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HW-SA-3197

CONF-279-1

PARTICLE SIZE DISTRIBUTION FOUND IN AIR
AT HANFORD PLUTONIUM FABRICATION FACILITIES

MASTER

by

B. V. Andersen
Radiological Development and Calibrations

September 12, 1963

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Prepared for Presentation at
Seventh Annual Western
Industrial Health Conference
San Francisco, California
September 27 and 28, 1963

Work Performed Under Contract No. AT(45-1)-1350
Between The Atomic Energy Commission
And General Electric Company

RADIATION PROTECTION OPERATION
HANFORD LABORATORIES

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B. V. Andersen

Radiological Development and Calibrations
Radiation Protection Operation
HANFORD LABORATORIES

ABSTRACT

The conventional sampling methods of filtration and impaction are generally utilized in the collection of air samples at Hanford. In addition, a Goetz aerosol spectrometer is available for special studies. Particle-size determinations are made by autoradiographic studies making use of both optical and electron microscopes. A supplementary tool sometimes utilized is the Royco Particle Counter. The study included an analysis of the potential usefulness and the limitations of these air sample study methods for plutonium aerosol particle size measurement. Preliminary results, from a continuing study of the particle size distribution in normal and abnormal air, show particle sizes ranging from hundredths to tens of microns. The studies include considerations of the usefulness of particle size data in planning biological experiments and in the application of these data to human internal deposition cases.

PARTICLE SIZE DISTRIBUTION FOUND IN AIR
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Plutonium and plutonium recycle research and development studies are principal products of the Hanford Atomic Products Operation. Each day, large quantities of plutonium and plutonium-containing materials are processed at the Hanford facilities. This processing is conducted in large sealed cells or processing hoods. Radiological design and engineering features provide for positive containment of plutonium materials at all times. In general, work with plutonium materials is performed remotely; however, maintenance and special processes do require the use of rubber hood gloves or plastic ports for specific operations. In general, these methods of handling plutonium have proven to be highly satisfactory. Glove ruptures or failures in hood seals occasionally result in minor amounts of plutonium being released to the building atmosphere. The very strict controls required on the concentration of plutonium in breathing⁽¹⁾ air and the very small maximum permissible body burden of plutonium, as shown in Slide 1, require that a dependable air sampling program be conducted in plutonium processing areas. To assure adequate protection and, in fact, to evaluate the consequences of plutonium releases to the breathing air, thorough analysis of plutonium air samples is required.

The current Hanford studies include a development program to provide air sample evaluation by characterizing plutonium particle sizes and size distributions as found in breathing air.

Air Sampling and Evaluation Methods

The routine air sampling program at Hanford consists of central vacuum systems with sufficient capacity to sample from 5 to 10 CFM of room air through 4 inch by 4 inch or 4 inch by 8 inch Hollingsworth and Vose Type 70 asbestos-based filter paper. The collection filters are changed according to the degree of interest on an eight-hour basis to as long as a weekly basis. There are several thousand such air samplers operated continually at Hanford. These filters provide the main source of air sample material on which particle-size analyses are made after cases of acute exposure to a plutonium aerosol. In a few locations judged to be of greater interest or greater potential hazard based upon work activities, the filter used is a 4 inch by 4 inch or 4 inch by 8 inch membrane-type filter. This membrane filter is more amenable to particle size determination work, especially for electron microscope viewing of particles.

All other air sampling methods discussed are utilized only in special studies and are not routinely operated.

Cascade Impactor

The cascade-type impactor sample was used extensively at Hanford to characterize particle sizes in stack effluent systems and to check effluent monitoring system efficiencies. However, it was not used in this study and is only mentioned here as a collection device that can be utilized.

A Goetz aerosol spectrometer⁽²⁾ is currently used at Hanford to characterize small particle diameters. The Goetz spectrometer process of separating the particles from the air-borne state is based upon the application of high centrifugal forces to a continuous laminar flow of the aerosol under conditions

which cause the fallout of particles to follow Stokes' law. The flow is confined to one pair of identical helical channels of rhomboidal cross-section on the periphery of a conical rotor; the axis of the rotation coincides with that of the helix. The air flow enters the channels near the top and leaves at the wide end of the rotor. The outer surface of the helix channel is the place where the air-suspended particles are deposited. A foil or filter paper is placed on this outer surface to collect the particles. The spectrometer is capable of collecting particles in a spectrum fashion from about 4 microns in diameter down to about 0.025 microns in diameter when tested with monodisperse latex spheres of unit density.

