PLUTONIUM AEROSOL PARTICLE SIZE DISTRIBUTIONS IN ROOM AIR

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ABSTRACT

The particle size of plutonium aerosols in employee work areas is an important consideration in the analysis of long-term continuous exposure received by radiation workers. This paper presents the particle-size distributions found in work areas by routine air sampling. Autoradiographic examination of the air samples was utilized to determine the equivalent spherical particle sizes in a range from about 0.05 to 10 microns. Aerosols consisting of essentially non-radioactive inert materials and plutonium oxide or plutonium carbide were examined. The plutonium particle distributions observed are compared to similar data in the literature on controlled oxidation or burning of plutonium with controlled atmospheres and conditions.
PLUTONIUM AEROSOL PARTICLE SIZE DISTRIBUTIONS IN ROOM AIR

INTRODUCTION

The distribution of plutonium particle size in work area aerosols is of interest to health physicists and to radiation biologists for a variety of reasons. Among these, the principal ones are evaluation of the adequacy of respiratory protection equipment, analysis of low level chronic internal exposure and the relationship of particle size to the biological excretion patterns. Extensive efforts have been spent in the study of radioactive aerosols and their effects under laboratory or controlled conditions. Similar data for aerosols in work areas in plutonium handling facilities is not commonplace. This paper describes the plutonium particle size distributions observed for plutonium oxide and plutonium carbide in work areas at Hanford.

Methods

Air Sampling

The air samples were taken with the routine air sampling program equipment which consists of a central vacuum system as the air mover coupled to 4" x 4" or 4" x 8" fixed filter samplers. The samplers are located about six feet above floor level and within three feet of the area of prime interest. Routine samples are collected on Hollingsworth and Vose Type 70 filter paper with samples changed on a shiftwise to weekly schedule.

The aerosol samples taken for the determination of the plutonium oxide particle size were taken with a 4" x 4" membrane filter paper which had a pore size of about 2 microns. The larger pore size permitted substitution of the membrane filter for the H-70 without greatly disturbing the sample flow rates at other locations on the central vacuum system. The membrane filter paper seemed to show good efficiency for submicron particles although the actual efficiency was not measured. This was indicated by the results of autoradiographic examination of the filters which indicated that the particles were generally collected on the surface of the filter. The aerosol samples used for plutonium carbide particle size determination were taken with H-70 filters.

Autoradiographic Exposure

The aerosol filters were mounted in exposure cameras consisting of standard 5" x 7" Kodak X-ray exposure holders which were altered so that the nuclear track film could be positioned reproducibly. The filters were contact-exposed to standard Kodak nuclear track plates, Type NTA, which have a 25 micron film emulsion mounted on a 2.5 x 7.6 centimeter glass slide. Film exposure times were from 12 minutes to 2,000 hours in decade increments. Standard darkroom procedures were employed in film plate development, fixing and washing. No protective covering was placed between the filter and film to permit the closest contact possible. No problems were encountered with contamination of the film.
Particle Sizing

The 2,000 hour exposure film was examined with an optical microscope at 430X power. The number of alpha tracks relating to each particle with total tracks between 5 and 50 was counted and recorded along with the slide location as measured with the stage micrometer. The tracks below 5 in number that seemed to be unrelated were ignored and those greater than 50 were estimated as to total number and their slide location recorded. The appropriate shorter exposure film slide was then utilized to determine the track number for these specific locations. The exposure camera and microscope stage micrometer permitted sufficient reproducibility in film location so that the particle in question could be readily relocated. In most cases, the alpha tracks from several hundred particles were counted and recorded, although in several cases, there were insufficient particles present and as few as 50 particles were examined for a specific aerosol sample.

The tedious, time consuming track counting determines the number of samples that can be examined. The American Society for Testing and Materials recommends sizing of about 1,000 particles or examination of one per cent of the total filter area. In the case of the 4" x 8" filter, the area examined was 2.7 per cent; for the 4" x 8" filter, about 1.3 per cent.

Particle Size Collection

The number of tracks counted per particle is converted to disintegration per minute using appropriate correction factors for geometry, and self absorption similar to techniques used by Yagoda. Calculations were made relating particle diameter to alpha activity in disintegrations per minute, assuming spherical particles of plutonium oxide and plutonium carbide with densities of about 11.5 and 13.5 respectively. A correction factor was included for the isotopic content of the plutonium aerosol in question. The plutonium aerosols examined were composed primarily of Pu239 but small amounts of Pu240, Pu241, and Pu242 were also present. Analysis of several possible mixtures have been reported by the Argonne National Laboratories. The relationship between particle size and alpha activity for pure Pu239 O2 is shown in Figure 1. In Figure 1, it is shown that particles of 0.01 d/m, 0.1 d/m and 1 d/m have diameters of 0.25, 0.52 and 1.1 microns respectively. Ten fold increases in radioactive content occur as particle diameters about double.

