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Abstract

A program is in progress to measure size characteristics and activity concentrations (source term) of plutonium aerosols generated by typical operations handling significant quantities of plutonium. This will provide a basis for generating similar aerosols in the laboratory to study the effectiveness of multiple stages of HEPA filtration. Initial data shows Pu concentrations ranging up to 16.5×10^6 dpm/m³, with the following aerodynamic size characteristics:

aerodynamic activity median diameter (aamd) = .52 to 5.5 μ m

geometric standard deviation (σ_g) = 1.5 to 6.60

I. Introduction

Increased concern regarding the potential release of radioactive particulates to the atmosphere has resulted in new stringent air cleaning requirements for facilities handling plutonium. These include proposals to require decontamination factors of 10^9 to 10^{12} . This can be attained by use of multiple High Efficiency Particulate Aerosol (HEPA) filters, which singly can provide decontamination factors of 10^3 to 10^4 . While extensive test data is available to substantiate this level of performance for individual HEPA filters, and filtration theory predicts multiple filter installations will provide the overall protection required, quantitative substantive data is not available. There is also concern that filter performance against laboratory test aerosols, such as DOP, is not fully representative of performance against plutonium aerosols. Because of this interest in realism, a field sampling program was designed to measure size characteristics and activity concentrations (source term) of plutonium aerosols generated by typical operations at several AEC facilities handling significant quantities of plutonium. Definition of these aerosols would provide a basis for generating similar aerosols in the laboratory to study the effectiveness of multiple stages of HEPA filtration.

Initially three plants, representing different production and research operations, utilizing both ²³⁸Pu and ²³⁹Pu, were selected to provide air sampling data to characterize plutonium source terms under a variety of operating conditions. Wide variations of activity concentration and size characteristics were expected due to: (1) differences in the amount of material handled, and mechanical and chemical operations at each site; and (2) aerosols incident on the main

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exhaust filters may undergo previous stages of filtration at the glovebox. Therefore, sampling data would be represented as ranges of values to describe aerosol concentration and size characteristics. Due to the nature of some operations performed in these plants, sampling locations and operational processes are identified only in these general terms:

1. research and development (r and d)
2. recovery (chemical operations)
3. production (mechanical operations)

The following discussion will primarily be directed at the size characteristics of plutonium aerosols, with only brief mention of activity levels. Data presented is preliminary, and was summarized only for presentation at the Round Table Discussion at this Air Cleaning Conference.

II. Sampling Procedures

The 8-stage Andersen impactor⁽¹⁾ was selected to be the primary sampler for particle size analysis. This device is simple, easy to operate in the field, provides size separation over a fairly wide range of particle diameters on the basis of the inertial properties of the aerosol, and its calibration is well documented.^(2,3) The impactor backup membrane filter and the eight impaction plates are alpha counted for analysis. Net count rate is proportional to the mass of particles deposited on each plate if two assumptions are made: (1) absorption of alpha particles in the particle (self-absorption) is negligible, and 2) specific activity (dpm/gm) is constant. A 5 MeV alpha particle has a range of at least 10 μm in PuO_2 ⁽⁴⁾ compared to a physical diameter of 3 to 4 μm collected on the first stage of the impactor. Therefore, an alpha particle emitted anywhere within a 4 μm PuO_2 particle should emerge from the particle with sufficient energy to be counted. Spectroscopy samples support this hypothesis for the particle size range of interest in this study. However, one of the three facilities discharges a mixture of ^{238}Pu and ^{239}Pu in highly variable ratios. Therefore, data obtained to date have only been defined in terms of aerodynamic activity median diameter (aamd) and geometric standard deviation (σ_g) rather than mass median diameter. The aamd probably relates most closely to the performance of an air cleaner against these aerosols.

