HEPA FILTER PERFORMANCE COMPARATIVE STUDY

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Abstract

Current products such as HEPA filters made without separators, with tapered separators and with mini separators have raised many questions for the Nuclear Ventilation System Design Engineer and/or the end user. The principal objective of this investigation is to report HEPA filter performance data and to compare the effectiveness of the various type HEPA filters for use in Nuclear Ventilation Systems with all tests run on the same equipment and under the same controlled conditions.

I. Introduction

As nuclear plants become larger and more numerous, great emphasis is being placed on improved, high performance ventilating systems. Under either accident situations or during routine operation, efficient and dependable air cleaning systems are required if nuclear release limits are to be met. As a result of the California referendum, increasing public awareness, and low as practical release limits, it is essential today to have a complete knowledge of HEPA performance to design a safe and reliable system.

The ability of HEPA filters to remove radioactive contamination in the form of particulates has been investigated by others. However these past studies have been limited to either specific removal systems using standard HEPA filters or have been conducted on well controlled manufactured units. This investigation was conducted on production models in which the tests were all conducted under essentially the same conditions.

The Nuclear Ventilating System Design Engineer needs to know if the type of HEPA filter he is going to use will function after exposure to postulated conditions which could occur in his particular system. The filter can be exposed to steam-air atmospheres for extended periods of time during a design basis accident; to heavy entrained water loadings as a result of spray activation; to excessive overpressures resulting from local tornadoes or inadvertent damper closing; to excessive heat due to fires, and to a possible seismic event. Based on this background and the many questions from designers and end users about filter performance, comparative tests were conducted on various types of filters to determine:

1. HEPA Filter integrity after exposure to abnormal conditions
2. HEPA Filter service life
3. Particle size penetration

HEPA filters were obtained at random from various vendors. The types of filters tested are shown in Figures 1.A through 1.E and can be described as follows:

1.A The Type A Separatorless Filter is formed from glass filter medium molded and formed into a corrugated shape and then folded back and forth on itself so that it becomes self-supporting. No separators are used. Estimated medium area is about 250 square feet. Manufacturer's rating is 1500 CFM @ 1" W. G. resistance.

1.B The Type B Separatorless Filter contains sixteen individual filter panels arranged in a vee shaped pattern and sealed into an outer metal retaining frame. The individual filter panels consist of a sheet metal frame containing pleated filter medium. Pleat depth is about 3/4 of an inch. Folds are spaced and retained in position by glass thread attached to the media surfaces in accurately located lines normal to the pleats so that when folded only string contacts string. Estimated filter media area is about 320 square feet. Manufacturer's rating is 1765 CFM @ 1" W. G. resistance.

1.C The Type C Tapered Separator Filter is constructed by folding glass filter medium over tapered aluminum separators. The separator is tapered from the front face of the filter to the rear of the fold. Net result is a filter medium pleat which is vee shaped. Estimated filter media area is about 250 square feet. Manufacturer's rating is 1320 CFM @ 1" W. G. resistance.

1.D The Type D Mini Separator Filter rated 1500 CFM is constructed by folding glass filter medium over standard design corrugated aluminum separators whose height has been decreased to permit the use of about 300 square feet of filter medium. Manufacturer's rating is 1500 CFM @ 1.2" W. G. resistance.

1.E The Type E Standard Separator Filter is constructed by folding glass filter medium over standard design and standard height separators. Filter media area is about 240 to 250 square feet. Manufacturer's rating is 1000 CFM @ 1.0" W. G. resistance.

II. HEPA Filter Integrity After Exposure to Postulated Abnormal Conditions

Heavy Entrained Water Loading Tests

Test conditions were maintained as follows:

	Test Conditions	Test Requirements
1.	Temperature	$95^{o} \pm 5^{o}F$
2.	Relative Humidity	95% Minimum
3.	Rate of Air Borne Water Droplets Flowing Toward the Filter	1-1/4 pounds per minute per 1000 CFM of nominal rated filter capacity
4.	Pressure Differential across filter	10.0 ± .02 inch water gage

5.	Time to Reach Maximum Pressure Differential	1-1/2 minute maximum
6.	Air Flow	That required to produce 10.0 ± .02 inch water gage differential

Figure 2 is a drawing of the test apparatus and test results are summarized in Table I.* The test duct is approximately 24" wide x 26" high by 48" long. Air flow is once thru. The required air flow is provided by a high pressure blower. Steam is fed into the blower intake to maintain temperature and RH requirements. Water is fed into the system through spray nozzles located an average distance of approximately 18" from the face of the filter under test. As a result, there is direct impingement of water droplets on the HEPA filter, an arrangement which allows for the most vigorous conditions which could be encountered.

The data shows that for heavy entrained moisture loadings, HEPA filters made with separators are structurally capable of performing under these conditions. These results are in agreement with others that corrugated separators add strength to the filter core.(1) Conversely both type separatorless filters failed. Type B failure was marginal or questionable because the initial penetration was .03% or borderline at the start. However, Type A was a definite failure and the results were the same on subsequent tests.

Pictures of the exposed filters are shown in Figures 3A through 3E. Figures 3C, 3D, and 3E show the corrugated separator styles in which there was no visible damage evident. Figure 3B shows the Type B separatorless filter. The circled area is where pin holes developed during the test. Figure 3A shows the Type "A" separatorless filters with heavy structural damage.

Excessive Pressure Tests

These tests were conducted on filters which had first been dust loaded to 4" water gage resistance with ASHRAE test dust** per ASHRAE Standard 52-68. Tests were then conducted in the high pressure test duct shown in Figure 4.

Cottrell precipitate was fed into the system at the rate of 454 grams per minute while maintaining air flow through the filter of 1000 CFM or 1500 CFM. Increase of resistance was noted and an insitu cold generated DOP penetration test was conducted after each 454 grams of dust was fed.(2) Dust loading was discontinued when visible rupture took place at which time a final DOP penetration was measured. Test results are summarized in Table II.

* All DOP penetrations reported in the heavy entrained water loading tests were obtained on the Q107 Penetrometer (136-300-175A).

** ASHRAE Standard 52-68 test dust is a specially compounded dust which is 72% by weight Standardized Air Cleaner Test Dust Fine, 23% Molocco Black and 5% ground linters.

An analysis of the data shows that a HEPA filter made with standard configuration separators will give the most reliable performance at pressure drops exceeding 10" water gage. The Type D Mini Separator Filter was dust loaded at 1500 CFM to 30.25" water gage with no sign of visible rupture and with only a small increase of .003 percent in DOP penetration. The Type E Standard Separator Filter was dust loaded at 1000 CFM to 31.25 inches water gage resistance before visible rupture was noted. Final DOP penetration was 6.0 percent; however, the last DOP penetration prior to rupture was .001%. Filter resistance when this measurement was taken was 29 inches water gage. The Type A Separatorless Filter dust loaded at 1000 CFM showed visible rupture at 20" W.G. resistance. DOP penetration was 1.5%. The Type A Separatorless Filter dust loaded at 1500 CFM showed visible rupture at 13.25" W.G. and DOP penetration was 0.06%.

Steam-Air Exposure Tests

These tests were conducted on a separator type filter and a separatorless type filter. Tests were conducted in the MSA Environmental Test Facility described in MSAR 71-45 and shown in Figure 5. (3)

Tests were conducted per the following parameters:

1.	Steam Air Volume Flow	1000 CFM
2.	Steam Air Temperature	270°F
3.	Steam Air Pressure	47 PSIG

The Type D separator type filter was subjected to the above conditions for 24 hours. The filter had an initial resistance of 0.90 inches water gage and an initial DOP penetration* of .001%. After 24 hours exposure the filter had a DOP penetration of .017% and a resistance of .91 inches water gage when tested at 1000 CFM.

The Type A separatorless filter was exposed to the same steam air conditions for 12 hours when visible rupture was noted. The filter had an initial DOP penetration of .006 and an initial resistance of .72 inches water gage at 1000 CFM. After exposure, the filter had a DOP penetration of 24 percent.

III. HEPA Filter Service Life

Filter life was evaluated utilizing the following dust loading tests:

1. ASHRAE 52-68
2. Sodium Oxide Collection

Curves were obtained relating clean filter pressure drop to air flow volumes. All data was obtained on ASHRAE 52-68 test duct. Results are shown in Figure 6.

*All DOP penetrations reported in the Steam Air Exposure Tests were obtained on the Q107 Penetrometer (136-300-175A).

Pressure drops at 1000 CFM ranged from .42 inches water gage for the Type B Separatorless Filter to .80 inches water gage for the Type A Separatorless Filter. Pressure drops at 1500 CFM ranged from .70 inches water gage for the Type B Separatorless Filter to 1.20 inches water gage for the Type D Mini Separator Filter and the Type A Separatorless Filter.

ASHRAE 52-68 Dust Loading

ASHRAE 52-68 dust loading curves are shown in Figures 7 and 8. The ASHRAE 52-68 Test Duct is shown in Figure 9. Test air flow is measured using a calibrated, long radius flow nozzle. Test air is brought in from the outside into a large mixing plenum. Dust is fed into the entrance between the large mixing plenum and the main test duct. A dust mixing baffle coupled with high air velocity between the mixing plenum and the test duct insures uniform air dust concentration. This test duct is designed on sound aero dynamic principals which insures accurate air volume measurement and consequent accurate filter resistance measurements.

An analysis of the curves reveals that filter dust holding capacity is determined by initial pressure drop, filter media area and the design configuration of the air passages. Considering those tests conducted at 1500 CFM, the Type B Separatorless Filter had the largest filter media area and the lowest initial pressure drop. However, because of the design of the air passage, its dust holding capacity was essentially the same as the Type D Mini Separator Filter and the Type A Separatorless Filter, both of which had higher initial pressure drops and less filter media area but had a better air passage design.

Sodium Oxide Tests

These tests were conducted to compare dust loadings on finer particulate because this type particulate can be encountered in Nuclear Power Plant Ventilating Systems. Sodium Oxide particles range in size from 1.4 to 2.5 microns mass median diameter as compared to the 4.2 microns mass median diameter of the ASHRAE test dust.[4] The tests were conducted on full size filters in the test duct shown in Figure 10.

A weighed amount of sodium* was placed in a stainless steel pan where burning was initiated by an acetylene torch in the presence of excess air such that the air and all of the resulting smoke was sucked from the pan and discharged through the filter into the atmosphere. Air flow was measured with a standard pitot tube and controlled with a butterfly damper during the test.

Test air volume rate was 1500 CFM. Filters were weighed before and after the test. All filters were loaded to a final resistance of 6 inches water gage. Results are summarized in Table III. The

*Sodium in the brick form (as shipped condition) with no attempt made to remove the oxide layer.

results confirm that dust holding capacity depends on initial pressure drop, filter media area and the design configuration of the air passage.

IV. Particle Size Penetration

Particle size penetration measurements were conducted at filter media flow rates ranging from 3 FPM to 28 FPM using homogeneous 0.56, 0.40, 0.30 and 0.16 micron diameter DOP particles. No comparative tests were conducted on the various filter medium because virtually all filter paper is manufactured with glass fibers which have the same spectrum of fiber diameters and lengths. Consequently most filter medium should show the same general particle penetration characteristics.

For the most part, the aerosols of importance to the design engineer are particles <10μ in size and perhaps the major portion are in the sub-micron range. For example, Whitby et al reported that in general 99% by count of atmospheric air particles are ≤1μ with the majority in the sub-micron range.[5] In addition to naturally occurring atmospheric particles, particles in the .005 to 0.1μ size can be present in nuclear ventilating systems due to the oxidation of plutonium, sodium, uranium, zirconium, etc. Langmiur and others have shown the most difficult particles to capture are in the general range of 0.1 to 0.3μ in size.[6]

Test aerosols were formed by the method outlined in the Handbook of Aerosols, AEC, 1950.[7] Figure 11 shows the apparatus used. Condensation nuclei (NaCl) are formed in the ionizer and mixed with DOP vapor produced in the boiler. The vapor condenses on the nuclei forming a homogeneous, uniform DOP aerosol. Particle size is controlled by varying the ratio of the mass of the condensable vapor to the number of nuclei.

Particle diameters were determined using the angular distribution of color and light polarization methods outlined in the Handbook of Aerosols. Penetration measurements were made using a NRL linear read out forward scattering photometer.[8]

Test results are shown in Figure 12. One can observe from the family of curves that if the filter medium velocity exceeds the AACC CS-1 recommended velocity of 5 FPM, particle penetration increases significantly.[9] Exceeding the 5 FPM medium velocity decreases the effect of diffusion and reduces collection efficiencies on sub-micron particles as shown in Figure 12, and as also reported by others.[10]

V. Conclusions

As a result of this study it can be concluded that:

1. Separator type filters are stronger than the separatorless type when tested under the same conditions as outlined in this report.

2. Overall HEPA filter performance is significantly improved by maximizing the amount of filter medium with the use of smaller height separators.

3. Particle collection efficiency for glass fiber HEPA medium in the range tested drops significantly when the filter medium velocity exceeds 5 FPM.

Filter Type	Mfg's Rate Flow	*Initial DOP % Pen.	*Final DOP % Pen.	*Increase DOP % Pen.	Test Period
Type "A" Separatorless	1500	.002	100	99.998	4 min.
Type "A" Separatorless Divider	1500	.005	6.0	5.995	5 min.
Type "B" Separatorless	1500	.030	.65	.62	62 min.
Type "C" Tapered Sep.	1500	.006	.030	.024	62 min.
Type "D" Mini Separator	1500	.005	.017	.012	62 min.
Type "E" Standard HEPA	1000	.010	.020	.010	62 min.

*DOP Penetration Tests conducted using Q107 Penetrometer (136-300-175A)

Table I Entrained water tests.

Filter Type	Air Flow CFM	Initial DOP % Pen.	Initial ΔP In. W.G.	DOP Pen. Prior to Visible Rupture %	Final DOP % Pen.	Max. ΔP In. W.G.
Type "A" Separatorless	1000	.001	4.0	0.30	1.5	20.0
Type "E" Standard Separator	1000	.001	4.2	0.001	6.0	31.25
Type "A" Separatorless	1500	.001	4.1	0.012	0.06	13.25
Type "D" Mini Separator	1500	.001	4.0	0.004	0.004*	30.25
					At Visible Rupture	

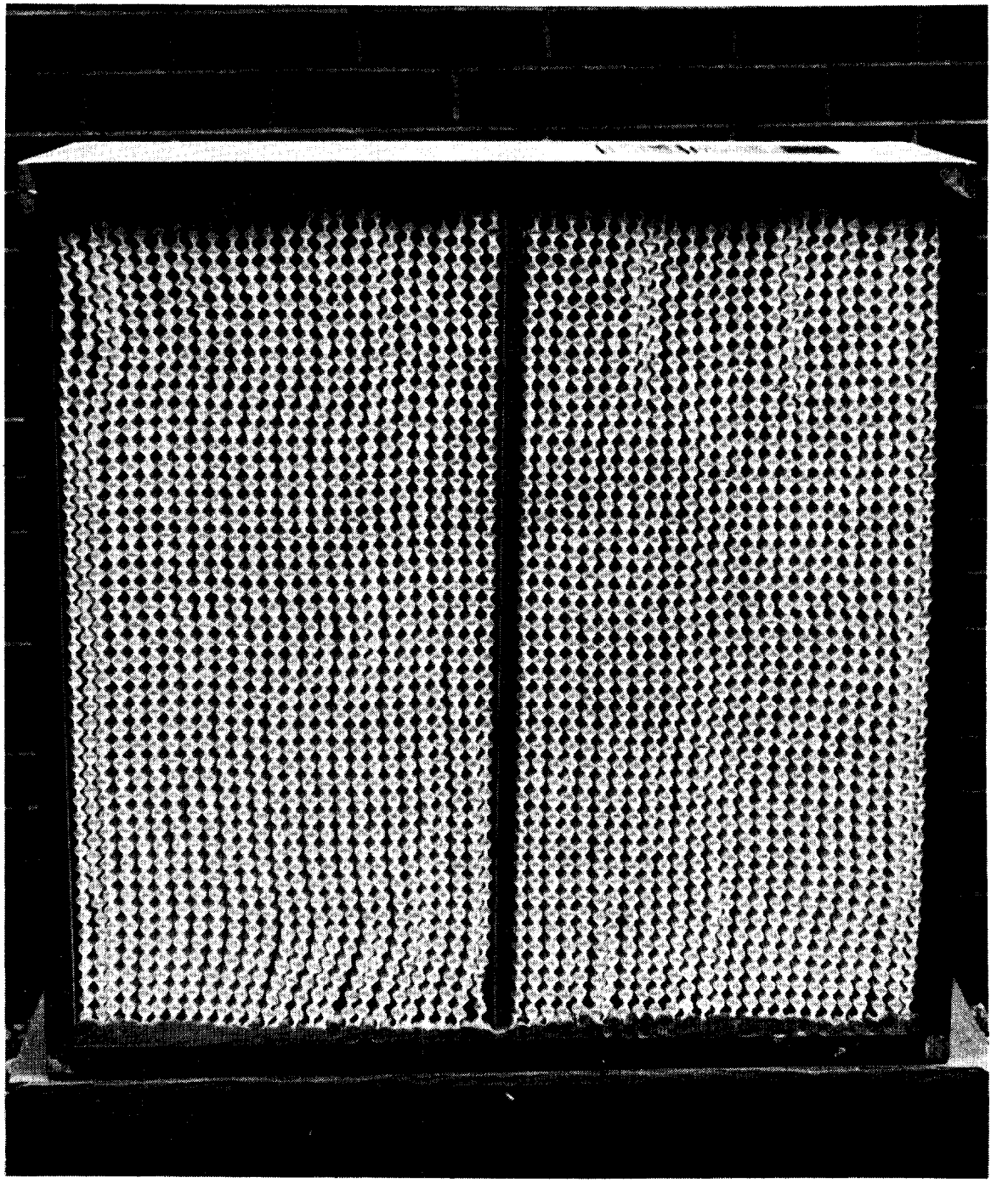
*No sign visible rupture.

In Situ Cold DOP Penetration Tests (2)

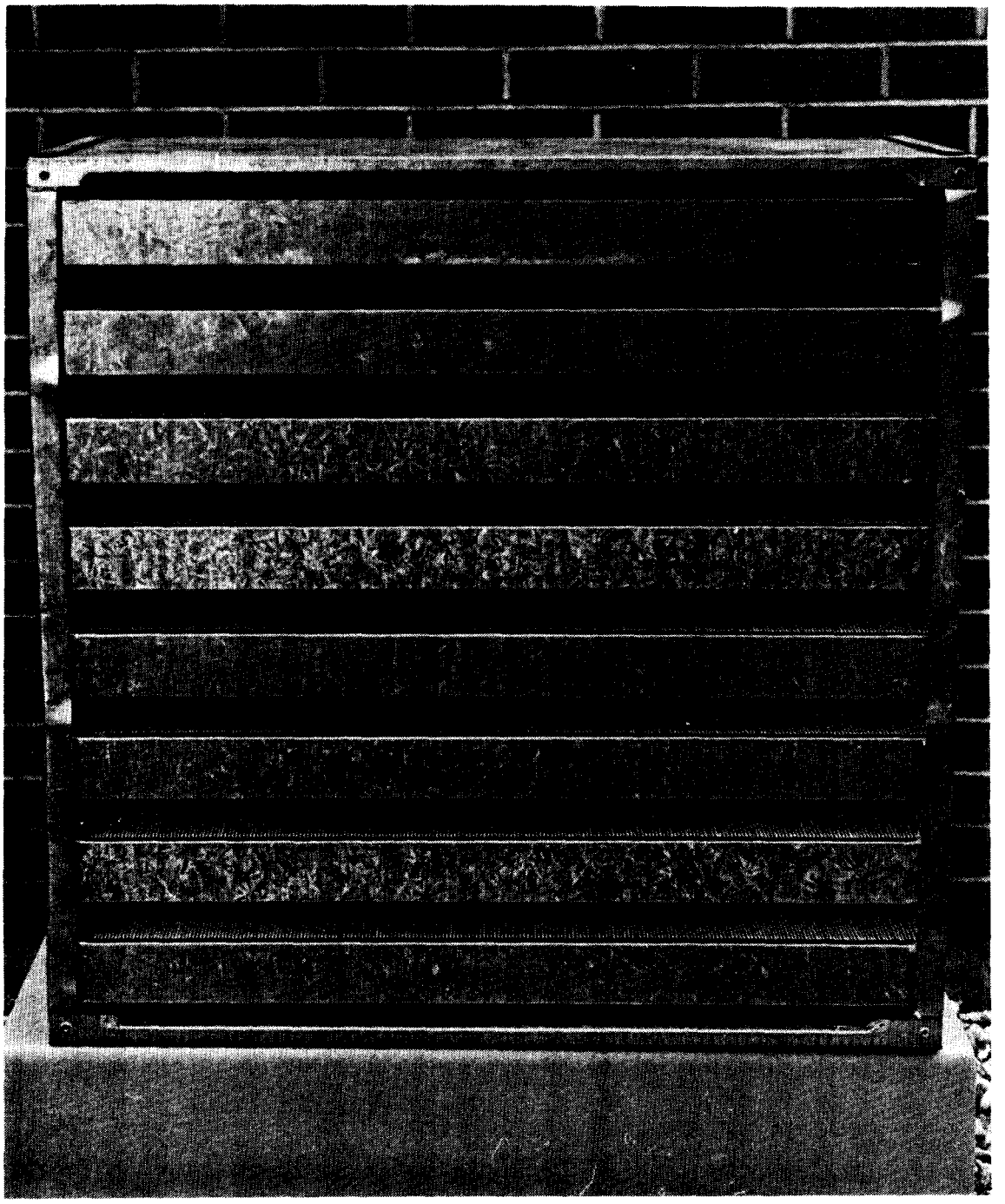
Table II Excessive pressure tests.

Filter Type	Flow CFM	Initial Wt. lb.	Initial Wt. oz.	Final Wt. Corrected to 6.0" W.G. lb.	Final Wt. Corrected to 6.0" W.G. oz.	Sodium Oxide Held lb.	Sodium Oxide Held oz.	Media Surface Area ft^2
Type "A" Separatorless	1500	30	14	32	6	1	8	250
Type "D" Mini Separator	1500	34	8	37	5-1/2	2	13-1/2	320
Type "C" Tapered Sep.	1500	36	6	37	3-1/2		13-1/2	246

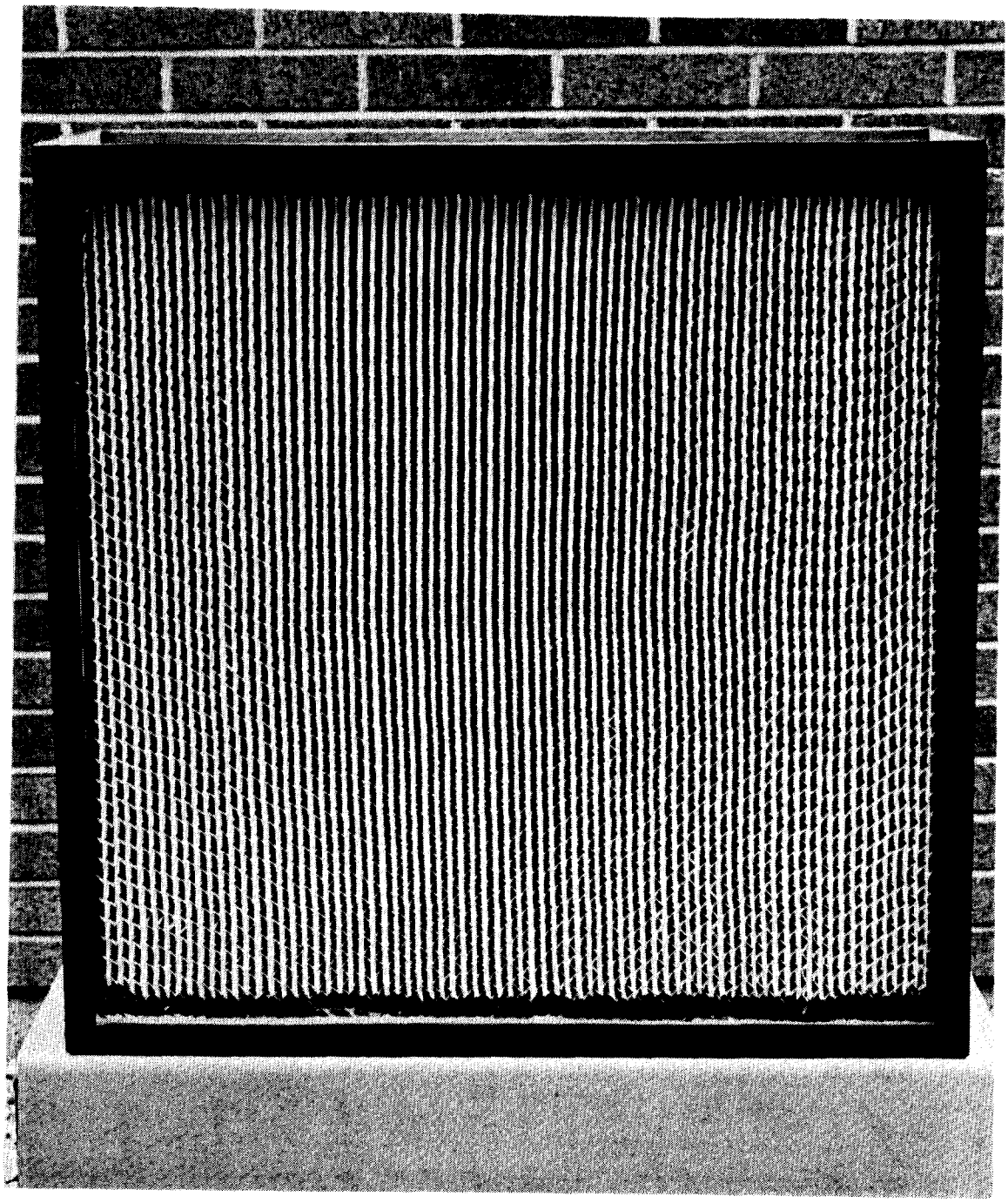
Table III Sodium oxide smoke loading.