Royco Particle Counter

A Royco Particle Counter PC-200-A is used to size non-radioactive particles in various building atmospheres at Hanford. Aerosol particles are passed in a stream through the sensing region of the optical system. This region is intensely illuminated by means of a light source. Particles entering it scatter incident light, a fraction of which proceeds to a photomultiplier tube. Electrical signals from the photomultiplier tube appear in the form of pulses which are related to particle sizes. The instrument has a particle size range from 0.3 to 8 microns and is not dependent on particle density. The instrument is suitable for dust counting in clean rooms and other dust counting applications. J. M. Selby has previously reported⁽³⁾ results obtained with this instrument and a dust particle distribution curve from his report is shown in Slide 2.

This type of instrumentation is not of use in plutonium particle size analysis because there are insufficient plutonium particles in breathing air even at maximum permissible concentrations to yield significant particle counts.

This sampler cannot differentiate between dust and plutonium particles. A comparison between the plutonium maximum permissible concentration in air and the number of particles required per cubic foot to produce this concentration is shown in Slide 3.

Electron Microscopy of Air Filters

Electron micrographs of particles deposited on membrane filter paper are used to measure particle sizes of air-borne plutonium aerosols. These samples were validated by applying a liquid nuclear track film emulsion to the specimens and examining the electron micrograph for alpha tracks. The plutonium particle is also shown on the same micrograph. However, when attempting to obtain a sample of ordinary room air, it was found that the dirt and other foreign substances on the membrane filter tend to obscure the plutonium particle and make the determination of particle size distribution very difficult and time consuming. Several micrographs are shown in Slide 4.

These micrographs are of the same filter area, with the only difference being that the second has had a coating of liquid nuclear track emulsion applied and exposed for about four days before development and examination with the electron microscope.

In the upper micrographs, it is apparent that the particles seen are not plutonium oxide since no alpha tracks are seen in the second micrograph which has the nuclear track emulsion present. In the lower-right micrograph, the alpha tracks are present but the many small particles seem to have vanished. They may have been dislocated during the filming process. The alpha tracks appear to be related to the single large (~1.5 micron) particle. The number

of disintegrations seen related to the film exposure time, indicates that the particle seen is probably a pure PuO_2 particle.

Autoradiographic Examination of Air Filters

The 4 inch by 4 inch or 4 inch by 8 inch asbestos type filters used routinely, supply a source of air samples that represent the normal and abnormal in plutonium air concentrations. If these are not utilized, special sampling is required to obtain materials for analysis. It is extremely unlikely that a special sample would be taken when an unexpected release of plutonium to breathing air occurs. For these reasons, the generally accepted method of autoradiographing the filters was employed to get a measure of the particle size distribution. In this method, previously reported by Leary⁽⁴⁾, the filter is exposed to nuclear track film and the number of tracks counted on a microscope for each particle detected. The plutonium particle size can then be calculated by assuming a particle density for the radioactive material of concern. The major limitations to the method are sensitivity and the tedious, time consuming, track counting. Since particles of all sizes are distributed randomly on the filter paper, an exposure time exists beyond which the tracks from larger particles tend to obscure the smaller ones. A practical range of sizes for detection of plutonium oxide spheres by this method seems to be from 0.1 micron to 10 microns.

Film exposures of each filter to the nuclear track film was performed in successive steps with times of 12 minutes, 120 minutes, 1,200 minutes, and 12,000 minutes employed. Each film slide was examined microscopically and the tracks counted for those particles which produced between 5 and 50 tracks on the film. This data was then utilized to calculate the plutonium oxide

particle diameter. It was assumed that the particles were spherical pure plutonium oxide with a density of 11.46. Radiomicrographs of the alpha tracks for some typical particles are seen in Slide 5.

The alpha track photos shown represent exposure time of 120 minutes, 1,200 minutes and 12,000 minutes for typical areas of one air sample. It is obvious that counting the tracks present in some cases would be difficult because of their large number and the overlap between alpha tracks from neighboring particles.

Results

The particle distribution data is presented as a plot of the cumulative percentage of number of particles up to various stated sizes against diameter on logarithmic probability paper. A straight line is usually found for most dusts, mists or smokes. This plot, as shown by Drinker and Hatch⁽⁵⁾ permits determination of the geometric mean diameter quickly and the geometric standard deviation can be calculated.

