected for your specific requirement. An example of this is, the Chemical Systems Laboratory uses a Hydrogen Flame Emission Detector (HYFED) to detect phosphorous-bearing toxic agents.

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CPP EXPERIMENTAL HEPA FILTER MEDIA

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At the Waste Calcining Facility located at the INEL, we have some rather challenging problems to overcome in the various filtration systems. Some have already been mentioned in other papers by Schindler, Loo, and Murphy.

One of the problems we are currently trying to overcome is that of having a filter media withstand the high NO_x concentrations, the high relative humidity and radioactivity. One of the major problems is that the present filter media has the organic binder destroyed after several days in the system due to the high (10,000 to 15,000 ppm) of NO_x and high Moisture loading on the filters, forming nitric acid.

We have asked Flanders Filters to develop a different binder in the filter media which would be more NO_x and moisture resistant. We have since tested three of such filters in the off-gas system of the Atmospheric Protection System (A.P.S.). Our first observation indicated that we had also achieved a further reduction in the Decontamination Factor (DF) of approximately 30. Before the filters were changed efforts were made to intentionally breach the filters by lowering the superheater temperature from approximately 170° F to 125° F for approximately twenty five minutes. There was no increase in the ΔP noted. These tests were duplicated and varied within system constraints, with little effect. No increase was seen on the stack monitor. Although we cannot be certain that moisture actually formed on the filter during these tests, it was felt by all concerned that it should have. Previous filters have been breached by too much moisture forming on the filters. Upon removal from the system a brief visual examination showed no apparent damage to the filters, and no sagging of the media. These filters are of the super-flow design - not having separators. Samples of the used filter media were taken to compare with new media and were to be remotely tested for the tensile strength using an Instron machine. However, personnel operating the Instron testing felt that the "hot" filter sample was too fragile. It had been an additional 6-7 weeks since removal and it was felt that further deterioration had likely taken place.

We are planning on experimenting with some additional filters in the A.P.S. off-gas system as well as the primary Waste Calcining Facility (WCF) off-gas filter systems.

One question arises that we have been unable to answer satisfactorily is that of the type and size of particles passing through or around the two HEPA filters in series at the WCF and collecting on the HEPA filters at the A.P.S. off-gas system. Are these particles formed by recondensing downstream of the WCF HEPA system or is it a wicking effect or what have you?

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DISCUSSION

FIRST: At the meeting that was held in Aix-en-Provence a year and a half ago we had a presentation from Argonne showing migration of plutonium through many filters in series.

COWAN: Digester offgas from acid digestion of combustible waste also caused failed filters due to condensate. The temperature was increased to 250-300°C so that the gas was dry and no problems are now found. Also, another filter on an HNO₃ concentrator is operated at 125-135°C without a problem. Increasing the temperature may solve your problem.

HANSON: On filter change, we noticed that we had liquid in the third set of filters downstream and it was at 170°. They tried to maintain the temperature in the 170-180°F range.

CALAN: We are operating in the 250°C range. We have another set of filters off the top of our fractionator where we're filtering nitric acid off the concentrator. We run these around 125-130°C and we are not experiencing difficulty there, either. But, if you look at the vapor pressure of nitric acid, a fairly small concentration of NO₂ can result in a rather high concentration of nitric acid forming in the filter itself. In the 40% range it condenses around 125°C.

BURCHSTED: Your problem suggests that the NO_x passed through the double bank of filters as a gas, then combined with moisture which condensed in those filters or in the duct downstream of them. These droplets then were trapped in the third stage filter. Condensation between filters in the first two stages did not occur because they were too close together; that is, they may have acted as a single filter in this respect.

HANSON: They are in the same housing and they are essentially one filter.

BURCHSTED: The first two?

HANSON: Yes.

BURCHSTED: There's apparently some condensation taking place between your double bank and your final bank down below.

HANSON: There could be. The gases go through superheaters but maybe this isn't sufficient.

FIRST: Our time for the Open-End has come to a conclusion. I think it was a very interesting session in terms of the variety of topics that we covered and the speakers were very kind and held their discussions to a very short time which was the intent. So, to all you gentlemen and to those who commented many thanks.

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SESSION XIV

FILTRATION

Thursday, August 10, 1978

CHAIRMEN: R. Dorman, H. Gilbert

LOADING CAPACITY OF VARIOUS FILTERS FOR SODIUM OXIDE/HYDROXIDE AEROSOLS

J. D. McCormack, R. K. Hilliard,
J. R. Barreca

PERFORMANCE OF A WET CELL WASHER FOR SODIUM FIRE AEROSOLS

W. C. Hinds, J. M. Price, M. W. First

ENHANCED FILTRATION PROGRAM AT LLL--A PROGRESS REPORT

W. Bergman, R. D. Taylor, H. H. Miller,
A. H. Biermann, H. D. Hebard, R. A.
daRoza, B. Y. Lum

STUDIES ON PROLONGING HEPA FILTER SERVICE IN CHEMICAL APPLICATIONS

R. W. Woodard, K. Terada, O. I.
Buttedahl

REPORT OF MINUTES OF GOVERNMENT-INDUSTRY MEETING ON FILTERS, MEDIA, AND MEDIA TESTING

W. L. Anderson

Appendix A A STUDY OF DIOCTYL PHTHALATE PARTICLES (DOP) GENERATED IN PENETROMETERS AND THE DEVICES USED CURRENTLY TO MEASURE THEIR SIZE

C. D. Skaats

Appendix B SIZE DISTRIBUTION OF "HOT DOP" AEROSOL PRODUCED BY ATI Q-127 AEROSOL GENERATOR

W. Hinds, M. First, D. Gibson, D. Leith

Appendix C FIBER DIAMETER DETERMINATION OF FIBER GLASS SAMPLES

S. Gross, C. Cain

EVALUATION OF DATA FROM HEPA FILTER QUALITY ASSURANCE TESTING STATIONS

J. T. Collins, R. R. Bellamy, J. R.
Allen

PERFORMANCE AND ENVIRONMENTAL CHARACTERISTICS OF A COMPACT, HIGH-CAPACITY HEPA FILTER DESIGN

C. E. Rose, R. D. Rivers

PERFORMANCE OF 1,000 AND 1800 CFM HEPA FILTERS ON LONG EXPOSURE TO LOW ATMOSPHERIC DUST LOADINGS

M. W. First, D. Leith

DECONTAMINATION OF HEPA FILTERS

J. W. Koenst, Jr., E. L. Lewis,
D. F. Luthy

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LOADING CAPACITY OF VARIOUS FILTERS FOR* SODIUM OXIDE/HYDROXIDE AEROSOLS

J. D. McCormack, R. K. Hilliard
J. R. Barreca

Hanford Engineering Development Laboratory
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Abstract

The loading capacity of commercially available prefilters and HEPA filters is reported for sodium oxide and sodium hydroxide fumes in moist and normal air. The test aerosol is characterized and is considered to be typical of that which would exist during accidents involving sodium fires in breeder reactors, plants, and laboratory facilities.

Sodium pool fires of 0.6 m² area in a closed 330 m³ room serve as an aerosol source to supply the recirculating filter test loop. The room and filter inlet stream is sampled to determine airborne particle size, mass concentrations, and dew point. Inlet mass concentrations of the test aerosol ranged up to 16 g/m³. The chemical forms were mixtures of Na₂O₂, NaOH, Na₂CO₃, and their hydrates.

Seven types of HEPA filters and seven types of prefilters were examined in six test series. The holding capacity of the filters ranged from 0.6 to 3.9 kg at 5 kPa pressure drop for aerosols generated in moist and normal air. The early time and moist aerosols showed the lowest loading capacities. Up to 5.1 kg of sodium carbonate aerosols were held. Flow and media efficiency had only a minor effect on the loadings achieved. No combination of prefilters tested was effective in providing increased holding capacity for the associated HEPA filter.

Introduction

Emergency air cleaning is an accepted means of accident mitigation for light water reactors⁽¹⁾. Similar air cleaning techniques for liquid metal cooled breeder reactors are still being developed and tested. A report to this conference two years ago reviewed the benefits of an Emergency Air Cleaning System (EACS) for breeder reactors and surveyed the probable usefulness of twenty-four air cleaning systems⁽²⁾. Such an air cleaning system would, of necessity, have to handle large quantities of sodium compounds, primarily the oxide and hydroxides, at high airborne concentrations and possibly under moist atmosphere conditions. Filter systems using commercial HEPA filters were well rated in the referenced evaluations because of the high degree of development, use, and reliability. These filters are known to be very efficient. However, information about loading capacity for the expected sodium compound aerosols is limited and in some cases conflicting results were reported⁽³⁻⁵⁾. Many of the early tests were

*Work performed under USDOE Contract EY-76-C-14-2170.

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made with very small sodium sources in dry atmospheres or at reduced oxygen contents. Today's understanding of the hypothetical core melt-down accident give airborne concentrations of sodium oxide within containment up to 50 g/m^3 . Further, water release from heated structural concrete is possible, allowing for the formation of NaOH and its hydrates. Also, if limestone aggregate concretes are used, the release of CO_2 gas with subsequent formation of sodium carbonate is possible. The objective of this work was to measure the loading capacity of several types of commercial HEPA filters and prefilters using sodium aerosols typical of those expected from breeder reactor core melt-through types of accidents.

Experimental Arrangements

Test Aerosol Generation and Sampling

The basic test facility consisted of a large closed concrete room (330 m^3 volume) containing a 0.6 m^2 heated pan for burning sodium.

The test aerosol generated in this smoke room was drawn to the filter through 13 meters of 25-cm diameter duct. The filtered air was returned to the smoke room by a blower capable of maintaining filter flow of $0.47 \text{ m}^3/\text{s}$ ($1000 \text{ ft}^3/\text{min}$) at 6 kPa (24 inches H_2O) pressure drop. The room atmosphere could be maintained at a slightly negative pressure to the outside by a $0.1 \text{ m}^3/\text{s}$ room exhaust to a ventilation system. Natural in-leakage of air maintained a satisfactory oxygen concentration for combustion. Steam and CO_2 gas could be fed to the room at controlled rates. Figure 1 is a schematic of the test arrangement. Figure 2 shows the test filter housing. The housing can accommodate $60 \times 60 \times 60 \text{ cm}$ HEPA filters and 60×60 prefilters up to 45 cm deep.

Samples of the test aerosol and atmosphere were taken in the smoke room through an airlock arrangement which allowed the sampling filter or cascade impactors to be inserted directly into the room, obviating the need for any inlet corrections. The mass concentration was determined using weighed filters. Sodium concentration was determined by titration of the filters with hydrochloric acid. Lower levels of sodium were determined on cascade impactors and a downstream sampler by flame emission spectrometry. Oxygen, CO_2 and water were measured by on-line instruments supplemented with grab samples analyzed by mass spectrometry and gravimetric means.

Cascade impactors were used for particle size measurement and electron microscope grids were exposed for particle shape information. Air flow, temperature, and pressure drop at the test filter were recorded by a 100-channel data logger on paper tape and magnetic tape for subsequent plotting and calculation.

Test Procedure

The general test procedure was to establish the initial room conditions and preheat the burn pan to 315°C . A preweighed test filter was installed in the housing, and sodium preheated to 200°C was delivered into the burn pan. Normally 50 kg of sodium was used. The room aerosol concentration was sampled and, when it was judged adequate for the test (about 3 g/m^3), loop flow was started. Flow was kept constant until the desired terminal pressure drop at the test

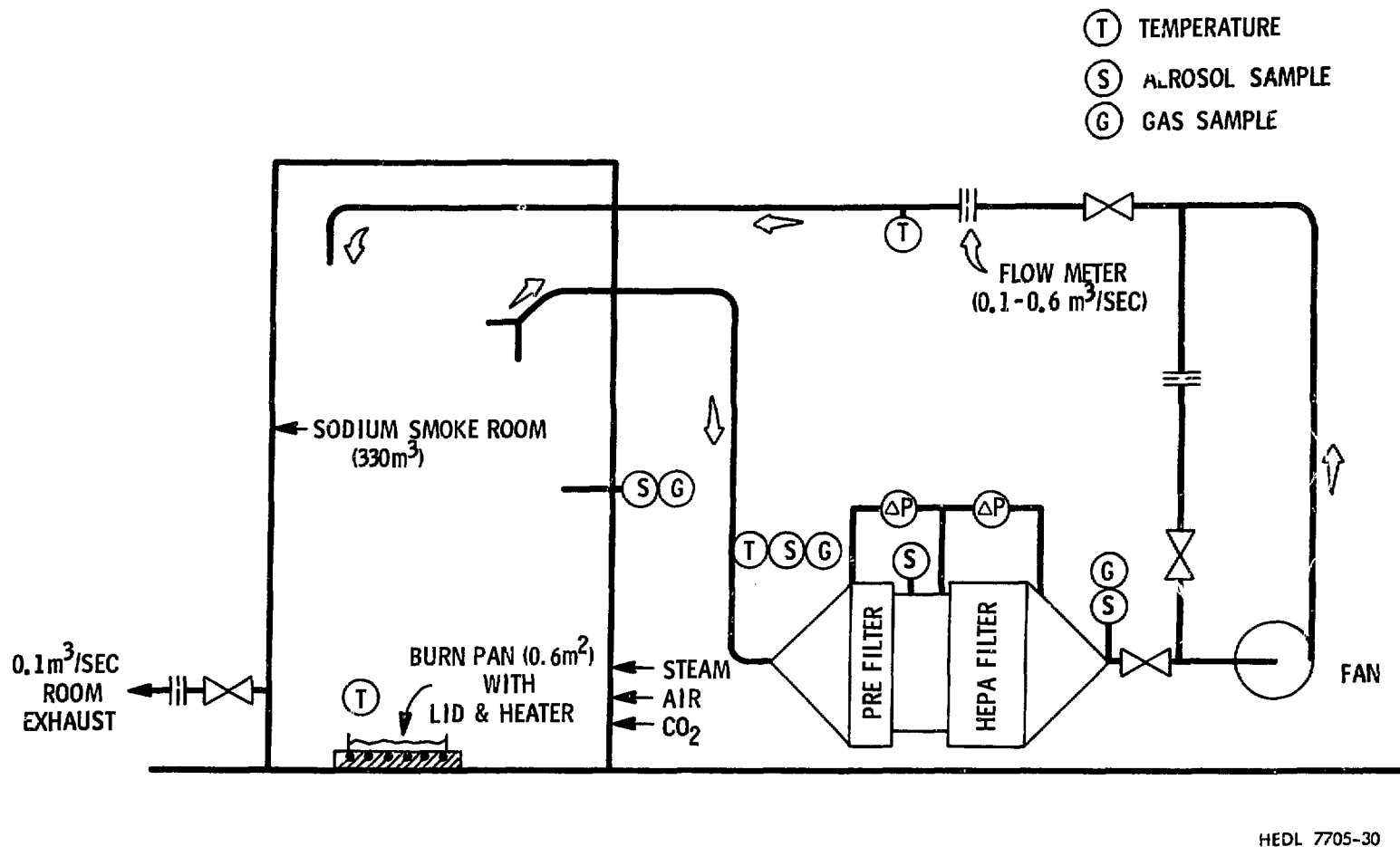


Figure 1. Filter loading loop schematic.

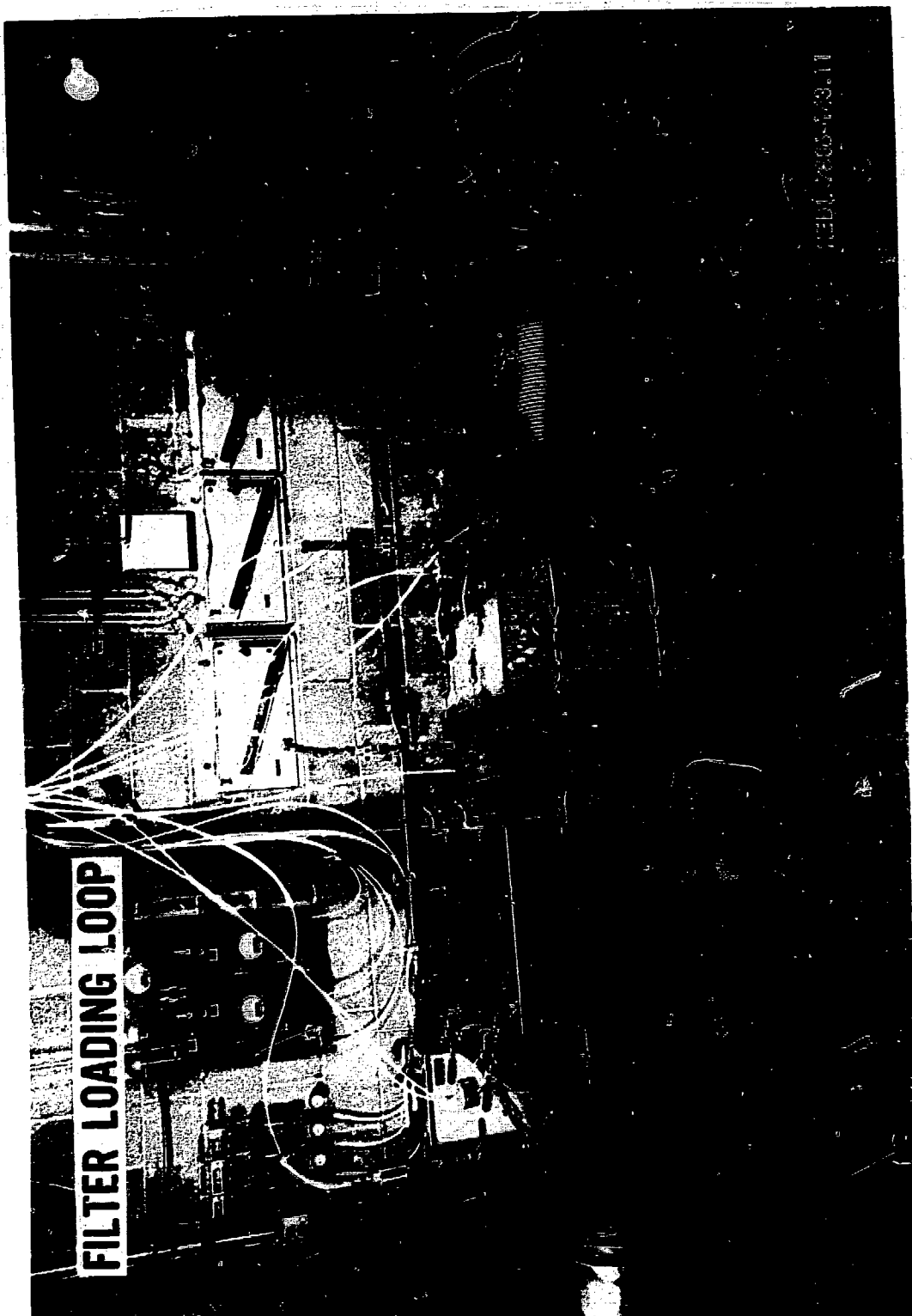


Figure 2. Filter loading loop.

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filter was reached. The flow was stopped; the filter rapidly removed, weighed, and a new filter inserted. Up to seven filters could be tested during one test series. For some test series, additional sodium was added to the burn pan to maintain a high aerosol concentration.

In two test series, the loop flow was started before the sodium fire to measure loading of fresh, lower concentration aerosols. This sequence duplicated the conditions expected from filters used in a ventilation system in service at the time the sodium fire starts.

Test Filters

A variety of test filters were obtained from commercial sources. These filters were selected to be typical of various manufacturing variables thought to be of interest, such as the media efficiency, media area, size of separators or absence of separators. All the test filters had a glass media suitable for high humidity use and all the filters were 60 x 60 x 30 cm size. The relevant details are summarized in Table I, where each filter type is identified with a key number which will be used in subsequent discussions for filter identification.

Test Prefilters

Several types of prefilters were also used. These prefilters had a 60 x 60 cm face area with various thicknesses. For the thinner 5-cm thick filters, two were used in a series. The filter characteristics and the key numbers are given in Table II. The metallic media filters were washed, dried, and reused. The others were discarded after use.

Filter Loading Test Variables and Sequence

Six filter loading test series were completed. Each test series consisted of a single sodium fire in a selected atmosphere condition. The sodium oxide/hydroxide aerosols generated were used to load up to seven test filters in sequence.

Primary variables examined in these runs were:

- Filter flow rate; 20%, 50%, and 100% of nominal,
- Generation of aerosol in normal and moist atmospheres,
- Filter construction, absence of separators,
- Filter media efficiency; 60, 90, and 99.97%,
- Prefilter construction, 7 types,
- CO₂ addition to the test atmosphere.

The initial test conditions, atmosphere, and primary test variables are given in Table III.

Experimental Results

The test results of most interest are the mass loading of the filters and final pressure drop. These results are summarized in Table IV for all the runs where 37 filter combinations were measured.

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Table I. Description of test filters.

| Test filter
key number | Vendor | Media
efficiency,
% | Estimated
media
area, m ² | Characteristics |
|---------------------------|---------|---------------------------|--|--|
| 1 | F | 99.97 | 23 | Waterproof glass media, Al separator. |
| 2 | F | 99.97 | 23 | Same as 1, w/o separators. |
| 3 | Various | 99.97 | Various | HPS type C; water resistant glass media, non-metallic separator, 90% RH, 49°C. |
| 4 | M | 99.97 | 30 | Glass media, mini Al separator, 1500 CFM. |
| 5 | F | 90 ^(a) | 17 | Glass media, no separator, 100% RH, 121°C. |
| 6 | C | 99.97 | 23 | Waterproof glass media, Al separator. |
| 7 | C | 60 ^(a) | 5.1 | Glass media, Al separator. |

(a) Per ASHRAE 53-68.

Table II. Description of test prefilters.

| Test filter
designator | Source | Thickness ^(a)
cm | Eff. % | Characteristics |
|---------------------------|--------|--------------------------------|-------------------|--|
| 20 | M | 13 | - | Knitted ss/fiber glass demister. |
| 21 | R | 10 | - | Crimped 14 and 18 mesh metal screen. |
| 22 | A | 5 | - | Progressive density; expanded metal and mesh layers. |
| 23 | A | 5 | - | Crimped metal screen, pyramid pocket design. |
| 24 | R | 10 | 30 ^(b) | Cotton fabric, nonwoven. |
| 25 | R | 30 | 50 ^(b) | Glass fiber mat, folded. |
| 26 | A | 5 | - | Crimped, expanded Al mesh, 3 layers. |

(a) All were 60 x 60 cm face.

(b) Atmosphere dust spot.

Table III. Filter loading test conditions

| Test No. | Initial Room Conditions | | | | Steam Addition
kg/hr | Final Room Conditions | | | Primary Test Variable |
|----------|-------------------------|--------------|-----------|----------------------|-------------------------|-----------------------|--------------|-----------|---------------------------------------|
| | Temp
°C | Dew Pt
°C | R.H.
% | CO ₂
% | | Temp
°C | Dew Pt
°C | R.H.
% | |
| FL 1 | 14.4 | -2.2 | 30 | Normal | None | 38.0 | -12.6 | ~3 | Flow Rate, filter type ⁽¹⁾ |
| FL 2 | 10.6 | 3.3 | ~60 | " | 4.6 | 33.0 | -12.0 | 4 | Same as FL 1 with steam |
| FL 3 | 16.6 | -1.7 | 26 | " | None | 38.0 | -13.0 | 3 | Filter construction |
| FL 4 | 21.1 | 6.7 | 39 | " | 9.1 | 43.0 | 8.0 | 12 | Prefilter types, with steam |
| FL 5 | 20.0 | 0.6 | 28 | " | None | 46.0 | -13.0 | 2 | Prefilter, dry |
| FL 6 | 25.0 | 8.9 | 36 | ~3 | 9.1 | 46.0 | 21.0 | 25 | CO ₂ addition with steam |

(1) with and without separators.

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Table IV. Summary of filter loading and pressure drop.

| Test No. | Filter F.F. | Type HEPA | Flow m ³ /sec | Mass on filter, kg | | P, kPa | | Aerosol Conc., g/m ³ |
|----------|-------------|-----------|--------------------------|--------------------|------------|--------|--------|---------------------------------|
| | | | | P.F. | HEPA Total | P.F. | Total | |
| FL1 A | None | 1 | 0.45 | None | 2.8 | None | 4.8 | 9.0 |
| | None | 1 | 0.23 | None | 2.6 | None | 4.6 | 7.2 |
| | None | 2 | 0.23 | None | 3.7 | None | 4.8 | 9.2 |
| | None | 2 | 0.45 | None | 3.0 | None | 4.8 | 9.4 |
| | None | 2 | 0.09 | None | 2.5 | None | 4.6 | 7.7 |
| | None | 1 | 0.09 | None | 2.7 | None | 4.7 | 6.5 |
| FL2 A | None | 1 | 0.47 | None | 0.63 | None | 5.0 | 10.7 |
| | None | 1 | 0.24 | None | 0.79 | None | 5.0 | 12.4 |
| | None | 2 | 0.24 | None | 2.5 | None | 4.5 | 16.5 |
| | None | 2 | 0.46 | None | 3.9 | None | 4.5 | 8.4 |
| | None | 2 | 0.09 | None | 1.8 | None | 4.5(1) | 10.4 |
| | None | 1 | 0.09 | None | 1.8 | None | 4.5 | 4.6 |
| FL3 A* | None | 3 | 0.45 | None | 0.62 | None | 4.5 | 1.8 |
| | None | 3 | 0.46 | None | 1.1 | None | 4.5 | 5.9 |
| | None | 4 | 0.46 | None | 2.0 | None | 4.6 | 5.0 |
| | None | 5 | 0.46 | None | 2.2 | None | 4.6 | 6.4 |
| | None | 6 | 0.46 | None | 1.7 | None | 4.5 | 4.9 |
| | None | 7 | 0.46 | None | 1.7 | None | 3.7 | 2.8 |
| FL4 A* | None | 3 | 0.46 | None | 2.0 | None | 4.6 | 6.7 |
| | 20 | 3 | 0.46 | 0.89 | 1.2 | 0.2 | 4.9 | 3.5 |
| | 21 | 3 | 0.47 | 1.1 | 2.2 | 0.05 | 4.7 | 6.9 |
| | 22(2) | 3 | 0.48 | 0.58 | 2.0 | 0.02 | 5.0 | 6.1 |
| | 23(2) | 3 | 0.48 | 0.51 | 1.5 | 0.02 | 4.8(3) | 7.0 |
| | 24(2) | 3 | 0.48 | 0.79 | 1.9 | 1.6 | 5.0 | 8.2 |
| FL5 A | 25 | 3 | 0.48 | 1.6 | 0.52 | 3.5 | 3.9 | 4.4 |
| | 20 | 3 | 0.47 | 0.51 | 0.44 | NM | % | 5.7 |
| | 21 | 3 | 0.47 | 0.31 | 0.36 | 0.02 | 6.0 | 4.0 |
| | 22(2) | 3 | 0.47 | 0.15 | 0.53 | 0.02 | 6.5 | 4.3 |
| | 23(2) | 3 | 0.47 | 0.89 | 0.81 | 5.0 | 6.2 | 4.2 |
| | 26(2) | 3 | 0.47 | 1.5 | 0.73 | 5.1 | 6.1(4) | 6.4 |
| FL6 A | 24 | 3 | 0.47 | 2.0 | 0.64 | 3.5 | 4.4 | 4.4 |
| | 25 | 3 | 0.47 | 3.6 | 0.21 | 5.3 | 5.8 | 6.1 |
| | - | 3 | 0.47 | - | 4.7 | - | 5.0 | 6.3 |
| | - | 2 | 0.47 | - | 5.1 | - | 4.9 | 4.9 |
| | 21 | 3 | 0.47 | 3.3 | 1.5 | 0.8 | 5.8 | 5.4 |
| | 25 | 3 | 0.47 | 1.4 | 0.17 | 5.1 | 5.6 | 7.6 |
| FL7 A | - | 3 | 0.47 | - | 4.0 | - | 5.0 | 4.8 |

* Flow started before sodium fire
 (1) Ruptured at 2.5 kPa P flow continued
 (2) Two filters used in series
 (3) Ruptured at 2.2 kPa
 (4) Ruptured at 4.5 kPa

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Table IV also lists the average aerosol mass concentration at the filter inlet, as calculated from the mass on the filter and the actual volume of air passing the filter.

Pressure Drop and Loading Capacity

For a pressure drop of about 5 kPa (20 inches H₂O), the HEPA mass loading ranged from a low of 0.6 kg to 3.9 kg for sodium aerosols generated in air. A maximum loading capacity of 5.1 kg was found for aerosols generated in a moist atmosphere containing CO₂ (test FL-6). In filter test series FL-4 and FL-5, prefilters were added to the system. The maximum combined loading was 3.8 kg. None of the prefilters tested were particularly effective in increasing the loading capacity of the following HEPA filter. In all cases the loadings and pressure drops were not evenly shared. The space used by the prefilter and HEPA filter in series would have been more effective for mass loading if two filters, with no prefilter, were arranged in parallel.

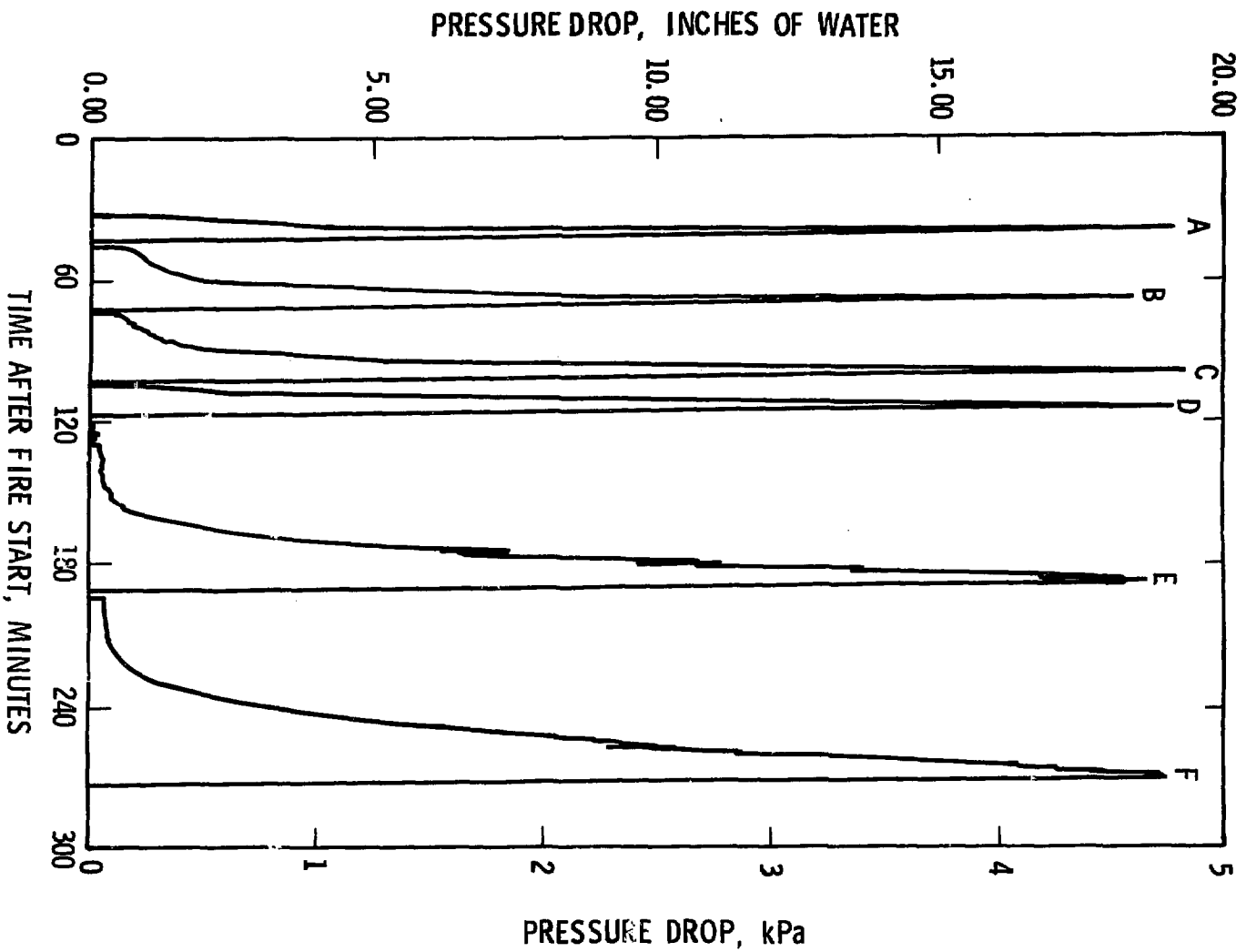
Pressure drop typically started low and then increased at an increasing rate until the filter ruptured or the test was terminated. This behavior is shown in Figures 3-7 where the pressure drops for all runs are given. Notice that filter 2-F ruptured at about 3 kPa. Figures 8 and 9 show typical filters after loading. Run FL-1 was in normal air. Steam was added during FL-2. The effect of moisture on the appearance of the filtered aerosol is especially noticeable in test series 2 where filter 2A and 2B show no visible deposit but were plugged by only 0.6 and 0.8 kg of material. As the test progressed, more deposit was visible and the later filters held from 1.8 to 3.9 kg. Although steam was added to the smoke room during test FL-2 at 4.6 kg/hr, the dew point, relative humidity, and mass/sodium ratio decreased with time. As the water content of the aerosol decreased, the filterability generally improved. Figure 10 is a plot of the mass to sodium ratio measured near the burn pan. This decreasing mass ratio behavior is typical of all the runs. The lower water content of the later aerosol samples is in accord with the reduced relative humidity of the smoke room atmosphere due to reduced dew point and increased temperatures.

Effect of Flow Rate and Filter Construction

The effect of flow rate on filter loading is shown by comparison of tests FL-1 and FL-2 for both dry and "wet" conditions. The effect of using HEPA filters without separators is also shown. These loadings are summarized in Table V.

From Table V it is apparent that flow has only a small effect on loading capacity and that except at 0.09 m³/sec (200 cfm), filters without separators had higher loading capacities. Moisture effects are also seen in Table V where FL-2, the wet run, results in generally lower filter loadings.

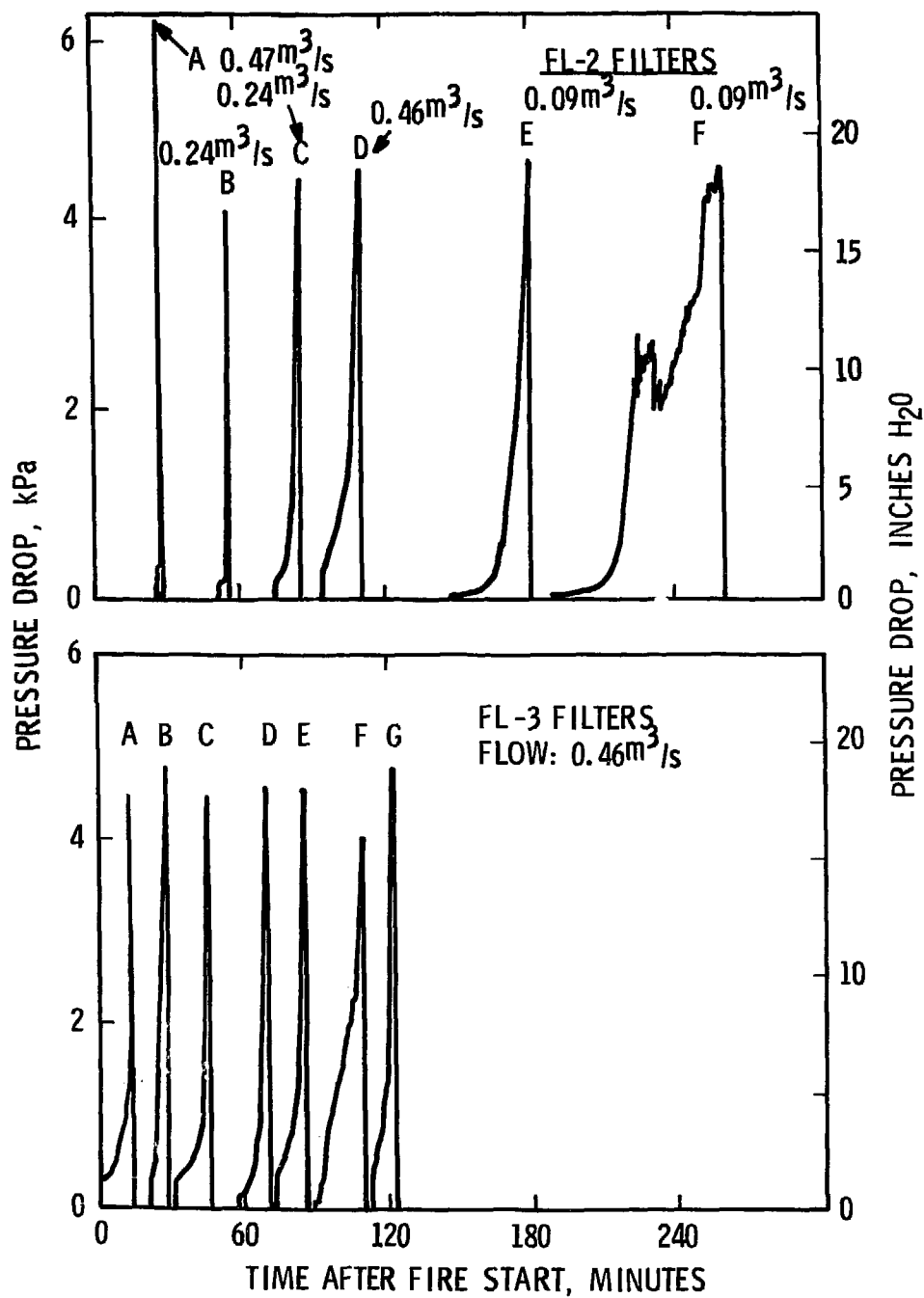
The effect of variations of filter construction and media efficiency are shown by the mass loadings of FL-3. This run also shows variations in mass loading caused by aerosol changes. This is seen by comparing the results of FL-3A, B, and G in Table IV. Identical



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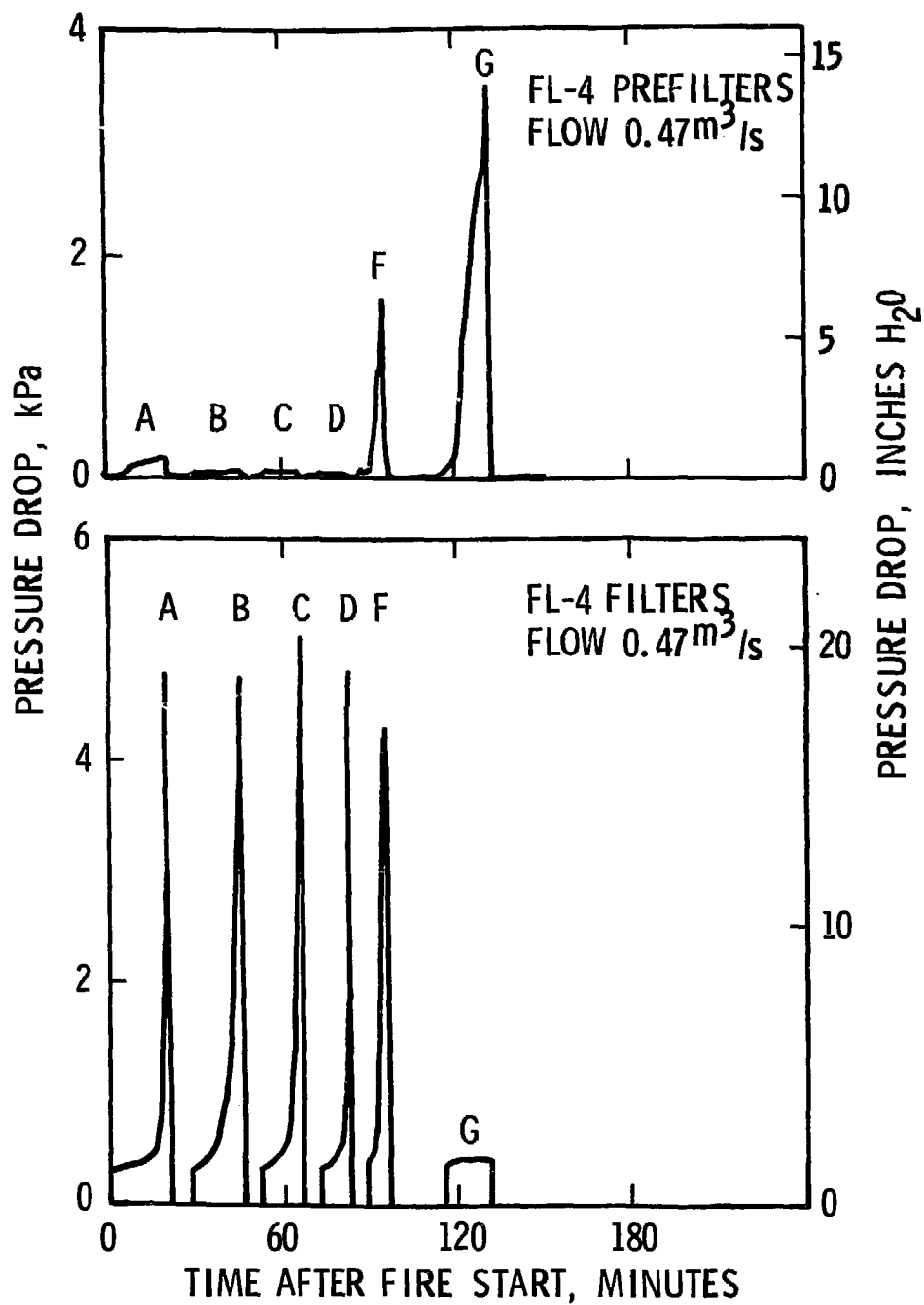
Figure 3. Pressure drop across filters, run FL-1.

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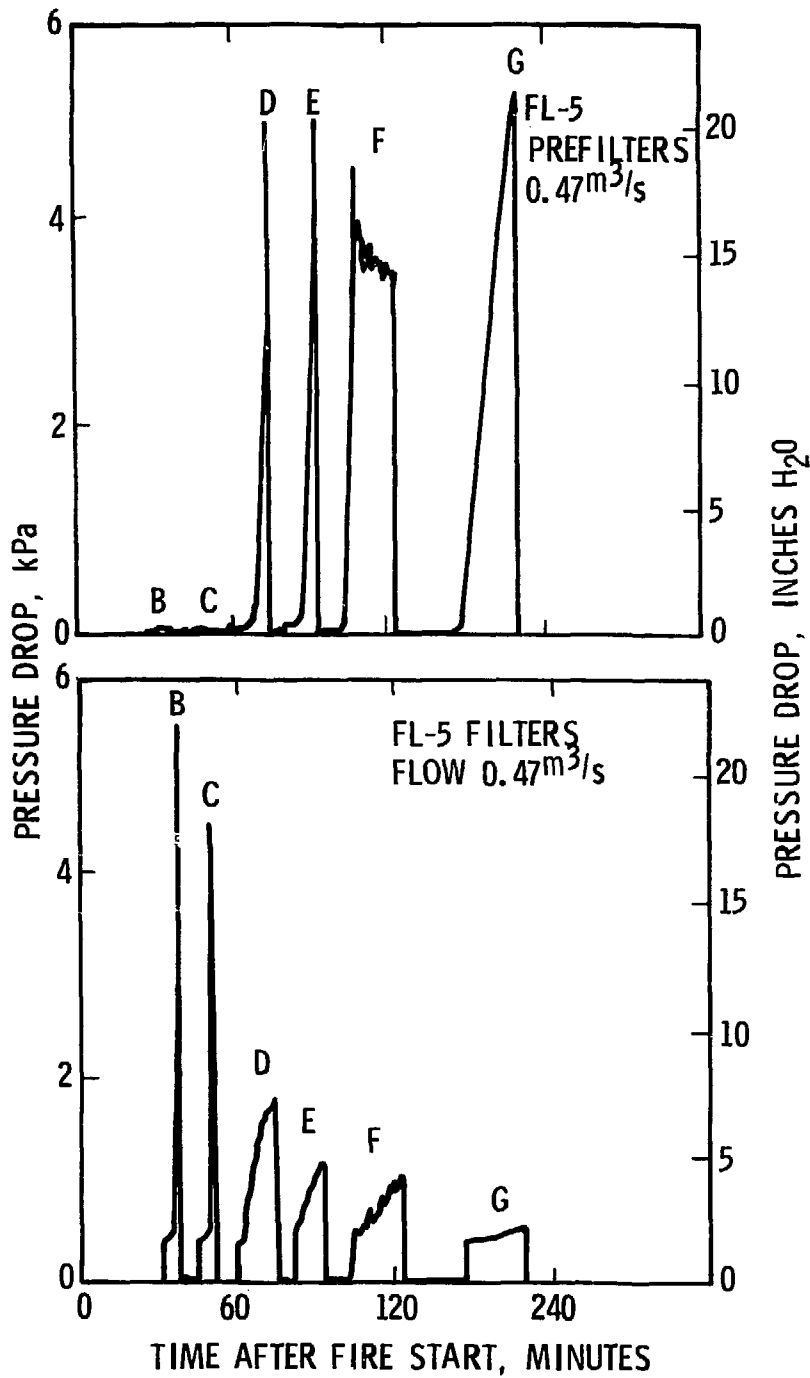
HEDL 7806-143.1

Figure 4. Pressure drop, runs FL-2 and FL-3.



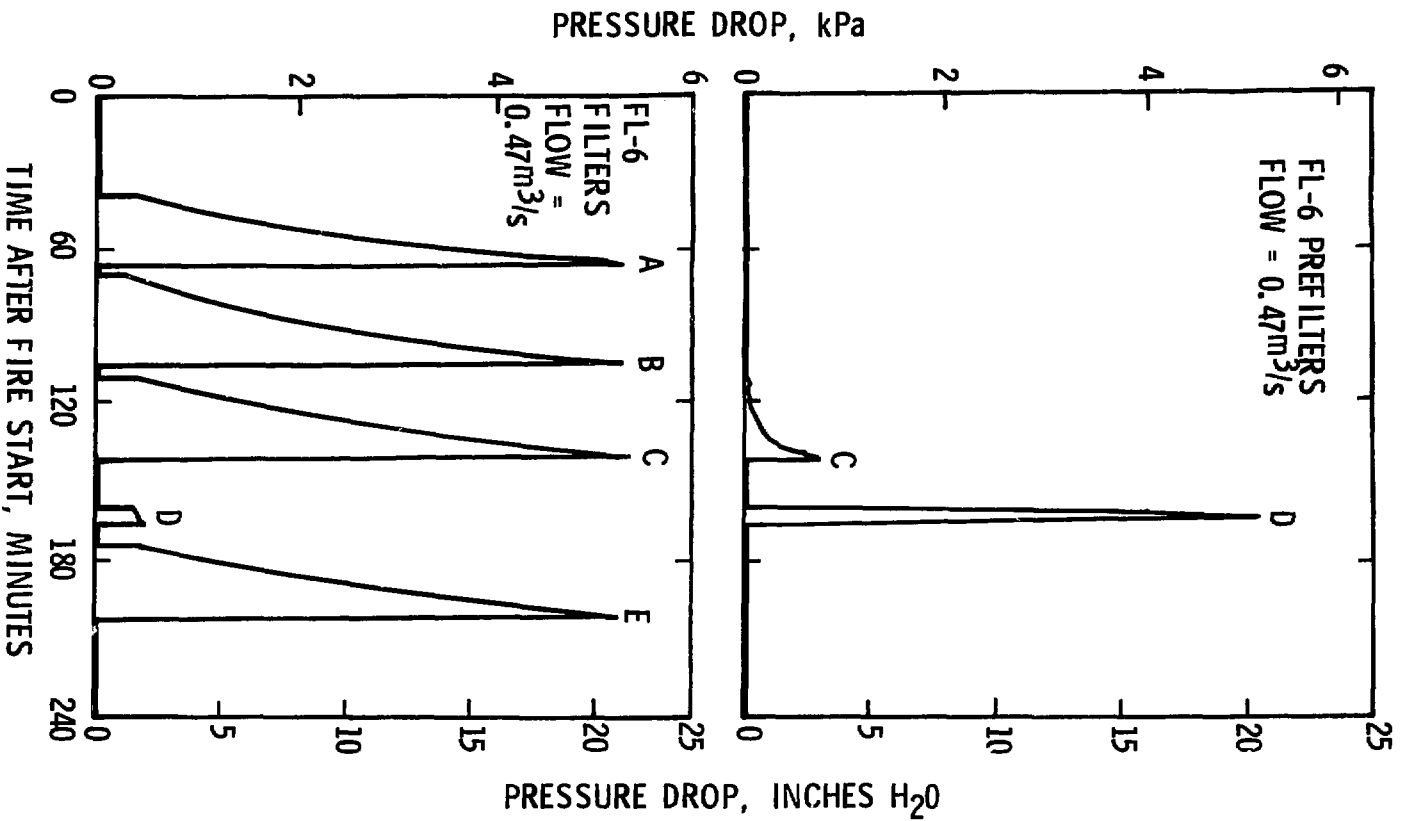
HEDL 7806-143.2

Figure 5. Pressure drop, run FL-4.



HEDL 7806-143.3

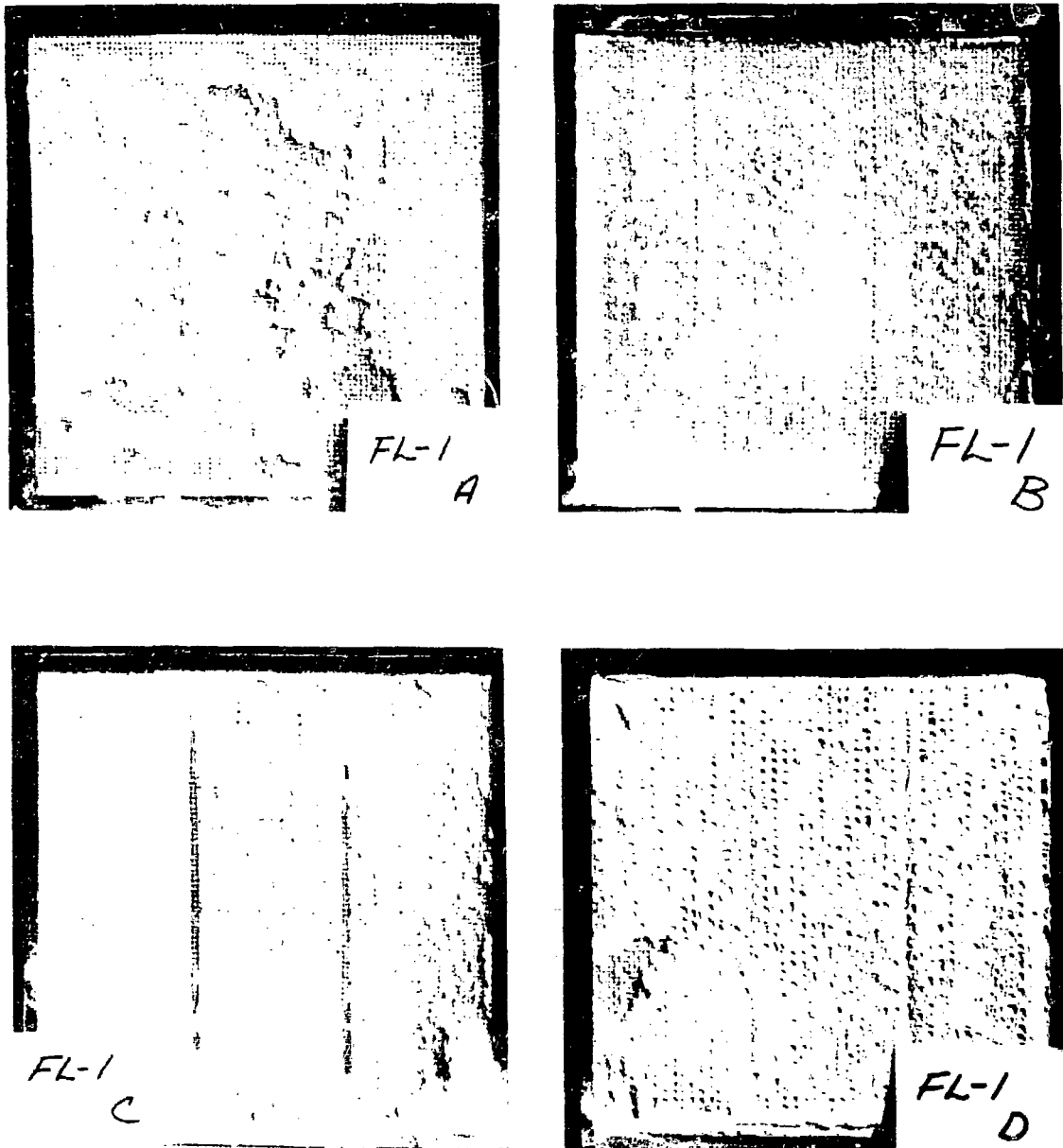
Figure 6. Pressure drop, run FL-5.



HEDL 7806-143.4

Figure 7. Pressure drop, run FL-6.

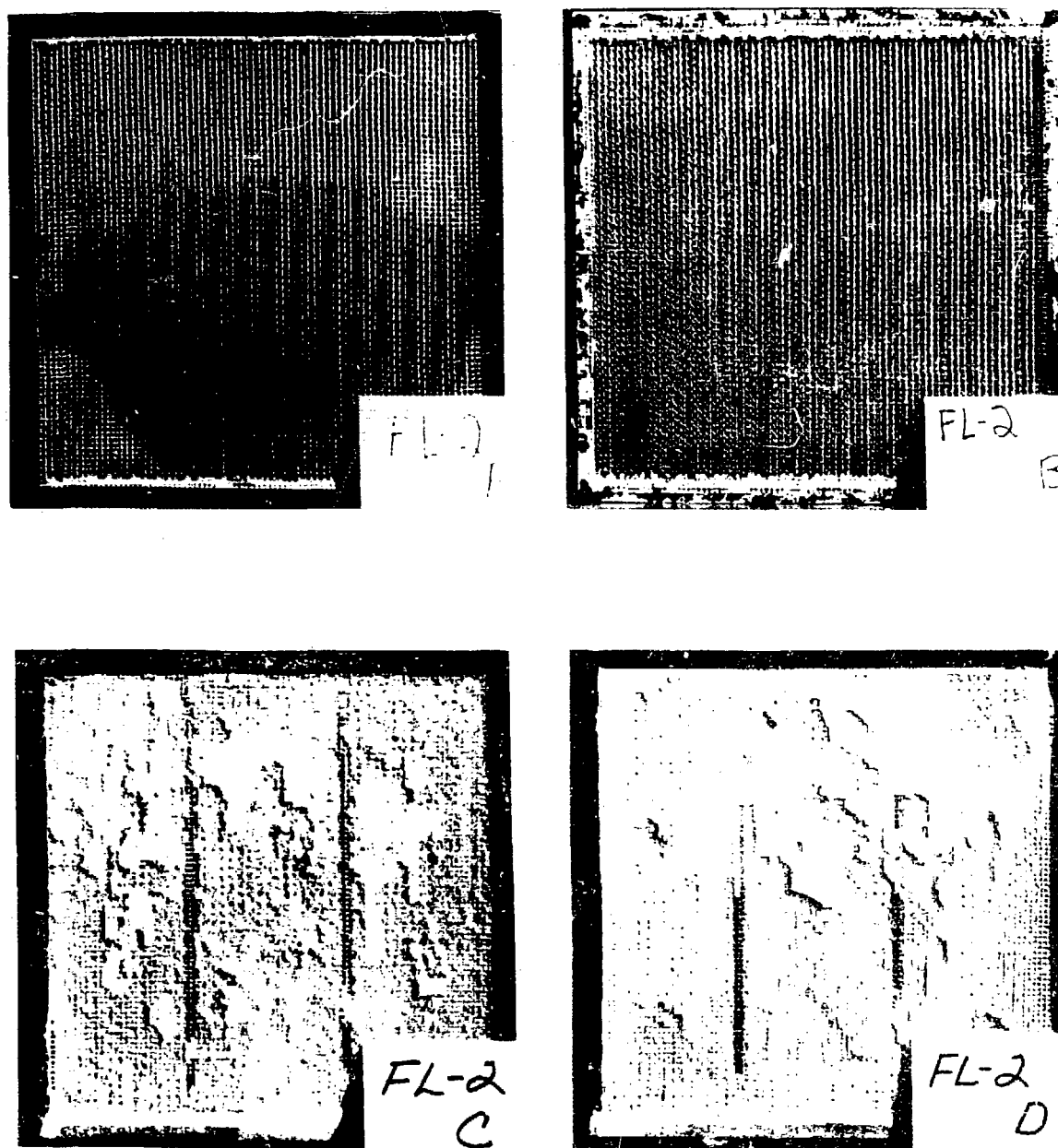
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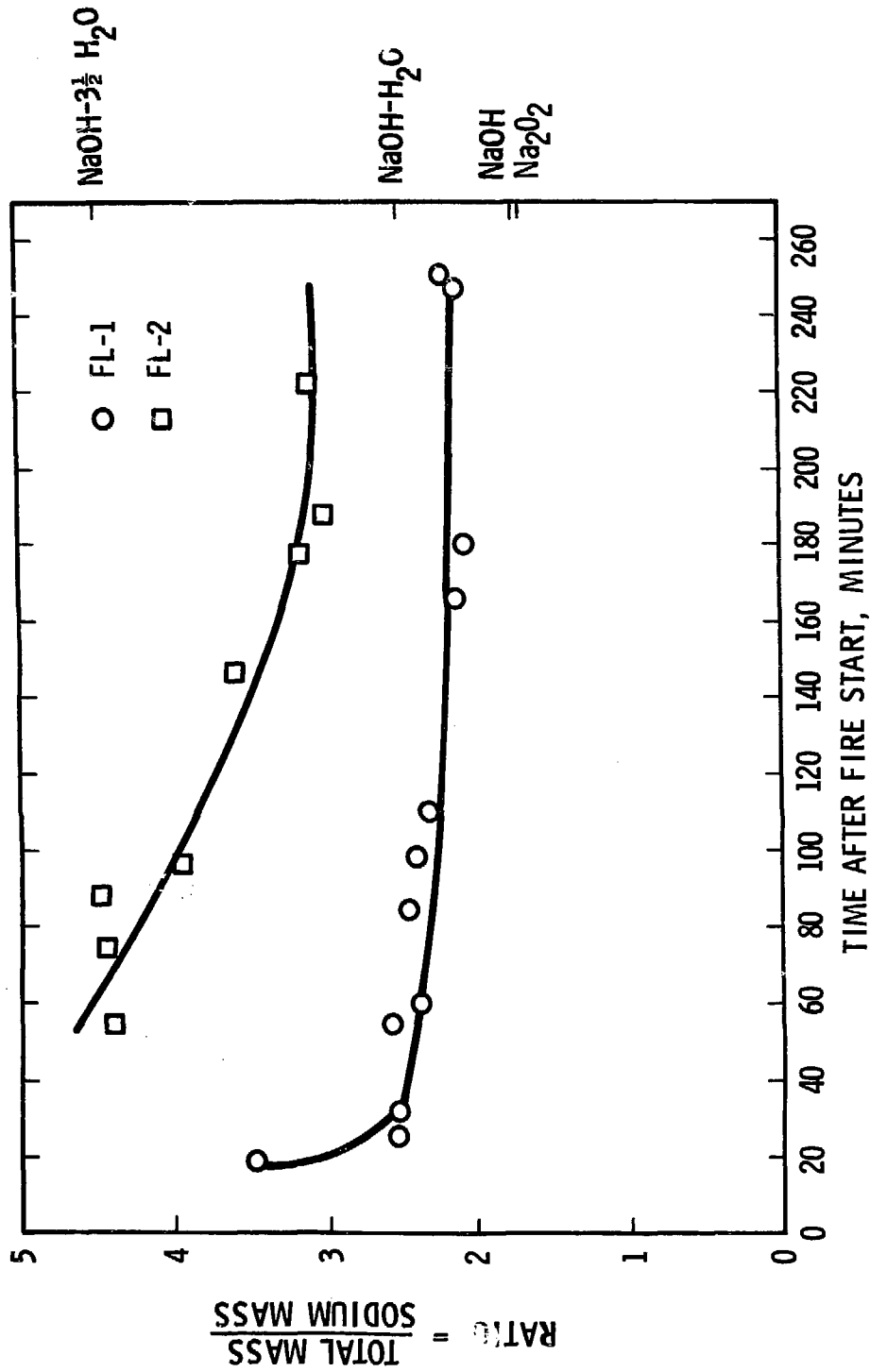
Figure 6. Filters after loading with dry aerosol.

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Figure 9. Filters after loading with moist aerosols.



HEDL 7806-143.6

Figure 10. Smoke room aerosol mass ratios for runs FL-1 and FL-2.

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Table V. Effect of flow and separators on filter loading.

| | | Mass loadings on filters, kg | | | | | |
|-----------------|-------------|------------------------------|------|------|-------------------------|------|------|
| Run | Atmosphere | w/separator | | | w/o separator | | |
| | | flow, m ³ /s | | | flow, m ³ /s | | |
| | | 0.09 | 0.24 | 0.47 | 0.09 | 0.24 | 0.47 |
| Filter Sequence | | F | B | A | E | C | D |
| FL 1 | Normal | 2.7 | 2.5 | 2.3 | 2.5 | 3.7 | 3.0 |
| FL 2 | Steam Added | 1.8 | 0.79 | 0.63 | 1.8 | 2.5 | 3.9 |

filters were used and loadings of 0.62, 1.1, and 2.0 kg were found. In test 3, the flow through the filter was started before the fire for A, and at 22 minutes and 113 minutes for B and G, respectively. The lowest loading was found for filtering at early stages of the fires. This is thought to be due to the high moisture content of the aerosol relative to the sodium.

Variations in loading for the other filters, where 60 and 90% efficiency filters and also 99.97% filters with "mini" aluminum separators were used are small, ranging from 1.7 kg to 2.2 kg. The highest loading was observed for the 90% (Key 5) filter. Again absence of separators appeared to help increase the loading capacity. Filter 3F ruptured at 2.2 kPa.

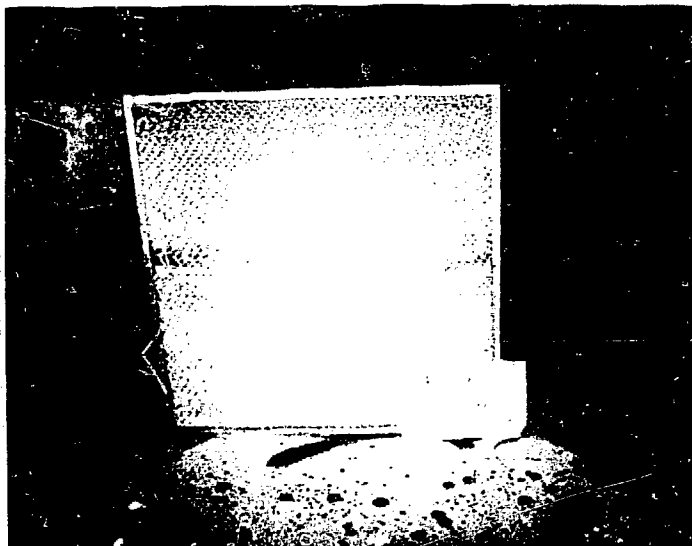
Prefilters

The use of prefilters was examined in runs FL4 and FL5 for aerosols generated in moist and normal humidity atmospheres. The best combination for the moist atmosphere conditions (test FL4-B) was the prefilter made of two sizes of screen mesh crimped into a herring-bone pattern (Key 21). The best combination observed for the normal humidity air was FL5-G (Key 25), a folded glass mat filter. These filters are shown in Figure 11. No combination tested was high in loading for both wet and dry test aerosols. The prefilter either removed most of the test aerosol or allowed a major fraction to penetrate, leading to loading of the backup HEPA filter. The folded glass mat prefilter (Key 25) had appreciable holding ability in both tests but is no more compact than the HEPA filters used. Prefilter FL5-F (Key 24) ruptured at 4.5 kPa.

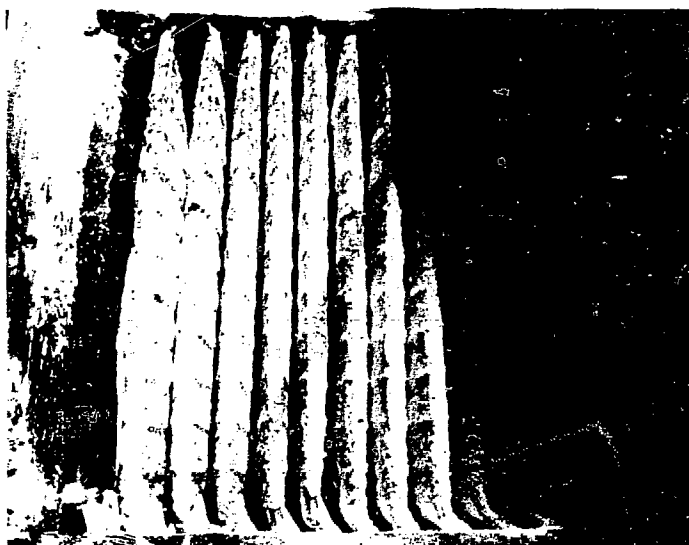
CO₂ Addition

Addition of CO₂ gas to the smoke room atmosphere and the filtering of the subsequent sodium carbonate aerosols was examined in run FL6 for several selected filter combinations. For these conditions, enhanced loading was found with over 5 kg being held at 4.9 kPa.

The smoke room air was kept at 3% CO₂ during the fire and steam at 9.1 kg/hr was added. Chemical and x-ray diffraction analysis of the aerosols deposited on all the filters showed nearly pure



RUN FL-4, PRE FILTER B-1



RUN FL-5, PRE FILTER G

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Figure 11. Two types of prefilters after loading.

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anhydrous Na_2CO_3 (see Table VI). Particle size was in the 1.4-1.7 μm aerodynamic mass median diameter (AMMD) range, as at the time as measured in the other runs. Particle shape changed earlier this run to shell-like clusters not previously observed in the tests using air. Figure 12 shows the particle shapes as seen by transmission electron micrographs.

Chemical Form of Deposits

The chemical form of the material removed from the filters, where duplicate deposits were collected, was determined by chemical analysis, and in some tests by x-ray diffraction. The amounts of Na_2CO_3 , Na_2O_3 , NaOH and total Na were found directly by analysis. The sodium not accounted for was assumed to be NaOH and the mass not accounted for was taken to be water. Sodium hydride was always less than 0.1% and sodium monoxide was not found. Table VI summarizes the analysis of these samples. The effect of adding moisture to the fire atmosphere is seen in runs FLB and FLc where the Na_2CO_3 content is reduced and the NaOH content increased. Run FLB is somewhat of an anomaly; no steam was added, but high NaOH was found. This may have resulted from some unknown moisture sources in the room or dust. This run, using nominally dry air, also had many of the characteristics of a "wet" atmosphere run in the appearance of the early filters and in the relatively low mass loadings on early filters.

In run FLH, of course, H_2O and moisture were added to the atmosphere and nearly complete conversion to anhydrous Na_2CO_3 was noted for all samples.

Aerosol Particle Size

Particle size was measured using cascade impactors in the smoke room and in the duct upstream of the filters. Two impactor types were used, one an 8-stage multijet type and the other a 6-stage radial slot type. Both impactors gave reasonably consistent data for the size being measured. Table VII presents the sizes found in the smoke room. These sizes are, however, smaller than reported by Hilliard et al.⁽⁷⁾ at this conference for measurements made in an 850-m³ vessel, probably because of the weaker aerosol source and the lower aerosol concentrations achieved in the present work. The sizes reported for the runs are not thought to vary by an amount which is significant to the filter loading tests.

Aerosol Release and Duct Losses

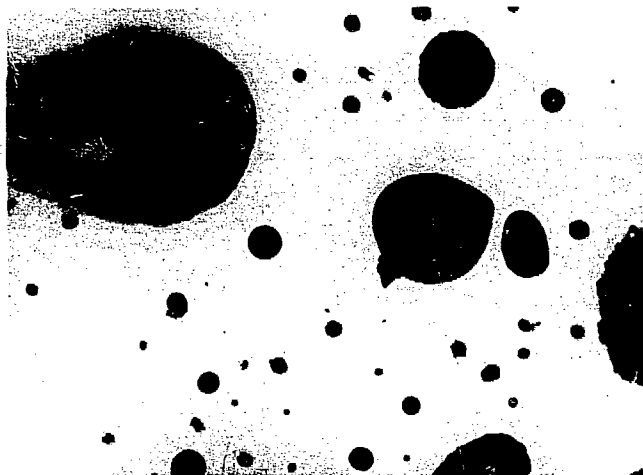
After each run the fire burned to completion and the ducts were washed to determine the sodium deposition in the duct and filter housing. For some runs the sodium aerosol on the smoke room floor was sampled and fallout determined. Table VIII presents the duct losses for each run, and also the fraction of the sodium aerosolized. The 25-cm diameter inlet duct has a 10-m horizontal length, a 3.4-m vertical length, two 90° ells and one 45° ell. The inlet was a 30° tapered section. No separate analysis of the deposition in the vertical and horizontal section was made. Duct losses are highest for the moist aerosols.

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Table VI. Typical chemical analysis of deposits on filters.