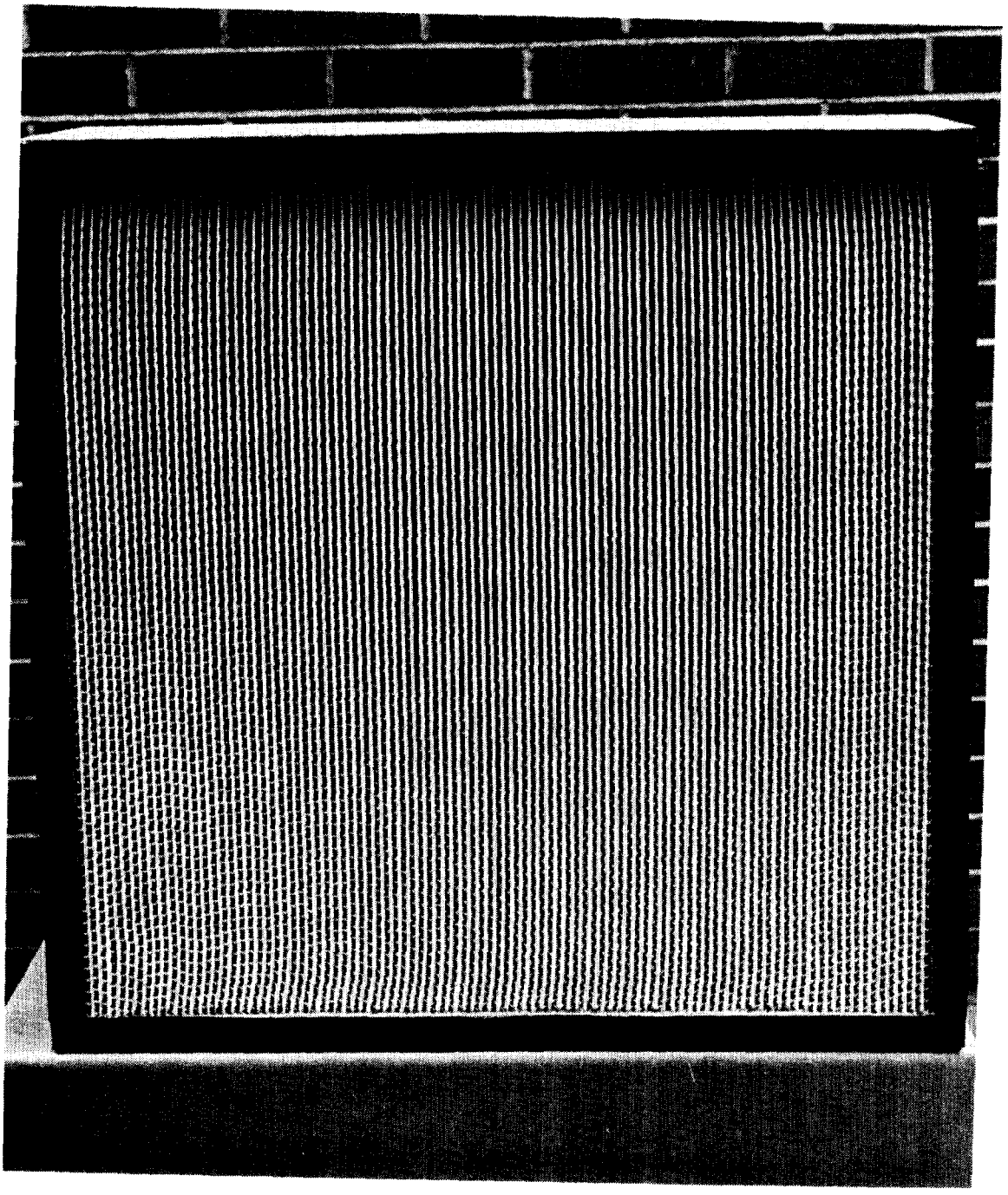
Type "A" - Separatorless Filter
"Figure 1A"



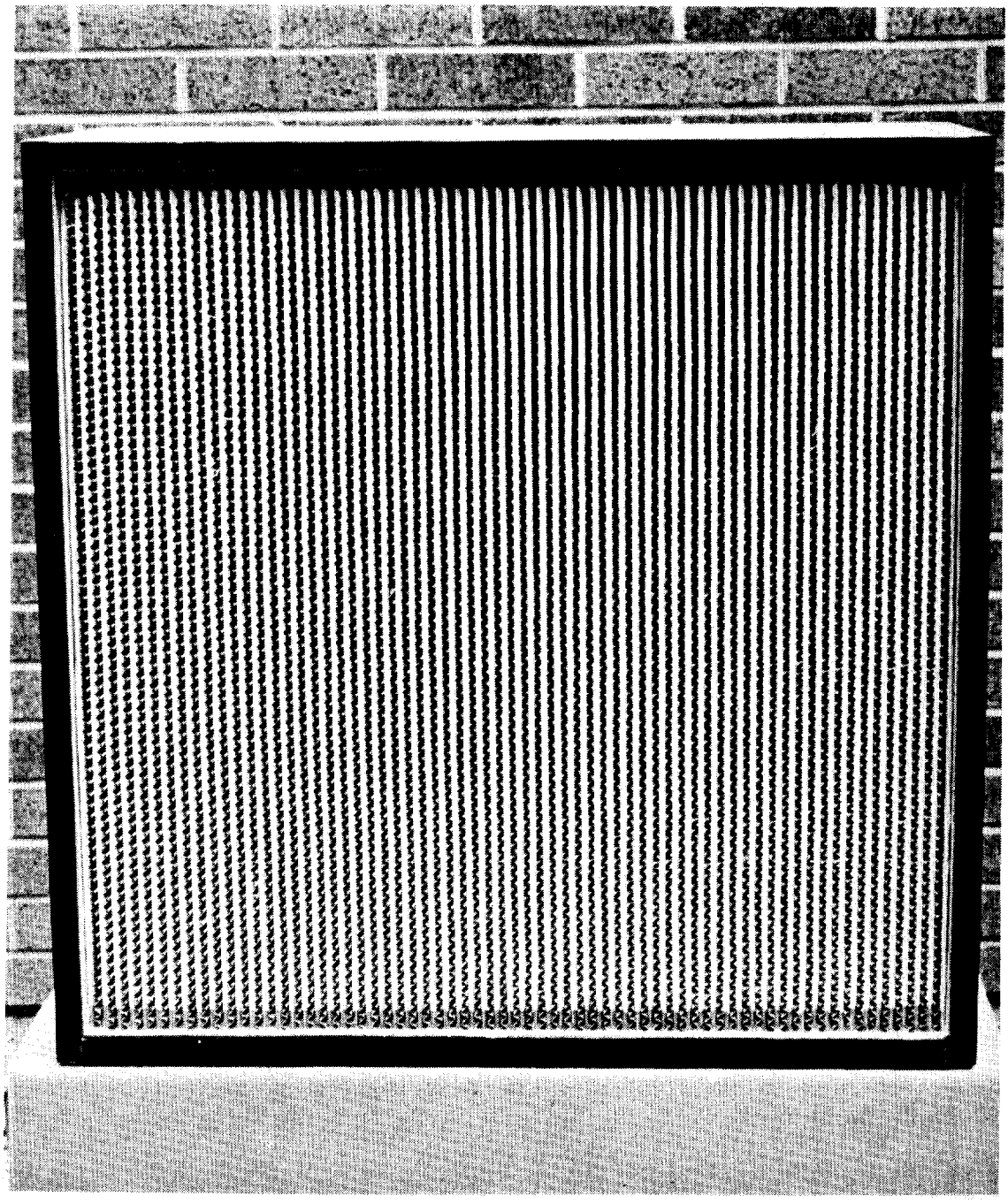
Type "B" - Separatorless Filter
"Figure 1B"



Type "C" - Tapered Separator Filter
"Figure 1C"



Type "D" - Mini Separator Filter
"Figure 1D"



Type "E" - Standard Separator Filter
"Figure 1E"

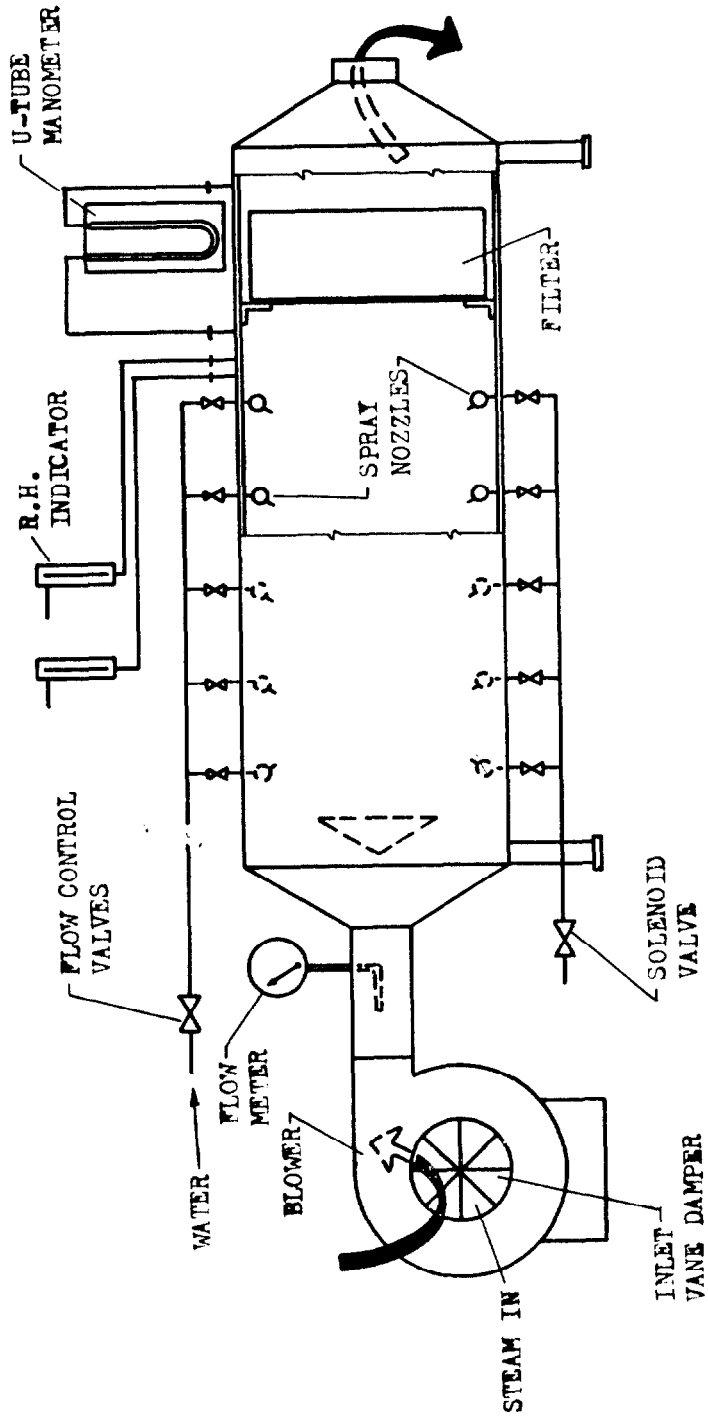
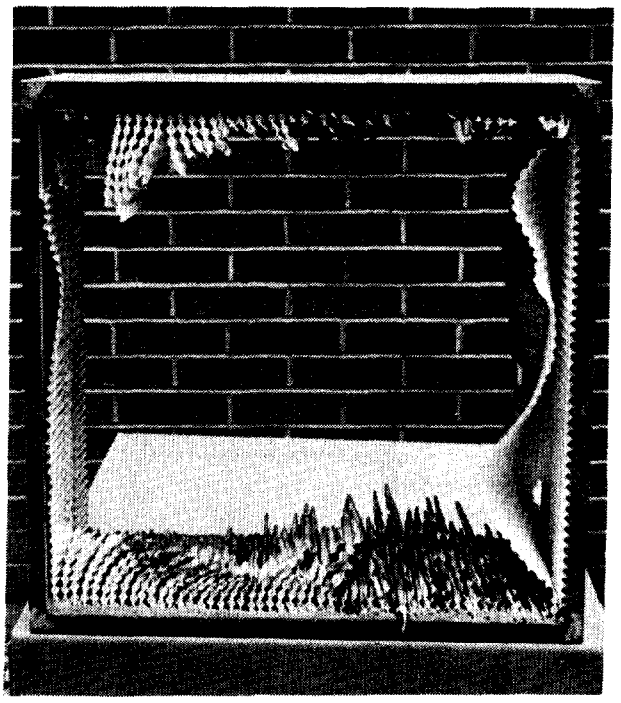
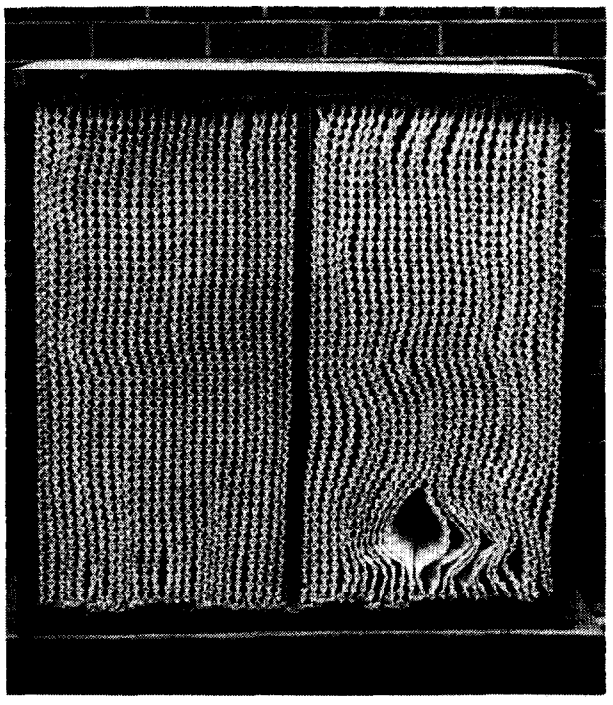


Figure 2 MSA moisture over pressure test system.

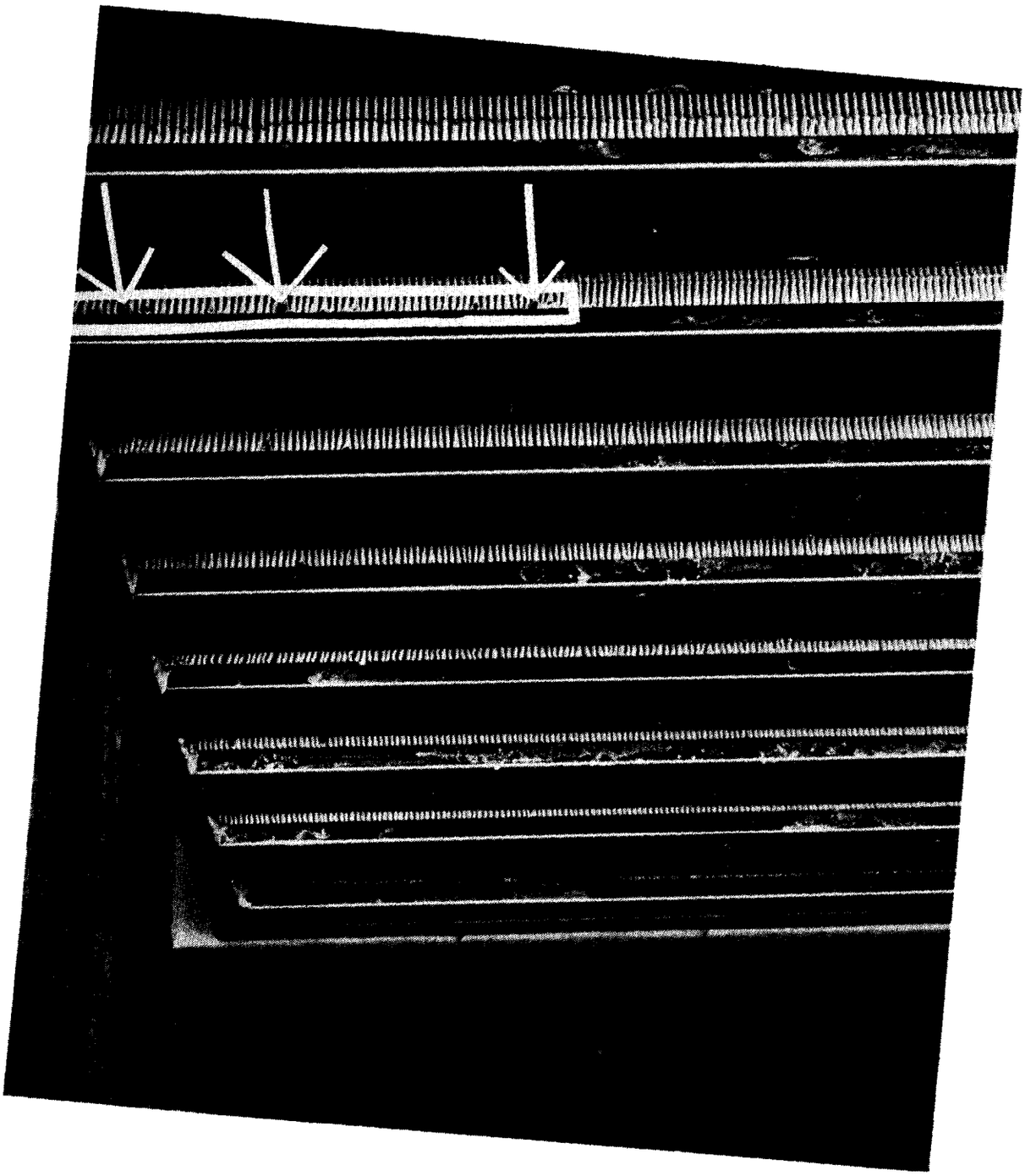


Shows complete blow-out when filter is not supported by hardware cloth and center separator.

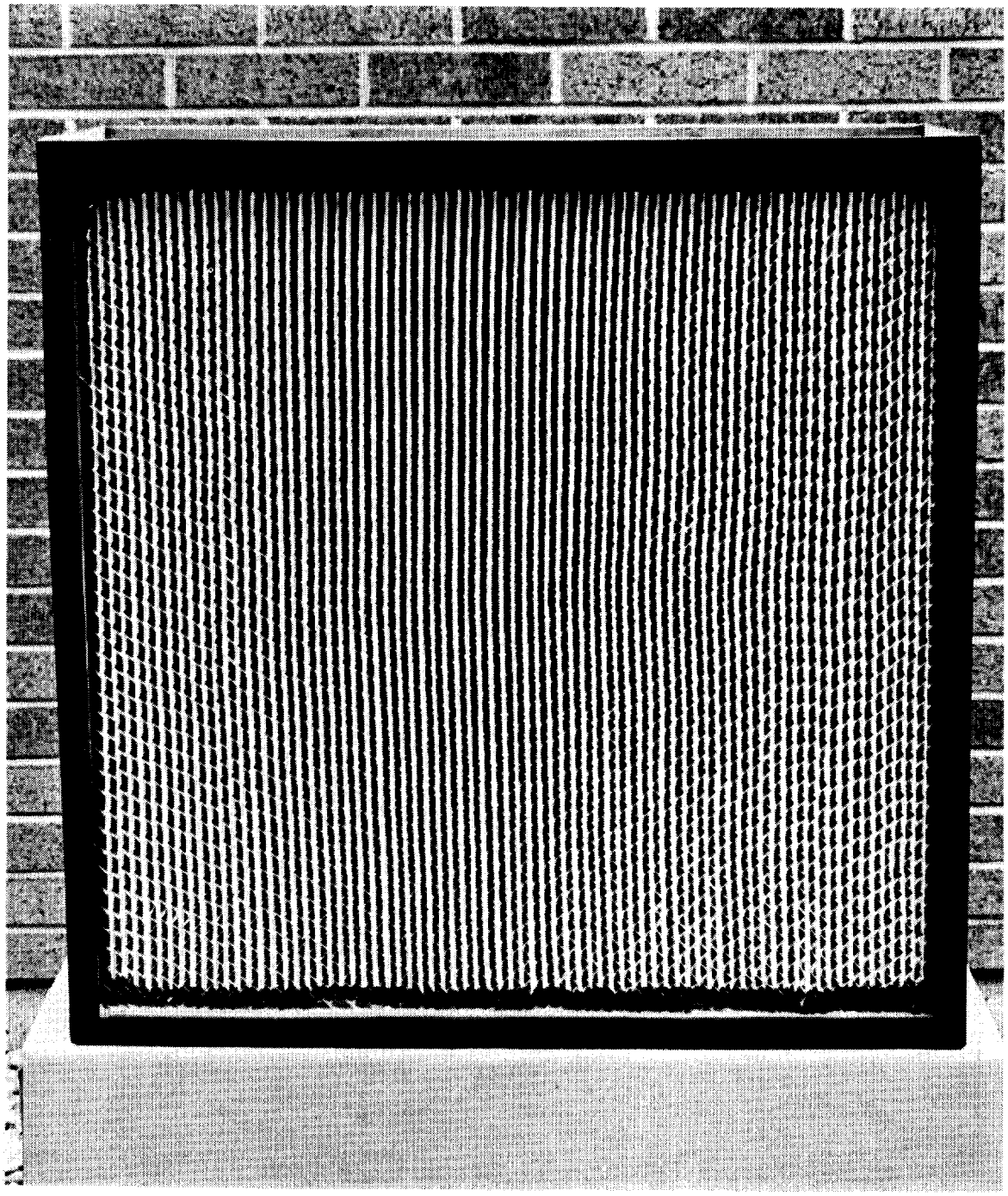


Shows heavy structural damage even when filter is supported by hardware cloth and center divider.

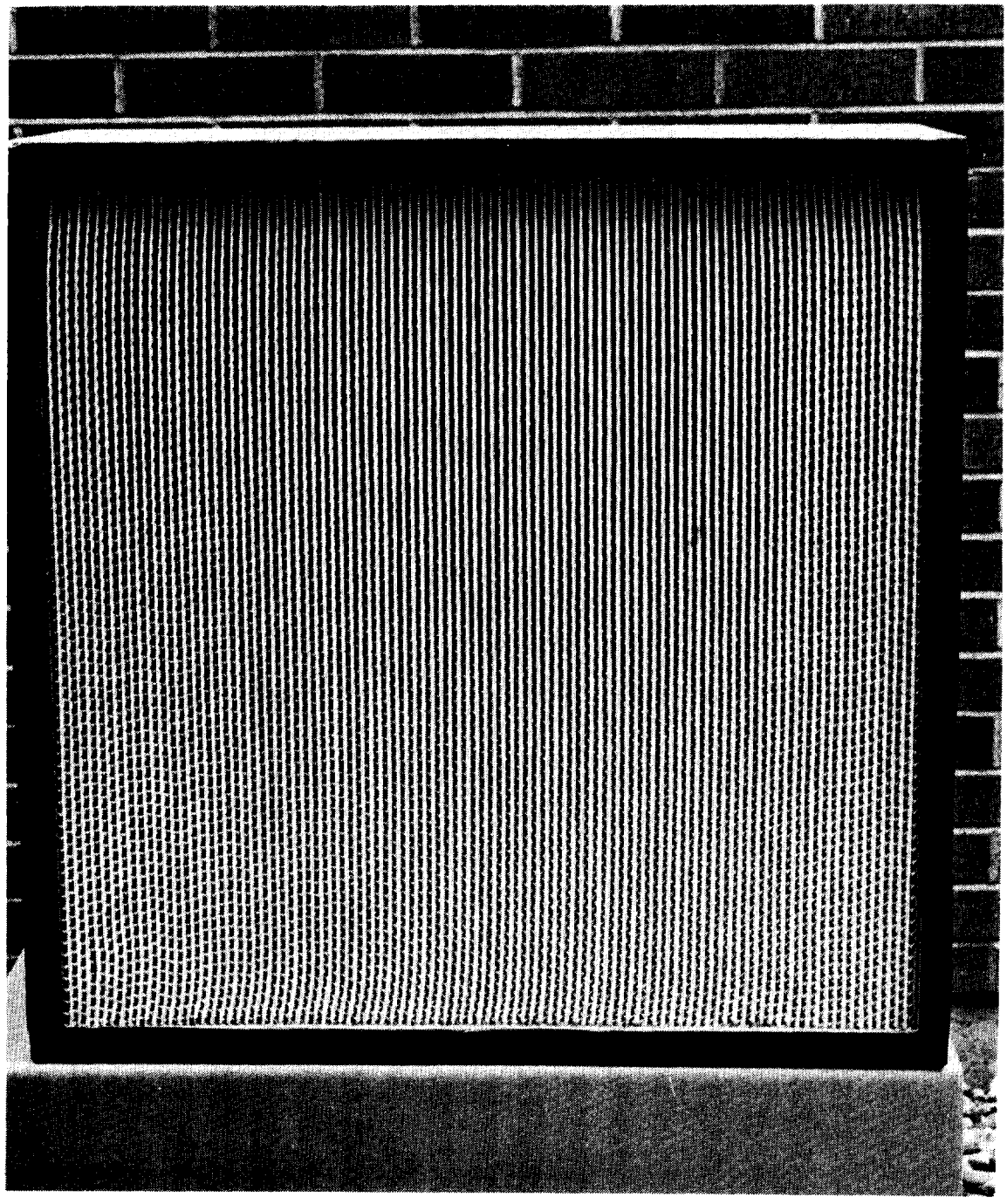
Type "A" - Separatorless Filter
"Figure 3A"



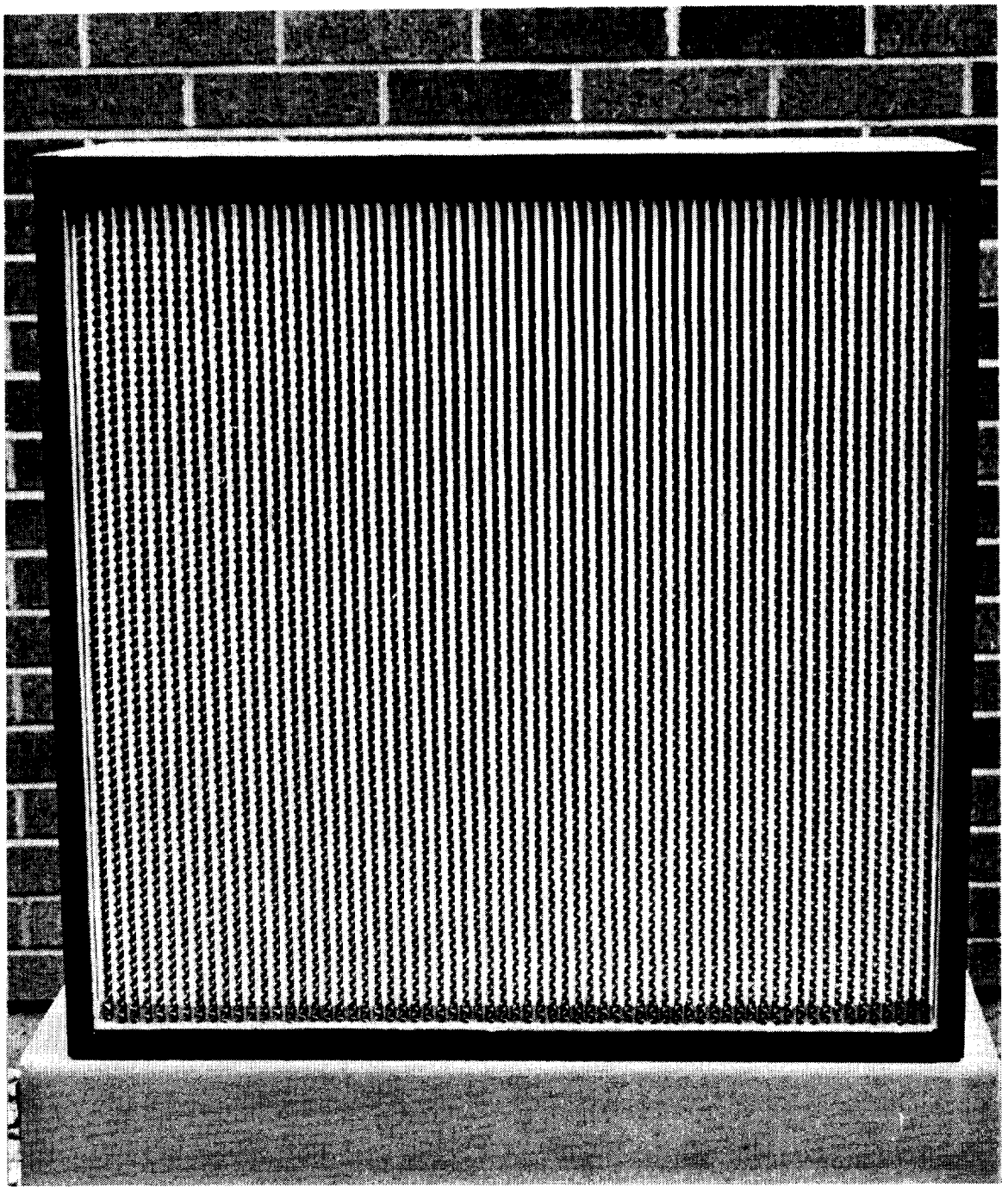
Type "B" - Separatorless Filter
"Figure 3B"



Type "C" - Tapered Separator Filter
"Figure 3C"



Type "D" - Mini Separator Filter
"Figure 3D"



Type "E" - Standard Separator Filter
"Figure 3E"

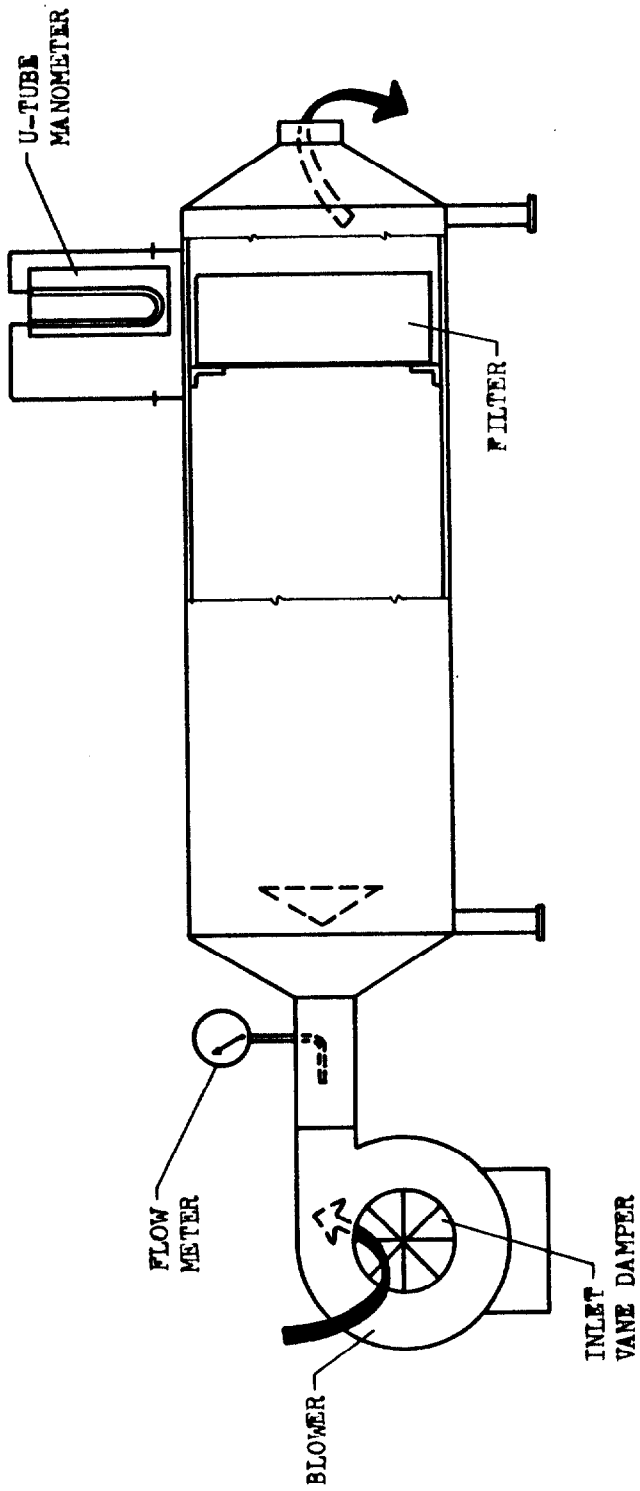


Figure 4 MSA high pressure test system.

