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PERFORMANCE OF MULTIPLE HEPA FILTERS AGAINST PLUTONIUM AEROSOLS

by

Manuel Gonzales, John C. Elder, Marvin I. Tillery,
and Harry J. Ettinger

ABSTRACT

Performance of multiple stages of high-efficiency particulate air (HEPA) filters has been verified against plutonium aerosols similar in size characteristics to those challenging the air-cleaning systems of plutonium-processing facilities. An experimental program was conducted to test each filter in systems of three HEPA filters operated in series against $^{239}\text{PuO}_2$ aerosols as high as 3.3×10^{10} dis/s·m³ in activity concentration and ranging from 0.22 μm to 1.6 μm in activity median aerodynamic diameter (amad). Mean penetration (ratio of downstream to upstream concentration) of each of the three filters in series was below 0.0002, but it apparently increased at each successive filter. Penetration vs size measurements showed that maximum penetration of $^{239}\text{PuO}_2$ occurred for sizes between 0.4- and 0.7-μm aerodynamic diameter (D_{ae}). HEPA filter penetration at half of rated flow differed little from full-flow penetration.

I. INTRODUCTION

Multiple stages of high-efficiency particulate air (HEPA) filters are used at most Energy Research and Development Administration (ERDA) nuclear facilities to control release of radioactive particulates. Although no emission standards have been established for radioactive particulate emission, ERDA Manual Chap. 0524¹ requires that emission be controlled so that effluent concentrations at the boundary between controlled and uncontrolled areas do not exceed specified amounts. This requirement limits soluble ^{239}Pu release to 2.22×10^{-3} dis/s·m³ at

the discharge point if no credit is allowed for atmospheric dilution between that point and the site boundary. Some operations exhaust plutonium concentrations as high as 4×10^9 dis/s·m³ into the air-cleaning system.² To reduce this concentration to that specified by ERDA the air-cleaning system must provide a decontamination factor (DF) on the order of 10^9 . Individual HEPA filters provide a DF of approximately 2×10^3 , and three in series theoretically could provide a DF of approximately 8×10^9 , if all filters performed equally well. However, each stage of filtration modifies the challenge aerosol so that particle diameters at each successive filter

stage approach a size of maximum penetration (SMP). Performance of the second and third filters against this most difficult-to-collect aerosol limits the overall system DF.

Published experimental results have indicated maximum penetration of fibrous filters over a range of submicron diameters, both above and below 0.3 μm .²⁻⁴ Monodisperse 0.3- μm dioctyl phthalate (DOP) was selected as a test aerosol for its proximity to the expected SMP and for its ease of generation.⁷ HEPA filters for use in ERDA facilities are quality-control tested by the manufacturer and by an ERDA quality-assurance station (QAS) to ensure ≤ 0.0003 penetration against 0.3- μm DOP.⁸

Problems associated with handling and installing these filters,⁹ and design characteristics that do not allow post-installation (in-place) testing of some older systems have prompted ERDA to establish the following tentative performance credit criteria: 0.0005 penetration for in-place testable stages and 0.0020 for stages not testable in-place.¹⁰ In-place testing in this case uses an 0.8- μm polydisperse DOP aerosol to test each HEPA filter stage.^{11,12} Individual filters and seals of untestable stages must undergo spot-leak testing with 0.8- μm DOP and repair of any detectable leak. These procedures have ensured high integrity of single filter stages. However, experimental data were not available to substantiate performance of multiple HEPA stages against plutonium aerosols that have a much higher density. To provide this information, an experimental program was conducted to: define the size characteristics of the source terms from major ERDA operations; simulate the more critical plutonium aerosols under laboratory test conditions; and evaluate the performance of multiple stages of HEPA filters against these laboratory aerosols. The possibility of obtaining adequate information by means of a field test program was discarded because existing multiple HEPA filter systems handling large quantities of plutonium did not permit testing of each individual stage. Field testing could not distinguish between plutonium aerosol penetration through filter media and penetration around improperly installed filters. The field sampling and laboratory testing described here quantitatively evaluated HEPA performance, excluding leakage around the filter, using plutonium aerosols representative of field conditions.

II. SOURCE TERM CHARACTERIZATION BY FIELD SAMPLING

Particle size distribution and alpha activity concentrations of plutonium aerosols immediately upstream of the exhaust HEPA filter banks at ERDA plutonium processing sites were determined in a field sampling study.³ Two locations at Mound Laboratories, two at the Rocky Flats Plant, and one at the Los Alamos Scientific Laboratory (LASL) were selected for sampling plutonium aerosols produced by typical research and production operations utilizing ²³⁹Pu and/or ²⁴¹Pu. Samples were collected during the most active periods of the working day, when activity concentrations could be termed "worst normal" and most operations were contributing plutonium aerosols to the process ventilation system. The many variables affecting size characteristics and activity concentration resulted in a range of aerosol size parameters for each facility. The relationships between some of these variables and conditions at the individual sampling sites are summarized in Table I. Although a detailed chemical analysis of each sample was not performed, the predominant aerosol chemical form exhausted into the filter system at each plant was reported to be PuO_2 .

Table I also summarizes the mean values for activity median aerodynamic diameter (amad) and geometric standard deviation (σ_g). Amad is a convenient unit because it is not affected by changes in isotopic ratio, particle shape, or particle density. Particle sizes were determined by radiometric analysis of each of the nine stages of Andersen impactors (eight impaction stages plus a backup membrane filter).¹³ Aerodynamic diameter was considered to be the aerosol parameter of concern in preference to geometric (microscopic) diameter, because inertial impaction is the chief mode of particle collection by HEPA filters operating at rated capacity against aerosols in the size range of interest.⁴

Aerosols from the two fabrication facilities had 2- to 5- μm amads; those from the two research and development facilities had 1- to 4- μm amads; and those from the recovery facility were consistently submicron, with a typical 0.3- to 0.5- μm amad. The recovery facility (Location D) also produced aerosols as small as 0.1 μm amad, had the highest mean activity concentration (1.5×10^8 dis/s $\cdot\text{m}^3$), and constituted the most difficult air cleaning problem.