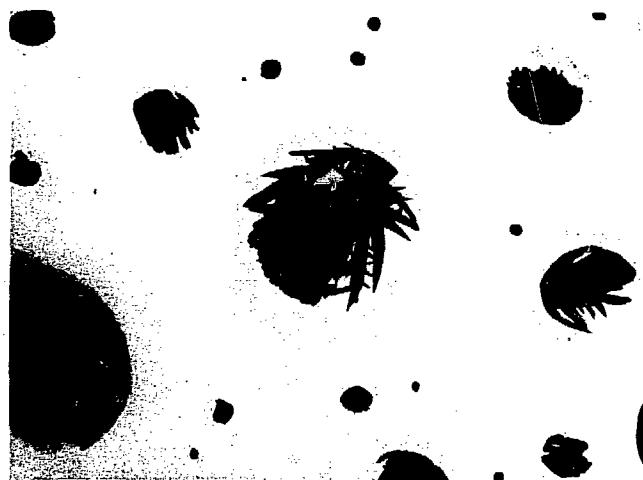
| Run and
filter no. | Time from
fire start,
min | Wt % | | | | Total
Na |
|-----------------------|---------------------------------|--------------------------|-------------------------|------|----------------------|-------------|
| | | Na_2CO_3 | Na_2O_2 | NaOH | H_2O | |
| FL1-CS2 | 125 | 3.2 | 31.8 | 44.0 | 21.0 | 44.5 |
| FL2-C | 76 | 0.6 | 8.6 | 61.4 | 29.4 | 43.0 |
| FL3-B | 22 | 1.2 | 19.5 | 33.7 | 30.6 | 40.0 |
| D | 58 | 1.0 | 26.6 | 26.9 | 26.0 | 43.0 |
| G | 113 | 0.9 | 27.7 | 33.5 | 17.4 | 47.0 |
| FL4-F | 89 | 1.8 | 12.2 | 45.9 | 40.1 | 34.4 |
| G | 108 | 3.6 | 2.3 | 56.1 | 38.0 | 35.2 |
| FL5-E1 | 82 | 1.3 | 10.0 | 74.6 | 14.0 | 49.7 |
| F1 | 104 | 3.0 | 12.0 | 69.2 | 15.7 | 48.5 |
| FL6 (A11) | 39-174 | 100 | 0 | 0 | 0 | 44.4 |

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→ 10 μ m ←

RUN FL-1 AT 117 MINUTES - DRY ATMOSPHERE



→ 10 μ m ←

RUN FL-6 AT 238 MINUTES - MOIST CO₂ ATMOSPHERE

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Figure 12. Aerosol particles, electron photomicrographs.

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Table VII. Aerosol particle sizes.

| Run | Aerosol particle sizes ^(a) | | | |
|-----|---------------------------------------|---------------------------------|---------------------------|----------------|
| | Sample time,
min | Impactor ^(b)
type | AMMD ^(c)
μm | σ _g |
| FL1 | 19 | A | 1.9 | 2.8 |
| FL2 | 21 | A | 2.5 | 2.1 |
| | 197 | S | 6.8 | 2.4 |
| FL3 | 10 | S | 2.9 | 4.9 |
| | 30 | S | 4.4 | 3.0 |
| FL5 | 20 | S | 1.6 | 2.9 |
| | 88 | A | 3.0 | 2.7 |
| | 105 | A | 3.4 | 2.7 |
| FL6 | 39 | A | 2.9 | 2.6 |
| | 71 | A | 1.4 | 3.7 |
| | 165 | A | 2.7 | 2.4 |
| | 183 | A | 2.6 | 2.4 |

(a) Sampled in smoke room

(b) A is multicircular jet, S is L-radial slot

(c) Aerodynamic Mass Median Diameter

Table VIII. Sodium aerosol release and duct losses.

| Run | Sodium in
pan, kg | Sodium,
Aerosolized
% | Duct deposition
% of intake |
|-----|----------------------|-----------------------------|--------------------------------|
| FL1 | 85 | - | 15 |
| 2 | 85 | - | 22 |
| 3 | 65 | 14 | 10 |
| 4 | 47 | 18 | 35 |
| 5 | 61 | 14 | 35 |
| 6 | 59 | 26 | 9 |

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The fraction aerosolized is based on the amount charged to the burn pan and the amount recovered from filters, duct work, and the estimated quantity deposited on the smoke room floor.

Filter Loading at Other Pressure Drops

These filter loading tests were run to rather high pressure drops. Emergency air cleaning systems are not expected to be used often, if at all. Hence, installation costs, not operating costs, are more important. The filter bank mass loading and therefore the size can obviously be reduced by running to the highest allowable pressure drop.

If the filter loading ability is required at lower pressure drops, the amounts reported can be prorated on a time basis. For most of the runs reported, the smoke room concentrations were nearly constant during the loading of a filter and the flow rate was held constant. Hence, the mass collected on a filter increased nearly linearly with running time. If a pressure drop at a shorter time is selected, the mass will be reduced by the same fraction as the time was reduced.

During loading of filters FL5-G and 6-F, the aerosol concentration decreased a significant amount and this accounts for the apparently linear increase of pressure drop with time.

Discussion and Conclusions

These tests show that the loading capacity of filters for sodium oxide, hydroxide and carbonate aerosols is quite variable, even when loaded under similar flow conditions. The amount held on nominal 0.47 m³/s (1000 cfm) HEPA filters ranged from 0.62 to 3.9 kg at 5 kPa for mixed Na₂O₂-NaOH aerosols. For dry sodium carbonate aerosols, up to 5.1 kg were held at 5 kPa. These values are in the same range of loadings as obtained in recent studies by other investigators. Gunn and Eaton in the 14th Air Cleaning Conference⁽⁶⁾ reported sodium oxide loading of 0.36 to 1.3 kg at 1.5 kPa (6" H₂O) for several filter types at 0.7 m³/s in normal humidity air. Our data for similar filters FL1-A, FL1-D, and FL3-C indicate 1.4 to 1.5 kg at 0.47 m³/s and 1.5 kPa which is quite comparable. Jordan⁽⁵⁾ reports 160 g/m² at 5 kPa for 2.6 m² fiberglass filters, or 1.6 kg for a HEPA containing 25 m² of media. Viles⁽⁴⁾ developed 20-cm thick graded media filter packs which held about 2.8 kg/m² of face area at 5.2 kPa pressure drop. The face velocities were 2 to 8 m/min which is lower than used during these tests. On a filter volume basis, this loading is equivalent to 1.6 kg on a standard-sized 60 x 60 x 30 cm HEPA filter. Again, this loading is comparable to that obtained on HEPA filters in this work.

Some general conclusions about the loading of HEPA filters with sodium oxide and sodium hydroxide aerosols can be made from the work reported here.

- Holding capacity at 5 kPa pressure drop ranged from 0.6 to 3.9 for a nominal 0.47 m³/s HEPA.
- Holding capacity is not strongly increased by using filters at lower than nominal flow.
- HEPA filters without separators have a high holding capacity.

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- Holding capacity is not strongly dependent on media efficiency.
- None of the prefilters tested were effective in increasing the combined filter holding capacity.
- The prefilter/HEPA filter combination which was best for dry aerosols was not the best for moist aerosols.
- The first fumes from a sodium fire give a higher pressure drop. This is thought to be due to a higher water/sodium content of the first aerosol produced.
- Sodium carbonate generated in a moist CO₂-containing atmosphere had higher loading capacities (up to 5.1 kg).
- The variation in the aerosol is more important than the HEPA filter construction or filter media in determining the loading obtained.
- Moisture and relative humidity in the fire atmosphere decreases the filter loading capacity. Further study will be required to determine the cause of this change.

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2. J.D. McCormack, R.K. Hilliard, A.K. Postma, and L.D. Muhlestein, "An Evaluation of Alternative Air Cleaning Systems for Emergency Use in LMFBR Plants," Proceedings of the 14th ERDA Air Cleaning Conference, CONF-760822, pg 989, February 1977.
3. R. Dennis, E. Kristal, and L. Silverman, "Fibrous Filters for NaK Fume Removal," NYO-4811, Harvard Air Cleaning Laboratory, May 1, 1960.
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DISCUSSION

BURCHSTED: Did I hear you say that the pressure loading across the HEPA filter itself went to 20 to 25 in.w.g.?

McCORMACK: Yes.

BURCHSTED: That's very interesting because we have been talking about 10 in. w.g. maximum. Did you notice any damage to the filters?

McCORMACK: Yes, at least one HEPA filter ruptured, one of the non-woven cotton fabric filters ruptured, and one of the low efficiency pleated filters ruptured in the range of 10-13 in.w.g.

BURCHSTED: Did you do any follow-up DOP tests to see if non-visible degradation had occurred?

McCORMACK: We did not, but we now have a 30 day test in progress. We have been loading the filter more gradually and DOP-testing it each seven days. So far, it has maintained its original efficiency.

BURCHSTED: What is the maximum ΔP you have reached so far?

McCORMACK: Around 12 in.w.g.

MURROW: Have you considered using prefilters or systems similar to those used at the Lawrence Livermore Laboratory for smoke removal? Do you think you might?

McCORMACK: We have not tried them. I have thought of using moving prefilters and they look appealing but we wanted to start with a more passive device.

MURROW: Perhaps some of the information from the LLL fire tests might cross pollinate with your studies.

McCORMACK: Yes, I am very interested in those tests.

DYMENT: In your conclusions, you find that HEPA filters without separators have a higher holding capacity. Is the increased capacity produced by a difference in the surface area of the paper in the two types of filter?

McCORMACK: I don't know for certain but I'm sure that increased area contributes to the increased holding capacity.

SCHIKARSKI: Concerning the behavior of fiber glass filters loaded with sodium fire aerosols, we have made a number of experiments at Karlsruhe which were reported at an earlier Air Cleaning Conference. We found loading capacities similar to those reported by Mr. McCormack. However, we also found, after the test, that loaded filters showed small "puffs" (little explosions) that destroyed the filter media. We believe that these explosions are due to the fact that not all sodium fire aerosols are completely oxidized before entering the filter. Therefore, small sodium droplets covered by sodium oxide will exist in the filter media which give rise to later reactions (or small explosions) with atmospheric moisture. This effect has led to the decision that, in German LMFBRs, fiber glass HEPA filters will not be used where sodium fires may occur.

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PERFORMANCE OF A WET CELL WASHER FOR SODIUM FIRE AEROSOLS

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Abstract

The suitability of 6 packing materials for removal of sodium fire aerosols in a wet cell scrubber was evaluated experimentally by measuring the efficiency and flow resistance of two 10.2 cm deep cells in series at air face velocities of 185 and 289 cm/sec, water flow rates of 0.210 and 0.387 cm³/sec/cm², with and without aerosol prehumidification. The most satisfactory material was found to be 50 μ m diameter stainless steel fibers at a packing density of 30 kg/m³ (porosity = 0.99). Two 10.2 cm deep cells in series gave a removal efficiency of 85% for typical sodium fire aerosols at a face velocity of 289 cm/sec. Measurement of efficiency as a function of particle size indicated a reduction in efficiency in the 0.4 to 1.1 μ m size range for all packing materials.

Introduction

One of the proposed safety systems for the LMFBR is an aerosol filtration system through which the reactor containment building atmosphere would be vented in the event of an accidental release of burning sodium. A proposed filter system consists of two stages: the first, a scrubbing system, containing a humidifying section, a wet cell washer, a water separator or demister, and a heater/dryer, and the second, consisting of a prefilter, a high efficiency particulate filter (HEPA), a zeolite bed, and another HEPA filter. Burning sodium metal in a normal oxygen atmosphere produces a dense aerosol composed of a mixture of sodium oxides, hydroxide, and carbonates. When combustion occurs at a relative humidity greater than 35%, the resulting agglomerated aerosol particles absorb water vapor and form sticky droplets that create difficult clogging and crusting problems for high efficiency air cleaning equipment.⁽¹⁾ Therefore, the scrubbing section would have to maintain greater than 90% removal efficiency without clogging for a period of many hours. The present study was conducted to develop performance data for the first stage scrubbing system.

Wet cell washers utilize beds of coarse fibers and are characterized by low pressure drop, high efficiency for particles above 5 μ m, and the ability to recirculate the scrubbing water for long periods. A principal advantage of this device for the application described above is its ability to resist clogging by sticky water soluble sodium aerosol particles.

Experimental Apparatus and Procedures

Figure 1 is a diagram of our experimental wet cell washer. It consisted of a humidification section followed by three cells, the first two wetted and the last, a non-wetted mist eliminator. A downstream centrifugal blower drew the aerosol from the chamber through

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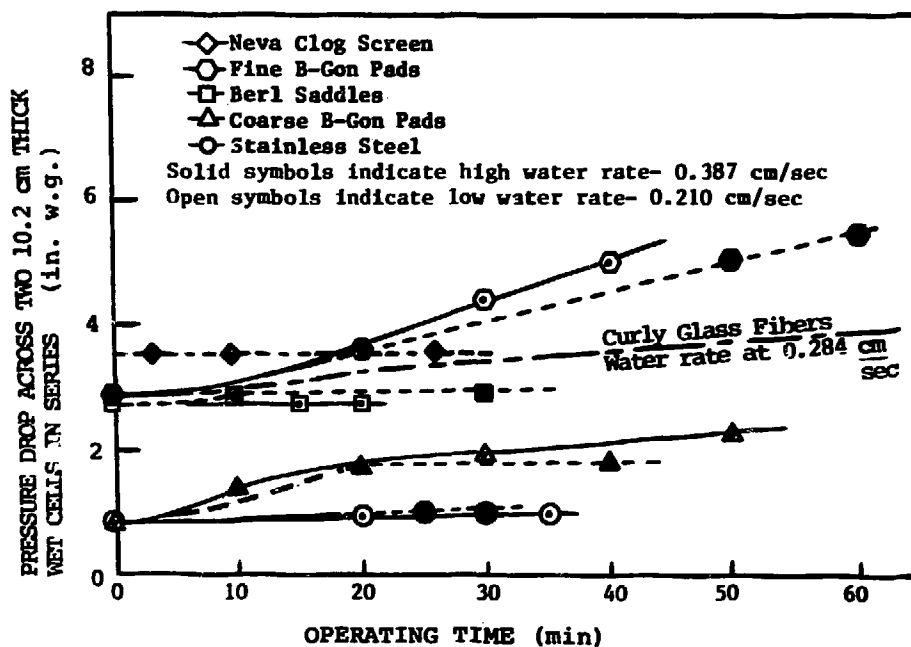
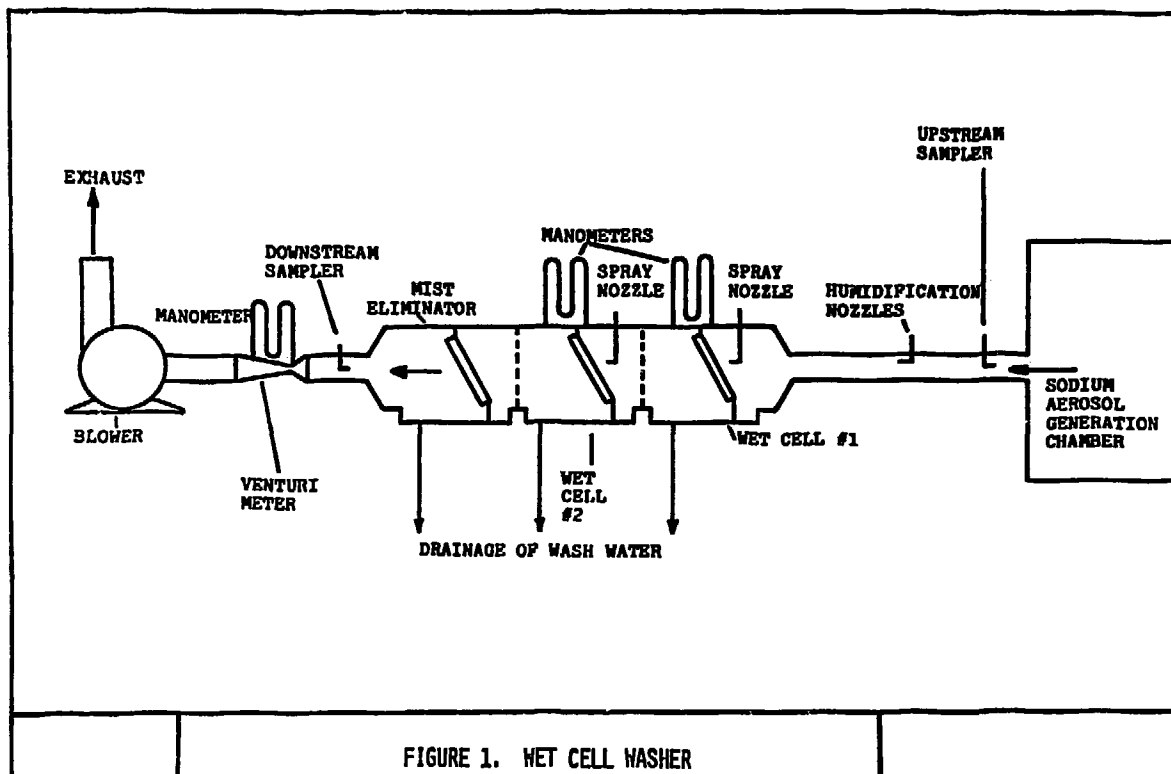


FIGURE 2. LOADING CHARACTERISTICS OF SIX PACKING MATERIALS AT 185 cm/sec

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the humidification section and the scrubber unit. A Venturi meter, located downstream of the scrubber unit, measured air flow rate and liquid manometers were used to measure pressure drop across the cells. The upstream sampling port was located between the 90 m³ aerosol generation chamber and the humidification section and the downstream sampling port was located after the mist eliminator cell. Cells up to 4" thick were mounted in a support frame sealed to the scrubber unit at a 45 degree angle to provide a 12" x 15" filter face area (820 cm² active area) in a 12" x 12" (30 cm x 30 cm) scrubber cross section. System air flow rates were 0.151 and 0.236 m³/sec, giving filter face velocities of 185 and 289 cm/sec.

Experimental variables were cell construction, cell face velocity, spray water flow rate, option of prehumidification, and aerosol concentration. Experiments were conducted using a factorial design for each filter medium and two levels for system variables. A total of 30 runs were made providing 151 efficiency and pressure drop measurements. Tests were done in a random order and analyzed statistically to show the significance of each variable or combination.

Cells were filled with the packing materials at the porosities indicated in Table I.

Table I. Characteristics of cell packings.

| <u>Cell packing</u> | <u>Fiber Size</u> | <u>Packing Density</u> | <u>Porosity</u> |
|------------------------|-------------------|------------------------|-----------------|
| curly glass fibers | 35 μ m | 16 kg/m ³ | .99 |
| stainless steel fibers | 50 μ m | 30 kg/m ³ | .99 |
| coarse B-GON* pads | 410 μ m | 26 kg/m ³ | .96 |
| fine B-GON* pads | 208 μ m | 31 kg/m ³ | .96 |
| ceramic Berl saddles | 1/4" nominal | 678 kg/m ³ | .72 |

In addition to packed cells, a Neva-clog** screen was tested. It is constructed from two thin perforated stainless steel plates each having an array of 0.025 cm diameter holes on 0.075 cm centers with the holes in one plate offset from the other so that aerosol flow must bend twice. This induces interception and impaction of aerosol particles on and between the plates.

Two Spraying Systems 1/4 J atomizing nozzles, each spraying 45 cm³/min, were used to humidify the sodium aerosol before it entered the first wet cell. These are two-fluid nozzles that use water and air pressure to produce a fine water spray with a high concentration of droplets in the 1-10 μ m range.⁽²⁾ Retention time in the humidification chamber was 1 sec.

Each wet cell is sprayed with a single Spraying Systems type 3/8" HH27W nozzle that produces a wide angle (120 degrees) solid cone

*Monofilament polypropylene fibers manufactured by Kimre Inc., Perrine, Florida 33157.

**Buffalo Forge Co., Buffalo, N.Y.

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spray that covered the entire cell face. Water flow rates were 170 and 320 cm³/sec to give a water rate per unit area of cell face of 0.210 to 0.390 cm³/sec/cm². Water drained to a sump at the bottom of each cabinet from which it could be recirculated or sent to waste. The final stage was an unsprayed, 2" thick mist eliminator cell to remove entrained water droplets.

Sodium aerosols were generated in a 90 m³ chamber. Elemental sodium was burned continuously in a heated steel pot to produce an aerosol of controlled concentration. A feed chute, 5 cm in diameter and 1.2 meters long and slanted 45 degrees from outside of the chamber to the sodium burning pot, was used to add small increments of sodium metal periodically throughout the course of each run. The rate of sodium addition was determined by monitoring the aerosol mass concentration in the chamber at ten minute intervals. In this manner, sodium fires were sustained for several hours and the sodium aerosol concentration maintained at 1 ± 0.2 gm/m³ by the addition of 35 to 50 g/min to the pool fire. For aerosol concentrations greater than 1.0 g/m³, sodium was added at the rate of 85 to 110 g/min and 1600 cm³/sec of compressed air was directed onto the surface of the burning sodium pool to increase the burning rate by providing additional oxygen and turbulent mixing. Thermocouples were used to monitor the temperature of the sodium in the burning pot. Initially the burning pot was heated electrically, but rapid heater element failures led to the later use of a gas burner.

Sodium aerosol was withdrawn from the 90 m³ aerosol chamber through a 25 cm diameter port that led to the prehumidification section upstream of the scrubber unit. Ports in the opposite chamber wall provided make up air. Other ports, located at mid-chamber height, were used to sample the chamber aerosol concentration. Chamber aerosol mass concentrations agreed with concentrations measured upstream of the scrubber unit, indicating negligible loss in the ductwork leading from the aerosol chamber to the scrubber unit.

Simultaneous isokinetic samples of the sodium aerosol concentration were collected up and downstream of the scrubber unit on all-glass absolute filter paper (MSA* 1106-B). After sampling was completed, and before the scrubber system was set to the next operating condition, the spray water rate to the cells was increased to about 0.5 cm³/sec/cm² to clean the packing until the pressure drop across the filter had stabilized.

The deposit on sample filters was dissolved in 100 ml of deionized distilled water and the solution analyzed for sodium content by atomic absorption spectrophotometry. Because the all-glass filter paper contains some leachable sodium, measurements were corrected for filter blank which was less than a 0.5% correction. By careful dilution to the most sensitive range of the atomic absorption spectrophotometer**, the sodium concentration was measured to an accuracy to $\pm$

*Mine Safety Appliances, Pittsburgh, PA

**Perkin-Elmer Model 360

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0.03 $\mu\text{g}/\text{ml}$. From dilution calculations and the precision of the analysis the accuracy of the total sodium collection efficiency measurements was determined to within $\pm 6\%$.

Eight stage Anderson 2000* cascade impactors were used at 28.3 lpm to determine the particle size distribution of the sodium aerosol upstream and downstream of the scrubber and the fractional efficiency for each cell packing. The cascade impactor measurements provided nine particle size ranges from 0.4 to 9.0 μm .

Results and Discussion

Pressure drop across two wet cells in series was measured at two water spray rates (0.210 and 0.387 $\text{cm}^3/\text{sec}/\text{cm}^2$) and at two face velocities (185 and 289 cm/sec). Pressure drop across the wet cells increased with increasing water rate, increasing face velocity, and, for some packing materials, with time of operation. Figure 2 presents pressure drop data at a face velocity of 185 cm/sec for each of the wetted cell packing materials as a function of exposure duration. The pressure drop of the fine and coarse B-GON pads increased more rapidly at the lower water spray rate than at the higher spray rate, but there was very little change in pressure drop at either water spray rate for the Berl saddles and stainless steel fiber packings.

In Figure 3, pressure drop across two wet cells as a function of scrubbing time is shown for the higher face velocity of 289 cm/sec . Again, the B-GON pads showed a greater pressure drop increase with time at the lower water spray rate, whereas Berl saddles and stainless steel fibers leveled off at a constant pressure drop after a small initial build up. Measurements of the pressure drop across individual cells indicated that an increase in pressure drop with time was a result solely of the increased resistance of the first cell.

Results of efficiency measurements for six packing materials are summarized in Table II in terms of the three dichotomous system variables: face velocity, water rate, and humidification. Except for one outlier, the efficiencies ranged from 59 to 92% with a maximum range for any packing material of 20%. Because of the narrow range and the high variability of the results, no clear patterns emerge. This fact was confirmed by the factorial analysis which failed to show significance for any of the system variables or interactions. The most important, though non-significant, effect came from face velocity, followed by water rate, and then, by humidification. The results for the stainless steel fibers, based on the largest number of measurements, showed consistent but modest increases in efficiency with face velocity and humidification. It is noteworthy that prehumidification had a weak and inconsistent effect on efficiency. Apparently, these hygroscopic particles grow so rapidly that they reach full growth in the region of the cell spray water and prehumidification is unnecessary. The curly glass fibers deteriorated

*Anderson 200, Atlanta, GA

Table II. Sodium removal efficiencies in percent for 7 cell packing materials.

| Cell Packing | 185 cm/sec Face Velocity | | | | 289 cm/sec Face Velocity | | | |
|---|--|----------|--|------------|--|--------|--|--------|
| | Water Rate | | | | Water Rate | | | |
| | 0.210 cm ³ /sec/cm ² | | 0.387 cm ³ /sec/cm ² | | 0.210 cm ³ /sec/cm ² | | 0.387 cm ³ /sec/cm ² | |
| | Humidification | | Humidification | | Humidification | | Humidification | |
| | No | Yes | No | Yes | No | Yes | No | Yes |
| Curly Glass Fibers | *86 (3) | *89 (17) | — | 89 ** (10) | — | — | — | — |
| Neva Clog Screen | — | — | 57 (4) | 70 (5) | — | — | 51 (4) | 65 (6) |
| Neva Clog Screen and Curly Glass Fibers | — | — | — | 90 ** (35) | — | — | — | — |
| Coarse B-GON Pads | 61 (1) | 63 (1) | 59 (1) | 70 (1) | 60 (1) | 66 (1) | 68 (1) | 73 (1) |
| Fine B-GON Pads | 89 (1) | 88 (1) | 89 (1) | 82 (1) | 92 (1) | 87 (1) | 88 (1) | 89 (1) |
| Stainless Steel Fibers | 70 (3) | 75 (4) | 70 (3) | 81 (5) | 81 (3) | 83 (3) | 81 (3) | 80 (7) |
| Porcelain Berl Saddles | 62 (1) | 49 (1) | 81 (1) | 68 (1) | 63 (1) | 68 (1) | 72 (1) | 61 (1) |

number of observations shown in parentheses

*Water rate was 0.210-0.284 cm³/sec/cm²

**Water rate was 0.318-0.340 cm³/sec/cm²

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Table III. Aerosol particle size characteristics.

| Scrubber
Run
| Chamber Aerosol
Concentration
(g/m ³) | Mass Median
Diameter
(μm) | Geometric
Standard
Deviation |
|----------------------------|---|---------------------------------|------------------------------------|
| 19 | 0.75 | 1.58 | 2.03 |
| 20 | 0.96 | 1.63 | 1.90 |
| 21 | 0.85 | 1.57 | 2.01 |
| 22 | 0.88 | 1.50 | 2.37 |
| 23 | 1.05 | 1.58 | 2.10 |
| 25 | 0.82 | 1.70 | 2.12 |
| 26 | 1.02 | 1.33 | 1.95 |
| 27 | 0.88 | 1.38 | 2.14 |
| 28 | 4.75 | 1.48 | 1.84 |
| 29 | 4.75 | 1.41 | 1.56 |
| 30 | 0.96 | 1.37 | 2.09 |
| Average of 11 measurements | | 1.50 | 2.01 |

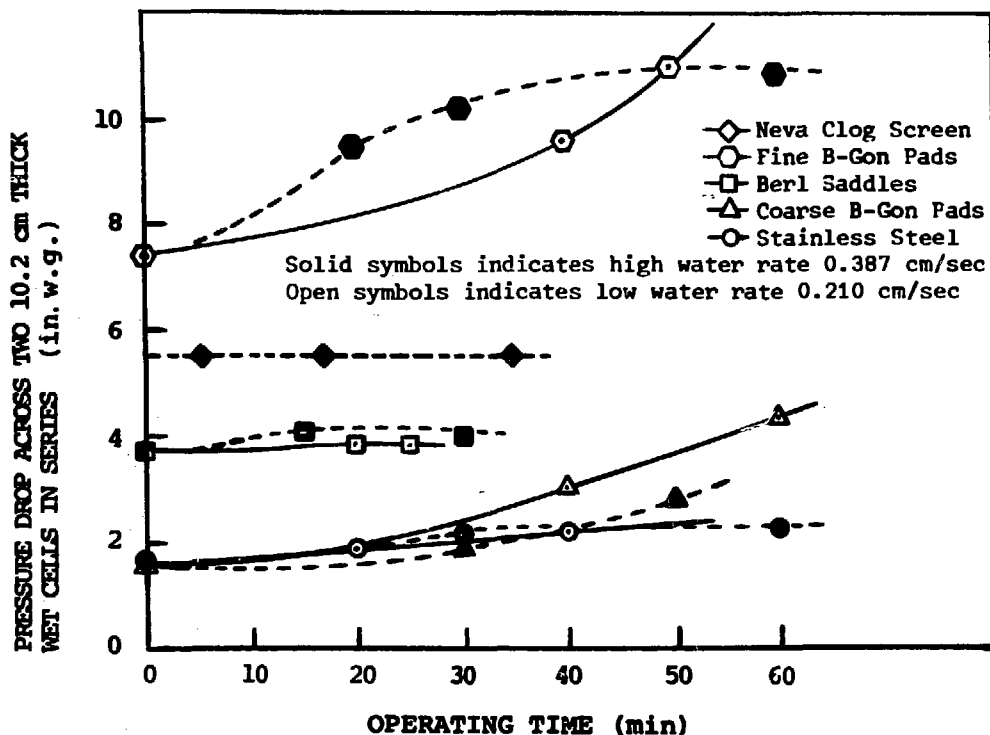


FIGURE 3. LOADING CHARACTERISTICS OF FIVE PACKING MATERIALS AT 289 cm/sec

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noticeably after 90 minutes of exposure to the strong caustic scrubbing solution whereas other materials showed no visible change after 8 hours of exposure.

A typical particle size distribution of the sodium fire aerosol upstream of the scrubber system is shown in Figure 4. All upstream size distributions showed a good fit to the log normal size distribution. Table III presents size distributions for several scrubber runs. The average Mass Median Diameter (MMD) was 1.50 μm with an average Geometric Standard Deviation (GSD) of 2.01. These particle size distributions are in reasonable agreement with data reported by Viles et al.³ (i.e., MMD = 1.26 to 1.74 μm and GSD = 2.65 - 3.36 in a nitrogen atmosphere) and Lauben et al.⁴ (MMD = 1.4 μm and GSD = 1.95).

Collection efficiency as a function of particle size was determined using up and downstream Anderson impactor sampling. Table IV presents the efficiency for 9 particle size ranges for each of the cell packing materials tested. In general, collection efficiency was near 100% for particle sizes larger than 2 or 3 μm . As shown in Figure 5, the lowest collection efficiency was found in the 0.40 μm to 1.1 μm particle size range. The increased efficiency for particles less than 0.5 μm shown in Figure 5 is not a result of diffusion of particles to collector surfaces. Although this mechanism becomes more effective as particle size decreases, it is comparatively weak for submicrometer particles. As shown in Table V, the predicted system efficiencies due to diffusion and impaction continue to decline as particle size decreases in the submicrometer range.

Table V. Calculated* scrubber efficiency due to diffusion and impaction for cells each packed with 10 cm of 50 μm diameter fibers.

| <u>Particle Size, μm</u> | <u>Eff. due to diffusion</u> | <u>Eff. due to impaction</u> | <u>Total eff.**</u> |
|--|------------------------------|------------------------------|---------------------|
| 0.25 | 2.0% | 0.05% | 2.0% |
| 0.55 | 1.1 | 4.5 | 5.5 |
| 0.9 | 0.8 | 34.7 | 35.4 |
| 1.6 | 0.6 | 98.6 | 98.6 |

*Efficiencies calculated using the methods described in reference (5).

**Assumes single fiber efficiencies for diffusion and impaction are additive.

The apparent increase in collection efficiency for particles less than 0.40 μm is thought to result from depletion of particle mass in this size range as a result of particle growth in the humid atmosphere rather than from collection. Growth by water adsorption occurs for all sizes of particles passing through the scrubber and shifts the size spectrum upward with a net reduction in the numbers of particles in the smallest sizes. This means that the smallest particles are depleted by growth to larger sizes and the larger particles are depleted by inertial removal, leaving an intermediate size

Table IV. Fractional efficiency of 5 packing materials

| Cell
Packing
Material | Face
Velocity
cm/sec | Aerosol Particle Size Range, μm | | | | | | | | |
|-----------------------------|----------------------------|--|---------|--------|---------|---------|---------|---------|---------|------|
| | | <.40 | 0.4-0.7 | .7-1.1 | 1.1-2.1 | 2.1-3.3 | 3.3-4.7 | 4.7-5.8 | 5.8-9.0 | >9.0 |
| Neva | 185 | 48 | 28 | 29 | 51 | 94 | 94 | 77 | 74 | 64 |
| Clog
Screen** | 289 | 67 | 58 | 49 | 67 | 67 | 100 | 67 | 83 | 95 |
| Coarse
B-GON | 185 | 68 | 51 | 22 | 43 | 55 | 98 | 100 | 89 | 75 |
| Fine
B-GON | 289 | 94 | 69 | 66 | 96 | 99 | 98 | 100 | 100 | 81 |
| Stainless
Steel | 185 | 62 | 22 | * | * | 96 | 96 | 100 | 100 | 100 |
| | 289 | 71 | 39 | 42 | 85 | 100 | 100 | 100 | 100 | 100 |
| Berl
Saddles | 185 | 95 | 57 | * | * | 95 | 100 | 100 | 100 | 100 |

* Unreliable data.

** Evaluated without mist eliminator stage.

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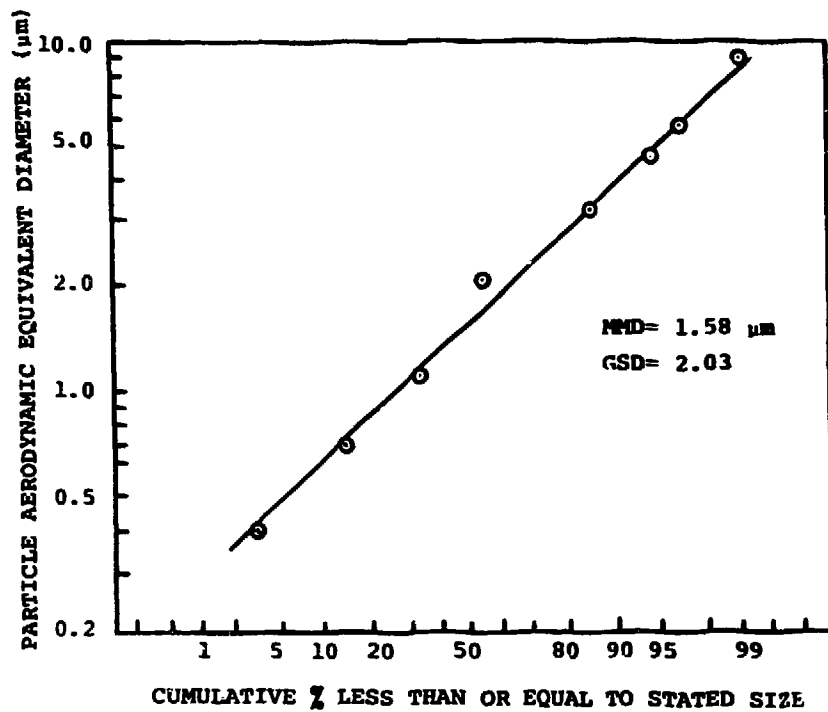


FIGURE 4. SODIUM FIRE AEROSOL INLET PARTICLE SIZE DISTRIBUTION

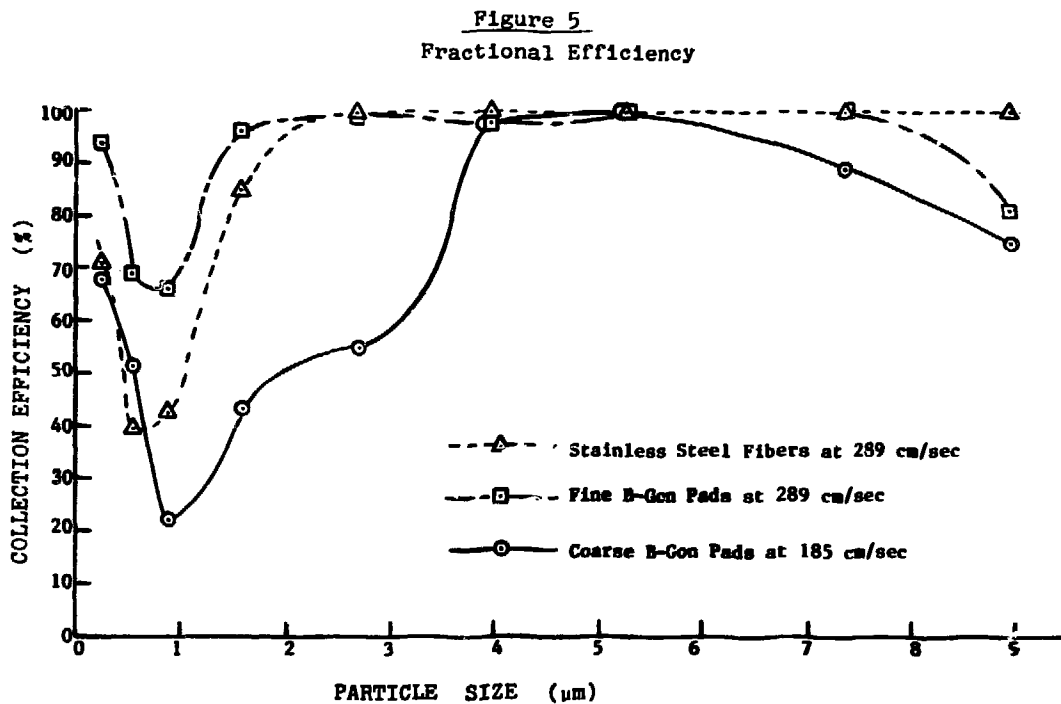


FIGURE 5. FRACTIONAL EFFICIENCY

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of maximum penetration. The size of maximum penetration depends on the inertial removal characteristics of the scrubber, the particle size distribution, and the particle size growth that occurs in the scrubber. For this particular system, i.e., a sodium pool fire in a normal atmosphere and a wet cell scrubber operated as described above, the apparent particle size for maximum penetration occurred between 0.5 and 1.0 μm .

Conclusions

Based on the results presented here, it may be concluded that it is feasible to use a wet cell scrubber upstream of a HEPA filter to reduce the loading of sodium combustion aerosol reaching the HEPA filter by a factor of ten or more (depending on scrubber size) for a period of many hours. A satisfactory design requires at least three cells in series, a face velocity of 200-300 cm/sec, and a water rate of 400 $\text{cm}^3/\text{sec}/\text{cm}^2$.

The most satisfactory single packing material studied was 50 μm diameter stainless steel fibers at a face velocity of 289 cm/sec. This material gave the lowest pressure drop and showed little tendency to clog when operated at a water rate of 0.387 $\text{cm}^3/\text{sec}/\text{cm}^2$. At this operating condition, typical sodium fire aerosols were removed by two 10 cm deep beds in series at efficiencies in the 80-90% range. Although the fine B-GON pads gave higher efficiencies than the stainless steel fibers, they were unsatisfactory because the flow resistance of the B-GON pads increased with time for all operating conditions as a result of incomplete wetting.

Combinations of packing materials may provide better performance than the use of a single material. Because the cell packings that gave the highest efficiencies tended to clog with time, they should be preceded by non-clogging, though lower efficiency, cells in a manner similar to a graded fiber diameter deep bed filter, i.e., the upstream cells would reduce the loading on the high efficiency cell so that the latter would never clog or would clog at an acceptably slower rate. Such a system might consist of a Neva-clog screen or $\frac{1}{4}$ " Berl saddles for the first cell (which would remove about 60% of the particulate mass but would not clog under any realistic conditions), a second cell of 50 μm stainless steel fibers which would capture an additional 25 to 30%, and a final cell of 35 μm stainless steel fibers which would ensure an overall efficiency in excess of 90%. Such a system would operate for 7 to 10 hours at a face velocity of 185 cm/sec and an inlet concentration of 1 gm/m^3 before it increased in resistance by one inch of water.

The curly glass fibers displayed inadequate chemical resistance to sodium fire aerosol after 90 minutes exposure, but other materials showed no visible change after 8 hours of exposure.

It was found that there is little advantage to using a prehumidification stage. This space could be used more profitably by installing a low resistance, non-clogging stage to reduce the possibility of clogging of subsequent stages.

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Packing materials that were non-clogging at an aerosol concentration of 1 gm/m^3 were also non-clogging at 5 gm/m^3 .

Because cells clog rapidly without continuous application of water, the water system must be designed to preclude even momentary interruptions of water and the water spray must be initiated prior to aerosol flow.

Although the use of a high water rate, $0.387 \text{ cm}^3/\text{sec}/\text{cm}^2$, had little effect on system efficiency, it prevented or reduced the rate of clogging.

The combined effect of particle growth by humidification and particle capture in the scrubber resulted in a most penetrating particle size. For the conditions evaluated, this size was between 0.5 and $1.0 \text{ }\mu\text{m}$.

Acknowledgement

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DISCUSSION

BURCHSTED: The efficiencies shown are mass efficiencies, are they not?

HINDS: Yes, they are sodium mass efficiencies determined by atomic absorption measurements of up- and downstream filters.

ALVARES: When you turned the water off and you got plugging, were you able to unplug by turning the water back on again?

HINDS: When we turned the water off, it wasn't intentional; the line broke and we paid complete attention to getting the line fixed. We ended up abandoning the whole test from then on. I don't think we ever made that particular test, but I believe it would be successful.

ALVARES: Did you ever run a HEPA filter downstream?

HINDS: No.

ALVARES: Why have no downstream HEPAs been installed to validate the performance of the scrubber?

HINDS: The objective of this study was to test only the performance of the scrubber. I believe tests such as you are asking for are planned at Hanford Engineering Development Laboratory.

HILLIARD: I suggest that your test aerosol was actually prehumidified in the test cell since high humidity air existed due to inflow and also to the gas-heated sodium system.

HINDS: We did allow room air to go into the chamber at about 40% relative humidity. Also, we had a burner that produced both CO₂ and water vapor, so there was significant moisture present.

HILLIARD: It looked from your slide as if you took the water exit streams and put them into a common pool. Did you ever measure the concentration in the liquid from the separate stages?

HINDS: We never measured it from the separate stages.

HILLIARD: I am interested in the carryover from the demister. Did you measure its efficiency as a function of the concentration in the water? Did you recirculate the scrubber liquid?

HINDS: No, we did not recirculate the scrubber liquid, except during some preliminary trials. For the rest, we used a once through water system and did not measure the sodium concentrations in the waste water downstream. I should mention that we measured efficiency by atomic absorption based on the amount of sodium atoms present, rather than by mass, to avoid the quantification problems associated with moisture pickup by the sodium oxide particles.

HILLIARD: I would comment that you were right when you said a wet scrubber can take a larger sodium oxide loading than a filter system. But, I think the loading is probably not infinite because you have to have a supply of water and even if you recirculate, there is a limit to solubility, especially for sodium carbonate. It requires a rather large water storage capacity.

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BROWN: Did you prepare a particle growth model which would account for the minimum in your curve or did you just hypothesize that particle growth occurred?

HINDS: We based the growth on work done by Clough and Garland (AERER 6460, 1970) in the UK in which they traced out curves of expected growth for different compositions of particles at different humidities from 50 to 100%. It is about a factor of three growth in diameter for our conditions.

BROWN: So the small particles grow at a rate so fast that it accounts for the minimum in the curve? I could see how they would change the shape of the curve, but the minimum seems unusual.

HINDS: The minimum is a result of the way we measure efficiency, i.e., measuring upstream and downstream of the whole system. The fact is that there aren't any of these little particles left because they have all grown to be big particles. They haven't been collected in the traditional sense.

BROWN: I understand that but I would also think that intermediate sized particles would grow at a somewhat slower rate.

HINDS: All particle sizes grow very rapidly and the difference in rate is not enough to conclude that the largest particles are not going to grow very much.

BROWN: Solution of the set of differential equations for lumped particle size groupings should be done to verify the hypothesis of a minimum in your collection efficiency. Does a mathematical model of particle growth predict the observed minimum in collection efficiency?

HINDS: Such an analysis was not done and is not necessary. The paper by Clough and Garland (AERER 6460 (1970)) shows that particles less than 2 μm in diameter, the particles of interest here, grow to an equilibrium size at a relative humidity of 99% in 0.01 sec. This growth is about three-fold for all particle sizes and is completed prior to a particle's arrival at the first wet cell.

Dup

ENHANCED FILTRATION PROGRAM AT LLL - A PROGRESS REPORT*

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Abstract

As part of Lawrence Livermore Laboratory's Enhanced Filtration Program, we are investigating the use of an externally applied electric field to improve the performance of fibrous filters. Our objective in this program is to develop filtration systems for the nuclear industry that will reduce the cost and volume of nuclear waste associated with present systems.

We have developed a new theory of the electrostatic filter that is consistent with experimental tests made during transient and steady-state conditions. For these tests, we used ac and dc electric fields, insulated and noninsulated electrodes, and conducting and nonconducting filter media; all tests were conducted in our small-scale 25 l/s test system, using sodium chloride aerosols. Our theory employs a new mechanism based on the attraction between charged particles and charged fibers in addition to the previously proposed mechanism based on the attraction between charged particles and polarized fibers. In this theory, fibers are charged when charged particles deposit on them.

We have also developed a theoretical model that explains the increase in filter efficiency and pressure drop when particles load on the filter. The filter loading tests we conducted to evaluate this model verify its accuracy to a remarkable degree. By using the model equations, we are thus able to explain the observed increase in filter efficiency as a function of particle mass loading, particle size, and particle-particle collection efficiency. The increase in filter pressure drop also agrees quantitatively with experiments involving different air flow velocities and particle sizes. We have also conducted experimental loading tests using external electric fields with insulated and noninsulated electrodes. In both cases, our test results showed that application of an electric field increases filter efficiency and reduces pressure drop compared to similar tests without an electric field.

Two systems we developed for use in the nuclear industry use electric fields to increase the performance of fibrous filters. One is designed for use inside glove boxes to control radioactive particles at their source; the other is designed for use in ventilation systems. Here we report the results of laboratory and field evaluations for the glove box system.

I. Introduction

The high-efficiency particulate-air (HEPA) filters used in the nuclear industry to remove radioactive airborne contaminants are extremely effective and practical devices and will continue to be so despite the large number of new control devices presently available. However, HEPA filters generate a significant volume of radioactive waste and are costly to purchase and operate. The actual cost of materials and labor to buy, change, test, and dispose of a HEPA filter is several times its initial purchase price.

*This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract No. W-7405-ENG-48.

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In an effort to reduce the HEPA's operational cost and the volume of radioactive waste it generates, the Nuclear Fuel Cycle and Production Division of the Department of Energy (DOE) has contracted with Lawrence Livermore Laboratory (LLL) to develop an enhanced filtration system that will extend the service life of HEPA filters. The approach selected by LLL consists of using an electrostatic prefilter to take the load off the HEPA filter since it appears to have a greater potential for success than such methods as scrubbers and electrostatic precipitators. The total cost of and volume of radioactive waste generated by the prefilter-HEPA filter system were assumed to be significantly less than the corresponding cost and waste from a system consisting of only a HEPA filter.

II. Previous Studies

Electrostatic filters are relatively old air cleaning devices that have been investigated since 1930. Although they can have a number of different configurations, all are based on charging or polarizing filter fibers to generate an electric force between the fibers and charged particles.

The first electrostatic filter was developed in 1930 by Hansen, who found that mixing a powdered resin with wool fibers greatly improved the resulting filter efficiency⁽¹⁾. The resin particles that coated the fibers were highly charged in the mixing process thereby, in effect, producing charged fibers. This type of electrostatic filter is widely used today, especially in respirators. Unfortunately resin filters are ruined when they are exposed to organic vapors that dissolve the resin or to ionizing radiation that neutralizes the resin charge.

Recognizing the problem of charge dissipation in the resin filter, a number of investigators have developed methods to continuously recharge the filter by generating charged aerosols upstream of the filter. In a method developed by Rossano and Silverman in 1954, an electric corona was used to charge particles before they entered the filter⁽²⁾. These charged particles were initially trapped in the filter by image forces until the fibers acquired a high charge density. Further particle trapping occurred primarily from Coulombic forces between charged fibers and charged particles. Reid and Browne have recently repeated these experiments and have shown high filter efficiencies⁽³⁾. Another method for continuously recharging the filter was proposed by Mazumder and Thomas in 1967⁽⁴⁾. Instead of charging the aerosols to be filtered, they injected a charged mist into the air stream in front of the filter. The charged mist would then be trapped by the filter fibers and effectively charge them. Oncoming aerosols would then be collected by Coulombic forces between the aerosols and the charged fibers. In another method, developed by Silverman et al, electrostatic charges were generated on fibers by rubbing the downstream side of the filter with a windshield wiper⁽⁵⁾.

Electrostatic filters can also be generated by polarizing the fibers with an external electric field. Although these electrostatic filters have been commercially available since the early 1950's, they have not been widely used. In 1954, Silverman et al evaluated several commercial electrostatic filters that consisted of glass fiber mats sandwiched between a high voltage and a ground screen⁽⁵⁾. Since that time, a large number of theoretical and experimental investigations have been made on the electrostatic filter. In 1961, Havlicek reported he was able to increase the filter efficiency for potassium dichromate particles from 55% to 99.98% by applying an electric field of 10 kV/cm⁽⁶⁾. The pressure drop across the filter at 20 cm/s was 150 Pa.

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An excellent review of electrostatic filters up to 1972 was given by Davies⁽¹⁾. The theoretical investigations showed the increase in filter efficiency with an applied electric field resulted from two electrical forces that arose from both polarized particles and fibers and from charged particles and polarized fibers. A good elementary description of these two forces was given by Rivers⁽⁷⁾.

In recent years, the number of studies dealing with electrostatic filters generated by an external electric field has significantly increased. These studies have become international with research groups in Russia (Kirsch⁽⁸⁾), Germany (Walkenhorst and Zebel⁽⁹⁾), Japan (Iinoya⁽¹⁰⁾), and the United States (Bogardus⁽¹¹⁾, Lamb⁽¹²⁾, Ariman⁽¹³⁾, Hill⁽¹⁴⁾, Frederick⁽¹⁵⁾, and Penney⁽¹⁶⁾) investigating various aspects of the electrostatic filter. Most of these studies are concerned with parameters that affect the well-established increase in filter efficiency due to the external electric field. However, Frederick reported in 1975 that the electrostatic filter also had a significantly lower pressure drop than the same filter without an applied electric field⁽¹⁵⁾. Lamb and Costanza⁽¹²⁾ and Penney⁽¹⁶⁾ have also observed large reductions in filter pressure drop with an external electric field. These results indicate electrostatic filters have a longer service life than standard filters.

Electrostatic filters have also been developed with a combination of particle charging and an external electric field. Thomas and Woodfin filled the space between plates of an electrostatic precipitator with glass fiber medium and observed a large increase in performance compared to the precipitator or to the filter alone⁽¹⁷⁾. In this configuration, applied high voltage generates an external electric field; it also generates ions in the same region by a corona discharge. This complicates system analysis because polarized and charged fibers now fill the normally open regions of an electrostatic precipitator.

Hybrid electrostatic filters can also be built by having separate regions for both particle charging and filtration. Inculet and Castle⁽¹⁸⁾ have evaluated a two-stage electrostatic filter having a concentric geometry. Particles charged by a corona discharge in the first stage are then trapped in a second stage that consists of a filter medium with a superimposed electric field.

Although any of the variety of electrostatic filters discussed above could be used in our Enhanced Filtration Program, we selected a method using a fibrous filter with a superimposed electric field because it, unlike the other methods, did not involve a corona discharge. Systems that use a corona discharge have a much higher energy consumption and pose a greater fire risk than the electrostatic filter we selected.

We have been investigating the use of applied electric fields on fibrous filters since October 1975 and presented the first results of our investigation at the 14th ERDA Air Cleaning Conference⁽¹⁹⁾. Similar to a previous study by Bogardus et al, our investigation dealt with the empirical effects of air flow, electric field strength, and different filter media on filter efficiency⁽¹¹⁾. We then made a more extensive analysis of the theoretical and experimental effects of face velocity, particle charge and size, filter void volume, and fiber size⁽²⁰⁾. We found most of our experimental measurements qualitatively agreed with Zebel's theory on charged particles and polarized fibers⁽²¹⁾. The results of our most recent work cover a period from January 1977 to June 1978 and are presented in this paper. Since the Enhanced Filtration Program at LLL is a continuing effort, many of the topics presented here are still under investigation.

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III. Theory and Small-Scale Experiments

A review of the test results from our early investigations shows we have treated the electrostatic filter as an oversimplified static system^(19,20) when it is, in fact, a very complex dynamic system that involves both mechanical and electrical forces. Perhaps the most well-known dynamic process is the increase in filter efficiency and pressure drop that occurs during filter clogging. A good understanding of the detailed filtration mechanisms that control filter efficiency and pressure drop is essential for designing an optimum electrostatic filter. In Table I, we qualitatively summarize the relative effort expended on and the level of understanding of the mechanical and electrical filtration mechanisms.

We have made separate entries for clean and clogged filters to emphasize the significant changes that occur when the filter becomes clogged.

Table I. State of the Art in Filtration

| Filter Condition | Filtration Mechanism | |
|------------------|-----------------------|--------------------|
| | Mechanical | Electrical |
| Clean | Extensively studied | Moderately studied |
| | Moderately understood | Poorly understood |
| Clogged | Moderately studied | Negligibly studied |
| | Poorly understood | Not understood |

Table I shows the level of understanding of filtration mechanisms reflects the extent of investigation.

Although we are concerned with developing an electrostatic filter, we must understand the mechanical and electrical mechanisms to optimize filter design. The effect of face velocity on filter efficiency illustrates this point. Increasing the face velocity increases filter efficiency for mechanical mechanisms (primarily inertia) but decreases filter efficiency for electrical mechanisms. Since the efficiency of an electrostatic filter depends on both mechanical and electrical mechanisms, both must be considered in developing an optimum design.

The objective of our theoretical studies and small-scale experiments was to understand the mechanical and electrical filtration mechanisms for both clean and clogged filters in sufficient detail to optimize the electrostatic filter. A survey of Table I shows the only reasonably well understood filtration mechanisms are the mechanical mechanisms for clean filters. Therefore, to reach our objective, we had to investigate and develop theories for the remaining filtration mechanisms shown in Table I. We have already met our objective for the electrical mechanisms for a clean filter and the mechanical mechanisms for a clogged filter. The results of these investigations are presented in this paper. We are currently investigating the electrical filtration mechanisms for a clogged filter.

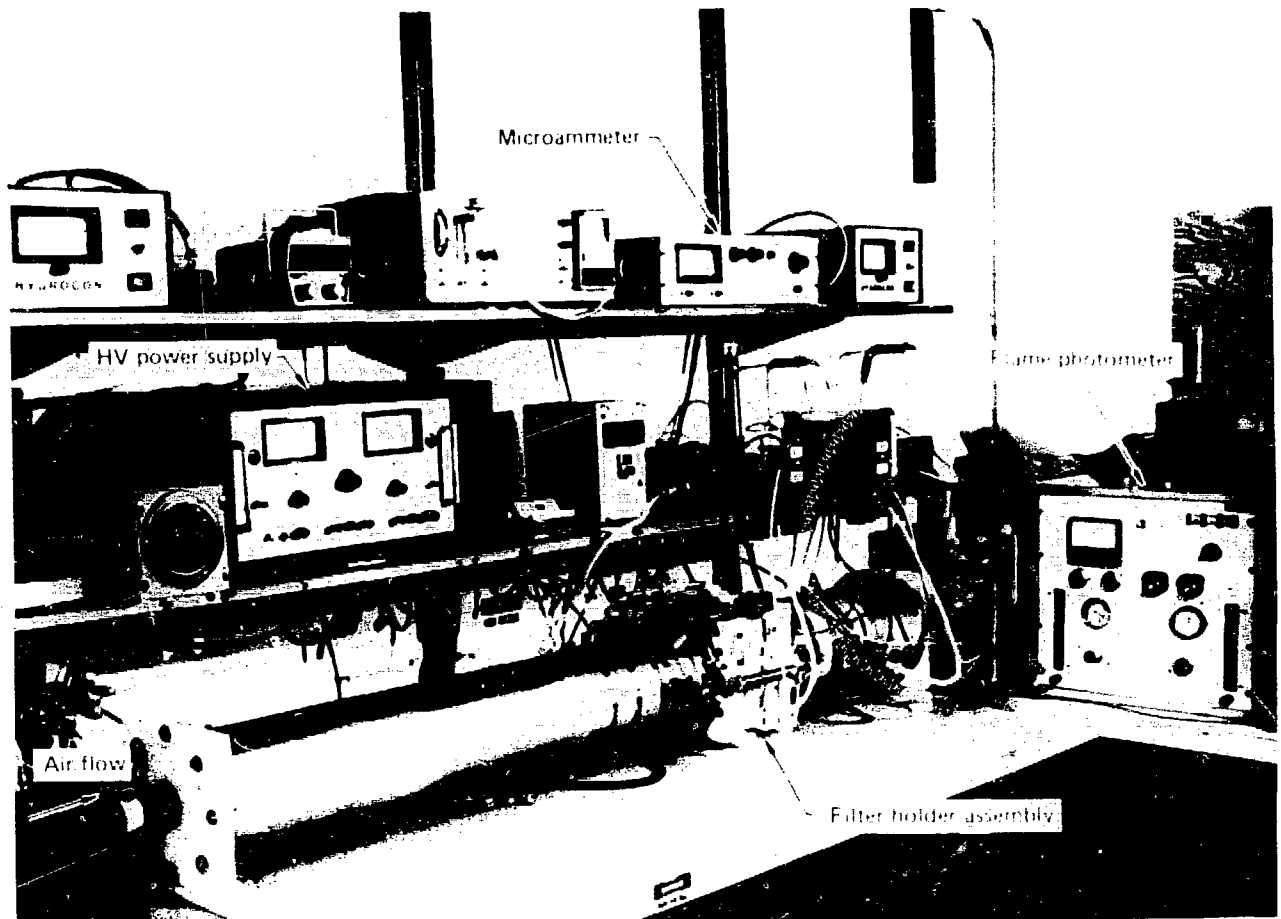


Figure 1 Small-scale filter test facility.

Experimental apparatus

The experimental apparatus used to generate the data presented in this section is shown in Fig. 1. Sodium chloride aerosols (ammd = 1.0 μ m, $\sigma_g = 2.0$) were generated using Wright nebulizers (not shown) and were passed through a flexible hose into a 20.3 cm-i.d. tube⁽¹⁹⁾. A Stairmand disk installed at the tube entrance promoted uniform mixing of the aerosols. The relative humidity of air in the duct generally fell between 40-60%. Measurements of air velocity and salt concentrations were taken across the duct and indicated a uniform distribution.

The electrostatic filter unit was mounted in the filter holder assembly and was energized with the high-voltage power supply. Efficiencies were determined by measuring the salt concentration before and after the filter with the flame photometer. Pressure drop measurements were also taken across the filter for each test. In certain experiments, a microammeter was used to measure current bleeding from the electrostatic filter. A more detailed picture of the filter holder assembly is shown in Fig. 2; here the filter holder is partially removed from the assembly to expose the electrode. Figure 3 shows the filter holder disassembled, with the perforated electrodes, glass-fiber filter, and spacer arranged in the same sequence as in the assembled unit. High voltage was applied to the rear electrode and the front electrode was grounded.

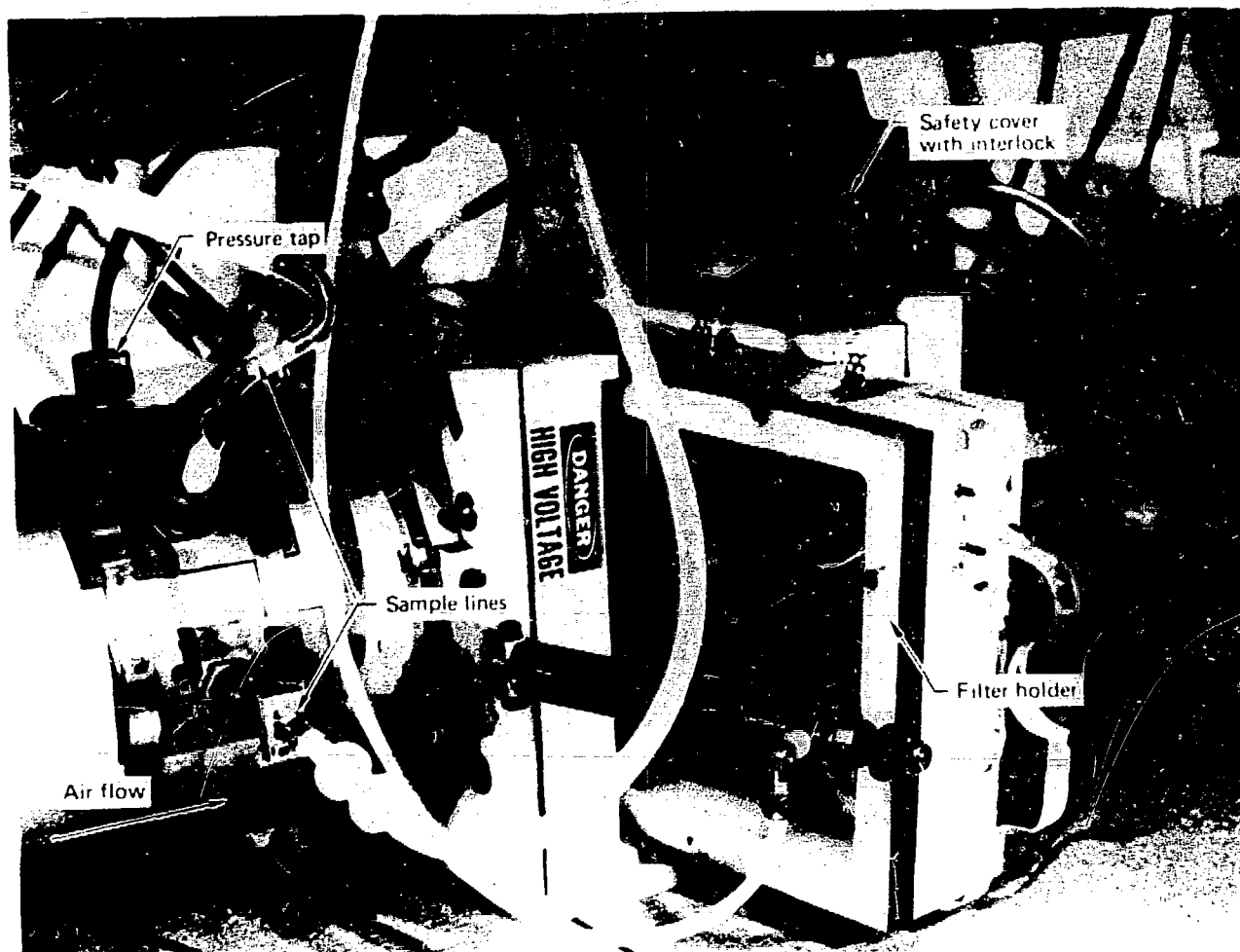


Figure 2 Filter holder assembly showing the filter holder partially removed.

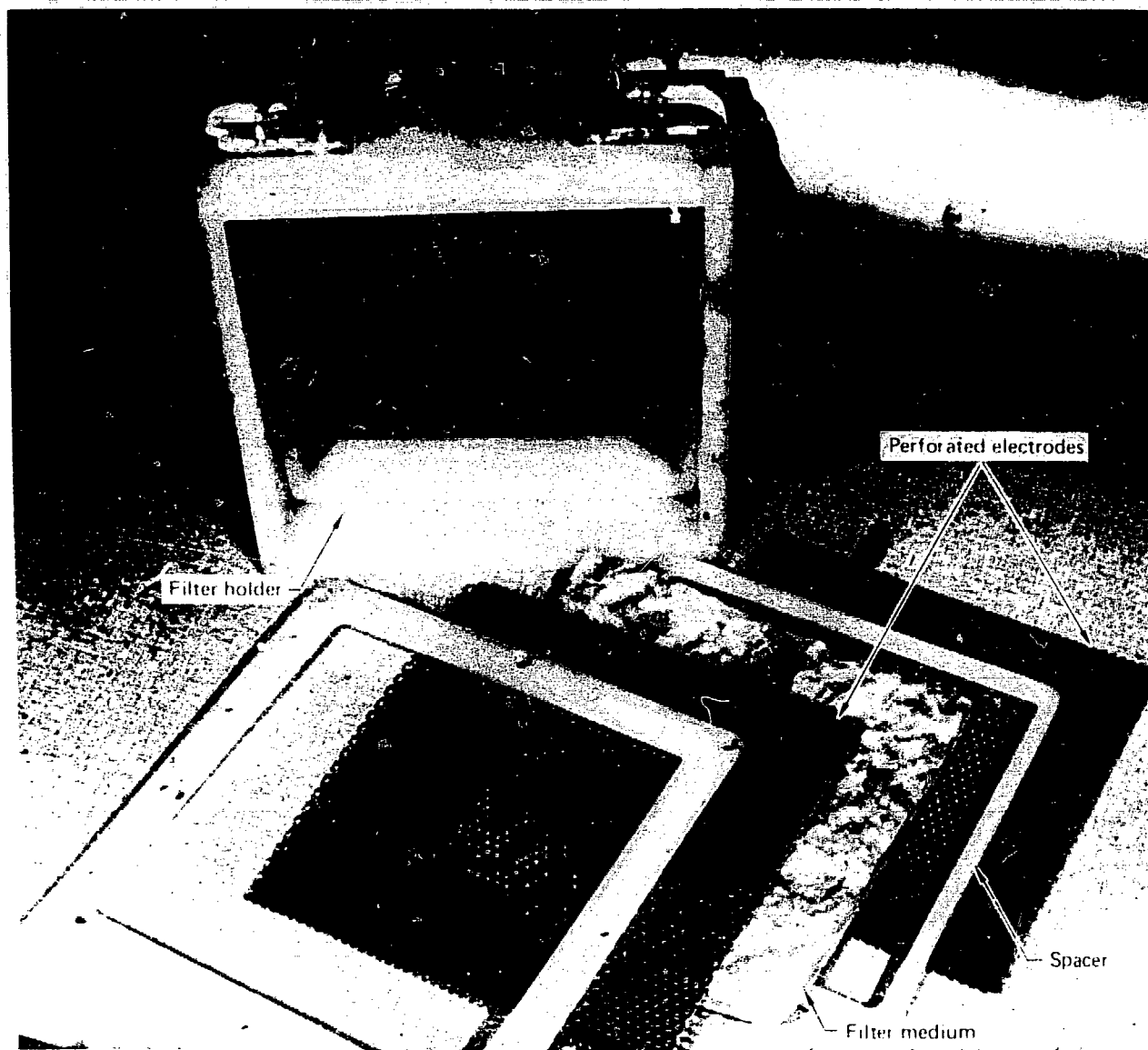


Figure 3 Disassembled filter holder.

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Extension of Filtration Theories for Fiber Size Distribution

Filtration theories are based on the premise that all filtration parameters such as fiber and particle size are monodisperse. However, in laboratory experiments most, if not all, filtration parameters have a distribution of values. The most common exception is particle size, which can be frequently treated as a monodisperse distribution. This is possible because modern particle size analyzers are able to measure small size intervals independently of other particle sizes. Measurements of filter efficiency within this size interval are, therefore, independent of efficiencies at other sizes. Efficiencies can also be determined with monodisperse aerosols because of commercially available latex particles in a wide range of monodisperse sizes. However, in spite of the availability of accurate particle size analyzers and monodisperse aerosols, most filter efficiencies are determined with heterodisperse aerosols using instruments that measure an unknown average of the size distribution.

The common practice is to take an arbitrary* average of the distribution for each parameter and treat this average as a monodisperse value. Even if the correct average were obtained, it may still be wrong to treat the average value of a parameter as a monodisperse value since products and quotients of parameters are generally involved in filtration equations. It is well known in mathematics that the average of a product is not equal to the product of the averages. Recognizing the problems that may occur when treating parameters having a distribution of values as a single value, we have been investigating how these distributions affect filter efficiencies and pressure drops.

Since fiber size is the most difficult filtration parameter to control in experimental systems, we began our distribution studies by determining the effect of fiber size distributions on filter pressure drop. Pressure drop was selected rather than efficiency because no particle parameters were involved in the equations. The objective of this study was to modify the pressure drop equation for monodisperse fiber sizes to allow for a distribution of fiber sizes.

Pressure Drop Theories

Although a number of pressure drop theories could be used as the basis for our heterodisperse model, we have considered only those theories that agree with Darcy's law, which states the pressure drop per unit flow velocity is a constant at any velocity⁽¹⁾. Theories based on flow around isolated fibers, like the frequently used Lamb's theory, do not agree with Darcy's law and were, therefore, not considered in our investigation. The problem with these theories is their failure to account for the influence of neighboring fibers on flow patterns.

Several models have been developed that include the interaction from neighboring fibers. The most popular of these models, e.g., the Kuwabara⁽²²⁾ and Happel⁽²³⁾ models, are called "cell models" because the streamlines form closed cells around each fiber within the filter mat. Spielman and Goren have developed a flow model wherein neighboring fibers create a dampening force on the fluid passing around each fiber⁽²⁴⁾. This model is less popular than the cell models presumably because Spielman and Goren's complicated equations involve Bessel functions.

* The average of the distribution selected is usually arbitrary because it is based on the device used to generate the distribution. For example, the average fiber size obtained from an electron microscopy study is normally chosen to be a number average without considering whether this average is the correct one for predicting filter efficiency or pressure drop.

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The pressure drop for all the models can be described by Eq. (1)

$$\Delta P = \frac{4\alpha\mu VX}{R^2} \Phi(\alpha), \quad (1)$$

where:

ΔP = pressure drop (Pa)

α = vol fraction of fibers

μ = viscosity (Pa·s) of gas

V = velocity (cm/s) of gas

X = filter thickness (cm)

R = fiber radius (cm)

$\Phi(\alpha)$ = function of α (varies with model)

The function Φ represents a dimensionless pressure drop that has been normalized for all variables except α .