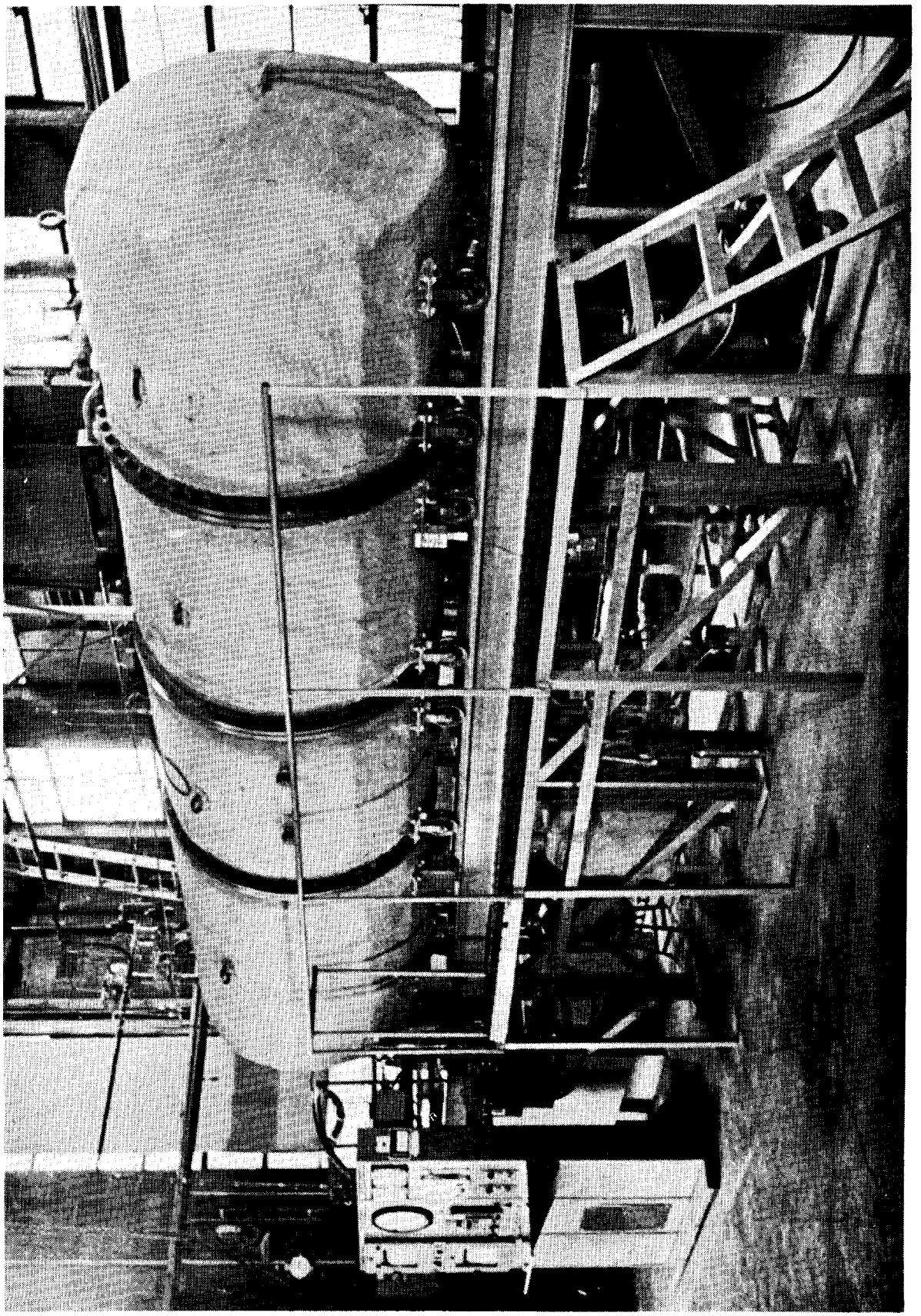


FIG. 5 - ENVIRONMENTAL TEST FACILITY (ETF), GENERAL VIEW

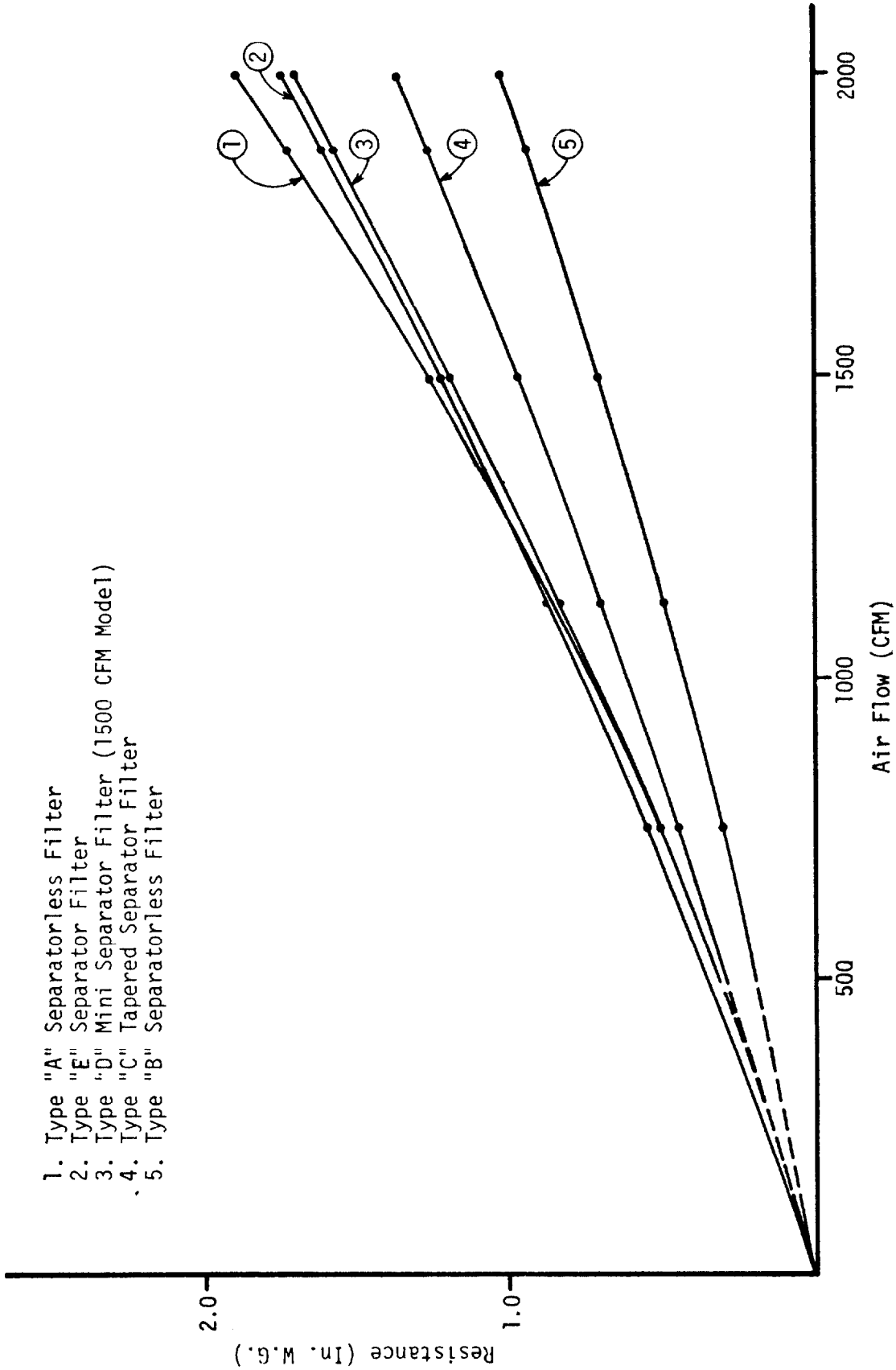


Figure 6 Clean filter air flow curves.

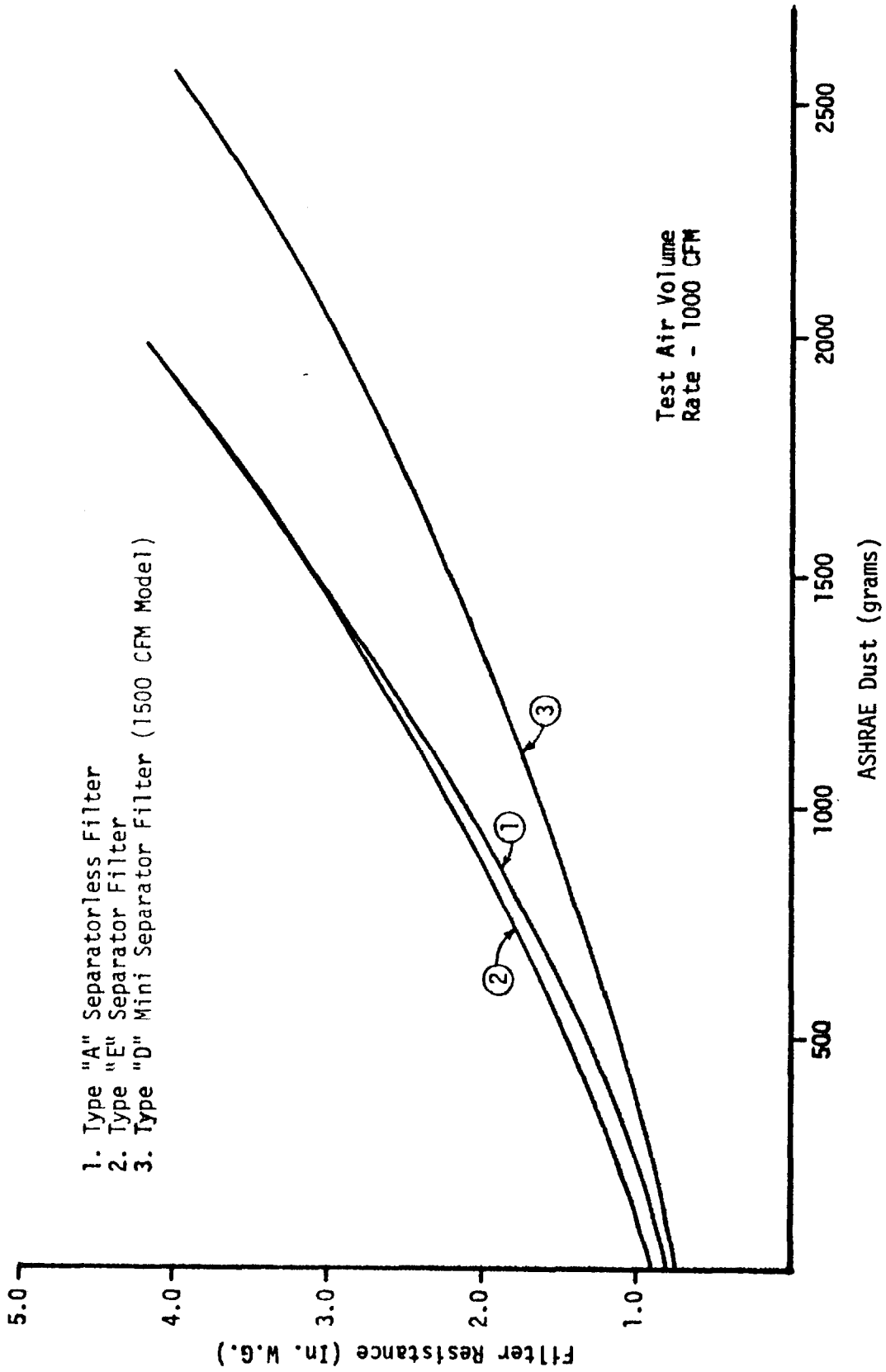


Figure 7 ASHRAE 52-68 dust loading curves.

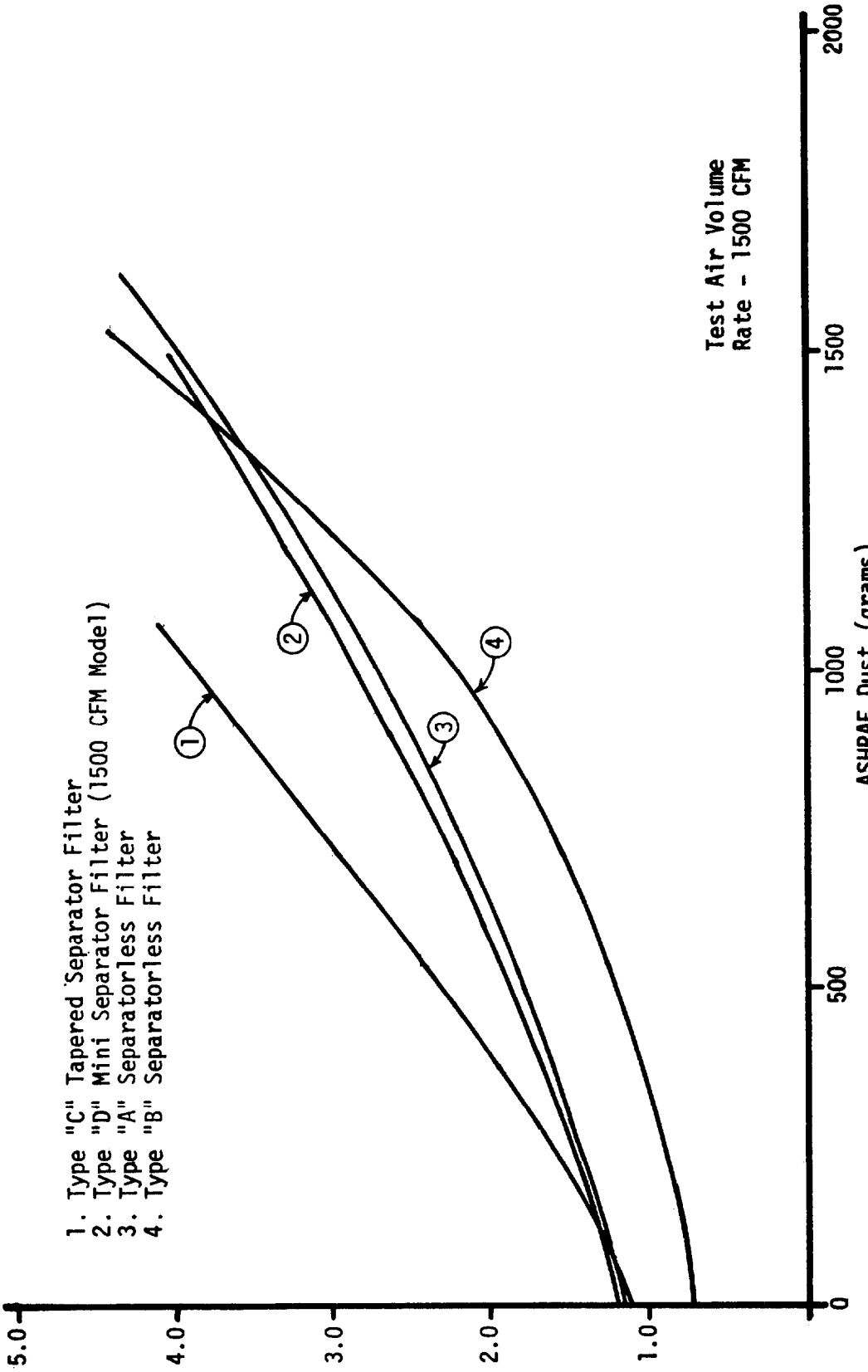


Figure 8 ASHRAE 52-68 dust loading curves.

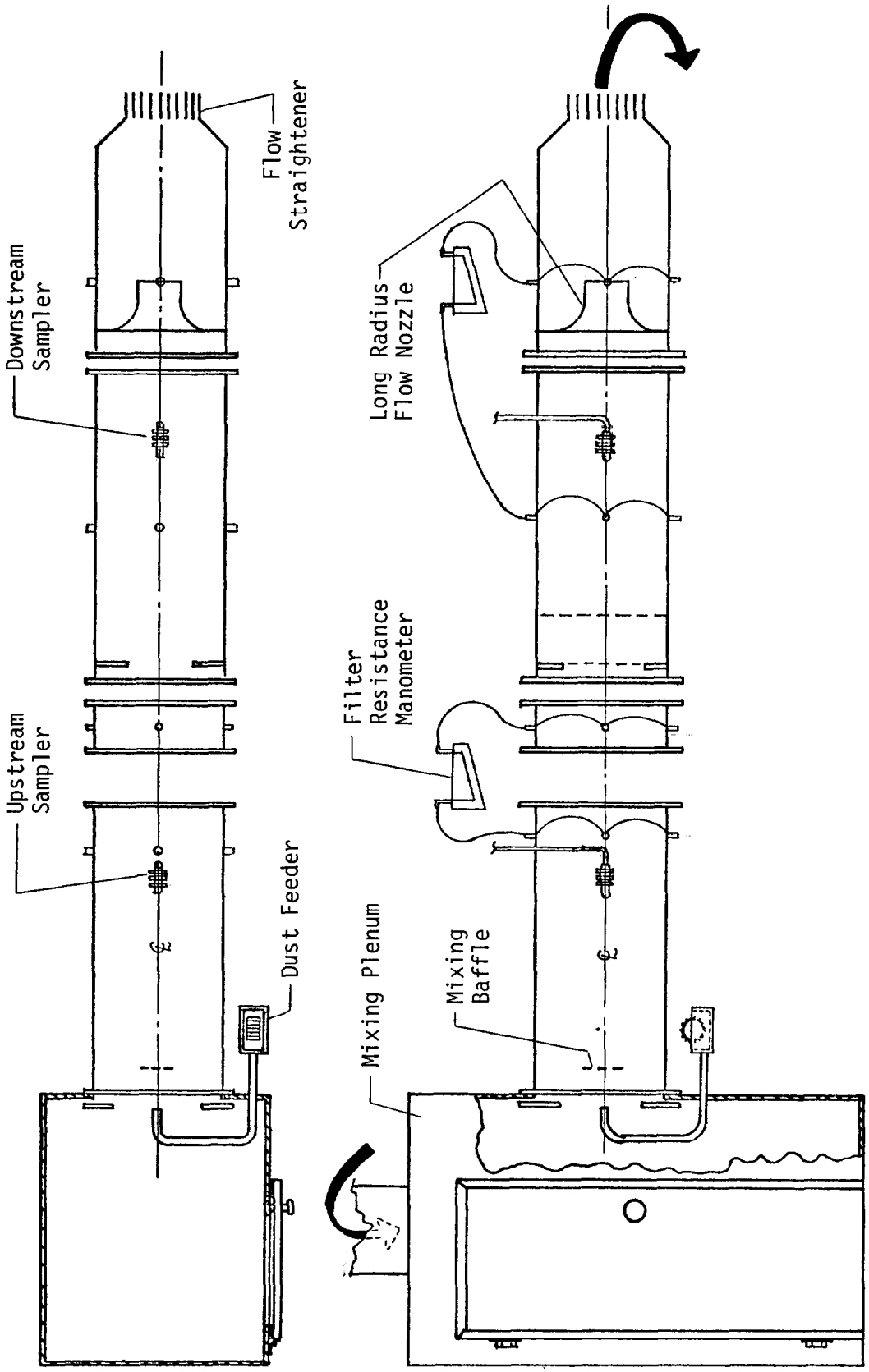


Figure 9 ASHRAE 52-68 test duct.

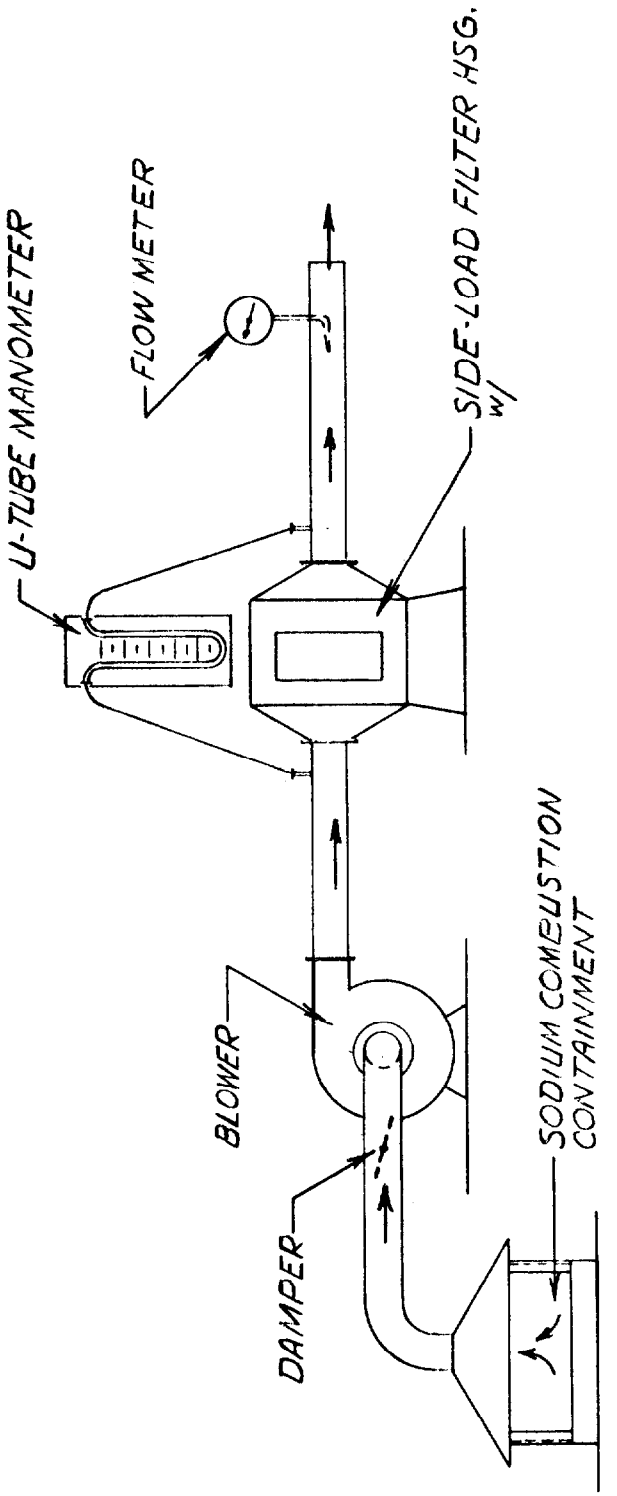


Figure 10 MSA sodium fume test duct.

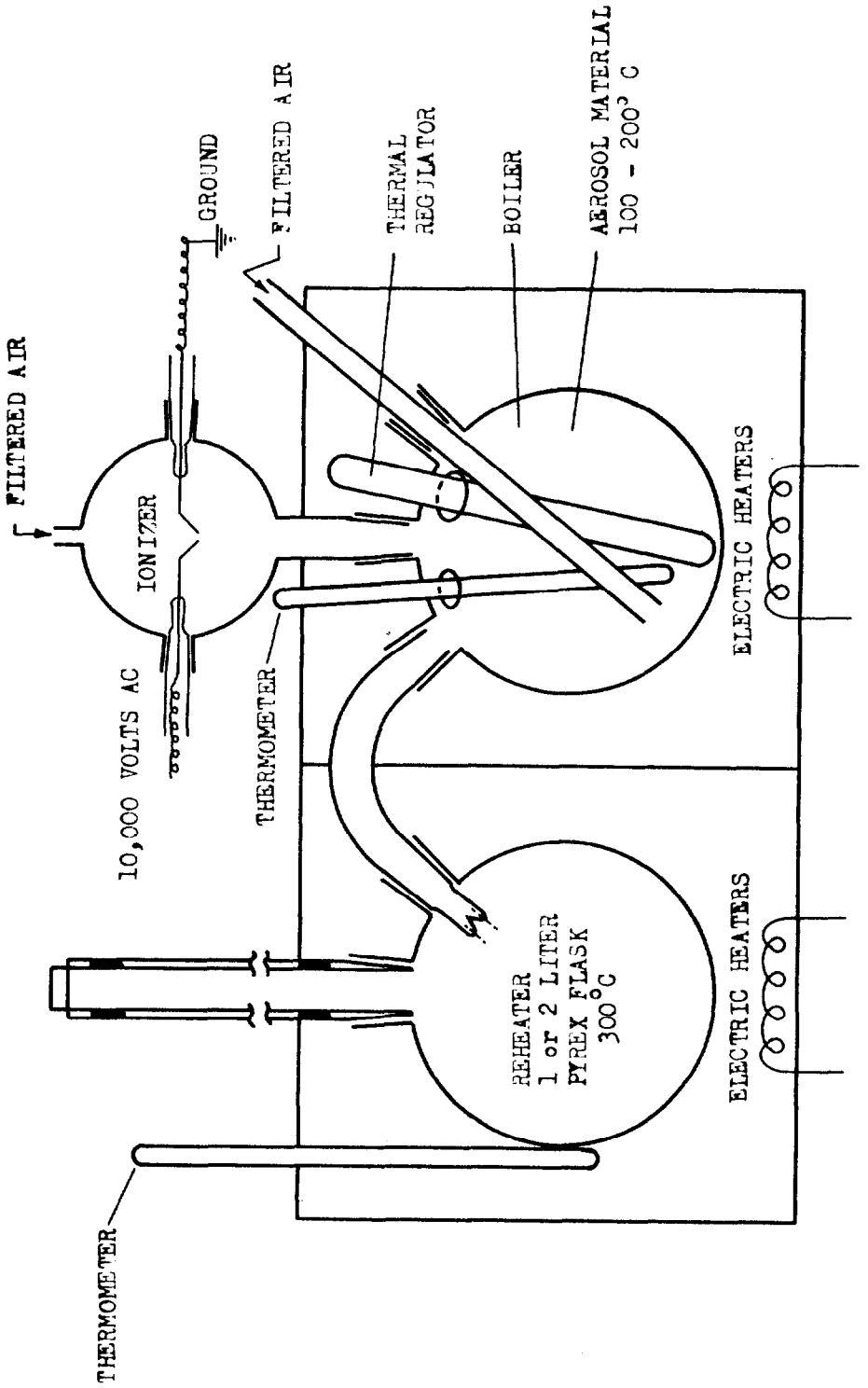


FIGURE 11 HOMOGENEOUS AEROSOL GENERATOR

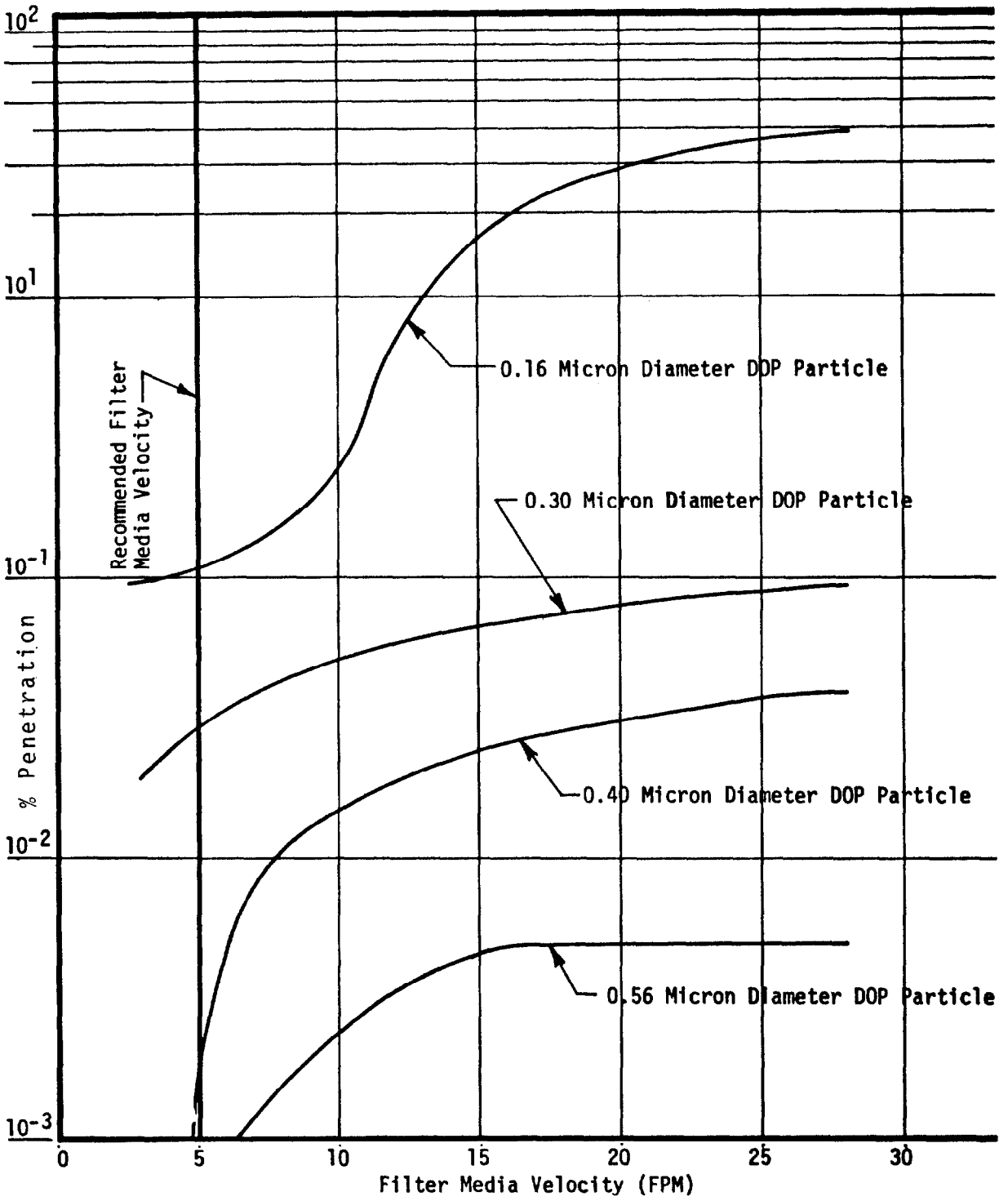


Figure 12 Particle size penetration curves.