TABLE I

MEAN PLUTONIUM AEROSOL SIZE CHARACTERISTICS^a AND ACTIVITY CONCENTRATIONS

Location	Type	Isotope	amad (μm)	σ_g	Activity Conc (dis/s·m ³)	
					Mean	Maximum
A	R & D	Both	1.9	2.1	2.0×10^2	8.4×10^2
B	R & D	238	2.9	3.0	2.0×10^3	2.0×10^4
C	Fabri- cation	238	4.1	1.7	1.0×10^3	1.1×10^4
D	Re- covery	239	0.5	3.9	1.5×10^5	4.0×10^5
E	Fabri- cation	239	2.6	2.9	2.7×10^4	4.3×10^5

^aAssuming that particle diameters are log-normally distributed.

III. EXPERIMENTAL APPARATUS AND TECHNIQUES

A. Multiple HEPA Test System

A HEPA filter test system was designed and constructed to test three filters in series against ²³⁹PuO₂ aerosols of sizes near the critical range defined in the field sampling. Performance was to be measured in terms of gross plutonium alpha activity penetrating each filter and of penetration as a function of aerosol aerodynamic diameter.

The test system was housed in two interconnected modules. Figure 1 shows the first module; a glovebox housing the aerosol generators, sampler 1, and HEPA filter 1. A hygrometer-thermometer mounted in the HEPA 1 inlet gave relative humidity and temperature data. Each test filter had a design flow rate of 0.012 m³/s (25 cfm) and its construction and filtration velocity were identical to those of typical 0.472-m³/s (1000-cfm) units used in most air-cleaning systems. The only difference was that the 0.472-m³/s units were generally open-faced, whereas the test filters were designed for in-line installation with 2-in. pipe nipples at each end.

The second module and its major components are shown in Fig. 2. These include sampler 2 followed by HEPA 2; sampler 3 followed by HEPA 3; the final sampler (4); and a vacuum blower. Air removed from the system by samplers 2 and 3 was returned to the system to maintain normal filter flow.

Samplers 1-3 were dual samplers that simultaneously collected a membrane-filter (MF) sample for measuring gross aerosol concentration and an eight-stage Andersen impactor sample for measuring aerosol aerodynamic size. The airstream entered these samplers through sharp-edged, right-angle probes sized for isokinetic sampling. The MF measurements determined total penetration in terms of radioactivity, whereas impactor data were used to calculate HEPA filter penetration as a function of plutonium aerosol aerodynamic size. This last calculation compared activity on both sides of the HEPA filter in a given impactor stage, allowing SMP determination. Sampler 4, consisting of nine 5.1-cm open-face glass fiber filters in a single housing, was designed to filter all air exhausting from the system. Very low activity levels at this point in the system precluded impactor measurements to define aerosol size characteristics downstream of HEPA 3.

The Andersen impactor impaction surfaces were covered with Type AA Millipore membrane filters to reduce particle rebound and to provide a convenient substrate for sample analysis. Although sticky substances thinly coating the impaction surface improve collection of lightly loaded samples, they could not be adequately controlled in these test conditions and were expected to interfere with radiometric analysis of samples. Limited studies of plutonium aerosol collection on the MF coating and

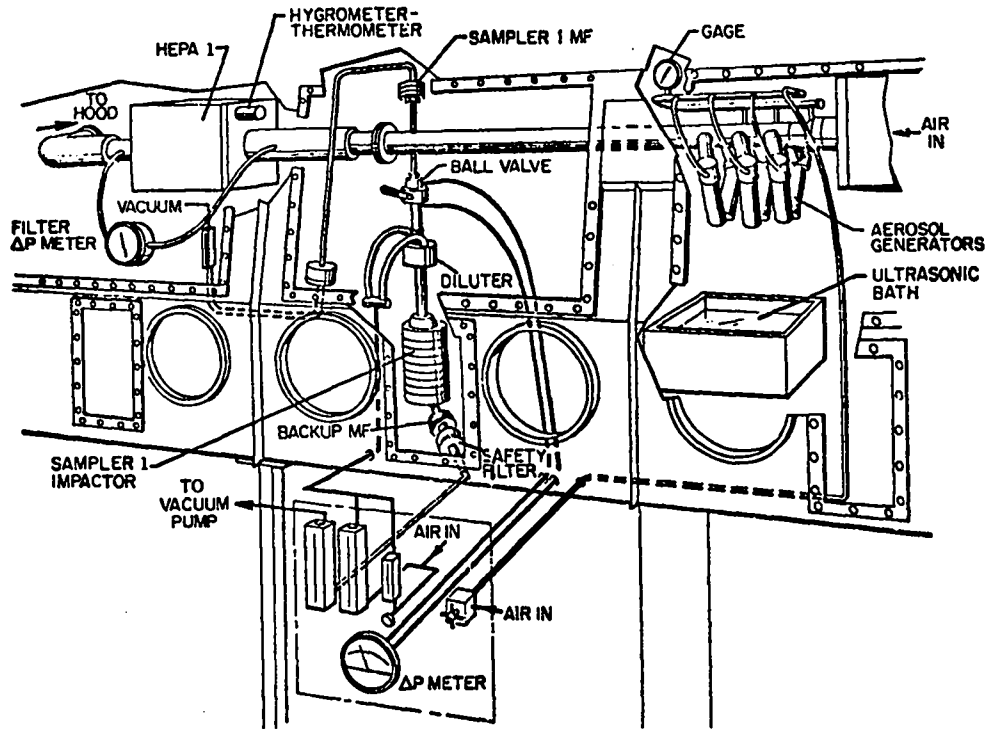


Fig. 1.
 HEPA filter test system in glovebox.