Pressure Drop Theory with Fiber Size Distribution

The pressure drop for a filter mat with a heterodisperse fiber size distribution can be derived by taking weighted averages of Eq. (1) subject to certain constraints. Although the specifics will be presented elsewhere, the physical basis for this derivation can be readily explained. Beginning with a filter mat having a uniform fiber size, one decreases and increases individual fibers until the desired distribution is obtained. The constraint in this process is that the total length of the fibers within the filter remain constant. In terms of the cell models, this constraint requires that the total number of flow cells remain constant. The weighting factors are then the fraction of the total fiber length or the fraction of the total number of flow cells having a given fiber size.

We have used an empirical pressure drop equation obtained by Davies to illustrate how the pressure drop changes for a distribution of fiber sizes⁽²⁵⁾. Davies' equation was chosen over other theories because it is simpler, obeys Eq. (1), and represents experimental data. Although any of the other theories could also be used as a starting point for the heterodisperse model, the resulting equation would be much more complex.

$$\frac{\Delta P}{\mu VX} = 16 \frac{\alpha^{3/2}}{R^2} \quad (2)$$

For a distribution of fiber sizes, we can show that

$$\frac{\Delta P}{\mu VX} = 16 \alpha^{3/2} \frac{\int_0^\infty N(R) R dR}{\left(\int_0^\infty N(R) R^2 dR \right)^{3/2}}, \quad (3)$$

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where $N(R)$ represents the number of fibers having a radius R . This equation is useful for calculating pressure drops when the distribution of fiber sizes is obtained from electron microscopy. By combining Eqns. (2) and (3), we can calculate the effective fiber radius R_{EF} if the number-size distribution is known.

$$R_{EF}^2 = \frac{\left(\int_0^{\infty} N(R) R^2 dR \right)^{3/2}}{\int_0^{\infty} N(R) R dR} \quad (4)$$

However, if the mass distribution of different size fibers is known, Eq. (5) is more useful.

$$\frac{\Delta P}{\mu V X} = 16 \left(\int_0^{\infty} \frac{\alpha dR}{R^2} \right)^{1/2} \int_0^{\infty} \frac{\alpha dR}{R} \quad (5)$$

Equations (3) and (5) are different forms of the same equations.

If the number (or mass) distribution of fibers is known to obey the log-normal distribution, we can integrate Eq. (3) or (5) analytically, yielding

$$R_{EF} = R_{NMR} \exp(1.25 \ln^2 \sigma_g) \quad (6)$$

where R_{NMR} is the number median radius and σ_g is the geometric standard deviation of the distribution.

Comparing Experimental Measurements with Theory

We took a series of electron micrographs of three filter media to determine fiber-size distributions. Figure 4 shows typical electron micrographs of filters AF-4, -11, and -18* at the same magnification. Note the AF-4 filter resembles tangled string while the AF-11 and -18 filters look like log jams. Using a Zeiss counter, we did a detailed size analysis of a large number of fibers and plotted the results on a log-probability graph. Since the data for AF-4 fell on a straight line, they can be represented by a log-normal distribution. The data for AF-11 and -18 did not fall on a straight line; however, a reasonably good straight line fit can be made for the central portion of the distribution. The number median diameters NMD and geometric standard deviations σ_g were obtained from the three lines.

An alternate method of displaying the fiber-size distribution is seen in Fig. 5, where the relative number of fibers per unit-size interval is plotted as a function of fiber diameter. The number median diameter D_{NMD} and the effective diameter D_{EF} (calculated from Eq. 6) are shown on the histogram. When distributions do not obey a standard equation, fiber-size statistics can only be determined from histograms (as shown in Fig. 5) or corresponding tabulated data.

* The filters designed at AF numbers represent commercially available filters from Johns Manville. The suffix number is a relative indication of the fiber size, which increases for larger numbers.



AF-4



AF-11

Figure 4
Electron micrographs of
filters AF-4, -11, and -18.



AF-18

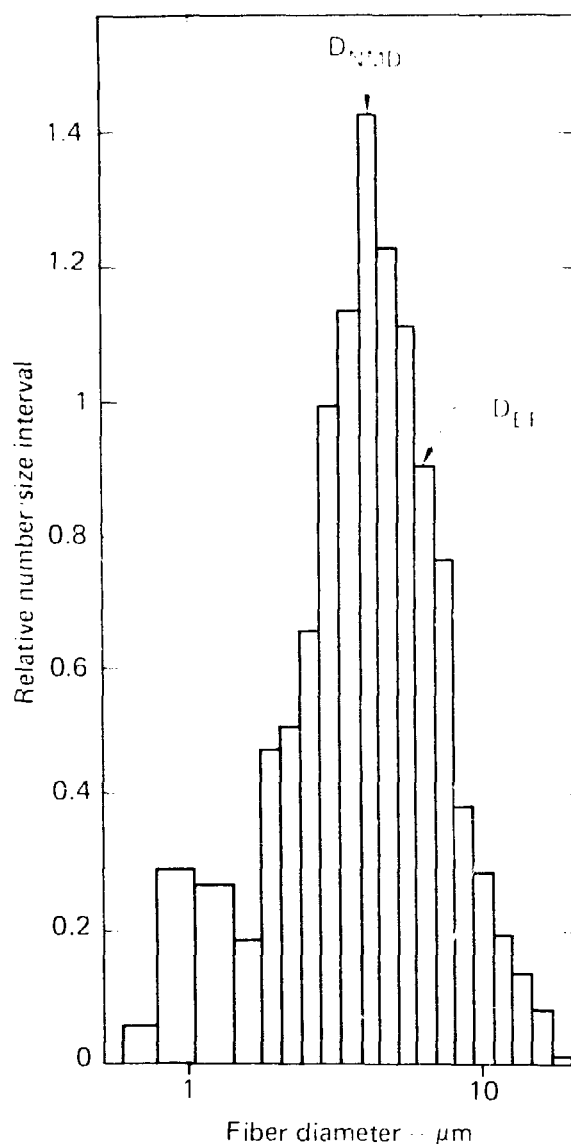


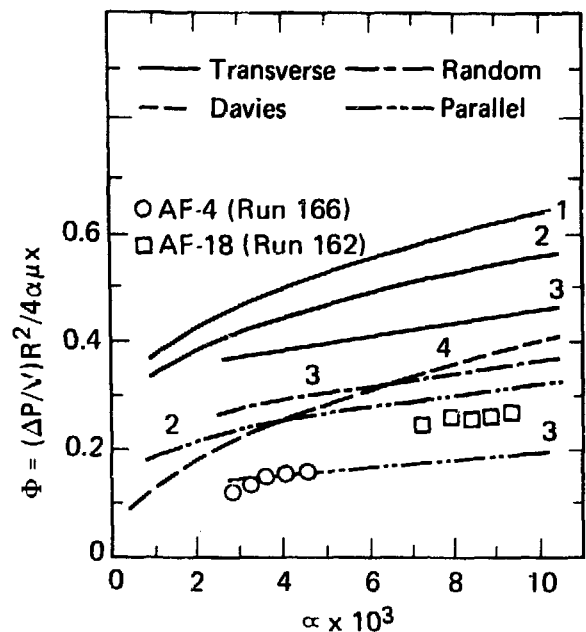
Figure 5

Histogram of number-size distribution as a function of fiber diameter for filter AF-11.

We then measured the pressure drop across the AF-4, -11 and -18 filters at various face velocities to compare the experimental values of Φ to the values predicted by Davies' empirical equation and the three theoretical models. The experimental values were determined from Eq. (3) using the fiber-size distributions obtained from electron microscopy. These data are shown in Fig. 6 as a function of α along with the Φ values from Davies' equation and the three flow theories. The Kuwabara and Happel theories are very similar cell models, while the Spielman and Goren theory is a fluid-dampening model. Curves for these models show fibers oriented transverse and parallel to the air flow. The Spielman and Goren model also shows a curve for a three-dimensional random orientation of fibers with respect to the air flow. As one would expect, the parallel orientation gives the lowest values for normalized pressure drop while the transverse orientation gives the highest values; the curve for the random orientation has an intermediate value. Note the curve representing a three-dimensional orientation almost matches Davies' empirical curve.

Figure 6

Comparison of 1) Kuwabara, 2) Happel, 3) Spielman and Goren, and 4) Davies Empirical flow theories with experimental filters AF-4, and AF-18 at flow rates of 12.3-65.4 cm/s. The normalized pressure drop, Φ , is plotted as a function of fiber volume fraction α .



The circles and squares in Fig. 6 represent experimental measurements at flow rates from 12.3 to 65.4 cm/s for filters AF-4 and -18, respectively. Each set of these data points show that α increases with increased flow velocity. This occurs because the filters compress to smaller thicknesses at higher flow rates, thus increasing the volume fraction of the fibers. If filter thickness remained constant at different flow rates, all points would fall on the same α value.

The most disturbing aspect of the experimental data points is the low values of the normalized pressure drop compared to Davies' empirical equation. The Φ values obtained from Davies' equation for AF-4 and -18 filters are approximately 40% and 28% higher than those obtained in our experiments. Although several possibilities for this discrepancy exist, the primary cause is believed due to the nonuniform distribution of fibers in the filter mat.

An equivalent method for comparing experimental pressure drop measurements with Davies' equation is based on measured and derived fiber diameters. A summary of fiber-size statistics for filters AF-4, -11, and -18 is given in Table II.

Table II. Comparison of fiber diameters calculated from pressure-drop equations

| Filter | D_{NMD} , μm | σ_g | D_{EF} [Eq. (6)], μm | D [Eq. (2)], μm | Increase, % |
|--------|---------------------|------------|-----------------------------|------------------------|-------------|
| AF-4 | 0.73 | 2.0 | 1.34 | 1.78 | 33 |
| AF-11 | 3.3 | 1.7 | 4.70 | 5.12 | 9 |
| AF-18 | 3.5 | 1.7 | 4.98 | 5.86 | 18 |

Using Eq. (6), we calculated the effective fiber diameter for each filter with the D_{NMD} and σ_g obtained from the log-probability graphs. The D_{EF} diameters should, in principle, have agreed with the diameters obtained from Davies' empirical equation,

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Eq. 2, that relates filter pressure drop measurements to fiber radius. However, a comparison of the two diameters in Table II indicates the diameters obtained from Davies' equation were always higher than the corresponding diameters calculated from fiber-size distributions. This discrepancy was seen earlier in Fig. 6 where the experimental Φ values for AF-4 and -11 were considerably lower than the values predicted from Davies' equation. We plan to investigate the cause of the discrepancy in our future studies.

Clean Filter Efficiency With an Electric Field

We have shown that previous investigators were able to greatly improve filter efficiency by applying an external electric field across the filter medium. The increased efficiency was believed to be the result of electrostatic forces arising from polarized particles and polarized fibers and from charged particles and polarized fibers. The former force was called dielectrophoresis by Bogardus et al⁽¹¹⁾.

A careful review of these previous studies has shown that dielectrophoresis makes a negligible contribution to increased filter efficiency. Fielding has acknowledged "the dielectrophoresis effect is minor" in discussing their paper⁽¹¹⁾. We also concluded in a previous study that the dielectrophoresis term is negligible⁽²⁰⁾. These conclusions were based on observations that the logarithm of the penetration was a linear function of the applied electric field rather than of the square, as predicted for dielectrophoresis. Two factors are responsible for dielectrophoresis' negligible contribution to the increase in filter efficiency with an applied electric field: 1) nearly all aerosols are charged, and 2) dielectrophoresis is only important for large particles. Since most filters already have very high efficiencies for large particles, any improvement in filter efficiency would not be seen. In contrast, small charged particles that normally penetrate the filter can be attracted to polarized fibers when an electric field is applied.

The previous studies on electrostatic filters were conducted under steady-state conditions, using noninsulated electrodes. A relatively simple static model developed by Zebel could explain nearly all of the experimental results from these studies⁽²¹⁾. Our previous results on the effect of electric field, face velocity, fiber volume fraction, and fiber size were in good qualitative agreement with the Coulombic force in Zebel's model⁽²⁰⁾.

However, our recent experiments with transient electric fields and insulated electrodes have shown the electrostatic filter is a complex dynamic system involving two electrostatic forces. One is the well-established Coulombic force between charged particles and polarized fibers; the other, while also a Coulombic force, involves charged particles and charged fibers. Although this additional force makes a large contribution to the increase in filter efficiency with an applied electric field, it had not been recognized before our investigation.

Theory of Electrostatic Filter

The electrostatic filter exhibits a very dynamic behavior with two opposing processes: the increasing fiber charge due to charged particles collected on the fibers, and the dissipation of this charge due to the conductivity of the fibers. We have incorporated these dynamic processes into our efficiency model for clean filters. The filter efficiency model used in our investigation is based on the standard single fiber efficiency⁽¹⁾. The differential form of the equation for this model is

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$$-\frac{\partial N}{\partial X} = \frac{2\alpha N \eta_T}{\pi R (1-\alpha)} \quad (7)$$

where:

N = conc (cm^{-3}) of particles

α = vol fraction of fibers

X = filter thickness (cm)

η_T = total single fiber efficiency

R = fiber radius (cm)

Equation (7) describes the fractional decrease in the number concentration, $-dN$, across a differential filter element, dX . The mechanical and electrical filtration mechanisms are incorporated into the total single fiber efficiency η_T .

One commonly used method for combining all the different filtration mechanisms is to calculate the single fiber efficiency from each mechanism separately and add these values together. Unfortunately, this procedure does not yield the correct results because it neglects the interaction terms between different mechanisms⁽¹⁾. Nonetheless, the summation of separate efficiency mechanisms has been used in our theoretical model because it provides trends and gives approximate values for this relatively simple model. We plan to develop a rigorous mathematical model of our electrostatic filter in future investigations.

The total single fiber efficiency used in our model is

$$\eta_T = \eta_M + \eta_{Pq} + \eta_{Qq} \quad (8)$$

where:

η_M = single fiber efficiency for mechanical mechanisms

η_{Pq} = single fiber efficiency for polarized fibers and charged particles

η_{Qq} = single fiber efficiency for charged fibers and charged particles

We have dropped the single fiber efficiency for polarized fibers and polarized particles (dielectrophoresis) used in our previous models because that term was insignificant. The term η_{Qq} represents a new term in our theoretical model and allows the charge to build up and dissipate on the fibers. Replacing η_T in Eq. (7) with Eq. (8) and substituting the expressions for η_{Pq} and η_{Qq} we have:

$$-\frac{\partial N}{\partial X} = \frac{2\alpha N}{\pi R (1-\alpha)} \left\{ \eta_M + \frac{qEC}{6\pi\mu rV} \left(\frac{1 + \frac{\epsilon_f - 1}{\epsilon_f + 1}}{1 + \frac{qEC}{6\pi\mu rV}} \right) + \frac{qQ}{3\pi\mu R rV} \right\} \quad (9)$$

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where:

q = particle charge (C)

E = electric field strength (V/cm)

C = Cunningham slip factor⁽¹⁾

μ = viscosity (Pa·s) of gas

V = velocity (cm/s) of gas

r = particle radius (cm)

ϵ_f = dielectric constant of fiber

Q = fiber charge (C)

The last term in braces represents the term η_{0q} . The fiber charge Q in this term is responsible for the dynamic nature of the electrostatic filter. Equation (10) gives the rate at which the fiber charge increases or decreases:

$$\frac{\partial Q}{\partial t} = -\frac{\gamma Q}{\epsilon} + q \frac{\partial m}{\partial t} \quad (10)$$

where:

γ = fiber conductivity (S)

ϵ = permittivity (F/m) of fiber

m = conc (cm⁻³) of particles in the filter

t = time (s)

This equation shows that increasing the fiber charge increases the rate of charge dissipation. Similarly, increasing the rate of particle accumulation in the filter increases the rate of charge accumulation on the fibers.

The rate of particle accumulation in the filter is given by:

$$\frac{\partial m}{\partial t} = \frac{2\alpha N V \eta_T}{\pi R(1-\alpha)} \quad (11)$$

This equation assumes that previously trapped particles cannot trap new particles.

Using Eqns. (9)-(11) and assuming a steady state for the fiber charge, we can show that

$$\ln P = \ln P_o - \frac{2\alpha q}{3 \pi^2 (1-\alpha) \mu R r} \left\{ \frac{N_o q \epsilon (1-P)}{R \gamma} + \frac{E C X}{2 V} \left(\frac{1 + \frac{\epsilon_f - 1}{\epsilon_f + 1}}{1 + \frac{q E C}{6 \pi \mu r V}} \right) \right\} \quad (12)$$

where:

P = particle penetration with mechanical and electrical mechanisms

P_o = particle penetration with only mechanical mechanisms

N_o = conc (cm⁻³) of particles at filter inlet

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Details of this derivation will be given elsewhere. Equation (12) allows us to calculate the filter penetration for our electrostatic filter under steady-state conditions. Since the filter efficiency equals $1-P$, we can also determine the filter efficiency.

Of the several observations that can be made from Eq. (12), the most prominent is the decrease in filter penetration due to the electrical terms. However, if the aerosols have no charge ($q = 0$), the filter penetration reduces to the penetration involving only mechanical mechanisms. This is an unlikely condition since almost all aerosols, either natural or artificially generated, have a particle charge. Equation (12) also shows that the charged fiber mechanism is favored by increased particle charge, whereas the polarized fiber term is favored by increased electric field. Increasing the air velocity decreases the effect of the polarized fiber mechanism but has no effect on the charged fiber mechanism. Another important observation from Eq. (12) is the decrease in the charged fiber mechanism due to increasing fiber conductivity. In fact, when the fiber conductivity increases sufficiently, the charged fiber mechanism disappears and Eq. (12) reduces to our previous electrostatic equation⁽²⁰⁾. Also, note that the filter penetration, P , occurs in two terms that cannot be combined. Solutions of the penetration in Eq. (12) therefore require numerical methods. However, for high filter efficiencies, $P < 0.1$, the P can be dropped from the right side of Eq. (12) with little error.

Transient Filtration Measurements

The theory of electrostatic filters presented in this paper can be used to explain the major experimental findings of our investigation. The experiments actually preceded our theoretical work and served as the basis for our model. Figure 7 shows the concentration of NaCl aerosols penetrating two electrostatic filters as a function of time. The major difference between the two filters was their conductivity. The filter on the left was relatively nonconducting (10^{10} ohm/cm) while the filter on the right was relatively conducting (10^8 ohm/cm). A dye, added to the filter on the right for identification purposes, was responsible for the increased conductivity. The two filters did not have the same steady-state penetration values because of slightly different fiber size and fiber volume fractions.

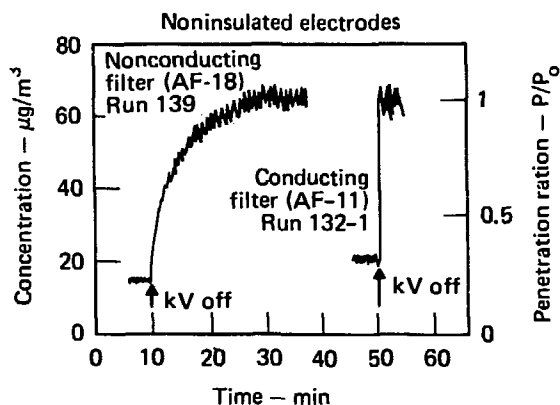


Figure 7

Aerosol concentration measurements downstream of the filter during transient conditions when the electric field is turned off for a nonconducting and conducting filter using noninsulated electrodes.

Figure 7 shows that when the electric field is turned off, the aerosol penetration increases very slowly for the nonconducting filter and very fast for the conducting filter. As these observations involve transients, we cannot interpret them using Eq. (12) because it only applied to steady-state conditions. The complicated transient equations will be presented elsewhere.

We can still make a qualitative explanation of the two transients using Eq. (10). For the nonconducting filter (small γ), a significant charge can build up on the fiber before the charge dissipation term ($-\gamma Q/\epsilon$) reaches equilibrium with the charge accumulation term ($q\partial m/\partial t$). However, the charge dissipation term for the conducting fiber (large γ) is so large that equilibrium between charge dissipation and accumulation is established with a negligible charge on the fibers. Since the conducting fiber cannot develop a charge, the penetration will instantly increase to the P_0 level when the electric field is turned off. In contrast, the nonconducting filter has a significant fiber charge which bleeds off very slowly when the electric field is turned off. As a rough approximation, the accumulation term in Eq. (10) becomes zero* and the resulting equation is integrated to yield

$$Q = Q_0 e^{-\frac{\gamma t}{\epsilon}}, \quad (13)$$

where Q_0 is the fiber charge before the electric field is removed. Although the polarized fiber mechanism disappears immediately when the electric field is removed, the charged fiber mechanism persists until all of the charge bleeds off.

In a similar experiment, we measured transient aerosol penetrations for the same nonconducting filter using electric fields generated by ac (60 Hz) and dc voltages. Figure 8 shows dc and ac electric fields generate the same filter behavior when the field is turned off as the nonconducting and conducting filters in Fig. 7, respectively. Although the results in Fig. 8 were obtained with insulated electrodes, similar results were obtained with noninsulated electrodes. The explanation for the transient behavior using a dc field was given previously for the nonconducting filter in Fig. 7. A comparison of the results using an ac field in Fig. 8 with those from the conducting filter in Fig. 7 suggests the same explanation would apply in both cases. This is not the case, since the conducting filter in Fig. 7 cannot support a fiber charge whereas the nonconducting filter in Fig. 8 can. However, the instantaneous increase in filter penetration for both cases when the electric field is shut off suggests that neither filter has developed a fiber charge.

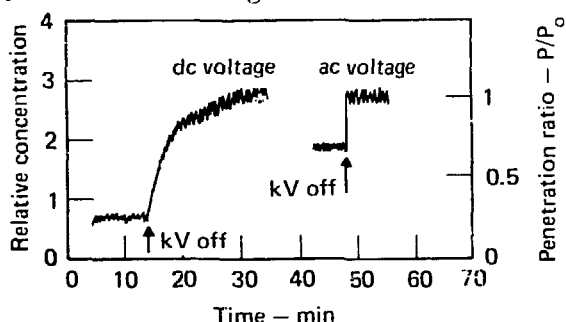


Figure 8

Aerosol concentration measurements downstream of a nonconducting filter during transient conditions when the electric field is turned off for a dc and an ac field using insulated electrodes.

We believe this behavior is due to the NaCl aerosols, which have equal positive and negative charges. When the electric field is in one direction, aerosols of a given charge are trapped on the fibers. When the electric field is reversed, aerosols of the opposite charge are deposited on the same fiber location, thereby cancelling the first charge. Repeated field reversals will continue this charge cancellation and, therefore, never develop a net charge on the fiber. Figure 8 also shows the aerosol penetration of a filter with an ac field is much higher than with a dc field. The higher penetration is due to the absence of the charged fiber mechanism, which lowers the penetration [see Eq. (12)].

* In a rigorous analysis, the accumulation term in Eq. (10) does not reduce to zero because it is a function of the fiber charge. This results in a longer time for the charge to bleed off.

The preceding explanation with ac fields provides a brief glimpse of the complex dynamics of the electrostatic filter. Except for the case involving ac fields, we have ignored the aerosol and fiber charge and have assumed in our simple model that we always have attractive forces. However, in a real electrostatic filter, both the aerosols and fibers have a distribution of plus and minus charges. Depending on the conductivity of the fiber, these charges may migrate and cancel. Clearly both attractive and repulsive forces are involved in this system.

Effect of Fiber Conductivity and Electrode Insulation on Filter Efficiency

The conductivity of the fiber is an important variable in electrostatic filtration since it determines the amount of charge that can be accumulated on the fibers and the associated increase in filter efficiency. In field applications, the fiber conductivity may increase as particles load up the filter or acid mists coat the fiber surface. Both effects may lower filter efficiency. The effect of fiber conductivity on filter efficiency also depends on the insulation of the high-voltage electrodes. Experimental results can only be interpreted when the effects of both fiber conductivity and electrode insulation are included in our theoretical model.

Figure 9 shows the results from efficiency tests on a conducting filter using both insulated and noninsulated electrodes. The logarithm of the penetration ratio is plotted against the applied electric field to linearize the resulting curve and eliminate the initial filter efficiency with no external field. A larger value of $-\ln(P/P_0)$ means a greater filter efficiency. We see that increasing the electric field improves the filter efficiency when noninsulated electrodes are used but has no effect when insulated electrodes are used.

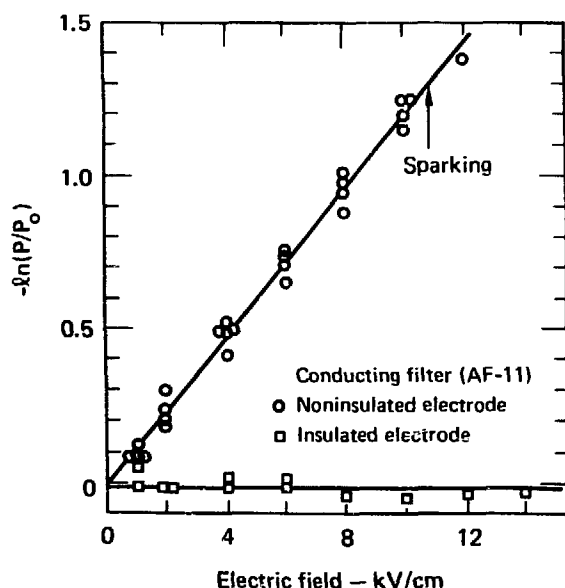


Figure 9

Effect of electrode insulation on the enhanced filtration for a conducting filter.

For tests on a less conductive filter (Fig.10), both insulated and noninsulated electrodes show an increase in filter efficiency when the applied electric field is increased. However, filter efficiency is lower when an insulated electrode is used instead of a noninsulated electrode. Based on Figs. 9 and 10, we would expect that in the limit of zero conductivity, the filter efficiency would be the same for either insulated or noninsulated electrodes.

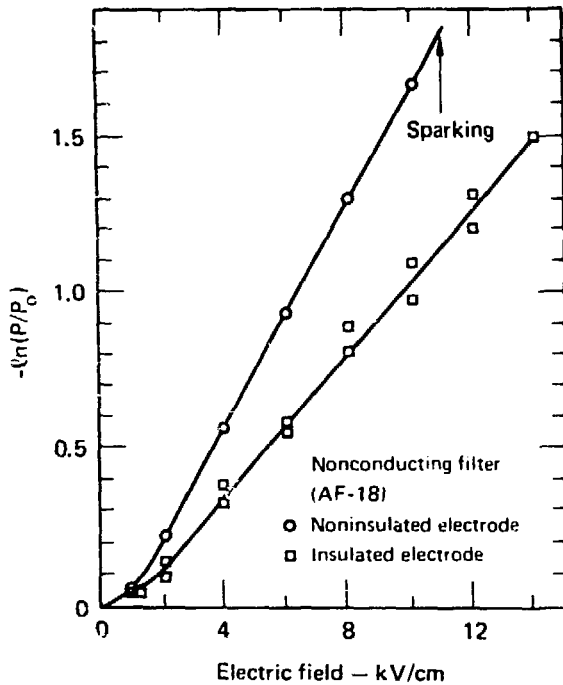


Figure 10

Effect of electrode insulation on the enhanced filtration efficiency of a nonconducting filter.

The conductivity of the fibers does not have to be very large for the filter to behave as seen in Fig. 9. The resistivities of the conducting and nonconducting filters were 10^8 and 10^{10} ohm/cm, respectively. Figures 9 and 10 also show that sparking across the electrodes occurs at 11 kV/cm for the noninsulated electrodes while no sparking occurs for the insulated electrodes up to 14 kV/cm.

The experimental results shown in Figs. 9 and 10 can be explained in terms of a charged fiber mechanism, using a schematic of our electrostatic filter system shown in Fig. 11. This figure shows a filter mat of fibers sandwiched between two electrodes that are connected to a high-voltage source. The high voltage produces an electric field E_0 within the filter bed that polarizes the individual fibers.

Charged aerosol particles will then be attracted to the polarized fibers and, if the fibers have a low conductivity, accumulate a net charge on the fibers. However, since all materials have a finite conductivity, the charge on the fibers will migrate in the direction of the electric field and dissipate over a time period according to Eq. (13). The positive and negative charges will migrate along the fibers to the electrode of opposite charge and build up an induced charge as shown in Fig. 11.

If the electrodes are not insulated, these induced charges will be neutralized on contact with the electrodes. However, if the electrodes are insulated, the induced charge cannot be neutralized. These induced charges will then generate an electric field that is opposed to the external field created by the high-voltage electrodes. The net effect is a reduction of the electric field within the filter medium.

The extent of this field reduction depends on the magnitude of the induced charge. For the filter shown in Fig. 10, the low conductivity allows only a small charge to build up next to the insulated electrode. This small charge buildup generates a small opposing electric field that consequently reduces filter

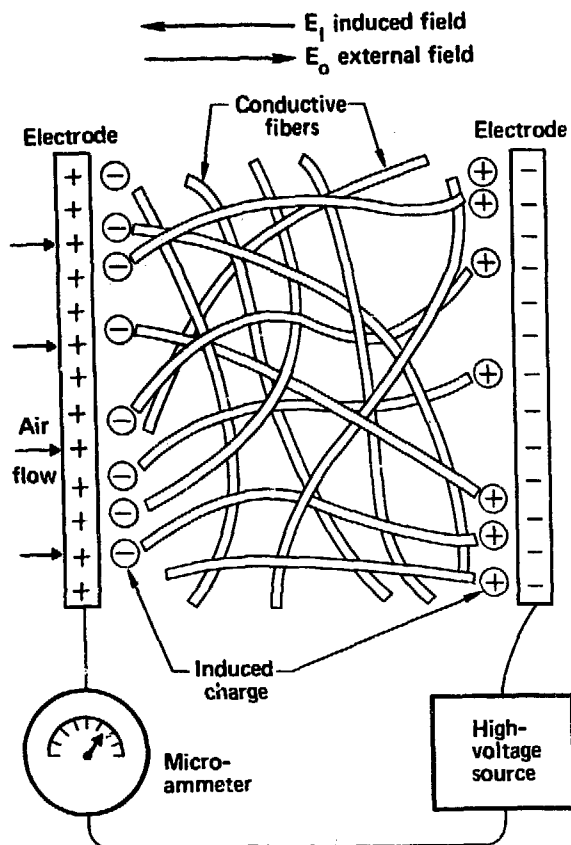


Figure 11

Schematic of the induced charge build-up that cancels the applied electric field when a conducting filter is used with insulated electrodes.

efficiency by only a small amount. In contrast, the conducting filter shown in Fig. 9 allows a significant charge to build up next to the insulated electrodes. The magnitude of this induced charge is large enough to cause a net cancellation of the external electric field within the filter medium. An applied electric field in this case does not increase filter efficiency.

Filter-Loading Model

We recently developed a filter-loading model⁽²⁸⁾ that avoids the problem of determining the detailed capture mechanism of particles in a loaded filter⁽²⁷⁾. Our approach is an extension of the theoretical treatment of filter efficiency and pressure drop for unloaded filters. It treats captured particles within the filter as new fibers. Filter efficiency and pressure drop are then derived in the same manner as for clean filters except that additional fibers are included to represent trapped particles. Details of the derivation will be presented elsewhere.

The expression for the concentration of particles penetrating a filter is given by

$$N = \frac{N_0 \exp - \left(\frac{2\alpha X \eta_F}{\pi R(1-\alpha)} + \frac{\pi V_{tr}^2 N_0 \eta_p}{2} \right)}{1 - \frac{\pi V_{tr}^2 N_0 \eta_p}{2} \exp - \left(\frac{2\alpha X \eta_F}{\pi R(1-\alpha)} + \frac{\pi V_{tr}^2 N_0 \eta_p}{2} \right)} \quad (14)$$

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where:

N = conc(cm^{-3}) of particles penetrating filter

N_0 = conc(cm^{-3}) of particles entering filter

α = vol fraction of fibers

X = filter thickness (cm)

η_F = single fiber efficiency

R = fiber radius (cm)

V = velocity (cm/s) of air

t = time (s)

r = particle radius (cm)

η_p = collection efficiency for one particle by previously trapped particles

Equation (14) can, without too much error, be approximated by the following equation:

$$N = N_0 \exp - \left(\frac{2\alpha X \eta_F}{\pi R(1 - \alpha)} + \frac{\pi V t r^2 N_0 \eta_p}{2} \right), \quad (15)$$

or, in terms of penetration (P),

$$P = P_0 \exp - \left(\frac{\pi V t r^2 N_0 \eta_p}{2} \right), \quad (16)$$

where $P = N/N_0$ at time t and $P_0 = N/N_0$ at time 0.

Since experimental data showing filter efficiency as a function of the mass of trapped particles are often available, the appropriate theoretical equation should relate efficiency or penetration to particle mass. This relationship is obtained from a particle mass balance. The rate of particle mass accumulation in the filter equals the rate of particle mass entering the filter times the filter efficiency or

$$\frac{dm}{dt} = (N_0 V A m_p) E, \quad (17)$$

where:

m = mass (g) of particles trapped in filter

A = filter area (cm^2)

m_p = mass (g) of a single particle

E = filter efficiency ($E = 1 - P$)

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The mass of particles trapped after a given time can be obtained by integrating Eq. (17) with respect to time and by using Eq. (16) for penetration. The result is

$$m = \left(\frac{8\rho l A}{3\eta_p} \right) (-\ln(P/P_0) + P - P_0) \quad (18)$$

where ρ = particle density (g/cm^3). If ψ represents the term $-\ln(P/P_0) + P - P_0$, then experimental data plotted as ψ vs m should fall on the same curve for any filter media and experimental conditions provided the aerosol properties remain constant.

If we consider a filter loaded with particles as a mixture of two types of fibers, one representing the original clean fibers and the other representing the trapped particles, we can use Eq. (5) to describe the pressure drop across loaded filters. The effect of neighboring fibers or particles is already included in Eq. (5). If we further assume all the original filter fibers are the same size and that trapped particles (now treated as fibers) also have a constant size, we can reduce Eq. (5) to

$$\frac{\Delta P}{\mu V X} = 16 \left(\frac{\alpha_F}{R^2} + \frac{\alpha_P}{r^2} \right)^{1/2} \left(\frac{\alpha_F}{R} + \frac{\alpha_P}{r} \right) \quad (19)$$

where:

- α_F = vol fraction of original fibers
- α_P = vol fraction of trapped particles
- R = radius (cm) of original fibers
- r = radius (cm) of particle fiber

Note the radius (r), of the particle fiber is not necessarily the same as the single particle radius. Equation (19) can be directly compared with experimental data.

Experimental Evaluation of Model

Although much additional work is still needed to develop accurate theoretical models of filter loading, our present model agrees reasonably well with experimental data, as can be demonstrated with the data shown in Fig. 12, where filter efficiency and pressure drop are plotted as a function of the particle mass trapped in the filter. These data were obtained in our small-scale test system, using sodium chloride aerosols. The filter media for both data sets in Fig. 12 is the same. The primary difference between the two is the airflow velocity used to load up the filter.

To test the accuracy of our theoretical equations, we plotted the experimental data as a dependent function of particle mass loading. The particular dependent function used was obtained from Eq. (18) for comparisons of filter efficiencies and from Eq. (19) for comparisons of filter pressure drops. A perfect agreement between theory and experiment was reached whenever curves of the particular dependent function vs particle mass loading coincided for widely different experimental conditions.

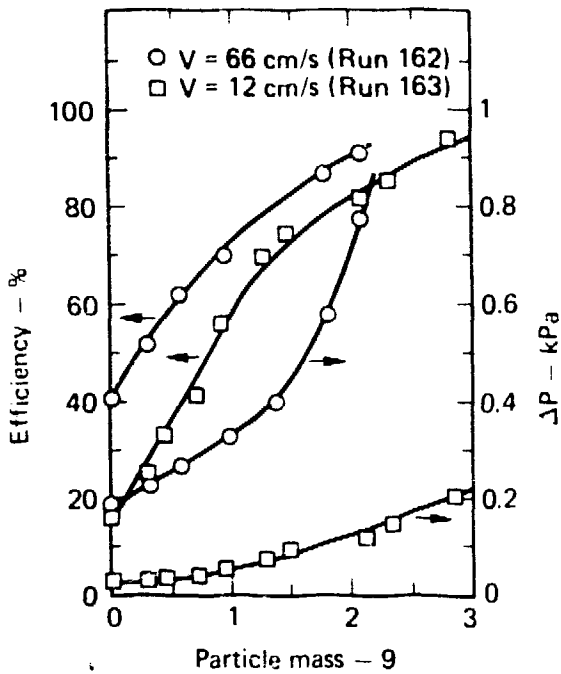


Figure 12

Efficiency and pressure drop as a function of particle mass loading for filter loading tests at different flow velocities.

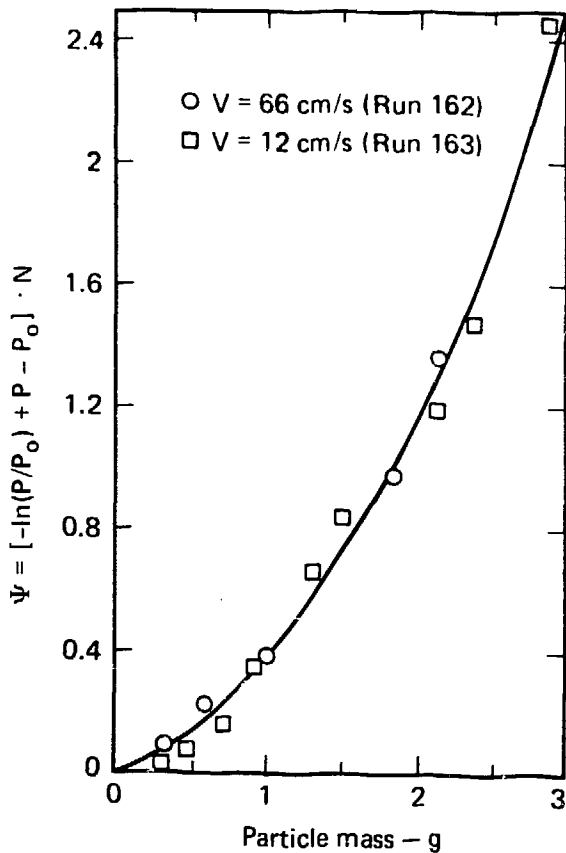


Figure 13

Normalized penetration Ψ as a function of particle mass loading for filter loading tests at different flow velocities.

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The efficiency data for the two experiments shown in Fig. 12 are shown re-plotted in Fig. 13, with the dependent variable ψ as a function of the particle mass. We multiplied the ψ values of the two experiments with a factor N to account for changes in particle-particle collection efficiency, η_p . The excellent agreement between the normalized ψ values seen in Fig. 13 indicates the accuracy of Eq. (18) for predicting filter efficiency values at different mass loadings.

A similar analysis of the pressure drop for the two experiments shown in Fig. 12 could be performed with Eq. (19). We plotted the data as $(\Delta P - \Delta P_0)/V$, where ΔP_0 is the pressure drop across the filter with zero mass loading, so the resulting curve would pass through zero at zero particle mass loading. The resulting data in Fig. 14 show good agreement between the two experiments. An even better fit could be obtained by normalizing both sets of data to the same value of filter thickness (X) and fiber volume fraction (α_F); however, the normalization is complicated by the compression of the filter at higher flow rates. This compression increases the value of $\Delta P/V$ by decreasing the filter thickness and increasing the fiber volume fraction.

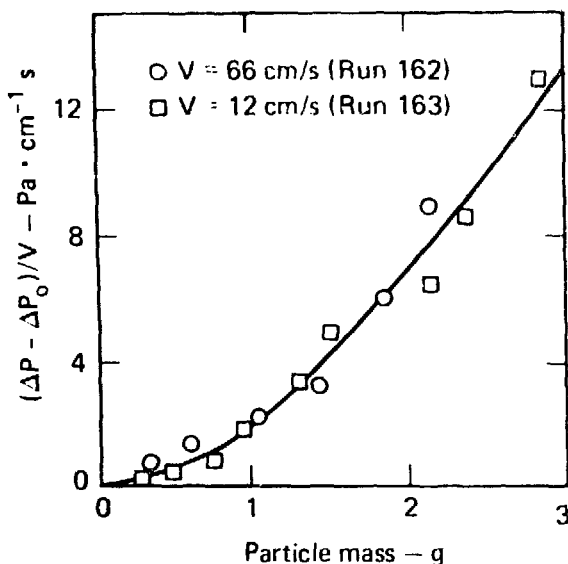


Figure 14

Pressure drop increase per unit flow velocity as a function of particle mass loading at different flow velocities.

Another illustration of the accuracy of Eq. (19) to predict the pressure drop for loaded filters can be made with experimental data obtained by Adley and Wisheart(29). They measured the pressure drop across a HEPA filter as a function of particle loading for three particle sizes. Their results are shown in Fig. 15(a). The equations developed to describe the very loose filters used in the Enhanced Filtration Program should be equally applicable for characterizing HEPA filters. The primary difference between HEPA filters and loosely packed filters is the higher fiber volume fraction (α_F) and the smaller fiber size (R) for HEPA filters. This difference allows us to simplify Eq. (19) by making a Taylor series expansion of the square root term and dropping small terms.

$$\frac{\Delta P}{\mu V X} = 16 \left(\frac{\alpha_F}{R^2} \right)^{1/2} \left(\frac{\alpha_F}{R} + \frac{\alpha_p}{r} \right). \quad (20)$$

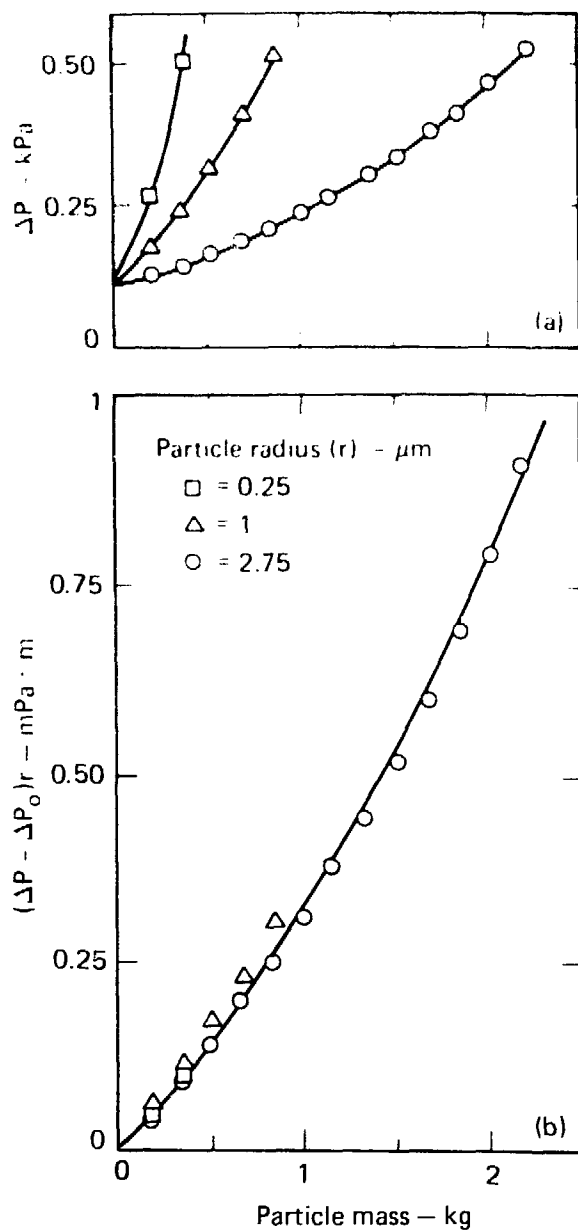


Figure 15

- (a) Loading test on HEPA filters using different size particles.
- (b) Loading data replotted to normalize pressure drop increase for different size particle diameters.

Then, we can subtract the initial pressure drop to yield

$$\frac{\Delta P - \Delta P_0}{\mu V X} = 16 \frac{\sqrt{a_1} a_p}{R r} \quad (21)$$

Equation (21) predicts the increased pressure drop due to particle loading will be inversely proportional to the particle size r . A plot of $(\Delta P - \Delta P_0)r$ will, therefore, be independent of the particle size and will increase in direct proportion to the particle loading or a_p . The data from Fig. 15(a) were replotted in this fashion in Fig. 15(b), where one can see all the data fall on a nearly straight line, thus confirming the validity of Eq. (21).

One series of experiments in our filter loading program is aimed at understanding how airflow velocity and the mass of trapped particles affect measured pressure drop. Figure 16 shows experimental pressure drop measurements versus particle mass at different air velocities for an AF-18 filter. Note the data in Fig. 16 were obtained in the same experiment (Run 162) shown in Fig. 12. However, the pressure drop measurements in Fig. 12 were taken at only one flow rate while Fig. 16 shows a complete map of the pressure drop resulting from various particle masses and flow rates.

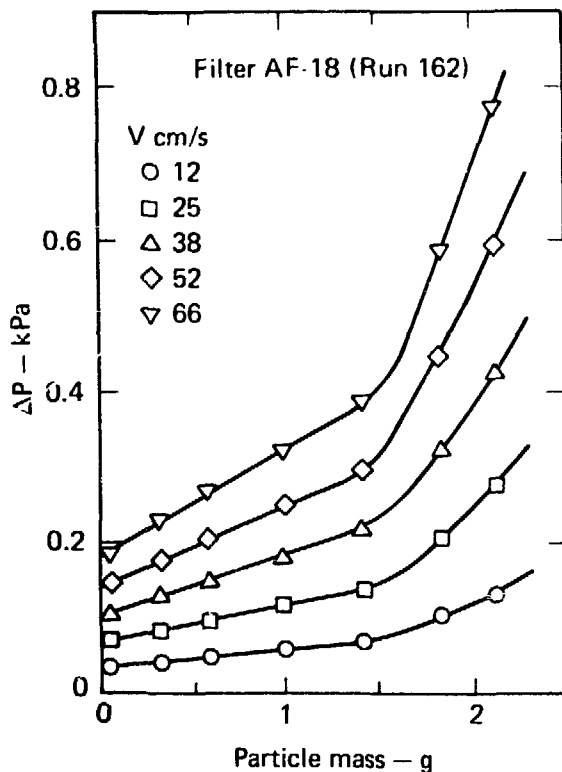


Figure 16

Pressure drop for filter AF-18 as a function of particle mass loading at various flow velocities.

If the same data are replotted as $\Delta P/V$ in Fig. 17, the primary influence of air flow velocity is removed. This agrees with Darcy's Law, which predicts that $\Delta P/V$ is a constant independent of the airflow rate⁽¹⁾. However, a detailed analysis of the data indicated $\Delta P/V$ increased slightly as the velocity increased. Moreover, this increase became greater for larger particle loadings. The reason for the observed increase in $\Delta P/V$ values with increasing velocity is primarily due to the compression of the filter at higher flow rates.

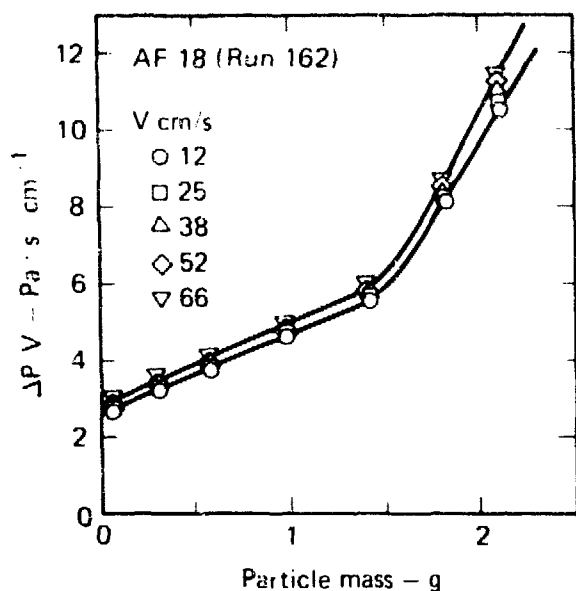


Figure 17

Pressure drop per unit flow velocity for filter AF-18 as a function of particle mass loading at various flow velocities.

Morphology of the Particle Deposits

Another important aspect of our filter loading study is the use of electron microscopy to characterize the particle deposit within the filter. We plan to study the morphology of the particle deposit under various conditions to discover if there is any relationship between the macroscopic observable properties and the microscopic nature of the deposit.

Figure 18 shows electron micrographs of the particle deposit on the top surface of an AF-18 filter, corresponding to a particle loading at 66 cm/s. The macroscopic properties of filter efficiency and pressure drop were seen earlier in Fig. 12.

Figure 19 shows electron micrographs taken on the surface of an AF-4 filter loaded with aerosols at a 66 cm/s flow velocity. Comparison of this figure with Fig. 18 shows the effect fiber size has on particle deposition.

The main difference between the particle depositions shown in Figs. 18 and 19 is the depth of the deposition. Although nearly the same mass of NaCl aerosol was trapped in both filters, the AF-4 filter had a surface deposition while the AF-18 filter had a major deposition at some depth within the filter. The deposition on HEPA filters would be similar to that seen in Fig. 19 since the fiber sizes would be comparable. Our theory predicts the concentration of deposited aerosols will be highest on the front face of the filter and will decrease exponentially with increasing filter depth. To account for the differences in particle deposition, we must add a sticking factor to our single fiber efficiencies⁽¹⁾.

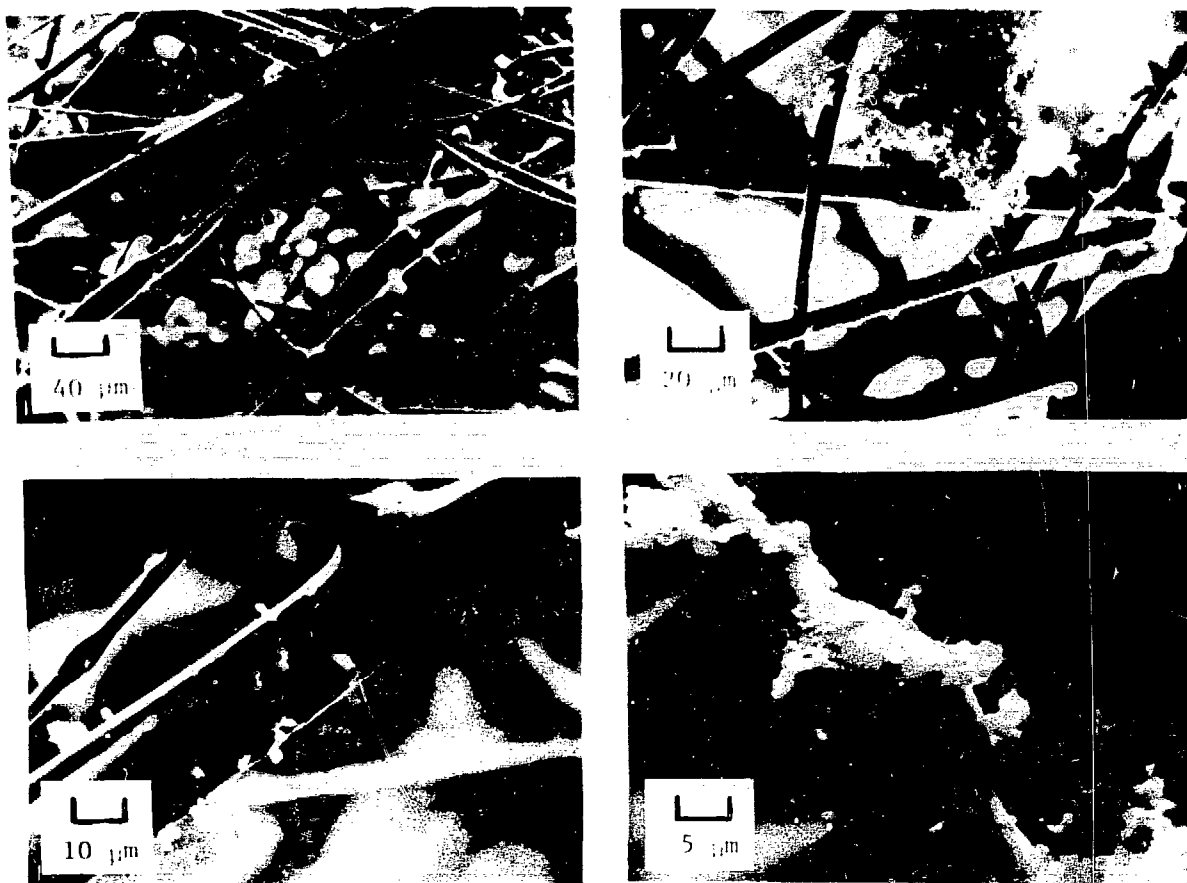


Figure 18 Electron micrographs of particle deposits on a AF-18 filter from a loading test conducted at a 66-cm/s flow velocity.

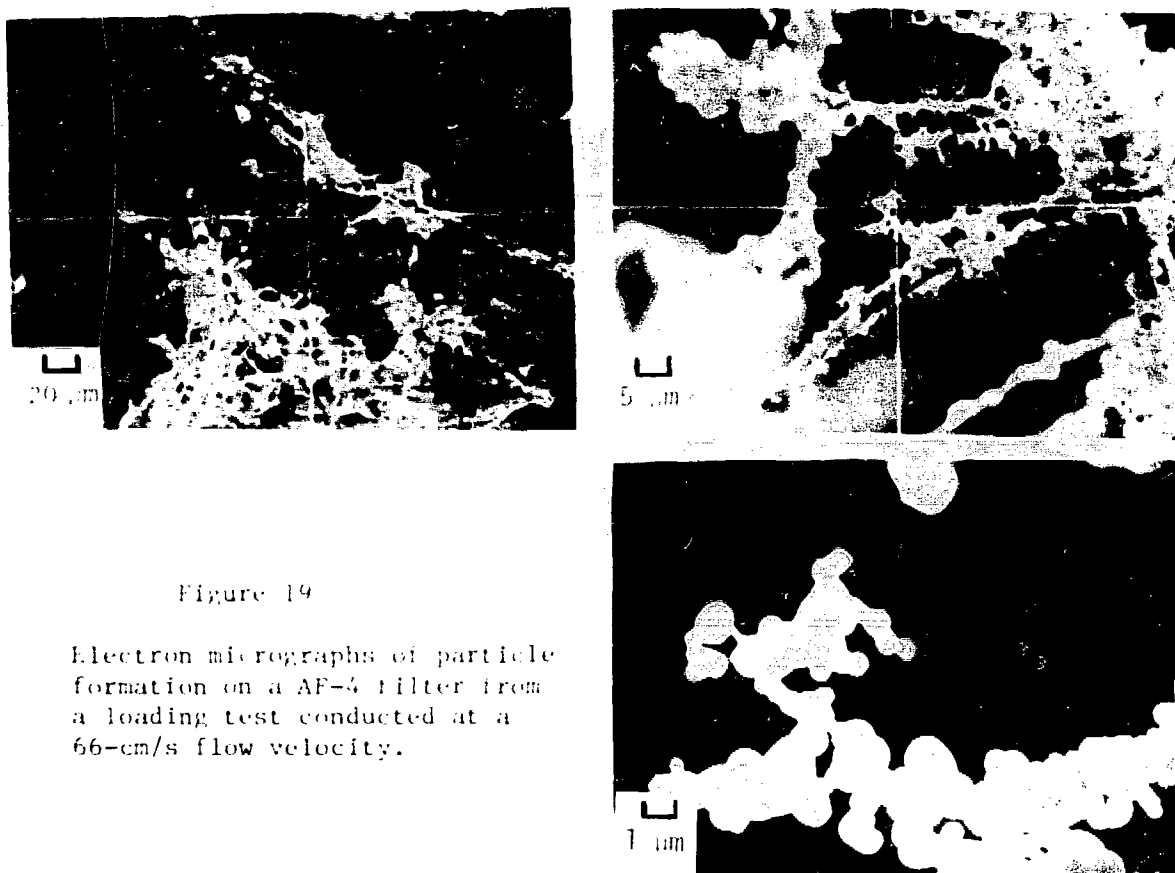


Figure 19

Electron micrographs of particle formation on a AF-4 filter from a loading test conducted at a 66-cm/s flow velocity.

Particle Loading Experiments Using Electric Fields

Test results from a preliminary filter-loading test with a 10-kV/cm applied electric field are shown in Fig. 20. Here an AF-18 filter was sandwiched between the front and rear screens used to generate the electric field. This figure shows three curves; one represents filter efficiency with an applied electric field of 10 kV/cm, one represents filter efficiency with no electric field, and the third represents pressure drop across the filter. We plotted these curves as a function of time instead of particle mass because we could not measure the mass until the end of the test.

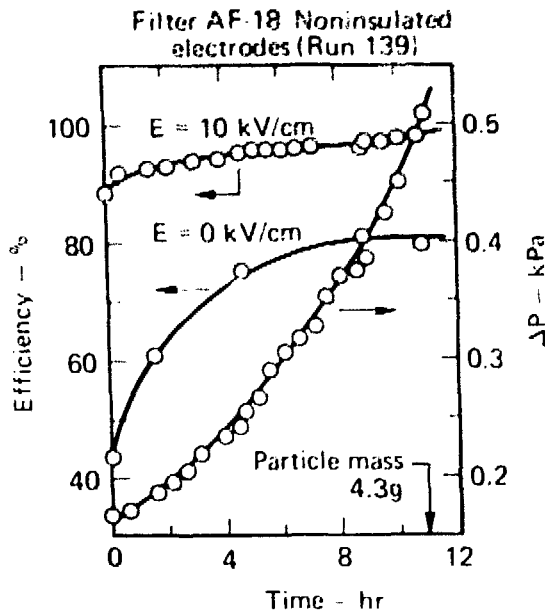


Figure 20

Filter loading test on an AF-18 filter using noninsulated electrodes to generate an electric field.

We have conducted three tests on the AF-18 filter to determine the effect of electric field and electrode insulation on the particle loading. These test results are summarized in Table III. All of these tests had the same initial pressure drop and were terminated when the pressure drop reached 500 Pa. The particle mass collected by the filters is shown in the last column.

Table III. Electric field and electrode insulation effects on filter efficiency and particle mass loading for the AF-18 filter.

| Test | Efficiency at zero particle mass loading, % | Efficiency at $\Delta P = 500 \text{ Pa}$, % | Particle mass loading at $\Delta P = 500 \text{ Pa}$, g |
|---------------------------------|---|---|--|
| No electric field | 41 | 84 | 1.70 |
| 10-kV/cm insulated electrode | 82 | 95 | 2.45 |
| 10-kV/cm noninsulated electrode | 89 | 98 | 4.30 |

Several important trends are seen in Table III. The filter efficiency increased as particles loaded the filter for all cases. Applying an electric field increased the filter efficiency at all particle loadings for both insulated and non-insulated electrodes. The slightly lower efficiency seen for the insulated electrodes resulted from the slight cancellation of the applied electric field by the induced charge as previously seen in Fig. 11.

Using theory presented in this paper, we are able to explain the increase in filter efficiency with particle loading and with an applied electric field. However, we have not yet developed a theory to explain the significantly higher loading capacity for a filter with an applied electric field. Instead of comparing the particle loadings at a given pressure drop, we can compare the pressure drops at a given particle mass loading. Such a comparison shows the electrostatic filter has a

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significantly lower pressure drop than the same filter with no electric field. Previous investigators have also reported significantly lower pressure drops with other electrostatic filters^(12,15,16).

We believe the reason for the lower pressure drop resulting from an electric field is due to a less restrictive particle deposit on the fibers. Since the electric field attracts particles to the fibers, the particle deposit would also tend to build up along the fibers. In contrast, ordinary filters do not have this electric force attracting the particles to the fibers. The particle deposit would thus form a greater number of dendrites to bridge between the fibers and block the air passage. Figure 21 illustrates this hypothesis. We are currently taking electron micrographs of particle deposits with and without an electric field at various stages of particle loading. Once we determine the nature of the particle deposit, we will then develop a model to explain the lower pressure drop for electrostatic filters.

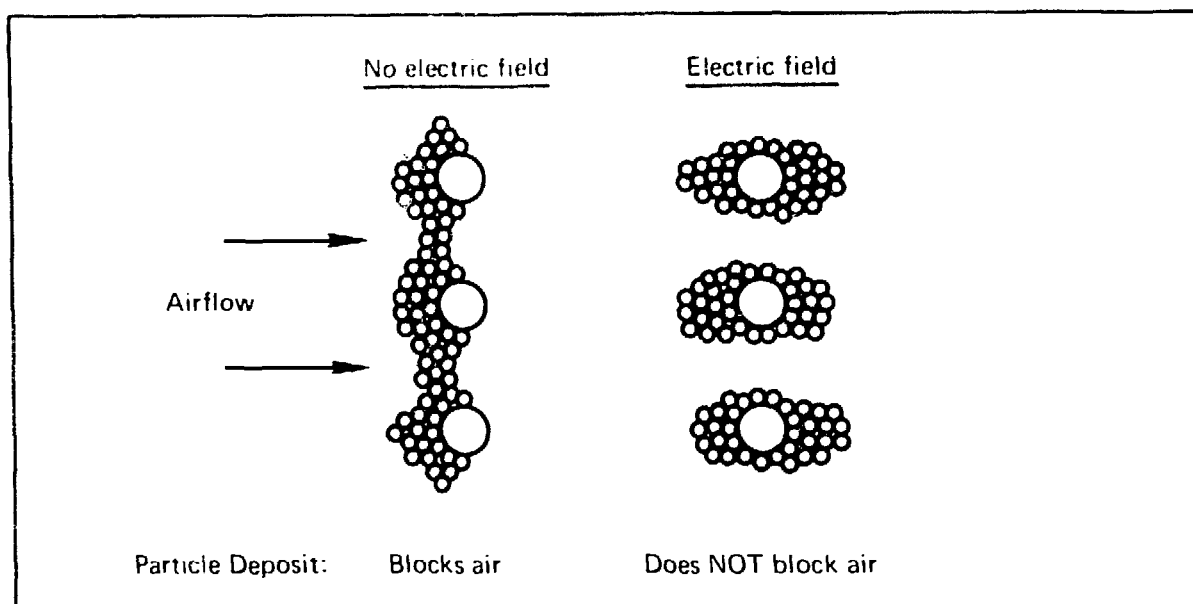


Figure 21 Schematic of possible explanation for decreased pressure drop observed when electric fields are used to enhance filtration.

IV. Application of the Electrostatic Filter in the Nuclear Industry

The objective of LLL's Enhanced Filtration Program is to develop an enhanced filtration system for HEPA filters that will extend their service life. To meet this objective, a large fraction of our effort during the past year has been devoted to developing practical hardware that can be evaluated in field environments. We have two prototype units under development: one for glove box installations and the other for installations in ventilation systems.

Application of electric filter concepts to glove boxes was not an original objective; however, the advantages from this spin-off contribute significantly to the management of radioactive waste and improve our ability to control radioactive particles at their source. Figure 22 shows a detailed schematic of our first prototype electric filter installed in a glove box. This prototype unit consists of a polyethylene main body supported near the ceiling of the glove box, a high-

voltage electrode mounted within the main body, a grounded electrode connected to a lever arm on the main body, and a handle that opens and closes the filter assembly for changing filters. Figure 22 shows the fiberglass filter medium in the filter holder; a tab allows for easier filter replacement.

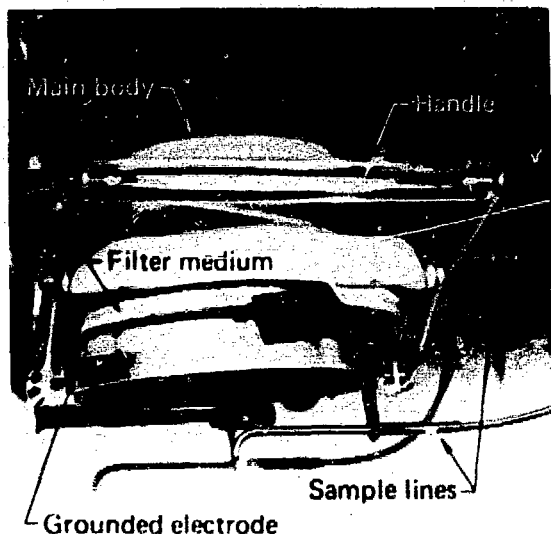


Figure 22

Prototype electric filter holder installed in glove box.

The design of the filter unit shown in Fig. 22 gives it several advantages over a conventional HEPA filter design. First, and most important, this prototype unit facilitates filter change. Figure 23 shows an operator removing the filter medium. Filter replacement takes about 30 s, can be done with one hand, and does not interrupt work inside the glove box. (We have assumed exhaust from the glove box is filtered by a HEPA filter at all times before being discharged into the atmosphere.)

The glove box unit's other advantages include a reduction of the contamination hazard normally encountered with filter changes and the possibility of recovering material from the filter. Use of this filter unit should also result in a reduction of nuclear waste volume since the typical filter medium has over 99% empty space and can be easily reduced in volume.

We have also examined the safety features of the prototype glove box filter and have incorporated the following features in it to reduce the possible high-voltage hazard:

- All high-voltage lines and electrodes have been well insulated to prevent current leakage.
- An interlock mechanism has been placed on the filter holder to shut off the high voltage when the filter holder is opened to replace filters.
- The high-voltage power supply has been set to automatically shut off when the current exceeds a preset level.

These prototype filter units will be installed in a number of glove boxes for radioactive material handling to evaluate their performance under field conditions.



Figure 23

Operator removing filter medium from the filter unit.

We have completed evaluating the electrostatic filter installed in a glove box used to grind chunks of UO_2/BeO into a fine powder. By taking filter samples of the aerosols before and after the electrostatic filter and by measuring the activity on a proportional counter, we determined filter efficiency. Beryllium analysis on the same filter samples gave us identical results. Figure 24 shows the sample filters mounted in a specially designed chamber that allowed us to measure filter efficiencies without contaminating the environment.

The efficiency of the electrostatic filter with no particle loading is shown in Fig. 25 for both UO_2 and NaCl aerosols as a function of the applied electric field. The tests with NaCl aerosols were conducted in a different glove box under controlled laboratory conditions and provided a reference for the field evaluation. Since efficiency measurements of the electrostatic filter with UO_2 aerosols would be determined using in-line filters, we used both in-line filters and a flame photometer in these reference tests. Figure 25 shows good agreement between these two measurement techniques. The NaCl tests also agreed with previous tests generated in our small-scale system in Fig. 1.

Compared to the NaCl tests, the measurements with UO_2 aerosols showed higher efficiencies and had considerable scatter. We found that the variation in filter efficiency was due to different operations within the glove box. Low efficiencies were obtained during grinding operations, and high efficiencies were obtained during powder handling operations. The variation in filter efficiency was probably due to differences in the aerosol properties during the different operations.

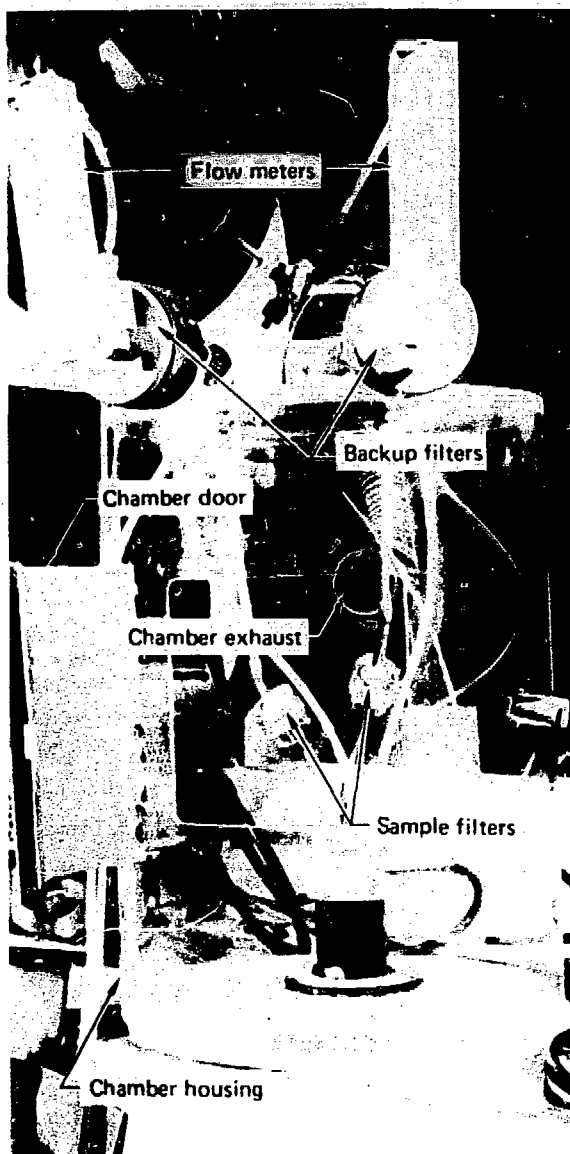


Figure 24
Sampling chamber containing filters
for efficiency measurements.

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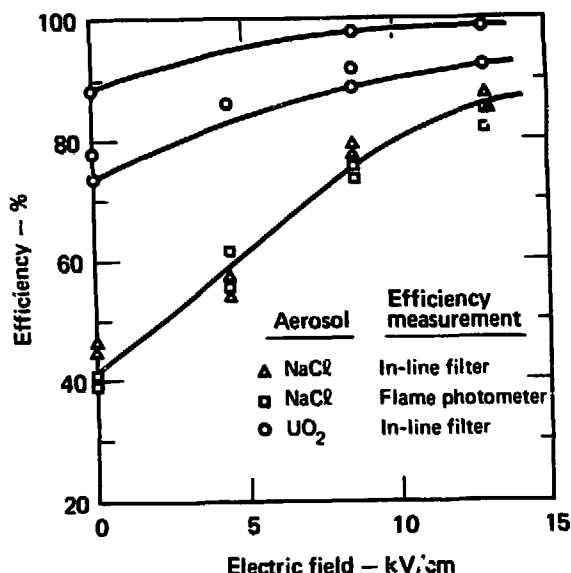


Figure 25

Efficiency of the prototype glove box filters for NaCl and UO₂ aerosols as a function of the applied electric field.