VI. References

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8. H. W. Kundson and L. White, "Development of smoke penetration meters", NRL Report P2642, (Sept. 14, 1945).

9. C. A. Burchsted, "Minutes meeting April 27, 1976, Institute of Environmental Sciences Nuclear Energy Committee", (Paragraph 4-b, page 2), (May 18, 1976).

10. J. Truitt, "Effect of velocity on efficiency of absolute filter media for removal of stainless steel - UO_2 Aerosol", ORNL Nuclear Safety Research and Development Program Bi-Monthly Report for January-February, 1971, USAEC Report, ORNL TM3342, Oak Ridge National Laboratory, (1971).

DISCUSSION

CADWELL: Did you do any prolonged service life tests on these filters to the extent of a year or more?

GUNN: No, we didn't. We started but we didn't finish.

CADWELL: How many filter units were tested to develop the data for the Type A Separatorless filter?

GUNN: Approximately 28 filters.

CADWELL: Measurements of effective filter media area would indicate areas different from those included in your study.

EDWARDS: How do you explain that the Type A Separatorless filter passed all QPL requirements, and the mini-separator filter did not?

GUNN: We found essentially no difference in test performances between our "mini type" and "mini type" as supplied by another who passed all QPL tests. However, it is my understanding that we also passed all tests except the media radiation requirement. Meeting or not meeting this requirement would not effect the results reported in this study.

EDWARDS: Why did your test criteria exceed the requirements of the military specification?

GUNN: We did not intend to verify the Mil-Spec in this study.

EDWARDS: Your opening statement implied a cooperative venture between MSA and other filter vendors when, in fact, Flanders Filters was not notified nor invited to submit filters for testing.

GUNN: I did not indicate anywhere in my speech and/or paper that this was a cooperative venture.

EDWARDS: How can you be sure that the products tested were intended for use in the nuclear industry?

GUNN: We can only go by what the vendors so indicated.

PENETRATION OF HEPA FILTERS BY ALPHA RECOIL AEROSOLS*

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Abstract

The self-scattering of alpha-active substances has long been recognized and is attributed to expulsion of aggregates of atoms from the surface of alpha-active materials by alpha emission recoil energy, and perhaps to further propulsion of these aggregates by subsequent alpha recoils. Workers at the University of Lowell recently predicted that this phenomenon might affect the retention of alpha-active particulate matter by HEPA filters, and found support in experiments with ^{212}Pb. Tests at Oak Ridge National Laboratory have confirmed that alpha-emitting particulate matter does penetrate high-efficiency filter media, such as that used in HEPA filters, much more effectively than do non-radioactive or beta-gamma active aerosols. Filter retention efficiencies drastically lower than the 99.9% quoted for ordinary particulate matter were observed with ^{212}Pb, ^{253}Es, and ^{238}Pu sources, indicating that the phenomenon is common to all of these and probably to all alpha-emitting materials of appropriate half-life. Results with controlled air-flow through filters in series are consistent with the picture of small particles dislodged from the "massive" surface of an alpha-active material, and then repeatedly dislodged from positions on the filter fibers by subsequent alpha recoils. The process shows only a small dependence on the physical form of the source material. Oxide dust, nitrate salt, and plated metal all seem to generate the recoil particles effectively. The amount penetrating a series of filters depends on the total amount of activity in the source material, its specific activity, and the length of time of air flow. Dependence on the air flow velocity is slight. It appears that this phenomenon has not been observed in previous experiments with alpha-active aerosols because the tests did not continue for a sufficiently long time. A theoretical model of the process has been developed, amenable to computer handling, that should allow calculation of the rate constants associated with the transfer through and release of radio-active material from a filter system by this process.

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I. Introduction

The phenomenon of alpha recoil or alpha "creep" has been recognized for many years. As early as 1910 Russ and Makower[1] proposed that the expulsion of aggregates of material by alpha recoil energy might account for the observed self-scattering of alpha active material.* In 1915 Lawson[2] investigated the phenomenon, confirmed that the proposed mechanism existed, and gave it the name "aggregate recoil." The work of Lawson was subsequently summarized by Rutherford[3]. Chamie[4] and Jedrezezowski[5] used photographic methods to investigate and confirm aggregate recoil processes. A variety of alpha emitting materials including radium, polonium, and thorium "emanation" were used in these experiments. In 1973 Vento[6] studied aggregate recoil particles of lead-212 and its daughter products by autoradiographic methods and determined that the particles showed a size distribution with an average of about 10^3 atoms/particle (diameter ≈ 0.003 μm), a size for which the expected HEPA filter efficiency should be higher than 99.97%. Alpha creep has been familiar to many persons working with polonium, plutonium, and the transplutonium isotopes.

About 1973 Ryan, Skrable, and Chabot at Lowell Technological Institute proposed that this process of expulsion of aggregates from the surface of alpha-active materials by alpha emission recoil energy, and perhaps further propulsion of these aggregates by subsequent alpha recoils, might affect the retention of alpha active materials by HEPA filters. Using ^{212}Pb plated on needles as sources** of aggregate recoil particles, they obtained experimental confirmation of this hypothesis[7]. They demonstrated, by a combination of counting and autoradiographic methods, that recoil aggregates were generated from the source and were found on downstream filters in amounts greater than would be expected if the particles were being retained with the rated filter efficiency.

This paper presents the results of subsequent work at Oak Ridge National Laboratory in which lead-212, einsteinium-253, plutonium-238, and plutonium-239 were used as sources of aggregate recoil particles for filter penetration tests.

II. Experimental Methods

All tests were on a laboratory scale. The filters used were 47-mm diameter Gelman, Type A. According to the manufacturer's literature,[8] these filters have a minimum retention of 99.98% for 0.3-micron sized particles, and have physical characteristics that are similar to those of commercial HEPA filters. For testing, the filters were held either in a Gelman in-line filter holder (product No. 1235) in which the filters were in physical contact, or in a device of our construction in which the filters were held separate. There was no significant difference in results with the filters in contact or separate. These methods of holding the filters and of mounting sources of alpha-active materials are shown in Figure 1. Two types of sources were used, (1) metallic deposits plated either electrostatically or electrolytically on stainless-steel or gold needles, or (2) metal oxides deposited in the fibers of a filter disc. The ^{212}Pb sources were prepared by electrostatic plating in the device shown in Figure 2. The needle was placed in

*It can easily be shown that the daughter-atom of an alpha decay event will have approximately 100 keV of energy or roughly 10^5 times that necessary to break chemical bonds.

**Daughters of ^{212}Pb (^{212}Po) provide the alpha emissions necessary to propel ^{212}Pb aggregates containing both ^{212}Pb and equilibrium amounts of these daughters products.

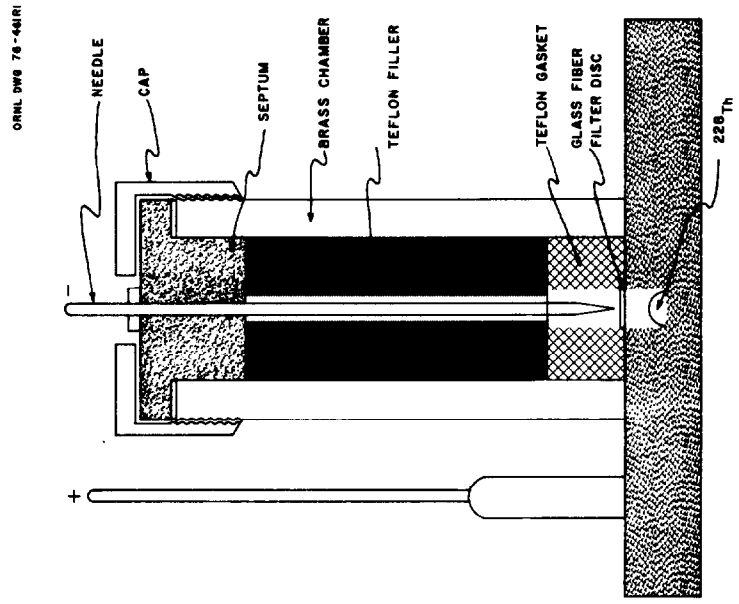

Figure 2 Lead-212 generator.

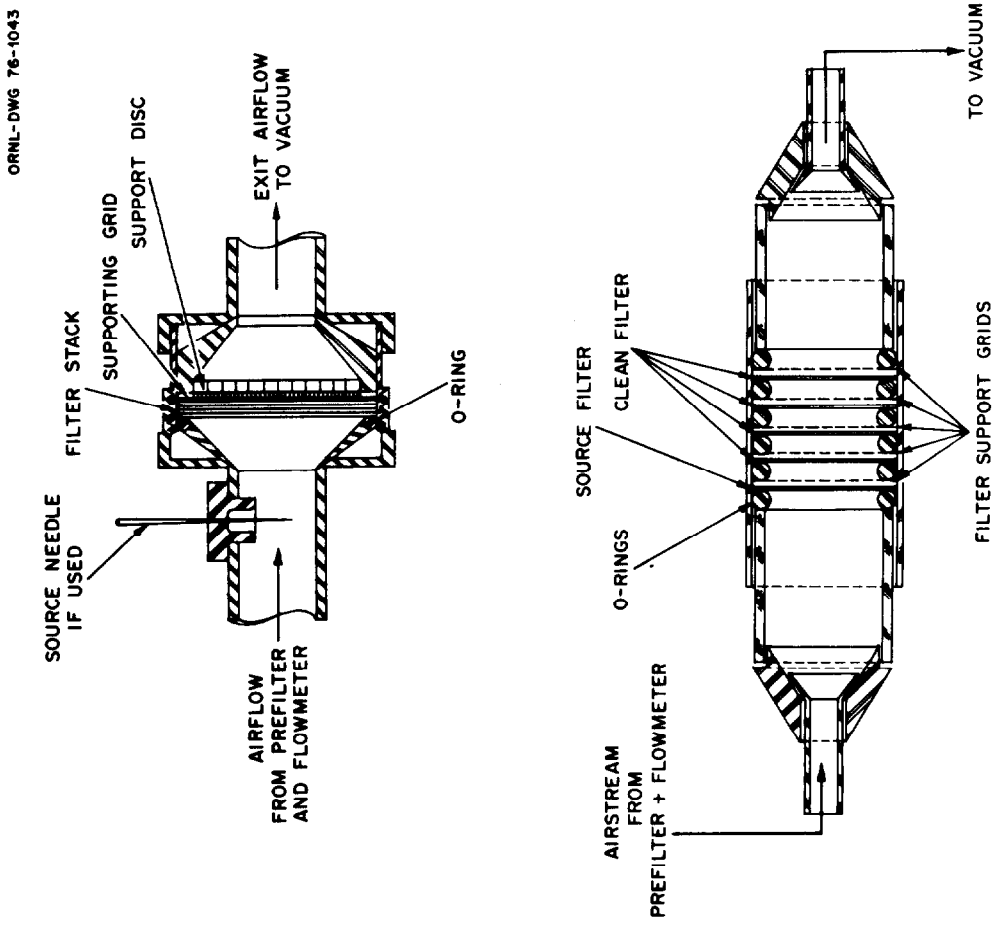

Figure 1 Experimental filter units.

proximity to a ^{228}Th source and maintained at a potential of about -9 volts. The 55-sec ^{220}Rn escaped through the filter stack and decayed to 0.16-sec ^{216}Po and then to 10.64-hr ^{212}Pb. Positive ions of both these species were collected on the needle. Sources of 0.5 to 25.0 μCi were prepared in this way. Sources of ^{253}Es on needles were prepared by conventional electroplating methods. Sources on filter discs were prepared by placing 25 to 100 microliters of a nitrate or chloride solution of the element on a filter disc, adding formic acid or ammonium hydroxide (to destroy the nitrate and/or hydrolyze the metal salt), and drying under infared lamps for approximately 30 minutes. These sources were presumed to be either metal oxides or oxy salts (MOX). Sources of 10^7 to 10^8 dpm were usually prepared in this way.

The lengths of the runs were determined by the half-life of the nuclide used and the length of time necessary to accumulate a meaningful amount of activity on the downstream filters. The half-lives of the nuclides, their specific activities, and the lengths of typical runs are listed in Table I. Initially many single-experiment runs were made; however, it became obvious that with runs of several

Table I Nuclides examined for aggregate recoil filter penetration.

Nuclide	$T_{1/2}$	g/Ci	Lengths of Runs
^{212}Pb	10.4 hrs	7.18×10^{-7}	4-30 hrs
^{253}Es	480.0 hrs	4.0×10^{-5}	2-20 days
^{238}Pu	87.4 y	5.8×10^{-2}	20 days
^{239}Pu	24,413 y	16.3	20 days

days duration efficient use of time demanded multiple-experiment runs, and the device shown in Figure 3 was built. In all cases, individual flow meters monitored the air flow through each filter stack. Flow rates were initially varied from 10 to 34 linear feet per minute (LFM), but later experiments, including all those with ^{238}Pu and ^{239}Pu, were run at 5 LFM, consistent with the usual flow rate through HEPA filter media. No measurable dependence of transfer on flow rate was observed in the experiments where it was varied. Each type of filter assembly used was tested for retention of a di-n-octyl phthalate mist by standard methods (DOP test) and found to perform as expected.

Liquid scintillation counting was the primary analytical method used. Source needles were leached and a portion of the solution was added to the scintillator, or the needle itself was immersed in the scintillator. (Tests indicated that the addition of a needle to the sample did not affect counting results.) Filter-disc sources were dissolved in dilute HF and diluted to a known volume, and an aliquot of this solution was added to the scintillator solution. Downstream filters were counted by immersion of the entire filter in the scintillator. Tests with known amounts of various alpha emitters dried on filter discs indicated this method of counting to be 100% efficient and reliable and reproducible within counting statistics. Studies of the decay rate for ^{212}Pb and of alpha and gamma spectra for the other isotopes confirmed that the activity seen on downstream filters originated from the source.

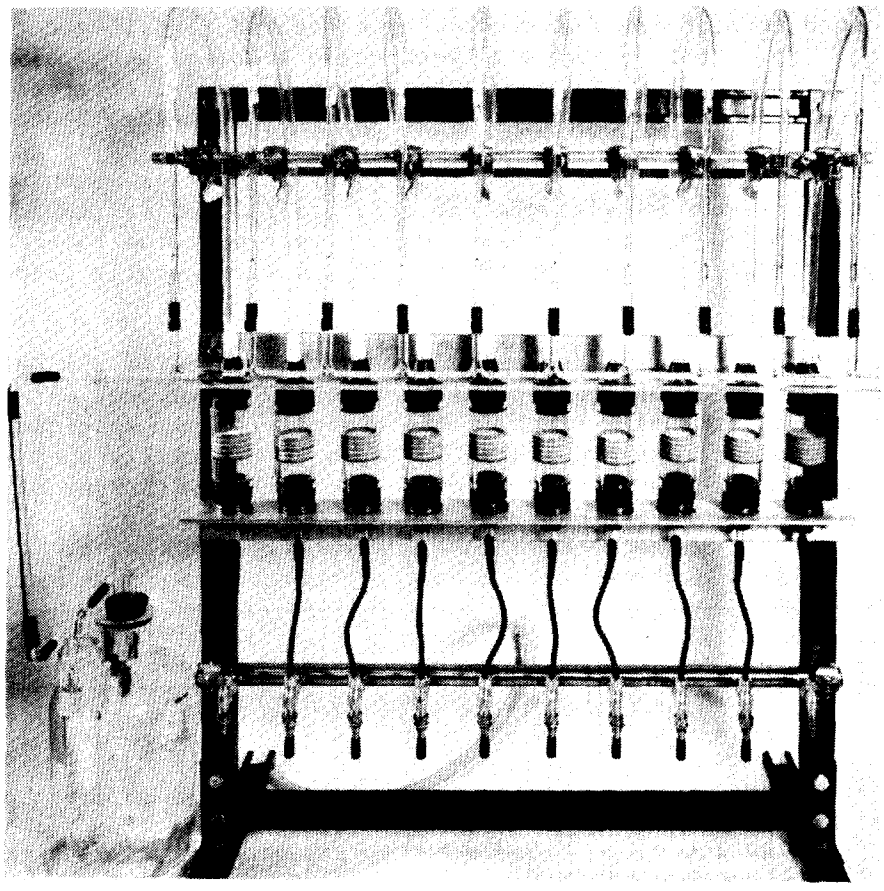

Figure 3 Ten-unit filter manifold.

III. Results and Discussion

General Observations

It should be emphasized in the beginning that in all these tests, the source material was firmly fixed to the needle or filter support. No normal dust or aerosol was generated to challenge the filter system. However, in all tests using sources containing alpha emitters, positive evidence of migration of the source material through a set of 4 or 5 filters in series was observed.

The fact that all of the alpha-active preparations produced migration of the source material through the filter stack is consistent with the idea of the expulsion of aggregate recoil particles from the source material and subsequent propulsion of these particles by alpha recoil energy. It should be noted here that in the case of ^{212}Pb, the migration of the non-alpha-active ^{212}Pb itself was observed, indicating that an aggregate of atoms is indeed expelled and propelled.

Tests in which only the beta-gamma active material $^{152,154}Eu$ was used showed no evidence of migration, but later tests with $^{152,154}Eu$ mixed with ^{238}Pu showed migration of the $^{152,154}Eu$. This again indicates the existence of aggregates.

The data obtained for aggregate recoil penetration of the filters by particles from the various sources are shown in Table II. The amount of material on downstream filters is small in all cases and represents only a minute fraction of the

source activity. However, the fraction of activity on the second and each succeeding downstream filter is much larger than would be expected from established filter efficiency and the amount on the filter immediately preceding it and does not represent a rate of decrease of activity consistent with filter efficiency. Further, it should be remembered that what we are observing appears to be a continuous process of release and transport, dependent on the amount of activity on the filter rather than on the concentration of any challenging aerosol. High-efficiency filters are, indeed, expected to perform with their rated efficiency with normal aerosols of alpha active materials over short periods of time, as has been shown by the extensive tests of Ettinger, et al.[9] The fact that the effect of aggregate recoil was not seen in those tests is attributed to the relatively short duration of the tests. Conversely, we believe that our observations can be explained only by the operation of an aggregate recoil mechanism as proposed above continuing over a relatively long period of time.

Mathematical Model

The change, with time, in the number of active atoms, N, on an alpha-active source can be described by the equation

$$\frac{dN_s}{dt} = -(K_s + \lambda)\, N_s(t), \tag{1}$$

in which λ is the usual radioactive decay constant, and K_s is the rate constant for transfer of atoms by the aggregate recoil process. If aggregate recoil particles are resuspended in the air stream and re-collected on the next downstream filter, then the change in activity on the first downstream filter after the source filter can be described by

$$\frac{dN_1}{dt} = K_s N_s(t) - (K_1 + \lambda)\, N_1(t). \tag{2}$$

The rate of change of activity on any downstream filter, m, is thus

$$\frac{dN_m}{dt} = K_{m-1} N_{m-1}(t) - (K_m + \lambda)\, N_m(t);\ m = 2,3,\ldots,M. \tag{3}$$

If it is assumed that $K_m = K_{m-1}$, etc., and $K_m > K_s$, this family of differential equations may be solved[10] to obtain an expression for the amount of activity on any filter at any given time,* giving:

$$N_m(t) = \frac{N_s(o) K_s k^{m-1}}{(K-K_s)^m}\, e^{-(\lambda + K_s)t}\, P(m,\ (K-K_s)t), \tag{4}$$

where:

$$P(m,x) = \frac{1}{\Gamma(m)} \int_o^x e^{-u} u^{m-1} du. \tag{5}$$

A computer code for fitting the data in Table II to equation 4 by a nonlinear least-squares analysis has been written, and values of K_s and K have been obtained for each set of data. Values of the transfer rate constants are listed in Table III. The errors for these transfer rate constants may be estimated by

*It is possible to solve the separate differential equations for the source and each filter obtaining K_s, K_1, K_2, etc., but this requires experimental values for N_s at t = 0 and t = t more accurate than can be reasonably obtained for successful application.

Table II Aggregate recoil filtration data

Source	Source Activity (dpm)	Run Time	Airflow (L.F.M.)	Activity on Filter (dpm) 1	2	3	4
Lead-212	9.6×10^6 (a)	4 h	34	123	22	107	50
	1.16×10^6 (a)	4 h	10	6,040	105	54	40
	5.48×10^7 (a)	4 h	10	509	259	45	99
	4.10×10^7 (a)	8 h	10	632	211	221	292
	2.69×10^7 (a)	4 h	10	1,650	23	23	18
	1.31×10^7 (a)	22 h	10	156	9	18	11
	4.93×10^7 (a)	4 h	8	162	32	17	8
	4.05×10^7 (a)	4 h	16	45	5	4	2
	1.82×10^7 (a)	4 h	24	61	4	5	8
	8.60×10^6 (a)	8 h	16	28	3	6	5
	2.14×10^7 (a)	8 h	24	53	22	21	20
	1.84×10^7 (a)	8 h	8	57	52	28	12
	2.60×10^7 (a)	16 h	8	56	17	16	15
	2.67×10^7 (a)	16 h	16	74	7	3	6
	2.19×10^7 (a)	16 h	24	378	31	11	6
	2.87×10^7 (a)	30 h	8	43	2	3	6
	5.18×10^6 (a)	30 h	16	29	4	2	3
Plutonium-238	3.7×10^8 (b)	20 d	5	1,373	16	10	3
	3.5×10^8 (b)	20 d	5	1,754	21	13	20
	3.6×10^8 (b)	20 d	5	1,175	18	7	71
	3.5×10^8 (b)	20 d	5	3,680	116	22	23
	3.7×10^8 (b)	20 d	5	637	22	17	30
	3.6×10^8 (b)	20 d	5	675	29	7	8
	3.6×10^8 (b)	20 d	5	2,598	30	17	9

Table II (Contd.) Aggregate recoil filtration data

Source	Source Activity (dpm)	Run Time	Airflow (L.F.M.)	Activity on Filter (dpm) 1	2	3	4
Einstein-ium-253	1.52×10^{8} (b)	10 d	23	3,454	36	7	5
	1.69×10^{7} (b)	48 h	10	839	22	19	7
	5.0×10^{7} (b)	11 d	10	5,110	29	17	99
	1.32×10^{7} (a)	11 d	10	348	11	7	3
	9.0×10^{6} (a)	20 d	10	5,134	205	100	46
	7.48×10^{7} (b)	20 d	10	5,511	14	9	7

Table III Summary of Transfer Rate Constants

Nuclide	Source Rate Constant (K_s)	Downstream Rate Constant (K)
^{238}Pu	$2.44 \times 10^{-6} d^{-1} \pm 9.38 \times 10^{-8} d^{-1}$	$8.59 \times 10^{-3} d^{-1} \pm 3.58 \times 10^{-3} d^{-1}$
^{253}Es	$1.16 \times 10^{-6} h^{-1} \pm 4.68 \times 10^{-7} h^{-1}$	$2.44 \times 10^{-3} h^{-1} \pm 7.87 \times 10^{-4} h^{-1}$
^{212}Pb	$4.17 \times 10^{-8} h^{-1} \pm 3.37 \times 10^{-8} h^{-1}$	$2.81 \times 10^{-3} h^{-1} \pm 1.53 \times 10^{-3} h^{-1}$

propagation of the known counting errors of each filter. The standard deviations associated with these transfer rate constants are included in Table III.

Release calculations. From the model above, the release from filter m during time interval t is

$$R_m(t) = \int_0^t N_m(\tau)\, d\tau = N_s(o)\left(\frac{K_s}{K_s + \lambda}\right) \frac{K}{K + \lambda}^{m} \left\{ P(m, (K + \lambda)t) - e^{-(K_s + \lambda)t} \left(\frac{K + \lambda}{K - K_s}\right)^m P(m, (K - K_s)t) \right\} . \quad (6)$$

Evaluation of this equation by computer methods[10] allows calculation of amounts of activity released by aggregate recoil under various conditions. Figure 4 shows the total release in μCi as a function of time for ^{238}Pu. The conditions assumed for these calculations are: (1) a set of four filters in series and (2) at time zero the first filter collects a unit amount (1 Ci) of the alpha active material. In each case the activity released is significantly greater than that expected from normal filter penetration of particles in the aggregate particle size range. Because of the design of the experiments and the method of calculation, the curve shows release due to aggregate recoil only.

