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CHARACTERISTICS OF THE AEROSOL PRODUCED FROM BURNING PLUTONIUM

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ABSTRACT

Safety analysis of sodium-cooled, plutonium-fueled, fast reactor plants must be concerned with the possibility of fires involving these materials. Design of an air cleaning system for such a facility requires basic data defining the aerosol characteristics of sodium and plutonium released during a fire.

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Size characteristics of the aerosol produced during plutonium fires were determined for different atmospheres ranging from 20.8% oxygen, 79.2% nitrogen to 0.5% oxygen, 99.5% nitrogen. Plutonium-cobalt-cerium alloy, and alpha- and delta-phase plutonium metal were burned. Data were obtained to estimate the fraction of plutonium alloy airborne during a plutonium fire. Fires simulating a reactor accident involving both fuel and coolant defined the relative airborne concentration of plutonium and sodium.

I. Summary

1. The aerosol collected directly above burning chips of plutonium-cobalt-cerium alloy (57.7% plutonium) showed the following size characteristics.

Count Median Diameter (C.M.D.) : 0.04 - 0.09 microns (μ)

Geometric Standard Deviation (σ_g): 1.24 - 1.54

Mass Median Diameter (M.M.D.) : 0.05 - 0.14 μ

2. The aerosol collected directly above burning metallic plutonium turnings (alpha- and delta-phase) showed the following size characteristics.

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C.M.D.: 0.02 - 0.06 μ

σ_g : 1.24 - 1.76

M.M.D.: 0.03 - 0.13 μ

3. Oxygen concentration of the burning atmosphere does not affect the particle size of the aerosol produced from burning plutonium metal or alloy.

4. When plutonium-cobalt-cerium alloy is burned under reduced oxygen conditions, the fraction airborne ranges from 1.8×10^{-7} to 38.8×10^{-7} depending on burn temperature and oxygen concentration.

5. Fires involving plutonium alloy and sodium produce airborne particulates with a plutonium-sodium ratio ranging from 0.34 to less than 0.008%. The high initial ratio is rapidly reduced as the molten sodium blankets the release of plutonium.

II. Introduction

Safety analysis of sodium-cooled, plutonium-fueled, reactor plants must consider the possibility of a fire involving these materials during the maximum credible accident (M.C.A.). To provide design criteria for the plant air cleaning system and to evaluate any potential hazard due to release of radioactive particulates, basic data defining the aerosol characteristics of sodium and plutonium produced during an M.C.A. fire are required.¹

A test program determined the size characteristics of the aerosol produced during plutonium fires. Since a system of this type may be operated at reduced oxygen conditions, aerosol parameters were determined for fires in which the atmosphere consisted of various oxygen-nitrogen mixtures ranging from 20.8% to less than 0.5% oxygen.

Data were also obtained defining the percent of burned plutonium airborne during a fire, and the relative concentrations of airborne sodium and plutonium resulting from a fire involving both reactor fuel and coolant.

III. Plutonium Aerosol

A. Background.

Several investigators have attempted to determine the aerosol characteristics of airborne plutonium produced from fires. Carter^{2,3} found an aerodynamic M.M.D. of 8.5 to 9.7 μ when sampling with a cascade impactor. He confirmed these values by autoradiography of fallout samples. Using cascade impactors, Stewart⁴ found M.M.D.'s (unit density) ranging from 1 to 16 μ . He suggested a single particle size distribution to define the airborne plutonium aerosol. This aerosol had an M.M.D. of approximately 1.5 μ . Sherwood⁵ used autoradiographic techniques on filter paper samples to evaluate the particle size of airborne plutonium in radiochemical laboratories. He found the airborne plutonium generally attached to inactive particles of dust. Using autoradiography, Moss⁶ found an M.M.D. of 0.14 to 0.65 μ when sampling for plutonium outside the glove boxes in metal fabrication areas.

They estimated the size of plutonium aerosols by relating the number of tracks arising from a particle of plutonium to the mass. It was evident from these data that the best way to view and characterize these small plutonium aerosols was by electron microscopy.