Previous evaluation of the Andersen impactor has indicated a potential error due to particle rebound.^(5,6) If the particle does not adhere to the first plate it contacts, but deposits on the next stage, or rebounds from all succeeding stages to be collected by the backup filter, the indicated aerosol size characteristics are in error. Use of a "sticky" impaction surface would interfere with alpha counting procedures and was not acceptable. An alternate technique is the application of membrane filter media to the surface of the impactor plates.⁽⁶⁾ In tests now under way, adjacent impactor sampling streams have been installed at one sampling site, and samples obtained covering the plates of one impactor with vinyl metrical

membrane filters (MF) leaving the plates of the other impactor bare. By alternating the MF coating on each impactor each day, data indicating rebound (or no rebound) can be developed despite possible slight differences between the two sampling streams. Alpha spectroscopy has provided information on the potential error associated with absorption of the alpha particles by the filter media applied to impactor plates as an anti-rebound agent. Preliminary analysis of the vinyl metrical media showed very little absorption and indicate its suitability as an impactor anti-rebound agent.

Sampling locations at each plant were selected to monitor the more contaminated air streams in each plant, and sampling systems were designed to provide isokinetic sampling conditions. Samples are collected for varying lengths of time, depending on the activity in the duct. For convenience of handling, counting, and shipping, activity per sample is limited to about 10^5 dpm, therefore, the time required to collect a sample of this order of magnitude varies greatly for each sampling location. Sampling periods are selected at times when normal activities in the building are underway; i.e., not at lunch time or break time or near quitting time.

A gas flow proportional counter is used to count alpha activity on each sample. This counter is approximately 33% efficient for bare samples and 22% efficient for standard sources covered with a .00025" mylar film. The mylar film is used to cover each impactor plate and filter to minimize contamination problems. Net count rate is determined for each sample and utilized as previously discussed in determining cumulative percent smaller than a stated aerodynamic particle size. Net count rates and the corresponding effective cutoff diameters (ECD)⁽⁷⁾ are submitted for computer analysis as a data set of 8 values to be fit by minimum chi-squares method⁽⁸⁾ and plotted on a log probability grid.⁽⁹⁾ This analysis defines aerodynamic activity median diameter (aamd) and geometric standard deviation (σ_g) for the aerosol, and the deviation of each data point from the best fit line. The latter value can be utilized in a test to confirm or reject the assumption of log normality of the distribution. A range of aamd and (σ_g) describes variations in the size characteristics of typical Pu aerosols.

To provide a frame of reference, Table 1 summarizes the operational characteristics associated with each sampling site. Table 2 indicates the variations in total activity at each sampling site. Concentration data was obtained using both Andersen impactors and adjacent membrane filters. Major variations between week-day and week-end samples are apparent for site A. Even greater variations between site A and sites B and C are, also indicated.

III. Aerosol Size Characteristics

Size characteristics of the Pu aerosols defined by the Andersen impactors are summarized in Table 3, which also indicates the significance of the rebound problem. Arithmetic mean values of aerodynamic activity median diameter (aamd) and geometric standard deviation (σ_g) are listed along with their extremes. Variations are apparent, which is not too surprising considering the large number of different

aerosol-producing operations conducted in these plants.

Somewhat surprising was the size distribution of the Pu aerosol at Location B. Size characteristics of the aerosol measured here are distinctive due to the high percentage of very small particles in the distribution. In the 14 impactor measurements made to date, the cumulative percent smaller than $0.43\text{ }\mu\text{m}$ (final impactor stage ECD) has averaged 60%; that is, 60% of total activity measured in the impactor sampler passed through the impactor and was collected on the backup membrane filter. A distribution of this type is not readily characterized by the Andersen impactor, and these results have not been considered in this presentation.

Figures 1 through 6 are included as log probability representations of typical plutonium size distributions, and the extremes. Figure 1 represents a sample which closely approximates the mean aerosol at Location A ($aamd = 1.70\text{ }\mu\text{m}$ and $\sigma_g = 2.33$). Figures 2 and 3 represent two extremes of σ_g for this sampling location. Figures 4 through 6 show typical size distributions for locations C, D and E. In each figure a best fit line defined by the chi-squares method⁽⁸⁾ is provided. Examination of the data in Table 3 reveals no significant particle rebound. Had the uncoated plates displayed a consistently lower $aamd$ and higher σ_g than coated plates, or larger amounts of activity on the backup filter, a rebound problem would be suggested. Preliminary results of rebound tests conducted with twin impactors has also indicated no rebound problem.