We measured the size distribution of the UO₂ aerosols with an Andersen impactor and found the activity median aerodynamic diameter (AMAD) was 5.4 μm . Figure 26 shows the size distribution of the UO₂ and NaCl aerosols with the cumulative percent of mass or activity plotted as a function of the aerodynamic diameter. The NaCl distribution was determined from mass measurements, while the UO₂ distribution was determined from activity measurements. The larger size of the UO₂ aerosols accounts for the higher efficiency compared to the NaCl aerosols in Fig. 25.

The effect of the electric field on filter efficiency is hard to see in Fig. 25 because both the mechanical and electrical filtration mechanisms contribute to the observed efficiencies. Figure 25 implies that the electric field has a much greater effect on the NaCl test than on the UO₂ test. However, replotting the data for UO₂ and NaCl as $\ln(P/P_0)$ versus the electric field would remove the mechanical mechanisms and show the electric field has the same effect on both aerosols. The decrease in the average filter penetration is 72% for both UO₂ and NaCl aerosols when the electric field is increased from 0 to 12.8 kV/cm.

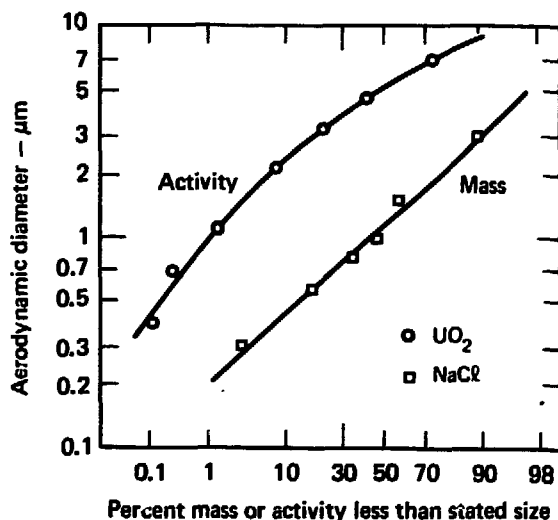


Figure 26

Particle size distribution for UO₂ and NaCl aerosols.

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The same decrease in filter penetration for both UO_2 and NaCl aerosols with increasing electric field appears to contradict our previous statement that the electric field (via dielectrophoresis) has no measurable effect on the filtration of large particles. This apparent contradiction can be resolved by noting in Fig. 25 that 80-90% of the UO_2 particles are trapped by the filter with no electric field. Most of the trapped particles are large, whereas those penetrating the filter are small. This does not contradict our previous statements since the electric field decreases the penetration of the small UO_2 particles.

We also conducted filter loading tests on the electrostatic filter installed in the UO_2 glove box. Figure 27 shows the filter efficiency and pressure drop for two loading tests, with and without an applied electric field, as a function of the particle mass trapped on the filter. These field data illustrate all the major trends seen in the laboratory tests with NaCl aerosols summarized in Table III. Figure 27 shows that filter efficiency increases with particle mass and with an applied electric field. The lower pressure drop at the high mass loading is also seen for the filter with an electric field. Although Fig. 27 implies there is little difference in filter efficiency with and without an electric field at a high particle mass, this is not true. The UO_2 penetrations at 6 g are 0.22% with and 0.72% without an electric field. Thus, the electric field significantly reduces penetration.

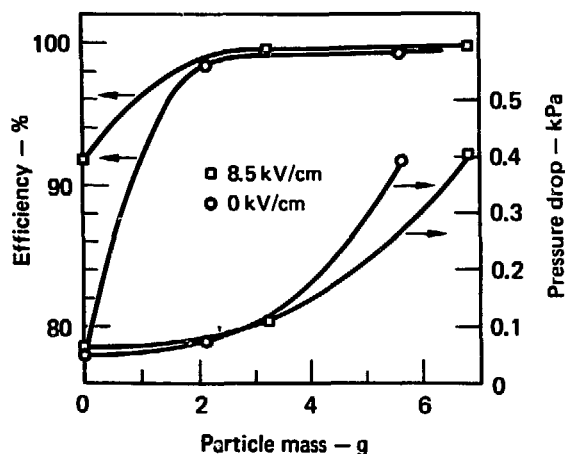


Figure 27

Filter efficiency and pressure drop for the prototype glove box filters during filter loading with and without an applied electric field.

An important finding of our field evaluation was the recovery of material normally lost in the ventilation system or in a HEPA filter. We found over 70% of the particle mass trapped on the filter could be recovered by gently shaking the filter and reusing it in normal operation. The pressure drop on this semi-clean filter was only slightly higher than the initial value. We successfully completed two cycles of filter loading and cleaning and believe repeated loading and cleaning may be possible.

In addition to our glove box applications, we are developing an electrostatic filter unit for installation in typical ventilation systems in the nuclear industry. To accomplish this objective, we have completed construction and calibration of the large-scale test system shown in Fig. 28. This system is designed to test prototype electrostatic filters at flow rates up to $0.472 \text{ m}^3/\text{s}$ (1000 cfm). Our test results with the flat screen design and NaCl aerosols are in good agreement with comparable tests in our small-scale and glove box systems. We are building and will soon evaluate extended area prefilters using electric fields.

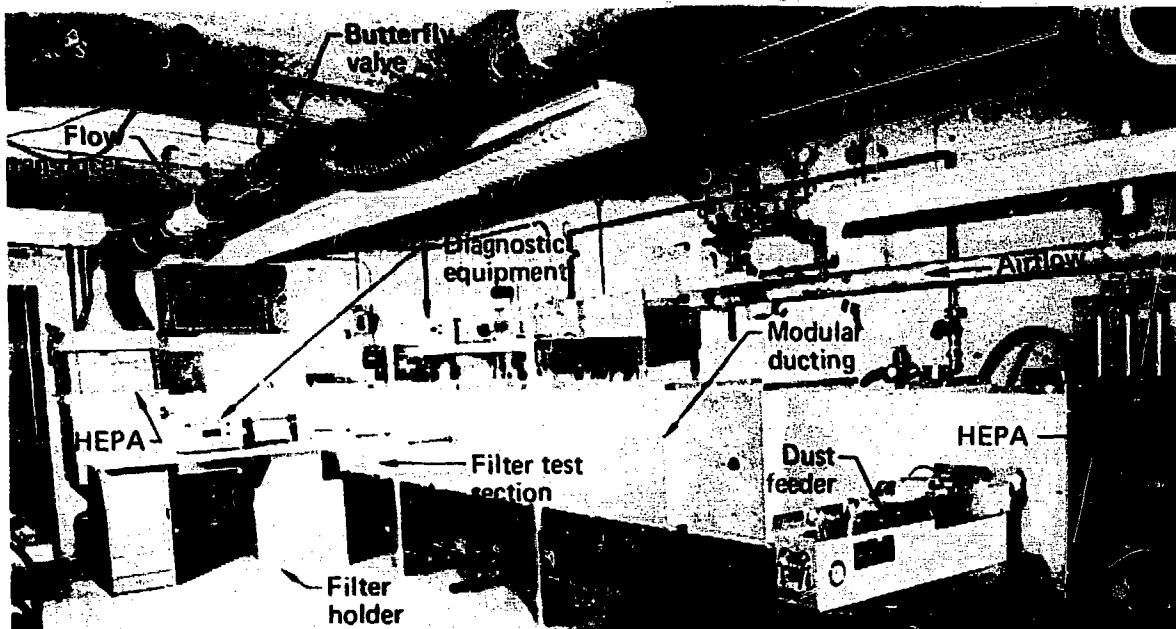


Figure 23 Large-scale test system.

V. Conclusions

The objective of LLL's Enhanced Filtration Program is to increase the service life of HEPA filters, thereby reducing filtration costs and the volume of nuclear waste. To reach this objective, we used an electrostatic filter to take the load off the HEPA filter. The electrostatic filter consists of a fibrous filter mat having a superimposed electric field generated by high-voltage electrodes that sandwich the filter. Investigation of this electrostatic filter is continuing, and the results presented in this paper represent a progress report of most of our work from January 1977 to June 1978. A number of additional investigations were not sufficiently advanced to be presented here. The major findings of this investigation were:

- Dielectrophoresis (the electric force between polarized particles and polarized fibers) makes a negligible contribution to the observed increase in filter efficiency with an applied electric field.
- Pressure drop theories for fibrous filters were extended to allow for distributions of fiber size. The predicted fiber diameter values for filter media were 9-53% lower than those calculated from pressure drop measurements and Davies' empirical equation⁽¹⁾.
- A new filtration mechanism based on the interaction of charged fibers and charged particles was responsible for much of the increased filter efficiency with an applied electric field.
- The electrostatic filter is a complex, dynamic system involving the buildup and dissipation of charge on the fibers.
- Fiber conductivity and electrode insulation have a large effect on filter efficiency. A filter with conducting fibers (10^8 ohm/cm) and insulated electrodes

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had no improvement in filter efficiency with an applied electric field. The same filter showed a large increase in filter efficiency when noninsulated electrodes were used.

- A theoretical model was developed that successfully explains the electrostatic filter efficiency.

- A theoretical model was developed that successfully explains the increase in filter efficiency and pressure drop that occurs during filter loading. Experimental data with different face velocities, particle size, and particle mass loading agreed with theoretical predictions.

- Filter loading tests with an applied electric field showed higher efficiencies and lower pressure drops than comparable tests without an applied electric field.

- Prototype electrostatic filters for glove box applications were built and evaluated in both laboratory and field tests. The evaluations were very successful.

VI. Acknowledgements

The authors take great pleasure in acknowledging the help of D. E. Salmi, H. L. Tasto, W. J. Kadi, and S. W. Wilson of LLL for the design and construction of the filter test facilities and prototype electrostatic filters shown in this report.

We are also indebted to Dr. J. C. Dempsey of DOE for his suggestions and for important information on new developments in the filtration field.

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DISCUSSION

BURCHSTED: Are the efficiencies you cited mass or number efficiencies?

BERGMAN: They are mass efficiencies.

BURCHSTED: An observation: we may be developing a nomenclature problem in calling these electrostatic filters, perhaps causing confusion with the better known plate-and-electrode electrostatic precipitator. I suggest the term "charged/polarized fiber filter" or "electrostatically charged/polarized fiber filter."

BERGMAN: I agree that there may be confusion. I like Dr. Rivers' term: "a nonionizing electrostatic filter." George Lamb, at Princeton Textile Institute, has called them "electrostatic filters, ESF."

RIVERS: Early in your talk you mentioned that the dielectrophoretic force was considerably less than the coulombic. I agree, and since this is the case, it would seem advantageous to increase the coulombic force by pre-charging the aerosol. Since all the elements for an ionizer (dust charger) are present, and the additional power consumption is quite trivial, have you considered adding an ionizer? In my experience, this results in sharply increased efficiency over the non-ionizing case.

BERGMAN: I mentioned a couple of references in the paper where people such as Don Reed (14th ERDA Air Cleaning Conference) have used aerosol precharging instead of just having an aerosol deposit naturally by image forces and charged fiber-charged particle interactions. You can also increase performance by having an electric field superimposed. So you see, you can use quite a variety of different combinations. We did not attempt to precharge the aerosols because electrical means require a high power consumption and a corona discharge. The system I described employs approximately 15 kilovolts and less than one microamp of current. This will give you an idea of how little power consumption is involved. We envisage an entire HEPA filter bank being operated by the equivalent of the power of a light bulb.

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RIVERS: I agree that an ionizer requires a lot more current, but it is still trivial.

T. F. ALLEN: We did some work on polarizing DOP and we experienced a lot of arcing in the system. You mentioned the use of polypropylene in the frame material of the filter. Polypropylene is a combustible material, and would not be allowed in a ventilating system. I wonder about this.

BERGMAN: This is our first prototype unit. We have a joint program with a group at Rocky Flats to explore additional versions. We don't consider the materials that we are using now to be the final design. In other words, we are primarily interested in insulation and preventing sparking and we have not seen sparking when we insulate the electrodes. When we do not insulate the electrodes, we do see significant sparking.

ALLEN: What voltage are you using?

BERGMAN: We are using 10,000 volts.

DYMENT: This is a fascinating paper and especially authoritative in respect to the fact that you use sodium chloride aerosols for your evaluations. Seriously, I have two questions; first, you state in your paper that for a uniformly thick filter medium, assuming particles stick at the first impact, one would expect the particle deposition density to tail away exponentially from the upstream face. Do any of your observations support this?

BERGMAN: It did and it did not. The smaller fibers show a heavy deposit on the surface which is what you'd expect from exponential decay by the standard filtration theories. For the filters composed of larger fibers, the heaviest deposit is somewhere in the center of the filter, rather than one the front surface. This is due to inefficient adhesion between particle and fiber after a collision. So, the theoretical models have to incorporate a sticking efficiency factor, whether you use van derWaal's forces or some other appropriate sticking factor. Thus, in some cases, you do see exponential decay whereas in others, you do not, but these differences can all be explained by rational means.

DYMENT: The second question: have you investigated the performance of your enhanced filter using a standardized test method such as ASHRAE 52-76 so that performance characteristics can be compared directly with commercially available filters of other types?

BERGMAN: The report shows our large scale 1000 CFM system. We have a standard ASHRAE dust feeder, we have a standard UK sodium chloride generator, and DOP generators, and we plan to make a thorough comparison with all of these aerosols.

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Dup

STUDIES ON PROLONGING HEPA FILTER SERVICE IN CHEMICAL APPLICATIONS

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Energy Systems Group
Rocky Flats Plant
Golden, Colorado
Work Funded by the Department of Energy

Abstract

The report presents information on activities directed toward gaining a better understanding of the performance of materials used in the construction of HEPA filters. The report discusses experiments in which materials were exposed to nitric and hydrofluoric acid vapors under controlled conditions in a specially constructed test chamber. In addition, observations are presented on filters tested in actual service. Finally, work on cleanable prefilters is reviewed.

I. Introduction

High Efficiency Particulate Air (HEPA) filters are relied upon to clean the air from chemical glovebox operations used in plutonium recovery. It is the objective of the work on this project to acquire information which will permit development of improved filters for chemical service.

II. Description of Chemical Exposure Tests

Test Chamber

A test chamber has been designed and constructed for the exposure of filter media to corrosive chemical vapors. The chamber is a stainless steel glovebox measuring 2 1/2 x 3 x 7 ft. with an atomizing nozzle and filter sample holders at opposite ends. A solution containing about 50 ppm hydrofluoric acid (HF) by weight and 8000 ppm of nitric acid (HNO₃) is atomized into the chamber atmosphere and drawn through the filters. These concentrations were selected in order to obtain an exposure level of about 50% relative humidity. There are holders for four filter handsheets, each with exposure dimensions of 8 x 3 inches. Flow rate through each filter was adjusted at 2 cfm to approximate the maximum flow rate of 1000 cfm through a 2 x 2 x 1 ft. HEPA filter. Dry air was added to the atomized solution to achieve the desired atmospheric concentrations.

A schematic diagram of the test chamber is shown in Figure 1. The amount of acid sprayed into the chamber is controlled by the air pressure in the atomizer. Dry instrument air is piped into the chamber to maintain the 50% relative humidity at 68°F. The relative humidity is allowed to fluctuate with temperature but the atomizing pressure is kept constant to maintain a steady rate of acid flow. Very little change in relative humidity is recorded as the laboratory temperatures in the past have ranged from a maximum of 77°F to a minimum of 68°F. Normal daily fluctuation range is about 4°F.

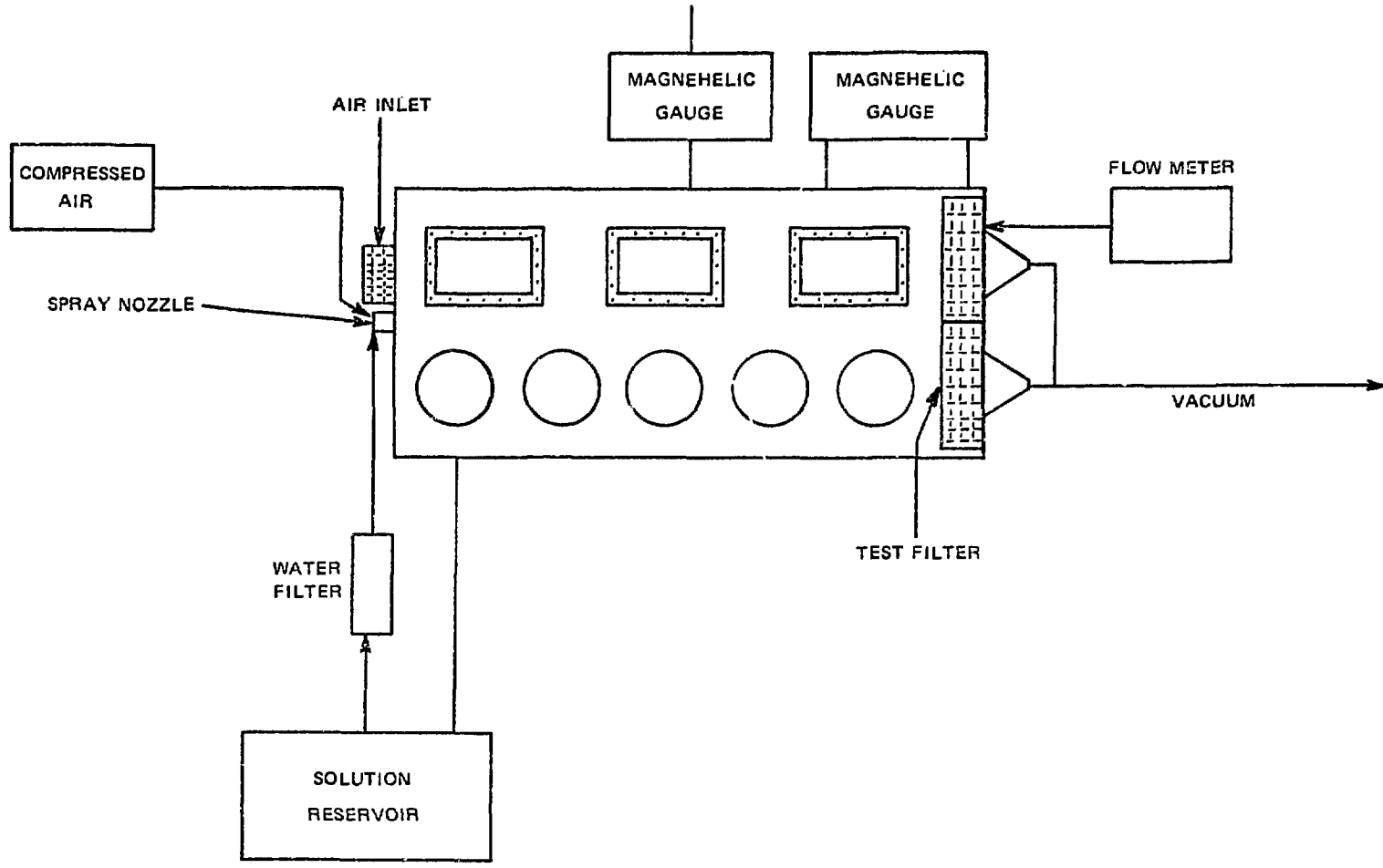


Figure 1. Schematic Diagram of Test Chamber

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Air is drawn through the chamber and filter media using the house vacuum line. An automatic control valve was placed in the line to compensate for any change in pressure.

A data acquisition system was installed to record information every hour. Data are currently being gathered on room humidity, room temperature, test chamber atmosphere temperature, house vacuum, NO_x concentration and total flow rate. Figure 2 shows a schematic diagram of the instrumentation of the chamber. The wet and dry bulb hygrometer data for the test chamber humidity is recorded on a separate chart. Thus far, attempts to interface a hygrometer with the data acquisition system have failed because of the corrosiveness of the chamber atmosphere.

Chemical Exposure of Filter Media

The initial experiments in the chamber were conducted to determine whether the small amount of crocidolite asbestos (~5%) improves the chemical resistance of the filter medium. Handsheets containing different concentrations of asbestos were exposed in the test chamber. Table I shows the data obtained in a series of tests. The results show that media with asbestos have greater retention of filtration efficiency after chemical exposure than the media without any asbestos. Under the conditions of the test there was no loss of chemical resistance when the asbestos contents were reduced to 4 and 3%.

When the asbestos was eliminated there was a significant increase in penetration. This is demonstrated where filter handsheets with and without asbestos were exposed to the same acid atmosphere simultaneously and for the same period.

Table I. HF-HNO₃ Exposure Tests on Filter Media
Exposure Time - 2 Weeks

| *Sample # | % Asbestos | *** Penetration | |
|-----------|------------|-----------------|-------|
| | | Before | After |
| 57 | 4 | 0.008 | 0.006 |
| 58 | 4 | | 0.006 |
| 59 | 4 | | 0.005 |
| 60 | 4 | | 0.008 |
| 61 | 3 | 0.009 | 0.004 |
| 62 | 3 | | 0.006 |
| 63 | 3 | | 0.008 |
| 64 | 3 | | 0.010 |
| 69 | 0 | 0.008 | 0.036 |
| 70 | 0 | | 0.080 |
| 71 | 4 | 0.008 | 0.006 |
| 72 | 4 | | 0.010 |
| 73 | 0 | 0.005 | 0.012 |
| 74 | 0 | | 0.060 |
| 75 | 4 | 0.005 | 0.012 |
| 76 | 4 | | 0.008 |
| 79 | 0 | 0.010 | 0.044 |
| 80 | 4 | 0.006 | 0.006 |

* Samples in brackets were run simultaneously.

**The penetration test is destructive and the "before" tests were on

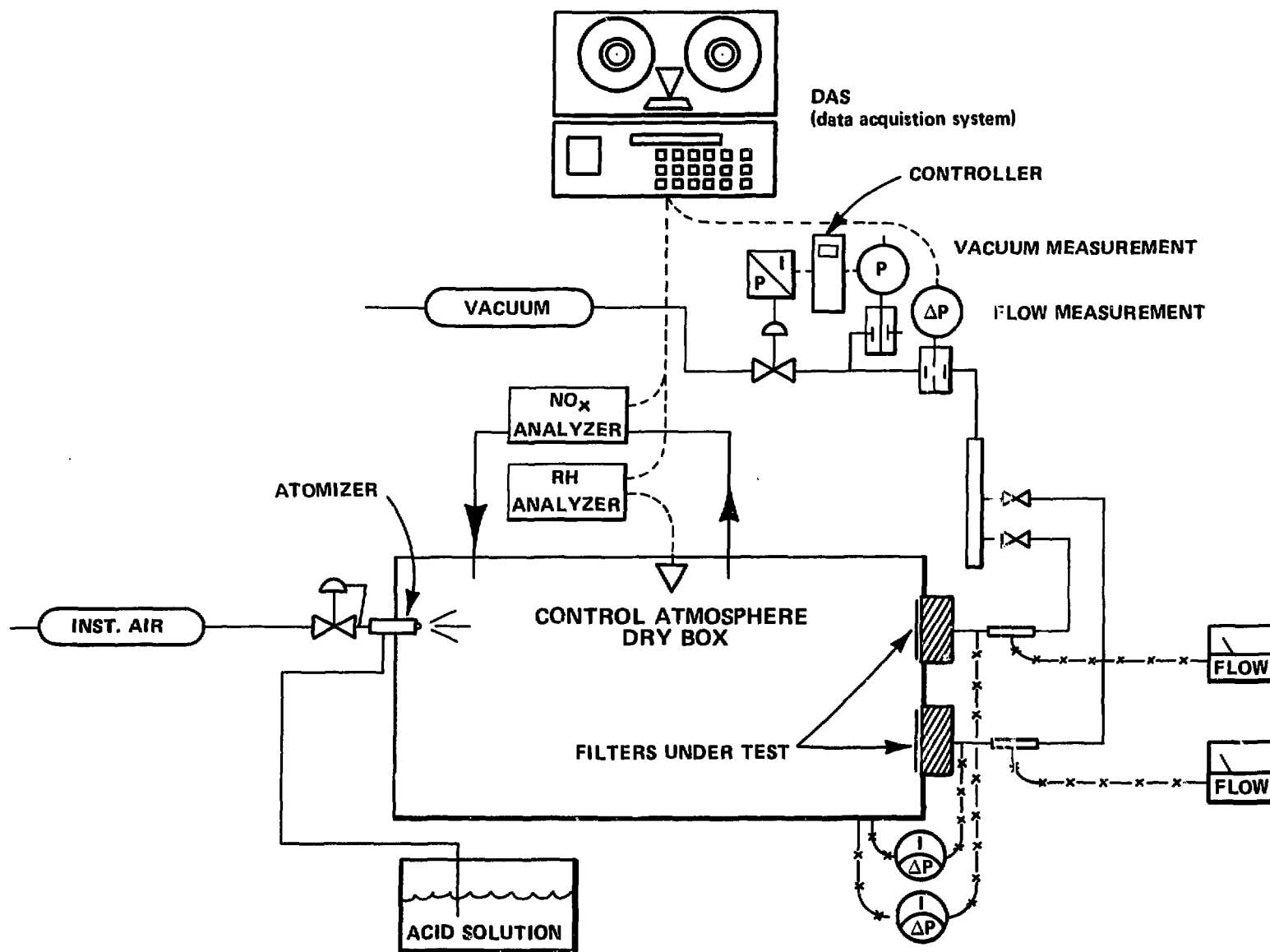


Figure 2. Diagram of Test Chamber Instrumentation

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handsheets made at the same time the test sheets were prepared but were not exposed to the acid atmosphere in the chamber.

Further tests to determine the optimum amount of asbestos to maximize corrosion resistance of the filter media have been suspended because of the health concerns regarding the handling of asbestos. Filter manufacturers are converting to asbestos-free filter media which has made the effect of asbestos in filter media a moot question.

Scanning electron photomicrography of the filter media exposed to the acid atmosphere in the test chamber has shown the presence of two different types of formations resulting from chemical attack of the filter media. They were web-like films connecting two glass fibers and bead-like structures strung along individual fibers. Electron microprobe analyses showed iron, nickel, and chromium present in the beads. Figure 3 is a scanning electron photomicrograph at 5000 magnification of a filter medium exposed to acid vapors in the test chamber. The numbers on the photograph refer to the areas which were analyzed with an energy dispersive spectrometer (EDS). Figures 3 a, b, and c are photographs of CRT displays of EDS scans of areas 1 - 3 respectively. The scans were over a range of 0 to 10.230 Kev in 50 seconds. Energy dispersive analyses show that areas 1 and 3 have typically a glass fiber composition. In the bead (area 2), although the concentration of silicon is less than that in the fiber, it is sufficiently great to indicate the HNO_3 -HF solution had attacked the glass fiber and left silicate beads as it dried. The presence of iron, nickel, and chromium is attributed to the mist of acid solution carrying a small concentration of corrosion products of the stainless steel acid reservoir. Acid mist, along with acid transported in the vapor state, corrodes the fiberglass. The finer fibers are preferentially corroded away resulting in loss of filter efficiency. Degradation of filters due to chemical exposure is likely to be basically by this mechanism.

Future work in the test chamber will be to establish whether some of the commercial, asbestos-free, acid resistant filter media are truly more chemically resistant and longer lived. Other experimental filter media which show a good filtration efficiency and other physical properties will be tested to evaluate their potential use in HEPA filters.

Chemical Exposure of Separators

The establishment of asbestos as a carcinogen has forced filter manufacturers, as well as users, to consider means to eliminate asbestos. Most of the asbestos used in HEPA filters is in the separators which serve to keep the filter media apart to maintain a free airflow.

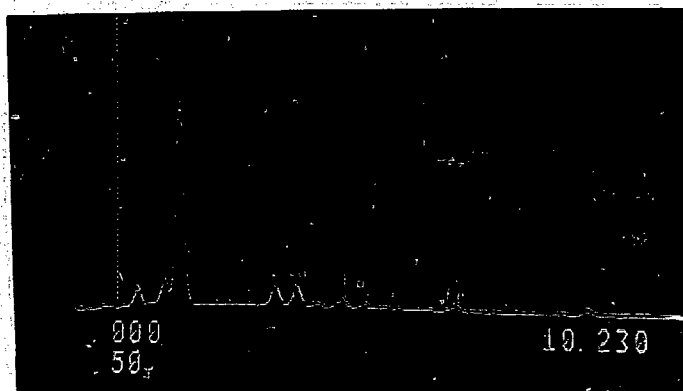
There are several approaches to this problem with regard to separators. One approach is to design the filter so that separators are not required. A manufacturer already is marketing a separatorless filter which is being evaluated in use at Rocky Flats. Another manufacturer is producing filters using tapes or strings as separators.

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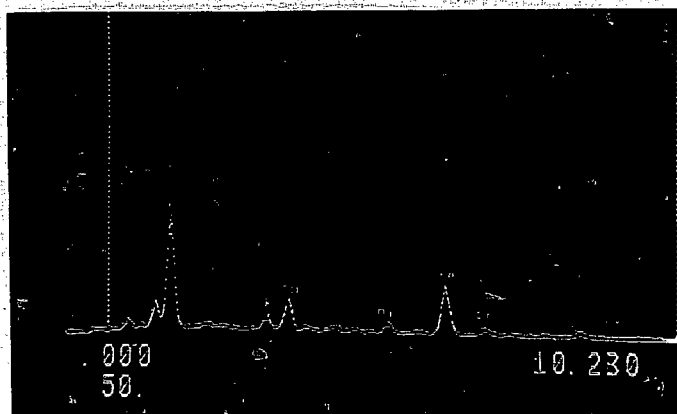


Figure 3. Scanning Electron Photomicrograph
of Filter Medium after Chemical
Exposure - 5000X

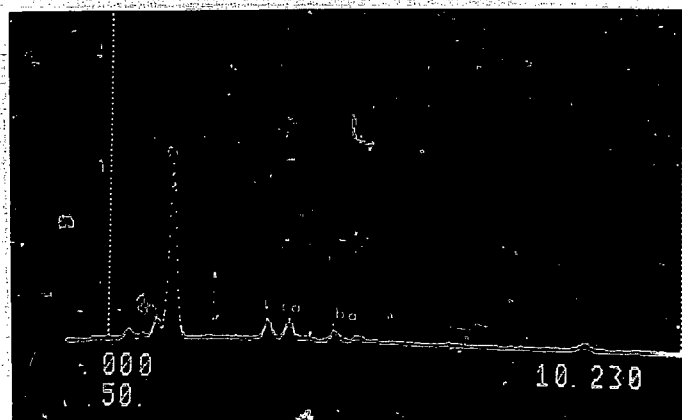
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AREA 1



AREA 2



AREA 3

Figures 3a, 3b, 3c. Cathode Ray Tube Displays of Energy Dispersive Spectrometer Scans of Areas 1, 2, and 3 shown in Figure 3.

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To insure a regular supply of filters which meet our specifications, Rocky Flats is investigating the possibility of using substitute materials to replace the asbestos separators.

Two general types of material were investigated or substituted for asbestos. They were plastics and metals. Three criteria used to screen candidate materials were flammability, chemical resistance, and cost.

It was found that the cost of plastic separators were at a minimum several factors higher than asbestos. For example, a polyvinylchloride (PVC) separator, 7 1/2 mil thick as shown in Figure 4, costs roughly 55¢ each in quantities in excess of 5000. Plastic material other than PVC tends to increase considerably above this cost. The cost of each asbestos separator is about 13¢. Another problem associated with the corrugation of plastics is that it can have memory which tends to return them to their original shape. This tendency can be reduced or eliminated by heating the material during corrugation. The memory of some polymers can also be destroyed by exposure to gamma irradiation. Gamma rays can break molecular chains and rearrange them so the material retains its new shape. The separators were fabricated from 7 1/2 mil thick PVC sheets by heating them in a mold. While the sheets exhibit little or no memory at room temperature, at about 150° they become pliable and sag.

Of the metal separators, aluminum was judged to be the most suitable. Its major shortcoming is a lack of acid resistance, especially to hydrofluoric acid. Attention was directed toward methods of protecting it from direct exposure to acid vapor. Two methods are being considered: one of them is the use of organic coatings such as fluorocarbon, epoxy, and phenolics which have good flame resistance as well as chemical resistance; the other is lamination with polymer films.

Polyvinylidene chloride and polyvinyl chloride laminated to aluminum were obtained from suppliers and suspended in the filter media test chamber for several months. Some delamination was more pronounced with the polyvinylidene chloride than the polyvinyl chloride. Exposure of the specimens whose edges were sealed with RTV silicone adhesives prevented any delamination or decomposition indicating that permeation of acid through the 0.5 and 1.0 mil polyvinylidene and polyvinyl chloride films is not a problem. Each vendor apparently uses his proprietary adhesive and will not reveal the formulation. One supplier's adhesive required that the aluminum foil be degreased before application and they did not have a practical process to accomplish this. When heat was used to degrease the aluminum, the heat treatment caused the aluminum to lose its temper. The alloy approved for use in the fabrication of HEPA filter separators requires a hardened grade.

Aluminum laminated with PVC by an aluminum company exhibited the greatest promise. Polyvinylidene laminate might be suitable if the plastic films were allowed to extend beyond the edge of the aluminum and sealed together. However, it was decided to use the PVC laminate for prototype studies since it exhibited only minor delamination in the test chamber.



Figure 4. Polyvinyl Chloride Separators

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Preliminary work with fluorocarbon and silicone coatings and a polyimide laminate indicates lack of adequate adhesion in the coatings and excessive cost for the laminate.

Stainless steel sheets for separators appear to be suitable except the cost of the steel would exceed \$500 per filter.

Future work planned is to continue testing and evaluation of coatings for aluminum.

III. In-Service Filter Testing

Chemical Studies

Chemical studies are made on components of HEPA filters which have been in service. The samples are taken from HEPA filters used in plenums to filter air exhausted from Production chemical glovebox lines. Figure 5 shows the location of the filters in the exhaust system. Note that six stages of filtration are used in booster plenums and two more stages in the main plenum. Most "in-service" tests are carried out by placing the test filters in the first stage of the four-stage booster.

The filters in the first stage bear the brunt of particulate and chemical vapors. The filters are highly contaminated during service and chemical tests can only be performed in a properly outfitted laboratory. However, in one instance, a filter removed from the sixth stage was not contaminated and could be handled without special precautions. Observations relating to the filter removed from the sixth stage are presented in Table III.

Table III. Data on HEPA Filter Removed from 6th Stage

Filter: Size 5, glass asbestos medium, asbestos paper separators
Manufacturer: Flanders Filters, Inc.

Tests on filter prior to service: Hanford Certification Lab(3/18/74)

Penetration: 100% flow .006% (.78" W.C. Δ P)
20% flow .004%

Gross weight (estimated): 21.4 kg (47 lbs.)

Tests after one year of service: Rocky Flats Filter Certification Lab

Penetration: 100% flow .004%
20% flow .020%

Gross weight: 30.4 kg (67 lbs.)

Test on medium taken from used filter:

Resistance: 42 mm W.C. @ 3.2 lpm thru 100 cm²

Tensile strength: Machine direction 7.42 lbs. per sq. in.

Cross direction 5.27 lbs. per sq. in.

Bulk: (thickness) .023 inch

The weight of the filter as noted in Table III was 67 lbs. A new filter averages about 47 lbs. The increase in filter weight, estimated at 20 lbs., is attributed primarily to reaction of ingredients in the separator with nitric acid and/or NO_x fumes, along with

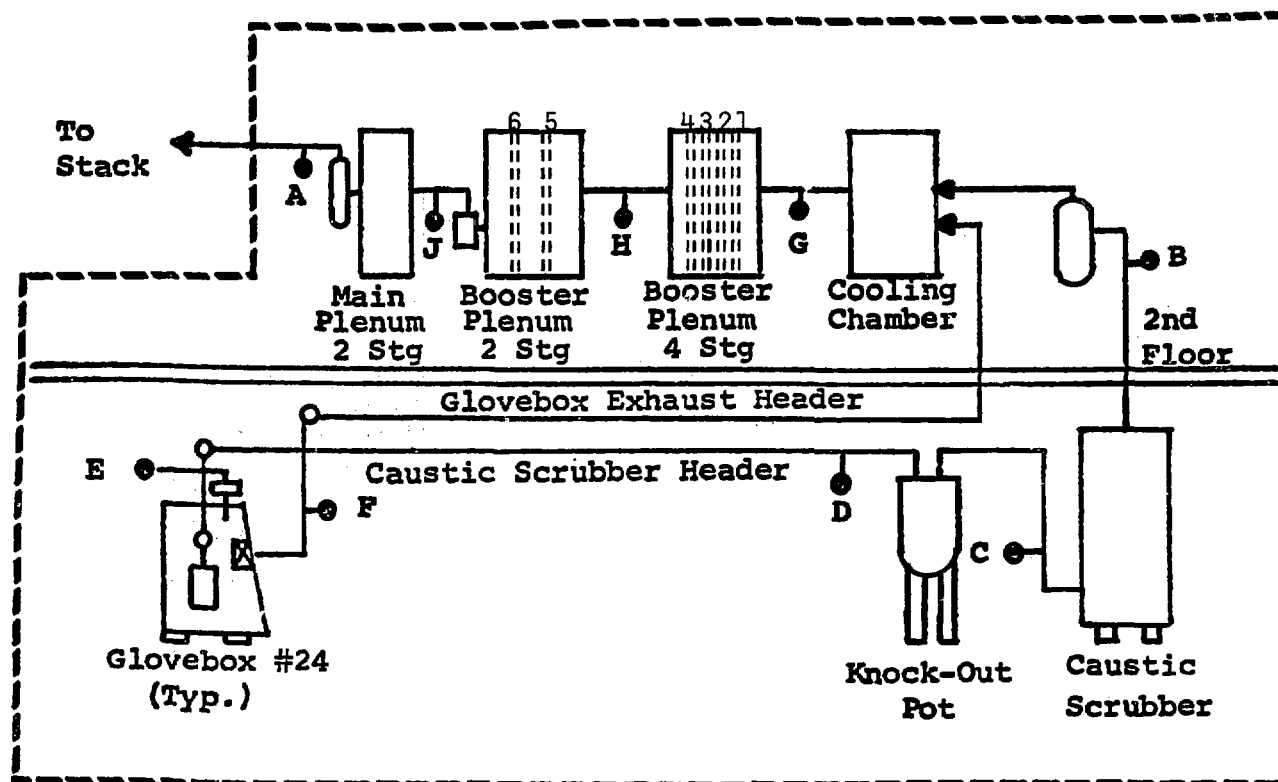


Figure 5. Exhaust System - Pu Chemical Recovery

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absorption of water. Evidence of the nitration reaction is depicted in the photograph shown in Figure 6 where crystals of magnesium nitrate are evident on the surface of separator material viewed under low power magnification.

Analytical data on the sixth stage filter components are presented in Table IV.

Table IV. Chemical Analyses of HEPA Filter
Removed from Sixth Stage

| <u>Sample Description</u> | <u>Wt. Loss_o</u>
<u>(%) to (C)</u> | <u>Water</u>
<u>Soluble (%)</u> | <u>Fluoride</u>
<u>(%)</u> | <u>Nitrate</u>
<u>(%)</u> |
|---------------------------|--|------------------------------------|-------------------------------|------------------------------|
| Medium | 3.6 950 | 2.7 | .10 | 1.0 |
| Separator | 61.0 840 | 68.3 | .07 | 35.8 |

The analyses support the observation that the filter, particularly the separator, became heavily nitrated.

Separatorless Filters

In-service tests have been made on separatorless filters on three occasions. The filters were Flanders Super-Flow[®] (Super-Flow is a trademark of Flanders Filters, Inc., Washington, N.C.) and constructed using a medium identified as Y100 with additive.

The filters have been mounted in the first stage of the plenum and the length of the in-service test ranged from 3 1/2 to 8 months. There has been no visible sign of failure of these filters. In one instance, one of the separatorless filters removed from service was transferred to a glovebox and the frame cut away to permit closer inspection of the medium. The medium was dark and dusty on the upstream face but clean and white on the downstream side. The medium was pliable. Penetration tests of the medium could not be performed because of plutonium contamination.

Chemical Resistant Filter

Contacts were established with the C. H. Dexter Division of Dexter Corporation and Mine Safety Appliances Company in regard to constructing special filters. The filters requested were to be fabricated using Dexter 1236 medium and asbestos paper separators. Both the medium and the separators to be treated by Dexter's patented process (1).

Samples of the Dexter medium to be used in the filters were subjected to analyses to evaluate its properties. The details of the thermogravimetric analysis appear in Table V.

Analysis by mass spectrometry indicated the volatiles included water, diethyl phthalate, and organics suggestive of cellulose (CH₃OH, etc.) Infrared analysis of the residue obtained from an extraction of the medium by boiling carbon tetrachloride indicated polyphenylene oxide (PPO).

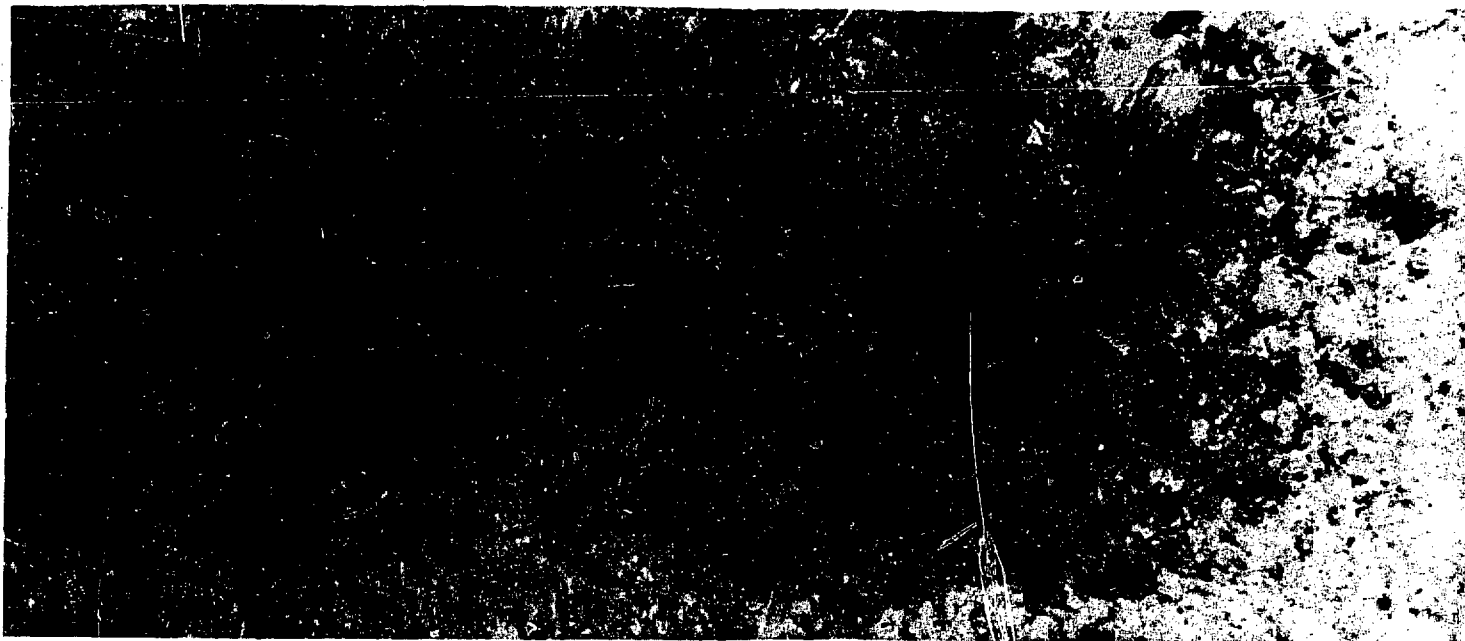


Figure 6. Photomicrograph (15X) of Magnesium Nitrate Crystals
on Separator Sample from 6th Stage Filter

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Table V. Thermogravimetric Analysis
of Dexter 1236 Medium

| Temperature
Range (°C) | Weight Loss
(%) | Comment |
|---------------------------|--------------------|--|
| 25-180 | .43 | Loss of H ₂ O |
| 180-330 | 1.1 | Attributed to loss of diethyl-
phthalate but may include some
PPO degradation. |
| 330-490 | 2.8 | Decomposition and oxidation of
PPO |
| 490-800 | .08 | Oxidation of residual carbon |

Four special-built HEPA filters (2' x 2' x 1') were received from Mine Safety Appliance Company (MSA). The filters were constructed by MSA using Dexter 1236 medium and asbestos paper separators. Both medium and separators had been treated with polyphenylene oxide (PPO) as described in Dexter's patent (1).

The filters were tested at the Rocky Flats Filter Certification Laboratory. Test results were as follows:

| Filter Serial No. | Resistance
Water Gage (in.) | DOP Penetration (%) | |
|-------------------|--------------------------------|---------------------|----------|
| | | 100% Flow | 20% Flow |
| 66553 | .97 | .016 | .014 |
| 66554 | .97 | .014 | .014 |
| 66555 | .98 | .016 | .014 |
| 66556 | .98 | .016 | .012 |

The filtration efficiency is better than the required minimum of .03% penetration by DOP aerosol even though the filters were fabricated using special medium and separators.

Two of these filters were placed in service for five months. Samples of the exposed medium and separator were chemically analyzed. Analyses were also performed on a standard glass/asbestos filter exposed alongside of the "chemical resistant" filter. Results appear in Table VI.

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Table VI. Chemical Analyses of Regular and Chemical Resistant Filters After 5 Months Service

| <u>Sample Description</u> | <u>Weight Loss (%) to</u> | <u>(°C)</u> | <u>Water Soluble (%)</u> | <u>Fluoride (%)</u> | <u>Nitrate (%)</u> |
|---------------------------|---------------------------|-------------|--------------------------|---------------------|--------------------|
| Standard glass/asbestos: | | | | | |
| Medium | 15.13 | 960 | 33.8 | 1.67 | 6.1 |
| Separator | 49.82 | 970 | 47.8 | .22 | 18.7 |
| Chemical Resistant: | | | | | |
| Medium | 16.71 | 970 | Lost | 1.30 | 6.4 |
| Separator | 44.51 | 970 | 50.1 | .12 | 22.9 |

The performance of the chemical resistant filter under the conditions of this test show it to be as good as the standard glass/asbestos filter with which it was compared. Additional in-service testing is under way relating to the effect of exposure on penetration.

Test of Aluminum Separator Protected with Polyvinyl Chloride Film Laminate

Corrugated aluminum is frequently used as a separator in construction of HEPA filters. While aluminum is satisfactory in most applications, it is more vulnerable to attack by chemical vapors; e.g., nitric acid in the presence of hydrofluoric acid. Encouraging laboratory results on protection of aluminum with a polyvinyl chloride (PVC) film laminate was extended to an in-service test.

An aluminum company laminated about 200 yards of 10 1/2-inch wide and 0.0015-inch thick aluminum with 0.001-inch thick PVC film on both surfaces. The alloy conformed to Military Specification 51068C for aluminum separators. A filter manufacturer constructed the prototype filter and experienced no difficulty in the fabrication. Tests at the Rocky Flats Filter Certification Laboratory on the finished filter showed the following results:

Penetration

| | |
|-------------|-----------------------------------|
| At 1000 cfm | 0.024% (0.030% maximum allowable) |
| At 200 cfm | 0.032% (0.030% maximum allowable) |

For normal use, this filter would have been rejected on failure to meet the penetration requirements at 200 cfm. However, it was placed in the first filter bank of a plutonium processing facility where HEPA filters are exposed to the most corrosive chemicals on the plant site. It was possible to install this filter for in-service tests despite its nonconformance to specifications because there are six banks of filters in the filtration system where only four are required. Also, the cause of nonconformance of the filter was independent of the laminated aluminum separators and significant

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information could be obtained by the in-service test. The filter was installed in the filter bank for over five months. Examination of the filter after its removal from the plenum showed no visible sign of degradation of the separators. However, a fire test at Edgewood Arsenal, N.J., where a prototype filter was exposed to a flow of hot air up to 750°F, demonstrated that the laminate was too flammable. Investigation of more temperature resistant materials is under way.

Cleanable Prefilters

An objective of the Filter Development Program is to increase the life of HEPA filters. The use of prefilters in conjunction with HEPA filters is one means whereby HEPA filter life can be extended. Many types of prefilters are available and incorporate a variety of fibrous materials such as glass, mineral, metal, and plastic.

Prefilters are usually constructed with a filter mat of rather loosely packed fibers. While in service, particulate matter collects in the fibrous medium and after the filter becomes sufficiently loaded the prefilter is removed and replaced. The used prefilter is discarded.

A logical objective in developing prefilter systems used in plutonium processing applications is to identify a means of cleaning the prefilter. A cleanable prefilter would offer the advantages of increased prefilter life and returning the plutonium to the process stream. Two materials have been investigated to determine the feature of cleanability.

Waterweb®

Waterweb® (Waterweb is the trademark of Heat Systems-Ultrasonics, Inc., Plainview, L.I., N.Y.) is a material developed for use as a demister and consists of plastic coated fiberglass screen which has been compressed into a rigid slab. Use of Waterweb as a prefilter is a departure from the use for which it was originally designed.

The first tests using this material were carried out by cementing a one-inch thick piece of Waterweb on a stainless steel frame which could be mounted in a holder normally used for a HEPA filter. Installation was made in a glovebox in which plutonium residues are leached. Exposure conditions include fine dust, moisture, hydrofluoric, and nitric acid fumes.

During the test the procedure was followed of exhausting the box through the prefilter. The surface of the prefilter became coated with a layer of dust approximately 1/8-inch thick and the pressure drop rose to as high as eight inches water column (W.C.) At this time the prefilter was valved out of service and the box was ventilated through an adjacent filter. The surface of the medium was vacuumed off using a 3/8-inch plastic hose connected to a vacuum receiver.

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After vacuuming, the prefilter was returned to service. Table VII presents data observed over a two-week test period.

Table VII. Performance of Prefilter
with Waterweb Medium

| Elapsed Time
(Days) | Pressure Drop
Across Prefilter
(Inches WG) | Comment |
|------------------------|--|-------------------------------|
| 0 | 1.2 | |
| 1 | 3.5 | |
| 2 | 5.0 | |
| 3 | 5.5 | Before vacuuming filter face. |
| 3 | 1.0 | After vacuuming filter face. |
| 4 | 3.0 | |
| 8 | 8.5 | Before vacuuming filter face. |
| 8 | 1.0 | After vacuuming filter face. |
| 9 | 3.0 | |
| 10 | 7.6 | |
| 15 | 8.5 | |

Observations were continued over a two-month period during which time the medium was vacuumed seven times. The medium was then removed from service and washed with water; no sign of degradation was observed.

An improved holder for the medium was designed and constructed. See Figure 7. Holders of this design are being evaluated in production service. Figure 8 illustrates how the prefilter holder also doubles as a hold-down for the HEPA filter which it precedes. The "watchcase" feature allows the prefilter to be removed without disturbing the HEPA filter.

ACS Demister Medium

Another type of demister medium being tested as a prefilter is Mistermesh® (Mistermesh is a trademark of ACS Industries, Woonsocket, R.I.). Mistermesh medium is fabricated from multifilament glass with parallel knit polypropylene. This material is not normally used as a filter for dry particulate.

Stainless steel frames, shown in Figure 9, were constructed to accommodate different thicknesses of the demister medium. The medium is friction fitted into the frame and held in place by a slip frame. When the unit is assembled it is approximately six inches thick and is used in place of a HEPA filter at the glovebox exhaust.

One prefilter, employing a pad of Mistermesh 8" x 8" x 3", was installed at the exhaust of a glovebox used for plutonium recovery.



Figure 7. Prefilter Holder

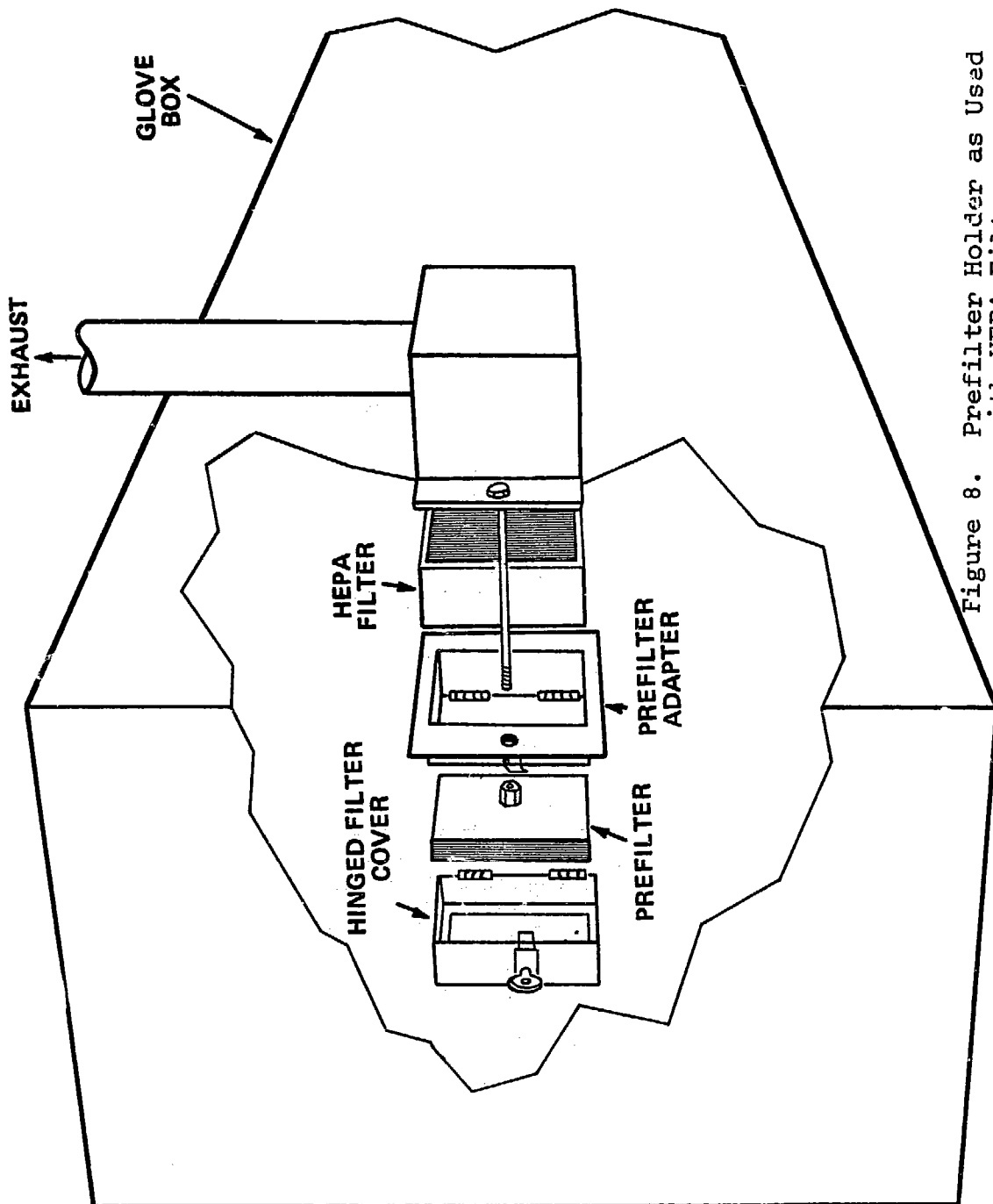


Figure 8. Prefilter Holder as Used with HEPA Filter

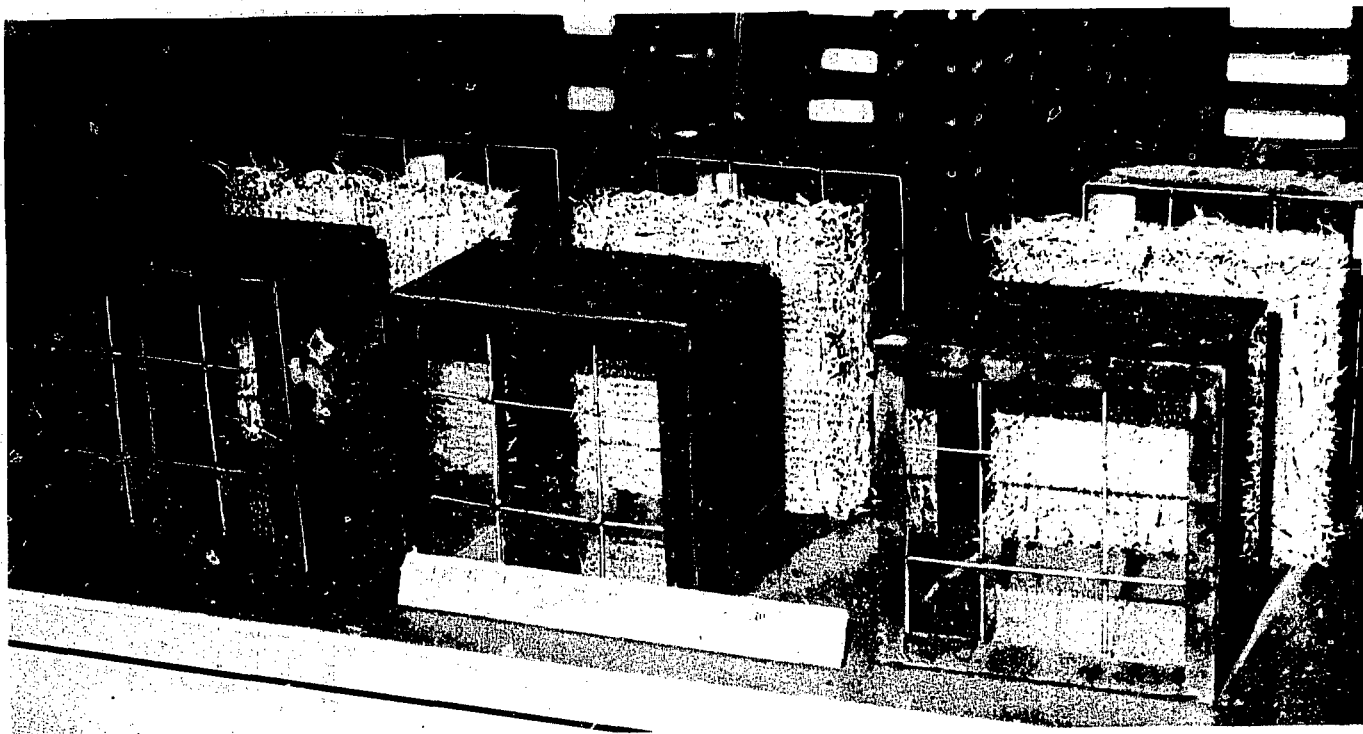


Figure 9. Prefilters Constructed of Stainless Steel Frames and ACS Medium

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After thirty days of service, the prefilter had plugged. The prefilter assembly was removed, the medium was taken from its holding frame and washed in the glovebox enclosure using water. Although the medium was rinsed repetitively, it could not be cleaned completely. After draining, the medium was allowed to dry and then returned to service.

Later on during the test period, the medium compacted in the holder to the extent that an additional pad, one-inch thick, was added to supplement the volume originally occupied by the three-inch pad. The ACS medium and its holding frame were dismantled after five months of intermittent service under production conditions. Although the efficiency of this type of medium is good for coarse particulate and droplets (2), it does not lend itself well to cleaning under the washing procedure tested.

IV. Summary

For summary, work toward the objective of prolonging the life of HEPA filters is proceeding along several lines. We have shown that asbestos does impart chemical resistance to HEPA filters and a suitable substitute should be found since use of asbestos in HEPA filters is being phased out.

Also, chemically resistant materials are being investigated for the replacement of asbestos separators.

The use of cleanable prefilters to reduce loading of glovebox HEPA filters appears to not only prolong the life of the filters but also reduces the amount of plutonium discarded as nuclear waste.

In-service tests of new commercial HEPA filters thus far indicates that they have service lives at least equal to those that are normally in current use.

Acknowledgement

Technical assistance was provided by J. F. Capes, W. D. Gilbert, K. J. Grossaint, and C. D. Skaats.

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- (1) W. T. Hense, U. S. Patent 3,594,993 (1971)
- (2) M. W. First, D. H. Leith, "ACS Entrainment Separator Performance for Small Droplet-Air-Steam Service," Report No. ACS-1 (1975), ACS Industries, Woonsocket RI

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DISCUSSION

BURCHSTED: We had an opportunity to see three experimental HEPA filters that had been stored in the attic of an AEC contractor's facility a few years ago. One had PVC separators, the others had unidentified plastic separators. The separators had softened and the filter pack of all three filters had sagged. I would suggest, therefore, that you view any filter with plastic separators with great caution.

GILBERT: About 16 or 17 years ago, filter purchasers, filter makers and filter papermakers, and fiber makers began to get together before each Air Cleaning Conference to talk about their mutual problems in an informal way. This nucleus of some 16 or 17 people made drafts of specifications which formed the basis for the filter that we know today. The deliberations of this informal group have been reported at each Air Cleaning Conference by Mr. Anderson, who is at the Naval Surface Weapons Center at Dahlgren, Virginia. By this conference, the original group of 16 or 17 has grown to about 125. This will be this year's report of the former informal filtration group that is now known as The Government-Industry Meeting of Filters, Media, and Media Testing.

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GOVERNMENT-INDUSTRY MEETING ON FILTERS, MEDIA, AND MEDIA TESTING

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Many of the accomplishments of the air filtration programs achieved thus far have been due to the efforts of an informed working group concerned with high efficiency filters. The existence of this group has now spanned eight air cleaning conferences and has shown successive growth and participation at each one. From the original handful of participants and their open discussion mode of operation, the meeting has progressed to an invited audience with a prepared agenda and, at this meeting, selected technical papers. At this, the 15th conference, 110 persons comprised the assembled body; 79 from industry, 15 from government, 10 contract investigators and 6 international guests.

The most recent session of this working group was held this past Monday morning and was devoted to a series of discussions on subjects of current interest. This session, following the precedents of earlier meetings, related the operating requirements placed on the high efficiency filter and the capacity of industry to meet them. To this end, the collected talents of the assembled body were unified toward the solution of the problems of the particulate filter, its components and methods of test. Representatives of all facets of the industrial complex were present, from the basic fiber suppliers, through the media producers, and finally to the filter unit fabricators. Research organizations from R&D government laboratories and academic institutions contributed status reports on work currently underway. Users at various levels expressed their problems and actively participated in the discussions.

The following review of the discussions may seem to be an agglomeration of information and show little continuity of subject. It is my intent to review for you, in abstract form, the items of committee deliberation. The items will be addressed in order of their discussion. At the recent session, six separate subjects were discussed. These were:

Revised Standards for Testing HEPA Filter Media

At the 14th Air Cleaning conference in Sun Valley, it was proposed that existing standards for testing of HEPA filter media be reviewed and that a single standard procedure be adopted. It was reasoned that this action would result in uniform requirements, specified techniques, and standardized equipment for testing that would produce comparable results at the different sites. In order to achieve this goal, a committee task force was established. This committee has met a number of times over the past two years and has completed a summary document titled "Efficiency and Flow Resistance Testing of HEPA Filters and Filter Media". A draft of this document (dated 7-4-78) was distributed at the meeting for comment. Since it is proposed to issue the final document as an ANSI standard, the chairman established a deadline of September 30, for receipt of

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comments and/or proposed revisions. Final publication will be contingent on the receipt and resolution of these comments and revisions.

A second action item from the Sun Valley conference was the determination of the need for a reference laboratory for purpose of consultation and calibration verification. The task force contacted filter paper manufacturers, filter fabricators, and users of these products and determined that the need for a reference laboratory was well established and would be invaluable for the correlation of tests results. In addition, the task force investigated a number of potential candidates for this role and conducted a technical/political/fiscal comparison of the proposed groups. Based on this study, it was recommended to the assembled body that a reference laboratory be established and that the Rockwell group at Rocky Flats be the selected site. A showing of hands signified the acceptance of both recommendations with only three dissenting votes out of the total assembly. Specific details of the mode of operation, as well as availability of services for the reference laboratory will be included in the proposed ANSI standard on filter and media testing. A point of contact at the Rocky Flats reference site is Mr. C. D. Skaats, Rockwell International, P. O. Box 464, Golden Colorado 80401.

Size Characteristics of DOP

Recent experience in the operation of the homogenous DOP generators used for filter media testing has raised the question as to the accuracy of the particle size and/or the size distribution of the generated aerosol.

Chuck Skaats of Rockwell International, Golden Colorado, reported on his work with the LAS-200 Laser Aerosol Spectrometer (PMS Co. of Bolder, Colorado) and its use for particle size measurement and distribution. Using this device, Skaats was able to draw some conclusions about the operation of the generating system:

1. The size distribution of the generated test aerosol was dependent on the system physical parameters. Pot temperature and variations in this temperature were critical in determining both size and distribution. Pre-conditioning of the air of spaces containing the instruments is essential for complete particle size control.
2. Disagreement exists between particle sizes measured by the photoelectric owl and size determined by the LAS-200.
3. Precise temperature measurement and controls necessary for stated particle size specifications will require improved instrumentation over what is currently being used. A complete copy of the Rockwell paper is attached to this report as Appendix A.

Confirmation of the Rockwell conclusions have been obtained by a more comprehensive investigation at the Harvard Air Cleaning Laboratory (HACL). William Hinds (HACL) reported to the group on their measurements of particle size and size distribution of the "hot DOP" aerosol. Using a PMS model ASASP-300 laser photometer

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for their studies, they compared particle sizes of various aerosols as determined by the photoelectric owl and the laser photometers. The LAS-200 photometer of Rockwell was also compared against the same aerosols. In addition, Hinds related his results with some theoretical work of Sinclair and reached the following conclusions:

1. The geometric standard deviation (GSD) of the Q-127 generator is about 1.15. This is sufficiently large to affect the interpretation of data from tests using the monodispersed aerosol.
2. The disagreement between the particle size measured by the photoelectric owl and the laser photometer is the result of aerosol polydispersity.
3. Variations in GSD's are believed to be the cause of inter-lab variability in penetration measurements.

Accepting these conclusions as factual, Hinds made a series of recommendations that included several options; these options ranged from changing the overall test size to conform more closely with the maximum particle size to using current equipment and technology and applying a correction factor based on a known GSD of a reference source. A complete copy of the Harvard paper is attached to this report as Appendix B.

Several comments on the two preceding papers are in order. It is acknowledged that particle sizes determined by the two instruments (owl vs. laser) are significantly different. However, it has not been demonstrated that this is a real phenomenon rather than a function of the calibration of the laser photometer. Such areas as refractive index differentials, types of median diameters determined and/or reported, physical calibration assumptions, etc. should all be considered before one assumes which device gives the more representative diameters. Further evaluations of interlab variability should be conducted before one concludes that GSD differential is the reason for experimental data variations. Parameters of flow, concentration, technique, leakage, etc. all must be considered and evaluated as part of the determination. It is expected that some on-going work at the reference lab and at Harvard will clarify some of these areas.

Glass Fiber Diameter Determinations

For many years the average diameter of microglass fibers has been determined by either an air permeability method (NRL) or a liquid drain time method (Williams Freeness). In general, both techniques gave comparable results at the larger microfiber diameters; deviations had been noted at the lower diameters especially those below 0.6 microns. In addition, neither technique supplied specific information on fiber diameter distributions. Cliff Cain, Johns-Manville, Denver, Colorado, presented a paper on their recent efforts to modify these existing techniques to make the test more responsive to both fiber diameter and diameter distribution. Using gas surface area measurements (BET) as the most accurate and reproducible data source, Cain compared the air permeability, liquid drain concepts as proposed by the NRL, Williams, Darcy,

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Kozeny-Carman, and Davies theoretical equations. From this work, we concluded that:

1. The porosity of the fiber mat must be accounted for in determining the volume-surface average fiber diameter using fluid flow techniques (air permeability or liquid drain).

2. Neither the NRL nor the Williams techniques adequately accounts for the variation in fiber diameter distribution inherent in production fiber samples.

3. Evaluations using the Davies modification of the Darcy equation does account for the variation of fiber diameter distributions.

4. The physical constants (Frazier Number) in the Davies equation can be correlated graphically with the BET derived volume-surface average fiber diameter.

Based on the results of the study, JM has derived a modified Williams Freeness vs. average fiber diameter calibration curve and uses this as a means of production fiber sizing. This has resulted in a more precise determination of the average fiber diameter and a better tool for quality control of the fiber production process. Test procedures are described in JM document FG-436-209. Sales literature (FF-91B 10-77) has been modified to reflect the code change-fiber diameter relationship in the standard fiber sizes (codes 100-112).

A complete copy of the JM paper is attached to this report as Appendix C.

QPL and Mil-Spec Status

George Smith, Chemical Systems Laboratory, Aberdeen Proving Ground reported on the status of the Qualified Products Listing for filter units currently managed by the Army. A listing (dated 23 December 1977) has been issued listing all manufacturers that have qualified their products in the various sizes and/or configurations. Copies of this listing were distributed at the meeting. A new QPL program will be conducted in FY 79 to supplement the existing lists with the new qualifiers. It is anticipated that the announcement for this FY 79 effort will be issued in November 1978, with testing to commence about the first of the following year.

Mil-Spec 51068, Revision E, on filter units is out for review and expected to be issued soon. Mil-Spec 51079, Revision C, on filter media is likewise out for review and expected to be issued if certain issue points can be resolved. Considerable debate was conducted on the specific issue of having two grades of filter media, with a color coding distinguishing between the two. Users questioned the technology gain/cost savings stated by the Army as the reason for the two media. Additional discussions were addressed to the length of time required for specification issue and the lack of response of the Army to the DOE input to the media specification. The committee chairman will work with Aberdeen and the filter media

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manufacturers in an attempt to resolve the remaining point of disagreement.