The mass mean diameter was determined by a similar plot of radioactive content, which is directly related to mass for pure PuO_2 , against diameter. The geometric standard deviation is the same for mass mean diameter and count mean diameter. A bar graph is also shown for each sample comparing the percentage of particles and radioactivity in the various diameter groups examined.

The first air sample results shown in Slides 6 and 7 are from a routine air sample taken when a can containing plutonium oxide powder was opened and a visible puff of powder was distributed throughout a room. It is seen in the illustration that the count mean particle diameter was about one micron

with a considerable number of particles in the 1 to 10 micron diameters present. The mass mean diameter is 6.3 microns. The number of large particles present on this filter prevented detection of any particles below 0.5 microns although there were probably a significant number present.

Results from another sample are shown in Slides 8 and 9. The air sample analyzed was a routine air sample taken when a small plutonium fire occurred in a hood, burning the hood gloves sufficiently to release some plutonium oxide to the room.

The log normal plot indicated a count mean particle diameter of about 0.4 microns which is considerably smaller than observed for the plutonium powder puff and with no particles detected larger than about 4 microns. In this case although the majority of the particles are of the smaller sizes, the radioactivity is still found to be mostly in larger-sized particles. The mass mean diameter is 1.5 μ .

The particle size distributions shown in Slides 10 and 11 were taken from a routine air sample that indicated a high plutonium air concentration for no known reason. It is noted that the count mean particle diameter from the probability plot is about 0.15 microns with the maximum particle diameter detected of about 2 microns. The majority of the radioactivity in the sample still is found in the relatively minor number of larger particles present.

The last typical particle distribution is presented in Slides 12 and 13. This air sample was taken in an exhaust duct after the hood air was filtered by two high efficiency Cambridge type filters but before final filtration and release to the atmosphere. The count mean particle diameter from the probability plot is about 0.24 microns with 99 per cent of the particles less

than about 0.9 microns in diameter. In this case, the hood is used for ball milling plutonium oxide, sizing the powder and then blending it with other materials in the development of various reactor fuel materials. Thus, the loading on the first absolute filter is quite high. These results are shown to emphasize the need for several filtration steps in hood systems when excessively contaminated atmospheres exist. It would also appear that the presence of an absolute filter or two does not give one free license to create highly contaminated atmospheres within a hood.

The few examples shown are believed to be typical of the particle-size distributions found in the routine air sampling program in our plutonium metal fabrication and experimental facilities. Further studies will be conducted on routine air samples involving other mixtures or compounds of plutonium such as plutonium carbide and plutonium and uranium oxide mixtures.

Goetz Spectrometer and Autoradiographic Particle Size Determination

Preliminary work on application of the Goetz spectrometer to determine particle size of PuO_2 in breathing air was performed. Since PuO_2 has a density of 11.46 and the spectrometer is calibrated with unit density latex spheres, the actual size range that the spectrometer will collect in a dispersed fashion is considerably smaller than the calibration minimum of 0.025 microns. A table comparing particle diameter for materials of several densities is shown in Slide 14. In the Goetz spectrometer, the PuO_2 particles in a pure state should be removed and distributed along the path length with particle diameters from 0.005 microns to about 2 microns.

Unfortunately, there is one more complicating factor that should be considered in the PuO_2 particle size distribution so that results can be

applied to the respirability of particles. This is the question of whether the PuO_2 particle is actually traveling by itself or whether it may be attached to a dust or other foreign particle present in the air stream. Long exposure autoradiographs of the Goetz spectrum will permit us to locate the path length of the particle and provide one with an estimate of the apparent Stokes diameter. Then track counting techniques should enable us to determine the apparent real diameter based upon a spherical particle of pure PuO_2 . From these measurements, one should be able to logically deduce whether or not the plutonium oxide particle is in fact traveling by itself or attached to a foreign particle. This work is currently in progress and will be reported in the literature when complete. This type of analysis involves long term film exposure and tedious microscopic counting of tracks. Film exposures for periods up to 3 months may be required to detect the smaller particles of PuO_2 .