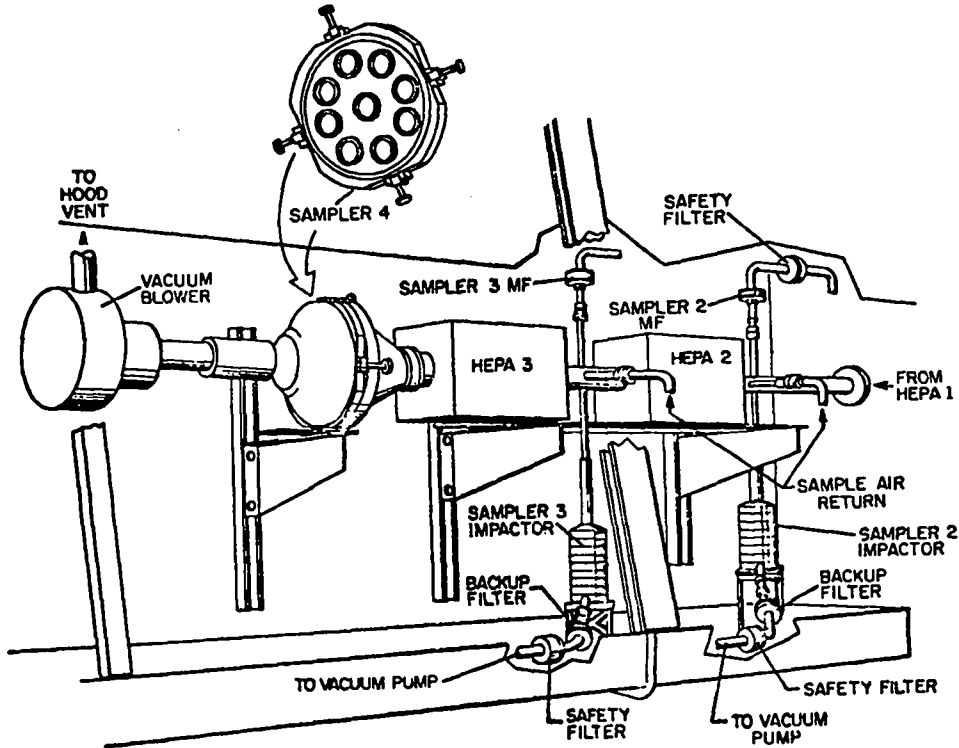


Fig. 2.
 HEPA filter test system in hood.

on a glass-fiber filter coating showed minimal difference and no indication of gross rebound such as high deposition on the backup filter.

To give enough alpha activity in samples taken downstream of the third HEPA filter in series, the aerosol-generating system had to produce activity concentrations of $\sim 10^{10}$ to 10^{11} dis/s \cdot m³. This high activity concentration upstream of the first HEPA filter produced activity levels on the first impactor that were too high to count with the equipment available. To alleviate this problem, a relatively small sample flow (2.36×10^{-5} m³/s) was drawn into the sampling probe and diluted with 4.48×10^{-4} m³/s of filtered air. Even after 20:1 dilution, some samples required additional serial dilutions before being counted. (Sample preparation and counting techniques are described later.) Andersen impactors downstream of the first and second HEPA filters did not need dilution systems because the lower activity concentrations at those positions permitted direct counting of samples.

Sampling times at each sampling position varied; 0.5 or 1.0 min was enough upstream of HEPA 1, and up to 2 h was necessary downstream of HEPA 2. Because of these gross differences in sampling times, it was necessary to take frequent samples upstream of HEPA 1 during each run and use a time-weighted average for the upstream concentration.

Plutonium aerosol was generated from a water suspension by a compressed-air-operated Retec nebulizer,¹⁴ slightly modified for higher output. Six nebulizers attached to a central duct (Fig. 1), with a generator suspension concentration of up to 8.0 mg/ml ²³⁹PuO₂, yielded aerosol concentrations of 1.0×10^9 to 3.3×10^{10} dis/s \cdot m³. Concentrations in the upper part of this range were required to test three HEPA filters in series. The aerosol was dried by adding heated dilution air through an inlet HEPA filter. Maximum temperature and humidity conditions entering HEPA 1 were 30°C and 50%, respectively, well within the ranges encountered in typical field applications.

To approximate the plutonium aerosols with 0.1- to 5- μ m amads measured in the field study, ²³⁹PuO₂ powders were dry ball-milled for various times and suspended in water to give 2.5- to 8.0-mg/ml concentrations. Ultrasonic agitation of the suspension broke up agglomerates, and addition of an anionic surfactant kept the suspensions well dispersed. Selective ball-milling kept the size within the range

of interest, except for some limitations at either end. By adjusting the ball milling time, it was possible to produce aerosols with 0.7- to 1.6- μ m amads, with σ_g 's ranging from 2.1 to 2.9. Even extensive dry ball milling did not produce an aerosol with an amad smaller than 0.7 μ m. However, the 0.7- μ m aerosols contained a significant fraction of < 0.4 - μ m particles, the smallest size fraction that the Andersen impactor can characterize when operated at its normal sampling rate of 0.47×10^{-3} m³/s.

To provide aerosols like those measured at the chemical recovery facility (Location D; amad as low as 0.1 μ m), a centrifugal ball mill was used to mill batches of ²³⁹PuO₂ for varying times. This mill gave smaller sizes in a reasonable time because of its higher rate of energy input. Milling was done using a carrier liquid to reduce agglomeration. Initially, ethanol was used, but high pressures generated within the mill enclosure necessitated a change to water as the carrier liquid. Further problems were encountered because alpha activity produced H₂, O₂, and H₂O₂, that created high pressures and explosive mixtures within the mill jar. A continuously vented mill enclosure was developed to eliminate these problems. The new milling procedures yielded 0.22- to 0.66- μ m aerosol amads in 44 to 167 h of milling. Though it did not reach the desired 0.1- μ m amad, 10-30% of this material was in the < 0.22 - μ m size range of interest.

The sampling system upstream of each HEPA stage was modified to allow impactor sampling at 1.4×10^{-3} m³/s. As in the field sampling program at Location D, operating the Andersen impactor at higher flow rates lowered the effective particle size range to include the lower limit of the range of interest (0.1 μ m). Calculated and experimentally measured effective cutoff diameters¹⁵ for 1.41×10^{-3} m³/s flow rates agree well enough to permit characterization of the test aerosol by this technique.