The cumulative mass and number fractions were then plotted on logarithmic probability paper on the assumption that the particle distribution was log normal in nature. This assumption should be valid and a straight line obtained although a tailing off of the upper and lower ends is expected since the method does not permit measurement of the smaller particles and the larger particles will not appear because of the nature of the source and the particle removal by settling. The mass mean diameter, geometric mean diameter and standard deviation were determined from the best straight line from the log normal plot. An excellent review of particle size measurement and interpretation of distributions and other applications may be found in a recent book by Irani and Callis.
FIGURE 1

Calculated Relationship Between Diameter and Alpha Activity for Pu$^{239}$O$_2$
Results

A series of aerosol filters taken during a 16-day period at a work location adjacent to a large hood containing large amounts of plutonium oxide were examined for particle size distribution. A membrane type filter media was utilized. The mass and geometric log normal distribution plots from one of these filters is shown in Figure 2. The particle size distribution plots for the other samples are shown in Appendix A at the end of this report.

A summary of the particle size distribution data resulting from these filters is shown in Table I.

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Sample Time Hours</th>
<th>GMD Graph</th>
<th>GMD Calc</th>
<th>MMD</th>
<th>( \sigma_g )</th>
<th>Particles Examined</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>72</td>
<td>.06</td>
<td>.05</td>
<td>1.3</td>
<td>2.8</td>
<td>303</td>
</tr>
<tr>
<td>2</td>
<td>24</td>
<td>.05</td>
<td>.04</td>
<td>0.11</td>
<td>1.8</td>
<td>147</td>
</tr>
<tr>
<td>3</td>
<td>24</td>
<td>.10</td>
<td>.11</td>
<td>0.6</td>
<td>2.1</td>
<td>413</td>
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<tr>
<td>4</td>
<td>24</td>
<td>.05</td>
<td>.16</td>
<td>1.6</td>
<td>2.4</td>
<td>161</td>
</tr>
<tr>
<td>5</td>
<td>24</td>
<td>.06</td>
<td>.04</td>
<td>0.7</td>
<td>2.7</td>
<td>404</td>
</tr>
<tr>
<td>6</td>
<td>72</td>
<td>.07</td>
<td>.07</td>
<td>1.1</td>
<td>2.6</td>
<td>514</td>
</tr>
<tr>
<td>7</td>
<td>24</td>
<td>.04</td>
<td>.35</td>
<td>4.2</td>
<td>2.5</td>
<td>378</td>
</tr>
<tr>
<td>8</td>
<td>24</td>
<td>.06</td>
<td>.005</td>
<td>0.2</td>
<td>3.0</td>
<td>371</td>
</tr>
<tr>
<td>9</td>
<td>72</td>
<td>.06</td>
<td>.21</td>
<td>1.1</td>
<td>2.1</td>
<td>875</td>
</tr>
<tr>
<td>10</td>
<td>24</td>
<td>.06</td>
<td>.30</td>
<td>0.8</td>
<td>2.0</td>
<td>244</td>
</tr>
</tbody>
</table>

MMD - Mass Mean Diameter
GMD - Geometric Mean Diameter
\( \sigma_g \) - Geometric Standard Deviation

Log GMD (calc) = Log MMD - 6.91 Log²\( \sigma_g \)

No known incidents occurred during the 16 days of continual sampling. The hood has many glove ports and some undetected deterioration of hood gloves may have occurred.

Table I includes a calculated geometrical mean diameter based upon log normal distribution for comparison purposes. It appears that the agreement between calculated values and those obtained graphically is good enough to indicate that particle distribution is essentially log normal in nature. If it were possible to measure the geometric particle size below 0.01 microns, we would probably find that the actual geometrical mean diameter (GMD) would actually be less than the 0.04 to 0.1 microns measured. Other investigators (8,9,10) have shown that atmospheric dust has a geometric mean diameter of about 0.028 microns; sodium oxide from burning sodium metal has a GMD of 0.04 microns, and separations process stack effluent has a GMD of about 0.05 microns.
FIGURE 2
Plutonium Oxide Particle Distribution at Work Site

Cumulative Mass/Number Fraction - Percent

Diameter of Equivalent Spherical Particle Microns

MMD = 1.1 μ
GMD = 0.07 μ
σg = 2.6
Six routine air samples taken at work locations where plutonium carbide was being processed were examined for particle size distribution. The summary results are shown in Table II with the actual distribution curves shown in Appendix B.

