**PLUTONIUM RELEASE STUDIES
III. RELEASE FROM HEATED
PLUTONIUM BEARING POWDERS**

J. MISHIMA, L. C. SCHWENDIMAN
AND C. A. RADASCH

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ABSTRACT

Four plutonium compounds in powder form--the oxide, partially oxidized oxalate, oxalate, and fluoride--were subjected to high temperature in air drawn up and around the material at various velocities. Under the conditions of these experiments, the greatest amount, at a rate of 0.90 wt%/hr, was released by plutonium oxalate at 700 °C in an upsweep of air at 100 cm/sec. The partially oxidized oxalate, the most readily entrained, showed significant release rates at all air velocities at 1000 °C, and at all temperatures in air moving at 100 cm/sec. Release rates ranged from 0.25 to 0.82 wt%/hr at 1000 °C and 0.057 to 0.82 wt%/hr in air with a velocity of 100 cm/sec. The smallest fraction was released by the fluoride during the runs, 0.05 wt%/hr at 1000 °C in an air velocity of 100 cm/sec. The residue remaining from the overheating of the fluoride appeared susceptible to further entrainment.

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INTRODUCTION

Many plutonium-bearing powders are involved in the production of high purity plutonium⁽¹⁾ and in the fabrication of reactor fuels.⁽²⁾ Powders subjected to the proper conditions of temperature, atmosphere and gas velocity may oxidize and the fine particulate matter formed may become airborne. The characteristics and fraction of the airborne particles depend upon many variables, i.e., physical division and characteristics of the starting material, airflow, the stresses to which the material is subjected prior to entrainment, etc. When considered in terms of a fire involving plutonium-bearing powders, the degree to which the oxide or other compounds are released to the atmosphere is of great importance in the evaluation of the accident consequences.

A review of available data on the oxidation rate and fractional release of plutonium during overheating was previously reported.⁽³⁾ This review also noted the need for further study of the behavior of plutonium and its compounds in overheating situations. The results of the experiments using small metallic plutonium pieces⁽⁴⁾ and bulk metal⁽⁵⁾ have already been reported. Further studies are planned to provide additional information for use in evaluating the hazards of fires to plutonium and its compounds.

Various studies have reported the amounts of airborne plutonium found in room air in plutonium processing areas.⁽⁶⁻¹⁰⁾ The amounts present [usually under the ICRP value for a 40-hr week, 2×10^{-12} $\mu\text{Ci}/\text{cm}^3$ of air⁽⁷⁾] and

the particles measured [MMD 4.2 to 0.1 μ ⁽⁸⁾] are small. These particles escape through small leaks in the protective systems (generally small punctures in rubber gloves). Passage by diffusion is a selective process and should result in only the smaller particles escaping. Hunt⁽¹¹⁾ has calculated the effects of various factors upon the attenuation of a source by its leak path.

There remains a need for quantitative information on the particle size distribution of the oxides produced under various circumstances, and on the amount and size distribution of airborne particles for plutonium hazards evaluation.⁽¹²⁻¹⁴⁾ Several authors have described the behavior of various plutonium-bearing materials. Drumond and Welch⁽¹⁵⁾ describe the dioxide produced by the ignition of air or oxygen of various materials.

TABLE I. Physical Properties of Plutonium Dioxide Ignited to 870 °⁽¹⁵⁾

Source	Color	Appearance
Sulphate	Yellow-green to green	Bulky powder
Nitrate	Dull yellow	Bulky solid
Chloride	Dull yellow	Powder
Fluoride	Khaki, traces of black	Granules
Oxalate	Yellow-buff	Bulky powder
Iodate	Buff	Very bulky
Hydroxide	Black, traces of yellow	Dense shiny particles

Myers⁽¹⁶⁾ described the decomposition of Pu(IV) oxalate and characterized the oxalate, dioxide, and fluoride produced. He found the oxalate to be tan in color with particles ranging from 5 to 7 μ in diameter. The color of the dioxide produced depended on the temperature of calcination, and ranged from an olive green at 300 °C to yellow green at 620 °C. The particle size was 2 to 5 μ in diameter.

The plutonium tetrafluoride obtained by hydrofluorination of the oxalate or oxide was salmon pink in color with particles 2 to 10 μ in size. Particle size was by X-ray analysis.

Potter⁽¹⁷⁾ reported the average particle size of oxide produced by calcination, as determined by gas permeability measurements, to increase with Increasing calcination temperature. Particle size ranged from 2.0 μ at 400 °C to 7.8 μ at 1200 °C. Oxide of 3.0- μ size originally calcined at 650 °C was found to increase to 5.7 and 6.2 μ when reheated to 800 and 1000 °C.