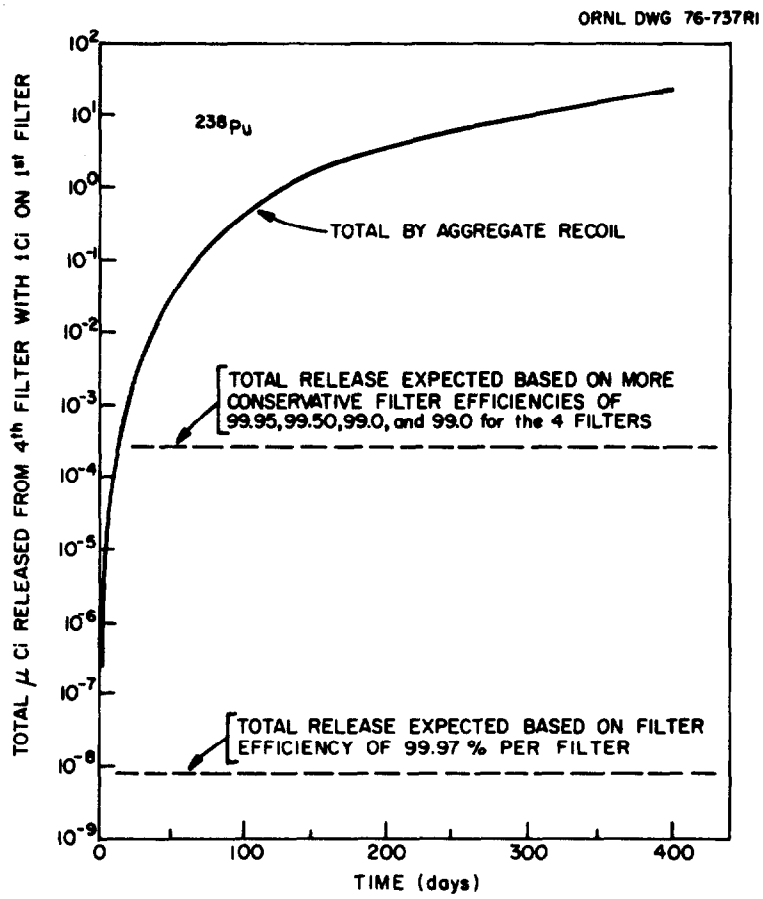

Figure 4 Calculated release of ^{238}Pu from a set of four HEPA filters, with 1 Ci initially collected on the first filter, as a function of elapsed time.

Figure 5 shows total releases calculated for three nuclides, ^{212}Pb, ^{253}Es, and ^{238}Pu, as a function of elapsed time in units of half-life. The release rates appear to increase rather sharply for 1 to 2 half-lives. Nuclides with longer half-lives eventually release a larger fraction of their source activity because the recoil-promoted migration through the filter system continues for a longer period of time.

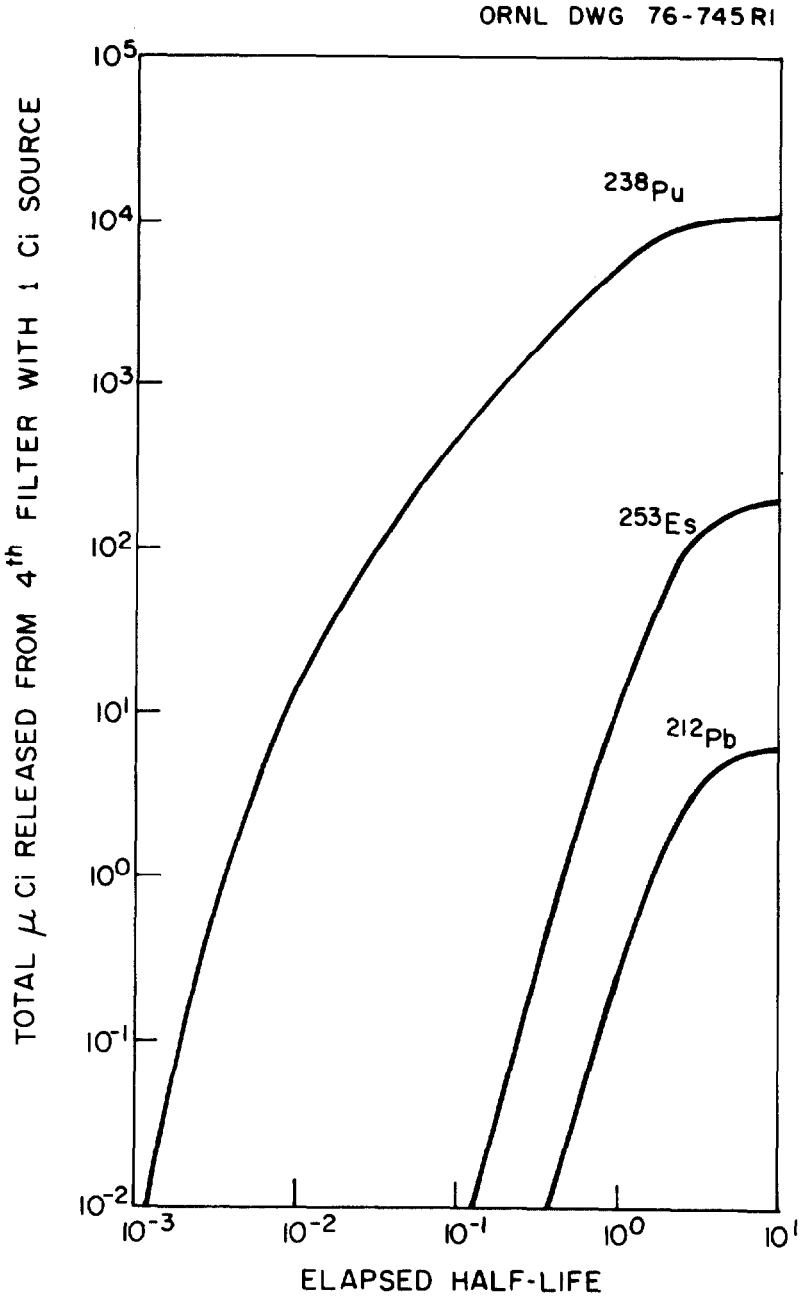

Figure 5 Calculated release of nuclides from a set of four HEPA filters, with 1 Ci initially collected on the first, as a function of elapsed half-lives.

Figure 6 allows comparison of the effectiveness of the aggregate recoil process in the penetration of a four-filter system for nuclides of different half-lives. Here we assumed all the nuclides to have approximately the same atomic weight and transfer rate constants as ^{238}Pu and calculated the total release to be expected during 300 days with initially 5 grams of the hypothetical nuclide on the first filter. Penetration by long half-life nuclides is slight because of their low specific activity (not much activity in 5 grams), and very short half-life nuclides do not penetrate effectively because much of the nuclide disappears through radioactive decay before it can migrate through the filter system. The Maximum effectiveness for the conditions selected appears to occur at about $T_{1/2}$ = 150 days. It is interesting to note in this connection that $^{210}Po(T_{1/2}$ = 138 days) is particularly notorious for its alpha "creep" behavior.

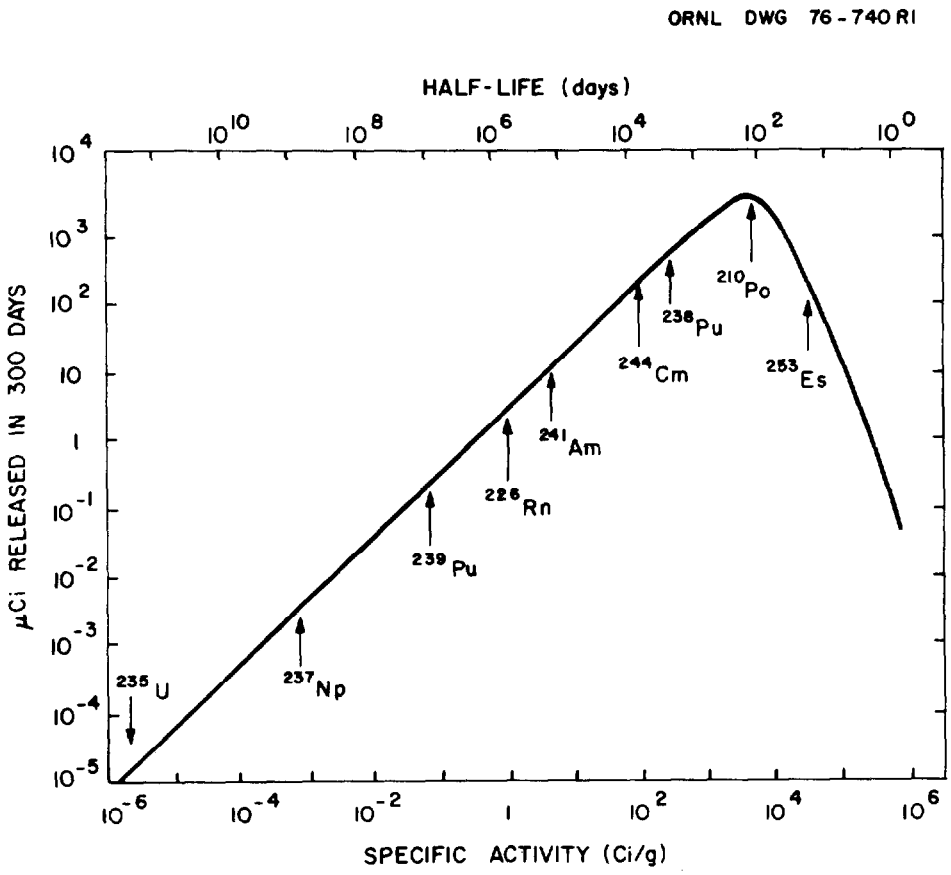

Figure 6 Calculated μCi of activity released in 300 days from 5 g each of hypothetical nuclides (physically resembling Pu), initially collected on the first of four filters, as a function of specific activity.

By appropriate application of the release equations, it is possible to calculate concentrations of airborne material released from a filtration system. Figure 7 shows a plot of the calculated air concentration of ^{238}Pu released by aggregate recoil from a series of 4 standard, 1000 CFM, HEPA filters. Two loading situations are evaluated: (1) with 0.25 Ci placed on the first filter at time zero and (2) with the load accumulated over 1 year at 0.042 Ci/month (0.25 Ci load at 6 months). Real situations would probably be included within these extremes of one-time or continuous loading. Concentrations being released at the end of one

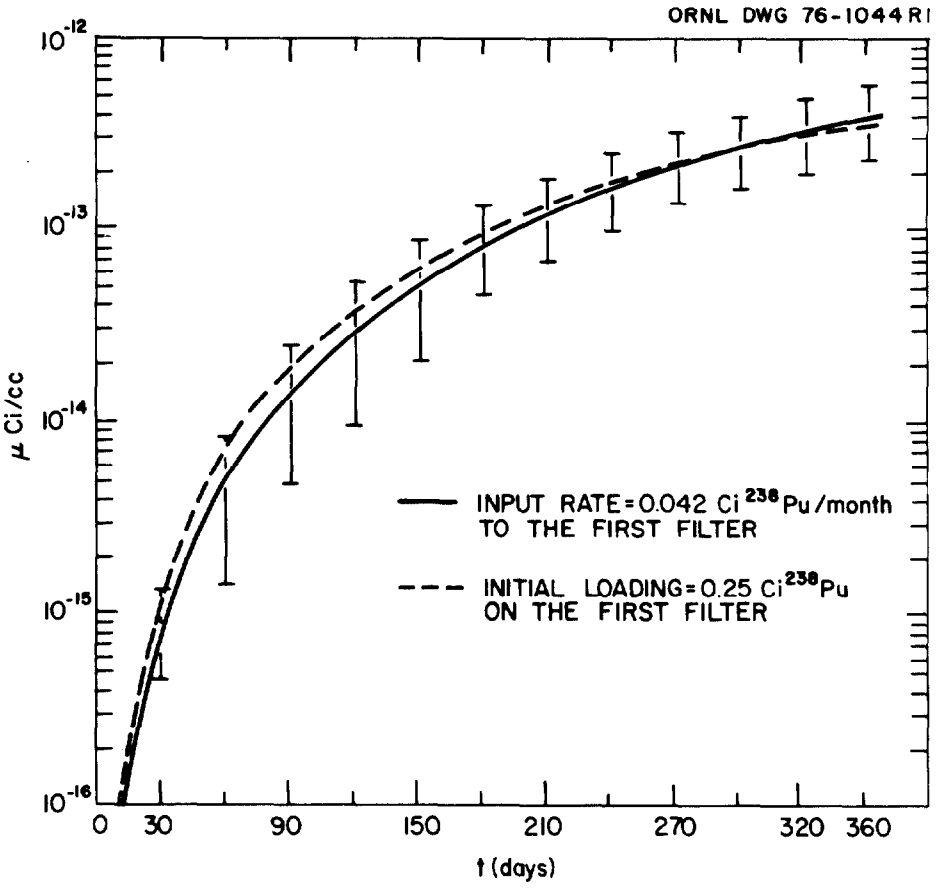

Figure 7 Calculated concentration (μCi/cc) of ^{238}Pu in air released from four 1000 CFM HEPA filters in series.

year are the same for both cases, within the accuracy of the calculations, and amount to about 3.5 x 10^{-13} μCi/cc. Preliminary work with ^{239}Pu indicates that, with loadings of the same amount of activity, releases would be similar. These concentrations are sufficiently large to warrant attention by those responsible for containment of alpha-emitting radioactive material.

Consequences and Prevention

The conclusion should not be drawn from this work that more alpha-active material has been released through air filters than was known. Exit air streams have been monitored and releases are, in general, well documented. However, there have been reports of unexplained slow increases in alpha activity in exit air from alpha processing facilities. These instances were usually attributed to development of leaks in the filter system, and new filters were installed. This work suggests, instead, that the observed increases in exit-air alpha activity were due to the aggregate recoil particle penetration of the filter system. It is thus appropriate to consider how this phenomenon might be prevented. Since the mechanism of aggregate recoil requires the residence of a significant amount of alpha activity on a filter for an extended time, any procedure that prevents long-term build-up of activity in the filter system will prevent the excessive migration

of alpha recoil particles. Tactics that immediately suggest themselves include frequent filter changes (perhaps of only the first filter, guided by a knowledge of the amount of activity on the filter), a washable filter, or precleaning of the air by some method that continuously removes particulates from the system. Many additional tests on both laboratory and engineering scale are needed to develop effective and economical methods of preventing aggregate recoil penetration of HEPA filters.

IV. Conclusions

Aggregate recoil particles from a source of alpha activity appear to penetrate HEPA filters much more effectively than would be expected on the basis of filter efficiency for similar sized stable aerosols. It is probable that an important portion of the alpha activity now released in air streams from alpha material processing facilities is due to the alpha aggregate recoil phenomenon. Significant total releases and significant exhaust air concentrations due to this mechanism are possible. Preventative measures are desirable and appear feasible.

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DISCUSSION

ETTINGER: The model you use does not include the function size. Very possibly, size is very important. We find from experimental work we've done at Los Alamos and from operating data at plutonium facilities (where the aerosols are realistic in terms of what comes out of these facilities) that we do not see the effect. It is very possible that alpha recoil is a real physical phenomenon but, in relation to the type of aerosols that come out of real plutonium situations, it is not a significant factor. Your test data indicate, at least in the text, that the plus or minus value on K is just a counting error and not an overall error based on a least squares fit to the data. Looking at the tabulated data in your report, I had a feeling that the plus or minus value might be fairly high because there seem to be no trends in counts when going from filter two to three to four. Sometimes, filter 4 had more activity and, in fact, I think the slide you showed, using an average of all of these tests, showed that filter 4 had, I think, two or three times as much activity as filter 3. I wonder if you've looked at that trend, or lack of trend, between filters 2, 3 and 4?

MCDOWELL: We've been concerned about this lack of trend. The explanation that we've come up with, which we believe is right, is that it is probably a matter of particle statistics. A very few particles could account for the activity that we're seeing on the filter. Therefore, counting statistics, in this situation, are probably more accurate than particle statistics. Whether you've got two particles or three particles on the filter could make the difference in the uncertainity in the count that you observe, whereas your count of what is there might be pretty accurate. I did not do the statistical evaluation of the K's. This was done by someone in our math division and they tell me that it is an estimate of the goodness of the fit to the equation derived from the uncertainties in counting and the scattering of the data.

STAFFORD: I wanted to reemphasize Harry's comment that we have not seen this type of decrease in efficiency much of the time when we've looked at our plutonium process exhaust filters. They are changed, generally, because of an increase in pressure drop. Reviewing our data, we have not seen a decrease in efficiency over the past several years. Did you look at the particle size distribution of your plutonium 238 aerosol?

MCDOWELL: No, we did not. In fact, we didn't generate an aerosol. Our aerosol generated itself. I'd like to reemphasize that, in all cases, we started out with a solid material which spontaneously produced particulate material that migrated and that we saw this with alpha active material and with alpha-beta-gamma active material. We know nothing about the particle size except to assume that it's similar to what had been previously mentioned with lead 212 which I spoke about.

BALSMEYER: I'm wondering if some of your statistical variation was related to the experimental setup. For instance, the distance between your filter media? Did you look at that at all?

MCDOWELL: No, we did not. We tried to look at relative humidity. That didn't seem to have any effect. The one thing that we were able to see some variation in that we had some control over was the way the source was prepared. This seemed reasonable to use because a source with a much finer particle size will have more surface area and the production of aggregate recoil particles from such a source ought to be more efficient. We don't have good data on it but there does seem to be a trend in that direction, i.e., that sources produced in such a way that they have a high surface area, produce more material traveling downstream.

BURCHSTED: I'd like to ask John Geer if Rocky Flats has anything to add to this since Rocky Flats operates systems with multiple filter banks that trap materials in this category.

GEER: I don't think we have seen anything that we would recognize as being due to the phenomena you're speaking of. Dick Woodard has done some work that indicates that there might be a few particles getting through, but we're not able to relate that to these phenomena. However, we have noted in the past that apparently there is some penetration, some slight penetration. We're trying to find a mechanism for it and we're interested in your comments.

KNOX: I'm addressing my question to Harry Ettinger. The phenomena that Jack reported here depend on time as well as on quantity. Did you have your multiple filters in test long enough to give time effects a chance to appear?

ETTINGER: I sent the data to Jack about a month ago. We sent data that showed no trend of penetration through HEPA filters that had been in place, I think, over a two year period. In addition, we did some laboratory work in which we put plutonium aerosol on a small HEPA filter, eight by eight by six inches, and put it away for approximately one year. At the end of one year, we tested it to see whether the plutonium on it had any effect. Although this is a somewhat different situation, we could see no effect. Again, the field data over a two year time span should show the effect that Jack has seen in his work.

MCDOWELL: I'd like to say that what we were observing is an effect produced by the alpha active material. It's not an effect of the filter. We don't think there's any degradation of the filter. So, putting a filter away with plutonium on it and testing it later by any kind of test should show no effect. As I understand it, the other data on penetration that came out of the operating data, involved a second filter in the system. There was a glovebox filter, a first bank and a second bank filter. Looking at the data I just showed and reflecting on this problem, it's true that what Harry had sent me showed no increase that could be attributed to alpha recoil. But, if we had looked at the second filter, we would not have seen any increase either. It's only apparent when you look at several filters. I think you'd have to have about three in series before you saw the effect, or at least three sampling stations. You can't see it just by looking across one filter.

EXHAUST FILTRATION ON GLOVEBOXES USED FOR AQUEOUS PROCESSING OF PLUTONIUM

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Abstract

The report covers information obtained during the study of ventilation of glovebox systems used for wet process operations associated with plutonium recovery. Analytical data are presented on:

- concentration of chemical components in exhaust air
- concentration of radioactive material
- chemical species deposited on, or found in, HEPA filters in the exhaust systems.

I. Introduction

Air exhausted from glovebox lines, in which plutonium is processed by "wet" chemical operations, is filtered using high efficiency particulate air (HEPA) filters to reduce the plutonium activity to a level acceptable for discharge to the atmosphere. Although HEPA filters are effective in this service, they require frequent replacement, thus adding to the volume of waste which must be handled.

An opportunity exists at the Rocky Flats Plant to observe large scale filter systems associated with chemical process operations. It is the objective of one phase of a project funded by the Energy Research & Development Administration (ERDA) to utilize plant conditions to find better ways of using HEPA filters and acquire data for development of improved HEPA filters and prefilters for chemical service. Successful achievements of these objectives would reduce the quantity of waste associated with disposal of contaminated filters.

II. Description

Figure 1 depicts the flow of air through the system used in chemical process operations. Room air enters a typical glovebox line through HEPA filters at point (1), and into the glovebox where it becomes contaminated with chemical fumes and radioactive particulate. Exhaust filters, point (2) filter out most of the particulate matter from the air leaving the box. Air and gaseous components

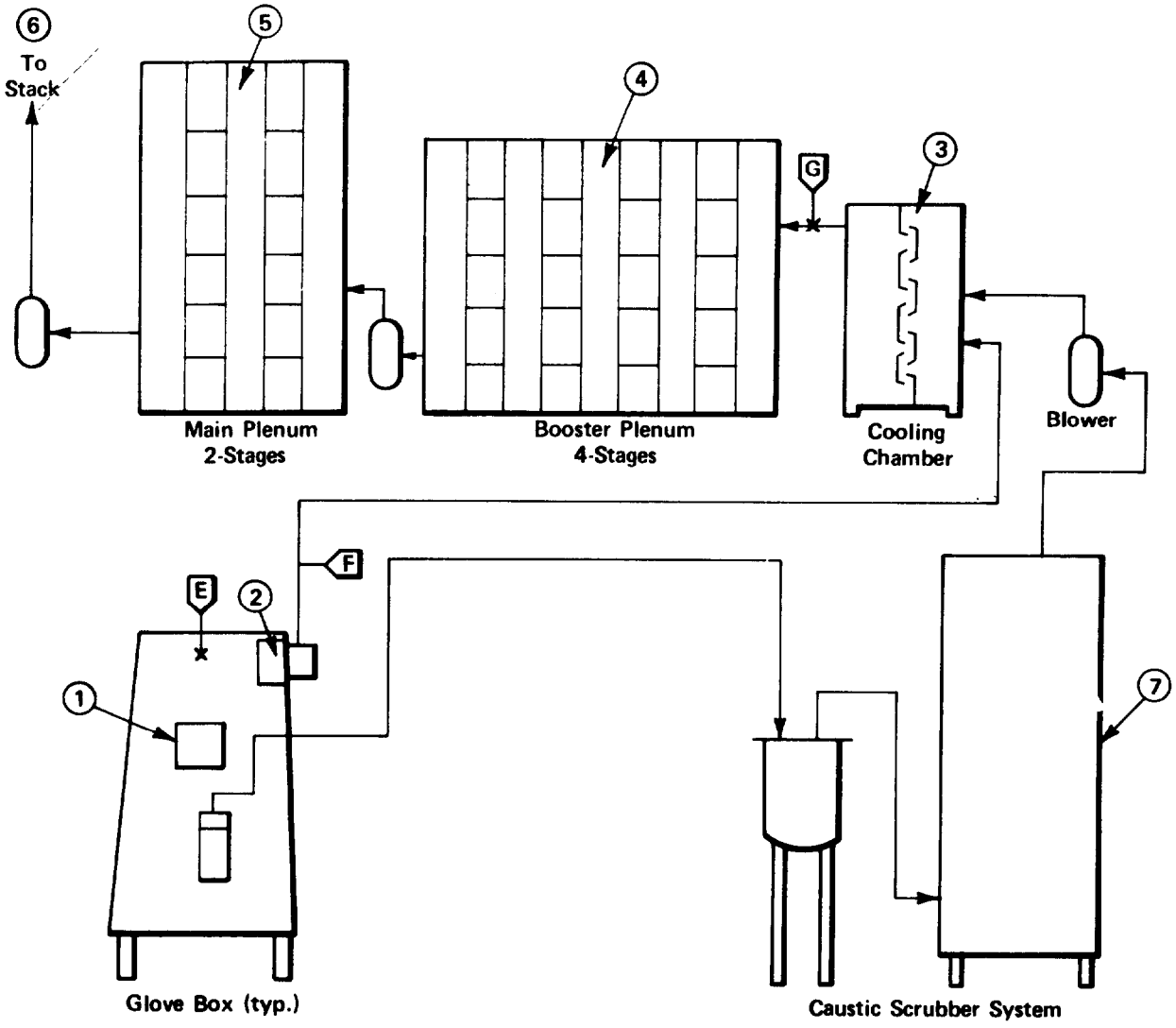

FIGURE 1
VENTILATION SYSTEM
CHEMICAL OPERATIONS

passing through the filter are collected by headers and flow through a cooling chamber (3), a multi-stage booster plenum (4), the building's main exhaust plenum (5), and then discharged through an exhaust stack (6). Some glovebox lines are provided with a supplemental exhaust system which passes the air vented from highly corrosive operations through a scrubber in which potassium hydroxide is circulated (7). The scrubbed exhaust air is then combined with the main exhaust air at the cooling chamber.

Chemical attack has been observed on all stages throughout the filter system but is most severe at the glovebox exhaust filter.

Figure 2 shows a glovebox in the chemical recycle facility. The volume of this box is 5.66 m^3 (200 $ft.^3$) and airflow is on the order of 2 m^3/min. (70 $ft.^3$/min.), thus giving about 20 air changes per hour. The process carried out in the box is leaching of solids which contain plutonium. The leach solution is 9 M nitric acid containing .1 to .2 M fluoride ion. Vapors from this mixture attack components of HEPA filters so vigorously that the filters at the box exhaust must be changed every few weeks. An example of a severely degraded glovebox exhaust filter is shown in Figure 3. Note how the media has become weak and is sluffing away from the upper edge of its frame. Collection headers, Figure 4, remove air exhausted from the glovebox lines.

A view of the caustic scrubber is shown in Figure 5. As mentioned previously, the scrubber system handles corrosive vapor drawn from processes in a limited number of boxes.

Figure 6 shows the cooling chamber. Headers which collect the exhaust from the various chemical process lines and the caustic scrubber converge at this point. The cooling chamber contains no filters but is equipped with heavy steel baffles and sprays for cooling, if required.

A 30-inch diameter line carries exhaust from the cooling chamber to a filter plenum, Figure 7, which houses four stages of HEPA filters, 30 filters per stage. Approximately 425 m^3/min. (15,000 $ft.^3$/min.) of air flow through the plenum. The 1st stage of HEPA filters is changed every two to three months. Subsequent stages in the plenum last six months to a year.