Filter Testing Program

A number of years ago, a "round-robin" filter test exchange was conducted between various laboratory, industry, and academic groups. The task force deliberations indicated that a renewal of this effort would be desirable and commissioned the reference laboratory, Rocky Flats, to initiate such a program. A variety of filter media has been collected from various media suppliers and a test group of filters assembled for the test program. These samples have been packaged in metal containers and forwarded to the various participants. Instructions with the samples requested that the test program be conducted using the standard operating procedure of the Mil-Spec 51079B and the resultant data forwarded to the reference laboratory. Analyses of the submitted data will be conducted by the reference laboratory and W. L. Anderson of the Naval Surface Weapons Center, Dahlgren, Virginia. Summarized data will be coded and forwarded to all participants.

Certification Program

Discussions at the 14th Air Cleaning conference expressed the need for a recognized certification program for in-house test personnel. Mel First (Harvard) reported to the group on his efforts to achieve this goal. A preliminary draft of proposed criteria for qualification was prepared and the Society of Mechanical Engineers was approached for possible endorsement and inclusion with their other certification programs, a rejection by this group has been received. The society's position that this special area of expertise was not in accordance with their overall policy of certification needs required another approach. After further deliberations, it was concluded that an alternate method would be to incorporate the program in an ANSI standard format. Following this approach, a draft of the standards for qualification has been prepared and distributed. Dr. First requested that comments, additions, and/or revisions be submitted by September 30, so that further actions can be expedited.

In conclusion, it should be re-emphasized that this informal working group, with its diversified representation, provides a means for a comprehensive and expedient solution to the problems of the filtration industry. The total effort has proven invaluable because it permits the surfacing and exposure of problems that might otherwise be lost in the quagmire of bureaucracy and management. The meetings are intended to be and actually are, a working level distribution of data and expertise as well as a progress report of ongoing projects in the particle filtration areas. To this end, we feel that we have been successful and future sessions are contemplated.

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Dup

Appendix A

A STUDY OF DIOCTYL PHTHALATE PARTICLES (DOP) GENERATED IN PENETROMETERS AND THE DEVICES USED CURRENTLY TO MEASURE THEIR SIZE

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The present method of measuring the 0.3 micrometer particle has been of concern to many users of Monodispersed DOP Penetrometers. Verification of the 0.3 micrometer is totally dependent upon the photoelectric particle-size meters (owls)* and the capabilities of the individual calibrating the owls and associated electronics.

Results from research work and from the study of the size of particles generated by a Monodispersed DOP Penetrometer indicate that what has been previously considered factual is now questionable.

In early 1977, the Test Facility at Rocky Flats purchased a specially designed Laser Aerosol Spectrometer (LAS 200) capable of measuring particle sizes in sixteen (16) separate channels from 0.1 to 1 micrometer at intervals of 0.055 micrometers. The results of each channel are printed as total count or shown visually on an oscilloscope. Other features were designed into the LAS 200 including portability.

One question deals with the use of the Laser to calibrate a DOP Penetrometer. The Laser itself is calibrated with Dow Latex spheres. The refractive index of the spheres is 1.508, which is near the refractive index of DOP, 1.485. We considered these refractory indexes close enough that the Laser would produce reliable results.

The Laser was first used to monitor particle size in a Q127 with no regard to concentration of DOP, a temperature of 156°C and with mechanical and visual "Owl" reading of 29°.

Verifying that the electronics and the mechanical "Owl" were calibrated according to standard procedures, samples of the DOP aerosol were taken from the mechanical "Owl" through the Laser at a flow of 0.2CFM with a printout every ten seconds. The results were also visible on an oscilloscope.

* "Owl" refers to the NRL Photoelectric particle-size meters. Visual means of measuring the intensity variation of each field in the mechanical analyzer.

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The printout and visual reading indicated that the particle size ranged from 0.3 to 0.6 micrometers. The curve displayed on the oscilloscope showed that the curve did not peak at 0.3 micrometers as was expected, but was very flat from 0.3 to 0.6 micrometers. Calculations from the printout indicated that only 10 to 15% of the total particles were in the 0.3 micrometer range. The concentration of DOP was approximately 130 micrograms per liter.

Operating Procedures 136-300-138, Edgewood Arsenal, recommends that the operating temperature should be 168 to 172°C to produce the 0.3 micrometers. We had previously found that these temperatures were too high to produce the concentration levels of 100 ± 10 micrograms. Therefore, temperatures were reduced until the level of concentration was obtained.

The first step was to reduce the heat, in 5° increments. Observing the reading on the oscilloscope during each 5° change, we could observe the change in the curve and regulate the heat to increase the percentage of the 0.3 micrometer range. At the same time, a definite drop in concentration levels was observed.

The temperature was continuously monitored by a Ralco digital thermometer which is certified to $\pm 1^\circ\text{C}$. At 145°C, the concentration level was approximately 75 micrograms per liter. Calculations indicated that 50% of the particles were in the 0.330 micrometer range. Eighty-four percent of the particles were less than 0.450 micrometers with a Geometric Standard Deviation of 1.36. Several attempts were made to reduce the temperature below 145°C. In each instance, no measurable aerosol was detected. For reasons undetermined, just prior to disappearance of the aerosol, the 0.3 micron particle size increased to 65% of the total count for approximately 20 seconds.

Each day for approximately two months, the Q127 was operated at the temperature of 145°C and measured by the LAS 200. After carefully studying the results, we concluded that the 145°C temperature would produce the best results.

During the entire study, no particular attention was given to the mechanical or visual "Owl". All measurements were solely dependent upon the LAS 200. Aligning the mechanical "Owl" and checking with the visual "Owl" and the particle size meter registering zero, we found the analyzer read 45°. Our original setting was 29° at 156°C. A joint test program was performed on March 23, 1978, at Harvard University on particle size versus the degree-of-angle (mechanical analyzer). The results of the latter study will be reported at the Government-Industry meeting.

Studies were also performed on the Q107 and Q76 Penetrometers. We found these units much easier to control; also, each unit produced a larger quantity of 0.3 micrometer particles for a concentration of 80 micrograms per liter.

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We found the Geometric Standard Deviation of the Q107 to be 1.34 with 84% of the particles less than 0.47 micrometers. The Geometric Standard Deviation of the Q76 was 1.28 with 84% of the particles less than .425 micrometers. Several attempts were made to reduce the Geometric Standard Deviation on the Q107 to 1.28. Some success has been realized with present temperature controllers.

A second study was performed on a polydispersed generator with a single 0.080" Alaski nozzle. The generator was installed in a chamber 13'x7'x8', and it produced a concentration level of 25 mg/m³. This particular chamber is used for respiratory fitting. The median of the observed particle sizes ranged from 0.383 to 0.400 micrometers with a Geometric Standard Deviation of 1.33 to 1.36. Previous information indicated the particle size was in the 0.7 micrometer range.

After a six-months study of the Q127, Q76 and Q107, the results have been analyzed and the following conclusions were reached:

1. Temperature measurement and control requires improved instrumentation.
2. Closer control of the ambient air and intake air of the Q76 and Q107 is required between 68 and 72°F.
3. Re-evaluate the mechanical analyzer and the 29° setting.
4. Consider changing the required 0.3 micron to 0.2. Our study indicated this is easier to produce than 0.3 micrometers and still permit the use of the 29° setting in the analyzer.

We definitely feel that other factors such as atmospheric pressure, humidity, and temperature of operating air, influence the generation of the 0.3 micron particle.

We also evaluated DOP from different manufacturers and found that some are easier to control at lower temperature and will still produce the 0.3 micron particles.

Should further study confirm the Harvard and Rocky Flats findings, the next question is what affect, if any, will this have on the media and filter industry.

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Appendix B SIZE DISTRIBUTION OF "HOT DOP" AEROSOL PRODUCED BY ATI Q-127 AEROSOL GENERATOR

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Abstract

Measurements of the particle size distribution of the "Hot DOP" aerosol produced by the ATI Q-127 aerosol generator at mechanical analyzer settings of 25°, 29°, 35°, 40°, and 45° were made using a high resolution optical counter, PMS Model ASASP-300. Size distributions were approximately log normal. At mechanical analyzer settings of 29° and 45° the generator produced an aerosol with CMD's of 0.23 and 0.30 μm , respectively, and GSD's of 1.15. These CMD's are 23% and 13% less than the sizes expected from Sinclair's³ curve of analyzer angle versus diameter for monodisperse aerosols. The differences between measured CMD's and expected sizes for the given analyzer settings are shown to be due to the polydispersity of the aerosol and the fact that polarization ratio is an extremely strong function of diameter (proportional to $d^{0.1}$).

Experimental

Measurements were made at Harvard School of Public Health of the size distribution of the aerosol droplets produced by our ATI Q-127 "Hot DOP" aerosol generator. Figure 1 shows the airflow system used to make particle size measurements. A dust-free dilution system was needed to reduce aerosol concentrations to a level that could be accommodated by the particle size measuring instruments used. Room air passed through a standard 1000 cfm HEPA filter to remove dust particles which might otherwise have confounded measurements of DOP concentration and size distribution. The filter was tested in place with cold DOP and found to have penetration less than 0.05%.

Downstream of the HEPA filter, DOP aerosol from the Q-127 was introduced through a tube at the duct centerline. The aerosol then passed a Stairmand disk which mixed the concentrated DOP with dust-free air to produce a uniform concentration within the duct. Approximately six duct diameters downstream of the Stairmand disk, aerosol samples were withdrawn through a sampling port flush with the inside of the duct wall. The remaining aerosol passed through a Venturi flowmeter and flow regulating damper to an exhaust fan.

The Q-127 generator was operated at the recommended air rate of 100 liters per minute. The flow of DOP to the air dilution system was controlled by the fraction of the total that was passed to waste through the chuck; the balance passed to the filtered air dilution system. By changing the volume rate at which DOP entered the dilution air system, changes in DOP concentration were effected.

Size measurements were made with a PMS Model ASASP-300 active scattering aerosol spectrometer probe connected to a DAS-32 data control system and a Wang 2200 minicomputer. Samples were drawn through the spectrometer with its own sampling pump. Because all

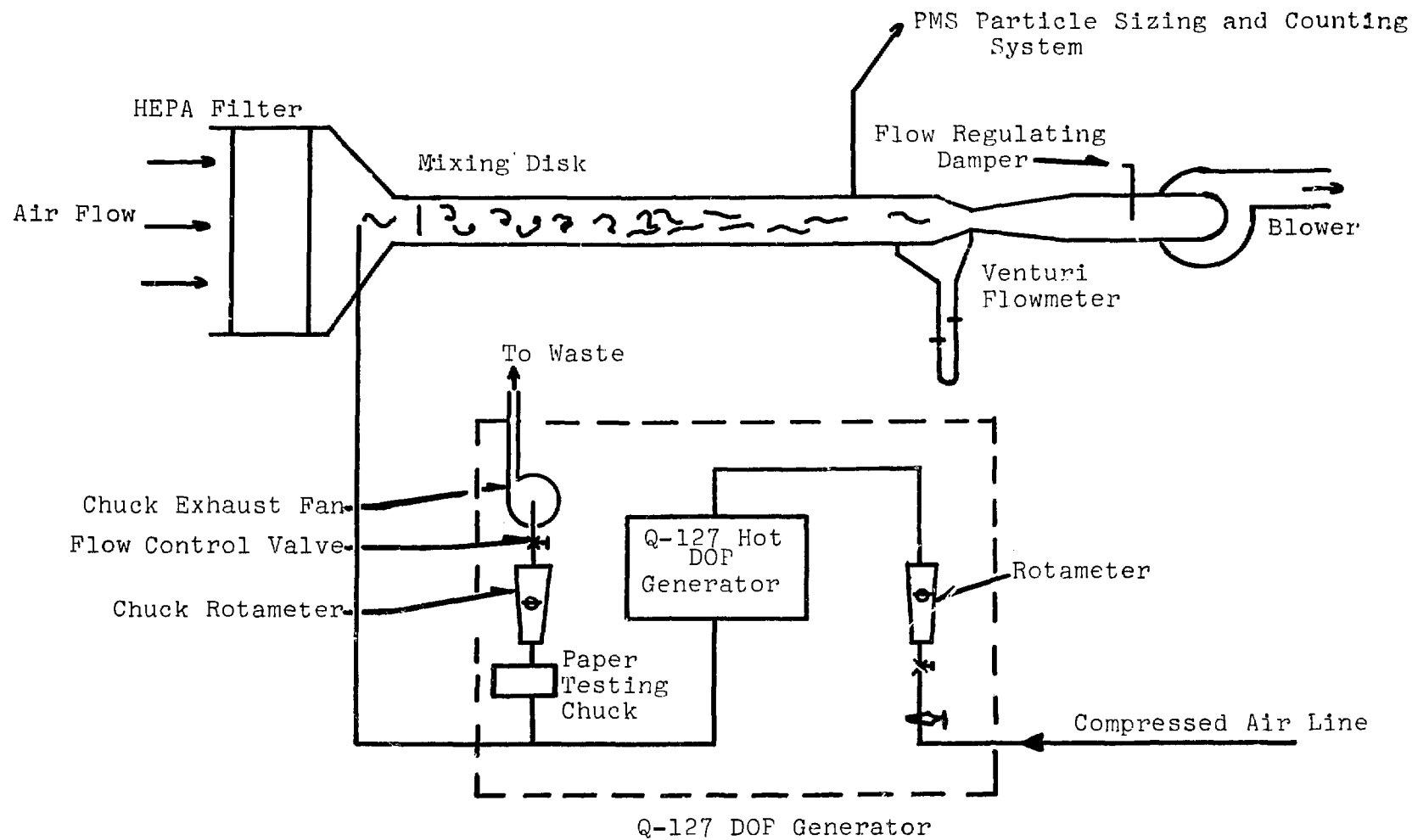


Fig. 1. Schematic drawing of "Hot DOP" air dilution system

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the DOP particles were submicrometer, no attempt was made to sample from the duct isokinetically. The relevant size ranges of the instrument are shown in Table 1. Because the spectrometer uses two independent instrument settings to cover the size range of interest, a check was made to ensure that these ranges overlapped properly. Results, shown in Figure 2, show good agreement in the overlapping region.

Immediately prior to sampling, background size distribution was measured and instrument size calibration verified using an aerosol of uniform diameter, 0.206 μm , polystyrene latex (PSL) spheres. The measured background particle counts are shown in Table 1. Sampling time for the background counts was 10 minutes but two minutes was used for the high concentration DOP measurement runs. The PSL spheres used for calibration were labeled 0.234 μm by Dow Chem. Co. but were found to be 0.206 μm by Porstendorfer and Heyder.¹ The light scattering instrument manufacturer claims that instrument accuracy is equal to plus or minus one channel width and the primary peak shown in Figure 3 is within this range.

Table 1. Ten minute background counts for test system.

| PROBE A RANGE 3 | | | | PROBE A RANGE 2 | | | |
|--------------------|-------|-------|-------|----------------------|-------|-------|-------|
| .15 TO .3 BY 0.010 | | | | .23 TO .605 BY 0.025 | | | |
| I | MIN | MAX | COUNT | I | MIN | MAX | COUNT |
| 1 | 0.150 | 0.160 | 20 | 1 | 0.230 | 0.255 | 5 |
| 2 | 0.160 | 0.170 | 2 | 2 | 0.255 | 0.280 | 2 |
| 3 | 0.170 | 0.180 | 4 | 3 | 0.280 | 0.305 | 0 |
| 4 | 0.180 | 0.190 | 2 | 4 | 0.305 | 0.330 | 1 |
| 5 | 0.190 | 0.200 | 2 | 5 | 0.330 | 0.355 | 0 |
| 6 | 0.200 | 0.210 | 3 | 6 | 0.355 | 0.380 | 1 |
| 7 | 0.210 | 0.220 | 3 | 7 | 0.380 | 0.405 | 0 |
| 8 | 0.220 | 0.230 | 1 | 8 | 0.405 | 0.430 | 0 |
| 9 | 0.230 | 0.240 | 1 | 9 | 0.430 | 0.455 | 0 |
| 10 | 0.240 | 0.250 | 0 | 10 | 0.455 | 0.480 | 0 |
| 11 | 0.250 | 0.260 | 1 | 11 | 0.480 | 0.505 | 0 |
| 12 | 0.260 | 0.270 | 1 | 12 | 0.505 | 0.530 | 0 |
| 13 | 0.270 | 0.280 | 1 | 13 | 0.530 | 0.555 | 0 |
| 14 | 0.280 | 0.290 | 0 | 14 | 0.555 | 0.580 | 0 |
| 15 | 0.290 | 0.300 | 2 | 15 | 0.580 | 0.605 | 0 |

Measurements were recorded in a format similar to Table 1 and results plotted on log probability graph paper as cumulative size distributions. Count median diameter and geometric standard deviation were obtained by eye fit of the best straight line through points between cumulative counts of 10 and 90 percent. The first set of DOP measurements was taken at an "OWL" analyzer angle of 29°, the "standard setting" for the Q-127 generator.⁴ Measurements were made at two concentrations differing by a factor of 6.4 to determine the significance of coincidence effects on particle size measurements. Results for a single measurement at each concentration are summarized in Table 2. Although the CMD's are within experimental error, the GSD is slightly higher at the higher concentration, consequently all subsequent measurements were made at a DOP aerosol concentration of 75 $\mu\text{gm}/\text{m}^3$.

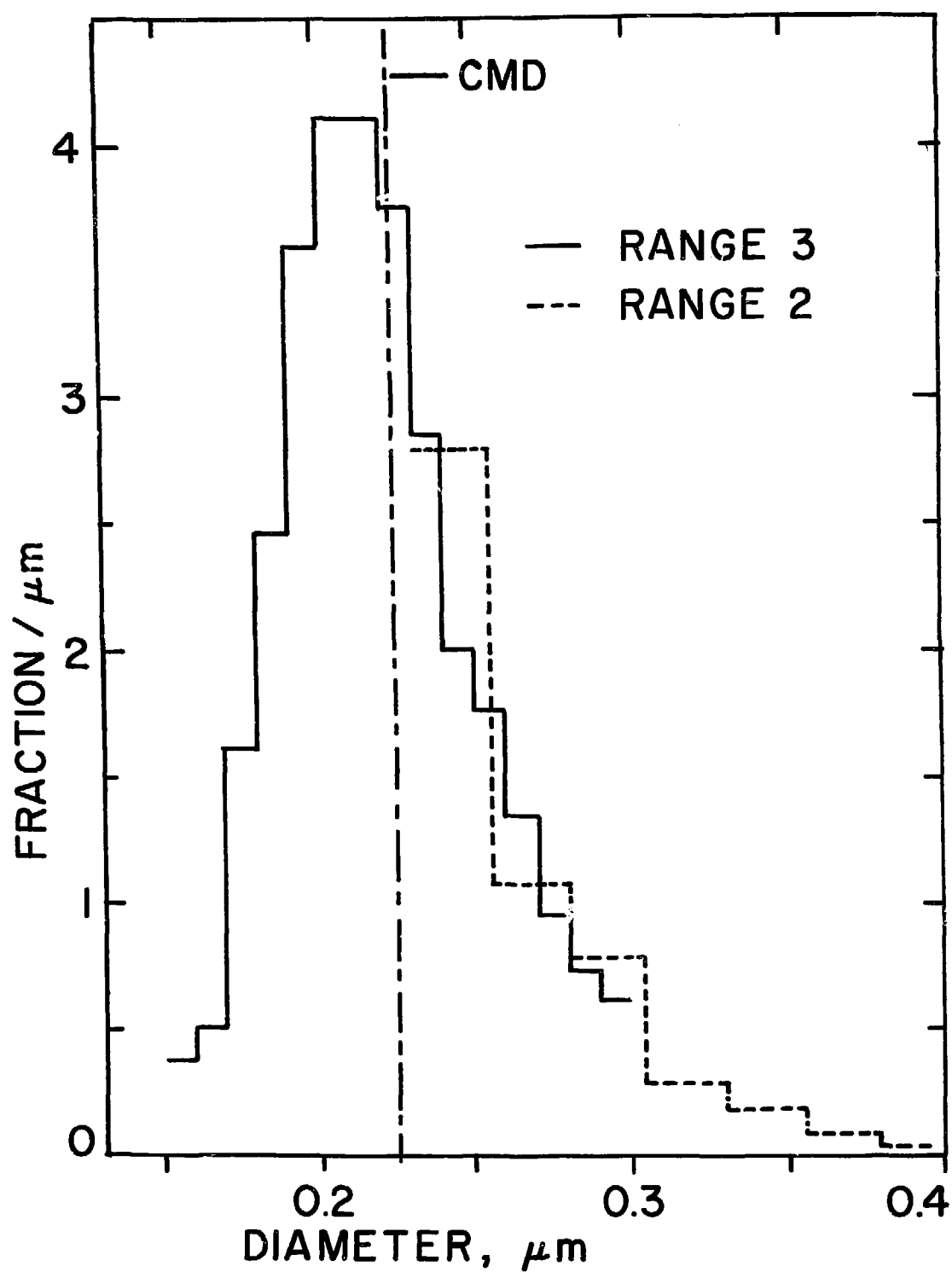


Fig. 2. Size distribution of "Hot DOP" showing overlap of PMS size ranges 2 and 3.

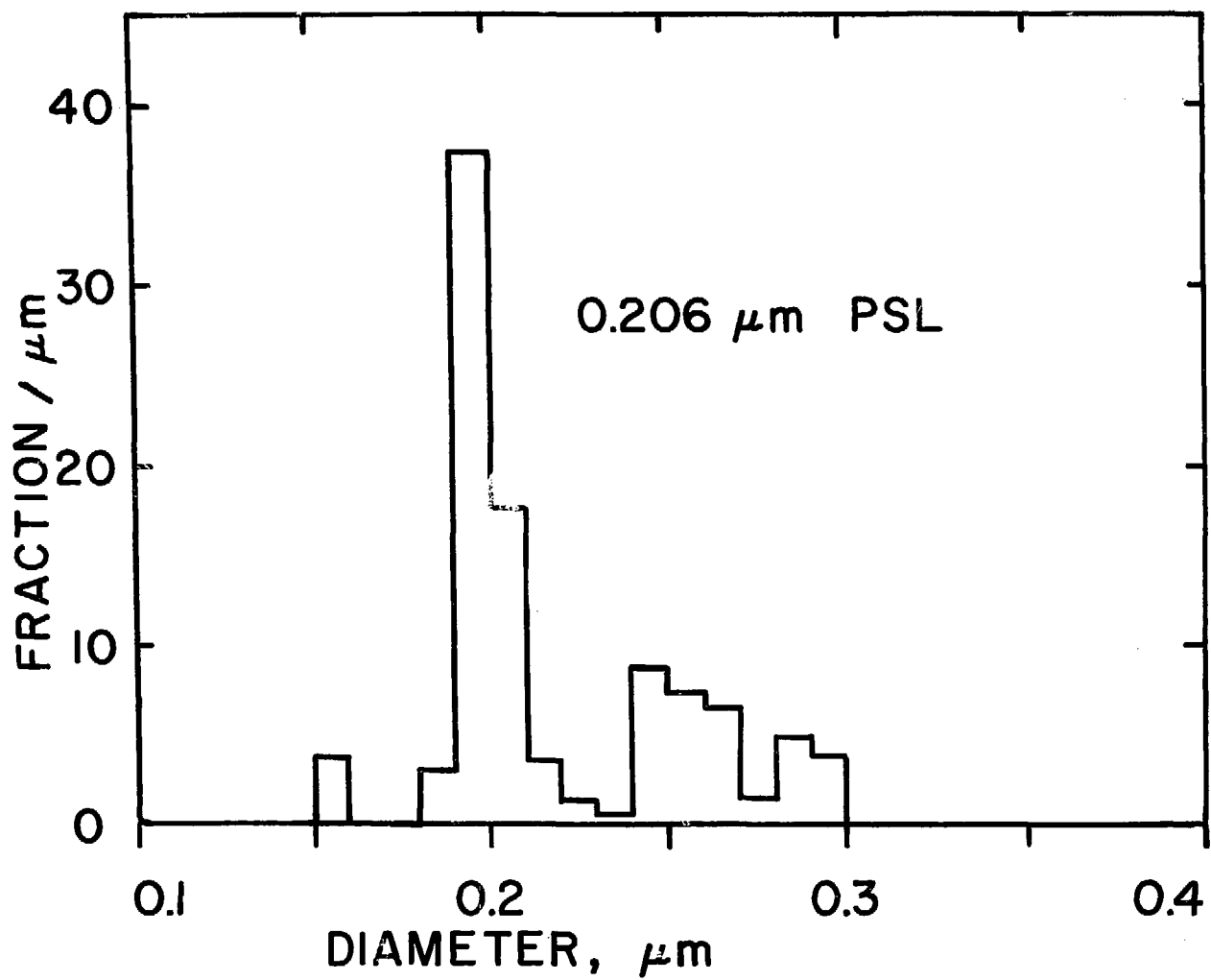


Fig. 3. Calibration of PMS with 0.206 μm PSL

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Table 2. Size distribution parameters measured by the PMS instrument at two aerosol concentrations for an analyzer setting of 29°.

| <u>DOP Conc.</u> | <u>CMD</u> | <u>GSD</u> |
|-------------------------------|--------------------|------------|
| 75 $\mu\text{gm}/\text{m}^3$ | 0.23 μm | 1.18 |
| 480 $\mu\text{gm}/\text{m}^3$ | 0.24 | 1.24 |

A second set of measurements was made to determine the size distribution for different generator operating conditions corresponding to OWL analyzer angles of 25°, 29°, 35°, 40°, and 45°. Results are shown for 29° and 45° in Figures 4 and 5, and all results are summarized in Table 3. As expected, particle size increased with analyzer angle setting and GSD remained constant for all analyzer angle settings. For analyzer angles of 29° and larger, a secondary peak was observed in the one or two smallest size channels. This secondary peak was not observed by Tillery² using a different PMS spectrometer with 0.01 μm channel width over the range of 0.08 to 0.23 μm . For the purpose of this analysis the small diameter secondary peak was considered to be an artifact of our instrument and was ignored in the size analysis. Figure 6 shows a comparison of Sinclair's curve of size vs. polarization ratio for monodisperse aerosols³ and the count median diameters we obtained at the five analyzer angle settings.

Table 3. Size distribution parameters measured by PMS instrument at six analyzer settings.

| <u>Analyzer Setting</u> | <u>CMD</u> | <u>GSD</u> |
|-------------------------|------------|------------|
| 25° | 0.205 | 1.18 |
| 29° | 0.232 | 1.15 |
| 35° | 0.250 | 1.15 |
| 40° | 0.265 | 1.15 |
| 45° | 0.300 | 1.15 |

It was suggested that the difference between measured size and theoretically derived size of 0.3 μm might be related to a difference in index of refraction (IR) between PSL calibration particles and DOP particles produced by the Q-127. Table 4 shows IR values for PSL and for DOP derived from several sources: "New DOP" was removed from the shipping containers in which it was received, "Used DOP" was drained from the reservoir of the generator after a number of hours of operation, and "Aerosol DOP" was collected by electrostatic precipitation of the output aerosol of the Harvard Q-127. The differences in IR of the several DOP samples is considered to be insignificant with respect to the response of the PMS analyzer and the same is true for the differences in IR between DOP and PSL. Knollenberg⁷ has discussed the insensitivity of the PMS analyzer to changes in IR of aerosol particles.

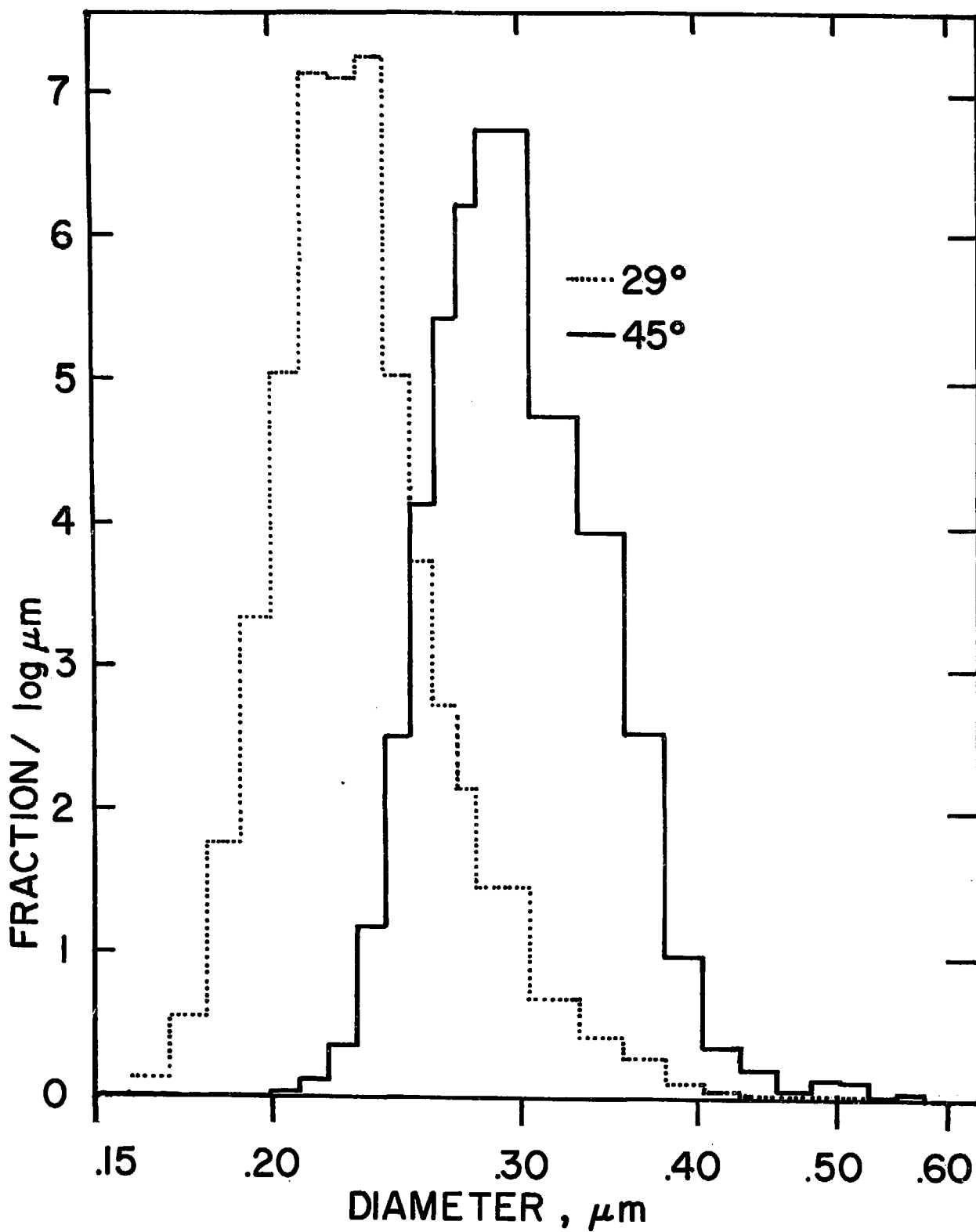


Fig. 4. Size distribution of DOP aerosol produced by Q-127 generator at analyzer settings of 29° and 45°.

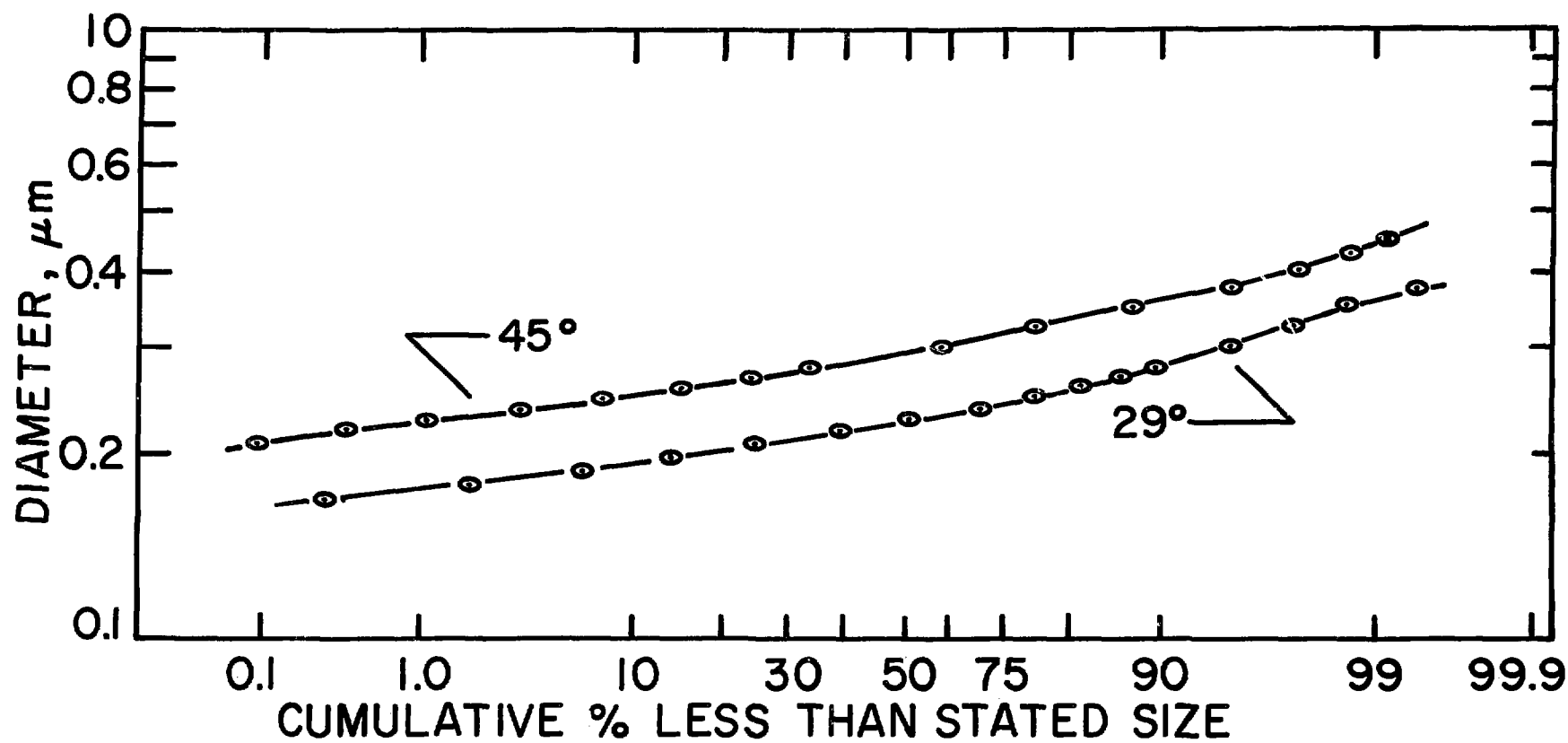


Fig. 5. Cumulative size distributions of DOP aerosol produced by Q-127 generator at analyzer settings of 29° and 45°.

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Table 4. Refractive index of PSL and DOP from four sources at 589 nm.

| <u>Source</u> | <u>Refractive Index at 22°C</u> |
|------------------|---------------------------------|
| PSL | 1.5905 |
| "New DOP " BASF | 1.4850 |
| "New DOP " Kodak | 1.4845 |
| "Old DOP" | 1.4858 |
| "Aerosol DOP" | 1.4864 |

Discussion

Table 5 shows filter penetration results for identical filter papers tested at 5 different laboratories.⁵ All measurements were made with the mechanical analyzer set to 29° except for the measurements shown in the first column for which the analyzer was set to 45°. There is significant variability in results between laboratories except that Dexter and Hollingsworth and Vose show close agreement. The penetration results obtained at Harvard when using an analyzer setting of 45° are consistently less than the results at 29°. The average ratio of penetration at 29° to 45° is 2.50 with a range from 1.84 to 3.25. For both analyzer settings, the test aerosols have the same geometric standard deviation but the median size differs by 0.07 µm. Thus a 23% reduction in median particle diameter produces a 250% increase in penetration for the DOP size range between 0.23 and 0.3 µm.

The decrease in penetration with increasing particle size suggests that impaction is the primary removal mechanism. If the test aerosol particles were to be reduced to a size close to that for maximum penetration, the effect on penetration of small size variations would be minimized.

With one or two exceptions, the two Harvard readings for analyzer settings of 29° and 45° bracket the penetration measurements made at other laboratories. This suggests that if the test aerosols used at these other laboratories had the same GSD as Harvard's (i.e., 1.15) the differences in CMD between laboratories would have to cover a range as large as 30%. As will be discussed later, these differences in CMD at constant GSD can be produced only at analyzer settings significantly different than 29°. Inasmuch as the standard operating condition for the Q-127 is an analyzer setting of 29°, the observed differences in filter penetration must be a result of differing geometric standard deviations of the test aerosols.

The Harvard data points in Figure 6 show approximately the same shape as the theoretical line calculated by Sinclair³ for a refractive index of 1.483 except that the particle diameters are 13 to 29% less at the same analyzer angle. It must be kept in mind, however, that the Harvard data points represent count median diameters of polydisperse aerosols whereas Sinclair's theoretical polarization ratio line was determined for perfectly monodisperse aerosols. The aerosols produced by the Q-127 cover a significant range of sizes, e.g., for a GSD of 1.15, having a two standard deviation range of +35% and -24% of the CMD. Therefore, the mechanical analyzer is measuring an

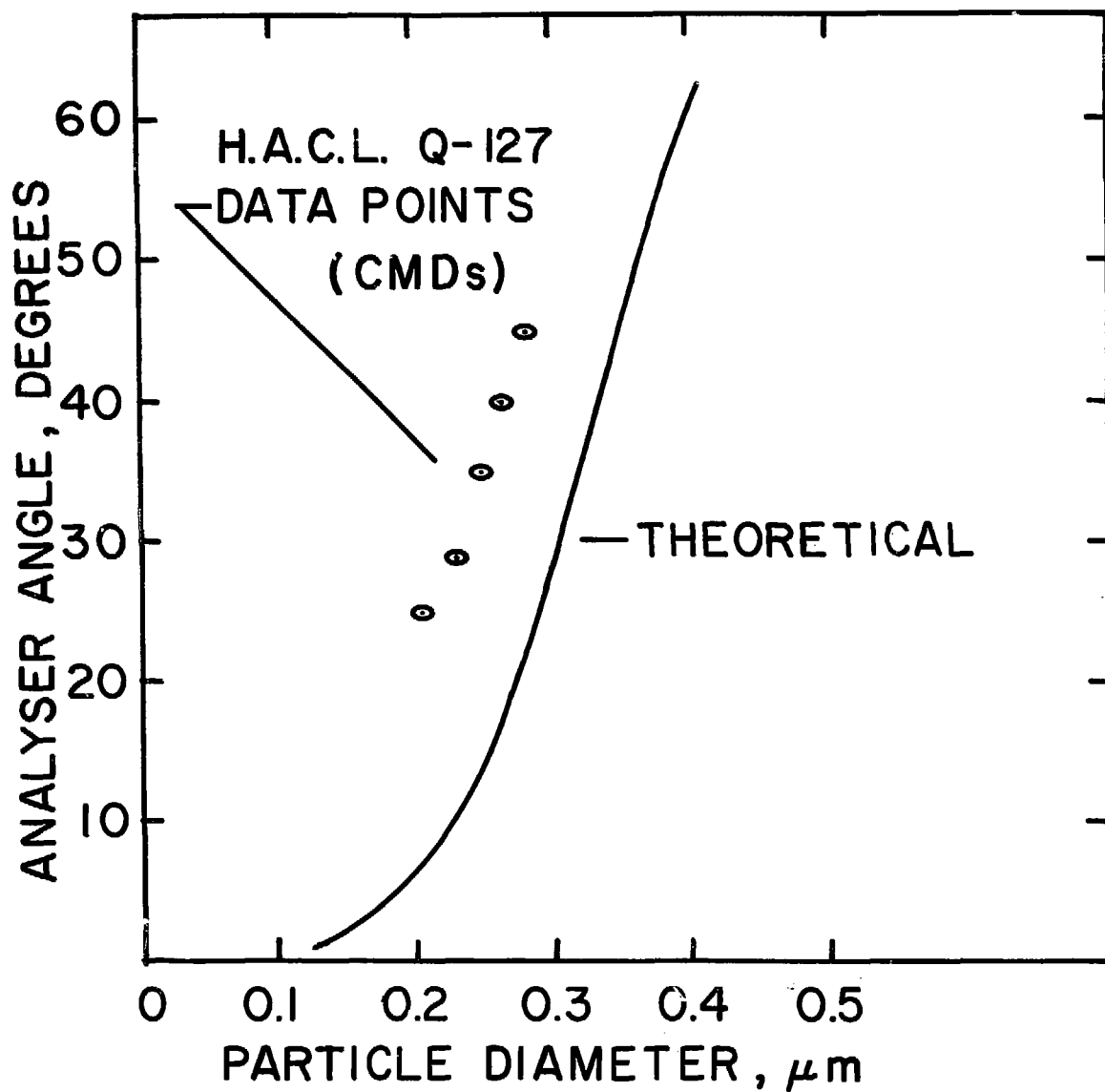


Fig. 6. Comparison of theoretical and measured particle size versus polarization ratio. (Theoretical curve from Sinclair,³ interpolated for $m = 1.483$.)

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average polarization ratio for these polydisperse aerosols. To determine the significance of polydispersity for this measurement, a graph of polarization ratio, i_2/i_1 , versus particle size was constructed in Figure 7 by converting analyzer angle, ϕ , as given by Sinclair³ and shown in Figure 6, to polarization ratio by the relationship,

$$i_2/i_1 = \tan^2 \phi. \quad (1)$$

Over the size range 0.15-0.4 μm , polarization ratio can be approximated by a constant power function of diameter,

$$i_2/i_1 = 4730 d^{8.1}, \quad (2)$$

for diameter in μm .

Table 5. DOP penetration measured at five laboratories.⁵

| Sample | Harvard
(45°) | Harvard
(29°) | Dexter
(29°) | H&V
(29°) | Fiber
Process
(29°) | Rocky
Flats
(29°) |
|--------|------------------|------------------|-----------------|--------------|---------------------------|-------------------------|
| 18 | .018 | .041 | .028 | - | - | .022 |
| 30 | .016 | .038 | .028 | - | - | .0215 |
| 14 | .010 | .027 | .021 | - | - | .015 |
| 11 | .004 | .012 | .009 | - | - | .006 |
| 23 | .001 | .002 | .002 | - | - | .001 |
| 25 | .012 | .022 | .019 | - | - | .014 |
| 34 | .004 | .012 | .009 | - | - | .008 |
| 12 | .007 | .018 | .013 | - | - | .009 |
| S 20 | .035 | .068 | .046 | .049 | .053 | - |
| S 21 | .007 | .016 | .013 | .014 | .015 | - |
| S 22 | .025 | .052 | .038 | .039 | .042 | - |
| S 23 | .005 | .010 | .007 | .008 | .009 | - |
| S 24 | .006 | .014 | .013 | .014 | .019 | - |
| 1A | .008 | .018 | .013 | .015 | - | - |
| 6A | .004 | .013 | .009 | .010 | - | - |
| 20A | .010 | .032 | .028 | .027 | - | - |
| 21A | .005 | .016 | .011 | .012 | - | - |
| 23A | .003 | .009 | .008 | .008 | - | - |
| 24A | .010 | .024 | .017 | .017 | - | - |
| 25A | .012 | .029 | .021 | .021 | - | - |
| 28A | .006 | .016 | .011 | .012 | - | - |
| 29A | .002 | .005 | .005 | .004 | - | - |
| 30A | .016 | .035 | .025 | .026 | - | - |

Analyzer setting shown in parenthesis.

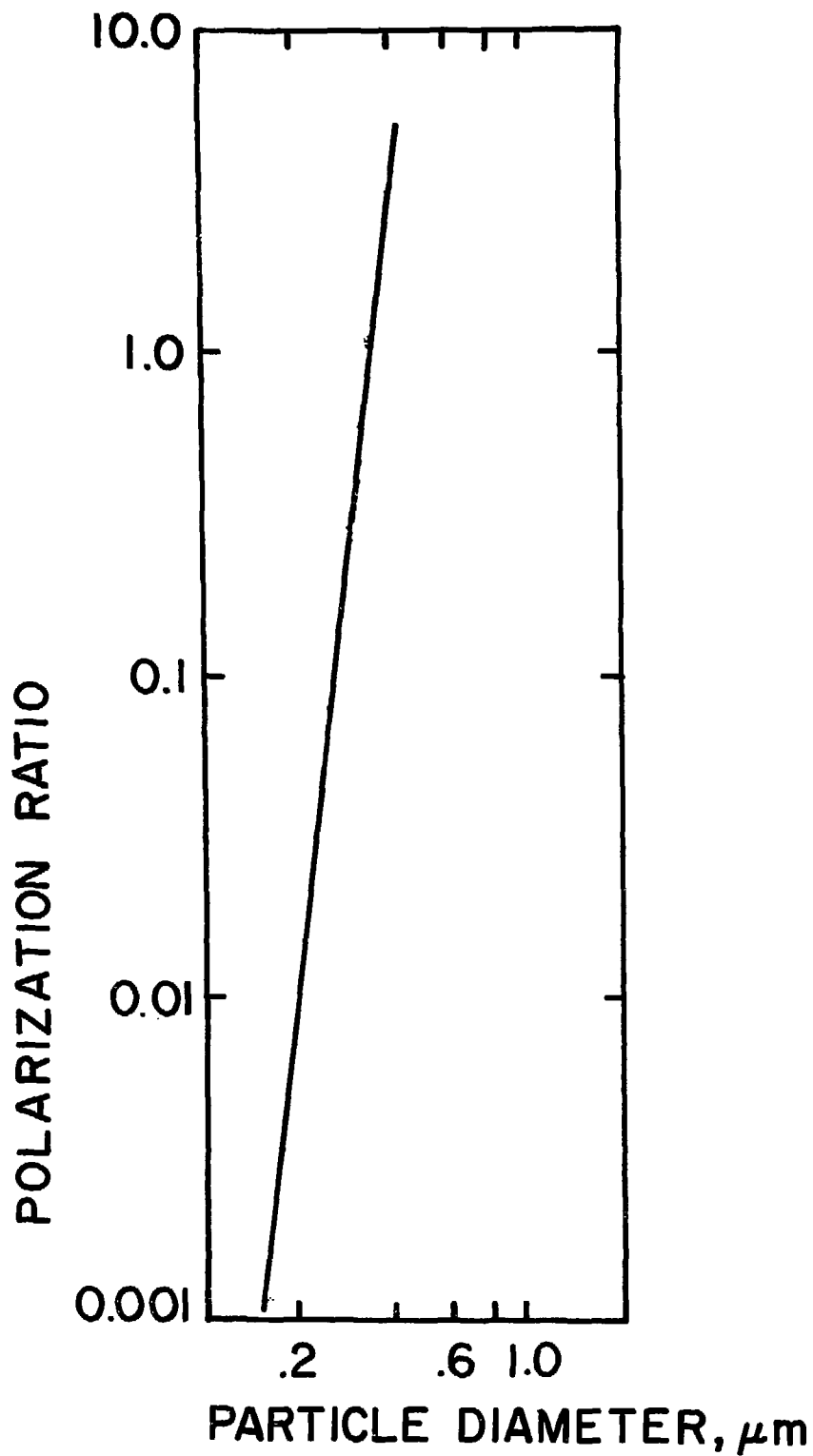


Fig. 7. Polarization ratio versus particle diameter - calculated from Sinclair's curve of analyzer angle vs. diameter³ for $m = 1.483$.

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The mechanical analyzer measures the average polarization ratio which is interpreted in practice as representing the diameter of aerosol particles of uniform size using Sinclair's³ curves of analyzer angle versus monodisperse particle size.

The average polarization ratio is defined as the ratio of the intensities of the horizontal and vertical polarization components of 90° scattered light. For a polydisperse aerosol the average polarization ratio is given by,

$$\overline{PR} = \frac{I_2}{I_1} = \frac{\sum n_j(i_2)_j}{\sum n_j(i_1)_j} \quad (3)$$

and the diameter of average polarization ratio by,

$$d_{\overline{PR}} \approx \left[\frac{\sum n_j(i_2)_j}{\sum n_j(i_1)_j} \right]^{\frac{1}{8.1}} \quad (4)$$

where n_j is the number of particles in the j th size category, i_2 and i_1 are the intensity parameters for horizontal and vertical polarization, respectively, and the summations are for all particle size categories. Values for i_1 and i_2 were calculated with a computer program⁽⁸⁾ and integrated over scattering angles from 88.5° to 91.5° and wavelengths defined by the emission characteristics of the light source (tungsten filament), the transmission of a number 59 wratten filter, and the sensitivity of the S-4 photodetector. Results of calculations using equation (3) are shown in Table 6 for hypothetical aerosols all of which may be produced at an analyzer setting of 29° and interpreted as $d_{\overline{PR}} = 0.3 \mu\text{m}$. Table 6 shows that at a GSD of 1.05, the condition for which Sinclair originally used the OWL ($0.3 \mu\text{m} \pm 10\%$ is taken to mean ± 2 standard deviation), the CMD and $d_{\overline{PR}}$ are indistinguishable.

Table 6. CMD of aerosols with log normal size distribution produced by HOT DOP generator at a mechanical analyzer setting of 29°.

| <u>GSD</u> | <u>CMD</u> |
|------------|------------|
| 1.00 | 0.300 |
| 1.05 | 0.294 |
| 1.10 | 0.286 |
| 1.15 | 0.274 |
| 1.20 | 0.260 |
| 1.25 | 0.246 |
| 1.30 | 0.230 |

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The size distributions measured at Harvard with the PMS analyzer had GSDs of about 1.15. Reference to Table 6 indicates that the GSD does not fully account for the entire difference between the average polarization size of $0.3 \mu\text{m}$ and the measured size of $0.23 \mu\text{m}$. However, an examination of the size distribution for analyzer angles of 29° and 45° , Figures 4 and 5, reveals that the distributions are not perfectly log normal but were slightly enriched at the large particle end of the distribution. Consequently, a more accurate estimate of the effect of polydispersity was made by numerical integration of equation (4) for the actual size distribution obtained at analyzer settings of 25° , 29° , 35° , 40° , and 45° . Results, shown in Table 7, show good agreement between particle size obtained from Sinclair's curve (Figure 6) and the diameter of average polarization ratio calculated from actual size distributions by numerical integration.

Table 7. Results of numerical integration of measured size distributions to obtain diameter of average polarization ratio.

| Analyzer Setting | Measured CMD | Calculated Dia. of Average Polarization Ratio | Dia. from Sinclair's Curve, Figure 6 |
|------------------|--------------|---|--------------------------------------|
| 25° | 0.205 | .270 | .290 |
| 29° | 0.232 | .278 | .302 |
| 35° | 0.250 | .290 | .318 |
| 40° | 0.265 | .302 | .334 |
| 45° | 0.300 | .323 | .346 |

One can conclude from this that the differences between $0.3 \mu\text{m}$ and the measured values of CMD are due primarily to the effects of polydispersity, which is significant, even at these narrow distributions, because the light scattering weighted diameter of average polarization ratio is such an extremely strong function of particle size (i.e., $d^{0.1}$).

Conclusions

- 1) The GSD of DOP aerosol produced by a Q-127 generator has been measured to be about 1.15. The GSD is sufficiently large to affect the interpretation of data from tests designed to use monodisperse aerosol.
- 2) The fact that the CMD is less than $0.3 \mu\text{m}$ when the analyzer setting is 29° is the result of aerosol polydispersity.
- 3) Filter penetration is sensitive to particle size because filter operating conditions are in the impaction region. It was thought when $0.3 \mu\text{m}$ was selected for the test aerosol for HEPA filters that this diameter was the size for maximum penetration and that the sensitivity of penetration measurements would be little affected by small size changes. Later studies showed the size for minimum penetration to be $0.08 \mu\text{m}$ and the size-penetration curve to be very steep in the region of $0.3 \mu\text{m}$.

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- 4) Therefore, when a DOP test aerosol is produced at an analyzer setting of 29°, small variations in GSD produce significant variations in filter penetration unless GSD is less than 1.09. Inasmuch as measured GSD's are closer to 1.15, variations in GSD are believed to be the cause of interlab variability in penetration measurements.
- 5) The following steps would reduce interlab variability:
 - a) Use a test aerosol of smaller particle size that corresponds more closely to the size for maximum penetration.
 - b) Continue to use the 29° polarization ratio but restrict GSD to <1.09. This corresponds to a CMD of 0.285, within 5% of 0.30 μm but means that a method must be devised to produce such an aerosol and to verify that the GSD is <1.09.
 - c) Continue to use the 29° standard but specify that the GSD must be within $\pm .01$ of 1.15. This also means that a method must be devised to monitor GSD accurately at each laboratory.
 - d) Continue to use the standard 29° setting on DOP generators and apply a correction factor to penetration readings that are based on a comparison of filter readings with a single reference lab that all testing laboratories employ for this purpose.

Acknowledgement

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Appendix C FIBER DIAMETER DETERMINATION OF FIBER GLASS SAMPLES

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Abstract

The Kozeny-Carmen equation is the basis of present practice in relating fiber diameter to handsheet ΔP in quality control of fiber glass papers used for filtration. The Naval Research Laboratory developed the following equation relating volume-surface diameter to the measured ΔP :

$$D = \frac{2.3}{\left\{ \frac{\Delta P}{L} \right\}^{1/2}}$$

$$\Delta P = \text{mm H}_2\text{O}$$

$$L = \text{inches sheet thickness}$$

$$D = \text{Volume-surface diameter in microns}$$

This equation assumes a constant linear velocity, a constant porosity in the glass sheet and a constant shape factor in the Kozeny-Carmen equation. We have found that porosity and shape factor vary considerably depending upon preparation techniques and fiber diameter distribution. Thus, direct correlations of handsheet ΔP to fiber diameter can result in considerable error by present practice.

The BET surface area provides us with the most accurate and reproducible and probably the most useful technique for fiber diameter determination. Using this method of surface area measurement, we have found good correlation between handsheet ΔP and surface area only if the effects of porosity are taken into consideration.

By using the Frazier air permeability tester, the volume surface fiber diameter (BET) is correlated to air flow and porosity using the following equation:

$$D = C \left\{ \frac{Q^2 W^3}{L} \right\}^{1/4}$$

where:

Q = flowrate of air (STP) $\text{ft}^3/\text{ft}^2/\text{min}$

W = handsheet sample weight in grams

L = handsheet caliper in mils

C = a constant for this test (0.176)

Introduction

The single most important parameter of glass microfiber products is the average fiber diameter of a given lot. The various codes are designated by ranges of fiber diameter and are priced accordingly. There are several different definitions for "average" fiber diameter, but the one of most consequence to our customers is the volume-surface average diameter. This is defined as the diameter that a theoretical fiber would require so that it would have the same surface area per unit weight as an actual fiber sample. Consequently, all of our reference fiber diameters were derived from

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surface areas obtained using BET krypton adsorption.

Currently, the Williams Freeness Test is used to run regular Quality Control checks on microfiber production. This method empirically relates the drainage time of a fiber slurry of known pH, temperature and concentration to its volume-surface average diameter. Several recent refinements on this test procedure have perfected it to a point where results on any given sample of fiber can be reproduced within ± 1.0 percent.

In spite of the precision in this method, it has become strikingly obvious that two different samples can have identical Williams Freeness drain times yet have different volume-surface average fiber diameters. This point indicates that liquid drainage, in the form of the Williams Freeness Test, is dependent on more than just the volume-surface fiber diameter. We believe that some of the error involved in drainage tests could either be accounted for or eliminated in a refined ΔP test, using air permeability as an indirect measurement of surface area. This concept is already in use in various test equipment, such as the Fisher Sub-Sieve Sizer, the Blaine Permeability Tester, and the Scheffield Micronaire Tester. The ΔP test is also used as a Quality Control Test for air filtration blankets. The intention of the paper is to refine the basic principle of these ΔP tests into a form that will give a true indication of the volume-surface average fiber diameter.

Discussion

The basic law governing one-dimensional flow through a permeable media is given by Darcy to be

$$\frac{Q}{A} = \frac{B_o}{u} \frac{\Delta P}{L}$$

Q = flowrate

A = area of sample

B_o = permeability coefficient

u = fluid viscosity

L = media thickness

This equation is applicable in cases of viscous streamline and laminar flow of liquids, and applies to gases when ΔP is small when compared to P , the ambient pressure. B_o , however, is not a constant, and is generally expressed as a function of E , the porosity, S_o , the surface area per unit volume of the sample, and k , an empirical constant. There have been numerous solutions to this function but we shall deal with only two.

The most famous treatment of this equation is the Kozeny-Carman form. This is a theoretical formulation based on a relationship analogous to the Poiseuille law of flow through a circular pipe, except that the hydraulic radius is used in the equation. The hydraulic radius is defined as the volume of sample filled with liquid divided by the wetted surface. Using this principle, the Kozeny-Carman equation takes the form:

$$B_o = \frac{1}{K S_o^2} \frac{E^3}{(1-E)^2}$$

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$$\text{and } \frac{Q}{A} = \frac{\Delta P}{K S_o^2 u L} \frac{E^3}{(1-E)^2}$$

For non-porous fiber samples, $S_o = 4/D$, where D equals the volume-surface average fiber diameter. Therefore, the Kozeny-Carman equation can be re-written in the form

$$D = \left[\frac{16 Q k u L (1-E)^2}{A \Delta P E^3} \right]^{1/2}$$

where D is determined solely in measurable quantities.

There are certain limitations to this equation, however. First, the equation was derived for randomly packed samples. Fibers in most samples tend to be oriented, often severely so. Also, this treatment assumes a uniform pore size, whereas there is actually a distribution of pore sizes. But of most importance is the relationship of k to the porosity, E . For oriented fiber beds, k remains constant only when E is less than 0.8. As E approaches 1.0, k rises rapidly. Table I gives some typical values of E versus k .

The Naval Research Laboratory developed a modification of this approach. A handsheet of the fiber in question is subjected to air with a constant linear velocity Q/A (14.2 cm/sec) and the resulting pressure drop is measured. By assuming a constant E for identically prepared samples, NRL reduced the Kozeny-Carman equation to:

$$D = \frac{2.3}{\left\{ \frac{\Delta P}{L} \right\}^{1/2}} \quad \begin{array}{ll} \{P\} & = \text{mm H}_2\text{O} \\ \{L\} & = \text{mils} \\ \{D\} & = \text{microns} \end{array}$$

This equation in effect assumes that the combination of terms $k(1-E)^2(E)^{-3}$ is constant. Looking at the third column in Table I, we see that this is not the case. This indicates that the NRL equation is valid only when the porosity of the sample is the same as the assumed value. However, even identically prepared samples can have varied porosities.

There is also an empirical solution to the Darcy permeability coefficient. Davies has determined that for beds of fibers with porosities between 0.7 to 0.99:

$$B_o = \frac{D^2}{64 (1-E)^{1.5} \{1 + 56 (1-E)^3\}}$$

Furthermore, for E greater than 0.9, B_o is closely approximated by:

$$B_o = \frac{D^2}{64 (1-E)^{1.5}}$$

This equation does not contain K . When combined with the Darcy equation we obtain:

$$D = \left\{ \frac{64 Q u L (1-E)^{1.5}}{A \Delta P} \right\}^{1/2}$$

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Since this is an empirical solution, its accuracy depends on the method of obtaining the reference diameters. We found that a better correlation with BET - determined fiber diameters is obtained by replacing the "64" with "36." The diameter is then given by:

$$D = \left\{ \frac{36 Q u L (1-E)^{1.5}}{A \Delta P} \right\}^{1/2}$$

This equation allows calculation of the fiber diameter from strictly measurable quantities by fixing either Q or ΔP and measuring the other. The Naval Research Laboratory chose to fix the flowrate and measure ΔP . However, we chose to fix the pressure differential and measure the flowrate, using a Frazier Permeability Tester adapted with a 1 square inch, wire mesh-supported opening. There are several reasons for this decision:

1. The calculated diameters are very sensitive to error in pressure readings at low values of ΔP (less than 50 mm H₂O). This sensitivity decreases as ΔP increases. Using the Frazier Tester, a ΔP can be chosen that is above this sensitive region. For this work we chose a ΔP equal to 5 inches (127 mm) H₂O, which is roughly in the middle of the range of ΔP for the NRL test. The wide range of flowrates encountered can be measured readily on the Frazier Tester by changing the calibrated orifices in the tester.
2. The Darcy equation for flow through porous media needs a correction factor for compressible gases,

$$\text{Correction Factor} = \frac{P_1 + 1/2 \Delta P}{P_1}$$

(the average pressure through the medium divided by the ambient pressure), which is usually assumed to be negligible for small ΔP . By fixing the ΔP at a specified value, this term becomes a constant and the effects of ignoring it in the theory can be accounted for in an equipment calibration constant.

3. The wide range of measurement possible using the different orifices allows considerable latitude in specifying other test parameters. The most notable of these is that thicker samples were used (approximately four times that of a standard handsheet) to minimize the effects of error in measuring L, the sample caliper.

The test samples were prepared by running a revised William Freeness Test on each sample of fiber. After measuring the Williams Freeness time, the samples were allowed to finish draining, and then were removed from the screen using blotter paper, pressed with a roller similar to a British Handsheet roller, and then allowed to dry. The actual test samples were 2.25 inch diameter disks that were punched from the center of the Williams Freeness pads to minimize the effect of fiber build-up around the outside of the Williams Freeness Tester. The samples were weighed, measured for caliper on a bigfoot micrometer with light downward pressure to minimize compression, and then measured on the Frazier tester. The data was analyzed using several methods that are discussed in the next section. An outline of our

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procedure is included in Appendix I.

Results

The primary difference between the Kozeny-Carman (NRL modification) and the Davies solutions is how they deal with the effects of porosity. The NRL solution assumes all fiber samples would have the same porosity if prepared in identical manner, regardless of the average diameter. However, the packing of fiber is greatly affected by the fiber diameter distribution. Samples with broad distributions should pack more tightly than those with narrow distributions. Therefore, if the resulting change in porosity is not accounted for, the results of a ΔP fiber diameter determination would be in error.

It was necessary to test whether the fiber diameter distribution has a real and significant effect on ΔP measurements. The actual fiber diameter distribution is difficult to measure, so we prepared a series of samples with progressively broader relative distributions. Samples of Codes 100, 106 and 110 fiber were chosen so that a blend of Codes 100 and 110 would have the same volume-surface average fiber diameter as the Code 106. Samples of 106 with increasing proportions of 100/110 mixture were prepared so that they would have the same volume surface fiber diameter with progressively broader distributions. Table II shows the results of these tests using both equations to evaluate the data.

BET determined fiber diameters confirmed that the blends used in this series did in fact have the same average diameters. The effect of fiber diameter distribution on the porosity, E , is shown. As expected, E decreases as the distribution broadens. This is reflected in the Williams Freeness data. Our samples had diameters of 0.70 microns, but the Williams Freeness diameters for the artificially broadened samples dropped as low as 0.65 microns. Also, it must be noted that our sample of 100% Code 106 had a diameter distribution of its own, so that it is quite possible that a 0.70 average diameter fiber sample with a narrower distribution could appear to be even coarser than 0.70 microns on a Williams Freeness Tester. Therefore, unless there is additional information about fiber diameter distribution or porosity, the Williams Freeness Test is only valid as an approximation.

The same samples were then checked using the conditions specified by the NRL method. The resulting fiber diameters are also listed in Table II. They show the same trend as the Williams Freeness data; narrow distributions cause the NRL ΔP test to indicate larger diameters than do broad distributions, even though the samples have the same volume-surface average diameter. Therefore the NRL test is susceptible to the same variation in sample diameter distribution as the Williams Freeness Test.

The last column shows the effect of accounting for the change in porosity caused by varying diameter distributions. This data, analyzed using the Davies equation, does not exhibit the same distinct relationship between diameter and distribution as the first two methods. (Figure I). The scatter among the data points appears to be more pronounced for the Davies equation, probably due to the

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additional measured parameters. However, the standard deviation was 2.8 percent for the Williams Freeness data, 6.3 percent for the NRL ΔP data, and 1.5 percent for the data analyzed with the Davies equation. Clearly, the possible increase in experimental error using the Davies equation is more than offset by reducing the error caused by the assumption of constant porosity. It should be noted that these results are consistent with results obtained on handsheets made on a British mold and a Woods and Noble mold, but that use of Williams Freeness disks produced more consistent data.

To determine how well the Kozeny-Carman and the Davies equation correlate with BET results over a wide range of fiber diameters, uniform samples of Codes 100, 102, 104, 106, 108A, 108B, and 110 were prepared by dispersing small bits of each code in a turbulent airstream. Pandom samples of each were measured on BET equipment to determine their volume-surface fiber diameters and to confirm their homogeneity. Triplicate samples of each grade were prepared on the Williams Freeness Tester and then tested on the Frazier Permeability Tester. The data was analyzed with the Davies equation and with the full Kozeny-Carman equation. Table III shows the mean fiber diameter of each code determined by these methods and their corresponding surface areas. ($d = 1.53/S.A.$). There does not appear to be a good correlation of either the Kozeny-Carman or the Davies diameters with the BET diameters. The reason for this can be seen by plotting surface areas for each against the BET surface areas. Both curves are linear to $1.5 \text{ m}^2/\text{g}$, which corresponds to about 1 micron. However, both fall off as the fiber samples become finer, suggesting that neither method of analyzing allows for direct calculation of surface areas and corresponding fiber diameter less than one micron. It is necessary to use a graphical correlation between air permeability diameters and BET diameters similar to the one relating Williams Freeness times to BET diameters. While either the Davies or the Kozeny-Carman equation could be used for this purpose, the simplified form (Appendix II) of the Davies equation was chosen because it is much easier to work with.

$$D = 0.176 \left\{ \frac{Q^2 W^3}{L} \right\}^{1/4}$$

D = fiber diameter, microns
Q = flowrate, $\text{ft}^3/\text{ft}^2\text{-min}$
W = sample weight, grams
L = caliper, inches

The bracketed term $\left\{ \frac{Q^2 W^3}{L} \right\}$ has a one-to-one correspondence to the

fiber diameter and is called the Frazier number. These numbers were calculated for the seven fiber Codes and are listed in Table IV. By plotting Frazier numbers against BET fiber diameters, a calibration curve is obtained (Figure III). Therefore, by measuring the weight and caliper of, and flowrate through, a prepared fiber sample, and calculating the corresponding Frazier number, the volume-surface average fiber diameter can be obtained graphically without error caused by differences in porosity and fiber diameter distribution.

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Conclusions

From this work we can conclude the following:

1. The porosity of a bed of fibers must be accounted for in determining the volume-surface average fiber diameter using fluid flow techniques (drainage or air permeability).
2. Neither the Williams Freeness Test nor the Naval Research Laboratories ΔP test adequately accounts for the variation in fiber diameter distribution inherent in production fiber samples.
3. Evaluations using the Davies modification of the Darcy equation does account for the variation in fiber diameter distribution.
4. The primary term in the Davies equation, $\frac{Q^2 W^3}{L}$, can be correlated graphically with the BET derived volume-surface average fiber diameters.

Appendix I

Scope

This method is used to determine the true volume-surface average fiber diameter unbiased by diameter distribution effects. This value can be compared to the Williams Freeness value obtained during the procedure.

Equipment

1. Williams Freeness Tester with Timer and 80 mesh screen plate (Williams Apparatus Co., Watertown, N.Y.).
2. Balance, sensitive to ± 0.01 g.
3. Waring Blender, Model 1120.
4. Thermometer.
5. Deionized water.
6. 10 ml graduate.
7. 0.1N NaOH.
8. Blotter paper, 4 in. x 4 in.
9. British Handsheet Roller or Equivalent.
10. 2.25 in. I.D. circular punch.
11. Micrometer, sensitive to 0.5 mils.

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12. Frazier High Pressure Differential Model Air Permeability Tester with 1 in.² adapter plates with open mesh screen support. (Frazier Precision Instrument Co., Gaithersburg, Maryland).
13. Small circle of 8 mesh screening to fit the bottom of the Williams Freeness Tester.

Procedure

1. Weigh 1.00 $\pm$ 0.01 gram of clean, dry fiber. For fiber diameters greater than 1.01 microns, weight out 2.00 $\pm$ 0.01 grams of fiber.
2. Pull the weighed fiber apart into several pieces and place in a clean empty Waring Blender jar.
3. Add approximately 500 ml. deionized H₂O to the jar, and then add 10 ml. of the 0.1N NaOH.
4. Turn blender on high speed and beat the fiber slurry for exactly two minutes.
5. Place the small screen in the cone of the tester. With the valve closed, fill the cone with deionized water until a meniscus forms above the base. Drop the screen plate onto the meniscus, taking care that no bubbles form beneath the screen, then close the tester and clamp in place.
6. Add approximately 100 ml. H₂O to the tester to cover the screen. Then transfer the fiber slurry to the tester, using small amounts of deionized H₂O to rinse all fiber from the Waring blender lid and jar. Add additional H₂O to the tester to bring the slurry level to the 1000 ml. mark.
7. Stir the slurry slightly to assure good dispersion of the fiber. Check slurry temperature (should be 68-72°F).
8. Reset timer to ZERO.
9. Open the drain valve rapidly. When slurry level reaches the zero mark, close the drain valve rapidly.
10. Record elapsed time noted on the timer. (This time can be converted to fiber diameter by use of calibration graphs).
11. Reopen valve to allow fiber slurry to finish draining. Open the tester and remove excess H₂O from the pad using blotting paper and hand pressure.
12. Separate the pad from the screen and place between fresh sheets of blotter paper. Press with the roller using three passes, then dry the samples (with blotter paper) in an oven. The fiber pads will separate from the paper as they dry.
13. Remove a disk from the center of the fiber pad using the 2.25 in. circular punch.