Another film technique that will be tried on particles of a size visible under the microscope is the use of a stripping film emulsion applied directly to the membrane filter material. The film and filter are then handled as a unit with the film development and examination of the alpha tracks and physical particle under the microscope conducted with the film and filter in precise alignment with each other. The result is that the alpha tracks can be assigned to definite physical particles and should provide a good indication of the state of attachment of PuO_2 to other aerosol particles. The method used will be similar to that reported by J. T. Quan in the American Industrial Hygiene Journal.⁽⁵⁾ The major problem with this method is the sizeable burden of ordinary dirt particles present on the filter compared to the few radioactive particles present. Similar work with glass fiber filter paper was published by D. C. Stevens in Harwell, England.⁽⁶⁾

Biological Interest in Particle Size

The latest summary of the respirability and deposition of particles based upon particle diameter is shown in Slide 15, as presented by the National Academy of Sciences in a special report on "Effects of Inhaled Radioactive Particles." (7)

The current understanding of particle retention is quite good down to particle diameters of about one tenth micron. Below this diameter, the problem was not studied as extensively since the methods of particle size analyses became more exacting and difficult. The study of actual aerosols at Hanford is designed to eventually determine the existing size distributions in routine operation. These results can be utilized in interpretation of animal experiments with plutonium aerosols. The Hanford Biology Laboratories are conducting significant experiments of this nature, many of which are reported in the literature. (8) They have concluded that particle size and chemical form cause marked differences in the fate of the inhaled plutonium in the body. They also have shown that the rate of urinary excretion of plutonium in dogs is quite dependent upon the characteristics of the inhaled plutonium aerosol. Since the currently accepted practice of determining body burdens of plutonium in man (9) is based upon urinary and fecal excretion rates, further studies of this type are needed.

Summary

The air sampling and particle size evaluation methods and equipment were described. The results presented indicate that PuO₂ particle size in breathing air is a direct function of the source of the particles. It is shown that in all cases the great majority of the total PuO₂ present is found in the larger

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diameter particles. All of the PuO_2 particles examined were in the size range that are respirable and deposit in significant quantities in the lungs.

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SLIDE 1

Characteristics and Limits
For Selected Radionuclides(1)

<u>Isotope</u>	<u>Half Life</u>		<u>MPC_{air}</u> <u>uc/cm³</u>	<u>MPBB</u> <u>.uc</u>
	<u>T_{Phys.}</u>	<u>T_{Biol.}</u>		
Pu-239	24,000 yrs.	175 yrs.	2×10^{-13}	0.04
Ra-226	1,622	2.5	3×10^{-11}	0.1
Sr-90	28	16	3×10^{-10}	2.0

*40 hour week

Slide 2

PARTICLE SIZE DISTRIBUTION 234-5 BUILDING

Slide 3

PuO_2 PARTICLE SIZE VS PARTICLES PER CUBIC FOOT FOR 1 MPC_A

SLIDE 4

Electron Micrographs of Membrane Filter Air Samples
On SiO_2 Substrate Pre- and Post Autoradiograph

| 10 Microns |

SLIDE 5

Plutonium Alpha Tracks
As Seen from Nuclear Track Emulsion

| 100 Microns |

SLIDE 6

PuO₂ Particle Distribution From Air Sample
Taken During Release of Cloud of PuO₂ Powder

SLIDE 7

PuO₂ Particle Distribution From Air Sample
Taken During Release of Cloud of PuO₂ Powder

SLIDE 8

PuO₂ Particle Distribution From Air Sample
Taken During Pu-239 Fire In Hood

SLIDE 9

PuO₂ Particle Distribution From Air Sample
Taken During Pu-239 Fire In Hood

SLIDE 10

PuO₂ Particle Distribution From Room Air Sample
Plutonium Metal Fabrication Building

SLIDE 11

PuO₂ Particle Distribution From Room Air Sample
Plutonium Metal Fabrication Building

SLIDE 12

PuO₂ Particle Distribution From Air Sample
Taken In Ventilation Duct Downstream From Two Absolute Filters

SLIDE 13

PuO₂ Particle Distribution From Air Sample Taken
In Ventilation Duct Downstream from Two Absolute Filters

SLIDE 14

Equivalent Particle Diameter for SiO₂ and PuO₂
Compared With Unit Density

Material	<u>H₂O</u>	<u>SiO₂</u>	<u>PuO₂</u>
<u>Density (g/cm³)</u>	<u>1.00</u>	<u>2.60</u>	<u>11.46</u>
Diameter (μ)	0.010	0.0073	0.0044
	0.025	0.018	0.011
	0.050	0.036	0.022
	0.10	0.073	0.044
	0.50	0.36	0.22
	1.00	0.73	0.44
	4.0	2.9	1.8

SLIDE 15

Deposition In Respiratory Tract