The present test was designed to characterize the aerosol present just above a plutonium fire. The air sampler selected collected the aerosol directly on E.M. grids and permitted the use of electron microscopy to evaluate any submicron aerosol produced.

B. Procedures.

Because of the toxicity of plutonium, all burning apparatus and sampling equipment had to be located in a glove box, and all sampling manipulations carried out through glove ports. Approximately 50 to 100 grams of plutonium or plutonium-cobalt-cerium alloy were burned in a tantalum dish which fitted inside the copper burning pot shown in Fig. 1. Special oxygen-nitrogen gas cylinders were prepared, and each cylinder analyzed using a gas chromatograph. The various oxygen-nitrogen gas mixtures were supplied just above the tantalum dish holding the plutonium, to evaluate the effects of oxygen concentration on aerosol characteristics. Thermocouple holes in the copper base plate permitted measurement of the burning pot temperature. Relative humidity within the glove box was consistently below 25 ppm.

Since the air sampler could not be used outside the glove box after completion of the experiment because of contamination, a relatively inexpensive sampler was desirable. The all-glass, point-to-plane electrostatic precipitator (E.S.P.) shown in Fig. 2 was used. This sampler permits aerosol collection directly on carbon-coated electron microscope (E.M.) grids. The unit is operated at a flow rate of 5 liters per minute and a high voltage potential of 9,000 to 10,000 volts a.c. provided by a conventional neon-lamp transformer. Previous work⁷ has shown that this sampler has representative aerosol collection characteristics comparable to those defined by several thermal and electrostatic precipitators. Figure 3 shows the E.S.P. just above the plutonium burning pot inside the glove box. The distance between the burning pot and the E.M. grid was 23 centimeters.

After sampling, the E.M. grid was removed from the glove box and monitored for alpha activity to estimate the quantity of plutonium collected. The grids were then alpha-counted and shadowed with chromium or platinum at 30°. Electron micrographs were obtained from random grid openings.

All electron micrographs (enlarged photographically to 50,000X) were sized on the basis of equivalent area (projected diameter)⁸ using the Zeiss Particle Size Analyzer,* and the data obtained were plotted on log-probability paper.⁹

Count median diameter (C.M.D.) and geometric standard deviation (σ_g) were determined graphically. Mass median diameter (M.M.D.) was calculated using the Hatch and Choate¹⁰ equation which also assumes a log normal distribution, $\text{Log M.M.D.} = \text{Log C.M.D.} + 6.9 \text{ Log}^2 \sigma_g$.

*Carl Zeiss (mfgr.) Particle Size Analyzer, Model TGZ-3.

C. Results.

Table I summarizes the aerosol characteristics from burning plutonium-cobalt-cerium alloy (57.7% plutonium) and plutonium metal (alpha- and delta-phase). In all instances the C.M.D. was below $0.1\ \mu$ and the M.M.D. below $0.15\ \mu$. Aerosol size was independent of oxygen concentration. The aerosol C.M.D. produced from burning alpha- and delta-phase plutonium was somewhat smaller than that resulting from burning the alloy.

The small particle size (C.M.D. of 0.02 to $0.09\ \mu$) shows that careful attention must be given to the design of air cleaning systems if these materials must be handled.

Figure 4 shows the typical aerosol collected when plutonium alloy or metal was burned at four different oxygen concentrations. Since the plutonium aerosol C.M.D. was as small as $0.02\ \mu$, it was feared that the platinum or chromium deposited on the E.M. grid during shadowing might present an artifact. Figure 5 shows a grid platinum shadowed on both sides at a magnification of 50,000 times. The single large particle (approximately $0.5\ \mu$) is due to background material within the glove box. This was verified by air samples obtained when no burning was taking place. The material deposited during shadowing presents no background problem.

Several of the samples collected during burning of delta-phase plutonium showed small amounts of a relatively large chain-structured aerosol. This aerosol was not present in samples taken when burning plutonium-cobalt-cerium alloy or alpha-phase plutonium. The possibility that gallium present in delta-phase plutonium produced this chain-structured aerosol was investigated. Figure 6 shows the aerosol produced from burning gallium metal. Figure 7 shows the chain-structured material present when delta-phase plutonium was burned. Because of this similarity, the chain-structured material was omitted when sizing the aerosol collected from burning delta-phase plutonium.