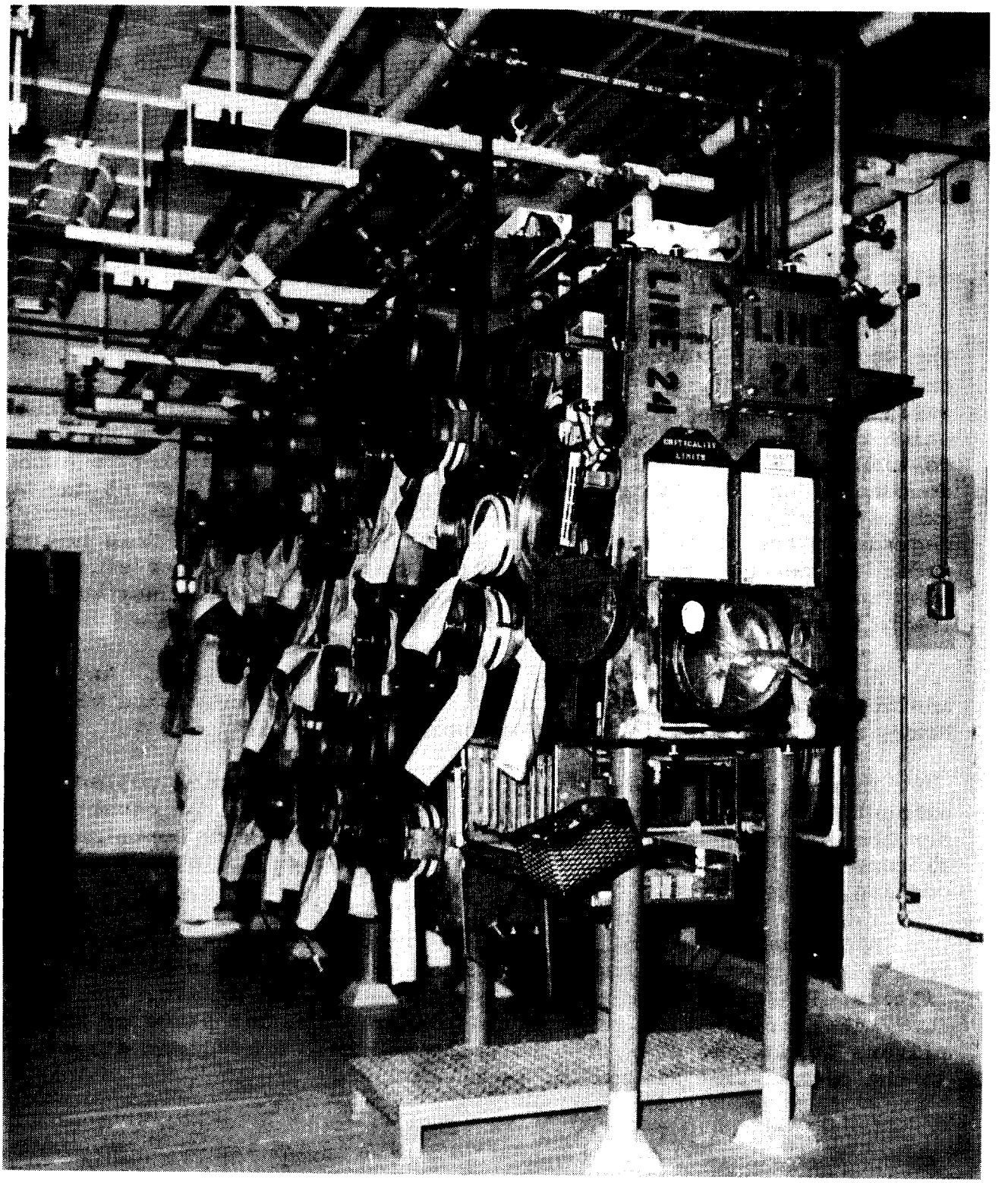

Figure 2. Glovebox Plutonium Recovery

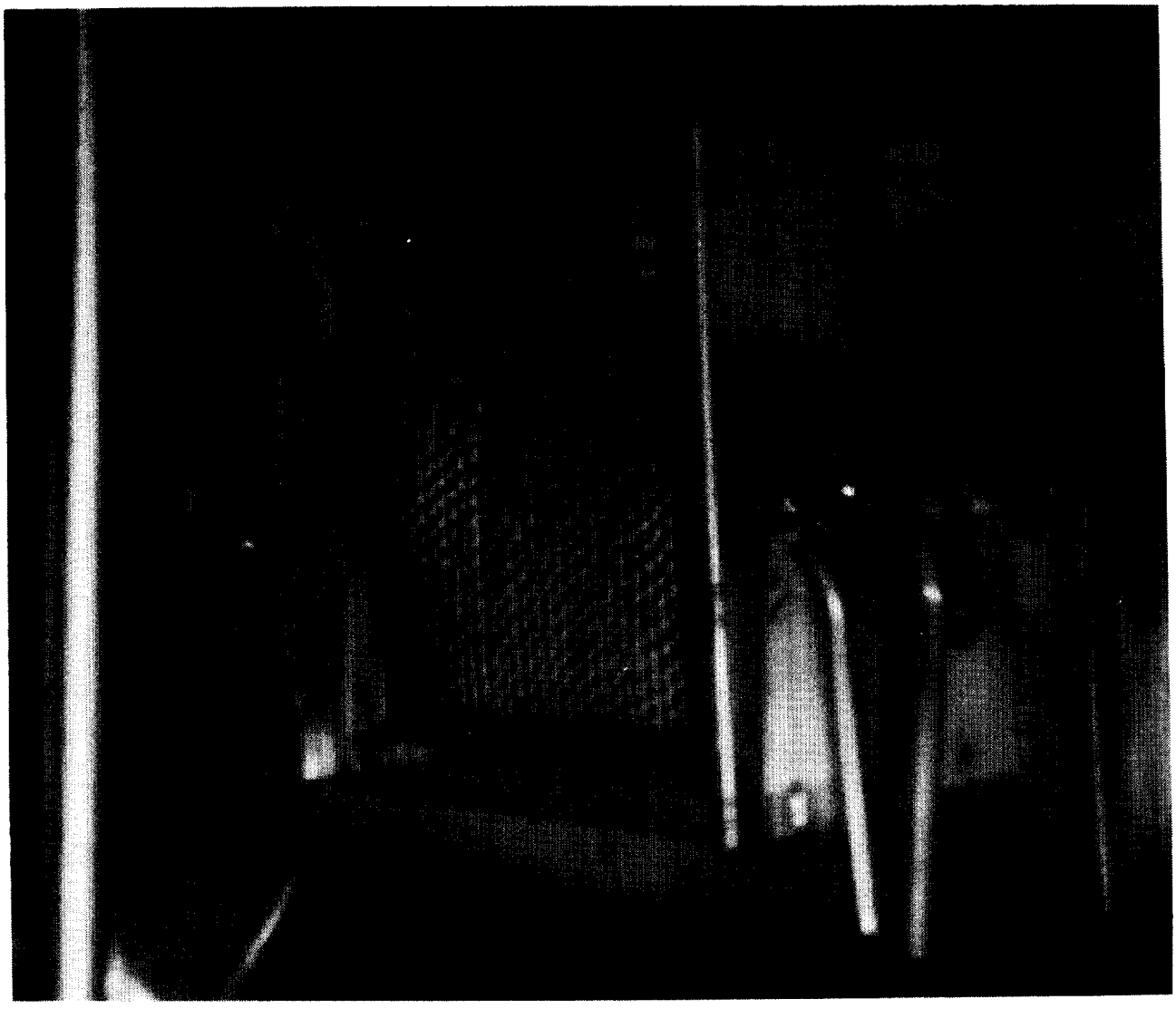

Figure 3. Glovebox Exhaust Filter
Degraded by Chemical Attack

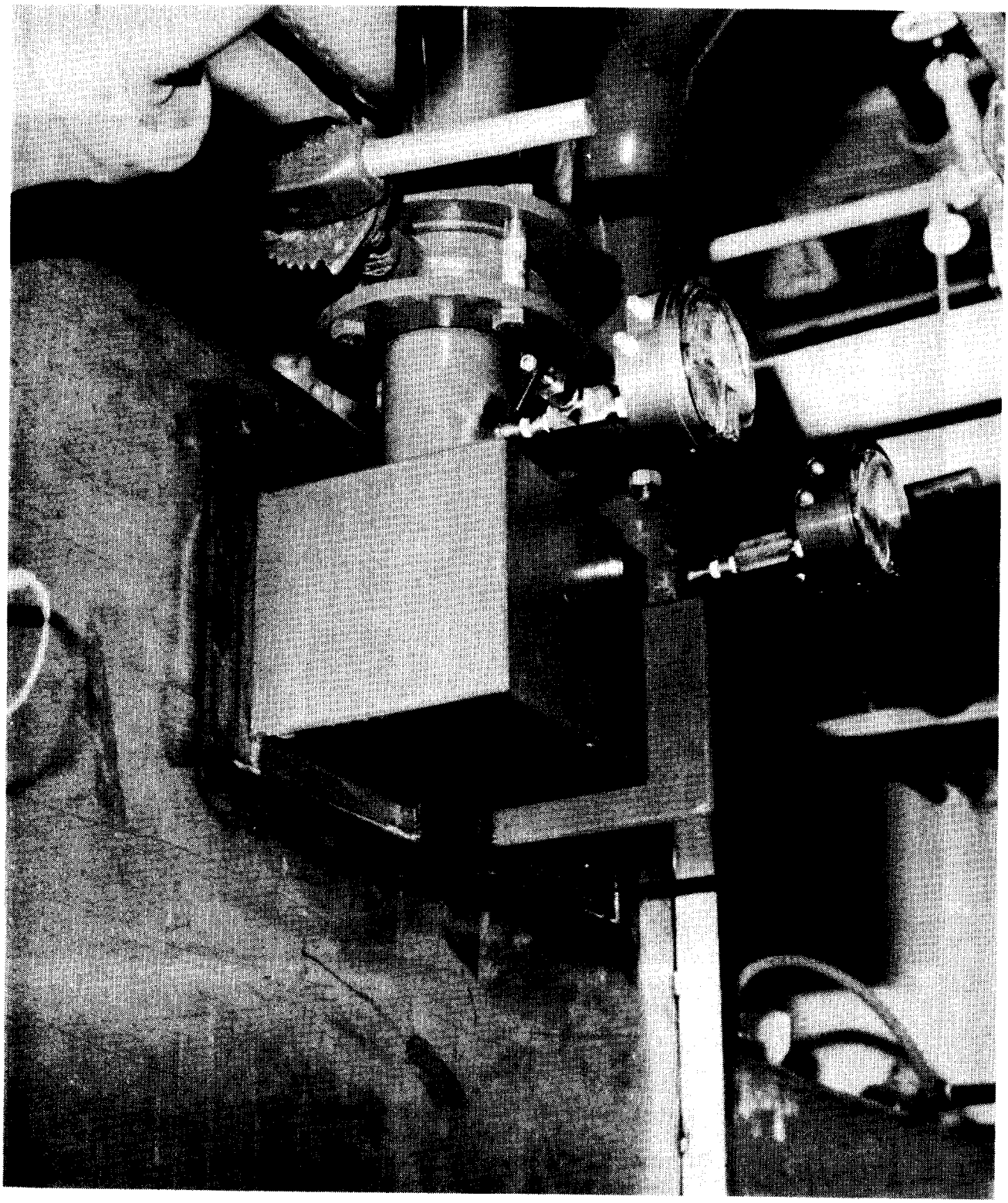

Figure 4. Collection Headers at Glovebox

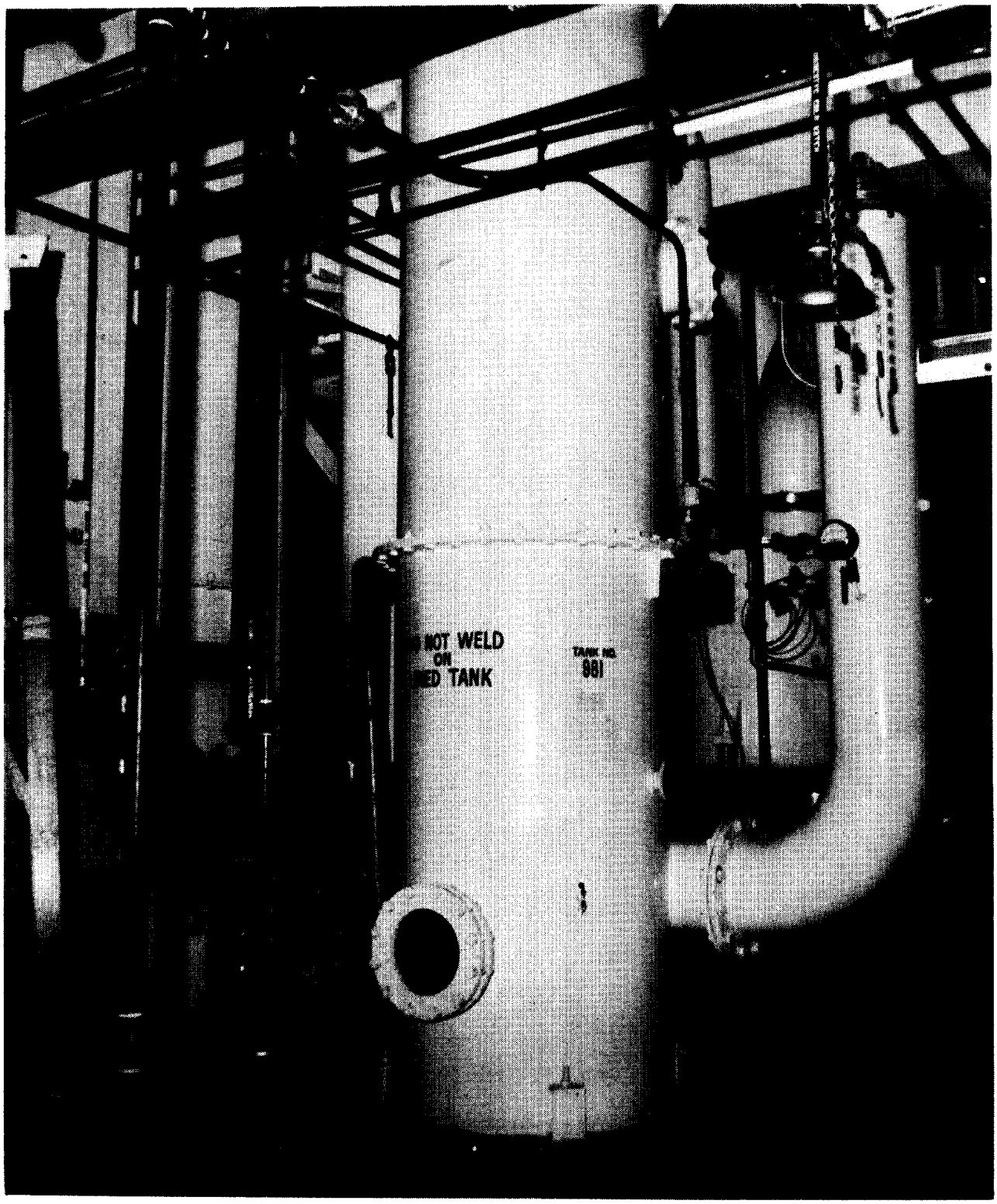

Figure 5. Caustic Scrubber

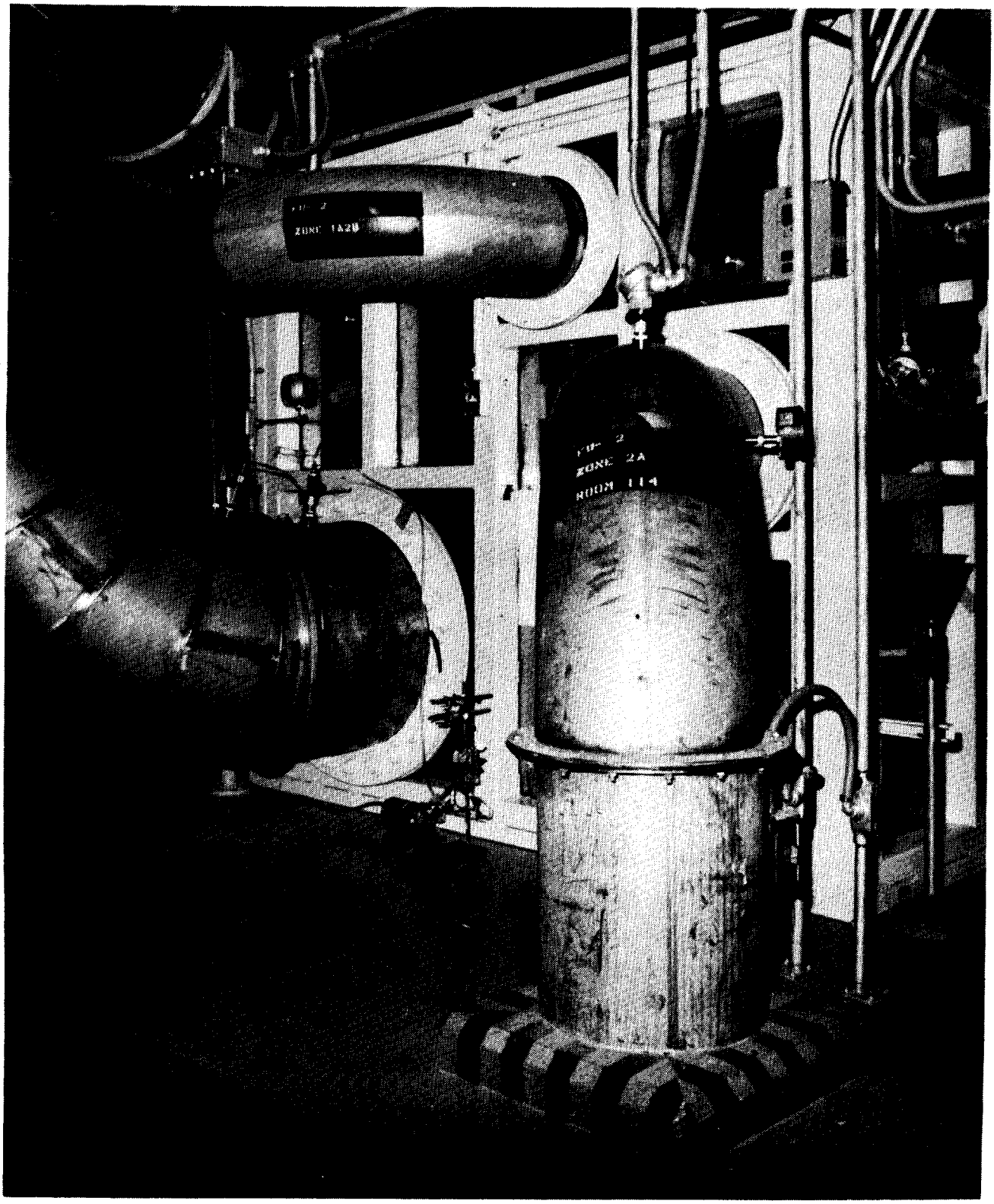

Figure 6. Cooling Chamber

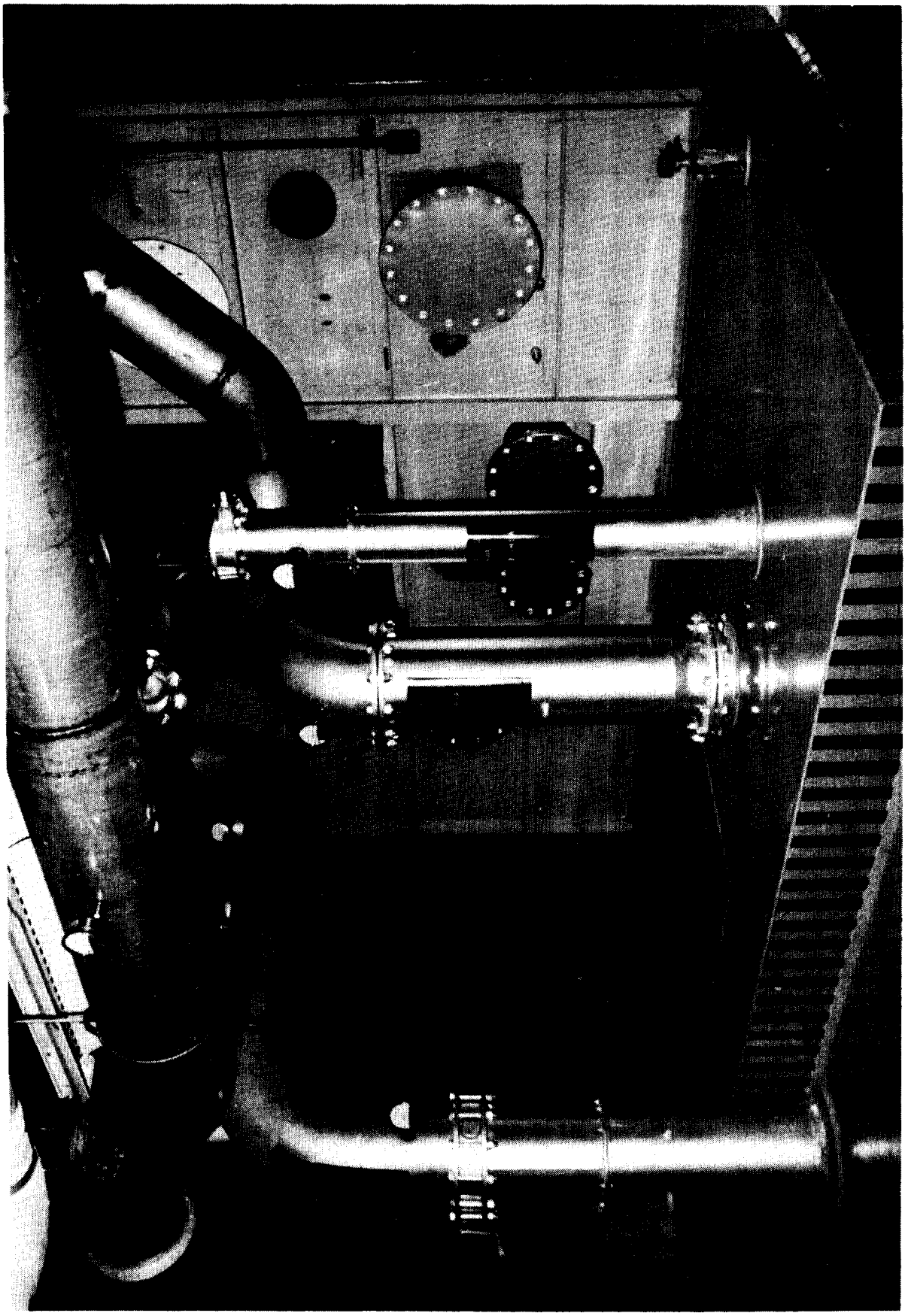

Figure 7. Collection Headers and Four-Stage Filter Plenum

Sampling of Exhaust Gases

A sampling program was begun to obtain background information on the concentration of various chemical and radioactive components of the glovebox air effluents. This program is still under way.

The sketch shown in Figure 1 also indicates the locations at which samples have been taken. Location E is a sample point in the interior of a glovebox in which plutonium is being leached from a refractory residue. Samples taken at this point would be expected to be highest in concentration of radioactive particulate and chemical components. F is a sample point located just a few feet downstream of the glovebox exhaust filter. G is located in the 30-inch diameter duct leading from the cooling chamber.

Sampling experience on radioactive particulate point E in the box, and point F downstream of a HEPA filter (Table I) shows the plutonium alpha activity observed upstream (E) and downstream (F) of the glovebox filter.

TABLE I

Alpha Activity Upstream and Downstream of Glovebox Exhaust Filter

"E" (Box) $d/m/m^3$	"F" (Header) $d/m/m^3$	Filter Eff. (%)
3.65×10^7	1.7×10^5	99.54
7.52×10^6	4.0×10^4	99.47
5.73×10^6	2.0×10^3	99.97
3.14×10^7	4.0×10^4	99.87
1.58×10^7	2.8×10^4	99.82
Average: 1.94×10^7	6.2×10^4	99.68

Membrane filters Millipore®* (.8 μ AA) were used to collect samples at points E and F.

The distribution of particle size of material taken from the membrane filters used in the glovebox atmosphere at point E was determined using a Quantimet 720 particle size analyzer which combines optical and electronic counting and sizing systems.

* Millipore® is the registered trademark of Millipore Corporation, Bedford, Mass., USA.

The distribution of particulate grouped in size intervals is shown in Table II.

TABLE II

Distribution of Pu Particulate Sampled in Glovebox (E)

Size of Interval (μ)	% of * Total
< 0.3	10.2
0.4 - 0.7	23.2
0.8 - 1.3	36.7
1.4 - 3.4	23.1
3.5 - 6.8	5.4
6.9 - 13.6	1.3
>13.7	.1

* Average of 8 determinations

A log probability plot, Figure 8, of the particle size distribution by count shows 50 percent of the total number of particles are 0.9 micrometers or less in size. Calculations show this would account for less than 0.5 percent of the total mass.

Chemical Components

Samples of air taken at the glovebox locations mentioned above were analyzed for chemical components (Table III). NO_x concentration observed in the box enclosure (point E, Figure 1) ranged from 0 to 171 parts per million (ppm) by volume. The wide range of NO_x concentration is attributed to the types of operation and level of activities in the glovebox. Exhaust air samples from the box (point F) ranged from 0 to 160 ppm NO_x.

Using liquid nitrogen to freeze condensible components, a sample taken of air in the box was calculated to contain 0.8 ppm by volume of fluoride in the box and ranged from 0.2 ppm to 2.5 ppm in the exhaust air from the box. The concentration of water varied but was usually in the range of 1500 to 3000 ppm. Acidity (H^+) found in the gas stream is attributable, in part, to nitric acid vapor.

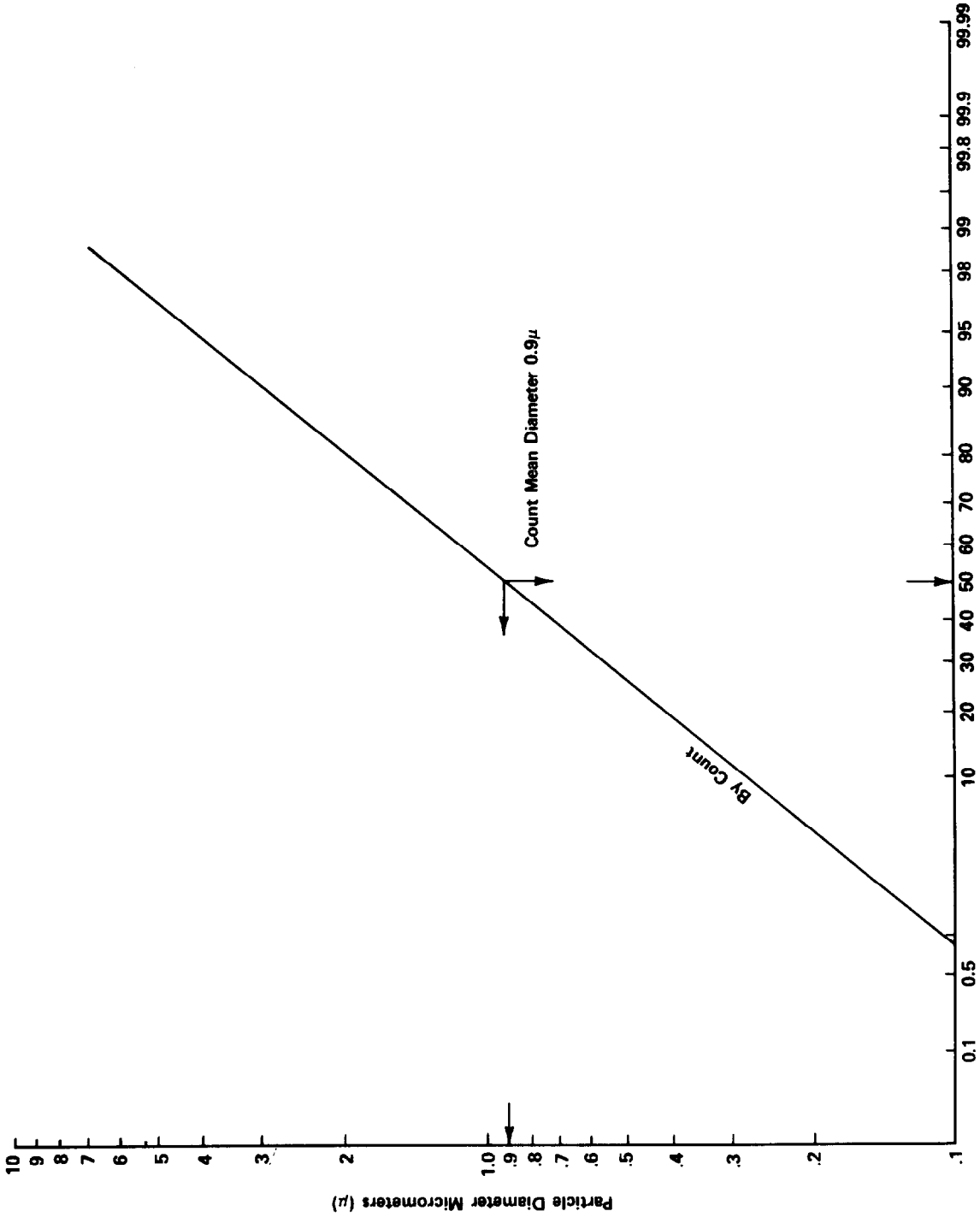

FIGURE 8. PARTICLE SIZE DISTRIBUTION

TABLE III

Concentration of NO_x, F, H_2O, H^+

Component	Vol. ppm at Sample Location *		
	In Glovebox (E)	Glovebox Exhaust (F)	Cooling Chamber Exhaust (G)
NO_x	0 - 171	0 - 160	.50 - 6.0
F	.8	.2 - 2.5	.14 - 0.6
H_2O		1500 - 3000	
H^+	1.9	4.2	

* Samples obtained over a four-month period under varied operating conditions.

Samples taken at the cooling chamber exhaust (point G, Figure 1) over a four-month period were found to be low in NO_x concentration and averaged about 1.0 ppm by volume, although readings as high as 6.0 ppm were observed. Fluoride at sample point G ranged from below the limit of detection, .14 ppm, to as high as .63 ppm. The analytical data indicate that the concentration of chemicals in the exhaust air, especially fluoride, is not high but their effect on HEPA filters is evident.

Samples of components of HEPA filters which have been in service in the booster plenum have been compared with unused component materials using a variety of analytical techniques.

Using thermogravimetric analysis and covering the temperature range to 1000°C, the volatile content of new media is about 4.0 percent (Table IV). After a filter is used in the 1st stage of the plenum, the volatiles increase to about 18 percent after a two and one-half month exposure. Volatiles on the 3rd stage media measured 4 percent even after an exposure of ten months. Volatiles in new asbestos base separator material are usually in the 25 percent range. After two and one-half months of use the volatile content measured in a 1st stage sample is close to 50 percent and after ten months exposure in the 3rd stage, increased to 61 percent.