NUMEC (The Nuclear Materials and Equipment Corporation, Appollo, Pennsylvania) has conducted a continuing series of studies on Plutonium-Bearing Nuclear Fuel Materials.⁽¹⁸⁻²⁵⁾ Their data indicate particle size of the oxide produced by calcination of plutonium oxalate to depend upon conditions under which the precursor and oxide are prepared and upon the means of measurement.

Three methods used to measure particle size were air permeability (a measurement of void spaces and therefore the average size of the basic particle), air, and liquid sedimentation. The air sedimentation apparatus uses high pressures to deagglomerate and blow the particles into the system.⁽²²⁾ Under these conditions, the basic particle size is measured and agrees well with the air permeability values.⁽²²⁾ The particle size determined by the liquid sedimentation was dependent upon the means used to disperse the particles⁽²¹⁾ and probably indicated the degree of deagglomeration taking place.

The median mass diameter (air sedimentation) of the plutonium oxalate ranged from 3.8 to 6.5 μ , and oxide produced by calcining the oxalate at 740 °C ranged from 1.7 to 2.9 μ .⁽²⁰⁾ Oxide particles were 26 to 68% of the size

of their precursor. The size of the basic particle (air permeability), although much smaller in size, increased with calcination temperature⁽²⁰⁾ as noted by Potter.⁽¹⁷⁾ The MMD (liquid sedimentation) of the aggregates showed no correlation with temperature and ranged from 8 to 10 and 10 to 12 μ . The particle size was also sensitive to initial plutonium concentration used in preparing the oxalate,⁽²⁴⁾ and washing procedure.⁽²⁵⁾ Continuously precipitated oxalate produced coarser oxide (13 to 14 μ MMD--liquid sedimentation) than that from batch precipitated oxalate (MMD 1.7 μ). All size distribution curves indicated more than one slope^(21,22,24) and were similar to those obtained for the oxide produced by unalloyed metal above ignition temperatures.⁽⁴⁾

Four plutonium materials (in the form of powders) commonly found in the Hanford processing area were selected for this study. The powders were:

- Plutonium oxide--the 15 to 40 μ fraction obtained from the oxidation of metallic plutonium above ignition temperature.
- Partially oxidized plutonium oxalate made by allowing plutonium oxalate from the production process to oxidize in dry, glovebox air (dewpoint of the air was less than minus 70 °F) for 6 months.
- Plutonium oxalate from the processing line.
- Plutonium fluoride from the processing line.

In the work to be reported, small samples of the several materials held in a shallow dish were heated to temperatures to 1000 °C. Air was introduced in an annular space around the rim of the dish and rose upward in a chimney over the dish. The fraction airborne per unit time was determined for several air velocities in the chimney section.

SUMMARY

Plutonium compounds as powders are found in many nuclear applications. The characteristics of the material released in an overheating incident are of consequence to the evaluation of hazards arising from the incident. To supply data required for evaluating hazards involved in powders commonly encountered and used in processing facilities, four plutonium-bearing powders were subjected to temperatures of 400, 700, and 1000 °C. The amount and characteristics of the fine particulate material entrained in air at upsweep velocities of 10, 50, and 100 cm/sec were determined.

The results show:

- The amount re-entrained from a mass of finely divided oxide ranged from 5×10^{-6} to 0.025 wt%/hr and is in the range anticipated from published re-entrainment factors.

The material produced during the heating of partially oxidized plutonium oxalate was the most readily airborne at the lower flow rates of the powders studied. Observable rates were found with the three air velocities used at 1000 °C and at elevated temperatures in air at 100 cm/sec velocity. Rates ranged from 0.057 to 0.82 wt%/hr.

- The fractional release from "fresh" plutonium oxalate was a marked function of the air velocity used. Observable release rates were found only at the higher velocities. Rates ranged from 0.007 to 0.90 wt%/hr--the highest rate measured during this study.
- Although the plutonium fluoride contained the largest fraction of fine material in the original powder, the chemical and physical changes induced in plutonium

fluoride by heating resulted in generally lower release rates than were found for the partially oxidized and "fresh" plutonium oxalates. They ranged from 0.007 to 0.05 wt%/hr. The residues were free-flowing and subject to entrainment under the proper conditions.

- The latter three powders, more readily airborne than either the metal or oxide, may pose a greater potential hazard during overheating situations.

EXPERIMENTAL

RELEASE RATE APPARATUS

In order to obtain semiquantitative data useful in predicting plutonium material behavior in an accident situation, the material should be subjected to the predominant processes governing its release under controlled conditions. Since each accident situation is atypical (otherwise the accident would not have occurred) the conditions prevailing during accidents are unique and a wide range of conditions can be postulated. It is difficult to propose a single experimental apparatus that will simulate all accident conditions. Further limitations are imposed by the physical restrictions of working within the confines of an enclosure (glovebox). Therefore, a workable experimental arrangement was somewhat arbitrarily chosen in which powder specimens could be heated in a stream of moving air and the entrained particles collected.