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14. Weigh the disk to the nearest 0.01 gram and measure its caliper to the nearest 0.5 mil.
15. Place disk over the wire mesh-backed 1 square inch opening in the Frazier Tester. Clamp tightly.
16. Install an orifice in the tester. The appropriate size orifice is determined by trial and error for each grade of fiber.
17. Start the motor and increase its speed by means of the Variac control until a reading of 5.0 inches H₂O is obtained on the left hand manometer, while making sure that the right hand manometer does not overflow. The right hand manometer should read between 4 and 20 inches of manometer oil (Sp. Gr. =0.827) for most accurate results. If not, a different orifice should be selected.
18. Once the right hand manometer reading is obtained, its corresponding flow rate is obtained from the orifice calibration tables.
19. Calculate the following value.

$$N = \frac{\text{Flow rate min-ft}^2 \left\{ \frac{\text{ft}^3}{2} \right\}^2 \text{ weight (g)}^3}{\text{Caliper (inches)}}^3$$
20. Refer to Figure III to determine the average fiber diameter corresponding to the calculated value, N.

Notes:

1. A separate calibration curve should be prepared relating BET diameter to ΔP results for each specific set of test equipment.
2. All openings on the Frazier Tester should be sealed, the orifices should be tight, the tester should be level and the manometers should be filled to zero points before running samples.

Appendix II Derivation of final forms of Kozeny-Carman and Davies equations.

The difference between the Kozeny-Carman equation and the Davies equation lies in their expression of B_o , the permeability coefficient in the Darcy equation.

1. Kozeny-Carman $B_o = \frac{d^2}{16K} \frac{E^3}{(1-E)^2}$
2. Davies $B_o = \frac{d^2}{36 (1-E)^{1.5} \{1+56 (1-E)^3\}}$

As E, the porosity, becomes large, above 0.9, the Davies expression is closely approximated by:

3. $B_o = \frac{d^2}{36 (1-E)^{1.5}}$

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The porosity is defined as the ratio of the volume of free space in a sample to the total volume of the sample. This can be expressed as:

$$4. \quad E = \frac{AL - W/\rho}{AL}$$

A = area of sample

L = thickness of sample

W = weight of sample

ρ = density of sample material

Substituting equation 4 into 1 and 3 we obtain:

$$\text{Kozeny-Carman} \quad B_o = \frac{d^2 (AL - W/\rho)^3}{16 K A L (W/\rho)^2}$$

$$\text{Davies} \quad B_o = \frac{d^2}{36} \left(\frac{AL}{W/\rho} \right)^{1.5}$$

When substituted into the Darcy equation and solved for the diameter, we obtain:

$$\text{Kozeny-Carman} \quad d = \left\{ \frac{16 Q K A L^2 \eta (W/\rho)^2}{A^1 \Delta P (AL - W/\rho)^3} \right\}^{1/2}$$

$$\text{Davies} \quad d = \left\{ \frac{36 Q \eta L}{A^1 \Delta P} \frac{W/\rho}{AL} \right\}^{1.5} \quad 1/2$$

where d = diameter of fiber, microns

Q = volumetric flowrate, ft³/ft² min

K = constant (Kozeny-Carman only)

A = area of sample, 25.65 cm²

L = caliper of sample, inches

W = weight of sample, grams

ρ = density of sample material, g/cc

η = viscosity of air at room temperature, 1.84 x 10⁻⁴ poise

A¹ = area of sample tested for air permeability, 1-in.²

ΔP = pressure drop across the sample, 5-in. H₂O

Using the numerical values specified in the above list and appropriate conversion factors, these equations reduce to:

$$\text{Kozeny-Carman} \quad d = 9.05 \left\{ \frac{Q W^2 L^2}{6.45 L - (W/\rho)} \right\}^{1/2}$$

$$\text{Davies} \quad d = 0.362 \left\{ \frac{Q^2 W^3}{L \rho^3} \right\}^{1/4}$$

For 475 glass, ρ equals 2.61 g/cc. Therefore, when applied to our glass microfibers, the Davies equation can be written:

$$d = 0.176 \left\{ \frac{Q^2 W^3}{L} \right\}^{1/4}$$

Notes:

1. The background for these equations can be found in The Flow of Gases Through Porous Media; Carman, P.C., Academic Press, Inc.

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N.Y., 1956.

- The value of K used in the Kozeny-Carman equation was 4.25, designated as the "shape factor" in the NRL adaption.

Appendix III Table I Typical k versus e for porous media

| E | K | $\frac{K(1-E)^2}{E^3}$ |
|------|------|------------------------|
| 0.6 | 4.3 | 3.18 |
| 0.7 | 4.4 | 1.15 |
| 0.8 | 5.4 | 0.422 |
| 0.9 | 8.8 | 0.121 |
| 0.95 | 15.2 | 0.044 |
| 0.98 | 32.8 | 0.014 |
| 0.99 | 61.0 | 0.006 |

Table II

| Composition | | | Porosity | Willaims | Freeness | NRL ΔP | Davies |
|-------------|-------|-------|----------|--------------|----------|----------------|------------|
| % 100 | % 106 | % 110 | E | Diameter | | Diameter | Diameter |
| 0 | 100 | 0 | 0.928 | 0.705microns | | 0.805microns | 0.780 mic. |
| 9 | 80 | 111 | 0.924 | 0.690 | | 0.771 | 0.772 |
| 18 | 60 | 22 | 0.919 | 0.680 | | 0.739 | 0.802 |
| 27 | 40 | 33 | 0.916 | 0.665 | | 0.727 | 0.795 |
| 36 | 20 | 44 | 0.911 | 0.660 | | 0.684 | 0.802 |
| 45 | 0 | 55 | 0.908 | 0.650 | | 0.673 | 0.780 |

Table III

| Code | BET | | Kozeny-Carman | | Davies | |
|------|----------|------------------------|---------------|------------------------|----------|------------------------|
| | Diameter | Surface Area | Diameter | Surface Area | Diameter | Surface Area |
| 100 | 0.343 | 4.46 m ² /g | 0.41mic. | 3.71 m ² /g | 0.50mic. | 3.06 m ² /g |
| 102 | 0.401 | 3.81 | 0.43 | 3.59 | 0.54 | 2.83 |
| 104 | 0.520 | 2.94 | 0.52 | 2.94 | 0.65 | 2.35 |
| 106 | 0.697 | 2.19 | 0.62 | 2.54 | 0.78 | 1.96 |
| 108A | 0.992 | 1.54 | 0.76 | 2.00 | 1.00 | 1.53 |
| 108B | 1.86 | 0.82 | 1.36 | 1.12 | 1.83 | 0.84 |
| 110 | 3.93 | 0.39 | 2.74 | 0.56 | 3.58 | 0.43 |

Table IV

| Code | BET Fiber Diameter | Frazier Number |
|------|--------------------|----------------|
| 100 | 0.34 microns | 65.6 |
| 102 | 0.40 | 88.0 |
| 104 | 0.52 | 186 |
| 106 | 0.70 | 380 |
| 108A | 0.99 | 1050 |
| 108B | 1.86 | 11,600 |
| 110 | 3.87 | 169,000 |

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Figure I Distribution versus fiber diameter davies, NRL ΔP , Williams Freeness

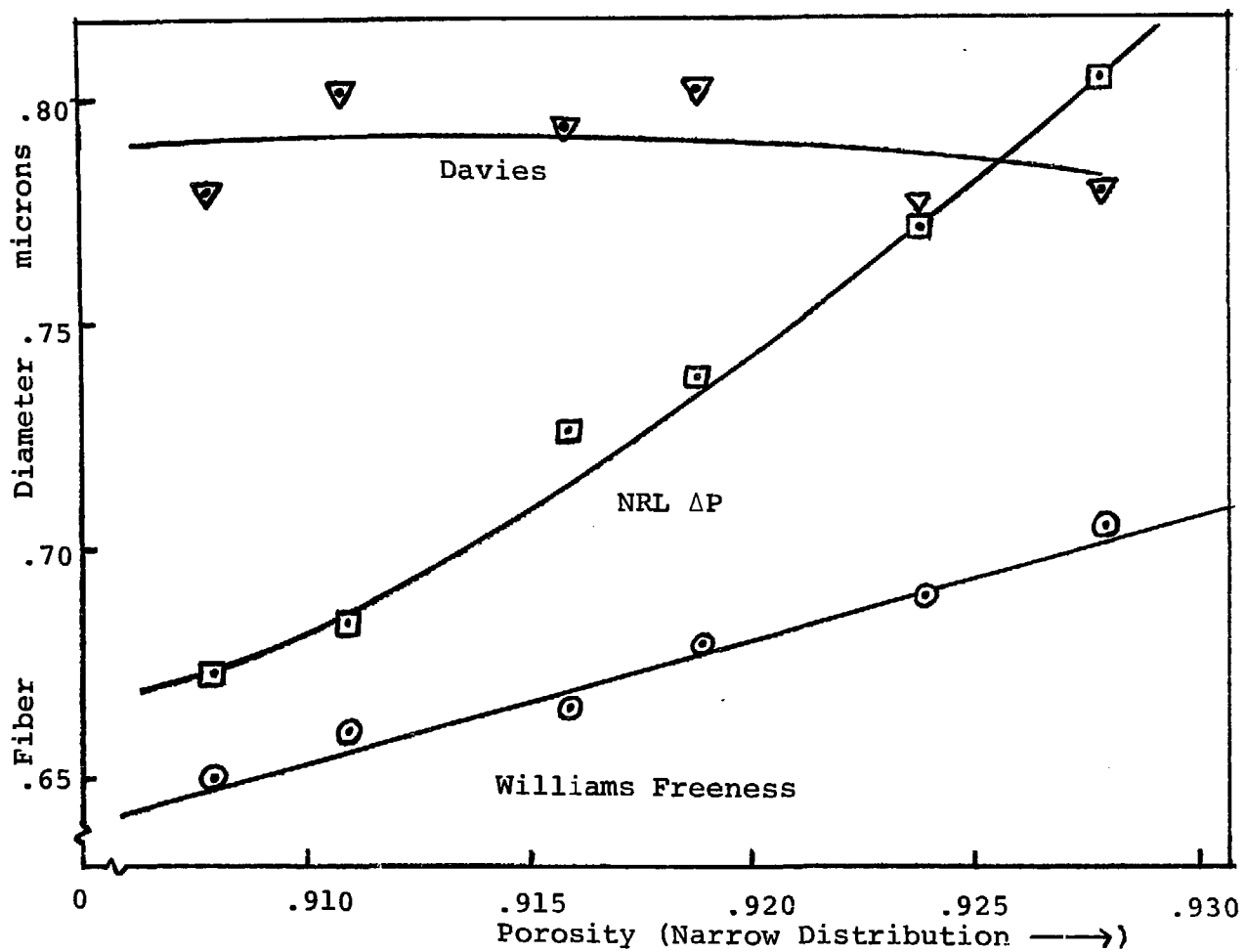


Figure II Surface Area, BET versus air permeability

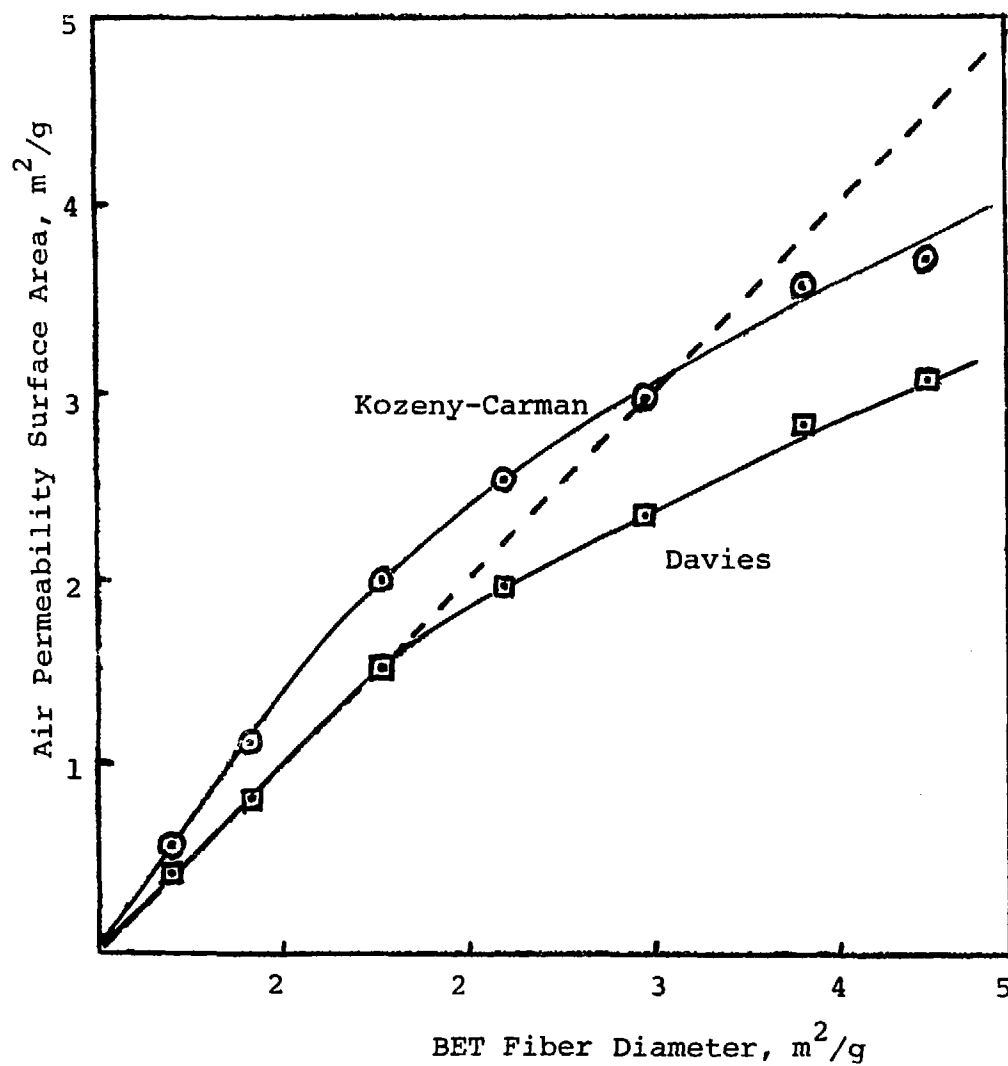
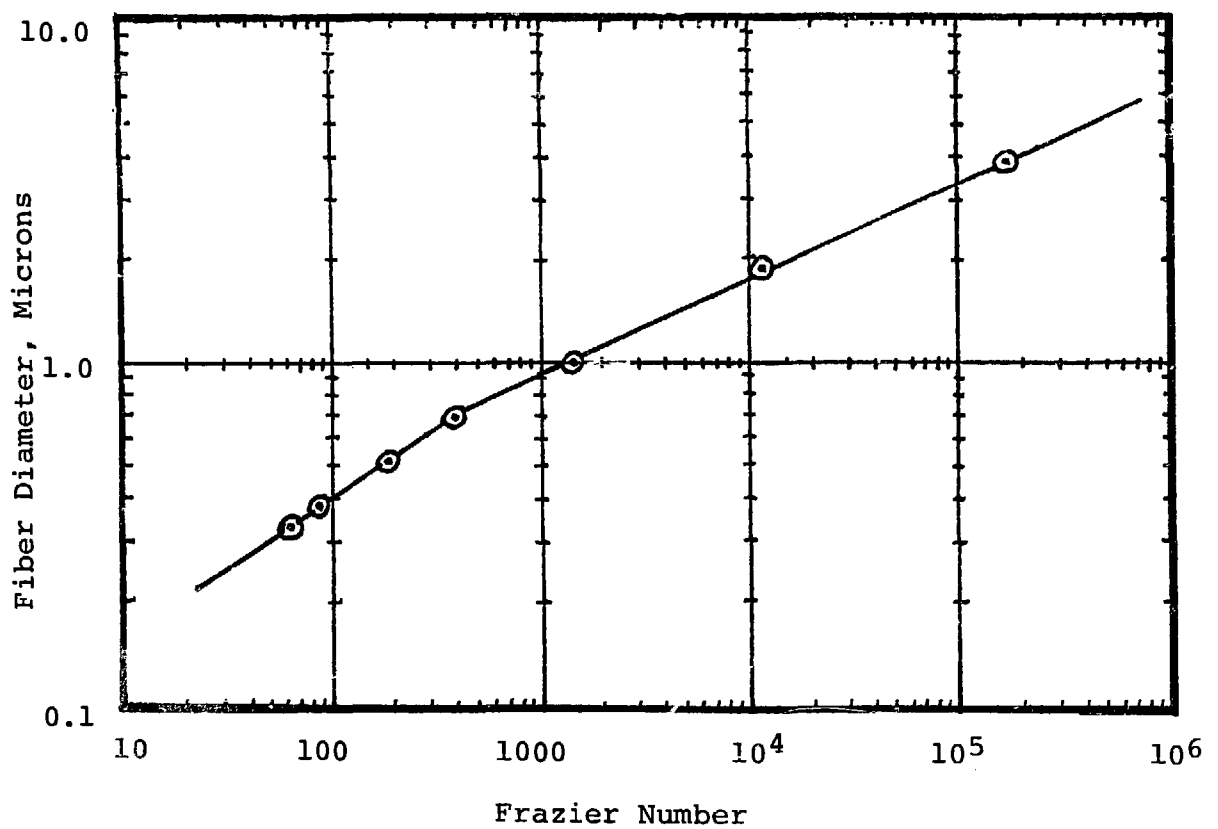


Figure III Frazier number versus fiber diameter



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EVALUATION OF DATA FROM HEPA FILTER QUALITY ASSURANCE TESTING STATIONS*

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In Revision 1 to Regulatory Guide 1.52, issued in July 1976, the NRC recommended that high efficiency particulate air (HEPA) filters for use in engineered safety feature (ESF) atmosphere cleanup systems be visually inspected and dioctyl phthalate (DOP) tested at either of two Department of Energy (DOE) operated QA Filter Testing Stations prior to their installation and use in commercial nuclear power plants. This recommendation was predicated on the practice initiated by DOE and its predecessor, the Atomic Energy Commission (AEC), in the early 1960's, and remains in effect today for all HEPA filters designated for use in DOE facilities. This practice was initiated because filter vendors were unable to consistently provide a HEPA filter that would meet the stringent requirements established by DOE and NRC and its predecessor the AEC.

In 1977, the NRC staff undertook a program to revise Regulatory Guide 1.52 to reflect recently issued industry standards (e.g., ANSI N509 and N510) and current industry practices. Revision 2 to Regulatory Guide 1.52 was formally issued in March 1978. In conducting this review, we reevaluated the recommendation that HEPA filters, intended for use in ESF systems in commercial nuclear power plants, be routinely tested at the DOE-QA Filter Testing Stations. As part of this evaluation we conducted a detailed analysis of the filter test results recorded by the two QA Testing Stations during the period 1971 to 1977. This paper will summarize the results of our analysis and explain the rationale for deleting the requirement that all HEPA filters intended for use in ESF systems be tested at the QA Testing Station prior to installation in nuclear power plants. Revision 2 to Regulatory Guide 1.52 (Section 3.d) recommends that HEPA filters be tested for penetration of DOP in accordance with the provisions of MIL-F-51068 and MIL-STD-282. In addition, it stipulates that the quality assurance requirements of Appendix B, "Quality Assurance Criteria for Nuclear Power Plants and Fuel Reprocessing Plants," to 10 CFR Part 50, be applied to all activities affecting the safety-related functions of HEPA filters. In effect, this revision places greater reliance on the filter vendors who already have the capability to construct HEPA filters in accordance with the requirements of ANSI N509 and to test their filters in accordance with the military specifications noted above.

During the period 1971 to 1977, the Oak Ridge and Hanford QA Testing Stations tested over 88,000 HEPA filters of all sizes and of this number, on the average, approximately 6% were rejected. Table 1 shows the number of filters tested of the four principal filter manufacturers, the number and percent of filters rejected, and the causes for these rejections. Of the filters tested, the principle cause of rejection was the failure of the filter to meet the penetration requirements. The number rejected because of excessive penetration ranged from a low of 16% to a high of 47%. What is not known, but would be most helpful, would be the measured penetration value compared to the acceptance criteria. This would permit an assessment of how far the penetration values exceed the acceptance criteria. To our knowledge, no error bar has been established for these tests, so the absolute value

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is imposed as the criteria for acceptance or rejection. Perhaps this is something that should be considered, since there certainly must be some error associated with the calibration and operation of the penetrometers.

As shown in Table 2, of the 88,000 filters tested, 30,966 filters had a rated capacity of 1000 cfm and less than 4% were rejected (1,167). Of these filters, less than 1,600 were specifically intended for use in ESF systems. Table 3 shows the number of filters tested from the four principal vendors, the number and percent of filters rejected, and the causes of these rejections. Of the 1,600 filters tested, approximately 1.5% were rejected due to failure to pass the penetration test or because of defective medium.

Table 4 shows the number of all types of filters tested at either DOE-QA Testing Stations, the percent rejected and the causes of these rejections. It is interesting to note that of all the filters tested, the 1000 cfm filters performed better than other filters tested (3.8% vs 6.4%).

Table 5 shows the total number of filters tested at Hanford for the four principal vendors, while Table 6 shows the results of 1000 cfm filters tested at this facility. Table 7 shows the total number of filters tested at Oak Ridge for the four principal vendors, while Table 8 shows the results of the 1000 cfm filters tested at this facility. Table 9, 10, 11, and 12 are included to show the semi-annual totals of the four filter manufacturers. For the period January 1, 1971 to September 30, 1977, rejection rates ranged from a low of 2% for vendor D to a high of 5.3% for vendor A for the filters tested at Oak Ridge, and from a low of 7.3% for vendor C to a high of 13.8% for vendor A for filters tested at Hanford. The reason for the higher rejection rate for filters tested at Hanford is not known. During this reporting period, Oak Ridge tested approximately 42,000 filters, while Hanford tested approximately 46,000. Since both facilities tested essentially the same number of filters, one could speculate that perhaps Hanford has a more rigorous inspection program or that there may be a difference in the acceptance criteria used by each facility. Whatever the reason, the wide difference should be determined.

In our opinion, the results of this evaluation show that the mandatory confirmatory tests at the DOE-QA Filter Testing Stations are not required for filters intended for use in ESF systems in nuclear power plants. We plan to monitor the test results from these facilities on a periodic basis to look for trends that may indicate a reversal in the presently low rejection rates.

In addition to the testing results, we also considered the cost to the industry of having these filters inspected at the QA Testing Stations prior to use in commercial nuclear power plants. For a typical power plant, we considered three ESF systems (e.g., BWR-standby gas treatment system, fuel handling system, and control room system) each having a rated capacity of 10,000 cfm and each system redundant. We considered that the cost of the filters are increased by approximately 30% (\$125 to \$160). Assuming once-a-year change out of these redundant systems (60 filters) each reactor could realize a savings of \$2,100 or \$63,000 over the expected 30 year operating life of the plant. Equivalently, for a projected 200 operating reactors in 1985, a savings of \$420,000 per year to the nuclear industry can be predicted. Based on the small number of filters rejected as shown previously, the additional cost associated with testing of these filters to assure they satisfy MIL-51068D before installation is questionable. As a result of this evaluation, the NRC has deleted the requirement that all filters be tested at a DOE-QA Filter Testing Station prior to use in ESF systems installed in nuclear power plants. We would like to emphasize that this revision affects only NRC licensed nuclear power reactors and does not have an impact on any DOE facilities.

TABLE 1 - All Filters Tested At
Either DOE Filter Test Facility

| | | | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|---------------|------------------------------|--|--------|----------------|-------|--------|---------------|
| MANUFACTURER | NUMBER TESTED | NUMBER REJECTED (% REJECTED) | PENETRATION | MEDIUM | CARRIER DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 13068 | 1594 (12.2%) | 264 | 249 | 76 | 617 | 254 | 134 |
| B | 10973 | 636 (5.8%) | 258 | 107 | 41 | 47 | 106 | 77 |
| C | 46701 | 2462 (5.3%) | 1121 | 432 | 59 | 193 | 418 | 239 |
| D | 17536 | 1089 (6.2%) | 132 | 499 | 72 | 165 | 21 | 200 |
| TOTAL | 88278 | 5781 (6.5%) | 1775 | 1287 | 248 | 1022 | 799 | 643 |

**TABLE 2 - Filters of 1000 CFM Capacity
Tested At Either DOE Filter Test
Facility**

| MANUFACTURER | NUMBER
TESTED | NUMBER
REJECTED
(% REJECTED) | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|------------------|------------------------------------|--|--------|-------------------|-------|--------|---------------|
| | | | PENETRATION | MEDIUM | CARRIER
DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 3573 | 371
(10.4%) | 106 | 72 | 38 | 45 | 44 | 66 |
| B | 1920 | 105
(5.5%) | 69 | 12 | 17 | 2 | 1 | 4 |
| C | 18061 | 424
(2.3%) | 191 | 80 | 21 | 66 | 9 | 57 |
| D | 7412 | 267
(3.6%) | 5 | 112 | 14 | 29 | 3 | 104 |
| TOTAL | 30966 | 1167
(3.8%) | 371 | 276 | 90 | 142 | 57 | 231 |

TABLE 3 - Filters of Capacity Greater Than or Equal to 1000 cfm for Nuclear Applications Tested at Either DOE Filter Test Facility

| | | | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|---------------|------------------------------|--|--------|----------------|-------|--------|---------------|
| MANUFACTURER | NUMBER TESTED | NUMBER REJECTED (% REJECTED) | PENETRATION | MEDIUM | CARRIER DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 133 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| B | 21 | 1
(4.8%) | 0 | 0 | 0 | 0 | 0 | 1 |
| C | 1053 | 22
(2.1%) | 12 | 10 | 0 | 0 | 0 | 0 |
| D | 383 | 1
(0.3%) | 1 | 0 | 0 | 0 | 0 | 0 |
| TOTAL | 1590 | 24
(1.5%) | 13 | 10 | 0 | 0 | 0 | 1 |

**TABLE 4 - All Filters Tested At
Either DOE Filter Test Facility
Differentiated by Filter Category**

| FILTER
CATEGORY | NUMBER
TESTED | NUMBER
REJECTED
(% REJECTED) | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|-----------------------|------------------|------------------------------------|--|--------|-------------------|-------|--------|---------------|
| | | | PENETRATION | MEDIUM | CARRIER
DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| ALL | 85050 | 5472
(6.4%) | 1715 | 1209 | 246 | 974 | 685 | 643 |
| 1000 CFM | 30966 | 1167
(3.8%) | 371 | 276 | 90 | 142 | 57 | 231 |
| ≠ 1000 CFM | 23375 | 1183
(5.1%) | 628 | 219 | 47 | 25 | 34 | 230 |
| ≥ 1000 CFM | 36620 | 1506
(4.1%) | 524 | 316 | 103 | 141 | 125 | 297 |
| ≥ 1000 CFM
NUCLEAR | 1590 | 24
(1.5%) | 13 | 10 | | | | 1 |

TABLE 5 - ALL FILTERS TESTED AT
HANFORD

| MANUFACTURER | NUMBER TESTED | NUMBER REJECTED
(% REJECTED) | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|---------------|---------------------------------|--|--------|----------------|-------|--------|---------------|
| | | | PENETRATION | MEDIUM | CARRIER DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 10632 | 1464
(13.8%) | 192 | 237 | 74 | 617 | 252 | 92 |
| B | 2417 | 270
(11.2%) | 101 | 60 | 13 | 37 | 54 | 5 |
| C | 22841 | 1659
(7.3%) | 552 | 385 | 54 | 190 | 400 | 78 |
| D | 10442 | 950
(9.1%) | 127 | 378 | 65 | 164 | 21 | 195 |
| TOTAL | 46332 | 4343
(9.4%) | 972 | 1060 | 206 | 1008 | 727 | 370 |

TABLE 6 - Filters of 1000 CFM
Capacity Tested At Hanford

| | | | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|---------------|------------------------------|--|--------|----------------|-------|--------|---------------|
| MANUFACTURER | NUMBER TESTED | NUMBER REJECTED (% REJECTED) | PENETRATION | MEDIUM | CARRIER DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 1983 | 292 (14.7%) | 64 | 63 | 37 | 45 | 44 | 39 |
| B | 273 | 36 (13.2%) | 16 | 5 | 12 | 2 | 1 | 0 |
| C | 3492 | 225 (6.4%) | 50 | 61 | 18 | 63 | 6 | 27 |
| D | 2973 | 253 (8.5%) | 4 | 103 | 12 | 28 | 3 | 103 |
| TOTAL | 8720 | 806 (9.2%) | 134 | 232 | 79 | 138 | 54 | 169 |

TABLE 7 - All Filters Tested At
Oak Ridge

| MANUFACTURER | NUMBER
TESTED | NUMBER
REJECTED
(% REJECTED) | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|------------------|------------------------------------|--|--------|-------------------|-------|--------|---------------|
| | | | PENETRATION | MEDIUM | CARRIER
DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 2436 | 130
(5.3%) | 72 | 12 | 2 | 0 | 2 | 42 |
| B | 8556 | 366
(4.3%) | 157 | 47 | 28 | 10 | 52 | 72 |
| C | 23860 | 803
(3.4%) | 569 | 47 | 5 | 3 | 18 | 161 |
| D | 7094 | 139
(2.0%) | 5 | 121 | 7 | 1 | 0 | 5 |
| TOTAL | 41946 | 1438
(3.4%) | 803 | 227 | 42 | 14 | 72 | 280 |

**TABLE 8 - Filters of 1000 CFM
Capacity Tested at Oak Ridge**

| MANUFACTURER | NUMBER
TESTED | NUMBER
REJECTED
(% REJECTED) | NUMBER REJECTED FOR EACH REJECTION CAUSE | | | | | |
|--------------|------------------|------------------------------------|--|--------|-------------------|-------|--------|---------------|
| | | | PENETRATION | MEDIUM | CARRIER
DAMAGE | FRAME | GASKET | MISCELLANEOUS |
| A | 1590 | 79
(5.0%) | 42 | 9 | 1 | 0 | 0 | 27 |
| B | 1648 | 69
(4.2%) | 53 | 7 | 5 | 0 | 0 | 4 |
| C | 14569 | 199
(1.4%) | 141 | 19 | 3 | 3 | 3 | 30 |
| D | 4439 | 14
(0.3%) | 1 | 9 | 2 | 1 | 0 | 1 |
| TOTAL | 22246 | 361
(1.6%) | 237 | 44 | 11 | 4 | 3 | 62 |

TABLE 9 - Filters Manufactured By Vendor A. (Semi-Annual Totals)

| DATE | OAK RIDGE | | | HANFORD | | | COMBINED | | |
|-----------------------|-----------|----------|--------|---------|----------|--------|----------|----------|--------|
| | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. |
| Jan 1 to Jun 30, 1971 | 441 | 24 | 5.4% | 287 | 45 | 15.7% | 728 | 69 | 9.5% |
| Jul 1 to Dec 31, 1971 | 273 | 12 | 4.4% | 262 | 23 | 8.8% | 535 | 35 | 6.5% |
| Jan 1 to Jun 30, 1972 | 805 | 62 | 7.7% | 861 | 115 | 13.4% | 1666 | 177 | 10.6% |
| Jul 1 to Dec 31, 1972 | 659 | 19 | 2.9% | 1210 | 137 | 11.3% | 1869 | 156 | 8.3% |
| Jan 1 to Jun 30, 1973 | 69 | 8 | 11.6% | 67 | 6 | 9.0% | 136 | 14 | 10.3% |
| Jul 1 to Dec 31, 1973 | 1 | 0 | | 327 | 69 | 21.1% | 328 | 69 | 21.0% |
| Jan 1 to Jun 30, 1974 | 2 | 0 | | 263 | 23 | 8.7% | 265 | 23 | 8.7% |
| Jul 1 to Dec 31, 1974 | 0 | 0 | | 148 | 33 | 22.3% | 148 | 33 | 22.3% |
| Jan 1 to Jun 30, 1975 | 15 | 2 | 13.3% | 72 | 3 | 4.2% | 87 | 5 | 5.7% |
| Jul 1 to Dec 31, 1975 | 4 | 0 | | 2110 | 281 | 13.3% | 2114 | 281 | 13.3% |
| Jan 1 to Jun 30, 1976 | 84 | 0 | | 2776 | 499 | 18.0% | 2860 | 499 | 17.4% |
| Jul 1 to Dec 31, 1976 | 83 | 3 | 3.6% | 1349 | 181 | 13.4% | 1432 | 184 | 12.8% |
| Jan 1 to Sep 30, 1977 | - | - | - | 899 | 49 | 5.4% | 900 | 49 | 5.4% |
| Total | 2436 | 130 | 5.3% | 10632 | 1464 | 13.8% | 13068 | 1594 | 12.2% |

TABLE 10 - Filters Manufactured by Vendor B. (Semi-Annual Totals)

| DATE | OAK RIDGE | | | HANFORD | | | COMBINED | | |
|-----------------------|-----------|----------|--------|---------|----------|--------|----------|----------|--------|
| | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. |
| Jan 1 to Jun 30, 1971 | 229 | 21 | 9.2% | 133 | 17 | 12.8% | 362 | 38 | 10.5% |
| Jul 1 to Dec 31, 1971 | 550 | 60 | 10.9% | 191 | 28 | 14.7% | 741 | 88 | 11.9% |
| Jan 1 to Jun 30, 1972 | 245 | 35 | 14.3% | 204 | 7 | 3.4% | 449 | 42 | 9.4% |
| Jul 1 to Dec 31, 1972 | 514 | 21 | 4.1% | 40 | 5 | 12.5% | 554 | 26 | 4.7% |
| Jan 1 to Jun 30, 1973 | 439 | 30 | 6.8% | 104 | 12 | 11.5% | 543 | 42 | 7.7% |
| Jul 1 to Dec 31, 1973 | 803 | 64 | 8.0% | 154 | 27 | 17.5% | 957 | 91 | 9.5% |
| Jan 1 to Jun 30, 1974 | 370 | 8 | 2.2% | 76 | 8 | 10.5% | 446 | 16 | 3.6% |
| Jul 1 to Dec 31, 1974 | 1078 | 30 | 2.8% | 8 | 0 | | 1086 | 30 | 2.8% |
| Jan 1 to Jun 30, 1975 | 717 | 6 | 0.8% | 31 | 4 | 12.9% | 748 | 10 | 1.3% |
| Jul 1 to Dec 31, 1975 | 1190 | 6 | 0.5% | 417 | 76 | 18.2% | 1607 | 82 | 5.1% |
| Jan 1 to Jun 30, 1976 | 1174 | 11 | 0.9% | 324 | 11 | 3.4% | 1498 | 22 | 1.5% |
| Jul 1 to Dec 31, 1976 | 1247 | 74 | 5.9% | 566 | 39 | 6.9% | 1813 | 113 | 6.2% |
| Jan 1 to Sep 30, 1977 | - | - | - | 169 | 36 | 21.3% | 169 | 36 | 21.3% |
| Total | 8556 | 366 | 4.3% | 2417 | 270 | 11.2% | 10973 | 636 | 5.8% |

TABLE 11 - Filters Manufactured By Vendor C (Semi-Annual Totals)

| DATE | OAK RIDGE | | | HANFORD | | | COMBINED | | |
|-----------------------|-----------|----------|--------|---------|----------|--------|----------|----------|--------|
| | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. |
| Jan 1 to Jun 30, 1971 | 1765 | 10 | 0.6% | 1032 | 44 | 4.3% | 2797 | 54 | 1.9% |
| Jul 1 to Dec 31, 1971 | 3800 | 78 | 2.1% | 1042 | 31 | 3.0% | 4842 | 109 | 2.3% |
| Jan 1 to Jun 30, 1972 | 2654 | 78 | 2.9% | 1547 | 119 | 7.7% | 4201 | 197 | 4.7% |
| Jul 1 to Dec 31, 1972 | 1998 | 159 | 8.0% | 1680 | 92 | 5.5% | 3678 | 251 | 6.8% |
| Jan 1 to Jun 30, 1972 | 1651 | 119 | 7.2% | 1773 | 366 | 20.6% | 3424 | 485 | 14.2% |
| Jul 1 to Dec 31, 1973 | 3022 | 110 | 3.6% | 1055 | 129 | 12.2% | 4077 | 239 | 5.9% |
| Jan 1 to Jun 30, 1974 | 1733 | 44 | 2.5% | 2621 | 51 | 1.9% | 4354 | 95 | 2.2% |
| Jul 1 to Dec 31, 1974 | 603 | 11 | 1.8% | 3157 | 145 | 4.6% | 3760 | 156 | 4.1% |
| Jan 1 to Jun 30, 1975 | 309 | 3 | 1.0% | 1608 | 89 | 5.5% | 1917 | 92 | 4.8% |
| Jul 1 to Dec 31, 1975 | 1672 | 46 | 2.8% | 1327 | 91 | 6.9% | 2999 | 137 | 4.6% |
| Jan 1 to Jun 30, 1976 | 1573 | 32 | 2.0% | 2735 | 110 | 4.0% | 4308 | 142 | 3.3% |
| Jul 1 to Dec 31, 1976 | 3080 | 113 | 3.7% | 1357 | 190 | 14.0% | 4437 | 303 | 6.8% |
| Jan 1 to Sep 30, 1977 | - | - | - | 1907 | 202 | 10.6% | 1907 | 202 | 10.6% |
| Total | 23860 | 803 | 3.4% | 22841 | 1659 | 7.3% | 46701 | 2462 | 5.3% |

TABLE 12 - Filters Manufactured by Vendor D (Semi-Annual Totals)

| DATE | OAK RIDGE | | | HANFORD | | | COMBINED | | |
|-----------------------|-----------|----------|--------|---------|----------|--------|----------|----------|--------|
| | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. | TESTED | REJECTED | % REJ. |
| Jan 1 to Jun 30, 1971 | 1268 | 101 | 8.0% | 38 | 0 | | 1306 | 101 | 7.7% |
| Jul 1 to Dec 31, 1971 | 187 | 0 | | 58 | 0 | | 245 | 0 | |
| Jan 1 to Jun 30, 1972 | 483 | 1 | 0.2% | 14 | 0 | | 497 | 1 | 0.2% |
| Jul 1 to Dec 31, 1972 | 144 | 5 | 3.5% | 3796 | 292 | 7.7% | 3940 | 297 | 7.5% |
| Jan 1 to Jun 30, 1973 | 462 | 12 | 2.6% | 2226 | 222 | 10.0% | 2688 | 234 | 8.7% |
| Jul 1 to Dec 31, 1973 | 498 | 3 | 0.6% | 1565 | 170 | 10.9% | 2063 | 173 | 8.4% |
| Jan 1 to Jun 30, 1974 | 625 | 3 | 0.5% | 1156 | 37 | 3.2% | 1781 | 40 | 2.2% |
| Jul 1 to Dec 31, 1974 | 799 | 1 | 0.1% | 135 | 17 | 12.6% | 934 | 18 | 1.9% |
| Jan 1 to Jun 30, 1975 | 1283 | 1 | 0.1% | 304 | 21 | 6.9% | 1587 | 22 | 1.4% |
| Jul 1 to Dec 31, 1975 | 393 | 2 | 0.5% | 211 | 18 | 8.5% | 604 | 20 | 3.3% |
| Jan 1 to Jun 30, 1976 | 454 | 0 | | 478 | 65 | 13.6% | 932 | 65 | 7.0% |
| Jul 1 to Dec 31, 1976 | 498 | 10 | 2.0% | 209 | 86 | 41.1% | 707 | 96 | 13.6% |
| Jan 1 to Sep 30, 1977 | - | - | - | 252 | 22 | 8.7% | 252 | 22 | 8.7% |
| Total | 7094 | 139 | 2.0% | 10442 | 950 | 9.1% | 17536 | 1089 | 6.2% |

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DISCUSSION

CADWELL: Any attempt to save money and streamline operations is appreciated. My interpretation of your data is that the Quality Assurance Stations have done an effective job in keeping the quality of the filters high. Our fear is that, if you take away the threat of rejection at the retest stations, there will be a gradual deterioration of the quality of HEPA filters with a resultant deterioration in the reactor safety program.

COLLINS: I have to take issue with you. I don't believe that the Quality Assurance Station by itself has been the primary reason for continuing good performance of the filters. I think that the nuclear industry has grown to a point where it realizes the need to produce a product comparable to other components of the nuclear power plant. I think the initiative and the efforts of the ANSI standards people and the industry itself to regulate the quality of filters have been the principal reasons, rather than the Quality Assurance Stations.

CADWELL: I agree with that but I am afraid that if you take away the threat of financial loss due to rejection at the Stations you open the door of compromise between the manufacturer and the buyer. That could become a potentially dangerous situation.

COLLINS: That could be a result but, as I indicated, we certainly are going to continue to monitor the program. Nevertheless, I think that the HEPA filter is only one component in the total nuclear power plant and if you take the kind of approach you are advocating, it could be extended to all other components, particularly to those in the pressure boundary systems. If we started to impose the practice of Quality Assurance Stations for all other components, I believe the nuclear industry would be on our back. I believe that we, as regulators, have a responsibility, when we see data that show trends, to indicate that testing is no longer required for government installations, i.e., that the industry can regulate itself, to "de-ratchet" not to "ratchet" for higher controls.

MURROW: Information on manufacturers A through D is impressively good but manufacturers "E" through "X" have no existence and no past history. How can we be assured they will produce as good filters when they enter the market?

COLLINS: I can appreciate that; the data that were available to us showed only the four principal vendors. The only answer I can give is that, if the rest of the alphabet comes along and if they can meet the requirements of Appendix B and can test filters in accordance with the ANSI standards and the Mil-Specs, more power to them. They certainly would be acceptable.

BURCHSTED: I would like to make five statements. (1) Past is not prelude; quality generated by the industry in the past has no bearing on quality of non-monitored filters in the future. (2) There is a fundamental quality assurance principle that the workman does not inspect his own work. (3) The proposed monitoring of the filter test stations has little bearing on filters supplied to the commercial power industry which will no longer require confirmatory quality assurance testing. (4) It is possible that abandonment of confirmatory test-station testing may lead to the development of two grades of filters, those subject to confirmatory testing, and those not subject to such testing. It is significant that Mr. Cadwell, a filter manufacturer, apparently believes the same. (5) As a very minimum, I would suggest that the Nuclear Regulatory Commission should obtain occasional samples of the filters furnished to power stations and have them tested by the filter test stations.

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COLLINS: I cannot argue with some of your statements but if you would realize the number of filters intended for use in commercial nuclear power plants was 1600 and that of that number only 1.5% were rejected, that is pretty good quality.

BURCHSTED: I accept that, but just because this happened in the past, it doesn't mean it will happen in the future in the absence of tests by Quality Assurance Stations.

COLLINS: That is very true. I think the only thing that we can say is that, in my opinion and in the opinion of the staff, the past is something that we should pay attention to. The experience we have gained is something that we must use. If it turns out that we were wrong, we were wrong, and it won't be the first time, but we try not to let it happen often.

KOVACH: I have two comments. One is that we at NUCON have not seen a difference between failure of installed components which were or were not tested at a QA station. Our observation is that damage to, or the failure of, HEPA filter systems is much more often caused by sloppy installation than a failure caused by poor filter manufacture or by excess shipping or by QA testing of the filters. This confirms your conclusions.

At the same time, some periodical and random checks on the manufacturers would be very helpful to assure maintenance of the quality of a fragile component of ESF and other systems. Maybe this is aiming at a compromise, but I believe it may be advantageous for the users to keep track of the manufacturers.

COLLINS: I appreciate the comment. What you have recommended has been proposed to us and I am giving it very serious consideration.

DAVIS: Will there be a continued requirement for in-place testing of HEPA filter systems at power plants, and is this a factor in the decision to discontinue the requirement for QA testing at the DOE test station?

COLLINS: Oh, yes! We would never eliminate the requirement for in-place testing. In-place test requirements are imposed on a licensed facility by its operating license as part of the technical specifications.

DAVIS: Is that a part of your decision then?

COLLINS: No, that has always been in the technical specifications and the tests will continue.

DAVIS: I mean, is that a part of the decision to dispense with the test station requirement.

COLLINS: It certainly had an impact on our decision.

FRY: Can you tell me what the frequency of filter sampling was? The reason I ask is that the situation you describe is a classic QC (quality control) problem. The quality of the product is good at the current frequency of sampling. In such a case, one simply reduces the frequency of sampling until a problem is identified and then the frequency of sampling is increased again. This is, in fact, exactly what you are doing by continuing to "monitor" product quality.

COLLINS: That is correct.

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STEINBERG: The arbitrary decision to cease QA testing of filters and subsequent closing of the QA Stations is a giant leap backward. Now, the filter suppliers will take advantage of the relaxed requirements and the possibility of installation of filters with defects will inevitably increase.

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PERFORMANCE AND ENVIRONMENTAL CHARACTERISTICS OF A COMPACT, HIGH-CAPACITY HEPA FILTER DESIGN

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Abstract

An improved version of the HEPA filter has been developed and evaluated for conformity with MIL-F-51068. In this design, the filter paper (conforming to MIL-F-51079) is pleated to a depth of only 18 mm. Ribbons essentially the same material as the filter paper form inter-pleat spacers, providing a crest-to-crest spacing of 3 mm. The resultant filter has the same DOP penetration level as the standard MIL-F-51068 HEPA filter, but about half its pressure drop at the same airflow. Materials of construction, adhesives, gaskets and faceguards match MIL-F-51068. Both flat-panel and zig-zag configurations of the filter meet the rough handling test of MIL-F-51068. Preliminary indications are that both filters will meet the overpressure tests of MIL-F-51068D and the flammability requirements of UL 586; these tests are in progress. Filters can be supplied to interchange with existing HEPA filter sizes. The improved pressure drop characteristic of these filters can be used to provide longer intervals between filter changes, or equal life at greater capacity.

I. Introduction

The basic design of HEPA filters used in American nuclear facilities has changed little since the original development of this filter during the 1940's. In this design, a web of paper is formed into pleats of depth from about 70 mm to about 300 mm, depending on the desired gas flow velocity through the completed filter. These pleats are held apart both upstream and downstream by corrugated spacers which provide a pleat pitch of about 9 mm. This media-separator pack is sealed into wood or metal cell sides to form a filter cartridge. The design has proved to be effective and reliable in a wide range of environmental conditions, when the proper component materials have been used. Component materials for both military and civilian nuclear applications are defined in MIL-F-0051079 (for the filter media) and MIL-F-0051068 (for the completed filter cartridge).

These specifications are definitive enough to give the impression that only minor deviations from the standard design are possible. The requirements imposed by MIL-F-0051079 on the filter media for example, essentially dictate a wet-laid glassfiber paper with a furnish of fibers predominantly less than 1 μ m in diameter. MIL-F-0051068 is even more restrictive in defining the design of the completed filter, since it specifies many construction details, as well as performance requirements. In both the "D" and "E" versions of this specification, however, wording exists allowing some material and construction deviations from the specified norms. Section 3.2.6, for example, permits the procuring agency to specify spacing means

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different from those stated in MIL-F-0051068. Filters made with such deviations must, of course, meet the environmental and performance requirements of the specification to be acceptable for nuclear service. One U.S. manufacturer of HEPA filters does produce a filter which has no separators at all, relying on a special double-pleating technique to provide the necessary air passages; this filter is accepted for nuclear applications. Thus it appears that deviations from MIL-F-0051068 can be permitted in U.S. nuclear applications, if reliability can be maintained, and if some advantage is obtained by the change.

A form of high efficiency particulate filter quite different from the U.S. design has been in use in Europe for several years. Figure 1 shows its design. Here the filter media is not pleated to the full depth of the filter cartridge in the direction of air flow. Instead, pleats are no more than 20 mm deep, and the crest-to-crest pitch is about 3 mm. Adjacent pleats are separated by ribbons of foam plastic, asbestos, or glued-on threads. A full size filter cartridge is fabricated from several panels of this construction arranged in a zig-zag fashion. This design allows considerably more area of filter media to be exposed to air flow in a given filter cartridge than is possible with the U.S. design. Such increased area provides a lower pressure drop at increased flow, or extended service life at normal flow.

Earlier European designs, like their American counterparts, did not pay great attention to flammability, radiation and humidity resistance. However, more recent versions have incorporated glassfiber filter media meeting MIL-F-0051079, nonflammable spacer strips, and adhesives of greater radiation resistance.

It appears entirely possible to qualify a filter of this European design under MIL-F-0051068. The discussion which follows describes the results of MIL-F-0051068-type tests on a European-style HEPA filter of U.S. fabrication. In addition, data on resistance versus air flow at various dust loadings are given. These tests indicate that the European filter design can equal the American design in environmental resistance, and offers substantially longer life for the same cartridge dimensions and pressure drop.

II. Filter Description

The filters tested have been of two forms: flat panels of pleated media (Fig. 2) and assemblies of such panels (Fig. 1). In both cases, the filter media was the same, with the following characteristics:

Media furnish: glassfiber, average fiber diameter $< 1 \mu\text{m}$
Caliper: 0.015 in (0.38 mm) minimum
Basis Weight: 55 lb/3000 ft² (89.5 g/m²)
Flat Sheet air flow resistance: 40 mm WG (392 Pa) at 320 cm/min
Dry Tensile Strengths (Typical)

| | |
|-------------------------------|-----------------------|
| Machine direction, uncreased: | 5.0 lb/in (8.76 N/cm) |
| Machine direction, creased : | 2.0 lb/in (3.50 N/cm) |
| Cross direction, uncreased : | 3.8 lb/in (6.65 N/cm) |

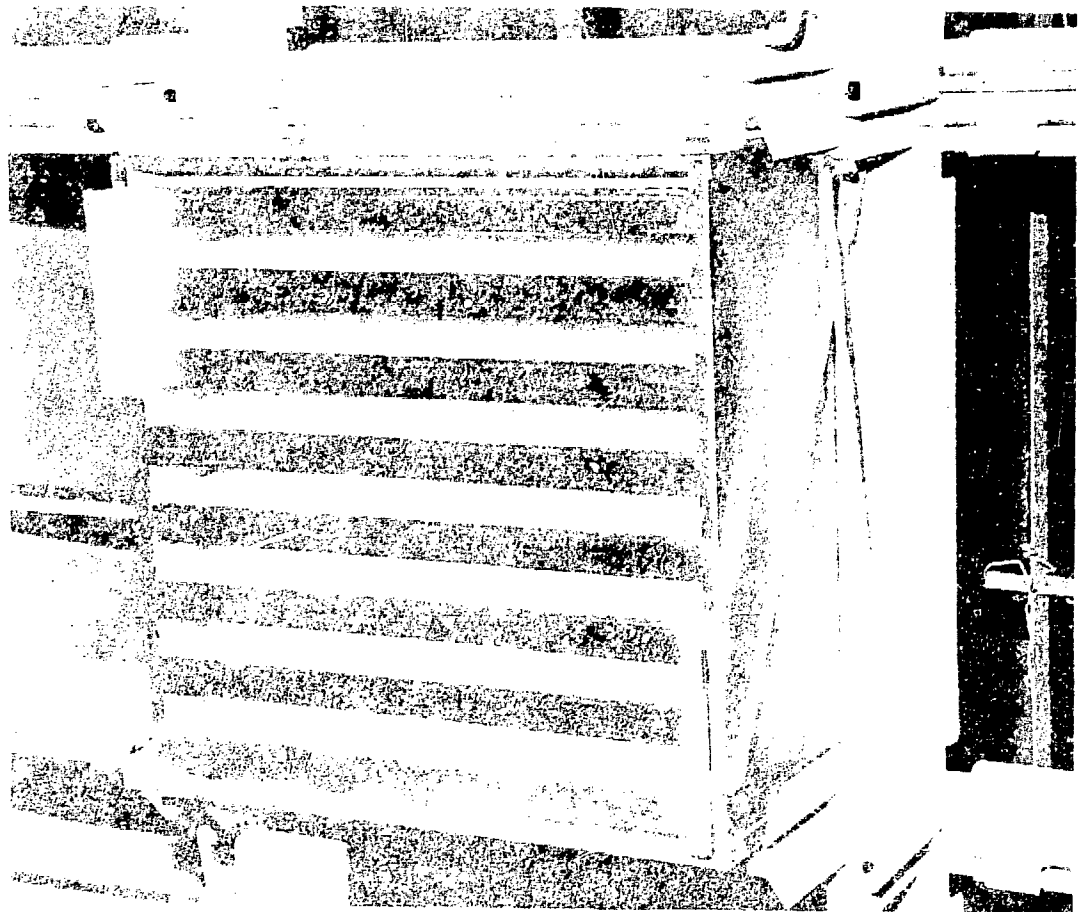


Figure 2. Close-up of Flat 1 and Flat 2 with cardboard and Styrofoam

The filter media is held in place by friction, rather than by a sealant. The flow is not entirely cut off from the media by the spacer material. In addition, the flammability and other characteristics of the spacer material is essentially irrelevant to the filter performance. (See Figs. 3 and 4 for construction details.)

In the case of conventional HEPA filter design, cell sides are made of various materials. For the flat panel filters, particle board, plywood, galvanized sheet steel and aluminum extrusions have all been used. The design described here is currently available only in plywood. For the zig-zag multipanel filter, European practice is to form cell sides from stainless, galvanized or painted sheet steel. The design described here is (at this writing) available in 20 ga. galvanized sheet steel only.

Several types of sealant have been used by European manufacturers to seal the edges of the packs to the cell sides: bitumen, silicones, polyvinyl chloride, and other synthetic resins and plastisols. The design described here is sealed with a two-part epoxy containing a fire retardant.

Faceguards, as shown in Fig. 2, are essential to the flat panel filter design for this service. The zig-zag design, however, is inherently protected by the channels which span the face of the filter and seal the upstream and downstream edges of the media panels. Expanded metal faceguards can be added to this filter also, of course.

Figure 3 shows a cross-section of the filter. The filter media is held in place by friction, rather than by a sealant. The flow is not entirely cut off from the media by the spacer material. In addition, the flammability and other characteristics of the spacer material is essentially irrelevant to the filter performance. (See Figs. 3 and 4 for construction details.)

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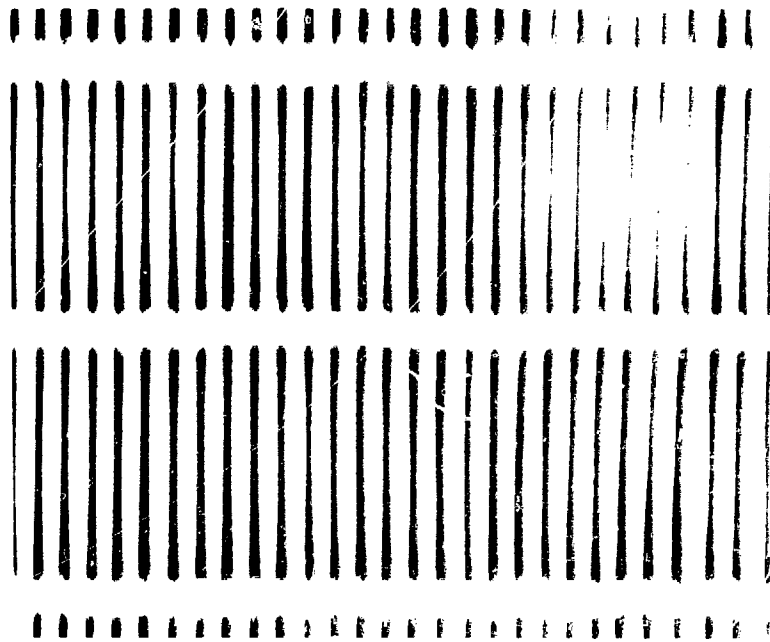


Figure 3. Close-Pleat Filter Media with Spacer Ribbons

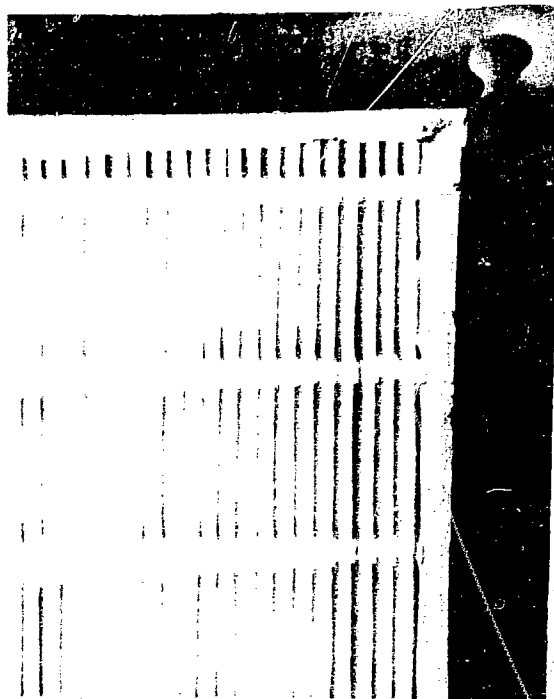


Figure 4. Sealant and Dovetail Gasket Joint, Flat Panel Filter

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[illegible]

1. *Journal of the American Medical Association*, 1990; 263: 1025-1026.

These findings indicate that the model under development allows for the possibility that the same individual may be involved in more than one type of generative experience. For example, an individual may be involved in a generative experience in which he or she is the subject of the experience, while at the same time being involved in another generative experience in which he or she is the object of the experience.

Fig. 12 is a plot of the zig-zag multilayer design at various air loadings and air flow. Comparison data for the standard HMA filter design are also shown. The lint fed was ASHRAE Standard Air Cleaner Test Dust minus its lint component. The standard design appears to build up resistance about three times as quickly as the zig-zag multilayer design.

Fig. 6 compares the resistance build-up for 500 ft³/min (850 m³/h) filters, the standard unit being 610x610x149 mm, and the close pleat design being 610x610x78 mm. The two filters track fairly closely up to 400 g dust load, after which the standard filter rises somewhat more slowly. However, the resistance at 400 g load was 3 in WG (747 Pa) which is quite high for normal service for this type of filter.

The smaller volume of the close-pleat panel filter, and its lesser weight (3.6 kg vs 9 kg for the standard design) undoubtedly reduce the waste disposal problem for this size unit, even if the two have equal lives.

IV. Environmental Tests

To date, we have been able to run only two of the environmental exposure tests specified in MIL-F-0051068D. The large capacity of the zig-zag multipanel unit means that a very large fan is needed to raise its resistance to 10 in WG (2490 Pa) as specified in MIL-F-0051068D. This test was simulated for a single slot of the

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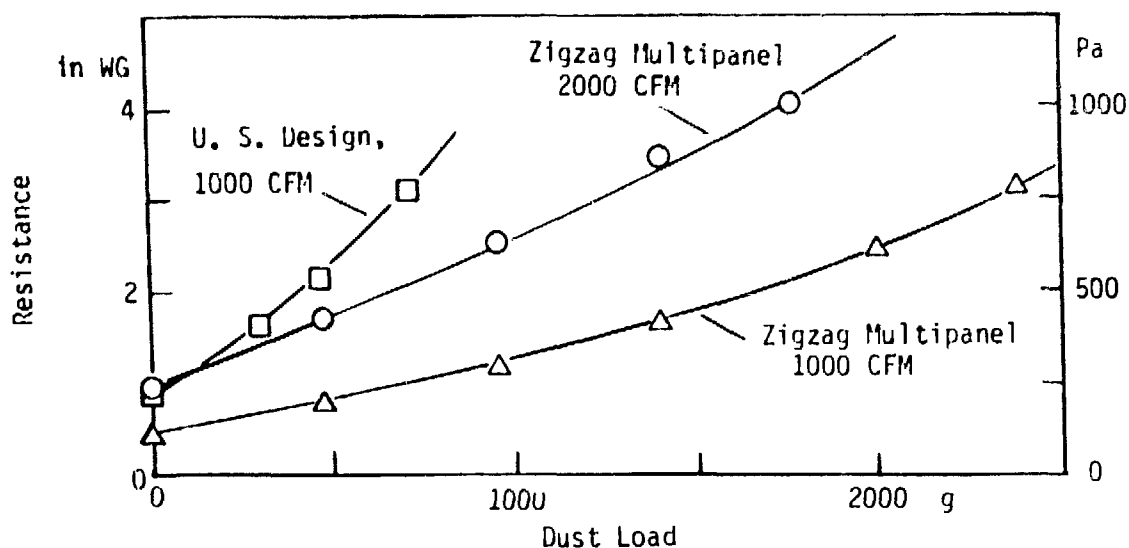


Figure 5. Resistance vs Dust Load for 610x610x292 mm Filters

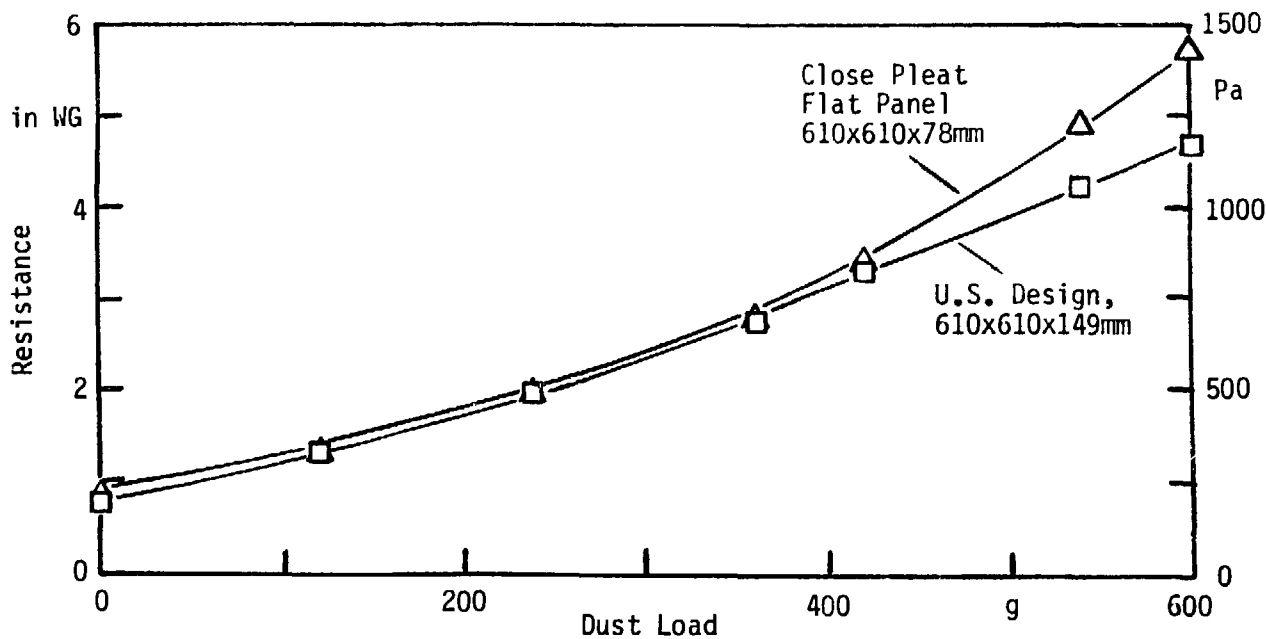


Figure 6. Resistance vs Dust Load for 500 ft³/min Filters

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zig-zag structure, and it was found that some downstream support between panels is needed to withstand the full 2490 Pa pressure. The full exposure, with DOP and resistance tests run before and after the overpressure exposure, was run on a close pleat flat panel unit, with the results listed in Table 1. The inherent structure of the media panel, plus the expanded metal faceguard and added crossbars (See Fig. 2) is apparently sufficient to qualify this filter.

Table I. Overpressure Tests.

Astrocel II Flat Panel Close-Pleat Filter

| Filter No. | DOP Penetration, % | | Pressure Drop At Rated Flow, mm WG | |
|------------|--------------------|----------------|------------------------------------|----------------|
| | Before Exposure | After Exposure | Before Exposure | After Exposure |
| 11 | 0.004 | 0.004 | 22.6 | 23.6 |
| 12 | 0.006 | 0.008 | 23.4 | 24.4 |
| 13 | 0.004 | 0.002 | 22.9 | 24.4 |

Filter dimensions 610x610x70 mm

Rated Flow: 500 cfm (850 m³/hr)

Exposure: Pressure Drop increased to 2490 Pa by increasing air flow. This condition was held for 15 minutes before flow was returned to rated level, and DOP Penetration and rated flow pressure drop "After" determined.

The rough handling test of MIL-F-0051068D, using the Q110 Vibrating Machine, was run on a zig-zag multipanel filter, with results listed in Table 2. This unit appears to survive this exposure without loss of performance.

A program exposing both filters to the remaining conditions of MIL-F-0051068D is now under way, as is the U.L. 586 series of flammability tests. The results of these tests will be reported when they are completed.

Table II. Rough Handling Tests
on 610x610x298 mm
Close Pleat Zig-Zag Multipanel Filter.

| | 1000 ft ³ /min | 200 ft ³ /min |
|---|---------------------------|--------------------------|
| DOP Penetration before rough handling | 0.018% | 0.017% |
| DOP Penetration after vertical rough handling | 0.017 | 0.020 |
| DOP Penetration after horizontal rough handling | 0.013 | 0.015 |

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Filter tested for DOP penetration "encapsulated" at 1000 cfm and 200 cfm.

Test sequence:

1. Rough handled with metal channels vertical on Q-110 rough handler - 200 cycles per min, 3/4 in. amplitude. 15 min. test
2. DOP test
3. Rough handle same filter with metal channels horizontal.
4. DOP test

References:

1. Rivers, R.D. (ed.) Design and Testing of Fan-Cooler-Filter Systems for Nuclear Applications. (USAEC Topical Report AAF-TR-7101) 1972

DISCUSSION

CADWELL: What will be the availability and relative price of the multi-panel, zig-zag filter?

RIVERS: The flat ones are available in any quantity, the multi-panel and zig-zag ones in small quantities. Our line isn't up to full speed yet.

CADWELL: Do you have any idea of the price yet?

RIVERS: No, I do not.

BURCHSTED: At the time we published the Nuclear Air Cleaning Handbook (ERDA 76-21), none of the European-design filters had met the U.S. requirements for nuclear-grade filters. John Dymont also commented that we had not discussed European practice in his review of the Handbook in "Filtration and Separation." When we re-issue the Handbook we would like to remedy this shortcoming, and I invite the overseas participants at this Conference to take steps to inform me accordingly,

RIVERS: I hope by the time of the 3rd Edition that we will have American experience, as well.

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PERFORMANCE OF 1,000 AND 1800 CFM HEPA FILTERS IN LONG EXPOSURE TO LOW ATMOSPHERIC DUST LOADINGS*

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Abstract

Comparative tests are in progress to determine the performance characteristics of European-design HEPA filters compared to U.S.-design units made with the same filter paper. European filters are being operated at their rated capacity of 1,800 cfm and at 1,000 cfm. The U.S.-design unit is operated at 1,000 cfm. Filters, installed on the roof of a 15-story building, are continuously exposed to an atmospheric dust aerosol of low concentration and small particle size. Plans are to continue these tests over a period of two or more years. Early results confirm that 1,800 cfm rated filters will have a very slow rate of pressure increase when operated at 1,000 cfm and an extended service life.

Introduction

Recent rapid increases in the price of nuclear solid waste disposal services have made it considerably more costly to dispose of a used HEPA filter than to acquire and install it. As there appears to be little immediate hope that important increases will take place in disposal productivity, total filter disposal costs can be lowered only by reducing the number of units discarded. A way that this desirable objective can be achieved without reducing the number of filters in essential service is to make them last longer. Obvious ways to accomplish this are to continue to operate HEPA filters until they reach a higher airflow resistance before replacement or to install more efficient (and higher resistance) prefilters. However, for existing systems, installed blowers and blower motors will be unable to deliver the required airflow at the higher filter resistance. Although the higher resistance option is open to designers of new filter systems, governmental requirements for energy conservation in new construction make it less attractive than it might have been at an earlier time.

Continental European filters have differed substantially in construction details from American and British HEPA filters. For many years, these European filters failed to find favor in the U.S. because the paper that was used in them was considered to be inferior. This no longer seems to be correct and interest in European filters has been rekindled because their design makes it possible to put sufficient effective filter paper into the standard 24-in. x 24-in. x 11 1/2-in. filter frame to handle 1,800 - 2000 cfm (instead of 1,000 cfm) at a clean filter resistance of 1-in. w.g. or less and to meet the maximum DOP penetration standard of 0.03% at these higher volumetric flowrates. The increase in air handling capacity is made possible by folding the filter paper into small pleats with very narrow air spaces between adjacent folds, thereby packing much more filter paper into the assigned space than is possible with the Amer-

*Work performed under U.S. Department of Energy Contract No. EY-S-02-3049 by the Harvard Air Cleaning Laboratory; Mr. J. Dempsey, Division of Waste Management, Contract Administrator.

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Irish and British filter design that uses wavy corrugated separators between large paper pleats. Flinders Filters, Inc. increased air-flow capacity of one of their HEPA filter products by going to a separatorless design (Super-Flow) and they still use relatively large pleats and have not been able to obtain as great an increase in air-flow capacity as have the European manufacturers who use smaller and shallower pleats that are separated from adjacent folds by glued-on thin strips of paper or braided string at approximately 1-inch intervals. Stacks of the shallow pleats are placed in the filter frame with the open ends faced perpendicular to the direction of upstream airflow, and with alternating inlet and outlet air channels extending into the filter frame between the stacks of pleated filter as shown in Figure 1. Alternate air channels are blocked on the upstream side and on the downstream side so that all air that enters must pass through the pleated paper to reach an exit air channel.

Because the resistance of HEPA filters is directly proportional to air flow rate, when an 1800 cfm rated filter is operated at 1000 cfm, its clean resistance will be only 55% of what it is at the higher flow rate (i.e., from 1.8 to 1.0 in. w.g.). On the assumption that an 1800 cfm rated filter has 1.8 times more effective filter paper surface than a 1,000 cfm rated filter, the dust accumulation on a unit area of paper of an 1800 cfm rated filter, when operated at 1,000 cfm, will be only 55% of the amount deposited when the filter is operated at its rated flow. This means that the airflow resistance buildup of a 24 x 24 x 11 1/2-in. 1800 cfm-rated filter, overated at 1,000 cfm, will be decreased proportionately by the lower unit dust deposit rate as well as by the lower airflow rate through both dust deposit and paper. Theoretically, the rate of resistance buildup will reflect the product of the two effects and in the case cited above, the rate of resistance increase should be only 0.3 as fast, i.e., $0.55 \times .55 = 0.30$. Put another way, when operated at 1,000 cfm, the period required for an 1800 cfm rated filter to reach its maximum rated resistance (usually 3-4 in. w.g.) will be three times as long and, as a result, the number of filters discarded to waste will be reduced by two thirds. This represents a very impressive potential saving in waste disposal costs should practise be shown to follow theory.