B. Sample Preparation and Radiometric Determination

High-activity samples (Samplers 1 and 2) required serial dilution to reduce counting rates within the capability of available alpha-counting equipment. The AA Millipore filter media (used as impaction surfaces in the impactors) and the backup MF sample were dissolved in acetone (up to 200 ml). An

aliquot withdrawn by precision syringe was plated on aluminum plates 8.25-cm diam, the plates were then wrapped in thin Mylar film (0.75 mg/cm²), and the sample was alpha counted with a 10- x 23-cm gas-flow proportional counter. Resolving-time corrections were based on 10- μ s resolving-time measurements. Alpha particle absorption in the Mylar film was accounted for by standardizing the counter with Mylar-covered sources. Self-absorption corrections were neglected because the 11- μ m range of an alpha particle (5.5 MeV for ²³⁹Pu) in ²³⁹PuO₂ is long, compared to the 3- to 4- μ m physical diameter of particles collected on the impactor's first stage. Alpha-spectrometry of energy peaks showed that absorption of alpha particles originating from ²³⁹Pu particles buried in the MF was insignificant. Low-activity samples (samplers 3 and 4) were counted on a low-background, high-efficiency system consisting of Sylvania Type 130 scintillator paper between the sample and a photomultiplier light pipe.

C. Methods of Statistical Analysis of Penetration Data

Activity concentrations were calculated using data from radiometric analysis of samplers 1-4, corrected for activity removed by upstream samplers. The corrected activity concentrations were then used to calculate penetration (the ratio of downstream and upstream concentrations). The resulting penetrations were analyzed by standard statistical methods to determine values of central tendency and variance. Where possible, the data were tested for Gaussian (normal) distribution to permit versatile presentation of data in terms of the mean and variance (standard deviation squared).

Several conditions encountered during the experiment indicated the need to delete data obviously in error due to identifiable instrument error. Some preliminary data were rejected because of sample contamination by radon and thoron daughters. This problem was eliminated by waiting several weeks between sample collection and counting, to permit radioactive decay of these short-half-life materials. Data from several complete runs were discarded because of major instrument problems. These included poor comparison of activity concentration indicated by sampler MF 1 and summation of impactor stages at the same location, unreasonably high σ_x

at sampler 3 (an indication of low flow in the impactor), and manufacturer error in one impactor sampler.

Variance of the measured penetrations was reduced in several instances by analysis in appropriate subgroups. Such analysis permitted the use of some data that otherwise would have been rejected as outliers by statistical testing. Grouping was particularly helpful in analyzing the HEPA 1 penetration data, since these filters were replaced because of loading after only one or two experimental runs. Random selection of replacement HEPA 1 filters resulted in higher variances relative to HEPA 2 or HEPA 3, which were replaced only occasionally. A similar variation occurred when filters of higher penetration by 0.3- μ m DOP (as indicated by QAS test) were purposely installed as HEPA 1 and 2 to increase the activity concentrations upstream and downstream of HEPA 3. Thereafter, it became advantageous to group the HEPA 1 and 2 filter penetrations into categories of low (0.00002-0.00015) or higher (0.00015-0.0003) QAS penetration. Grouped in this way, data previously rejected as outliers could be included in the calculation of mean and variance. Variance of data in each category was reduced, and confidence in the results was improved.

Testing for outliers provided a consistent technique for evaluating data that were unusual for unexplainable reasons. An outliers test usually assumes that the data are normally distributed and then rejects the maximum or minimum value if it deviates from the normal distribution by a fixed amount at a desired significance level. The Grubbs outliers test was used with the following test statistics as indices of deviation:

$$T_n = \frac{x_n - \bar{x}}{S} \quad \text{and} \quad T_1 = \frac{\bar{x} - x_1}{S}$$

where x_n is the largest and x_1 the smallest observation, \bar{x} is sample mean, and S is sample standard deviation (based on $n-1$ observations).¹⁶ If T_n or T_1 exceeded the tabulated critical value T_c for n observations at the 5% significance level, x_n (or x_1) was considered to be an outlier and was omitted from the reported data. This process was repeated until neither the largest nor the smallest data point was rejected as an outlier.

A count-rate criterion was established to ensure that penetration values were based on count rates significantly above average background (about 0.27 counts/min). Average sample and background counting times were 1400 min and 1000 min, respectively. The counting errors (± 1 std dev) corresponding to average gross count rates (GCR) at several factors of background (BG) count rate are listed below:

<u>Average GCR</u>	<u>Counting Error (%)</u>
2 x BG	9.5
1.5 x BG	17.5
1.1 x BG	73

A gross count rate ≥ 1.5 times background was considered to produce acceptable error levels when compared to other error sources in the experiments.

The distribution of penetration data from an individual HEPA stage can be described fully by the mean and standard deviation, if data can be shown to be normally distributed. Normality was evaluated by applying a statistical test of the Kolmogorov-Smirnov type (chi-square, Lilliefors, or W-test).¹⁷ If the test statistic computed from the data exceeded a critical value provided by the test at the desired significance level, the hypothesis of normality was rejected.

Each set of penetration data was subjected to the W-test at the 5% significance level, where a set was defined as a series of data gathered under unique conditions of flow, HEPA position, or DOP penetration category. Of the 53 sets tested, 30 did not deviate from the normal distribution. The natural logarithms of the data in 26 of the 53 sets were subjected to the W-test, and 14 sets were lognormally distributed. Five of the 26 sets were rejected as being neither normally nor lognormally distributed. Because no clear overall tendency to either distribution was indicated, the penetration data discussed in the following sections are expressed as sample mean, \bar{x} , and sample standard deviation, S, with the qualification that a majority of the sets were normally distributed.

When two sets of penetration data with high variances were compared but could not be assumed significantly different by simple inspection, a test of significance called the sign test was applied at the

5% significance level.¹⁸ In this test, the difference D_i between each paired observation X_i and Y_i was recorded as a + or - for each pair according to whether D_i was positive or negative. The number of times the less-frequent sign occurred was then compared to a tabulated critical value. If the number of times the less-frequent sign occurred was less than or equal to the critical value, the hypothesis that x and y had the same distribution was rejected. This test was particularly useful in examining penetration data as a function of particle size.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. HEPA Penetration at Full Flow

Penetrations of HEPA 1-3 by 0.2- to 1.6- μm amad $^{239}\text{PuO}_2$ aerosols in a 0.012-m³/s (25-ft³/min) air-stream are given in Table II. The sample mean penetration \bar{x} and sample standard deviation S are reported, along with the number of data points used in calculating \bar{x} and S, and the total number of data points available in that category. The number of data points available for each filter varies owing to deletion of data as described in Sec. III.C. or, in a few cases, to sampling intentionally not performed. The difference between these two values in the N column is the number of data points rejected as outliers.