IV. Airborne Plutonium

To fully evaluate the potential hazard from a plutonium fire, it is necessary to estimate what fraction of the material burned would be airborne. Stewart⁴ has found that from 2.1×10^{-2} to 6×10^{-5} of burned plutonium metal or alloy is airborne. His tests were conducted under oxygen concentrations at or above 20.8% with nitrogen constituting the remainder of the burn atmosphere. A limited test program was carried out at Los Alamos to obtain similar information for reduced oxygen conditions.

Plutonium-cobalt-cerium alloy (2.0 grams) was placed in the tantalum boat located in the center of the quartz tube shown in Fig. 8. Oxygen-nitrogen gas mixtures ranging from 0.4 to 2.76% oxygen flowed over the tantalum boat at a velocity of approximately 300 feet per minute. This gas velocity was selected to simulate conditions during a postulated M.C.A. No attempt was made to evaluate the effect of varying gas velocity past the burning plutonium alloy. After a combustion furnace was brought to temperature, the quartz tube was positioned in the furnace and the entire gas mixture passing over the plutonium was sampled with an AA Millipore filter. This closed-system test procedure guaranteed collection

of all airborne particulate material. There was no attempt to compensate for plate-out on any surfaces between the tantalum boat and the sampling Millipore filter. The Millipore filters were alpha-counted to determine what fraction of the original 2.0 grams of alloy was airborne.

Table II and Figure 9 summarize the results obtained. At 500°C the airborne fraction is about 2×10^{-7} and is independent of oxygen concentration. At 700°C the airborne fraction ranges from 4×10^{-7} to 4×10^{-6} with the larger value at the higher oxygen concentration.

V. Sodium-Plutonium Fires

A fire under M.C.A. conditions probably would involve both plutonium and sodium. To simulate an accident of this type, the rocking furnace shown partially assembled in Fig. 10 was used. Sodium metal and plutonium alloy were inserted into the vertical tube, heated to a molten state and thoroughly mixed by rocking the Tee-shaped furnace through an angle of 60°. During heating and rocking, a nitrogen atmosphere was maintained within the furnace. After being rocked at 550 to 600°C., the molten mixture was dumped into the burning pot previously described (Fig. 1). A 2.67% oxygen, 97.33% nitrogen gas mixture was supplied to the burning pot. Air samples were obtained using Millipore filter field monitors, and the relative concentrations of airborne plutonium and sodium were determined. Figure 11 shows the test apparatus in the glove box.

During the tests, the initial melt consisted of 45 grams of sodium and 20 grams of plutonium-cobalt-cerium alloy (57.7% plutonium). To minimize contamination from plutonium within the glove box, filter samples were removed from their plastic holder after removal from the glove box.

Table III summarizes the results obtained in two different test series. The "Reference Time" shows the time interval between dumping of the sodium-plutonium mixture out of the rocking furnace and the start of sampling. In Test Series B, the molten sodium-plutonium mixture was agitated after sample 4 and again after sample 8. The sampling period for each sample was one minute.

Excluding sample A-1, the relative concentration of airborne plutonium to sodium ranges between 0.34% and $< 0.008\%$. In all likelihood, the single high value (sample A-1) was caused by contamination of the Millipore filter during handling in the dry box. These data, with the exception of sample A-1, are plotted in Fig. 12 with Sample B-7 ($\text{Pu/Na} < 0.008\%$) assumed to be 0.001%. These data indicate that the sodium tends to blanket the release of plutonium after the simulated accident (dumping of the molten mixture from the rocking furnace), and that this trend is reversed when the mixture is agitated. This trend is shown by the three sets of data points in Fig. 12 (A-2 through A-4, B-1 through B-4, and B-5 through B-7). The remaining data points (B-8 through B-11) fall within the range of values similarly outlined but do not show any consistent trend.