TABLE IV

Volatile Content of Filter Components

Percent Weight Loss to 1000°C

Sample	New	1st Stage 2 1/2 Mo.	3rd Stage 10 Mo.
Glass Media	4	18	4
Asbestos Separator	25	50	61

The data indicate that filter media in the 1st stage adsorbed volatiles, including H_2O, HF, and HNO_3. These components would also react with particulate matter trapped in the media. Volatile content of the 3rd stage media showed minimal difference from unexposed media. In contrast, the weight of volatiles in the separator increased with length of service - even in the 3rd stage, and indicates material in the separator is reacting with gaseous component in the exhaust airstream.

Hot water leach of weighed filter samples were examined for acid and leachable ions using specific ion electrodes (Table V). New media was found to have a pH of 8.9, fluoride <.06 percent, and nitrate .09 percent. Leach solution of media exposed in the 1st stage of the filter plenum gave a pH of 3.9, fluoride 2.2 percent, and nitrate 9.8 percent. Media exposed in the 3rd stage of the filter plenum for ten months gave a pH of 3.15, fluoride .45 percent, and nitrate 1.6 percent.

Leaches performed on samples of filter separator material taken from the 1st stage filter after two and one-half months service, yielded a pH of 3.9, fluoride .22 percent, and nitrate 19.6 percent. A separator sample, after ten months exposure in the 3rd stage, yielded a pH of 3.4, fluoride .24 percent, and nitrate 36.5 percent. The soluble substance leached from the separator was identified to be predominately magnesium nitrate, along with soluble silicate(s).

TABLE V

Water Leach of Filter Components

Sample	New	1st Stage Filter After 2 1/2 Mo.	3rd Stage Filter After 10 Mo.
Media:			
pH	8.90	3.90	3.15
F	.06%*	2.20%	.45%
NO_3	.09%	9.80%	1.60%
Separator:			
pH		3.9	3.4
F		.22%	.24%
NO_3		19.60%	36.50%

* Weight percent based on weight of sample leached

The leach data correlate with the thermogravimetric analysis. It is of special interest that the separator in the 3rd stage had adsorbed more nitrate than that from a 1st stage sample, indicating continued reaction during the exposure period.

Mass Spectral outgas of media and separator samples provide another means of observing chemical effects on these materials (Table VI). With new filter media only small amounts of moisture, carbon dioxide, carbon monoxide, etc., were observed. On 1st stage filter media in service for two and one-half months, the major substances which outgased included silicon tetrafluoride, water, nitrogen oxides; minor components included hydrogen fluoride, ammonia, and carbon dioxide. Third stage filter media in service ten months outgased water as the major component and ammonia, silicon tetrafluoride, and nitrogen oxides in minor concentrations.

Outgas data taken on samples from new separators gave only water as a major component. Used separators from the 1st and 3rd stages gave major components: water, nitrogen oxides, and carbon dioxide with minor amounts of silicon tetrafluoride.

TABLE VI

Mass Spectral Outgas of Filter Components

Sample Source		Outgas Components	
Filter Media	- New	Major:	None
		Trace:	H_2O, CO, CO_2, Hydrocarbons
Separator	- New	Major:	CO_2, CO, H_2O
		Trace:	CH_3OH, CH_2O, Binder Decomp.
Filter Media	- Used in 1st Stage	Major:	SiF_4, H_2O, NO, CO
		Minor:	HF, NH_3, CO_2
Separator	- Used in 1st Stage	Major:	H_2O, NO, N_2O, CO_2
		Minor:	SiF_4
		Trace:	HF, Hydrocarbons, HCl, HNO_3
Filter Media	- Used in 3rd Stage	Major:	H_2O, NH_3, SiF_4, N_2O, NO, DOP
		Minor:	NO_2, HCl, CO, CO_2, Fluorosiloxanes
Separator	- Used in 3rd Stage	Major:	H_2O, NO_2, NO, N_2O, CO
		Trace:	SO_2, Hydrocarbons

III. Summary

In summary, the trends noted by the analytical work indicate that fluoride in the exhaust gas preferentially adsorbs on the glass filter media whereas the nitrate preferentially reacts with the chemicals associated with the asbestos separator. The partitioning is more pronounced in the 3rd stage filters where there is less deposition of particulate matter in the media. The concentration of fluoride in 3rd stage filter components is lower than on the 1st stage, even after longer exposure, suggesting depletion of fluoride as it passes through a number of stages of filter media.

The cumulative effect of fluoride compounds in the degradation of filters is undoubtedly quite complex. The properties of glass fibers may be greatly affected by alteration of the oxygen-bridged

polymeric silicate ions $(Si_xO_y)^n$ or formation of stable compounds such as CaF_2, SiF_4, and NaF.

The take-up of nitrate in media is relatively high only on the 1st stage filters where it combines with, or is a part of, the particulate matter being filtered. Nitrate is heavily absorbed by the asbestos separators in filters of all stages.

The sampling program will be continued as new components are developed and operational changes are made. An area that will receive special attention is at the glovebox line, the source of chemical and particulate contaminants.

It is intended to explore the use of prefilters at the glovebox exhaust of a type which are resistant to acid fumes and take out the larger particulate which should contain most of the plutonium. A cleanable prefilter would be attractive from both the standpoint of plutonium economy and reduction of the numbers of filters discarded.

Since asbestos base separators react with nitric acid vapor and oxides of nitrogen, attention will be given to variations of separator composition.

Acknowledgement

Determinations on the size of plutonium particulate were performed by J. K. Fraser of the Radiochemistry Laboratory at the Rocky Flats Plant.

ENTRAINMENT SEPARATOR PERFORMANCE

M.W. First and D. Leith
Harvard Air Cleaning Laboratory

Abstract

Clean and dust-loaded ACS entrainment separators mounted upstream of HEPA filters were exposed to a combination of fine water mist and steam at about 70°C from one to four hours. In every trial, the ACS entrainment separator prevented measureable deterioration of performance in the following HEPA filter. Droplet size-efficiency evaluation of the ACS entrainment separators showed that, within the accuracy of the measurements, they meet all service requirements and are fully equal to the best separator units available for service on pressurized water reactors.

I. Introduction

A loss of coolant accident in a PRW nuclear reactor imposes severe conditions on the air cleaning elements that comprise an important part of the engineered safeguards system. A major source of air cleaning system stress would be the very large amounts of condensed water in the form of mist and droplets that would be entrained with the containment vessel air, released fission products, and uncondensed steam. This condensate could flood the particulate filters and activated charcoal adsorption units in the absence of efficient liquid water separation devices. The detrimental effects of large amounts of condensed steam on particulate filters and activated charcoal adsorption units was recognized many years ago, and remedial measures have included increasing the water repellency and steam resistance of absolute filter papers (1) as well as the introduction of entrainment separators (also called moisture separators and mist eliminators) upstream of the principal elements of the air cleaning train.

Peters (2) investigated the mist separation characteristics of full-scale 2-in. thick mats of "Teflon" yarn wrapped over stainless steel reinforcing wire* when exposed for 10 days to a simulated loss of coolant atmosphere and found that these units were capable of preventing failure of downstream absolute filters while maintaining flow at or close to design values. Rivers and Trinkle (3) described a moisture separator that was developed for the Connecticut Yankee Atomic Power Plant and found to be capable of protecting downstream absolute filters for a minimum of 24-hours when subjected to design flow rates of a "saturated air-steam mixtures at pressures up to 40 psi and 261°F, the maximum predicted conditions." These moisture separators consisted of "three type M-105 phenolic-bonded glass fiber pads" mounted in a frame approximately 8-in. deep and placed downstream of a set of louvers.** MSAR 71-45 (4) described

*York Separator, Style 321. Otto H. York Co.
** American Air Filter Co., Louisville, Ky.

tests of five commercially available entrainment moisture separators and found that two of these units AAF Type T** and MSA Type G † were capable of removing greater than 99% of droplets in the 1-10 μm size range when handling a simulated PWR postaccident atmosphere at rated flow rate for a number of hours. The MSA Type G Separator is 5-in. deep and consists of multiple layers of 9 μm glass fiber and 0.006-in. diameter knitted wire pads.

The present study was undertaken to evaluate the usefulness of a new commercially available entrainment separator for service in PRW power reactor engineered safeguards systems.

II. Test Program

Ten separate trials were conducted in each of which an unused 1,000 CFM nominal capacity filter was protected by a new ACS entrainment separator of equal air flow rating located upstream of it. In each, the combination was subjected for a prolonged period to a high concentration of water droplets in the 1-10 μm diameter range suspended in a saturated steam-air mixture at about 70°C. Droplet concentration measurements were made upstream and downstream of each entrainment separator and droplet collection efficiency determinations were made for four important size ranges. Three new absolute filter and new separator pairs were tested for periods of one hour and three identical pairs were tested for periods of four hours each. Four additional pairs of new elements were tested for four hours after having been loaded with Cottrell-precipitated fly ash.

Before and after exposure to the droplet laden steam-air mixture for the prescribed length of time, the efficiency of each absolute filter was checked at an airflow rate of 600, 1000, and 1600 cfm using a dioctylphthalate (DOP) test aerosol having a mass median diameter of 0.6 μm and a geometric standard deviation of 1.65. In every case, ACS entrainment separators protected downstream absolute filters to the degree that no decrease in efficiency could be detected after prolonged exposure to the droplet-steam aerosol.

III. Test Facilities

A dimensioned schematic of the test facility designed for this program is shown in Figure 1. The steam-air mixture entered the filter and entrainment separator housing though a rectangular section where additional steam and water droplets were injected and where a thermocouple was located to record the temperature of the aerosol as it reached the entrainment separator. At the entrainment separator and absolute filter, the housing cross section enlarged from 20-in. square to 24-in. square to provide sumps for water drainage. Between separator and filter there was a 5-ft. long section equipped with Plexiglas view port.

**American Air Filter Co., Louisville, KY.
† Mine Safety Appliances Co., Pittsburgh, PA.

Downstream of the filter, there was an 8-in. diameter duct with a Venturi meter for measuring flow rate, a flow regulator, and a Buffalo Forge Co. centrifugal blower with a 48-in. diameter impeller and 25 HP motor, capable of moving 2000 cfm of air at 40-in. w.g. After the blower, the flow entered a switch that made it possible to send the flow to the waste air system or back to the test section for recirculation. The recirculation mode was used whenever elevated temperature steam-air-droplet trials were conducted but the entire stream was exhausted to the waste air system whenever filters were tested with DOP. All surfaces of the test apparatus except the duct leading from the switch to the waste air system were heat insulated with 3.5 in. of commercial fiberglas blanket covered with aluminum foil.

Each entrainment separator and absolute filter was mounted firmly in the test tunnel against 1-in.flat-ground flanges. Gasket material 1/8-in. thick, self-adhesive closed sponge rubber, was applied to the flanges to assure a good seal. The absolute filters were equipped by the manufacturer with Neoprene sponge gaskets. Each separator or filter was held firmly in position by eight clamps. In every case, before exposing separator or filter to the droplet and steam mixture, an in-place filter test was performed with DOP to demonstrate that the separator and filter were mounted satisfactorily.

Steam was injected in the system at 3 psi through a 3/4-in. pipe located upstream of the separator. A dense fog of water droplets was generated by a bank of 39 Spraying Systems Co.* 1/4 J nozzles. These are two-fluid atomizing nozzles that required compressed air as well as water. They were operated at a manifold pressure of 7.5 psi to the compressed air side and a water flow of 2.2 lpm. Air pressure was measured with a pressure gauge mounted on the inlet air manifold and water flow rate was measured with a rotameter located just upstream of the inlet liquid manifold. Figure 2 shows the bank of 39 nozzles as they were located in the injection section. During the steam-droplet tests, the aerosol was recirculated but, because injection of steam, water, and compressed air was continuous, it was necessary to bleed off a fraction of the gas stream to the laboratory waste air system to avoid overpressurizing the test facility.

Water drains were provided at four points along the housing as shown in Figure 1; one drain was upstream and one was downstream of the entrainment separator, other drains were upstream and downstream of the filter. The rate at which water drained from the sections upstream and downstream of the separator was measured using a graduated container and stopwatch. The liquid flow from the drains on either side of the absolute filter were so low that they could only be measured by noting the total amount of liquid collected over the course of a four-hour run.

* Spraying Systems Co., North Avenue at Schmale Road, Wheaton, IL.

Pressure taps were located up and downstream of the entrainment separator and absolute filter to measure pressure drop across these units and across the Venturi flowmeter to measure system flow. Pressure taps were connected to manometers to provide continuous visual observation of pressure drop at these three points in the system. In addition, the pressure taps were connected through a sequential switching system to a pressure transducer that made it possible to record pressures on a strip chart. Figure 3 is a photograph of the pressure drop and temperature monitoring instruments and strip chart recorder. The temperature in the test section was measured with an iron-constantin thermocouple pair. One side was placed in the injection section and the other was placed in an ice water bath. The thermocouple was recalibrated with boiling water prior to each separator test and the output was continuously monitored and recorded on a strip chart. All data pertaining to pressure drop, temperature, water flowrate, and compressed air were recorded in a lab book at ten minute intervals; water drain flowrates were recorded each twenty minutes.

Droplet concentration and size measurements were made with 4-stage May-Cassella cascade impactors located inside the test housing upstream and downstream of the entrainment separator. Each of the four impactor stages was fitted with a glass slide coated with freshly generated magnesium oxide (MgO) deposited by passing the slide through the fume rising from a burning magnesium ribbon. When the test atmosphere was drawn through the impactor, water droplets present in the gas deposited on successive stages according to decreasing droplet size. When a droplet hits the coated slide, a crater forms in the soft, smooth magnesium oxide that has the same diameter as the droplet. Crater diameters were measured under the optical microscope to establish droplet size parameters. During the sampling, the impactor was mounted inside the moisture eliminator-filter housing and operated to sample isokinetically. The characteristic droplet diameters collected on each stage were found to be: stage 1, 29 μm; stage 2, 12.5 μm; stage 3, 5.3 μm; stage 4, 3.1 μm.

A. ACS Entrainment Separator

The moisture separators used in these tests were manufactured by ACS Industries, Inc., Woonsocket, R.I. and designed specifically for this application. The manufacturer's description of the separators is as follows:

"They are composed of a 5-1/2" thick pad of knitted mesh enclosed in a welded frame of stainless steel sheet metal. The mesh is manufactured using a parallel knitted style composed of .006" T304 SS and multifilament fiberglass. The knitted composite mesh is crimped and the pad is constructed by building up layers of mesh, in such a way as to obtain a pre-determined density. Interspersed through the pad thickness are a number of layers of plain wire mesh to assist in the removal of entrained liquid from the interior of the mesh pad.

"The frame consists of a single length of 16 gage, T304 SS sheet formed with a 3/4" lip on both edges and finally formed into a 24" square. The pre-assembled mesh pad is inserted into the frame which

is closed and heliarc welded at one corner. Square cross grids made of 1/8" T304 SS rod on 5" centers are welded into place on both faces of the mesh pad. Drain holes are drilled in one side of the square frame and the unit is marked to indicate that the holes are on the bottom when it is installed. On the top two rows of horizontal grid members on both faces, 3-1 1/2" long pieces of 1/8" T304 SS rod are welded on each grid rod at an upangle (6 per side) to act as prongs to insure that the mesh pad doesn't settle or pack down and pull away from the top of the frame when installed upright.

"These units are designated as ACS Model 101-55." A drawing reflecting the above description is shown in Figure 4.

B. Absolute Filters

Catalog No. 7C83-L, size F filters were purchased from Flanders Filters, Inc. Face dimensions of these units were 24-in.square and depth, 11 1/2-in. Filters were fabricated from glass fiber paper, aluminum foil separators, plastic foam sealant, and double flanged chromatized steel frame. Rated efficiency for homogeneous 0.3 μm DOP is not less than 99.97%. Rated pressure drop is not more than 1.0 in. w.g. at 1000 cfm. Manufacturer's data, stamped on each filter, are listed in Table 1.

IV. Test Program

Ten individual tests were made with all-new moisture separator-filter pairs. Tests 1, 2, and 3 were each of one hour duration and were conducted with clean, new separators and filters. Tests 4, 5, and 6 were each of four hours duration and were conducted with clean, new separators and filters. Tests 7, 8, 9, and 10 were each conducted with new separators and filters which first had been loaded with dust. Each of these latter tests was of four hours duration. In test 7, the moisture separator-filter pair was loaded in tandem with 29 pounds of dust. For tests 8, 9, and 10, the separator-filter pairs were loaded in tandem with one pound of dust. The dust used for loading the moisture separators and filters prior to steam-water droplet testing was seived Cottrell-precipitated pulverized coal fly ash having a count median diameter of 0.6 μm and a geometric standard deviation of 3.0. This prepared coal fly ash test dust is equivalent to NBS dust without the addition of small amounts of cotton linters and carbon black that are sometimes added to shorten test time and to improve the readability of the discoloration papers. As neither addition serves any useful purpose for, or influences the results of the tests performed in this study, they were omitted.

After inserting a separator and absolute filter into the test tunnel, a DOP in-place filter test, carried out in conformity with the methods recommended in ORNL-NSIC-65 (5) was performed on the separator and on the filter at air flow rates of 600, 1000, and 1600 cfm to establish that the unit was undamaged and that there was no leakage around the mounting flanges. A TDA*, seven-nozzle hetero-

*Air Techniques, Inc., Baltimore, MD.

geneous (cold) DOP generator, operated at 30 psi, was used. Upstream and downstream DOP concentrations were measured with a TDA-2D* photometer. When leakage was found, the filter or separator affected was reseated the filter mounting clamps, retightened and additional DOP tests performed until it was established that no leakage occurred. Beakers were placed in the housing at distances of 1, 2, 3, and 4 feet downstream of the entrainment separator to indicate the rate at which droplets or condensation reached the floor of the dropout section at various distances downstream of the separator.

A run was started by feeding steam to the recirculation loop and water and air to the spray nozzles. The temperature inside the test section was monitored and the test period started after the unit reached a stable equilibrium temperature of about 70°C (15 to 30 minutes).

Water and airflow rates to the spray nozzles and flow rate through the entire system were carefully monitored and adjusted whenever necessary. Generally, once equilibrium conditions were established, these flows were stable. Visual observation of droplet carryover downstream of the separator was attempted during each trial but, because of the steam, the Plexiglas observation port was always fogged making meaningful observations impossible. Near the end of each trial, cascade impactor samples were taken downstream of the separator for droplet size analysis. At the conclusion of the trial, the water, air, and steam feeds were discontinued immediately and the water level in the four droplet collecting beakers on the floor of the tunnel was measured and recorded.

The efficiency of the absolute filter after exposure to the steam-droplet-air atmosphere was checked by performing once again the prescribed DOP in-place filter test at 600, 1000, and 1600 cfm. The test section was then opened, the separator and filter removed and examined visually.

The number concentration of droplets in each size range produced by the 39 nozzle spray manifold was calculated from single nozzle tests by applying a suitable multiplication factor. The results are given in Table 2. Droplet concentrations downstream of the entrainment separator and ahead of the absolute filter were measured at the conclusion of each test. After the size of droplets collected on each impactor stage had been determined, it was only necessary to count the number of droplets present on each stage to develop the downstream droplet size distribution and calculate droplet collection efficiency.

V. Test Results

Table 3 summarizes temperature and pressure drop measurements for each of the ten trials. Temperature and pressure drop were stable during all tests for both clean and dirty separators and filters.

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*Air Techniques, Inc. Baltimore, MD.

Table 4 summarizes water drainage measurements for each of the ten trials. Close to 99.9% came from the drains before and after the entrainment separator. Table 5 shows the results of in-place filter tests performed before and after each of the trials for entrainment separators and absolute filters. No degradation in filtering ability of the filters was observed after exposure to a steam-droplet atmosphere in any of the tests. In all cases, the entrainment separator successfully protected the absolute filter from damage.

Table 6 is a summary of droplet size vs collection efficiency data for the moisture separators determined from analysis of cascade impactor data. The results are plotted in Figure 5 with the curve for the MSA entrainment separator. They show that the collection efficiency vs droplet diameter relationship for the ACS entrainment separator is identical, within experimental reliability, with that of the MSA separator.

After each trial, between 1.5 and 2 ml of water was found in each beaker placed on the floor of the rectangular test section between moisture eliminator and filter regardless of beaker location or duration of test. This demonstrates that few large droplets were blown off the downstream face of the moisture eliminator.

VI. Summary and Conclusions

Ten ACS entrainment separators have been tested for their ability to protect absolute filters from damage when exposed to a droplet laden steam atmosphere similar to that expected during an accident at a PWR. The test aerosol contained high concentrations of water droplets in the 1 to 10 μm diameter range at temperatures in the vicinity of 70°C. Pressure was atmospheric.

In all cases, the separators protected the absolute filters from damage on the basis of acceptable in-place DOP filter tests conducted at 600, 1000, and 1600 cfm. The droplet diameter vs collection efficiency characteristics of the ACS entrainment separators were found to be comparable to those of the MSA entrainment separator, as reported in Reference 4.

The demister units that were tested met "qualification requirements similar to those found in MSAR 71-45 [4] and, as a consequence, may be presumed to be acceptable to the Nuclear Regulatory Commission staff as defined in Regulatory Guide 1.52[6].

Table 1 Absolute filters used for tests.

Test #	Serial #	Factory Measurements Airflow Resistance at 1000 cfm in. w.g.	Homogeneous DOP % Penetration
1	A445098	0.74	0.004
2	A445100	0.72	0.006
3	A445095	0.86	0.002
4	A445099	0.78	0.010
5	A445104	0.80	0.002
6	A445097	0.70	0.006
7	A445105	0.80	0.004
8	A445096	0.76	0.006
9	A445102	0.80	0.002
10	A445103	0.78	0.005

Table 2 Number concentration of droplets from a 39-Nozzle bank of Spraying Systems 1/4-J nozzles.

Stage Droplet Diameter micrometers	Number Concentration #/cm^3 of air
29.0	646
12.5	661
5.3	121
3.1	91

Table 3 Temperature and pressure drop at test conditions.

Test	Temp.°C	Pressure Drop, in. w.g. Separator	Filter
1	55	0.80	0.90
2	66	1.02	0.92
3	56	0.87	1.05
4	75	1.05	0.95
5	66	0.91	0.96
6	74	0.94	0.96
7	69	2.60	1.40
8	72	1.13	0.93
9	73	0.92	0.92
10	73	0.87	0.87
Average *		0.95± 0.10	0.94± 0.05

± = standard deviation
* Excluding test 7, in which 29 pounds of dust were fed to the separator and filter. All pressure drop data taken at 1000 cfm.

Table 4 Water drain rates.

Test	Separator Upstream	Separator Downstream	Filter Upstream	Filter Downstream
1	-- no data --		-- no data --	
2	1.77 lpm	0.335 lpm	0.0014 lpm	0
3	1.74	0.265	0.0011	0
4	1.90	0.340	0.00074	0.00015
5	1.99	0.139	0.00098	0.00023
6	1.93	0.087	0.00106	0.00031
7	1.90	0.035	0.00083	0.00008
8	1.93	0.160	0.00375	0.00052
9	1.93	0.093	-- no data --	
10	1.97	0.056	0.00478	0.00042
Average:	1.90 lpm	0.17 1 pm	0.0018 lpm	0.00028 lpm
% of total drained	99.9%		0.11%	

Table 5 DOP penetration for separators and absolute filters before and after steam and droplet exposure.

Test	Separator, %		Absolute Filter, %	
	Before	After	Before	After
1	80-90	100	<0.01	<0.01
2	98-100	87-100	<0.01	<0.01
3	90-100	98-100	<0.01	<0.01
4	95-96	93-97	<0.01	<0.01
5	93-97	94-98	<0.01	<0.01
6	93-95	93-95	<0.01	<0.01
7	95-97	93-95	<0.01	<0.01
8	90-93	90-95	<0.01	<0.01
9	78-87	92-97	<0.01	<0.01
10	84-90	90-100	<0.01	<0.01

Table 6 Droplet collection efficiency of entrainment separators.

Test	Droplet diameter, μm 29	12.5	5.3	3.1	
1	---------- no data ----------				
2	99.87%	99.95%	99.88%	99.99%	
3	99.89%	99.98%	99.93%	99.92%	
4	99.99%	99.96%	99.72%	99.91%	
5	99.92%	99.67%	99.20%	99.85%	
6	99.78%	99.67%	98.51%	99.91%	
7	99.79%	99.67%	97.58%	99.63%	
8	99.75%	99.91%	99.36%	99.46%	
9	99.30%	99.72%	---- no data-----		
(10 -no data-)	99.79%	99.82%	99.17%	99.81%	Average