Penetration of HEPA 2 was higher than that of HEPA 1 by factors ranging from 2 to 3. Very few runs exhibited higher penetration of HEPA 1 than of HEPA 2; those runs giving higher $^{239}\text{PuO}_2$ penetration of HEPA 1 also showed much higher DOP quality-control penetration of HEPA 1.

The measured HEPA filter penetrations were well within the present minimum ERDA performance guidelines for each stage (500×10^{-6} for the first stage and 2000×10^{-6} for succeeding stages). In fact, HEPA 2 penetration was always $< 200 \times 10^{-6}$ (Table II). HEPA 1 data varied more than HEPA 2 data owing to the frequent changeout of HEPA 1 described earlier.

Mean penetration of HEPA 3 exceeded that of HEPA 1 by a factor of 8 to 9, for filters with low DOP penetration rating. This increased penetration of HEPA 3 results from either reduced particle size or, more likely, from aerosol modification at both ends of the size spectrum as it passed each filtration

TABLE II
PENETRATION OF HEPA FILTERS OPERATING AT FULL FLOW
BY 0.22- to 1.6- μm amad $^{238}\text{PuO}_2$ AEROSOL

HEPA No.	Penetration Rating	Maximum Penetration ($\times 10^6$)	Mean Penetration \bar{x} ($\times 10^6$)	Std Dev S ($\times 10^6$)	N^c
1	Low ^a	21	8.3	7.1	20/20
1	Higher ^b	108	32	33	23/24
2	Low ^b	31	17	7.2	22/23
2	Higher ^b	159	106	38	15/15
3	Low ^a	78	71	10	6/8

^a20 to 150 $\times 10^{-6}$ by 0.3- μm DOP quality-assurance test.

^b151 to 300 $\times 10^{-6}$ by 0.3- μm DOP quality-assurance test.

^cNumber of data points passing outliers test/data points available.

stage. The aerosol amad would in the latter case approach the size of maximum penetration (SMP), and the σ_g would be reduced. Mean values for σ_g showed a 30-35% reduction across HEPA 1 and 4-5% reduction across HEPA 2. Conversely, an amad significantly below the SMP might increase slightly after passing a filtration stage.

These test results clearly show low penetration of HEPA 1, 2, or 3 by the polydisperse 0.22- to 1.6- μm amad $^{238}\text{PuO}_2$ aerosols. For instance, the highest mean penetration in Table II, 106 $\times 10^6$, and its corresponding standard deviation indicate the probability of these filters' exceeding 200 $\times 10^6$ penetration (99.98% efficiency) is <1%. The decontamination factor for three filters in series (based on highest mean penetrations from Table II) was 4 $\times 10^{12}$. A worst-case DF for three filters in series (based on the maximum penetrations measured for each stage) was 8 $\times 10^{11}$, which exceeds the DF required by the ERDA minimum performance guideline (DF = 5 $\times 10^8$).

B. Penetration as a Function of Particle Size at Full Flow

Full-flow data for the penetration of HEPA 1 and 2 by $^{238}\text{PuO}_2$ particles in the size intervals defined by the Andersen impactor operated at 0.47 $\times 10^{-3}\text{m}^3/\text{s}$ are given in Table III. The following tables and figures give the penetrations of HEPA filters

categorized into low and higher DOP penetration as defined earlier.

Table III shows that penetration of HEPA 1 was significantly higher in the 0.43- to 0.65- μm D_{ae} interval than in adjacent intervals for filters with low DOP penetration, and that equal penetration occurred in the 0.43- to 0.65- μm and 0.65- to 1.1- μm intervals for filters in the higher DOP category. The HEPA 2 result was not definite, owing to the absence of a maximum in any interval.

Full-flow penetration of HEPA 1 and 2 by particles in the size intervals defined by the Andersen impactor operated at a higher flow (1.42 $\times 10^{-3}\text{m}^3/\text{s}$) is given in Tables IV and V, respectively. At this higher flow rate, each impactor stage has a smaller effective cutoff diameter (ECD), and the last-stage ECD is reduced from 0.43 to 0.12 μm . HEPA 1 results given in Table IV were consistent with the low-penetration HEPA 1 results given in Table III. Maximum penetration for this category occurred in the 0.44- to 0.96- μm interval, which contains the 0.43- to 0.65- μm interval noted in Table III. Filters showing higher DOP penetration showed maximum $^{238}\text{PuO}_2$ penetration in the 0.22- to 0.44- μm and 0.44- to 0.96- μm intervals.

The HEPA 2 low-DOP category in Table V failed to show maximum penetration in a specific size interval (as noted in Table III). Although HEPA 2 filters with higher DOP penetration apparently exhibited unusually high penetration in the >5.4- μm interval and the <0.12- μm interval, the data were

TABLE III
HEPA PENETRATION VS PARTICLE SIZE
FULL-FLOW RUNS
SAMPLE FLOW 0.47×10^{-3} m³/s

Impactor Stage	Size Interval D _{ae} (μm)	HEPA 1						HEPA 2		
		Low Penetration ^a			Higher Penetration ^b			Low Penetration ^a Only		
		Mean Penetr. \bar{x} (x10 ⁶)	Std Dev S (x10 ⁶)	N ^c	Mean Penetr. \bar{x} (x10 ⁶)	Std Dev S (x10 ⁶)	N ^c	Mean Penetr. \bar{x} (x10 ⁶)	Std Dev S (x10 ⁶)	N ^c
0	>11.0	2.1	2.0	21/21	3.0	2.6	6/6	---	---	---
1	7-11	1.7	1.5	21/21	1.9	1.7	6/6	---	---	---
2	4.7 - 7	1.0	0.83	21/21	1.7	1.3	5/6	---	---	---
3	3.3 - 4.7	1.2	1.2	21/21	1.5	1.2	5/6	---	---	---
4	2.1 - 3.3	1.8	2.1	21/21	3.7	3.7	5/6	---	---	---
5	1.1 - 2.1	6.8	5.7	21/21	11	11	6/6	2.1	2.1	11/11
6	0.65 - 1.1	20	10	21/21	27	26	5/6	17	9.2	11/11
7	0.43 - 0.65	28	14	21/21	26	23	6/6	18	6.1	11/11
MF2	<0.43	16	11	21/21	13	11	6/6	26	13	11/11

^a20 to 150 x 10⁻⁸ by 0.3-μm DOP quality-assurance test.