### TABLE II

<table>
<thead>
<tr>
<th>Sample</th>
<th>GMD Graph</th>
<th>MMD</th>
<th>$\sigma_g$</th>
<th>Maximum Particle Diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.11</td>
<td>0.80</td>
<td>1.5</td>
<td>1.4</td>
</tr>
<tr>
<td>2</td>
<td>0.25</td>
<td>0.64</td>
<td>1.7</td>
<td>1.2</td>
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<tr>
<td>3</td>
<td>0.27</td>
<td>0.49</td>
<td>1.6</td>
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<tr>
<td>4</td>
<td>0.23</td>
<td>0.66</td>
<td>1.8</td>
<td>1.7</td>
</tr>
<tr>
<td>5</td>
<td>0.30</td>
<td>0.50</td>
<td>1.6</td>
<td>1.2</td>
</tr>
<tr>
<td>6</td>
<td>0.44</td>
<td>0.64</td>
<td>1.4</td>
<td>1.7</td>
</tr>
</tbody>
</table>

**Notes:**
- **GMD** - Mass Mean Diameter
- **GMD** - Geometric Mean Diameter
- $\sigma_g$ - Geometric Standard Deviation

The maximum film exposure time used in the plutonium carbide series was 200 hours. The results obtained were more consistent than those from plutonium oxide.

**Discussion**

K. Stewart has studied the particle size distribution as released from plutonium metal oxidized in air at room temperature at a number of humidities. He found that the mass mean diameter increased from about 0.5 microns under dry conditions to about 10 microns under very humid conditions. Our results show a mass mean diameter of about 1 micron in our relatively dry conditions at Hanford. The results in Table I range from 0.1 to 4.2 microns with most of the samples showing a mass mean diameter of around one. The low mass mean diameters were probably the result of no larger particles being airborne from the hood source or lack of employee traffic in the work area which would tend to keep the larger particles from being airborne.

The mass mean diameters found for the plutonium carbide aerosol all were below one micron. Practical experience by radiation monitoring people has indicated that the particle size tends to be smaller than plutonium oxide based upon the observation that in a number of instances when it was known that a plutonium carbide release had occurred, no severe contamination of horizontal surfaces resulted. The plutonium carbide was found in the absolute filters in the ventilation system.

The particle size data from the ten samples of plutonium oxide aerosol summarized in Table I was composited into a single set of data. The size distribution obtained
is shown in Appendix A. A total of 3857 particles were sized for the ten samples. The largest one per cent (38 particles) of the particles were tabulated for the composite sample and also for each of the ten samples. These numbers are shown in Table III.

### TABLE III

**Summary of Largest One Per Cent of Particles Measured in PuO₂ Aerosol Samples**

<table>
<thead>
<tr>
<th>Diameter Microns</th>
<th>Number Particles Deposited (Each Filter 1%)</th>
<th>Number Particles Deposited (Composite 1%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>2 - 3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>1.5 - 2</td>
<td>3</td>
<td>6</td>
</tr>
<tr>
<td>1.0 - 1.5</td>
<td>15</td>
<td>27</td>
</tr>
<tr>
<td>0.9 - 1.0</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>0.5 - 0.9</td>
<td>6</td>
<td>-</td>
</tr>
<tr>
<td>0.2 - 0.5</td>
<td>7</td>
<td>-</td>
</tr>
</tbody>
</table>

When considering all of the particles measured on the 10 PuO₂ filters, it was seen that about 85 per cent of the plutonium is found in the largest one per cent of the particles. These particles as seen in Table III ranged in size from 0.9 to 4 microns. All of the particles measured are in a size range that is commonly considered respirable and of interest in internal exposure evaluation. The data indicate that during routine working conditions the respirable air concentration is probably not over-estimated by filter samples and use of a two-stage sampler to remove large particles which are not likely to enter the lungs is not necessary. Filter samples examined for particle size during known releases have generally shown only small particle diameters, although in one instance where a carton of PuO₂ powder was spilled the maximum particle size found on a nearby air sample was on the order of 10 microns.