Spectroscopy of several samples has shown the presence at Location A of ^{238}Pu and ^{239}Pu in similar quantities which complicates measurement of size characteristics. The ^{238}Pu and ^{239}Pu particles contributing to the total alpha activity on each impactor plate probably originate at different unrelated operations resulting in two distinct size distributions as indicated in Fig. 7. Samples to investigate the makeup of composite distributions using alpha spectroscopy have been obtained, and are undergoing analysis. Impactor samples will provide ^{238}Pu to ^{239}Pu ratios for each stage, relating isotope concentration to aerodynamic size.

VI. Acknowledgment

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12th AEC AIR CLEANING CONFERENCE

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Table 1
Sampling Locations

<u>Location</u>	<u>Plutonium Isotope</u>	<u>Major Operation</u>
A	238 & 239	r & d
B	239	recovery
C	239	production
D	238	r & d
E	238	production & recovery

Table 2

Activity Concentrations of Pu Aerosols (dpm/m³)

<u>Description</u>	<u>Location</u>	# <u>Samples</u>	<u>A₁ (direct MF)</u>			<u>A_S (Impactor + MF₂)</u>		
			<u>Mean</u>	<u>Max</u>	<u>Min</u>	<u>Mean</u>	<u>Max</u>	<u>Min</u>
Weekdays	A	65	11,388	34,590	286	9,980	36,325	198
Weekends	A	15	988	4,579	51	1,505	4,798	44
Weekdays	B	19	8.10x10 ⁶	16.5x10 ⁶	3.87x10 ⁶	6.72x10 ⁶	15.2x10 ⁶	4.06x10 ⁵
Weekdays	C	26	1.27x10 ⁵	7.86x10 ⁵	1,814	0.98x10 ⁵	6.79x10 ⁵	2,611
Weekdays	D	12	12,020	0.1x10 ⁶	138	4,124	32,537	4
Weekdays	E	11	31,084	0.303x10 ⁶	12	24,913	2.43x10 ⁵	25

Table 3
Andersen Impactor
Size Characteristics of Pu Aerosol

<u>Description</u>	<u>Location</u>	# <u>obs.</u>	<u>aamd (μm)</u>			<u>σ_g</u>		
			<u>mean</u>	<u>max</u>	<u>min</u>	<u>mean</u>	<u>max</u>	<u>min</u>
Weekdays, no coating	A	63	1.70	3.30	.52	2.33	4.94	1.53
Weekends, no coating	A	12	1.92	3.32	1.03	2.37	3.20	2.03
All Samples, DM-800 coating	A	5	2.10	2.50	1.25	2.09	2.61	1.74
All Samples, AA coating	A	10	1.86	2.70	.92	2.38	3.37	1.83
Weekdays, AA coating	B	14	*					
Weekdays, no coating	C	6	2.83	3.53	1.96	3.03	6.40	1.98
Weekdays, AA coating	C	11	2.37	3.25	1.61	2.98	6.6	2.06
Weekdays, DM-800 coating	D	9	3.20	5.51	1.80	3.11	4.36	2.13
Weekdays, DM-800 coating	E	11	1.87	2.99	.57	2.21	2.63	1.58
All samples (excluding B)		127	--	5.51	0.52	--	6.60	1.53

*60% smaller than last impaction stage; not amenable to analysis using Andersen impactor.

DM-800-Gelman vinyl metrical membrane filters (.8 μm pore size)

AA-Millipore Filters (.8 μm pore size)

Figure 1: Size Characteristics of Pu Aerosol--
Representative of Mean aamd and σ_g .
LOCATION A

Figure 2: Size Characteristics of Pu Aerosol--
Representative of Distribution With High σ_g .
LOCATION A

Figure 3: Size Characteristics of Pu Aerosol--
Representative of Distribution with Low σ_g .

LOCATION A

Figure 4: Size Characteristics of Pu Aerosol --
Typical Distribution - Location C

Figure 5: Characteristics of Pu Aerosol --
Typical Distribution - Location D

Figure 6: Size Characteristics of Pu Aerosol --
Typical Distribution - Location E

Figure 7: Size Characteristics of Pu Aerosol--
Representative of Composite Distribution.