All the generated material should be collected and subjected to an undisturbed size distribution. Under the temperature conditions contemplated, a method for collecting all the particles directly at the source was not practicable. The airborne material was therefore carried some distance by the air to a collector.

The manner in which the air strikes the material will undoubtedly affect both the amount entrained and the size distribution of the particles airborne. Since the material could not be sampled at the source, the flow direction (vertically or horizontally) and travel distance also would affect these values. Accident conditions can be postulated to suit any arrangement of air flow and direction. For simplicity and ease of manipulation within the enclosure, a chimney arrangement with air flow around and over the sample material was selected. The upsweep vertical flow is more representative of convective air currents and minimizes variation in particle deposition by gravitational settling.

To minimize the potential for cross contamination from the air within the glovebox, the drawing of air directly into the bell-shaped section of the chimney through a side arm was initially attempted. Building compressed air, suitably filtered, was introduced at a flow 10% greater than that drawn through the chimney. A single inlet was used slightly above the annular space between the dish and chimney. This arrangement caused the incoming air to jet upon the powder and produce high release results (see Table II).

TABLE II. *Comparison of Release Rates Obtained for Partially Oxidized Plutonium Oxalate With and Without Uncontaminated Air Supply (wt%/hr released at stated air velocity)*

<u>Temperature</u>	<u>50 cm/sec</u>		<u>100 cm/sec</u>	
	<u>With</u>	<u>Without</u>	<u>With</u>	<u>Without</u>
Ambient	0.76	<0.003	--	0.33
1000 °C	0.72	0.007	7.7	0.18

In the runs using the uncontaminated air, the glass fiber filters were so heavily loaded that the collected material could be readily dislodged while removing the filter. During runs at ambient temperature and 100 cm/sec, 60 to 70% of the powder was lost from the dish during a one hr period. A large portion of this was carried away within the first few minutes of the run. Under the same conditions, but without this source of air, less than 0.4 wt% was removed from the dish.

Glovebox air samples taken during the experimental run showed the contamination resulting from the use of glovebox air as sweep air to be insignificant compared with anticipated release rates. Table III shows the activity found.

TABLE III. *Activity Found in Glovebox Air During Experimental Runs (Using membrane filters, 1 hr sampling time)*

<u>Air Velocity, cm/sec</u>	<u>Volume Collected, liters</u>	<u>Activity, dpm</u>
10	400	9×10^3
		1.27×10^4
		9×10^3
50	2000	4.7×10^4
		4.5×10^4
		4.7×10^4
100	4000	6×10^4
		7×10^4
		6×10^4

These levels are considerably below the contamination levels found normally from processing samples in the experimental and analytical gloveboxes. Use of uncontaminated air in this manner was undesirable because of the accompanying difficulties. Air drawn symmetrically from the glovebox air materially assisted in the operation.

A photograph of the experimental system chosen is shown in Figure 1, and a schematic diagram in Figure 2. The principal parts of the apparatus are:

- A 15 kW, 10 kHz induction heating unit*
- A water-cooled work coil of 1/4-in. copper tubing wrapped around an alundum thimble containing a graphite susceptor
- A stainless steel "furnace cap" which fits over the alundum thimble, and with a shallow cup cut into its upper surface to hold the powder specimens. A 1/8-in. diameter well was drilled into each cap from its rim to the centerline to hold a thermocouple for measuring heat run temperatures.
- A platinum-platinum 13% rhodium thermocouple
- A 44 mm OD quartz chimney to carry the entrained particles upward to the filter
- A Pyrex® 2-in. filter holder with a fritted glass support modified to minimize the pressure drop across the holder
- A temperature recorder^{e*}

Air, drawn up and around the stainless steel furnace cap, entered the quartz chimney via the annular space between the cap and bottom of the chimney (see Figure 3). The two pieces were spaced so that the nominal velocity at the opening was approximately twice the calculated velocity in the chimney. Entering particles were carried up into

* *Manufactured by the Ajax Magnethermic Corporation, Youngstown, Ohio.*

® *Registered trade name of the Corning Glass Company.*

** *Micromax Recorder manufactured by Leeds and Northrup, Philadelphia, Pa.*

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FIGURE 1. Release Rate Apparatus

FIGURE 2. *Apparatus for Measurement of Release Rates of Overheated Plutonium Powders*

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FIGURE 3. *Stainless Steel Furnace Cap
and Bottom of Quartz Chimney*

the chimney and either deposited on the 0.003-in. mild steel chimney liner or collected on a 2-in. glass fiber filter." The predetermined velocities within the chimney were produced through use of building vacuum. Air flows were measured by one of several calibrated rotameters after passing through a back-up high efficiency particulate filter.**

The values for the air velocities through the chimney were chosen as representative of the velocity range in accident situations. The lower value, 10 cm/sec, provided sufficient velocity in the chimney to carry a plutonium dioxide particle of 17 μ diam (see Figure 4). Thus, particles constituting an inhalation hazard would be collected. The upper velocity value, 100 cm/sec, was selected as a practical limit for the experimental arrangement.