^b151 to 300 x 10⁻⁸ by 0.3-μm DOP quality-assurance test.

^cNumber of data points passing outliers test/data points available.

TABLE IV
HEPA 1 PENETRATION VS PARTICLE SIZE
FULL-FLOW RUNS
SAMPLE FLOW 1.42×10^{-3} m³/s

Impactor Stage	Size Interval D _{ae} (μm)	Low Penetration ^a			Higher Penetration ^b		
		Mean Penetr. \bar{x} (x10 ⁶)	Std Dev S (x10 ⁶)	N ^c	Mean Penetr. \bar{x} (x10 ⁶)	Std Dev S (x10 ⁶)	N ^c
0	>5.4	0.44	0.34	7/8	13	13	14/14
1	3.4 - 5.4	0.36	0.37	7/8	13	12	13/14
2	2.3 - 3.4	0.23	0.18	7/8	14	13	14/14
3	1.5 - 2.3	0.20	0.13	7/8	30	30	14/14
4	0.96 - 1.5	0.74	0.51	7/8	39	33	13/14
5	0.44 - 0.96	5.8	5.4	7/8	64	57	13/14
6	0.22 - 0.44	2.8	1.7	7/8	71	59	14/14
7	0.12 - 0.22	2.7	2.2	7/8	54	50	14/14
MF2	<0.12	1.0	0.67	7/8	36	36	14/14

^a20 to 150 x 10⁻⁶ by 0.3-μm DOP quality-assurance test.

^b151 to 300 x 10⁻⁶ by 0.3-μm DOP quality-assurance test.

^cNumber of data points passing outliers test/data points available.

TABLE V
HEPA 2 PENETRATION VS PARTICLE SIZE
FULL-FLOW RUNS
SAMPLE FLOW $1.42 \times 10^{-3} \text{ m}^3/\text{s}$

Impactor Stage	Size Interval D_{ae} (μm)	Low Penetration ^a			Higher Penetration ^b		
		Mean Penetr. \bar{x} ($\times 10^6$)	Std Dev S ($\times 10^6$)	N ^c	Mean Penetr. \bar{x} ($\times 10^6$)	Std Dev S ($\times 10^6$)	N ^c
0	>5.4	---	---	---	61	53	3/4
1	3.4 - 5.4	---	---	---	22	14	3/4
2	2.3 - 3.4	---	---	---	3.0	1.3	3/4
3	1.5 - 2.3	---	---	---	4.2	2.0	3/4
4	0.96 - 1.5	5.1	4.9	5/5	24	19	4/4
5	0.44 - 0.96	11	3.6	8/8	126	82	4/4
6	0.22 - 0.44	13	3.6	8/8	92	24	4/4
7	0.12 - 0.22	12	4.5	8/8	82	31	4/4
MF2	<0.12	13	3.7	8/8	200	220	4/4

^a20 to 150×10^{-6} by 0.3- μm DOP quality-assurance test.

^b151 to 300×10^{-6} by 0.3- μm quality-assurance test.

^cNumber of data points passing outliers test/data points available.

few in number and failed to show statistically significant differences among size intervals.

Bar graphs of mean penetrations of HEPA 1 as a function of impactor size interval (Figs. 3, 4, 5, and 6) are helpful in visualizing the probable SMP location. Although the impactor size intervals are not uniform and are too large to permit a smooth curve through midpoints in the bar graph, the bar configuration provides a basis for predicting a size range that includes the SMP. This range, 0.4- to 0.7- μm , is larger than the 0.3- μm SMP commonly accepted for unit-density aerosols like DOP. A larger SMP for $^{238}\text{PuO}_2$ can be attributed to increased diffusion collection.¹⁹ Diffusion deposition varies inversely with geometric particle size (and velocity) and does not depend on particle density. Because a particle of given D_{ae} has a geometric particle size (D_p) proportional to $D_{ae}/\rho^{1/3}$, a high-density particle would have a smaller D_p than a unit-density particle with the same D_{ae} . Diffusion collection of these high-density particles would be enhanced and would yield an SMP somewhat above that for unit-density particles.

C. HEPA Penetration at Half Flow

As stated earlier, reduced velocity in HEPA filters reduces particle collection by impaction and

improves collection of small particles because of increased diffusion. Because some HEPA systems are operated at reduced flow to extend their useful life by delaying changeout necessitated by high differential pressure, a series of penetration measurements to determine how reduced flow affects HEPA performance was performed under test conditions described earlier, except that system flow was half of normal rated capacity.

Penetration data for HEPA 1-3 operating at 0.006 m^3/s (half of rated flow) are given in Table VI. These data are representative of HEPA performance against 0.23- to 0.6- μm -amad $^{238}\text{PuO}_2$ aerosols, a narrower range of sizes than that used to challenge filters at full flow. Primary observations were as follows: higher penetration at HEPA 2 than at HEPA 1, comparable to full-flow test results; a large variance associated with HEPA 1 data, similar to full-flow test results; and very similar penetration values for HEPA 1 and 2 in half-flow and full-flow test results.

Penetration could increase or decrease with reduced air velocity (flow), depending on where most of the aerosol activity lies with respect to a reference SMP. If the amad were greater than the SMP, penetration would probably increase because of reduced inertial collection; conversely, an aerosol whose amad was below the SMP would probably

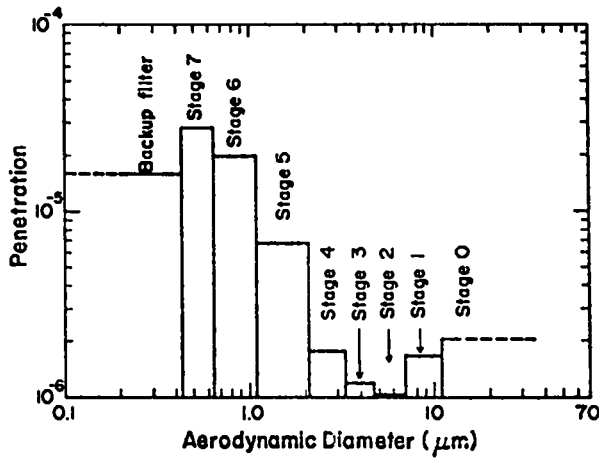


Fig. 3.