**Future Work**

The knowledge of plutonium particle size from a particular release may be of value in assessing the possible internal deposition incurred by exposed workers. Particle size may have a real effect on the observed excretion curve which may be quite different for workers receiving an acute exposure compared to the chronic exposure. The correlation of particle size to body burden and excretion pattern will be attempted if the opportunity arises. The Hanford Biology group has concluded that particle size and chemical form caused marked differences in the fate of inhaled plutonium aerosol.

The work presented in this paper assumes that the plutonium particles were not attached to dirt particles. The state of attachment of the plutonium probably depends strongly on the source. In our case, copious quantities of plutonium are generally involved during particle formation so that the effect of foreign particles is probably not extensive. The use of stripping film emulsion applied
directly to the membrane filters is planned in an attempt to look at the alpha
tracks and particle simultaneously according to the method reported by J. T. Quan. (10)
Similar studies were conducted by D. C. Stevens in Harwell, England. (11)

Conclusions

The airborne particle size distribution at work sites where large amounts of
plutonium oxide and plutonium carbide are processed was determined. The use of
a two-stage air sampling device is not warranted for routine plutonium aerosol
sampling. The results obtained indicate agreement with controlled oxidation of
plutonium and release as aerosols as studied by Stewart.

Acknowledgment

The author wishes to express his appreciation to Evelyn L. Skager who carried
out the time consuming alpha track counting of the autoradiographic exposures.
REFERENCES

(6) P. G. Stoddart, Argonne National Laboratory, ANL-6759 (1963).
(9) J. J. Fitzgerald, Knolls Atomic Power Laboratory, KAPL-1015 (1954).
Appendix A

Appendix A contains graphical presentations of the particle distributions for the individual aerosol taken of the plutonium oxide aerosol. It also includes a composite particle distribution for the ten samples examined.
**FIGURE A.1**
Size Distribution of Plutonium Oxide Aerosol At Work Site

**FIGURE A.2**
Size Distribution of Plutonium Oxide Aerosol At Work Site
FIGURE A.3
Size Distribution of Plutonium Oxide Aerosol At Work Site

FIGURE A.4
Size Distribution of Plutonium Oxide Aerosol At Work Site
**FIGURE A.5**

Size Distribution of Plutonium Oxide Aerosol At Work Site

- MMD = 0.65 µ
- σg = 3.20
- Membrane Filter
- Oxide Hood - 24 hours

**FIGURE A.6**

Size Distribution of Plutonium Oxide Aerosol At Work Site

- MMD = 1.1 µ
- σg = 2.5
- Membrane Filter
- Oxide Hood - 72 hours
FIGURE A.7
Size Distribution of Plutonium Oxide Aerosol At Work Site

MMD = 4.2 μ
σg = 2.5
Membrane Filter - Oxide Hood = 24 hours

FIGURE A.8
Size Distribution of Plutonium Oxide Aerosol At Work Site

MMD = 0.2 μ
σg = 3.0
Membrane Filter - Oxide Hood = 24 hours
FIGURE A.9
Size Distribution of Plutonium Oxide Aerosol At Work Site

MMD = 1.1 µ  
σg = 2.1
Membrane Filter - Oxide Hood - 72 hours

FIGURE A.10
Size Distribution of Plutonium Oxide Aerosol At Work Site

MMD = 0.8 µ  
σg = 2.0
Membrane Filter - Oxide Hood - 24 hours
FIGURE A.11

Composite of Figures A.1 through A.10
Size Distribution of
Plutonium Carbide Aerosol at Work Site
Appendix B

Appendix B contains graphical presentations of the particle size distributions for the individual aerosol samples taken of the plutonium carbide aerosol. It also indicates a composite particle distribution for the six samples examined.
FIGURE B.1
Size Distribution of Plutonium Carbide Aerosol At Work Site

FIGURE B.2
Size Distribution of Plutonium Carbide Aerosol At Work Site
FIGURE B.3
Size Distribution of Plutonium Carbide Aerosol At Work Site

FIGURE B.4
Size Distribution of Plutonium Carbide Aerosol At Work Site
FIGURE B.5
Size Distribution of Plutonium Carbide Aerosol at Work Site

FIGURE B.6
Size Distribution of Plutonium Carbide Aerosol at Work Site
FIGURE B.7

Composite of Figure B.1 through B.6

Size Distribution of

Plutonium Carbide Aerosol at Work Site