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TABLE I
PLUTONIUM AEROSOL CHARACTERISTICS

Material Burned	Oxygen Content (%)	Temperature (°C)	C.M.D. (μ)	σ_g	M.M.D. (μ)
Pu-Co-Ce	20.8	550-575	0.04	1.24	0.05
Pu-Co-Ce	8.8	500-550	0.08	1.45	0.13
Pu-Co-Ce	1.7	510	0.08	1.54	0.14
Pu-Co-Ce	0.49	505	0.09	1.41	0.13
None	20.8	25	---	---	---

Pu (α phase)	20.8	575	0.02	1.33	0.03
Pu (α phase)	8.8	525	0.05 0.03	1.72 1.49	0.13 0.05
Pu (α phase)	1.7	500	0.06 0.03	1.64 1.76	0.13 0.08
Pu (α phase)	0.49	495	0.06	1.46	0.09
None	20.8	25	~ 0.5-0.6	---	---

Pu (δ phase)	20.8	550	0.04	1.24	0.05
Pu (δ phase)	10.6	525	0.05	1.38	0.06
Pu (δ phase)	2.67	505	0.04	1.55	0.07
Pu (δ phase)	0.55	500	0.03	1.47	0.05

TABLE II
FRACTION PLUTONIUM AIRBORNE DURING FIRE

<u>Oxygen Concentration (%)</u>	<u>Temperature (°C)</u>	<u>Fraction Pu Airborne</u>
1.9	700	38.8×10^{-7}
0.4	700	4.2×10^{-7}
2.76	500	1.9×10^{-7}
0.5	500	1.8×10^{-7}

TABLE III

RELATIVE AIRBORNE CONCENTRATION OF PLUTONIUM AND SODIUM

<u>Test Series</u>	<u>Sample Number</u>	<u>Reference Time (min.)</u>	<u>Sodium Collected (mg)</u>	<u>Plutonium Collected (mg)</u>	<u>(Pu/Na (%))</u>
A	1	0	0.96	0.194	20.2
	2	1.5	1.08	0.002	0.19
	3	3.0	1.20	0.004	0.34
	4	7.0	9.34	0.007	0.08
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B	1	0	1.06	0.0017	0.16
	2	1.5	1.42	0.0008	0.06
	3	3.0	1.49	0.0005	0.03
	4	4.5	3.16	0.0002	0.01
	5	7.0	2.31	0.0014	0.06
	6	8.5	4.79	0.0005	0.01
	7	10.0	0.38	< 0.00003	< 0.008
	8	11.5	1.91	0.0004	0.02
	9	14.0	2.00	0.0001	0.01
	10	15.5	1.46	0.0007	0.05
	11	18.5	1.68	0.0005	0.03

CAPTIONS

- Fig. 1 Copper Burning Pot with Tantalum Dish.
- Fig. 2 Point-to-Plane Electrostatic Precipitator.
- Fig. 3 Plutonium Burning Apparatus and Sampler in Glove Box.
- Fig. 4 Aerosol Produced by Burning Plutonium Metal (α phase) in 1.7% Oxygen. Shadowed With Platinum at 30°. Typical for All Plutonium and Oxygen Conditions Tested.
- Fig. 5 Electron Microscope Grid Double Shadowed with Platinum.
- Fig. 6 Aerosol Produced by Burning Gallium Metal. Shadowed with Platinum at 30°.
- Fig. 7 Aerosol Produced by Burning Delta-Phase Plutonium. Shadowed with Platinum at 30°.
- Fig. 8 Quartz Tube Used to Determine Fraction of Plutonium Airborne During Fire.
- Fig. 9 Fraction of Plutonium Airborne During Fire as Function of Burn Temperature and Oxygen Concentration.
- Fig. 10 Rocking Furnace Used for Sodium-Plutonium Fires.
- Fig. 11 Test Setup in Glove Box For Sodium-Plutonium Fires.
- Fig. 12 Relative Airborne Concentration of Sodium and Plutonium as Function of Time After Dumping Melt from Rocking Furnace.

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Relative Airborne Concentration of Plutonium & Sodium as function of Time After Dumping

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