An induction heating unit was used in these experiments because of convenience, ease of temperature control, and for safety reasons. Heat was generated by induced currents within the graphite susceptor and stainless steel furnace cap in an alternating magnetic field. Heating in this manner at times produced some vibrations in the furnace cap which likely would have enhanced the release. This effect is believed to be small.

* Type A made by the Gelman Instrument Co., Ann Arbor, Michigan. The filter has a stated efficiency of greater than 99.7% for 0.3 micron DOP spheres.

** No. DZ-78001, Mine Safety Appliance Co., 201 N. Craddock, Pittsburgh, Pa.

FIGURE 4. *Equivalent Diameter PuO_2 Particle Lifted by Various Air Velocities*

PROCEDURE FOR DETERMINING RELEASE RATES

Plutonium amounts released by the various powders were determined by:

1. Placing a weighed amount of the material in the shallow depression in the stainless steel furnace cap,
- 2, Drawing air around the sample at the desired velocity while heating,
3. Determining the amount of plutonium deposited on and carried through the chimney.

A stainless steel furnace cap, a 2-in. membrane filter in a disposable holder, a pair of tweezers, a 2-rn. glass fiber filter, and a piece of 0.003-in. mild steel shimstock chimney liner in a Pyrex carrier were loaded into the glove-box prior to each run. Each item was packaged separately in a plastic bag to minimize contamination. A tare weight was obtained for the furnace cap (approximately 140 g), and the weight of the added specimen obtained by difference. Powder weights varied from 0.98 to 1.03 g.

The powder specimen was placed on the alundum thimble and the remainder of the apparatus assembled. New forceps were used in each run to handle the glass fiber filter. All glass portions of the apparatus were cleaned to a non-smearable level between runs. After the annular space between the bottom of the chimney and the furnace cap had been adjusted to the correct 3/16-in. distance with a spacer, air was drawn around the sample at the desired velocity for one hour to determine the release at ambient temperature.

The release at high temperature was then determined. The chimney liner and filter were changed and the air flow re-established. Temperature recording was initiated and the specimen heated at maximum output of the induction unit. Rates varied somewhat during each run due to

variations in geometry but were generally around 200 to 250 °C per minute. Figure 5 shows typical heating curves for the three temperatures used in this study. The Induction unit was turned off before the desired temperature was reached and the temperature allowed to drift to the desired level. The induction unit was turned on again to some predetermined setting to maintain the desired level (400, 700, or 1000 °C). Temperature was held at this level (within ±2%) for one hour. Sampling, to include material released during cooling, was continued for one hour after the heat was turned off.

The chimney filter and liner, and glovebox air sample were removed from the apparatus and were examined, labelled, and packaged for analysis. Chimney filters were placed on a clean 8-oz. jar lid and examined microscopically under 100X magnification prior to packaging. Residues remaining on the furnace caps after heating were also examined under magnification. The chimney liners were carefully slid into their Pyrex holders which doubled as dissolution vessels during analyses. All samples were sealed in plastic and removed from the glovebox for analysis. Special handling procedures were used during processing to minimize the effects of cross-contamination.

ANALYTICAL

The amount of plutonium present was determined by dissolving the material collected in nitric acid, isolating the plutonium by extraction, and counting the alpha particle emission of the samples. Weighted aliquots of the original powders were dissolved in nitric acid and the disintegrations per minute (dpm) per gram of powder determined. The values obtained are shown in Table IV.

FIGURE 5. Typical Time Versus Temperature Curves for 3 Experimental Temperatures Used in Release Studies

TABLE IV. *Activity of Powders Used as Sources for Release Rate Experiments (dpm/g of powder)*

Partially Oxidized Plutonium Oxalate	1.27×10^{11} dpm/g
Fresh Plutonium Oxalate	1.20×10^{11} dpm/g
Plutonium Fluoride	1.35×10^{11} dpm/g

Although the filter samples could be dissolved and a dilution mounted directly for counting, some difficulties were encountered with the high iron content of the dissolved chimney liner. In these samples, the plutonium was extracted with tri-iso-octylamine (TIOA) from an acid solution prior to mounting.

PARTICLE SIZE ANALYSIS

Particle size distributions were determined microscopically on the source material, and on some airborne material collected on the air sweep filters. Particles were grouped into 7 size fractions as follows: <5, 5-8, 8-12.5, 12.5-20, 20-32, 32-50, and >50 μ . Particles were compared with a calibrated glove and circle graticule inserted into the eye piece of a monocular microscope (located in the glovebox).