Accelerated life tests of HEPA filters have been conducted with simulated aerosols, such as NBS filter test dust, at particle concentrations several orders of magnitude greater than HEPA filters are ordinarily subjected to in nuclear service. These results have notoriously poor predictive value for filters exposed to low dust loadings containing particles of much smaller size. The disadvantage of performing filter life tests with realistic aerosol concentrations and particle sizes is the very long times that are needed to arrive at reliable conclusions. This is unfortunate, but unavoidable.

Test Arrangements

Atmospheric dust has been widely used for high efficiency filter testing but near ground level, particle size and concentration are still greater than HEPA filters used in nuclear service are likely to be exposed to. If, however, ambient air substantially above ground level is selected as the test aerosol, dust concentrations and dust

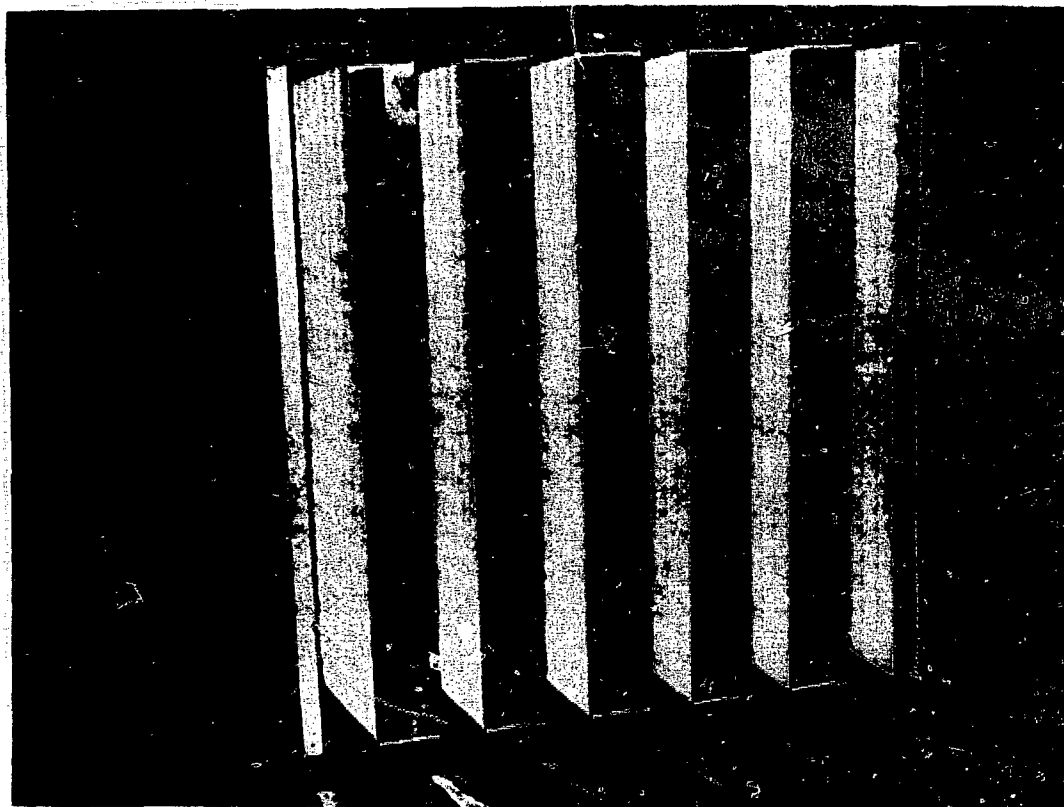


Figure 1. HEPA filter of continental European design.

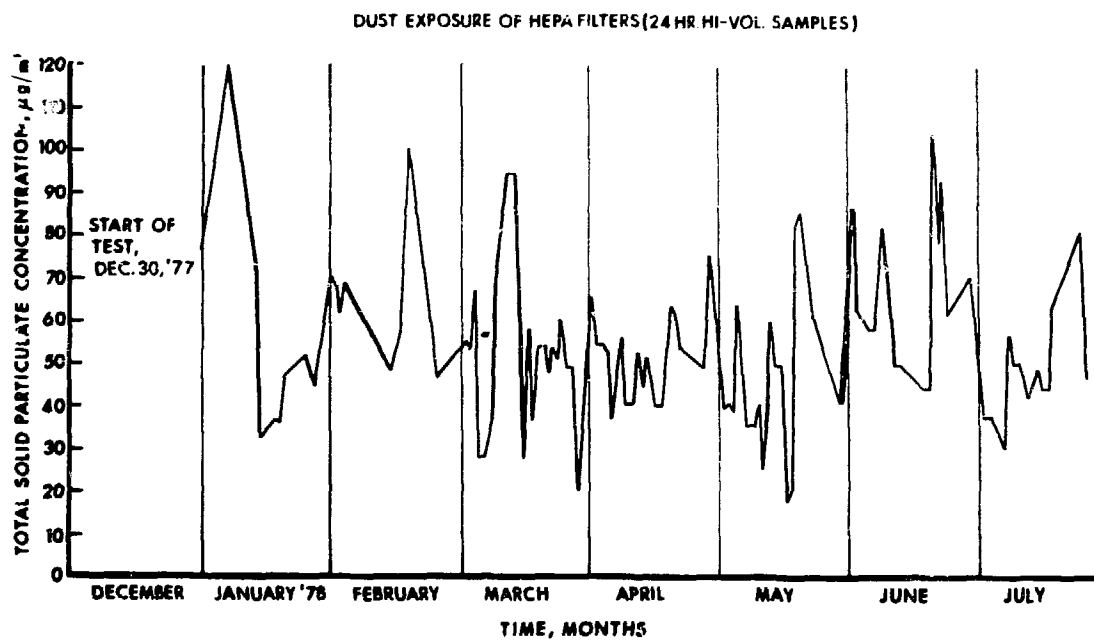


Figure 2. Dust exposure of HEPA filters (24 hr. Hi-Vol samples).

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size will be a close match for the aerosols these filters are likely to be exposed to in practice. Therefore, when a decision was made to test the life extension properties of European-design HEPA filters, a test location was selected on the open roof of the 15-story building that houses the Harvard Air Cleaning Laboratory. Dust concentrations at this site are measured daily with high volume air samplers using a technique published by the U.S. EPA and widely used throughout the world for 24-hour average total suspended particulated matter determinations. Figure 2 shows 24 hour average values since December 1977 at this location. Dust concentration fluctuates between 30 and 95 micrograms total dust per cubic meter of air ($\mu\text{g}/\text{m}^3$) with an average of $55 \mu\text{g}/\text{m}^3$ over a period of approximately 7 months.

The test is designed to compare the service life of an American-design filter operated at 1,000 cfm with a European-design filter operated at its rated flow rate of 1800 cfm and at a downrated flow-rate of 1000 cfm. For this purpose, three identical 24 x 24 x 11 5/8 in. filter test rigs have been constructed side by side on the roof, Figure 3. Each consists of (1) an inlet louvered grill to exclude snow and rain, (2) a straight 24 x 24 inch cross-section entry duct 4 feet long to straighten flow before entry to the filter and provide greater protection of the filter from the elements, (3) the test filter clamped between flanges with filter edge gaskets on both faces, (4) a flow regulating slide damper and, (5) an exhaust blower-motor set of appropriate size for the desired flow rate at resistance in excess of 4 in.w.g. across the test filter. Airflow resistance of the entry grill has been calibrated to measure airflow rate. The installation is shown in Figure 3.

European-design filters were purchased from Luwa Co. of Switzerland. They are designated "Luwa Standard Cells, Type N" and contain 35 m^2 of active glass paper filter surface. The filter paper used in these Luwa filters is manufactured by Dexter Corp., Windsor Locks, Connecticut and was not, then, used by any U.S. manufacturer of HEPA filters. Therefore, it was necessary to obtain a roll of this paper from Dexter Corp. and have it fabricated into U.S. filters corresponding to MIL-F-51068D. American Air Filter Co. of Louisville, KY fabricated our 1,000 cfm filters with aluminum separators, rubber base adhesive, and the Dexter/Luwa glass filter paper.

Results

The test filters were installed, tested in-place with DOP, and they have been in continuous operation since December 1977 at the pre-selected airflow rates. Filters are inspected daily, airflow readjusted, when needed, and filter pressure drop noted. At the same time, the 24-hour high volume air sampling filter is changed. High volume air sampling is maintained on a continuous basis over weekends and holidays. Figure 4 shows the starting (clean) resistance of each filter and the increase of resistance with time to the end of July 1978. It will be apparent from the figure that several reversible resistance increases have occurred over the test period; the most marked during January 1978. This peak resulted from a heavy snow fall that partially covered the louvered inlet grills and permitted snow to reach the filter inside, but after drying, airflow resistance returned to its former level. The later temporary

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Figure 3. HEPA filter test rigs on roof on 15 story building.

AIRFLOW RESISTANCE CHANGES WITH SERVICE TIME

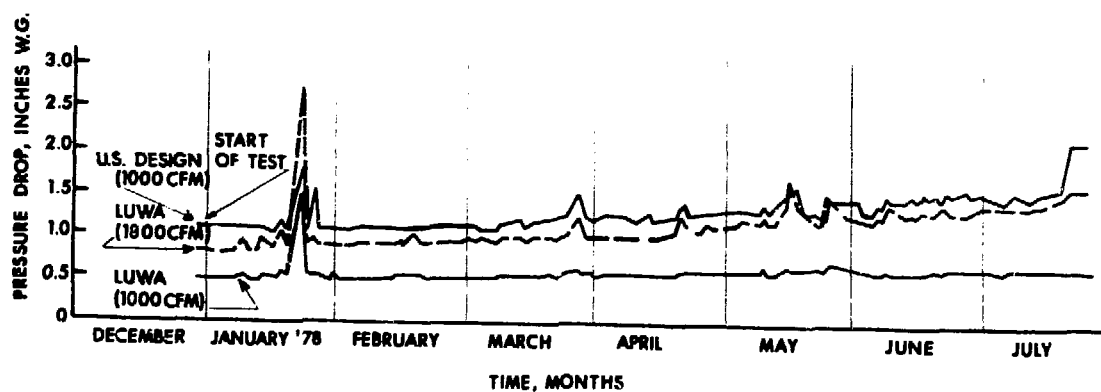


Figure 4. Airflow resistance changes with service time.

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peaks are related to periods of heavy rain.

A total of 459 grams (1 lb.) of atmospheric dust has entered each of the filters operated at 1,000 cfm and 826 grams (1.8 lb.) has entered the 1,800 cfm filter. One gram of dust has been recovered from the bottom of the entry duct leading to the 1,800 cfm filter and two grams from each of the entry ducts leading to the filters operated at 1,000 cfm. It is assumed that the remainder has deposited on the surfaces of the filters. The U.S.-design 1,000 cfm filter had an initial airflow resistance of 1.1 in. w.g. and this had increased to 2.2 in. w.g. by July 31, 1971. The 1,000 cfm Luwa filter (rated at 1800 cfm) started clean at 0.50 in. w.g. and has increased to 0.66 in. w.g. The Luwa filter operated at 1800 cfm started with a clean resistance of 0.83 in. w.g. and this has increased to 1.6 in. w.g. with the accumulation of 1.8 lb. of dust. From the figure, it will be clear that the U.S. filter and the 1800 cfm Luwa filter are responding in a parallel manner to the accumulation of dust but that the Luwa filter operating at 1,000 cfm is generating a resistance curve with a much flatter slope.

Future Plans

It is intended that this test will continue for the next two years and that the filters currently under test will be joined this month by three additional filters: one Sofiltra-Poelman unit of European design (Filtr A Air) operated at 1800 cfm and one operated at 1,000 cfm plus a new high capacity (2,000 cfm) filter manufactured by the American Air Filter Co. It is intended that these, also, will be operated for at least two years under the exposure conditions that have been described.

DECONTAMINATION OF HEPA FILTERS

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Abstract

Mound Facility, during many years of plutonium-238 experience, has recovered over 150 kg of plutonium-238. Much of this material was recovered from HEPA filters or from solid wastes such as sludge and slag. The objective of this task was to modify and improve the existing nitric acid leaching process used at Mound so that filters from the nuclear fuel cycle could be decontaminated effectively. Various leaching agents were tested to determine their capability for dissolving PuO_2 , UO_2 , U_3O_8 , AmO_2 , NpO_2 , CmO_2 , and ThO_2 in mixtures of the following: HNO_3 -HF; HNO_3 -HF- H_2SO_4 ; and HNO_3 -(NH_4) $_2\text{Ce}(\text{NO}_3)_6$. Adsorption isotherms were obtained for two leaching systems. In some tests simulated contaminated HEPA filter material was used, while in others actual spent glovebox filters were used.

The maximum decontamination factor of 833 was achieved in the recovery of plutonium-238 from actual filters. The dissolution was accomplished by using a six-stage process with 4N HNO_3 -0.23M (NH_4) $_2\text{Ce}(\text{NO}_3)_6$ as the leaching agent. Thorium oxide was also effectively dissolved from filter media using a mixture of nitric acid and ceric ammonium nitrate. Sodium carbonate and Na_2CO_3 - KNO_3 fusion tests were performed using simulated PuO_2 -contaminated filter media at various temperatures. Approximately 70 wt% of the PuO_2 was soluble in a mixture composed of 70 wt% Na_2CO_3 -30 wt% KNO_3 (heated for 1 hr at 950°C).

Introduction

Efforts were directed toward determining the dissolution parameters of PuO_2 , UO_2 , and U_3O_8 ** in various leaching reagents. The reagents used were various concentrations of HNO_3 , HNO_3 -HF, HNO_3 -HF- H_2SO_4 , HNO_3 -(NH_4) $_2\text{Ce}(\text{NO}_3)_6$, and HNO_3 - H_2SO_4 . Two types of plutonium dissolution tests were conducted with 8N HNO_3 . In the first test, PuO_2 was placed in 8N HNO_3 at boiling temperature. In the other test, HEPA filter media contaminated with PuO_2 were added to boiling 8N HNO_3 . The dissolution rates of PuO_2 as measured by the alpha activity of the acid media were determined for each test; as shown in Figure 1, the apparent dissolution rate was less in the test using PuO_2 -contaminated filter media. After 14 hr of heating, for example, there was a 3% difference (11% minus 8%) in dissolution rates. At this point, plutonium adsorption isotherms were developed to determine whether any of the dissolved plutonium was being adsorbed by the filter media.

*Mound Facility is operated by Monsanto Research Corporation for the U.S. Department of Energy under Contract No. EY-76-C-04-0053.

**The uranium oxides were depleted uranium-238, whereas the PuO_2 was a mixture of 80 wt% plutonium-238, 16 wt% plutonium-239, 2.5 wt% plutonium-240, 0.8 wt% plutonium-241, 0.2 wt% plutonium-242, plus small amounts of other actinides calcined at 750°C.

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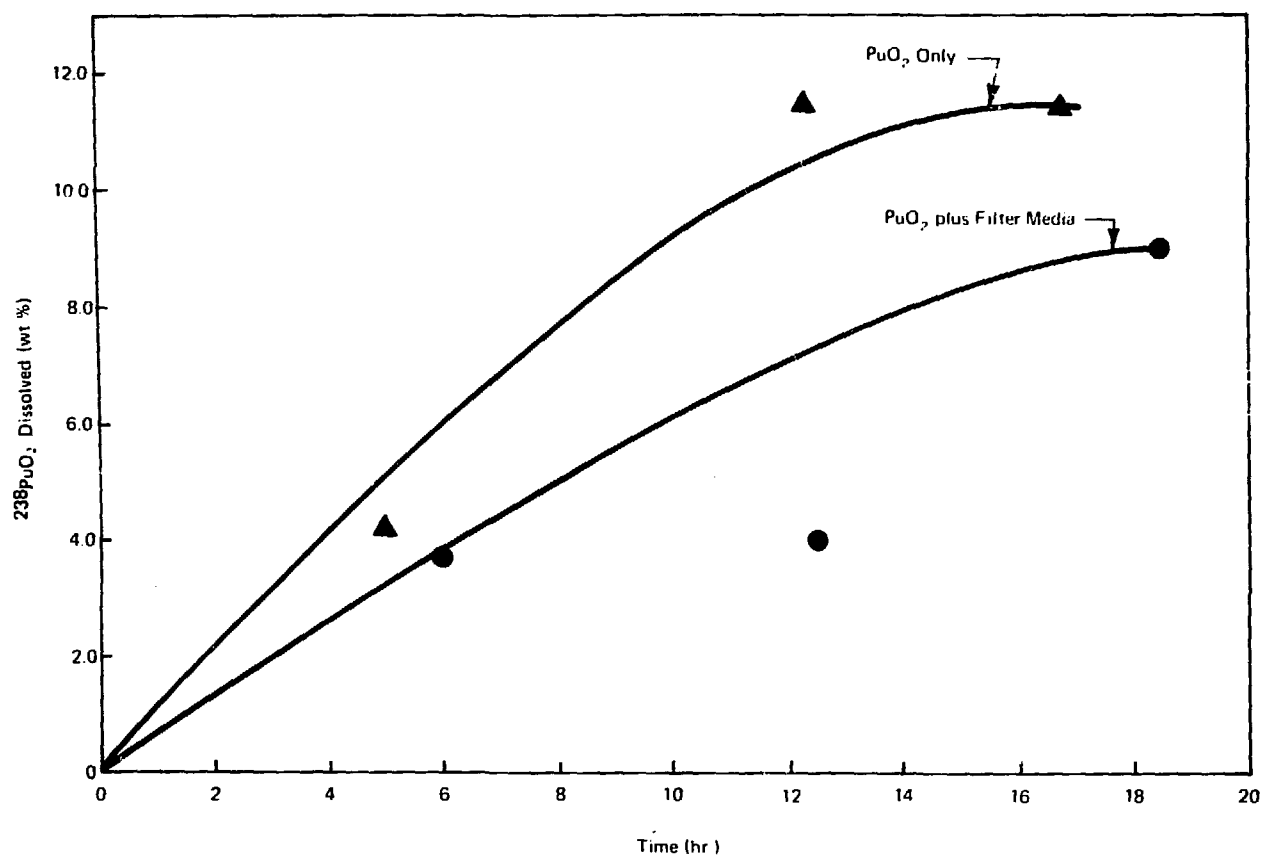


Figure 1 - Effect of filter media on dissolution rates of PuO_2 in boiling 8N HNO_3 .

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This is of great importance since the amount of adsorbed plutonium would limit the decontamination factor that could be achieved. Adsorption isotherms were determined for the Pu-HNO₃ and Pu-HNO₃-HF systems.

Efforts were also directed toward the determination of dissolution parameters in various reagents of PuO₂* and PuO₂-UO₂** solid solution. The reagents used were various concentrations of HNO₃, -HF-H₂SO₄, HNO₃-HF, HNO₃-(NH₄)₂Ce(NO₃)₆, and HNO₃-H₂SO₄. In one series of tests, simulated contaminated HEPA filter media was used. This material was prepared by mixing shredded filter media with actinide oxides. In another series, actual HEPA glovebox filter media contaminated with PuO₂ were used. Fusion tests were also completed using simulated PuO₂-contaminated filter media. Both Na₂CO₃ and Na₂CO₃-KNO₃ were investigated as possible fusion agents.

Dissolution parameters (in various reagents) of americium-241 and plutonium-239 oxide mixtures, uranium-233 oxide, neptunium-237 oxide, curium-244 oxide, thorium-232 oxide, and PuO₂ were determined. The reagents used were various concentrations of HNO₃-HF-H₂SO₄, HNO₃-(NH₄)₂Ce(NO₃)₆, HNO₃-HF and HNO₃. Both simulated contaminated HEPA filter media and actual glovebox filter media from spent filters were used. The maximum decontamination factor was 833, achieved using a six-stage dissolution process. Also, PuO₂ was fused with Na₂CO₃ various elevated temperatures, and a dissolution percentage was determined.

Experimental Work

Adsorption Studies

Standard plutonium solutions were prepared by dissolving PuO₂ in boiling 8N HNO₃. The HEPA filter media were shredded and ball-milled to <40 mesh. The filter media were placed in 100 ml of the prepared PuO₂-acid solution and stirred magnetically for several hours to reach equilibrium. A blank sample was run without filter media to correct for adsorption on the test beakers. All tests were made at ambient temperature (~20°C).

Figure 2 shows the adsorption of plutonium on filter media in different acids. The ordinate is x/m, where x = milligrams of plutonium-238 adsorbed and m = milligrams of filter media. The abscissa is C, which is the concentration of the solution in equilibrium with the

*The uranium oxides were depleted uranium-238, whereas the PuO₂ was a mixture of 80 wt% Plutonium-238, 16 wt% Plutonium-239, 2.5 wt% Plutonium-240, 0.8 wt% Plutonium-241, 0.2 wt% Plutonium-242, plus small amounts of other actinides calcined at 750°C.

**The uranium was uranium-238 while the plutonium was 85.0 wt% plutonium-239, 12.0 wt% plutonium-240, 1.7 wt% plutonium-241, 0.75 wt% plutonium-238, plus small amounts of other actinides.

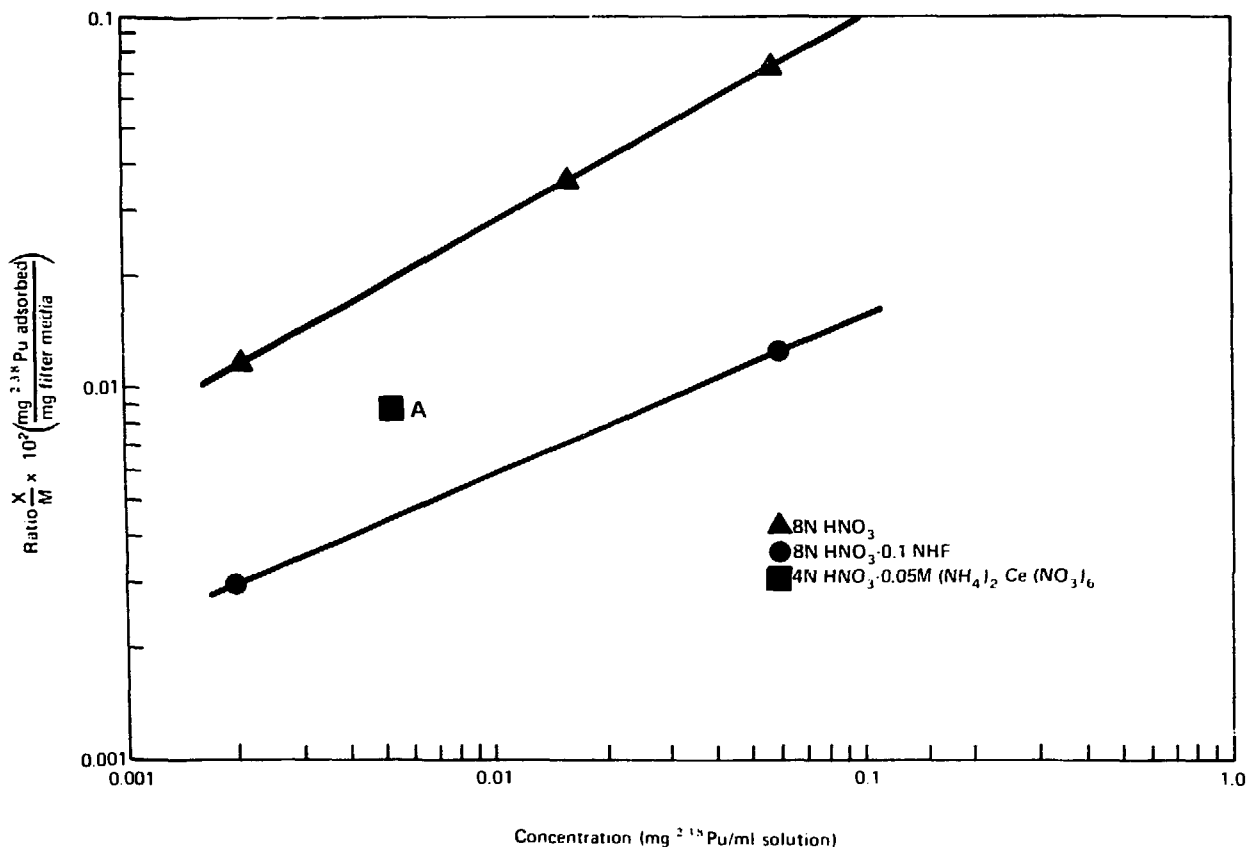


Figure 2 - Adsorption of ^{238}Pu on filter media at ambient temperature ($\sim 20^\circ\text{C}$) in different acids.

filter media (expressed in milligrams of plutonium-238 per milliliter of solution). The Freundlich equation relates x/m and C as follows:

$$x/m = kC^{1/n} \quad (1)$$

or

$$\log (x/m) = \log k + (1/n) \log C \quad (2)$$

The plot of $\log (x/m)$ as a function of $\log C$ should be a straight line with slope $1/n$ and $\log k$ the point of intercept. The straight lines in Figure 2 are such a plot. It should be noted that for any given concentration, the x/m ratio is smaller for the 8N HNO_3 - 0.1N HF system. This means that a larger decontamination factor could be achieved using the HNO_3 - HF leaching agent (assuming that the same percentage of PuO_2 is dissolved by the reagent in the dissolution step).

Tests were also performed to determine the solubility of the filter media in various dissolution reagents. It was determined that approximately 30 wt% of the filter media is soluble in 8N HNO_3 , and approximately 50 wt% of the filter media dissolves in 8N HNO_3 - 0.1N HF . Also, in another adsorption test it was found that the use of 4N HNO_3 - 0.05M ceric ammonium nitrate (see point A, Figure 2) would result in a decontamination factor between that obtained with 8N HNO_3 - 0.1N HF and that with 8N HNO_3 (assuming each leaching agent was equally effective in dissolving PuO_2).

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PuO₂ Dissolution Studies

Contaminated HEPA filter media were prepared by mixing PuO₂* powder with shredded filter media. The plutonium-238 concentration of the mixture was 11 mg/cm³ of prepared media. Small samples of this prepared mixture (about 3 g) were added to glass beakers containing 250 ml of leaching solution. All tests were performed at boiling temperatures. Samples were withdrawn periodically and the plutonium-238 concentration of the solution was determined. The contents of the beakers were stirred every 2 hr. The solution volume and concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid.

Table I lists results from dissolution tests for PuO₂ in various acid mixtures. The HNO₃-H₂SO₄ mixtures were relatively ineffective dissolution agents as illustrated in Figure 3. A fivefold increase in H₂SO₄ concentration increased the dissolution rate by only 3%. The maximum weight percent of plutonium-238 dioxide dissolved was only 18% after an 18-hr reaction time. Figure 4 illustrates the effect of initial cerium(IV) concentration on the dissolution rate. The 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ leaching agent was by far the most effective of the three HNO₃-cerium(IV) solutions, with 88% of the plutonium-238 dioxide dissolved in 3 hr.

The reaction mechanism for dissolving PuO₂ in HNO₃-cerium(IV) solutions is shown in Equations 3 and 4.



Because stoichiometric quantities of cerium(IV) are required for complete dissolution of the PuO₂, large amounts of cerium(IV) are required to dissolve large quantities of PuO₂. Therefore, it would seem advantageous to use small amounts of cerium(IV) and then add oxidizing compounds to oxidize the cerium(III) to cerium(IV). An example of this would be the addition of KMnO₄ to the depleted solution. This would cause the Mn⁺⁷ + 3e⁻ ⇌ Mn⁺⁴ reaction which would oxidize the depleted cerium as follows:



where one mole of manganese oxidizes three moles of cerium. This was done in acid tests 25 and 25A (see Table I). Approximately 1 g of KMnO₄ was added to a depleted 4N HNO₃-0.005M(NH₄)₂Ce(NO₃)₆ solution and the solution heated an additional 15 hr. In this period of time, the concentration of PuO₂ dissolved increased significantly from 12 wt% in the depleted solution to 26 wt% in the regenerated solution.

*The PuO₂ was fine powder having a composition of 80 wt% plutonium-238, 16 wt% plutonium-239, 2.5 wt% plutonium-240, 0.8 wt% plutonium-241, 0.2 wt% plutonium-242, plus small amounts of other actinides. It was calcined for 2 hr at 950°C.

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Table 1 - DISSOLUTION OF PLUTONIUM DIOXIDE IN VARIOUS ACID MIXTURES

| Acid
Test
Number | Reagent Composition ^a | Time
Heated
(hr) | PuO ₂
Dissolved
(wt %) |
|------------------------|---|------------------------|---|
| 19-1 | 4N HNO ₃ -0.05M CAN | 5 3/4 | 42.9 |
| 19-2 | 4N HNO ₃ -0.05M CAN | 12 1/4 | 41.4 |
| 19-3 | 4N HNO ₃ -0.05M CAN | 18 3/4 | 38.5 |
| 20-1 | 8N HNO ₃ -0.1N H ₂ SO ₄ | 5 3/4 | 14.4 |
| 20-2 | 8N HNO ₃ -0.1N H ₂ SO ₄ | 12 | 12.7 |
| 20-3 | 8N HNO ₃ -0.1N H ₂ SO ₄ | 18 | 15.9 |
| 21-1 | 12N HNO ₃ -0.1N HF-0.1N H ₂ SO ₄ | 4 3/4 | 115.2 |
| 21-2 | 12N HNO ₃ -0.1N HF-0.1N H ₂ SO ₄ | 10 1/2 | 107.1 |
| 22-1 | 12N HNO ₃ -0.1N HF | 1 3/4 | 87.0 |
| 22-2 | 12N HNO ₃ -0.1N HF | 6 3/4 | 94.1 |
| 22-3 | 12N HNO ₃ -0.1N HF | 11 1/2 | 107.9 |
| 25-1 | 4N HNO ₃ -0.005M CAN | 2 1/4 | 11.1 |
| 25-2 | 4N HNO ₃ -0.005M CAN | 7 3/4 | 13.4 |
| 25-3 | 4N HNO ₃ -0.005M CAN | 13 1/4 | 11.9 |
| 25-A-1 | 4N HNO ₃ -0.005M CAN- KMnO ₄ | 3 | 13.4 |
| 25-A-2 | 4N HNO ₃ -0.005M CAN- KMnO ₄ | 8 1/2 | 18.9 |
| 25-A-3 | 4N HNO ₃ -0.005M CAN- KMnO ₄ | 14 3/4 | 26.3 |
| 26-1 | 12N HNO ₃ -0.01N HF | 1 3/4 | 31.6 |
| 26-2 | 12N HNO ₃ -0.01N HF | 7 1/4 | 41.0 |
| 26-3 | 12N HNO ₃ -0.01N HF | 12 3/4 | 40.9 |
| 27-1 | 12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄ | 1 1/4 | 30.1 |
| 27-2 | 12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄ | 6 3/4 | 33.1 |
| 27-3 | 12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄ | 12 1/4 | 36.3 |
| 28-1 | 12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄ | 1 | 96.3 |
| 28-2 | 12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄ | 6 1/2 | 100.9 |
| 28-3 | 12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄ | 12 | 101.8 |
| 29-1 | 8N HNO ₃ -0.5N H ₂ SO ₄ | 5 1/2 | 15.8 |
| 29-2 | 8N HNO ₃ -0.5N H ₂ SO ₄ | 10 3/4 | 17.4 |
| 29-3 | 8N HNO ₃ -0.5N H ₂ SO ₄ | 15 1/4 | 17.4 |
| 30-1 | 12N HNO ₃ -0.05N HF 0.01N H ₂ SO ₄ | 3 | 95.9 |
| 30-2 | 12N HNO ₃ -0.05N HF 0.01N H ₂ SO ₄ | 8 1/4 | 88.0 |
| 30-3 | 12N HNO ₃ -0.05N HF 0.01N H ₂ SO ₄ | 14 1/2 | 97.7 |
| 31-1 | 4N HNO ₃ -0.1M CAN | 3 | 87.7 |
| 31-2 | 4N HNO ₃ -0.1M CAN | 8 1/4 | 88.0 |
| 31-3 | 4N HNO ₃ -0.1M CAN | 14 1/4 | 80.6 |
| 32-1 | 8N HNO ₃ -K ₂ S ₂ O ₈ | 3 | 0.86 |
| 32-2 | 8N HNO ₃ -K ₂ S ₂ O ₈ | 7 1/2 | 0.82 |
| 32-3 | 8N HNO ₃ -K ₂ S ₂ O ₈ | 13 1/2 | 1.89 |

^aCAN is abbreviation for ceric ammonium nitrate

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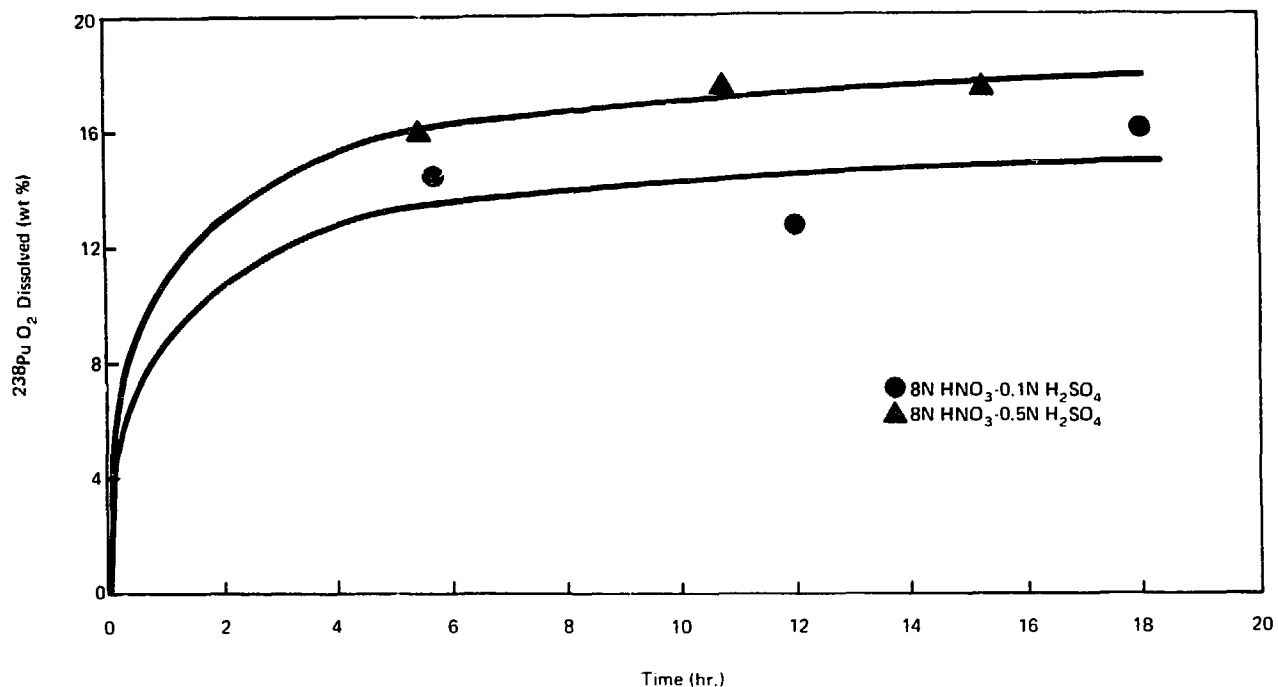


Figure 3 - Effect of H_2SO_4 concentration on dissolution rate.

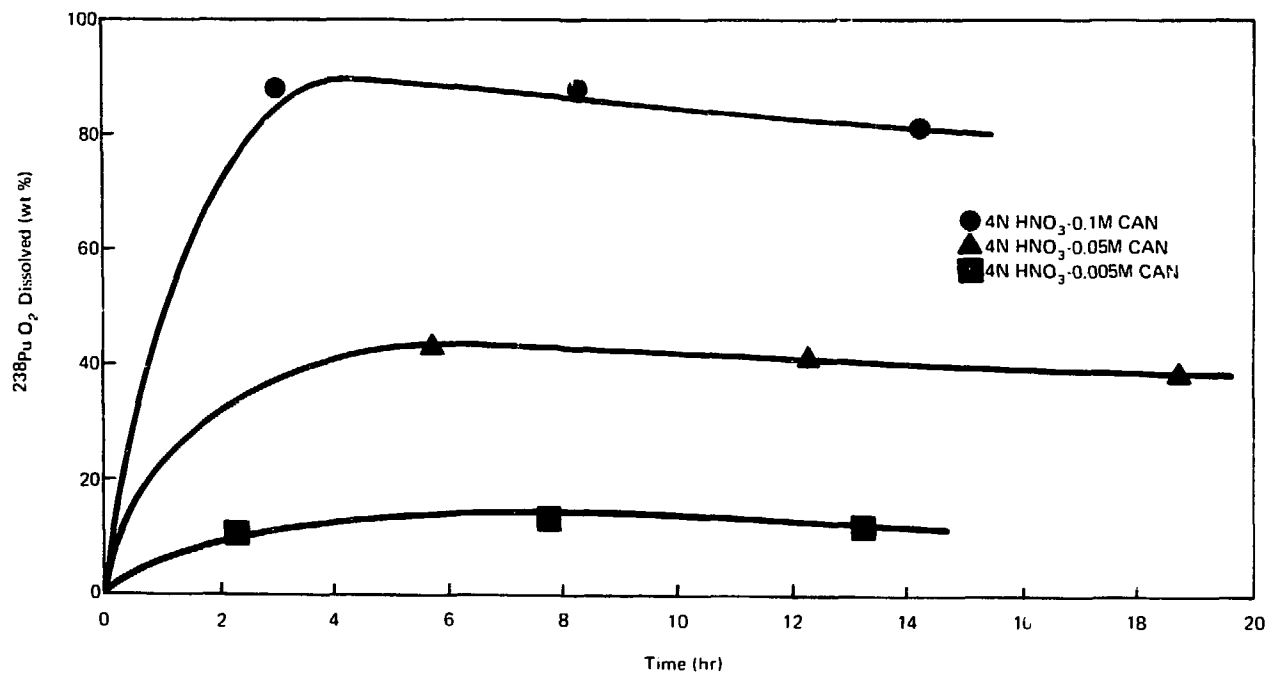


Figure 4 - Effect of ceric ammonium nitrate (CAN) concentration on dissolution rate.

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This method does, however, add manganese and potassium to the dissolved salts of the waste stream.

Another experiment was conducted using 8N HNO_3 and $\text{K}_2\text{S}_2\text{O}_8$ (potassium persulphate) as an oxidizing agent (see Table I, acid test 32). After $13\frac{1}{2}$ hr of reaction time at boiling temperature, only 1.89% of the PuO_2 had dissolved. Figure 1 shows that approximately 7.5% of the PuO_2 dissolved in $13\frac{1}{2}$ hr with no additional oxidizing agent present. Thus the $\text{K}_2\text{S}_2\text{O}_8$ actually decreased the dissolution rate of the PuO_2 .

Figures 5 and 6 show the effect of the HF concentration on the dissolution rate of PuO_2 . For the HNO_3 -HF system, a tenfold increase in the HF concentration increases the concentration of dissolved PuO_2 from about 40 wt% to approximately 100 wt% for 9 hr of reaction time (Figure 5). A twofold increase in the HF concentration for the HNO_3 -HF- H_2SO_4 system increases the concentration of dissolved PuO_2 from 94 wt% to 100 wt% for 9 hr of reaction time (Figure 6). As can be seen in Table I, any dissolution test in which the HF concentration was 0.05N, or greater, was successful in obtaining a 90 to 100% dissolution of the PuO_2 . The effect of sintering temperature on dissolution rate for various reagents can be seen in Figures 7 and 8.

UO_2 and U_3O_8 Dissolution Studies

Contaminated filter media samples were prepared by mixing 0.2 g of either UO_2 or U_3O_8 * with 2.8 g of shredded filter media. These samples were combined in a glass round-bottom flask containing 250 ml of the desired leaching solution. The flasks were attached to reflux condensers, and the contents were heated and refluxed at boiling temperatures. Samples were withdrawn periodically, and the uranium concentration of the solution was determined.

Table 2 lists results of dissolution tests for uranium oxide in various acid mixtures; in every test the uranium oxides dissolved rapidly. The percentage of uranium oxide dissolved was always greater than 90%; 8N HNO_3 performed as well as any of the acid mixtures, dissolving essentially 100% of the UO_2 or U_3O_8 after 6 hr of heating. As a comparison, the percentage of PuO_2 dissolved after 6 hr was ~4% (see Figure 1).

PuO_2 -75 wt% UO_2 Solid Solution Studies

The solid solution was composed of plutonium** and uranium-238 oxides which had been fired at 1600°C in a reducing atmosphere. Six dissolution tests were performed (see Table III). The powdered PuO_2 - UO_2 solid solution was mixed thoroughly with shredded filter media using a ratio of 1 g of solid solution to 12 g of filter media. A small sample of the mixture (approximately 3 g) was added to a glass beaker containing 250 ml of leaching solution. Samples were withdrawn periodically

*The U_3O_8 had been fired at 950°C for 2 hr.

**Assay of the plutonium was 85.0 wt% plutonium-239, 12.0% plutonium-240, 1.7% plutonium-241, 0.75% plutonium-238, plus small amounts of other actinides.

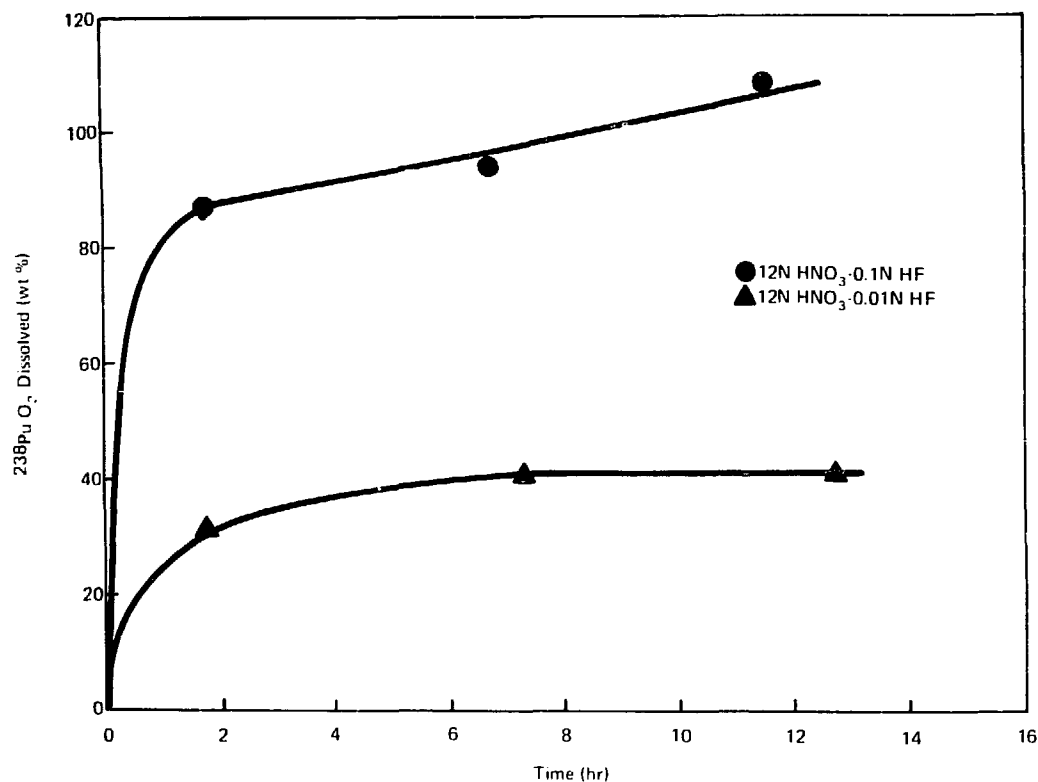


Figure 5 - Effect of HF concentration on dissolution rate of PuO₂.

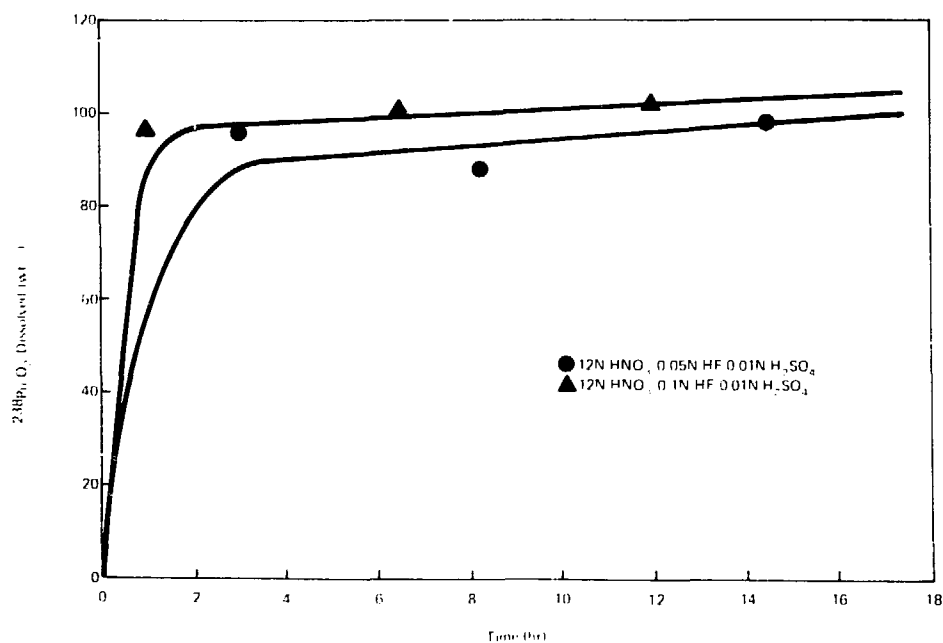


Figure 6 - Effects of HF and H₂SO₄ concentrations on dissolution rates.

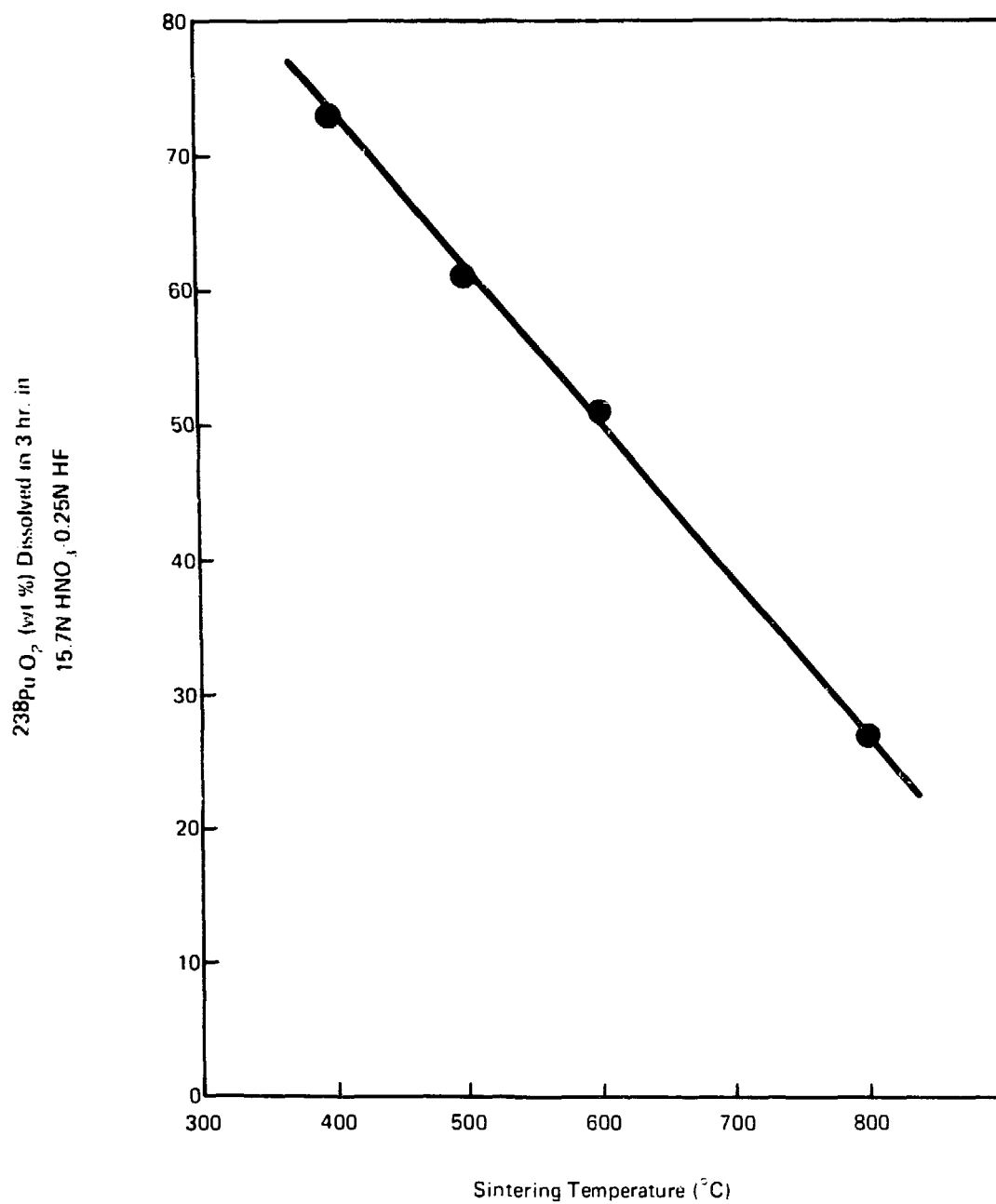


Figure 7 - Effect of sintering temperature on dissolution rate (2).

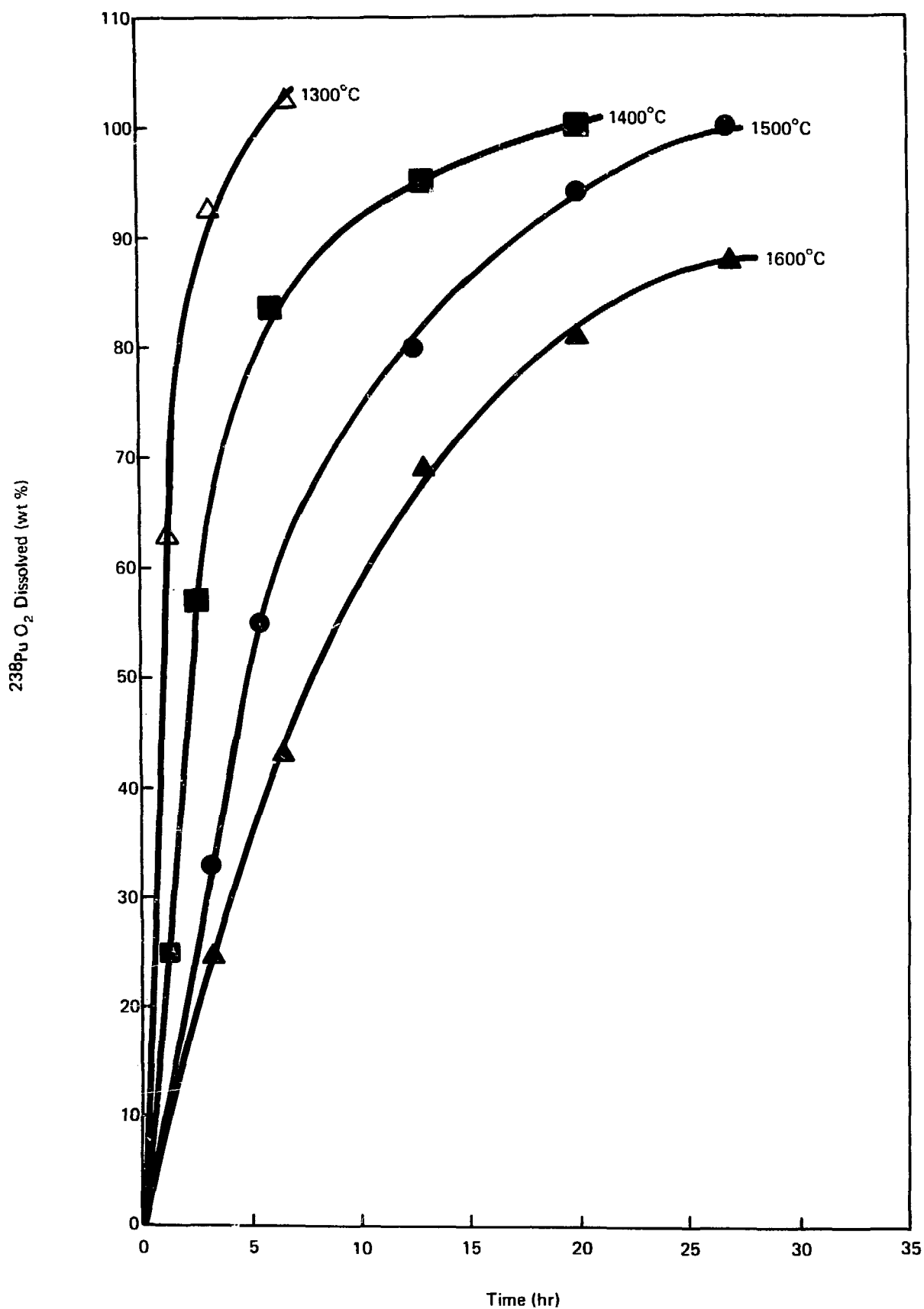


Figure 8 - Effect of sintering temperature on plutonium dioxide dissolution in 4N HNO_3 - 0.15M cerium(IV) (3).

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Table II - DISSOLUTION OF URANIUM OXIDES IN VARIOUS ACID MIXTURES

| Acid Test Number | Reagent Composition | Time Heated (hr) | UO ₂ or U ₃ O ₈ Dissolved (wt %) |
|------------------------------------|--|------------------|---|
| UO ₂ -1-1 | 12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄ | 1 1/2 | 92.8 |
| UO ₂ -1-2 | 12N HNO ₃ -0.01N HF-0.1N H ₂ SO ₄ | 6 1/2 | 94.1 |
| UO ₂ -2-1 | 12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄ | 1 | 91.0 |
| UO ₂ -2-2 | 12N HNO ₃ -0.1N HF-0.01N H ₂ SO ₄ | 7 | 99.6 |
| UO ₂ -3-1 | 12N HNO ₃ -0.1N H ₂ SO ₄ | 4 3/4 | 99.3 |
| UO ₂ -4-1 | 8N HNO ₃ | 5 3/4 | 102.8 |
| U ₃ O ₈ -5-1 | 8N HNO ₃ | 1 | 107.0 |
| U ₃ O ₈ -6-1 | 12N HNO ₃ -0.1N H ₂ SO ₄ | 1 | 103.0 |

and the percentage of solid solution dissolved was determined. The solution volume and concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid.

As can be seen in Table III, four reagents were successful in dissolving greater than 93% of the PuO₂-UO₂ in 2 hr. These were 12N HNO₃-0.1N HF, 12N HNO₃-0.05N HF-0.01N H₂SO₄, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 8N HNO₃. Both the 4N HNO₃ and the 4N HNO₃-0.1N H₂SO₄ were unsuccessful in attaining a 90% dissolution even when leaching times of 7 hr were used. Figure 9 shows the effect of HNO₃ concentration on the dissolution rate. Doubling the acid normality (from 4N to 8N) doubled the percent dissolved (after 2 hr of heating) from 46% to 93%. Therefore, it can be concluded that 8N HNO₃, 12N HNO₃-0.1N HF, 12N HNO₃-0.05N HF-0.01N H₂SO₄, and 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ would be acceptable leaching reagents for PuO₂-UO₂ solid solution whereas 4N HNO₃ and 4N HNO₃-0.1N H₂SO₄ are unacceptable.

Salt Fusion Studies

Sodium carbonate and Na₂CO₃-KNO₃ fusions were completed using PuO₂* contaminated filter media. Small samples were prepared by thoroughly mixing 0.1 g PuO₂ and 1.5 g of filter media. Approximately 13 g of salt (Na₂CO₃ or Na₂CO₃-KNO₃) was placed in a platinum crucible and 1.6 g of contaminated filter media was then added. This was mixed thoroughly and then heated slowly to 950°C. The crucible and contents were maintained at 950°C for 1 hr and then allowed to cool slowly to ambient temperature. The cooled melt was then removed from the crucible and dissolved in 4N HNO₃. The acid was maintained at boiling temperature for at least 1 hr and subsequently sampled for plutonium-238 concentration. Table IV tabulates the results achieved in these

*The PuO₂ was a fine powder, having a composition of 80 wt% plutonium-238, 16% plutonium-239, 2.5% plutonium-240, 0.8% plutonium-241, 0.2% plutonium-242, plus small amounts of other actinides, calcined at 950°C.

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Table III - $\text{PuO}_2\text{-UO}_2$ Solid Solution Dissolution

| Acid Test
Number | Reagent Composition | Time Heated (hr) | $\text{PuO}_2\text{-UO}_2$ |
|---------------------|--|------------------|----------------------------|
| | | | Dissolved (wt %) |
| Pu-U-1 | 12N HNO_3 -0.1N HF | 2 | 100.0 |
| Pu-U-2 | 12N HNO_3 -0.05N HF-0.01N H_2SO_4 | 2 | 97.6 |
| Pu-U-3 | 4N HNO_3 -0.1M CAN ^a | 2 | 94.8 |
| Pu-U-4A | 8N HNO_3 | 1 | 71.8 |
| Pu-U-4B | 8N HNO_3 | 2 | 93.2 |
| Pu-U-5A | 4N HNO_3 | 2-1/2 | 49.6 |
| Pu-U-5B | 4N HNO_3 | 4-1/2 | 54.2 |
| Pu-U-5C | 4N HNO_3 | 7 | 60.0 |
| Pu-U-6 | 4N HNO_3 -0.1N H_2SO_4 | 3-3/4 | 80.2 |

^aAbbreviation for ceric ammonium nitrate.

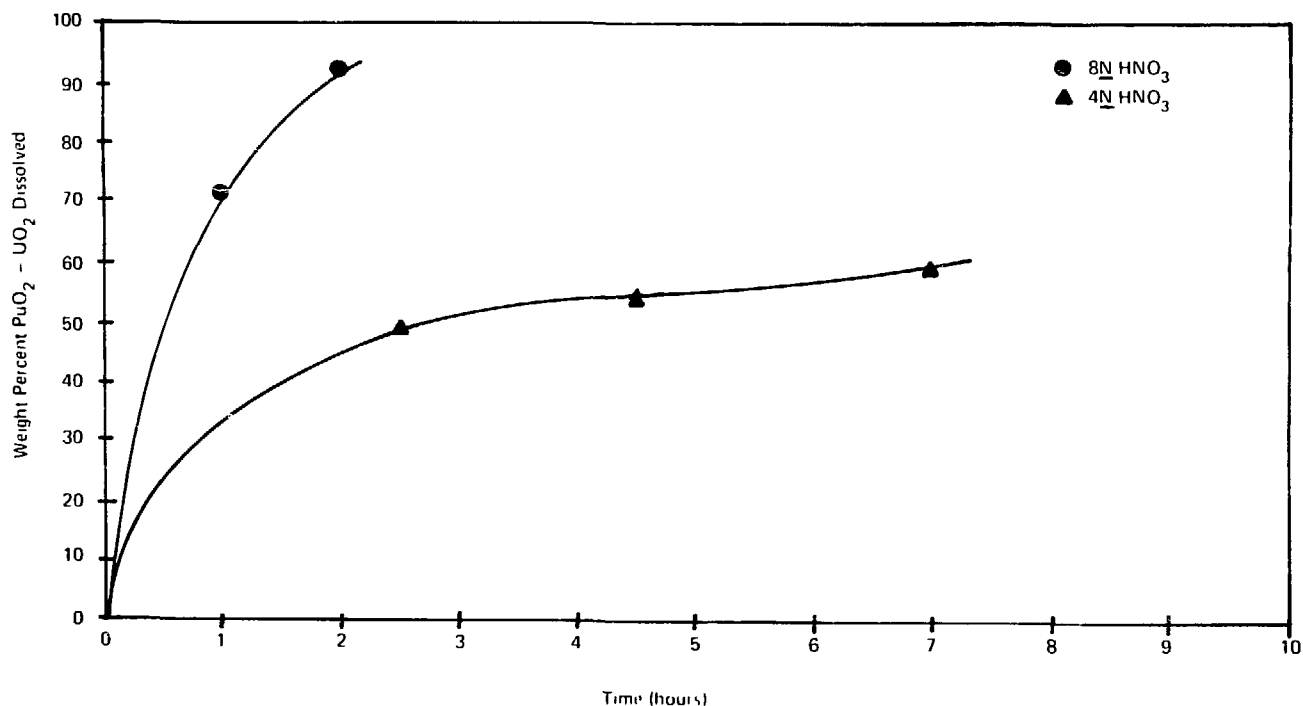


Figure 9 - Effect of HNO_3 concentration on dissolution rate of $\text{PuO}_2\text{-UO}_2$

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Table IV - Na_2CO_3 - KNO_3 Fusion of PuO_2

| Fusion Test Number | Salt Composition (wt %) | Time Heated (hr) | Operating Temperature ($^{\circ}\text{C}$) | PuO_2 Solubilized (wt %) |
|--------------------|--|------------------|--|-----------------------------------|
| Pu-38 | 100% Na_2CO_3 | 1 | 950 | 8.83 |
| Pu-39 | Na_2CO_3 -8% KNO_3 | 1 | 950 | 27.8 |
| Pu-40 | Na_2CO_3 -16% KNO_3 | 1 | 950 | 48.7 |
| Pu-41 | Na_2CO_3 -30% KNO_3 | 1 | 950 | 71.0 |

fusion experiments. As can be seen, the maximum percent of dissolution obtained was 71% using a salt mixture of Na_2CO_3 -30 wt% KNO_3 .

Figure 10 shows the effect of potassium nitrate composition on PuO_2 solubility. The effect is linear between zero and 16% KNO_3 and nonlinear at higher potassium nitrate concentrations. Also because of time limitations, only three Na_2CO_3 - KNO_3 fusions were performed and the maximum concentration of potassium nitrate used was 30 wt%. It should be noted that the greater the concentration (wt%) of potassium nitrate, the more vigorous the reaction.

The temperature must be raised slowly in order to prevent the contents of the crucible from spilling over. The 71% recovery obtained using Na_2CO_3 -30 wt% KNO_3 is considered unsatisfactory for our purposes. The dissolution must be greater than 90% in order to achieve large decontamination factors. Perhaps by using greater than 30% KNO_3 this result can be achieved.

Dissolution of Actual HEPA Glovebox Filter Media

Small test samples of approximately 3 g each were removed from an actual glovebox filter contaminated with PuO_2 *. Each sample contained approximately 55 mg of plutonium-238. These samples were placed in a glass beaker containing 100 ml of leaching reagent. The leaching agents used were 4N HNO_3 -0.1M ceric ammonium nitrate and 12N HNO_3 -0.05N HF -0.01N H_2SO_4 . The tests were performed at boiling temperatures. The contents of the beakers were stirred every hour, and samples were withdrawn periodically for plutonium analysis. The solution volume and acid concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid. Table V contains the results obtained in these experiments. The average PuO_2 dissolution was 92.5 wt% using 4N HNO_3 -0.1M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ and 93.3 wt% using 12N HNO_3 -0.05N HF -0.01N H_2SO_4 . These compare well with the average dissolution percentages found for simulated filter media samples listed in Table I (85.4% and 93.9% respectively). Figure 11 is a graph of the dissolution rate for PuO_2 in

*The PuO_2 was a fine powder, having a composition of 80 wt% plutonium-238, 16% plutonium-239, 2.5% plutonium-240, 0.8% plutonium-241, 0.2% plutonium-242, plus small amounts of other actinides, calcined at 950°C .

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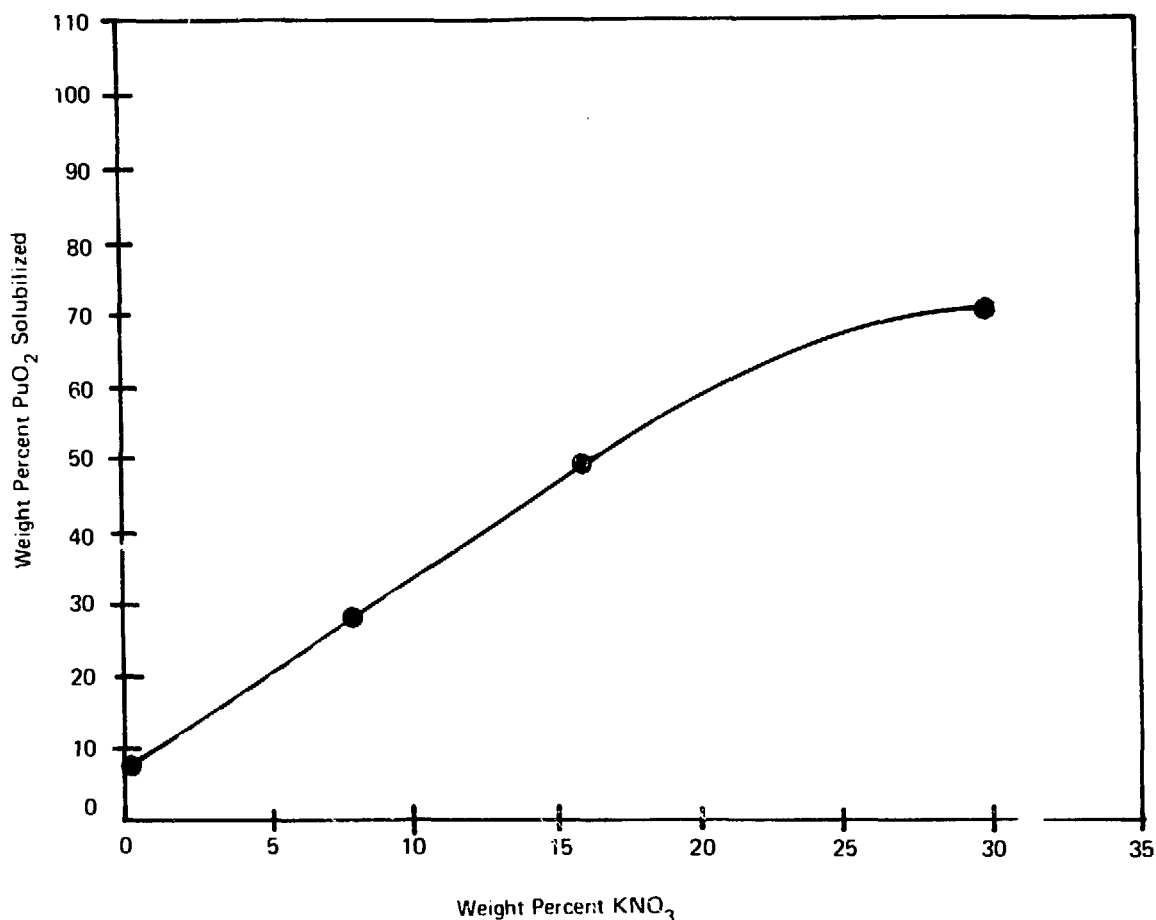


Figure 10 - Effect of salt composition on percent PuO₂ solubilized.

12N HNO₃-0.05N HF-0.01N H₂SO₄. It shows that the dissolution reaction is rapid, with approximately 90% of the PuO₂ dissolved in 4 hr.

Larger samples (40 g) of actual HEPA filter were treated with 12N HNO₃-0.05N HF-0.01N H₂SO₄. These experiments were conducted similarly to those previously described (using 3-g samples) except that different ratios of acid volume (liters) to plutonium-238 weight (grams) were used. Also the filter media were processed through several successive, identical stages in an attempt to obtain the greatest overall decontamination factor (see Table VI and Figure 12). As can be seen in Figure 6, a value of 0.67 for the ratio of acid volume:PuO₂ weight yields a 99.35% dissolution in five stages. On the other hand, a volume:weight ratio of 2.35 produces a 99.53% dissolution in three stages. Thus the decontamination factor is 154 for the lower volume:weight ratio and 213 for the higher ratio. These final dissolution percentages and decontamination factors were determined by calorimetry and gamma counting of the final filter media residues. All other dissolution percentages and decontamination factors were determined by alpha counting of solution samples.

The fact that a higher volume of acid:weight of PuO₂ ratio produces a higher dissolution is explained as follows: The HF in the

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Table V - Dissolution of Small Samples of Actual HEPA Filters

| Acid Test Number | Reagent Composition | Time Heated (hr) | PuO ₂ Dissolved (wt %) | Acid/Pu-238 Ratio (liters/grams) |
|------------------|---|------------------|-----------------------------------|----------------------------------|
| 3265-3A | 4N HNO ₃ -0.1M CAN | 3-1/2 | 103.2 | 1.84 |
| 3265-3B | 4N HNO ₃ -0.1M CAN | 5 | 103.3 | 1.84 |
| 3265-3C | 4N HNO ₃ -0.1M CAN | 11-3/4 | 81.9 | 1.84 |
| 3265-3D | 4N HNO ₃ -0.1M CAN | 14-3/4 | 81.5 | 1.84 |
| 3265-4A | 12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄ | 3-1/2 | 96.9 | 1.83 |
| 3265-4B | 12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄ | 5 | 89.1 | 1.83 |
| 3265-4C | 12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄ | 12-1/2 | 93.9 | 1.83 |

*Abbreviation for ceric ammonium nitrate - (NH₄)₂Ce(NO₃)₆.