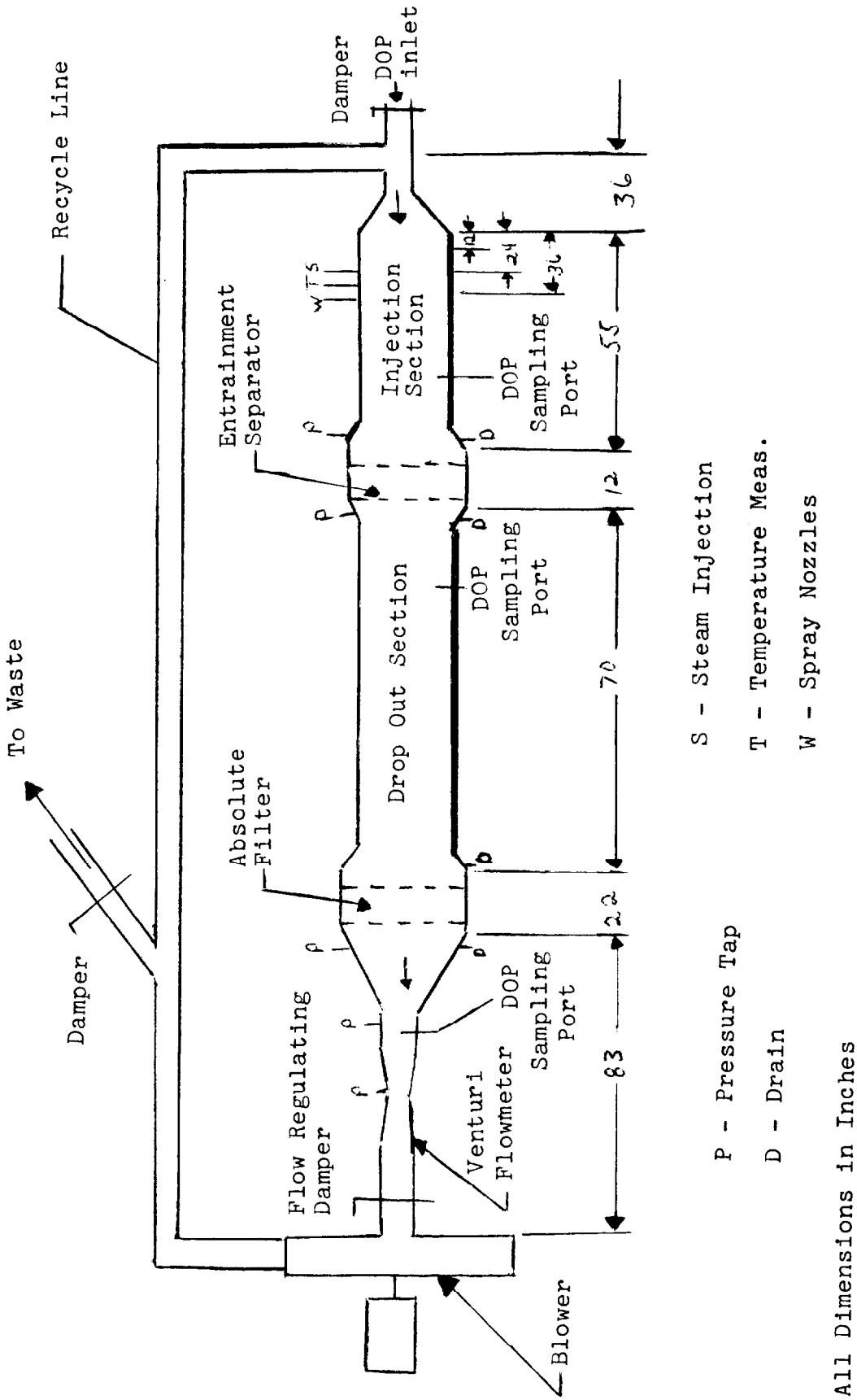

FIGURE 1 SKETCH OF APPARATUS

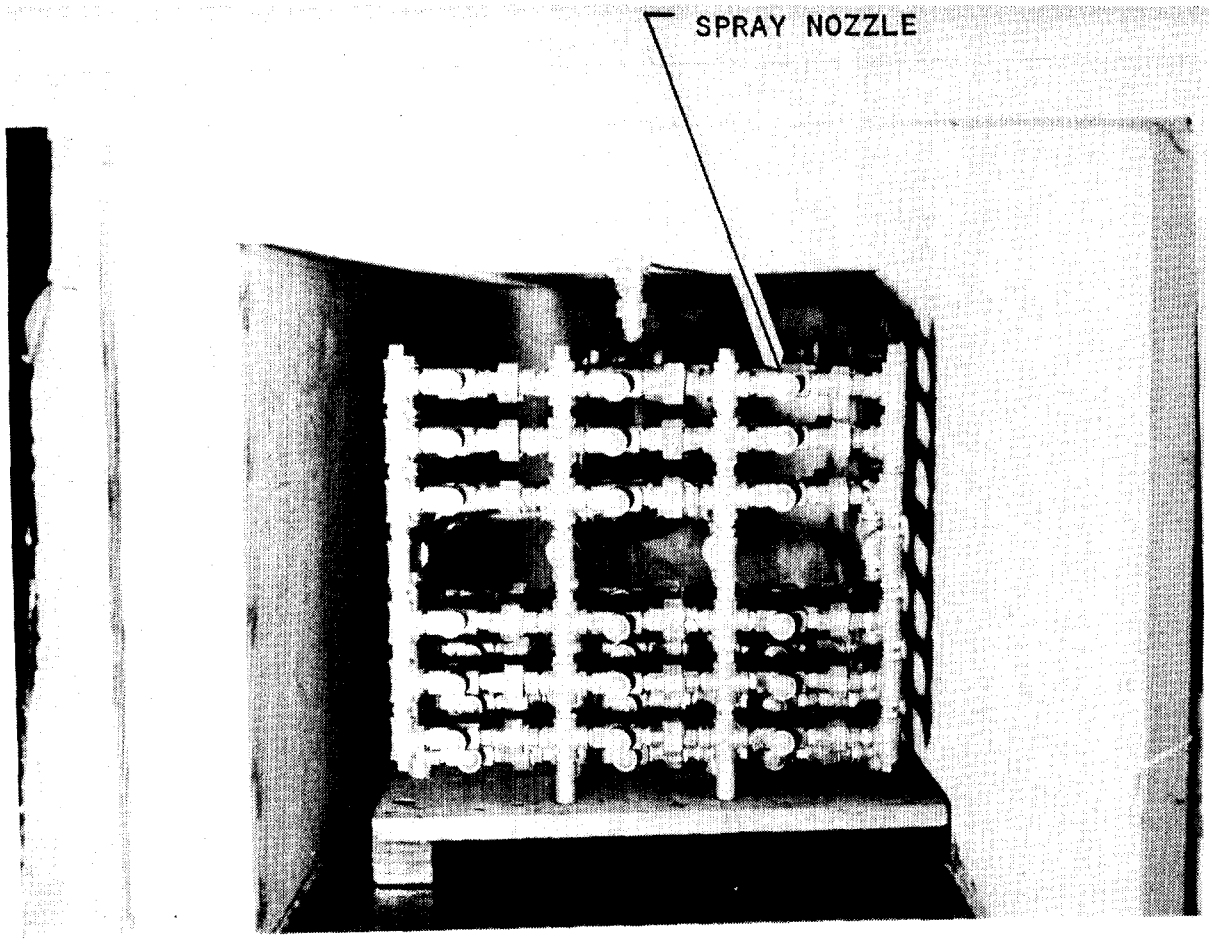

FIGURE 2

PHOTOGRAPH OF NOZZLES, INTERIOR OF INJECTION SECTION

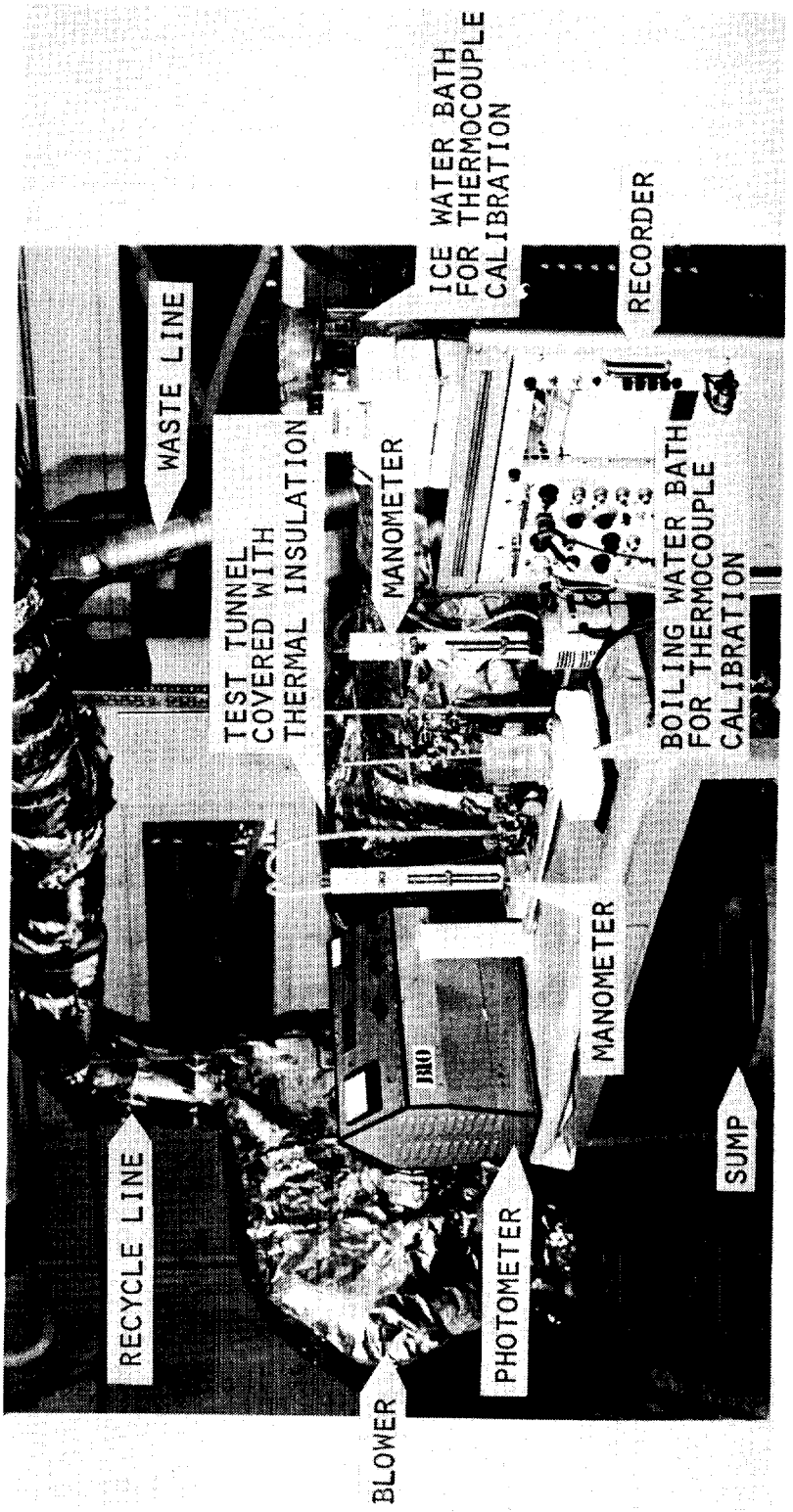

FIGURE 3

PHOTOGRAPH OF EQUIPMENT USED

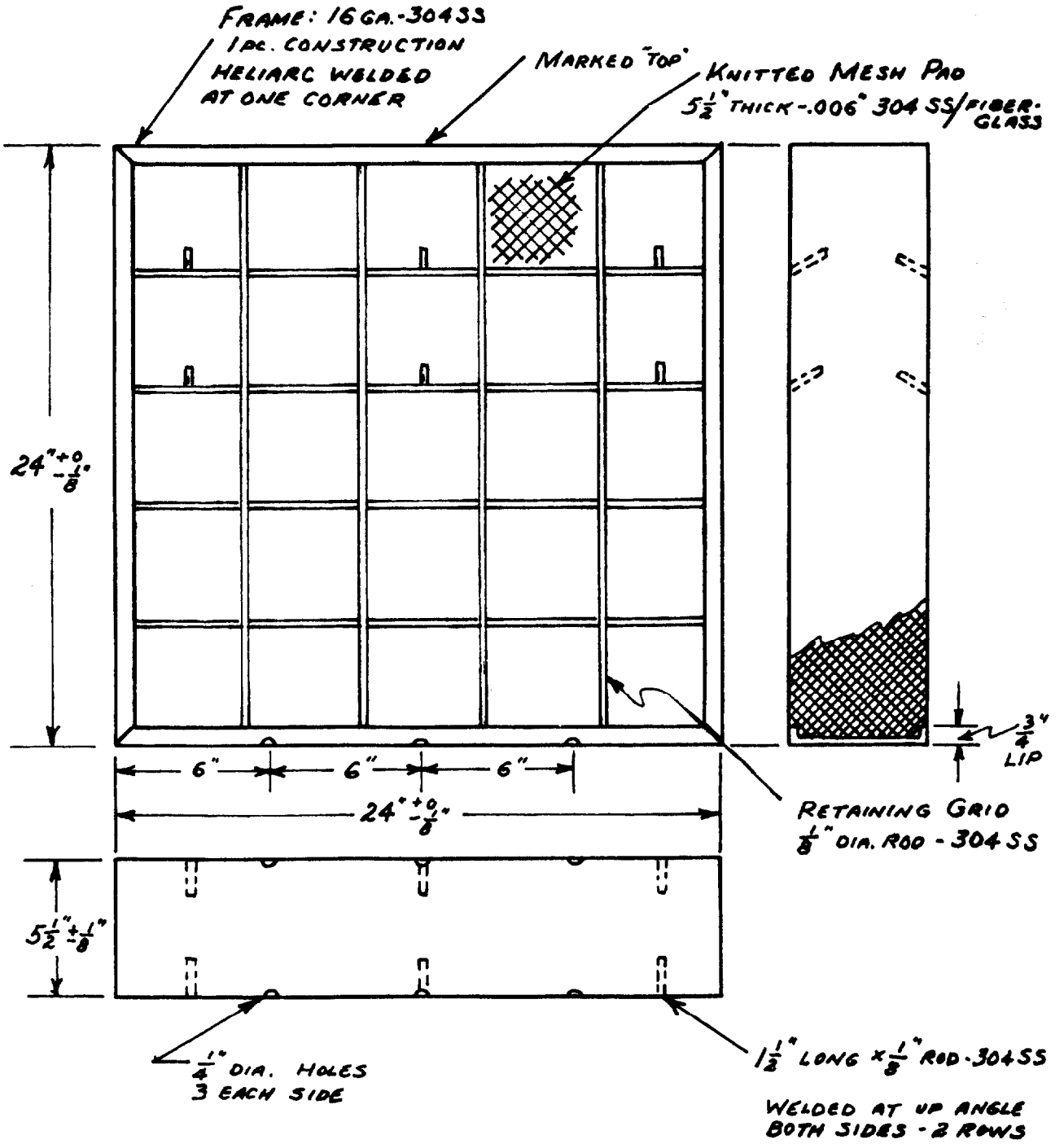

FIGURE 4 ACS ENTRAINMENT SEPARATOR

ACS INDUSTRIES, INC.
WOONSOCKET, RHODE ISLAND

Date JAN '75	Customer		Cust. Part No.
Scale 1" = 1'	Material AS NOTED	Part Name MOISTURE SEPARATOR	Part No. MODEL 101-55
Dr. By J.L.			

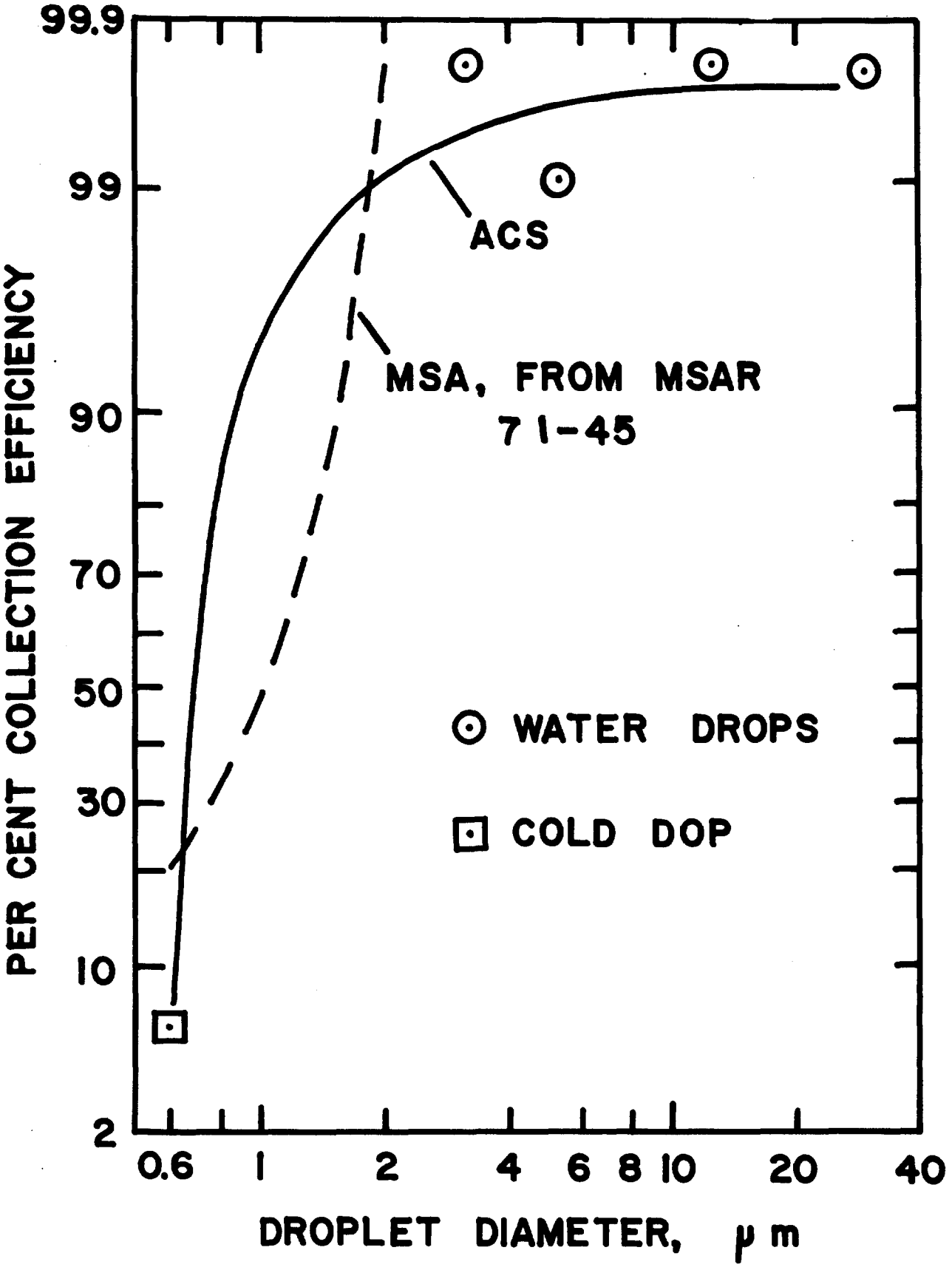

FIGURE 5 PER CENT COLLECTION EFFICIENCY VS DROPLET DIAMETER, MSA AND ACS ENTRAINMENT SEPARATORS

References

1. MIL-F-51079 B,"Filter Medium, Fire-Resistant, High-Efficiency," Military Specification, 4 April 1974.

2. Peters, A.H., Application of Moisture Separators and Particulate Filters in Reactor Containment, DP-812, Office of Tech. Services, U.S. Dept. of Commerce, Washington D.C., Dec. 1962.

3. Rivers, R.D. and Trinkle, J.L., Moisture Separator Study, NYO-3250 -6, American Air Filter Co., Inc., Louisville, Ky., June 1966.

4. MSAR 71-45, Entrained Moisture Separators for Fine Particle Water-Air-Steam Service: Performance, Development and Status, MSA Research Corp., Evans City, PA., March 1971.

5. Burchsted, C.A., Fuller, A.B., and Kahn, J.E., Nuclear Air Cleaning Handbook, ERDA 76-21 (ORNL NSIC 65, Rev. 1), Energy Research and Development Administration, Washington, D.C., 1976.

6. Regulatory Guide 1.52, Design, Testing, and Maintenance Criteria for Engineered-Safety-Feature Atmosphere Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants, USNRC.

DISCUSSION

GEER: Based on the MSA report, we are going to try the MSA filter as a prefilter to the poor one you saw in Woodard's paper. We hope to have it installed soon so we will have some operating data in a couple of months.

T. T. ALLAN: What is the purpose of the 7-ft distance between the entrainment separator and the HEPA filter as shown in the apparatus diagram? If the face velocity through the entrainment separator is evenly distributed, can this distance be reduced? By how much? This is important because of the limited space requirements in many installations.

FIRST: The 7-ft distance between entrainment separator and HEPA filter was chosen because that distance was used for the qualification tests cited in Reference 4. Obviously, this is not a very informative answer. The effect of this distance is to permit very large droplets that may accumulate and become detached from the downstream side of the entrainment separator to fall to the sump by gravity before reaching the HEPA filter. I don't know by how much this distance can be reduced because it has not been tested, to the best of my knowledge. It seems reasonable to conclude that the greater the distance between entrainment separator and HEPA filters, the better.

MARBLE: How can this test qualify a separator to meet "no visible downstream plume" when report says "windows fogged so we could not observe downstream conditions?"

FIRST: The windows fogged because they were not covered with thermal insulation ahd the inner surfaces were below the dew point when tests were conducted with warm, saturated air on the inside. When tests were conducted at ambient temperature, no visible downstream plume was observed even when air saturation was assured by recirculating the air through the spray banks in a closed system.

MARBLE: How did you determine that saturation was obtained, or was it only close?

FIRST: Steam was added upstream of the spray nozzles to heat and saturate the air. During passage through the spray nozzle banks, the temperature of the air was lower below its dew point and condensation occurred. This assured saturation as there was a small degree of cooling in the remainder of the test tunnel.

MARBLE: Do you have pressure rise data on the HEPA filters?

FIRST: These are reported in the paper. Briefly, pressure rise was insignificant in all cases.

GOVERNMENT-INDUSTRY MEETING ON FILTERS, MEDIA, AND MEDIA TESTING

W. L. Anderson
Naval Surface Weapons Center
Dahlgren, Virginia

An informal working group has been responsible for many of the accomplishments achieved thus far in the high efficiency filtration programs of ERDA. Originally constituted at the 7th Air Cleaning Conference, the group consisted of a handful of individuals meeting in a night session at a local motel to discuss current problems in a "bull-session" format. At the 10th Conference in New York, the group was officially recognized with Mr. Gilbert as chairman. Attendance has increased from the 32 who attended the organizational meeting to a level of about 60; this level has been maintained at the last several sessions by limiting attendance to invitation only. At the 13th Conference in San Francisco, leadership of the group was transferred from Mr. Gilbert to Dr. First; this change had been requested by Mr. Gilbert following his retirement from ERDA.

The most recent session of the working group was held this past Sunday afternoon (Aug 1) and was devoted to a series of discussions on subjects of current interest. It was the first meeting under the new leadership but followed the precedent of earlier meetings in that it related to the operating requirements placed on 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 problems of the particulate filter, its components and test methods. A prepared agenda listed six items with discussion leaders identified for each. The individual leaders were extremely well prepared and all items were discussed, evaluated, and resolved with dispatch.

At this the 14th Conference in Sun Valley, Idaho, a total of 63 persons comprised the assembled body; although the majority were from industry, representatives from government, government contractors, academic institutions, and four foreign nations were in attendance. Included were representatives of all the industrial complex; from the specifiers, the basic component suppliers, the unit manufacturers, and finally the end item users. 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.

It is my intent to review for you, in abstract form, the items of the committee deliberations. The items will be discussed in the order of their agenda listing. In accordance with the new format, prepared papers were reviewed and the intimate discussions and deliberations were conspicious by their absence.

1. Standardization of Test Methodology. This subject was reviewed for the group by William Heyse, Dexter Corporation, Windsor Locks, Conn.

Standards and instruction manuals are lacking in specific details to assure that comparable testing can be accomplished at all sites whether they be production control, quality assurance, and/or specification conformance. This is particularly true in resistance, penetration, test flow, and physical testing of both the media and the filter unit. The previous utilization of primary standards, round-robin testing, and/or referee laboratory evaluation to assure uniformity in testing was abandoned when the Naval Research Laboratory terminated their filter development programs in 1971. Attendees agreed that test methods and procedures should be updated and that some form of program be developed for coordinating testing at the concerned laboratories.

a. Recommendation. The Q127 Test Manual be reviewed and revised to bring it up to present day standards. Specific ASTM, ANSI, TAPPI, and other standards should be specified so that all groups are adhering to the same test conditions. The position/desirability of a referee laboratory be determined; if accepted, a laboratory should be so identified and begin to function as a point of contact and a single source of data comparison.

b. Action. The chairman will appoint a Ad Hoc group to up grade the test procedures, examine the referee lab position, and screen potential candidates for the final selection of the referee lab.

2. Microfiber Fiber Diameter Determination. The discussion of this subject was presented by Clifford Cain, Johns-Manville (JM), Denver, Colorado.

Glass fiber diameters have previously been determined by the Williams Freeness Test that was established and calibrated by the Naval Research Laboratory in the early 1950's. The original curve was considered valid for fibers down to just less than 1 micron diameter but in later years was extrapolated to include fibers as low as 0.1 micron diameter. Information developed over the years indicated that the calibration curve might be in error over this extrapolated portion. Mr. Cain reported on his findings that this reported fiber diameter discrepancy was a fact and that corrections should be made in the previously reported size ranges 0.05 to 0.75 micron diameter; simply stated these lower fiber diameters were actually larger than stated in the suppliers literature. R&D efforts to show effects of temperature, pH, dispersion and physical test techniques on the Williams method were presented. Preliminary evidence was also obtained to show a technique for estimation of meaningful fiber size distributions. A new Williams fiber diameter calibration curve was obtained and verified by surface area (BET) techniques.

a. Recommdnation. JM shall adopt the new calibration curve and use it for determining future fiber diameters. An effort should be made by JM to correlate the new calibration techniques to production facilities in an attempt to produce more uniform and reproducible fiber lots. The experimental data presented should be documented so that it is available for other research groups.

b. Action. JM will issue new sales literature stating the correct fiber diameters for codes 100, 102, 104, and 106. The data presented will be assembled in a manuscript form and offered to a technical journal for publication.

3. Fiber Migration from HEPA Filters. Discussions on this subject were presented by C. D. Skaats, Rockwell International, Golden, Colorado.

Industrial hygiene and medical authorities have been concerned about the release of glass fibers from HEPA filter units supplying air to ventilation systems. This concern was amplified when medical evidence indicated that glass fibers deposited in human lungs were potential sites for carcinoma development. Attendees were in agreement that binder additives to the media would enhance fiber retention and under standard operating conditions, no hazard should exist in their use. In the absence of existing experimental data, Mr. Skaats and Dr. Leineweber of JM reported on the operation of a typical system and their attempts to detect fiber migration from the HEPA unit. Sampling was conducted for 6 hours using the Nucleopore collection techniques with optical and electron microscopy for fiber identification. Since no fibers could be found on their samples, they concluded that no fibers were released from the HEPA units.

a. Recommendation. Attendees should endorse the results of the experiment and distribute the information to their customers and fellow workers. The results should be documented and published, perhaps as a letter to the editor. Some quantitative definition of the word no should be determined. Sensitivity of detection stated as fiber count per sample volume should be determined.

b. Action. Mr. Skaats will review the physical parameters of the experiment and report back on the quantitative value of fiber detection.

4. Qualified Products List (QPL) for HEPA Units. Max Negler of Edgewood Arsenal, Maryland presented the findings in this area.

After two years of testing the QPL for HEPA filter units is now available. Manufacturers have submitted samples for qualification to Edgewood, tests have now been completed, and certification is now imminent. It should be pointed out that the long time delay was at least partially attributed to a discrepancy of radiation test results

between Edgewood and Savannah River. Technical problems in determining radiation exposure conditions resulted in a reworking of the test conditions and a restructuring of the ultimate results. Since a total of five manufacturers had each submitted samples in 7 different categories, a comprehensive listing of acceptance or rejection was not possible. Some suppliers had all products accepted, some had partial acceptance, and some failed all qualifications. Contact with the various unit fabricators will be necessary before one can determine which of their units are listed on the qualified products roster.

a. Recommendation. QPL listings as presented be accepted and appropriate letters of qualification issued to the producers. It was further recommended that QPL testing be continued and those producers who failed to qualify on their first effort be encouraged to resubmit updated units.

b. Action. Edgewood issue 1st phase QPL approval/status letters to industry. Filter Media samples and/or units be resubmitted as soon as possible for follow-on QPL testing.

5. Draft Mil. Specs. on HEPA Media and Units. Discussion leader for revision of the Mil Specs. was Mr. Max Negler of Edgewood, Md.

Mil Specs F-51068 and F-51079 are under revision to include all changes made to date. The filter specification (51068) revisions are relatively minor and include physical and marking changes as well as inclusion of QPL requirements. The media specification (51079) was revised to a greater extent. The acidity determination was eliminated and media tensile strengths were lowered in several areas. The number and size of test specimens was also changed. In addition, provision for a QPL will be included in the filter medium specification, and the SPEC will be expanded to cover two grades of media. Grade A will meet all requirements of 51079; Grade B will meet all requirements of the specification except gramma irradiation testing, and will not have a minimum thickness requirement.

a. Recommendation. The revisions be adopted and the revised Mil. Spec. be issued as soon as possible.

b. Action. ERDA review the proposed tensile changes and decide whether the adopted values are adequate for their requirements. ERDA should issue a revised Bulletin 306 stating the requirement changes they deem necessary.

6. DOP Toxicity During Man Testing. Sam Steinberg of ATI, Baltimore, Maryland reviewed the present status of the subject.

Inquiries into the toxicity of DOP when used for man-testing of protective facepieces have increased over the recent past; many of these have originated from commercial sources. Since no maximum allowable concentrations (MAC) have been established for this specific compound, some concern exists over its relative toxicity. A search of the literature indicates that DOP is non-toxic and attendees agreed that in their opinion no hazard exists in the proposed use. Statements from a toxicity expert (Stokinger) has resulted in a similar opinion that this class of material is not considered to be toxic and as such no special MAC is required. After considerable discussion it was decided that the assembled group was not qualified to state an official position on the subject; NIH or NIOSH would be the appropriate government agency to issue such a position statement.

a. Recommendation. A task group be formed to solicit and collect information from a variety of sources (medical, toxicologists, producers) on the toxicity levels of DOP. This information would be submitted to the government agency responsible for establishment of MAC's; a request would be made for an acceptance level that could be applied to man test conditions.

b. Action. The chairman will appoint necessary membership for the task group to carry out the recommendations.

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 absolute filtration industry. The total effort has proven invaluable because it permits the surfacing and review 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.

CLOSING REMARKS OF SESSION CHAIRMAN: (C. A. Burchsted)

We had a number of papers on electrical effects and a number of papers on sand. Don Orth of Savannah River discussed the advantages and problems of selecting sand for sand filters at the Savannah River plant.

Mr. Schuster discussed the operation of electrical field air cleaners and Mr. Reid spoke about static collection on filter beds. This was followed by Gary Nelson who talked about electrostatic collection of filter beds and the electronic experiments at Livermore Laboratory.

Chuck Skaats reviewed his experiences with three filter manufacturers and discussed construction problems. I think his discussion demonstrates clearly that qualification tests of a filter are no substitute for the one-by-one, quality assurance testing that ERDA conducts for the Commission and makes available to commercial users on a cost basis.

After that, Charlie Gunn compared the performance of a number of HEPA filters of different construction under very adverse conditions. We don't see such conditions very often in real situations, but they can occur and are very important factors in the qualification of filters.

Jack McDowell gave the most controversial paper on the list. If there are questions that anyone would like to discuss with Jack on the alpha mechanism, I would suggest they do so.

Lastly, Mr. Woodard on chemical operations, Dr. First on the testing of demisters, and Anderson's summary of the filter meeting.