Generally, one scan was made across the sample. Additional scans at 45, 90, and 135 °C were taken when sufficient particles were not counted in a single scan. The number distribution of the particles was converted into a mass distribution by assuming the material to be homogeneous, a density of 11.4, and with the volume a cube function of the projected diameter. The data were plotted and the Median Mass Diameter (MMD) taken from the graph.

RESULTS AND DISCUSSIONRELEASE FROM PLUTONIUM OXIDE

The amount of material released from oxide was found to range from 3.2×10^{-6} to 0.56 wt%/hr and appeared to be a function of air velocity. Temperature did not appear to significantly affect the release rate. The amount released appeared to be similar to re-entrainment of particles from a hard surface.

The source powder used for these experiments was less than the 44 but greater than the 15 μ fraction of the oxide produced during the air oxidation of alpha plutonium above ignition temperature. The unalloyed relatively pure metallic plutonium was heated in a quartz tube and the residue separated into particle size fractions by a combination sieve-air elutriation-impactor technique. This 15 to 44 μ fraction comprised 0.60 to 0.80 wt% of the oxide produced. This material was composed of two types of particle--dark, elongated bars and yellow, finely divided spheres. A more complete description of the procedure and results can be found in reference (4).

The amounts released under the conditions used in these experiments are shown in Table V.

TABLE V. *Amounts of Plutonium Entrained in Air from the Heated Oxide (The 15 to 44 μ fraction from the air oxidation of unalloyed plutonium metal above ignition temperature.)*

	wt%/hr	
	<u>10 cm/sec</u>	<u>117 cm/sec</u>
Ambient Temperature	6.1×10^{-6}	0.56
800-900 °C	5.3×10^{-6}	0.025

The amount entrained in 10 cm/sec air was similar to the level elutriated by air with a nominal velocity of 8.2 cm/sec during the oxidation of metallic plutonium,⁽⁴⁾ and agrees well with the reported values for resuspension factors for plutonium dioxide.⁽²⁶⁾ The value found for release at 800 to 900 °C using 117 cm/sec air was found to compare with the amount entrained by passing air at 525 cm/sec over bare, ignited metal⁽⁵⁾ and re-entrained when both moving air and a transfer of momentum are involved.⁽²⁷⁾ The high release rate at ambient temperature appears to be anomalous with the 117 cm/sec air velocity. The explanation is not readily apparent, but may have resulted from inadvertent cross contamination during analysis, atypical mounting of the specimen, or flow conditions.

PARTIALLY OXIDIZED PLUTONIUM OXALATE

Of the four powders used in this study, partially oxidized plutonium oxalate was the most easily entrained in a flowing air stream. The entrained amount increased with both temperature and air velocity to a maximum value of 0.82 wt%/hr at 1000 °C and 100 cm/sec velocity--the maximum conditions used in these studies.

The partially oxidized plutonium oxalate powder used as source material in these experiments was produced by holding normal production-line oxalate for approximately 6 months in a dry air (dewpoint less than -70 °F) glovebox. This powder was tan, finely divided, and free flowing with an activity of 1.27×10^{11} dpm/g and found to be approximately 68% plutonium. Under magnification, the powder was composed of both cream colored and brownish spheres with an MMD of 32 μ . The size distribution is shown in Figure 6.

URE 6. *Size Distribution of Partially Oxidized Plutonium Oxalate Used in Release Runs (205 Particles Sized at 100X)*

The release rates, determined by the use of partially oxidized plutonium oxalate as a source at various temperatures in air at the three velocities, are listed in Table VI. The amounts carried through and deposited on the chimney are listed separately. The rate appears to increase somewhat between 10 and 50 cm/sec and increases linearly between 50 and 100 cm/sec (see Figure 7). The highest value, 0.82 wt%/hr, was obtained at the maximum temperature and velocity and is an average of two results, 0.54 and 1.1 wt%/hr. The rate initially increased sharply with temperature and then leveled off at the maximum temperature (see Figure 8). The increase in rate with temperature may result from a transfer of momentum to the particles from the stresses created in heating, or because of variations in the behavior of the specimen holder in the induction unit magnetic field.

TABLE VI. Partially Oxidized Plutonium Oxalate Release Rates (in wt%/hr)

Temperature, °C	Sample Type	Nominal Air Velocity Through Chimney		
		10 cm/sec	50 cm/sec	100 cm/sec
Ambient	A	0.051	$<4 \times 10^{-5}$	$<4 \times 10^{-5}$
	B	4.6×10^{-3}	$<4 \times 10^{-3}$	0.66
400	A	--	--	0.057
	B	--	--	0.0075
700	A	--	--	0.735
	B	--	--	0.041
1000	A	0.25	0.62	0.82*
	B	0.023	$<4 \times 10^{-3}$	0.057*

A *Particles carried through chimney (collected on glass fiber filter).*

B *Particles entrained but deposited on chimney walls (collected on 0.003-in. mild steel shimstock liner).*