²³⁹PuO₂ aerosol size vs penetration of HEPA 1 filters showing low DOP penetration (impactor sample flow was $0.47 \times 10^{-3} \text{ m}^3/\text{s}$).

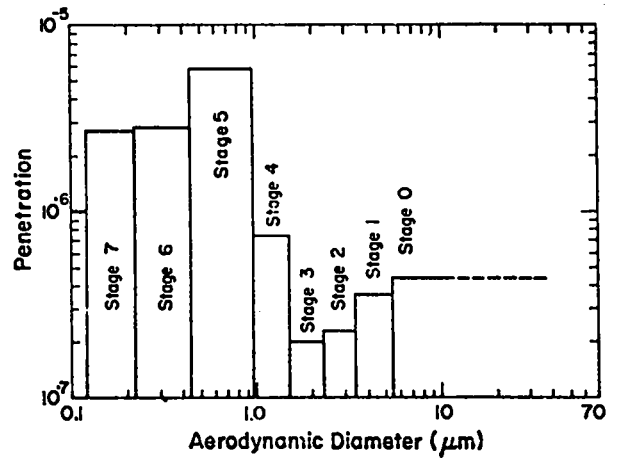


Fig. 5.

²³⁹PuO₂ aerosol size vs penetration of HEPA 1 filters showing low DOP penetration (impactor sample flow was $1.42 \times 10^{-3} \text{ m}^3/\text{s}$).

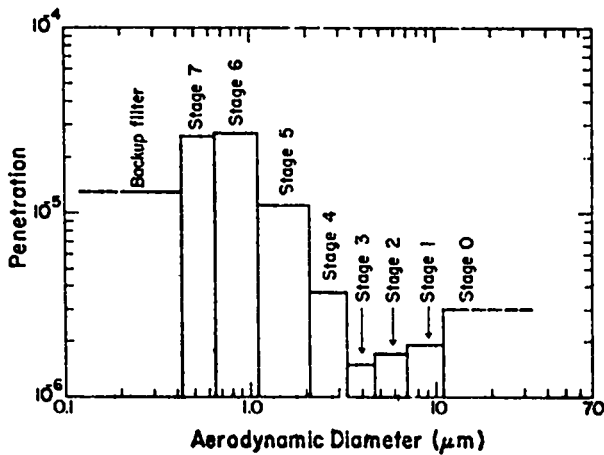


Fig. 4.

²³⁸PuO₂ aerosol size vs penetration of HEPA 1 filters showing higher DOP penetration (impactor sample flow was $0.47 \times 10^{-3} \text{ m}^3/\text{s}$).

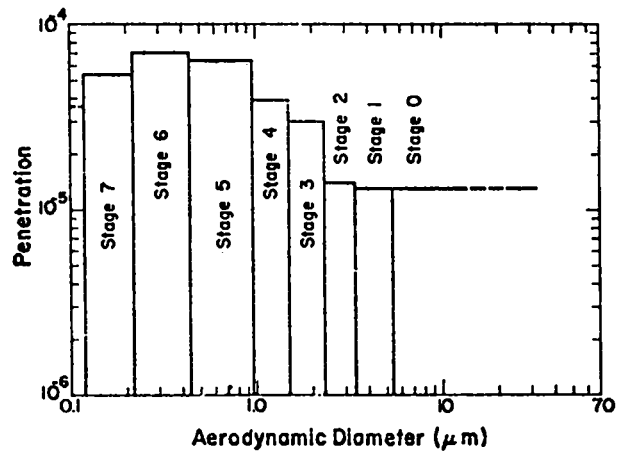


Fig. 6.

²³⁸PuO₂ aerosol size vs penetration HEPA 1 filters showing higher DOP penetration (impactor sample flow was $1.42 \times 10^{-3} \text{ m}^3/\text{s}$).

penetrate less owing to enhanced diffusional collection. Half-flow penetrations in Table VI and full-flow penetrations in Table II describe a "low amad and low flow" condition versus a "higher amad and high flow" condition, both of which should lead to low penetration.

Half-flow penetration of HEPA 3 in series with HEPA 1 and 2 is also given in Table VI. HEPA 3 penetration is higher at half flow than at full flow, but still far below 0.002. Size characteristics of aerosols challenging HEPA 3 at half and full flow were very similar, and DOP quality-control test results were identical for all the filters used in these tests; thus these two conditions are eliminated as possible causes of the higher penetration. If lower flow had been the cause, an equally large increase should have been observed at HEPA 2. Other than suggesting the possibility of damage in shipping or installation, we have no explanation to offer here.

The decontamination factor based on mean penetrations of three filters in series (Table VI) was 10^{12} . Worst-case DF based on maximum penetrations was 7×10^{10} , well above the 5×10^8 ERDA guideline DF. Although the worst-case criteria are very stringent and the situation is not likely to arise, the worst-case DF is an impressive illustration of the performance capabilities of a three-stage HEPA system.

D. Penetration as a Function of Particle Size at Half Flow

Table VII includes HEPA 1 penetration vs size results from half-flow testing. Maximum penetration from half-flow runs occurred in the 0.44- to 0.96- μm interval for low-DOP filters, and almost equally in the 0.44- to 0.96- μm and 0.96- to 1.5- μm intervals for the higher DOP filters. An indication of slightly larger SMP at half flow would be consistent with enhanced diffusion collection at the lower airstream velocity; however, the large S values associated with each penetration would make a small increase difficult to recognize.

E. Effects of High-Specific-Activity Particles on HEPA Filter Media

Thermal and radiation damage of HEPA filter media by high-specific-activity particles was investigated theoretically and experimentally. Heat-transfer calculations showed that 50- μm ^{239}Pu particles raise the surface temperature of 0.5- μm glass fibers negligibly above ambient temperature.²⁰ Heat removal by either conduction or radiation would provide the necessary cooling, exclusive of a convective contribution that may be even greater than that of radiation or conduction. Thus, high-specific-activity ^{239}Pu particles cannot heat filter fibers to the softening point.