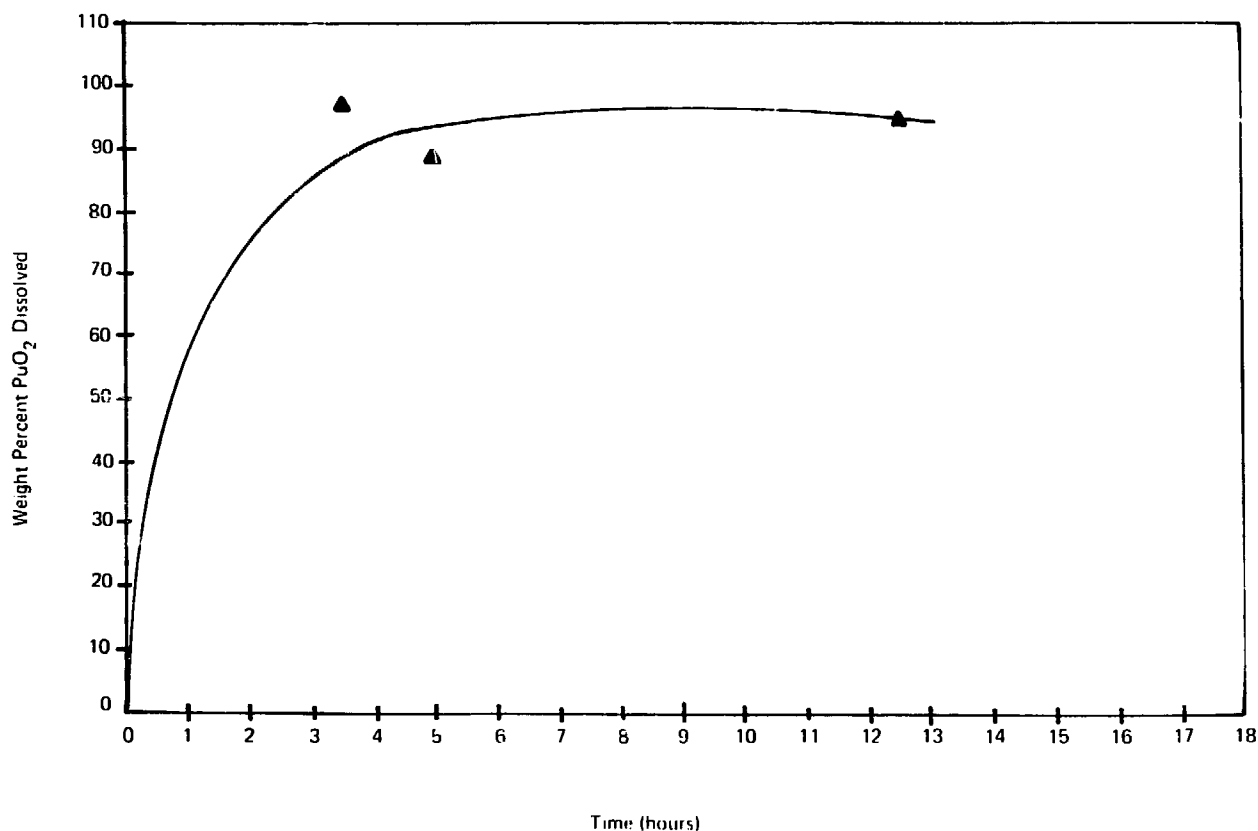


Figure 11 - Dissolution rate of PuO₂ in 12N HNO₃-0.05N HF-0.01N H₂SO₄.

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Table VI - 12N HNO₃-0.05N HF-0.01N H₂SO₄
Dissolution of Large Samples of Actual HEPA Filters

| Acid Test Number | Stage | Cumulative PuO ₂ Dissolved (wt %) | Acid/Pu-238 Ratio (liters/g) | Time Heated (hr) |
|------------------|-------|--|------------------------------|------------------|
| 3265-7-1 | 1 | 84.9 | 0.67 | 3-1/2 |
| 3265-7-2 | 2 | 88.9 | 0.67 | 5-1/4 |
| 3265-7-3 | 3 | 93.3 | 0.67 | 5-3/4 |
| 3265-7-4 | 4 | 95.8 | 0.67 | 7-1/4 |
| 3265-7-5 | 5 | 99.35 | 0.67 | 6-1/2 |
| 3265-9-1 | 1 | 94.2 | 2.35 | 10-1/2 |
| 3265-9-2 | 2 | 97.6 | 2.35 | 10-3/4 |
| 3265-9-3 | 3 | 99.53 | 2.35 | 7 |

leaching solution is reacting with both PuO₂ particles and glass present in the filter media. When a small amount of leachant is added to a relatively large amount of contaminated filter media, the fluoride ion concentration of the solution is quickly depleted as SiF₄ gas is produced. The remaining HF is not sufficient to dissolve 90% of the PuO₂ present, as desired. Conversely, if a large amount of leachant is added to the same amount of contaminated filter media, there is sufficient HF present to react with the glass and dissolve 90% (or greater) of the PuO₂. What is needed, therefore, is a larger amount of HF in the first stage of the process. This could be accomplished using a larger volume of 12N HNO₃-0.05N HF-0.01N H₂SO₄ or a higher concentration of HF in the acid mixture. For the subsequent stages, an HF concentration of 0.05N should be sufficient since most of the glass has reacted.

Dissolution of PuO₂* in 4N HNO₃-0.15M (NH₄)₂Ce(NO₃)₆

A contaminated filter media sample was prepared by mixing 0.16 g of plutonium dioxide with 3 g of shredded filter media. This sample was placed into a glass beaker containing 250 ml of 4N HNO₃-0.15M ceric ammonium nitrate. The beaker and contents were heated to boiling. Samples were withdrawn periodically and analyzed for plutonium concentration. The solution volume and concentration were kept constant by adding acid of proper concentration in order to replace evaporated acid. Figure 13 shows the effect of Ce⁺⁴ concentration on plutonium dioxide dissolution rate. The 4N HNO₃-0.10M (NH₄)₂Ce(NO₃)₆

*The PuO₂ was a fine powder, having a composition of 80 wt% plutonium-238, 16% plutonium-239, 2.5% plutonium-240, 0.8% plutonium-241, 0.2% plutonium-242, plus small amounts of other actinides, calcined at 950°C.

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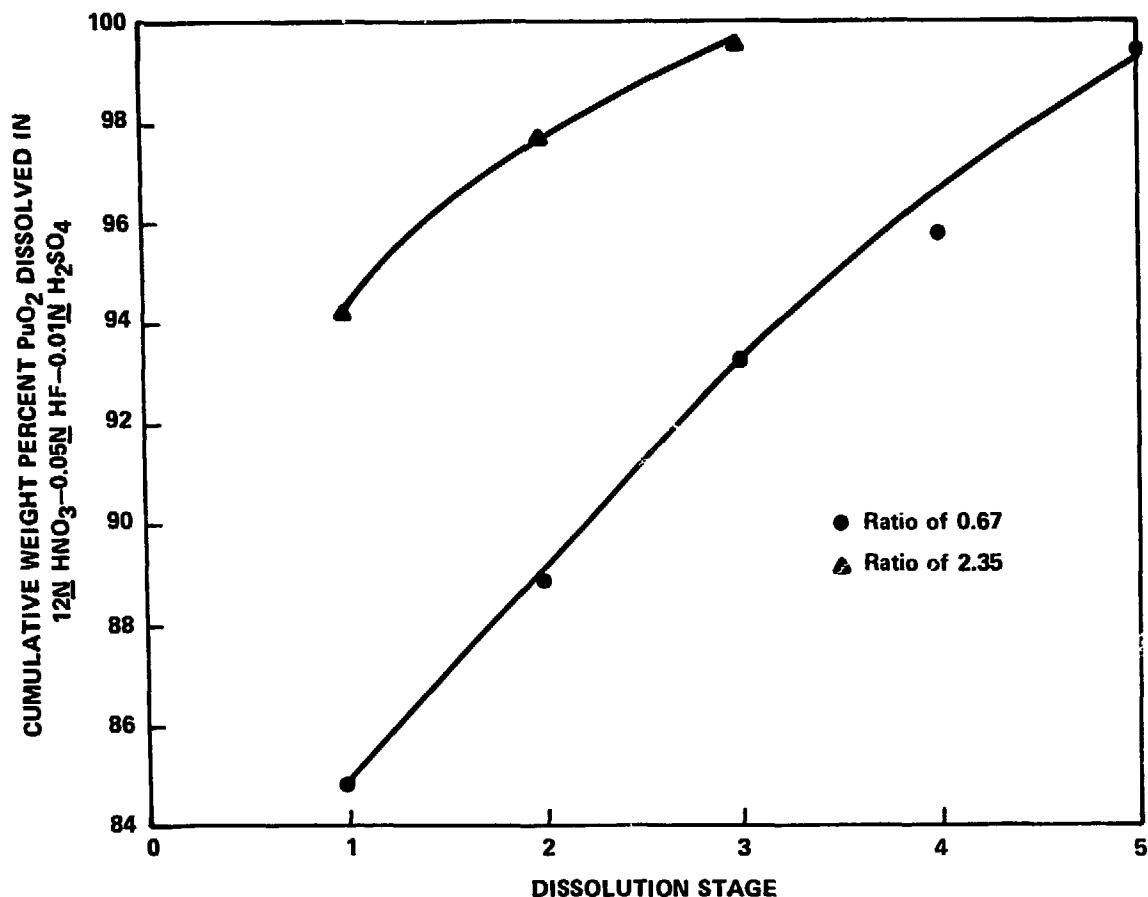


Figure 12 - Effect of acid volume:PuO₂ weight ratio on dissolution percentage.

curve is drawn from data contained in Figure 4. It should be noted that a 50% increase in the Ce⁴⁺ concentration results in a 10% increase in weight percent plutonium dissolved.

Americium-241 and Plutonium-239* Mixed Oxide Dissolution Studies

Contaminated filter media was prepared by mixing AmO₂-PuO₂ powder with shredded filter media. Small samples of this mixture were then treated with various leaching agents, including 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, 12N HNO₃-0.05N HF-0.01N H₂SO₄, and 8N HNO₃. A sample of filter media was placed in a beaker, 250 ml of the leaching agent was added, and the temperature was increased to the boiling point of the solution. Samples were withdrawn periodically and filtered through a 4-5 μm glass filter. The americium-241 and plutonium-239 concentrations were determined by alpha spectrometry. The solution volume and concentration were kept constant by adding acid of the proper concentration to replace evaporated acid. As can be seen in Table VII, the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ was an effective leaching agent, dissolving

* This material was approximately three parts plutonium-239 oxide mixed with one part americium-241 oxide by weight. It had been fired for 2 hr at 1000°C.

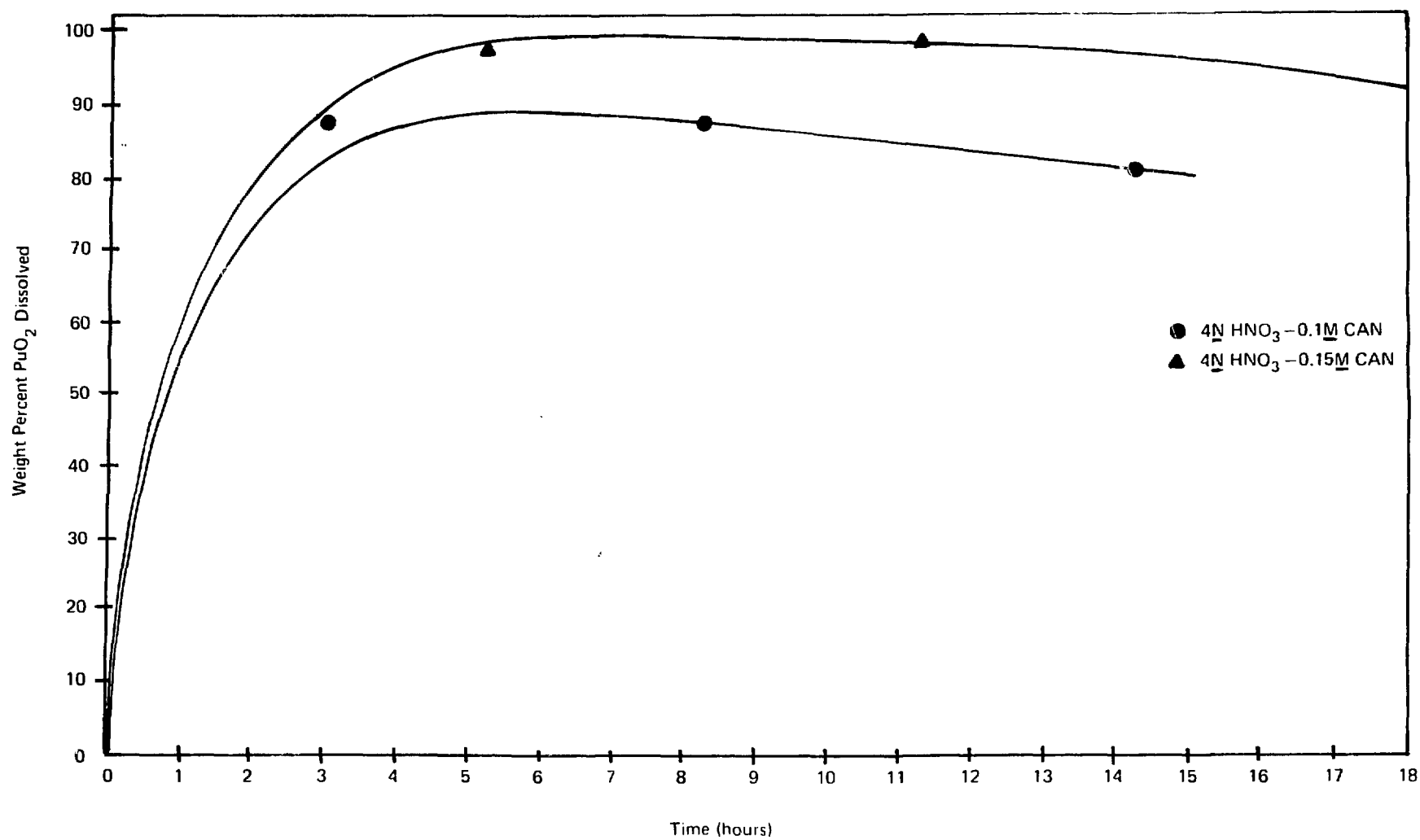


Figure 13 - Effect of ceric ammonium nitrate concentration on PuO_2 dissolution rate.

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Table VII - AmO₂-PuO₂ DISSOLUTION TESTS

| Acid Test Number | Reagent Composition | Time Heated (hr) | PuO ₂ Dissolved (wt%) | AmO ₂ Dissolved (wt%) |
|------------------|---|------------------|----------------------------------|----------------------------------|
| Am-Pu-1A | 4N HNO ₃ -0.1M CAN ^a | 5 3/4 | 72.9 | 63.2 |
| Am-Pu-1B | 4N HNO ₃ -0.1M CAN ^a | 11 3/4 | 93.7 | 111.8 |
| Am-Pu-2A | 12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄ | 5 3/4 | 68.9 | 72.4 |
| Am-Pu-2B | " | 11 1/4 | 80.2 | 90.4 |
| Am-Pu-4A | 8N HNO ₃ | 7 1/4 | 37.2 | 88.9 |
| Am-Pu-4B | " | 13 1/2 | 36.3 | 96.7 |

^aCAN is an acronym for ceric ammonium nitrate.

greater than 90% of both AmO₂ and PuO₂ in 11-3/4 hr. On the other hand, the 8N HNO₃ was not a satisfactory leaching agent since only 36% of the PuO₂ dissolved in 13½ hr, although 97% of the AmO₂ dissolved during this time (see Figure 14). The mixture of HNO₃-HF-H₂SO₄ was effective in dissolving the AmO₂ (90% in 11-1/4 hr) but not as effective in dissolving the PuO₂ (80% in 11-1/4 hr). Of the three reagents tested, the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ is the recommended leaching agent for mixtures of AmO₂-PuO₂.

Uranium-233 Oxide Dissolution Studies

These studies were conducted using U₃O₈ mixed with filter media. The oxide had been fired at 950°C for 2 hr. A small sample of the prepared filter media (approximately 3.2 g) was placed into a beaker containing 250 ml of boiling reagent. The reagents used were 8N HNO₃, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 12N HNO₃-0.05N HF-0.01N H₂SO₄. Samples were withdrawn periodically for alpha analysis. All samples were filtered through 4-5 µm glass filters. The solution volume and concentration were kept constant by adding acid of the proper concentration to replace evaporated acid. As can be seen from Table VIII, in all three cases the dissolution was very rapid and exceeded 90%. For example, the dissolution rate in 8N HNO₃ is depicted in Figure 16. All three reagents would be acceptable dissolution reagents for uranium-233 oxide.

Sodium Carbonate Fusion of PuO₂*

These studies were conducted in order to determine the effect of temperature on fusion efficiency. Small samples were prepared by mixing 64 mg of PuO₂ with 2.25 g of Na₂CO₃. The PuO₂-Na₂CO₃ mixture was then placed into a platinum crucible and contents were then heated to the desired operating temperature (1200°C and 1300°C) and maintained

*The PuO₂ was a fine powder having a composition of 80 wt% plutonium-238, 16 wt% plutonium-239, 2.5 wt% plutonium-240, 0.8 wt% plutonium-241, 0.2 wt% plutonium-242, plus small amounts of other actinides. It was calcined for 2 hr at 950°C.

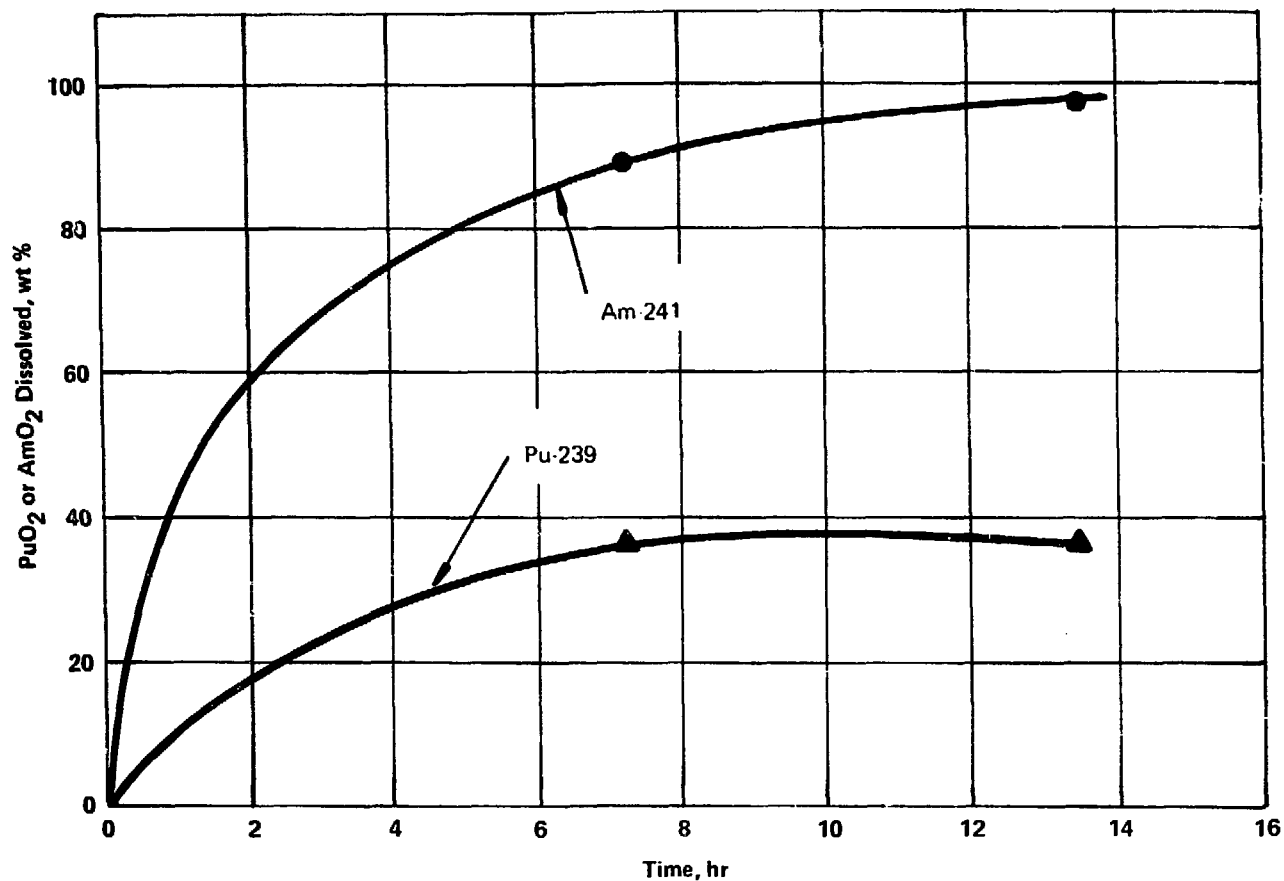


Figure 14 - Dissolution rate of $\text{Am}^{241}\text{-Pu}^{239}$ oxides in 8N HNO_3 .

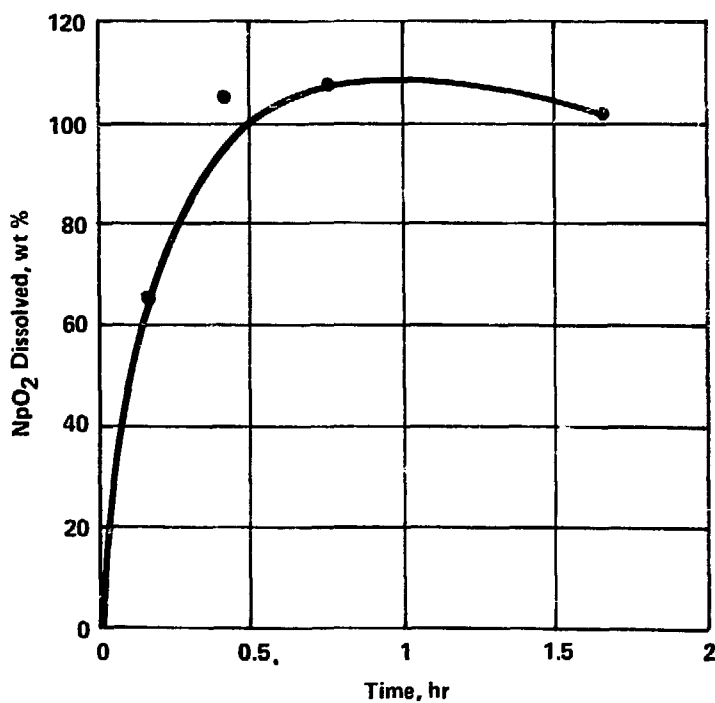


Figure 15 - Dissolution rate of U_3O_8 in $8\text{N HNO}_3\text{-}0.1\text{N HF}$

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Table VIII- U-233 OXIDE DISSOLUTION TESTS

| Acid Test Number | Reagent Composition | Time Heated (min) | U-233 Oxide Dissolved (wt%) |
|------------------|---|-------------------|-----------------------------|
| U-3-1 | 8N HNO ₃ | 5 | 97.4 |
| U-3-2 | " | 20 | 106.1 |
| U-3-3 | " | 65 | 93.8 |
| U-3-4 | " | 255 | 95.9 |
| U-4-1 | 4N HNO ₃ -0.1M CAN ^a | 5 | 89.7 |
| U-4-2 | " | 15 | 104.8 |
| U-4-3 | " | 60 | 95.2 |
| U-4-4 | " | 360 | 95.7 |
| U-5-1 | 12N HNO ₃ -0.05N HF-0.01N H ₂ SO ₄ | 5 | 100.9 |
| U-5-2 | " | 15 | 106.2 |
| U-5-3 | " | 50 | 100.1 |
| U-5-4 | " | 240 | 102.5 |

^aCAN is an acronym for ceric ammonium nitrate.

there for 1 hr. The crucible and melt were cooled to room temperature, and the melt was removed from the crucible and dissolved in 4N HNO₃. As can be seen in Table IX, there is not a significant difference (4.6%) in the two dissolution percentages and it is probable that the difference is due to experimental error. Therefore, it can be concluded that within the temperature range under investigation, the fusion efficiency is approximately 58% (average value).

Neptunium-237 Oxide Dissolution Studies

The NpO₂ was fired for 2 hr at 950°C. Samples of contaminated filter media were prepared by mixing 0.2 g of NpO₂ with 3 g of filter media. The procedure was identical to that used in the uranium-233 oxide dissolution studies. The following three leaching reagents were tested: 8N HNO₃, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 12N HNO₃-0.05N HF-0.01N H₂SO₄. Table X lists the results from these experiments. As can be seen, the 8N HNO₃ was unsuccessful in dissolving the NpO₂ rapidly with only 32% solubilized in 8 hr. On the other hand, each of the other two reagents dissolved the NpO₂ rapidly with greater than 90% dissolved after 1/2 hr of boiling (see Figure 15 for dissolution percentage as a function of time). Therefore, both the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and the 12N HNO₃-0.05N HF-0.01N H₂SO₄ acid mixtures would be recommended for dissolution of NpO₂ from filter media.

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Table IX - Na_2CO_3 FUSIONS OF PuO_2

| Fusion Test Number | Time Heated (hr) | Operating Temperature ($^{\circ}\text{C}$) | PuO_2 Solubilized (wt%) | Dissolution Reagent |
|--------------------|------------------|--|----------------------------------|---------------------|
| Pu-44 | 1 | 1300 $^{\circ}\text{C}$ | 56.4 | 4N HNO_3 |
| Pu-45 | 1 | 1200 $^{\circ}\text{C}$ | 61.0 | 4N HNO_3 |

Table X - Np-237 OXIDE DISSOLUTION TESTS

| Acid Test Number | Reagent Composition | Time Heated (min) | NpO_2 Dissolved (wt%) |
|------------------|--|-------------------|--------------------------------|
| Np-3-1 | 8N HNO_3 | 10 | 1.3 |
| Np-3-2 | " | 25 | 1.9 |
| Np-3-3 | " | 45 | 4.1 |
| Np-3-4 | " | 75 | 6.1 |
| Np-3-5 | " | 480 | 32.3 |
| Np-4-1 | 4N HNO_3 -0.1M CAN ^a | 10 | 93.7 |
| Np-4-2 | " | 25 | 97.2 |
| Np-4-3 | " | 40 | 88.9 |
| Np-4-4 | " | 60 | 89.1 |
| Np-5-1 | 12N HNO_3 -0.05N HF-0.01N H_2SO_4 | 10 | 64.7 |
| Np-5-2 | " | 25 | 105.2 |
| Np-5-3 | " | 45 | 107.4 |
| Np-5-4 | " | 100 | 102.3 |

^aCAN is an acronym for ceric ammonium nitrate.

Curium-244* Oxide Dissolution Studies

Contaminated filter media were prepared by mixing 13 mg of CmO_2 with 3 g of filter media. The experimental procedures were identical to those used for the uranium-233 dissolution studies. The two leaching reagents tested were 12N HNO_3 -0.05N HF and 4N HNO_3 -0.1M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. The results of these tests, listed in Table XI, show that both reagents dissolved the CmO_2 very rapidly (essentially 100% dissolution in 10 min). Figure 17 shows the dissolution rate for 4N HNO_3 -0.1M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. Both reagents are recommended for dissolution of CmO_2 from contaminated filter media.

*This was a powder which was approximately 94% curium-244 and 4% curium-246, the remainder being curium-245, -247, and -248.

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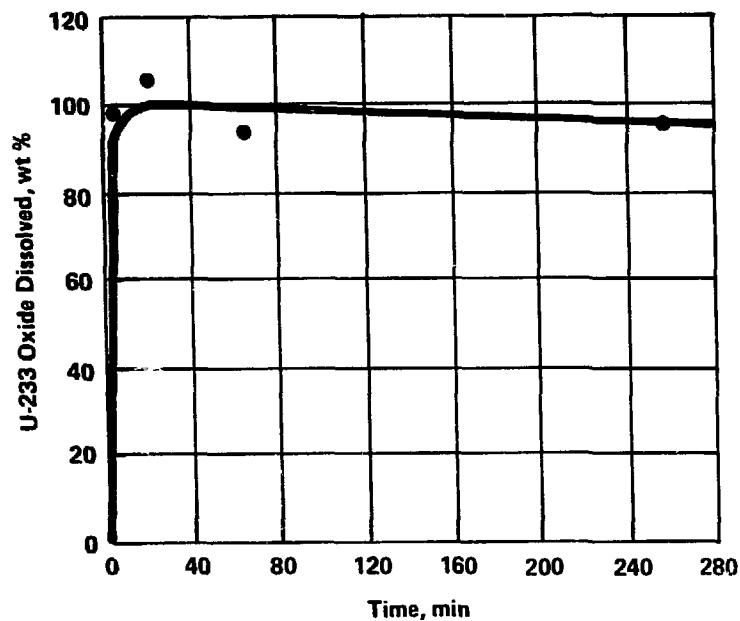


Figure 16- Dissolution rate of U-233 oxide in 8N HNO₃.

Table XI - Cm-244 OXIDE DISSOLUTION TESTS

| Acid Test Number | Reagent Composition | Time Heated (min) | CmO ₂ Dissolved (wt%) |
|------------------|--|-------------------|----------------------------------|
| Cm-2A | 12N HNO ₃ -0.05N HF | 10 | 103.3 |
| Cm-2B | " | 25 | 100.8 |
| Cm-2C | " | 60 | 105.0 |
| Cm-2D | " | 120 | 99.1 |
| Cm-2E | " | 180 | 104.5 |
| Cm-2F | " | 540 | 93.3 |
| Cm-2G | " | 540 | 99.0 |
| Cm-3A | 4N HNO ₃ -0.1M CAN ^a | 10 | 112.1 |
| Cm-3B | " | 25 | 110.0 |
| Cm-3C | " | 60 | 109.0 |
| Cm-3D | " | 120 | 107.6 |
| Cm-3E | " | 330 | 110.5 |

^aCAN is an acronym for ceric ammonium nitrate.

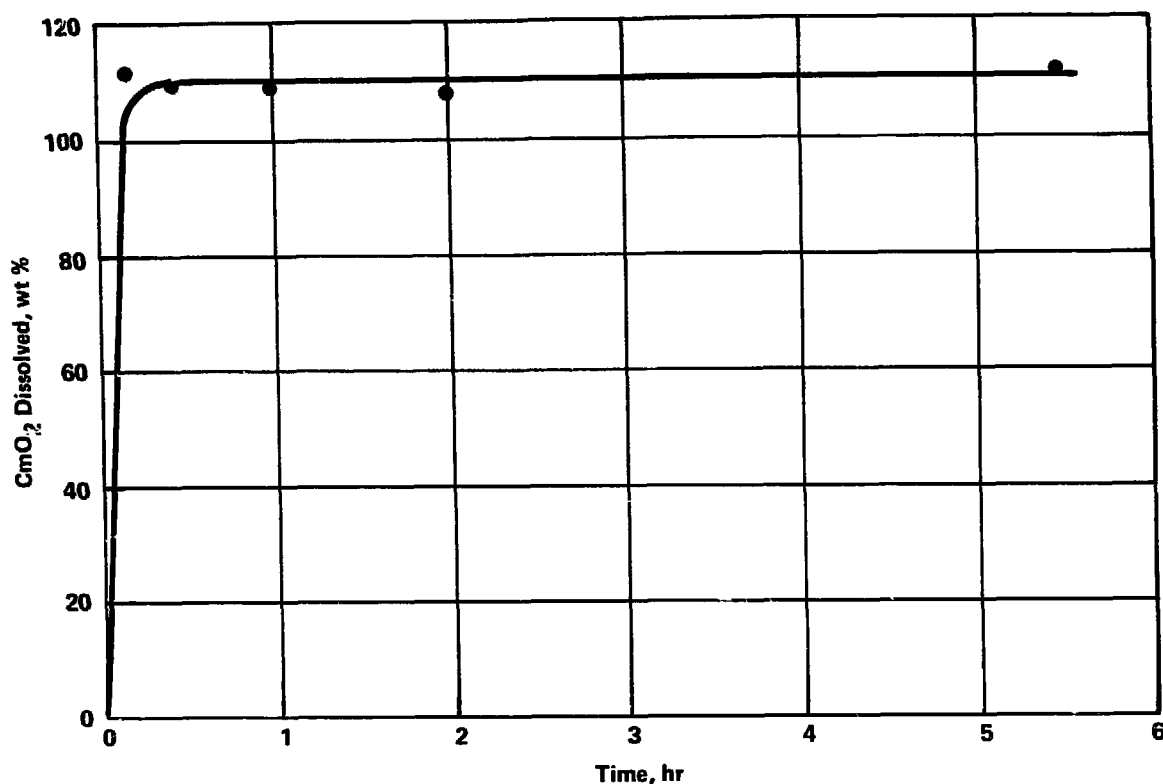


Figure 17 - Dissolution rate of CmO_2 in 4N HNO_3 - $0.1\text{M (NH}_4)_2\text{Ce(NO}_3)_6$.

Thorium-232 Oxide Dissolution Tests

Contaminated filter media were prepared by mixing 0.24 g of ThO_2 with 3 g of filter media. The experimental procedure was identical to that used in the uranium-233 oxide studies. The ThO_2 was a fine powder which had been fired at 600°C . The purpose of the experiment was to determine whether ThO_2 would dissolve in 4N HNO_3 - $0.1\text{M (NH}_4)_2\text{Ce(NO}_3)_6$ similarly to other actinide oxides previously tested (PuO_2 , CmO_2 , AmO_2 , U_3O_8 , and NpO_2). As shown by the data in Table XII and Figure 18, the dissolution rate was slow at first (0-7 hr), rapid from 7 to 9 hr, and slow again from 9 to 20 hr. The reason for this is not known at this time, but it is thought that it took 7 hr to attack the surface of the ThO_2 and open the pores so that rapid dissolution could begin. It therefore appears that 4N HNO_3 - $0.1\text{M (NH}_4)_2\text{Ce(NO}_3)_6$ would be an acceptable leaching agent for ThO_2 from filter media, although dissolution time is longer than for other actinides.

A point of interest is that in a similar experiment using just ThO_2 (no filter media), the dissolution percentage obtained was only 25% after 20 hr of boiling. The reason for the difference in dissolution percentages (25% versus 100%) is that the filter media contains 0.14 wt% F^- . This small amount of fluoride ion has been found to be sufficient to catalyze the dissolution reaction.

Dissolution of PuO_2 from Actual Glovebox Filters

These tests were conducted using samples of filter media removed from actual spent glovebox filters. The PuO_2 on the filters was the same type as that used in the fusion experiments.

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Table XII - ThO₂ DISSOLUTION DATA

| Acid Test Number | Reagent Composition | Time Heated (min) | ThO ₂ Dissolved (wt%) |
|------------------|---------------------------------|-------------------|----------------------------------|
| Th-1 | 4N HNO ₃ -0.1M CAN * | 5 | 0.6 |
| Th-2 | " | 10 | 0.4 |
| Th-3 | " | 20 | 1.4 |
| Th-4 | " | 35 | 2.4 |
| Th-5 | " | 60 | 5.1 |
| Th-6 | " | 180 | 4.7 |
| Th-7 | " | 300 | 12.5 |
| Th-8 | " | 440 | 31.7 |
| Th-9 | " | 440 | 29.3 |
| Th-10 | " | 470 | 42.1 |
| Th-11 | " | 545 | 82.3 |
| Th-12 | " | 905 | 100.5 |
| Th-13 | " | 1205 | 104.3 |

*CAN is an acronym for ceric ammonium nitrate.

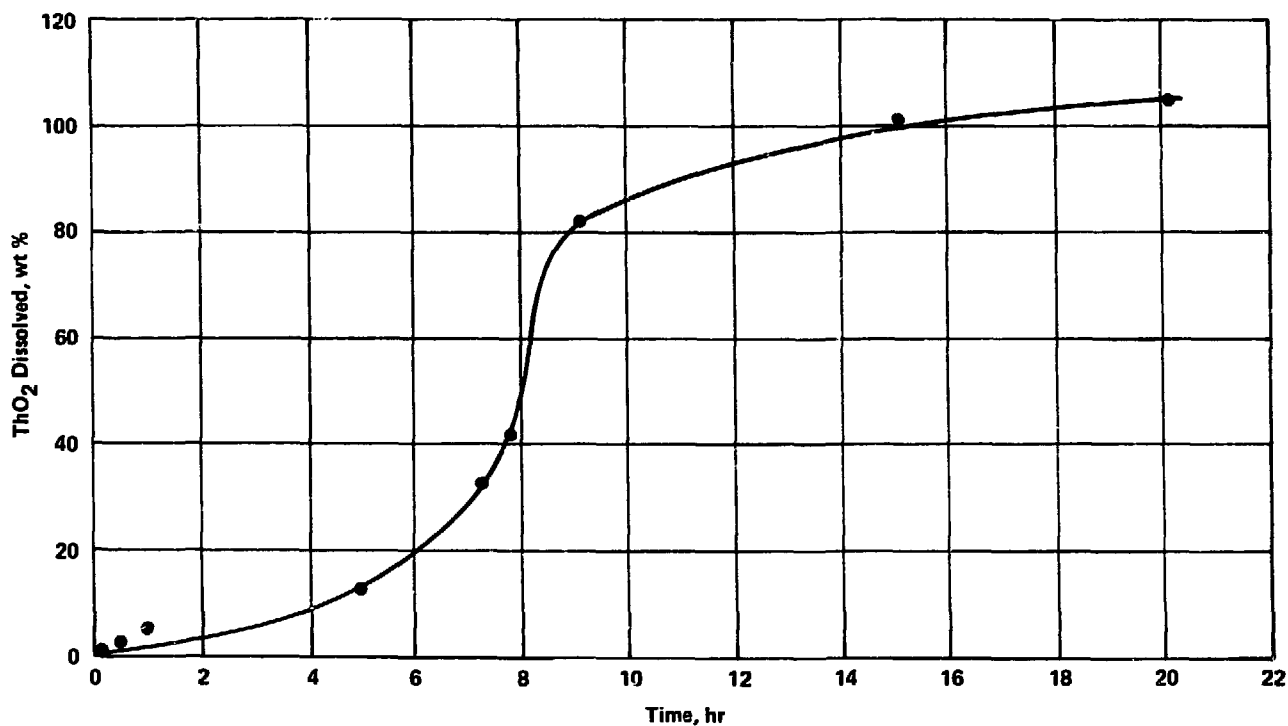


Figure 18 - Wt % ThO₂ dissolved in 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆.

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Experiment A

A 9.5 g sample of filter media containing 0.18 g of PuO_2 was dissolved in a six-stage dissolution procedure. The leaching reagent in the first five stages was 170 ml of 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$, and the sixth stage contained 12N HNO_3 -0.1N HF. The 12N HNO_3 -0.1N HF was used because the 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ had proved ineffective in the fifth stage of dissolution. The volume of 12N HNO_3 -0.1N HF used was 250 ml and all studies were done at boiling temperature. The solution was sampled periodically for plutonium concentration, and all samples were filtered through 4-5 μm glass filters. When the dissolution rate for a particular stage had become very slow (as indicated by α analysis of solution), the filter media were separated from the leaching solution by filtration and placed in the next stage of dissolution.

Results for this experiment are given in Table XIII. Figures 19 through 21 show the dissolution rate for each stage, and Figure 22 shows the cumulative PuO_2 dissolution percentage for the six-stage process. It should be noted that final dissolution percentage was 99.88% (a contamination factor of 833). This was determined by gamma ray spectrometry of the residual filter media and agrees well with the final value as determined by alpha counting of the solution samples (99.80%). It should also be noted that the 12N HNO_3 -0.1N HF solution used in the sixth stage was more than twice as effective as the 4N HNO_3 -0.23 M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ used in the fifth stage (0.079% dissolved versus 0.031%). The reason for this is not understood at this time. More studies will be done to investigate this phenomenon.

Experiment B

A 25.6 g sample of filter media containing 0.31 g of PuO_2 was dissolved in a four-stage dissolution process. The leaching reagent used in the test was 460 ml of 4N HNO_3 -0.3M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. The experimental procedure was identical to that used in Experiment A of this section. The results for this experiment are given in Table XIV; Figure 23 shows the cumulative PuO_2 dissolution percentage for the four stage process. The dissolution percentages in Table XIV were determined by alpha counting, but the cumulative percentages used in Figure 23 were based on a final gamma spectrometry value of 98.24%. This latter value compares well with the value of 97.62% obtained by alpha counting. The gamma value is, however, considered more accurate than the total obtained by simple addition of the individual alpha count values. The decontamination factor obtained using this gamma value is 88.5. The decontamination factor obtained after four stages in Experiment A of this section was 417.

A logical question would be: Why was the decontamination factor less when more ceric ammonium nitrate per mole of PuO_2 was used? It is known that the 25.6 g sample filter media contained a greater percentage of pack-to-frame sealant* than the 9.5-g sample. It is also

*This is the material that is used to seal the asbestos-glass filter pack to the wooden frame. It is a fire retardant polyurethane foam and a rubber base adhesive.

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Table XIII- PuO₂ DISSOLUTION DATA (9.5-g sample)

| Acid Test Number | Reagent Composition | Time Heated (min) | PuO ₂ Dissolved (wt%) | Process Stage |
|------------------|---|-------------------|----------------------------------|---------------|
| 256-1 A | 4N HNO ₃ -0.23M CAN ^a | 10 | 82.0 | 1 |
| 256-2 A | " | 20 | 79.5 | 1 |
| 256-3 A | " | 40 | 87.3 | 1 |
| 256-4 A | " | 60 | 80.9 | 1 |
| 256-5 A | " | 120 | 85.8 | 1 |
| 256-6 A | " | 210 | 90.6 | 1 |
| 256-1 B | " | 10 | 0.41 | 2 |
| 256-2 B | " | 20 | 1.17 | 2 |
| 256-3 B | " | 45 | 2.50 | 2 |
| 256-4 B | " | 65 | 2.97 | 2 |
| 256-5 B | " | 120 | 4.75 | 2 |
| 256-6 B | " | 360 | 7.30 | 2 |
| 256-7 B | " | 510 | 7.47 | 2 |
| 256-1 C | " | 30 | 0.29 | 3 |
| 256-2 C | " | 75 | 0.55 | 3 |
| 256-3 C | " | 135 | 0.87 | 3 |
| 256-4 C | " | 315 | 1.06 | 3 |
| 256-5 C | " | 435 | 1.09 | 3 |
| 256-6 C | " | 675 | 1.31 | 3 |
| 256-7 C | " | 675 | 1.39 | 3 |
| 256-1 D | " | 20 | 0.029 | 4 |
| 256-2 D | " | 45 | 0.063 | 4 |
| 256-3 D | " | 60 | 0.079 | 4 |
| 256-4 D | " | 135 | 0.139 | 4 |
| 256-5 D | " | 330 | 0.213 | 4 |
| 256-6 D | " | 645 | 0.222 | 4 |
| 256-7 D | " | 645 | 0.228 | 4 |
| 256-1 E | " | 15 | 0.011 | 5 |
| 256-2 E | " | 30 | 0.012 | 5 |
| 256-3 E | " | 60 | 0.015 | 5 |
| 256-4 E | " | 135 | 0.023 | 5 |
| 256-5 E | " | 195 | 0.025 | 5 |
| 256-6 E | " | 465 | 0.029 | 5 |
| 256-7 E | " | 600 | 0.031 | 5 |
| 256-8 E | " | 780 | 0.032 | 5 |
| 256-9 E | " | 780 | 0.025 | 5 |
| 256-1 F | 12N HNO ₃ -0.1N HF | 30 | 0.016 | 6 |
| 256-2 F | " | 60 | 0.030 | 6 |
| 256-3 F | " | 240 | 0.047 | 6 |
| 256-4 F | " | 585 | 0.079 | 6 |
| 256-5 F | " | 945 | 0.075 | 6 |

^aCAN is an acronym for (NH₄)₂Ce(NO₃)₆.

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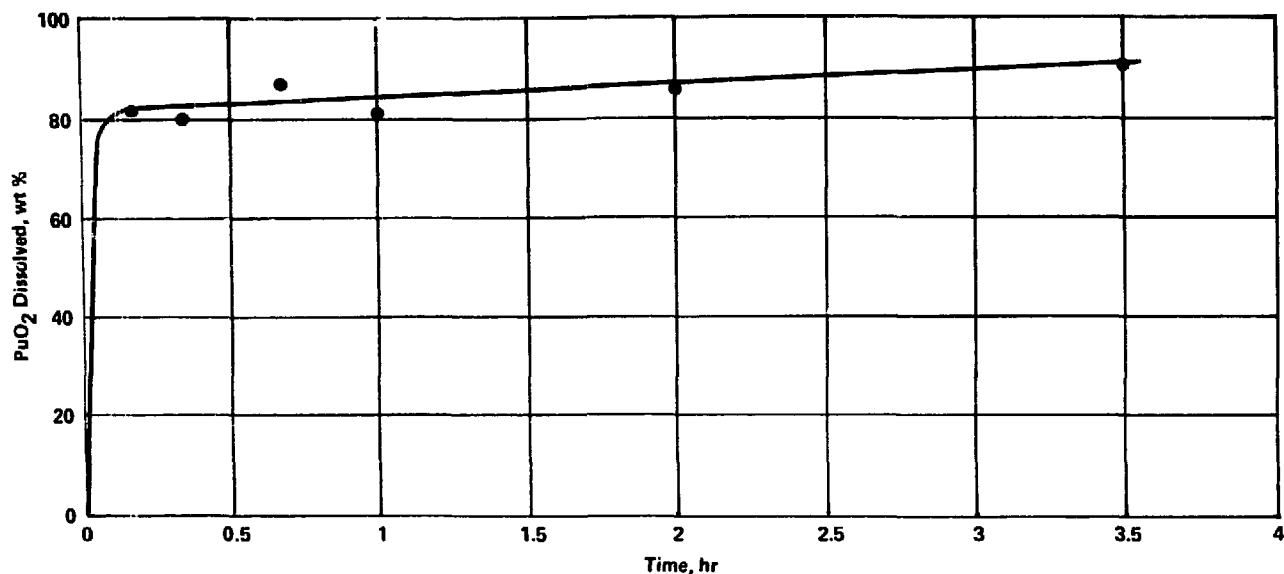


Figure 19 - Dissolution rate of PuO₂ from filter media (1st stage).

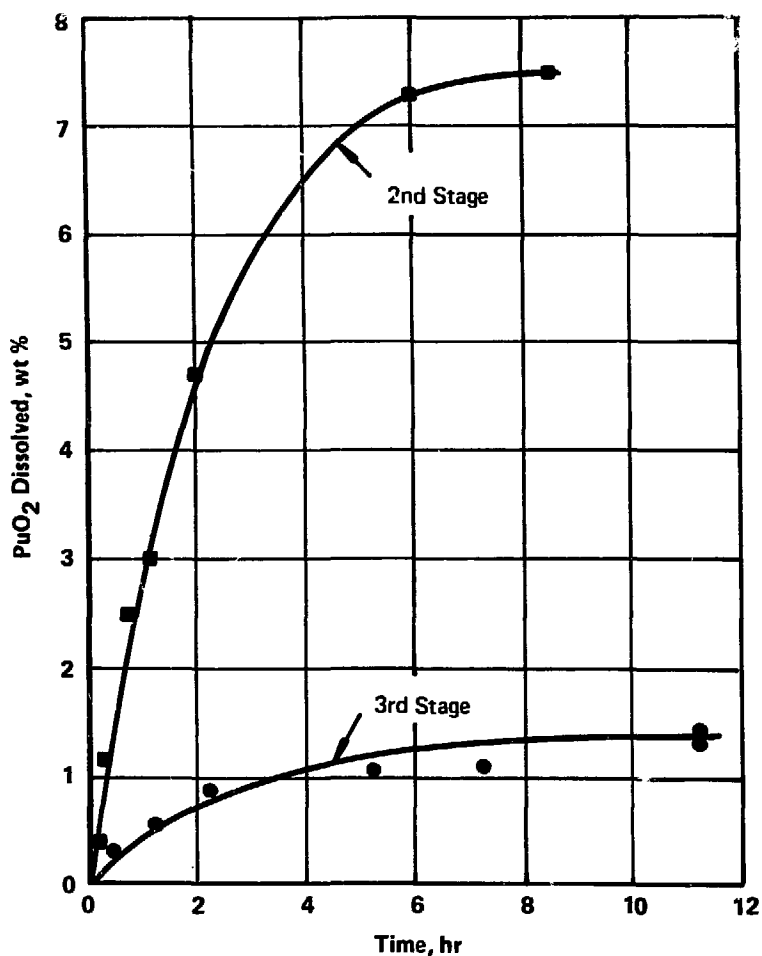


Figure 20 - Dissolution of PuO₂ from filter media (2nd and 3rd stage).

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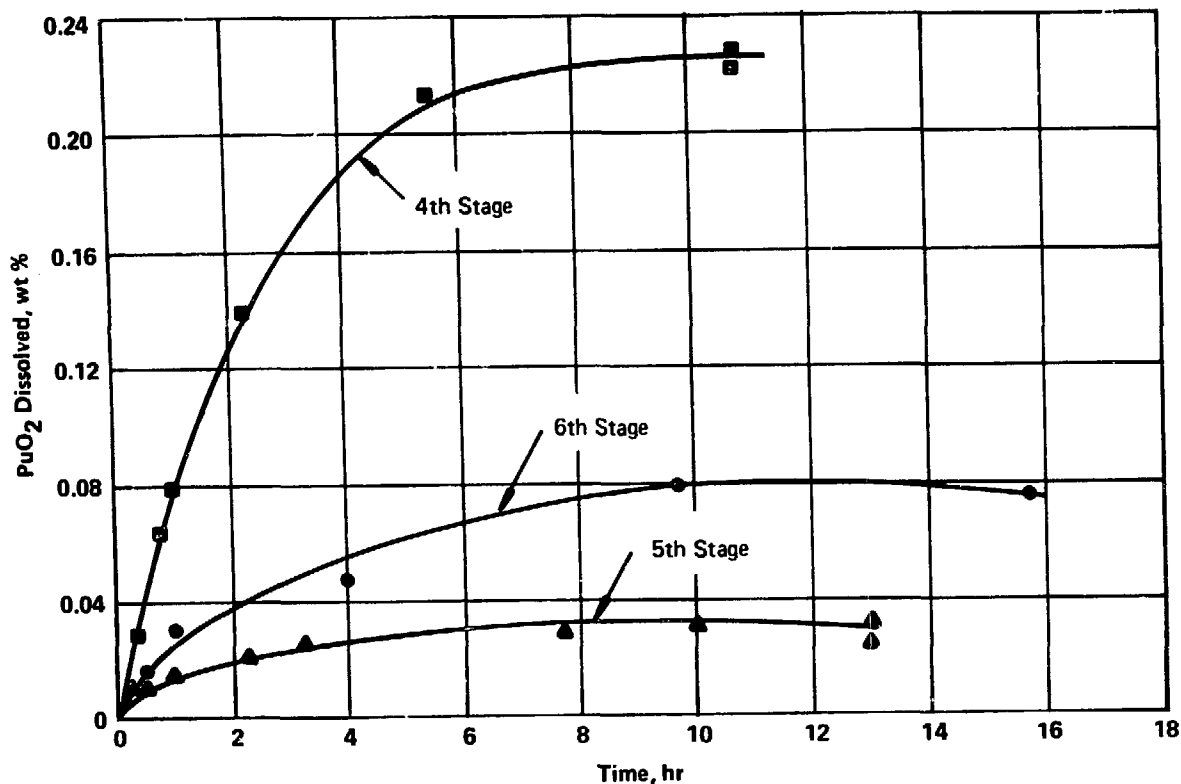


Figure 21 - Dissolution of PuO₂ from filter media (4th, 5th, and 6th stage).

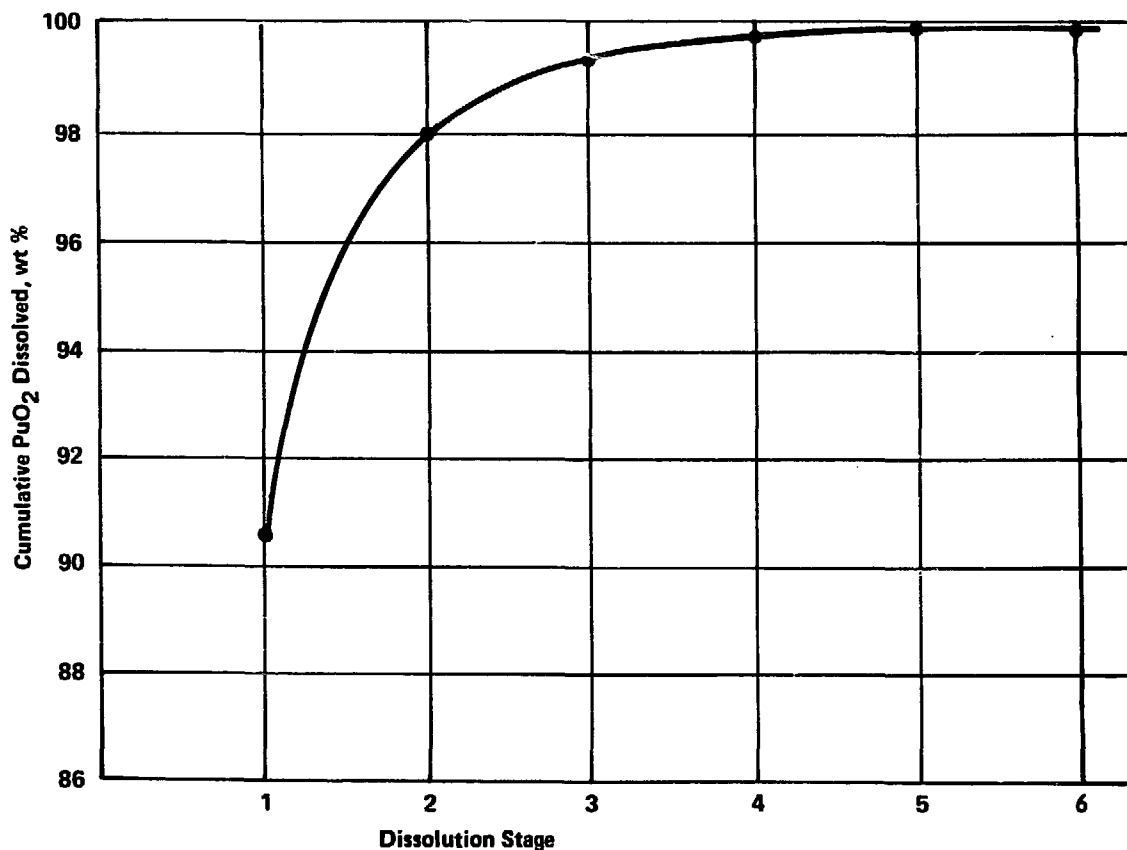


Figure 22 - Cumulative wt % PuO₂ dissolved during six-stage process.

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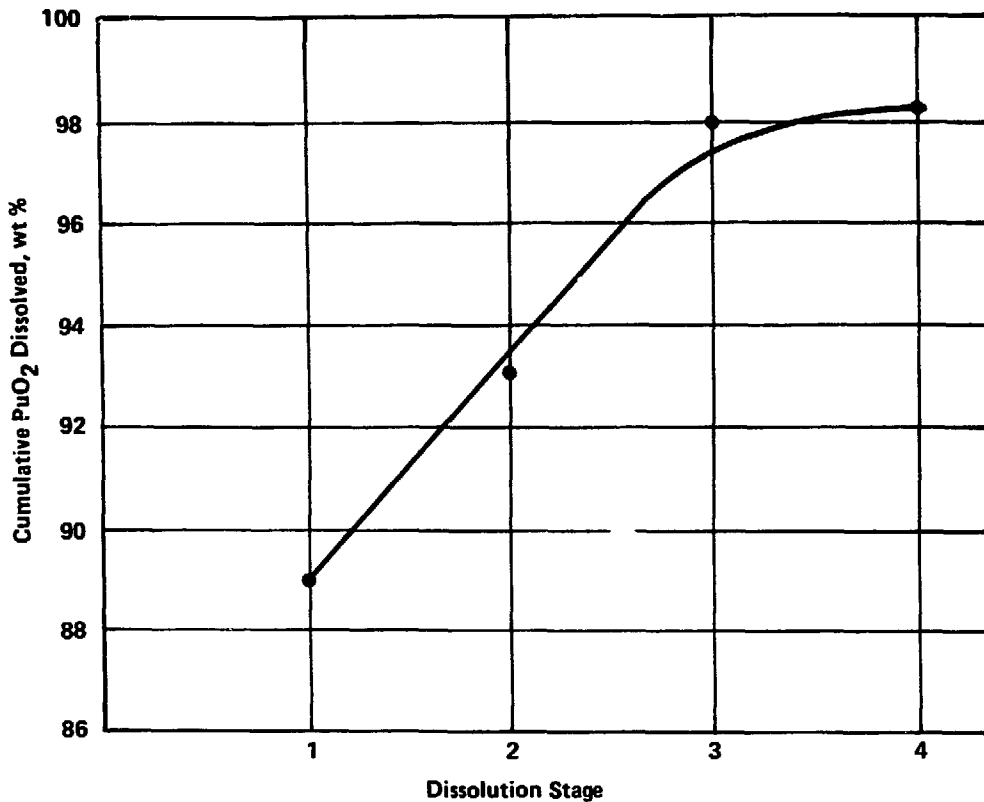


Figure 23 - Cumulative wt % PuO₂ dissolved during four-stage process.

Table XIV- PuO₂ DISSOLUTION DATA (25.6-g sample)

| Acid Test Number | Reagent Composition | Stage | Time Heated (hr) | PuO ₂ Dissolved (wt%) | Cumulative PuO ₂ Dissolved (wt%) |
|------------------|--|-------|------------------|----------------------------------|---|
| 12-1 | 4N HNO ₃ -0.3M CAN ^a | 1 | 6 1/2 | 88.40 | 88.40 |
| 12-2 | " | 2 | 15 | 4.04 | 92.44 |
| 12-3 | " | 3 | 7 1/4 | 4.91 | 97.35 |
| 12-4 | " | 4 | 7 | 0.27 | 97.62 ^b |

^aCAN is an acronym for ceric ammonium nitrate.

^bGamma ray analysis of remaining filter media indicated that 98.24% of the PuO₂ had been dissolved.

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known (from the color change of the leaching solution) that the Ce^{+4} ions were reduced much more quickly during the dissolution of PuO_2 from the 25.6-g filter media sample. Therefore, it is surmised that the reason for the smaller decontamination factor (smaller percentage dissolved) is that the Ce^{+4} ions were used mainly to oxidize the sealant thus leaving fewer Ce^{+4} ions to oxidize to Pu^{+4} . A simple solution to this problem might be to increase the amount of ceric ammonium nitrate when larger amounts of sealant are present with the filter media.

Summary

Plutonium adsorption isotherms were developed for the $Pu-HNO_3$ and $Pu-HNO_3-HF$ systems which proved that the filter media did indeed adsorb plutonium. The x/m ratio for the $Pu-HNO_3-HF$ system was less than that for the $Pu-HNO_3$ system at any given concentration. Thus, higher decontamination factors can be obtained with HNO_3-HF (assuming each leaching agent is equally effective in the dissolution of plutonium dioxide).

Dissolution tests were conducted on filter media contaminated with PuO_2 . The $HNO_3-H_2SO_4$ mixtures were relatively ineffective as leaching agents (see Figure 3). The best dissolution agents found were mixtures of HNO_3-HF and $HNO_3-HF-H_2SO_4$ in which the HF concentration was 0.05N or greater. These solutions dissolved greater than 90% of the PuO_2 in 15 hr (see Figures 5 and 6). Several tests were performed with $HNO_3-(NH_4)_2Ce(NO_3)_6$ solutions (see Figure 4). It was found that 4N $HNO_3-0.1M (NH_4)_2Ce(NO_3)_6$ was the best of these mixtures with 88% of the PuO_2 dissolved in 3 hr. Addition of $KMnO_4$ to 4N HNO_3 -cerium(III) dissolution mixtures was successful in oxidizing the cerium(III) to cerium(IV) and thus allowing more of the PuO_2 to dissolve.

Several dissolution tests were completed with UO_2 and U_3O_8 contaminated filter media. All leaching agents tested dissolved the uranium oxide rapidly (see Table II). Equally good results were obtained with 8N HNO_3 which dissolved ~100% of the UO_2 or U_3O_8 in 6 hr.

Neither Na_2CO_3 nor $Na_2CO_3-KNO_3$ was satisfactory as a fusion agent for PuO_2 . The greatest percentage of PuO_2 recovered was 71% using a Na_2CO_3-30 wt % KNO_3 salt mixture. Also there are tremendous corrosion problems associated with this process. The platinum crucibles and the furnace used in these experiments were corrosively attacked by the resultant vapors.

The $PuO_2-75\%$ UO_2 solid solution dissolved readily in 8N HNO_3 , 12N $HNO_3-0.1N HF$, 12N $HNO_3-0.05N HF-0.01N H_2SO_4$, and 4N $HNO_3-0.1M (NH_4)_2Ce(NO_3)_6$. These would be acceptable leaching agents since greater than 93% of the PuO_2 dissolved in 2 hr at boiling temperatures. Hueda (4) has reported that PuO_2-UO_2 can be dissolved in nitric acid alone (up to 35% PuO_2). Baehr and Dippel (5) reported that $UO_2-15\%$ PuO_2 fired at 1600°C will dissolve easily in 14M HNO_3 . These sources substantiate recent findings at Mound Facility.

Small and large samples of actual HEPA glovebox filters (contaminated with PuO_2) were treated with various leaching solutions. For small samples, 12N $HNO_3-0.05N HF-0.01N H_2SO_4$ proved superior to 4N $HNO_3-0.1M (NH_4)_2Ce(NO_3)_6$ with greater than 89% of the PuO_2 dissolved. For larger samples, it was found that an increase in the ratio of acid volume to PuO_2 weight increased the dissolution percentage. Figure 12

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compares a five-stage process (volume/weight ratio of 0.67) with a three-stage process having volume:weight ratio of 2.35. It should be noted that the latter process obtained a higher percentage of dissolution than the former.

Dissolution studies using 4N HNO₃-0.15M ceric ammonium nitrate dissolved greater than 93% of the initial PuO₂. Therefore, 4N HNO₃ in combination with (NH₄)₂Ce(NO₃)₆ is a recommended leaching reagent.

Filter media which were contaminated with americium-241 and plutonium-239 oxides were treated with three different leaching reagents to determine the effectiveness of each reagent in dissolving the actinide oxides. The three reagents used were 8N HNO₃, 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆, and 12N HNO₃-0.05N HF-0.01N H₂SO₄. It was found that the best leaching agent was the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ which dissolved greater than 90% of the AmO₂ and the PuO₂.

Another series of tests was conducted to determine the best leaching agent for uranium-233 oxide (U₃O₈). Simulated samples of contaminated filter media were prepared by mixing 0.2 g of U₃O₈ powder with 3 g of shredded filter media. The reagents used were the same three as were used for the AmO₂-PuO₂ studies. It was found that all three reagents were successful in dissolving greater than 90% of the U₃O₈ in a very short time (15 min).

Fusion studies were conducted using Na₂CO₃ and PuO₂. The purpose of these experiments was to determine the effect of temperature on fusion efficiency. It was found that there was not a significant difference (4.6%) in the PuO₂ solubilized within the temperature range under investigation (1200-1300°C).

Experiments were conducted using filter media contaminated with neptunium-237 oxide. The purpose of the tests was to determine which of the three reagents would be successful in dissolving the NpO₂. Results indicate that both the 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and the 12N HNO₃-0.05N HF-0.01N H₂SO₄ were successful in dissolving greater than 90% of the NpO₂ in 0.5 hr. The 8N HNO₃ on the other hand solubilized only 32% of the NpO₂ in 8 hr and therefore would not be recommended as a leaching agent for NpO₂.

In other series of experiments, filter media contaminated with curium-244 oxide were dissolved in leaching tests using both 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ and 12N HNO₃-0.05N HF. Both reagents dissolved essentially 100% of the CmO₂ in approximately 10 min and therefore would be recommended for dissolving CmO₂ from filter media.

Filter media contaminated with thorium-232 oxide were leached with a 4N HNO₃-0.1M (NH₄)₂Ce(NO₃)₆ solution in an attempt to dissolve the ThO₂. The rate of dissolution was slow at the beginning (0-7 hr), rapid from 7 to 9 hr, and slow again at the end (9-20 hr). Complete dissolution was achieved in 20 hr, considerably longer than for the other actinide oxides previously tested. A point of interest is that in a similar experiment using only 4N HNO₃-0.1M ceric ammonium nitrate and ThO₂ (no filter media), the amount dissolved was only 25% after 20 hr. The reason for this difference is that the F⁻ catalyst is present in the filter media. Therefore, it is concluded that although

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the dissolution time was much longer for the ThO_2 , it is still an acceptable leaching reagent for ThO_2 from filter media.

Several experiments were completed using PuO_2 contaminated filter media from spent glovebox filters. These tests were made in order to determine how much $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ is required for dissolution of PuO_2 from such filters. In one experiment, a 9.5-g sample of filter media was processed through a six-stage dissolution process. In the initial five stages, 4N HNO_3 -0.23M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ was used and in the sixth stage 12N HNO_3 -0.1N HF was used. The amount of $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ used in each stage was 60 times the initial moles of PuO_2 present in the filter media. The overall decontamination factor obtained for the six-stage process was 833. In a second experiment, a 25.6-g sample of filter media was processed through a four-stage dissolution procedure. The leaching solution was 4N HNO_3 -0.30M $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$. The amount of ceric ammonium nitrate used in each stage was 120 times the initial moles of PuO_2 present in the filter media. The overall decontamination factor achieved in the four-stage process was 55.8.

The amount of ceric ammonium nitrate required cannot be based exclusively upon the amount of actinide present in the filter media. The reason for this is that other reactants such as pack-to-frame sealant* and other organic materials are present in the spent filter media. A high percentage of sealant was present in the 25.6-g sample and this is perhaps why less plutonium was dissolved. In other words, even though more ceric ammonium nitrate was used per mole of plutonium present, a lower percentage of the plutonium dissolved because a large percentage of the Ce^{+4} ions was depleted by reaction with the organics present. A formula for calculating the $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$ required per gram of filter media has not been developed. More studies must be done before the relationship can be determined.

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*This is a fire retardant polyurethane and a rubber base adhesive.

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DISCUSSION

BURCHSTED: After hearing your presentation, I wonder if a more appropriate title would be, "Product Recovery from Used HEPA Filters." Certainly, such a title would make the subject matter more accessible after the Proceedings have been indexed in other publications.

LEWIS: Thank you for that suggestion.

GUEVARA: I second the suggestion made by Burchsted. ;If decontamination were the appropriate topic, we would have liked to have heard more about economics and the ultimate disposal that is intended for the waste.

LOO: Would you describe the condition of the filter medium after the leeching process is completed?

LEWIS: After leeching, the filter medium is a white powdery substance.

LOO: Is it still intact?

LEWIS: No, the individual pieces have disintegrated and it has become a fine white powder.

DORMAN: Thank you, Mr. Lewis. It must have been quite a monumental experimental task to get all those results.

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CLOSING REMARKS OF SESSION CHAIRMEN:

GILBERT: In our first paper, Mr. McCormack reviewed a number of filters with different design and different media, some with prefilters, some without. With sodium oxide and hydroxide aerosols, he found considerable plugging, up to 25 in.w.g. I might advise him that filter media are only tested up to 20 in.w.g. Along the same lines, Dr. Hinds told us about the use of packed beds with different packings to be used with sodium aerosols. They exhibited considerable removal efficiency except for particles in the 0.5-1.1 μm range. In his brief report of the Government-Industry Meeting on Filters, Media, and Media Testing, Mr. Anderson did not have enough time to tell you that Johns-Manville described their latest quality control methods for measuring glass fiber diameters. Accurate measurement of fiber diameters is very critical for making filter paper that will meet nuclear grade filter criteria. They described their new program for making air permeability measurements that can be related to fiber diameter but they will continue to use the older Williams Freeness test simultaneously until all the users are familiar with the newer designations. Pursuing a DOE objective of reducing the volume of solid waste from spent filters that must be placed in storage or long-term burial, Mr. Woodward from Rocky Flats described their efforts to extend filter life by filter redesign, particularly filters subjected to chemical applications. In this endeavor, they are collaborating with the Bergman project at the Lawrence Livermore Laboratory.

DORMAN: I would also like to comment on the paper by Dr. Bergman because in 1960 I was sent Havlicek's original paper (most of it was written in his own fair hand) to referee for submission to the Journal. I have seen only about two references to that paper in the eighteen years that have elapsed since then. I think there was something by George Fielding a year or so ago and there has been an odd passing reference, but it never seemed to have been followed up very seriously. On the point of electric type filters, I would like to say there is another filter which is now being made in Europe and Holland based on the electric principle. Sheets of plastic material are subjected to voltages and the sheet is then stretched and fibrillated. This procedure produces quite fine fibers that can be fabricated into filters for air conditioning service or for dust respirators. They are quite efficient and have low resistance. However, they suffer, like most electric type filters, from the effects of oil which either dissolves the outer layers of the fibers or forms a Faraday cage that stops the electric effects. In addition, electric type fibrous filters tend to decay in efficiency with increase in loading of dust. For instance, one of the reasons why we did not use DOP in England in the old days when resin-wool filters were in vogue was that oil wrecked the wool resin filter in about 3 or 4 minutes. DOP increase filter penetration from 0.001% to 5 or 10%. Now, we find that even sodium chloride degrades the efficiency of electrified media. This means that electric type fibrous filters are always used. The papers by Dr. First and by Dr. Rivers were concerned with saving filter volume and they are, in a way, complementary to the work which is being done on dismantling filters. My own experience of what is termed the "European design filter," based on the LUWA filter, is that it's a satisfactory filter when it is used against normal dusts. I have had no experience with it in the nuclear industry, nor do I know how it will stand up against much rough usage. But, I shall be very interested to keep in touch with the work at AAF and Harvard and to hear what happens. On the paper by John Collins, I think there are arguments both for and against what he proposes. And, I think I would be very wise, as a non-United States chairman, to keep out of your internal arguments because I shall get clobbered by one side or the other and I want to go home. The last paper, presented by Mr. Lewis, obviously represented a very comprehensive series of tests and I can only admire the detail that he has presented. Obviously, he and his collaborators have spent a long, long time at it and produced a very good paper.

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