* *Average of two runs.*

FIGURE 7. *Variations in Partially Oxidized Plutonium Oxalate Release Rates as a Function of Air Velocity (at 1000 °C)*

FIGURE 8. *Variations in Partially Oxidized Plutonium Oxalate Release Rates as a Function of Temperature (at 100 cm/sec air velocity)*

The weight percent per hour values for deposrtion in the chimney are much more scattered. These values are at the system lower detection limits as determined by control runs. The proximity of the lower end of the shimstock liner to the source (approximately 1/8 in.) made contamination control difficult. These values could readily be influenced by small amounts of contamination. Combining the filter and deposition values does not alter the release rate significantly. Assuming contamination did not significantly affect the chimney deposition rates, filter and deposition values average approximately 7% of the material carried through the chimney.

The particles collected on the filters were examined microscopically. They were cream-colored spheres ranging in size from 8 to 36 μ in diameter. At a temperature of 400 °C in air with a velocity of 100 cm/sec (see Figure 9) the size distribution of the particles entrained was very much like that of the finer fraction of the source material (see Figure 6). The MMD of the entrained material was 24 μ compared to 31 μ for the powder used as a source.

The physical properties of the material remaining after heating can also affect the hazards associated with an accident. The hazards associated with the handling of easily dispersed material are greater than for the same material sintered into a ceramic mass. The physical appearance of the residues from these experiments is given in Table VIP. For residues from the heatrng of partially oxidized oxalate, temperatures near 1000 °C were necessary to convert the powder into a form not easily dispersed. The residues' characteristics at 400 and 700 °C agree with the dioxide reported by Drummond and Welch.⁽¹⁵⁾ Both could be readily dispersed.

FIGURE 9. *Distribution of Particles Entrained in Air with a Velocity of 100 cm/sec from a Plutonium Oxalate-Oxide Mixture Heated to 700 °C. (1156 particle sized optically)*

TABLE VII. *Physical Appearance of Residue from the Overheating of Partially Oxidized Plutonium Oxalate*

Temp., °C	Air Velocity, cm/sec	Physical Appearance of Residue	Physical Appearance of Material Collected on Chimney Filter
400	100	Yellow-green, free-flowing powder.	Yellow to green spheres ranging from 6 to 46 μ in diameter. Surface of filter obscured.
700	100	Yellow-green cake easily dispersed.	Yellow to green spheres ranging from 8 to 36 μ in diameter. Surface of filter visible.
1000	10	Hard, black cake.	Yellow spheres ranging from 4 to 26 μ in diameter. Mostly black shapeless masses.
1000	50	Hard, black cake.	Dark crust composed of yellow spheres ranging from 4 to 26 μ in diameter, and black shapeless masses. Some translucent maroon flakes 600 to 1000 μ .
1000	100	Hard, black cake.	Dark crust of yellow spheres ranging from 6 to 40 μ in diameter, and black shapeless masses

RELEASE FROM PLUTONIUM OXALATE

Plutonium oxalate gave the highest release rate of the materials studied--0.9 wt%/hr at 700 °C in air with a velocity of 100 cm/sec. Release rates were lower at 400 and 1000 °C. A high air velocity was required to entrain the particles produced. Significant release rates were found only at 100 cm/sec.

The plutonium oxalate used in these studies was material freshly prepared in the plant process. The oxalate was moist and had the color, consistency, and physical appearance of brown sugar. A few large lumps approximately 1/8 in. in diameter were visible in the mass. The powder had an activity of 1.70×10^{11} dpm/g. Under magnification, the individual particles were spherical in shape with an MMD of 50 μ . The size distribution is shown in Figure 10. Over the 4-month period encompassing the study, the marked alteration in physical appearance of the oxalate to a free-flowing, olive-green powder was probably due to moisture loss to the dry glovebox air.

The release rates determined in air at 10, 50, and 100 cm/sec, and temperatures of 400, 700, and 1000 °C are shown in Table VIII. The amounts released, dependent on the air velocity, were of significant quantity only at 100 cm/sec air velocity. The dependence on velocity is not surprising considering the fairly steep slope of the size distribution curve (Figure 7). The high value, at 700 °C, 0.90 wt%/hr, probably results from a combination of maximum shrinkage and drying of the oxalate sphere without sintering. No significant release was found at lower air velocities at this temperature. Three to five percent of the material passing through the chimney deposited on the walls.

The physical appearance of the material collected on the filters and the residue remaining on the stainless steel dishes after heating are summarized in Table IX. The residues from all runs were caked and fragmented with light tapping. In most, only the surface layers were hardened and the powder underneath was free flowing. Probably because of the more rapid forming of the hard surface layer, the particles collected on filters appeared to

FIGURE 10. *Size Distribution of Plutonium Oxalate Used in Release Runs. (855 particles sized at 100X)*

decrease slightly on the upper size limit with increasing heat. Thus, heating rate would appear to be significant in the amount released.