TABLE VI
PENETRATION OF HEPA FILTERS
OPERATING AT HALF FLOW
BY $^{239}\text{PuO}_2$ AEROSOL 0.23 - 0.6 μm amad

HEPA No.	Penetration Rating	Maximum Penetration ($\times 10^6$)	Mean Penetration \bar{x}	Std Dev S	N^c
1	Low ^a	33	12×10^{-6}	12×10^{-6}	6/6
1	Higher ^b	90	20×10^{-6}	29×10^{-6}	15/16
2	Low ^a	34	22×10^{-6}	7.5×10^{-6}	6/6
2	Higher ^b	123	69×10^{-6}	53×10^{-6}	10/10
3	Low ^a	1370	740×10^{-6}	365×10^{-6}	5/5

^a20 to 150×10^{-6} by 0.3- μm DOP quality-assurance test.

^b151 to 300×10^{-6} by 0.3- μm DOP quality-assurance test.

^cNumber of data points passing outliers test/data points available.

TABLE VII
HEPA #1 PENETRATION VS PARTICLE SIZE
HALF-FLOW RUNS

Impactor Stage	Size Interval D_{ae} (μm)	Low Penetration ^a			Higher Penetration ^b		
		Mean Penetr. \bar{x} ($\times 10^6$)	Std Dev S ($\times 10^6$)	N ^c	Mean Penetr. \bar{x} ($\times 10^6$)	Std Dev S ($\times 10^6$)	N ^c
0	> 5.4	2.3	2.0	13/14	6.2	5.3	9/10
1	3.4 - 5.4	1.7	2.0	13/14	8.0	7.5	9/10
2	2.3 - 3.4	1.7	2.0	12/14	22	23	10/10
3	1.5 - 2.3	5.6	5.7	12/14	44	55	10/10
4	0.96- 1.5	10.0	12	14/14	61	85	10/10
5	0.44- 0.96	17	14	14/14	56	66	10/10
6	0.22- 0.44	5.7	5.2	12/14	45	53	10/10
7	0.12- 0.22	4.5	4.4	13/14	41	43	10/10
MF 2	< 0.12	1.9	2.0	14/14	35	50	10/10

^a20 to 150 $\times 10^{-6}$ by 0.3- μm DOP quality-assurance test.

^b151 to 300 $\times 10^{-6}$ by 0.3- μm DOP quality assurance test.

^cNumber of data points passing outliers test/data points available.

Radiation damage to filters by high-specific-activity particles was considered in terms of the integrated exposure of filter fibers in contact with a $^{238}\text{PuO}_2$ particle. Experimental work reported in the literature²¹ shows that damage to Pyrex borosilicate containers appeared as very fine cracks after exposure to an integrated 10^{20} - α/m^2 alpha flux. Silica was leached at exposures as low as 10^{18} α/m^2 . Alpha exposure of a glass fiber in contact with a 10 - μm $^{238}\text{PuO}_2$ particle was calculated to be 3×10^{20} $\alpha/\text{m}^2/\text{yr}$.²² Although this suggested that alpha radiation could cause HEPA filter efficiency loss, the probability appears rather small that enough fibers are being damaged to cause a significant reduction in filter collection efficiency.

Several filters that had been already tested with a sub-micron $^{238}\text{PuO}_2$ aerosol and loaded with from 0.5 to 2.5 g of aerosol $^{238}\text{PuO}_2$, were stored under nonflow conditions for approximately 1 yr and then retested against similar $^{238}\text{PuO}_2$ aerosols. Table VIII shows that all these filters exhibited <0.0001 penetration in both tests, indicating no significant loss of performance following loading and storage with $^{238}\text{PuO}_2$.

V. SUMMARY

Efficiency studies of multistage HEPA filter systems were performed to confirm design requirements of air-cleaning systems for several new plutonium-processing plants. Primary interest lay in demonstrating that filter penetrations that satisfied the ERDA guidelines of <0.0005 for in-place testable stages and <0.0020 for stages not testable in place were attainable. A laboratory study was initiated using $^{238}\text{PuO}_2$ as the test aerosol. A field study preceding the laboratory phase determined the aerosol size characteristics and activity concentrations for typical plutonium-processing operations at three ERDA plants. The performance of a three-stage HEPA test system was evaluated against similar aerosols.

Penetrations of three HEPA filters in series by $^{238}\text{PuO}_2$ aerosols were measured at the rated filter flow and at half the rated flow. Aerosol size ranged from 0.22- to 1.6 - μm amad.

Although penetration increased at each succeeding stage and the aerosol size distribution was modified to a more penetrating range, mean penetration of each stage remained generally below

TABLE VIII
PENETRATIONS OF HEPA FILTERS AFTER EXTENDED EXPOSURE TO $^{239}\text{PuO}_2$

Filter Number	Total Loading $^{239}\text{PuO}_2$ (g)	Exposure Time ^a (days)	Filter Penetration					
			Before Exposure			After Exposure		
			amad (μm)	σ_g	Penetration	amad (μm)	σ_g	Penetration
415	0.50	379	0.7	2.3	<0.0001	0.8	9.3	<0.0001
			1.3	2.9	<0.0001	0.7	5.2	<0.0001
			1.3	2.7	<0.0001	0.7	6.4	<0.0001
414	0.73	385	0.6	2.2	<0.0001	0.8	5.0	<0.0001
			0.8	2.7	<0.0001			
			1.6	2.7	<0.0001			
430	2.50	372	0.7	2.1	<0.0001	0.6	4.0	<0.0001
			0.8	2.2	<0.0001			

^aTime-lapse since first exposure.

0.0002 under half- and full-flow conditions. Maximum penetration in all tests was 0.0014 (observed at HEPA 3 in half-flow tests). This value meets the 0.0020 guideline for untestable second and third stages.

Decontamination factors calculated from the highest mean penetration values from all tests (half and full flow) equaled or exceeded 10^{12} . Worst-case decontamination factors calculated from the highest penetration data for each stage exceeded 10^{10} . However, as this study was done under ideal conditions to ensure that only aerosol penetration (not leakage around the filter) was monitored, proper installation of quality-control-tested HEPA filters is of prime importance to achieve the decontamination factors determined here.

Maximum HEPA filter penetration by $^{239}\text{PuO}_2$ aerosol was observed between 0.4- and 0.7- μm D_{50} . This most penetrating size, large compared to the 0.3 μm frequently cited for unit-density aerosols, reflects the influence of density on the filtration mechanism.

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