TABLE VIII. *Plutonium Oxalate Release Rates fin wt%/hr)*

Temperature, °C	Sample Type	Nominal Air Velocity Through Chimney		
		10 cm/sec	50 cm/sec	100 cm/sec
Ambient	A	<0.004	<0.004	<0.004 0.073
		<0.004	<0.004	0.38 0.54
	B	<0.004	<0.004	0.006 0.025
		<0.004	0.0096	0.023 0.036
400	A	--	--	0.48
	B	--	--	0.016
700	A	0.0044	<0.004	0.90
	B	<0.004	<0.004	0.047
1000	A	<0.004	0.007	0.25
	B	<0.004	0.005	0.075

A Particles carried through chimney (collected on glass fiber filter).

B Particles entrained but deposited on chimney walls (collected on 0.003 in. mild steel shimstock liner).

PLUTONIUM FLUORIDE

The amount of material released by plutonium fluoride when heated in moving air was lower than that found for partially oxidized plutonium oxalate and "fresh" plutonium oxalate. Release rates varied from 0.007 wt%/hr at 400 °C in air with a velocity of 100 cm/sec, to 0.056 wt%/hr at 1000 °C and 100 cm/sec (the maximum conditions imposed in these studies).

The plutonium fluoride powder used in these experiments was normal process material. As received, the powder was a green, finely divided, free-flowing powder containing a few large lumps. Microscopic examination showed the powder to

be composed of individual spheres and agglomerates with a median diameter of $26\ \mu$ and a MMD of $38\ \mu$ (see Figure 11). The activity of the powder was determined to be 1.35×10^{11} dpm/g.

TABLE IX. Physical Appearance of Material Collected on Filters and Remaining on Dish in the Overheating of Plutonium Oxalate Powder

Temp., °C	Air Velocity, cm/sec	Residue	Filter
400	100	Easily broken up, olive-green cake.	Individual green spheres and agglomerates ranging from 5 to $58\ \mu$ in diameter.
700	100	Easily broken up, olive-green cake, More dense than at $400\ ^\circ\text{C}$.	Brown to cream colored spheres from 5 to $50\ \mu$ in diameter.
1000	10	Same as above.	Green flecks from 5 to $35\ \mu$ in diameter. Very few spheres.
1000	50	Same as above.	Dark green flecks ranging from 5 to $20\ \mu$ in diameter.
1000	100	Same as above.	Combination of dark ash-like material, green spheres and agglomerates and broken spheres. Particles range from 5 to $35\ \mu$ in diameter.

The release rates determined in this study by heating plutonium fluoride powder to various temperatures in air velocities of 10 , 50 , and $100\ \text{cm/sec}$ are listed in Table X. Only at elevated temperatures and maximum air flow were detectable amounts of materials released. The rates, varying from approximately $0.007\ \text{wt\%/hr}$ to nearly $0.06\ \text{wt\%/hr}$, were lower than those found for the partially oxidized oxalate and oxalate.

FIGURE 11. Size Distribution of Plutonium Fluoride Used in Release Runs (347 Particles Sized at 100X)

TABLE X. *Plutonium Fluoride Release Rates (In wt% released per hour)*

Temperature, °C	Sample Type	Nominal Air Velocity Through Chimney		
		10 cm/sec	50 cm/sec	100 cm/sec
Ambient	A	$<4 \times 10^{-3}$	$<4 \times 10^{-3}$	$<4 \times 10^{-3}$ $<4 \times 10^{-3}$
	B	$<4 \times 10^{-3}$	$<4 \times 10^{-3}$	$<4 \times 10^{-3}$ $<4 \times 10^{-3}$
400	A	--	--	0.0073
	B	--	--	$<4 \times 10^{-3}$
700	A	--	--	0.022
	B	--	--	0.048
1000	A	$<4 \times 10^{-3}$	$<4 \times 10^{-3}$	0.05 0.02
	B	$<4 \times 10^{-3}$	$<4 \times 10^{-3}$	0.016 $<4 \times 10^{-3}$

A *Particles carried through chimney (collected on glass fiber filter..*

B *Particles entrained but deposited on chimney walls collected on 0.003 in. mild steel shimstock liner).*

Table XI lists the physical characteristics of the material collected on the chimney filter and that remaining in the dish from the overheating of plutonium fluoride powder. Particle characteristics are reported for all chimney filters, even those below the lower detection limits for the experiments. The lower limits had purposely been set at a sufficiently high level that normal variations in contamination level would not cause difficulties in the interpretation of results. A value of 4×10^{-5} wt% represents 40 μ g for the 1-g of powder used in these runs. Many 10- μ particles could be present and still be below the limits used. The particles observed were green spheres and agglomerates characteristic of oxides produced from

plutonium fluoride.⁽¹⁵⁾ That very few of these particles were detected on the filter from the run at 1000 °C in air swept past the powder at 10 cm/sec is indicative of the low entrainment at this velocity. The residues at all temperatures were easily dispersed and still present a hazard.

TABLE XI. Physical Appearance of Material Collected on Filters and Remaining on Dish in the Overheating of Plutonium Fluoride Powder

Temp., °C	Air Velocity, cm/sec	Residue	Filter
400	100	Easily dispersed green powder	Green spheres and agglomerates ranging from 5 to 50 μ in diameter,
700	100	Same as above.	Green spheres and agglomerates ranging from 8 to 40 μ in diameter.
1000	10	Same as above.	Dark ash-like particles. No spheres or agglomerates observed.
1000	50	Same as above.	Green spheres and agglomerates from 10 to 24 μ in diameter
1000	100	Same as above.	Green spheres and agglomerates from 7.5 to 29 μ in diameter

The particle size distribution of the material collected on the chimney filters during several runs was determined. At ambient temperature in air with a velocity of 100 cm/sec, the distribution (see Figure 12) indicates a finer size range than the source material (see Figure 11). As the temperature is increased, the size distribution indicating the entrainment of larger particles accounts for the higher release rates (see Figures 13 and 14). The total amounts entrained

FIGURE 12. *Distribution of Particles Entrained by Air with a Velocity of 100 cm/sec from Plutonium Fluoride at Ambient Temperature*

FIGURE 13. *Distribution of Particles Entrained in Air with a Velocity of 100 cm/sec from Plutonium Fluoride Heated to 700 °C*

FIGURE 14 *Particles Entrained in Air at 100 cm/sec from 1 g PuF_4 Powder at 1000 °C (303 Particles Sized at 100X)*

at both 700 and 1000 °C are similar (0.07 and 0.066 wt%/hr respectively) with less reaching the filter at 700 °C. The size distribution at 700 °C is coarser.

CONCLUSIONS AND FUTURE WORK

From a hazards evaluation viewpoint, all four of the examined powders possess varying characteristics which can affect the amount of oxide released.

The material released by the finely divided oxide, the 15 to 44-, fraction from the oxidation above ignition temperatures of unalloyed, metallic plutonium, is principally the process of re-entraining a solid into a flow gas stream. The amounts released, ranging from 5×10^{-6} to 0.025 wt%/hr, are in the range of published plutonium dioxide re-entrainment values.⁽²⁶⁾ If momentum can be transferred to the particles by some process such as varying thermal stresses within the mass of source material, etc., the release of particles is enhanced.

Of the four powders tested partially oxidized plutonium oxalate appeared to present the greatest potential for release during heating and was the most readily airborne. This powder showed an observable release at all air velocities at 1000 °C and ranged in rates from 0.25 to 0.82 wt%/hr. In air sweeping upwards around the powder at a velocity of 100 cm/sec, the release rates varied from 0.057 wt%/hr at 400 °C to 0.82 wt%/hr at 1000 °C. A very small fraction of the material collected on the chimney filter was less than 6 μ in diameter (see Figure 9). The partially oxidized oxalate cakes, on heating to 1000 °C, produce a tightly bound, not readily dispersed mass,

The fractional release from "fresh" plutonium oxalate was a marked function of velocity, and observable rates

were found only at the highest velocities used. The highest release rate observed during the experiments, 0.9 wt%/hr, was found for the oxalate at 700 °C in air sweeping upwards around the powders at 100 cm/sec. The rapid evaporation of moisture from this powder during heating undoubtedly aids in the entrainment process. Some hardening of the surface layer occurs when heat is added to form a barrier to further entrainment. Thus, the residue is a dispersable cake but the release rate is reduced at 1000 °C.

The plutonium fluoride release rates were lower than those found for the partially oxidized and "fresh" plutonium oxalate. The rates varied from 0.007 to 0.05 wt%/hr. Although the source material contained the largest fraction of fine material (see Figure 11), the chemical and physical changes induced by heating contributed less to the entrainment process in this material than in the other two powders. The residues did not cake and remained free-flowing. Thus, the surface layer does not constitute a barrier against additional entrainment. These residues present the greatest potential handling hazard of the four powders used,

The latter three powders are definitely more readily airborne than the metal or oxide produced from the metal. Even in air velocities 5 times as great, 525 cm/sec, the release rates in wt%/hr for oxides were less than those found with the powders at 1000 °C (0.05 wt%/hr). The residue after heating still presents the potential for additional dispersal. Conversely, **re-entrainment**, even from finely divided oxide, is low.

Additional work to characterize the amount and nature of the aerosol produced by burning various combustible materials mixed with these plutonium compound- is

contemplated, Also the study of low and high temperature plutonium release from plutonium nitrate solutions and its residue is in progress.

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