PLUTONIUM RELEASE STUDIES

IV. FRACTIONAL RELEASE FROM HEATING PLUTONIUM NITRATE
SOLUTIONS IN A FLOWING AIR STREAM

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ABSTRACT

The fraction of plutonium released as an aerosol upon heating solutions of plutonium nitrate or plutonium nitrate residues was determined. Three arrangements and heating regimes were investigated: 1. Low temperature air drying of shallow pools of concentrated plutonium nitrate solutions. 2. Evaporation of water from a small vessel containing plutonium nitrate at different vapor rates. 3. Heating solids remaining after evaporation of shallow pools of plutonium nitrate to near dryness. Releases during the air drying of plutonium nitrate solutions, at ambient temperature and with slow heating rates, were small (0.001 wt.% maximum) as were releases during the low temperature heating of the solid residues (3×10^{-6} wt.% maximum). Forced evaporation of a plutonium nitrate solution released measurable airborne plutonium under all conditions. During full rolling boil 0.18% of the plutonium nitrate was airborne during evaporation of 90% of the liquid in a small beaker. The solids remaining after evaporation of the solution had fractional releases ranging from 0.0046 to 0.012 wt.% during a 2-hour sampling period. Three temperatures -- 400°C, 700°C, and 1000°C -- were used with an up-sweep of air at three velocities -- 10, 50 and 100 cm/sec. One sample, N5a, was partially converted to oxide and released 0.12 wt.% during the 2-hour period. Median Mass Diameter of the particles airborne during this run was 20.5 microns.

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TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	1
SUMMARY	2
DISCUSSION AND RESULTS	3
A. AEROSOL RELEASED DURING THE AIR DRYING OF SHALLOW POOLS OF CONCENTRATED PLUTONIUM NITRATE SOLUTION	4
1. <u>Equipment and Procedure</u>	4
2. <u>Experimental</u>	7
3. <u>Results</u>	9
B. AEROSOL RELEASED DURING THE HEATING OF POOLS OF PLUTONIUM NITRATE IN A FLOWING AIR STREAM	9
1. <u>Equipment & Procedure</u>	9
2. <u>Experimental</u>	14
3. <u>Results</u>	17
C. THE AEROSOL RELEASED DURING THE HEATING OF THE SOLIDS REMAIN- ING AFTER THE EVAPOPATION OF PLUTONIUM NITRATE SOLUTIONS	18
1. <u>Equipment and Procedure</u>	18
2. <u>Experimental</u>	27
3. <u>Results</u>	29
CONCLUSIONS AND FUTURE WORK	33
REFERENCES	36

LIST OF FIGURES

<u>Fig. No.</u>		<u>Page</u>
1	Plutonium Nitrate Slow Drying System	5
2	Close up of Sample Holder in Plutonium Nitrate Slow Drying System	5
3	Diagram of Apparatus Used in Air Drying of Shallow Pools of Concentrated Plutonium Nitrate Solution	6
4	Apparatus used to Determine Fractional Releases During The Heating of Pools of Plutonium Nitrate Solutions	12
5	Diagram of Apparatus used to Determine Fractional Releases During the Heating of Pools of Plutonium Nitrate Solutions	13
6	Solids Release Apparatus	20
7	Apparatus for Measurement of Release from Overheated Solids from Plutonium Nitrate Solution Evaporation	22
8	Stainless Steel Furnace Cap and Bottom of Quartz Chimney	23
9	Equivalent Diameter PuO_2 Particle Lifted by Various Air Velocities	24
10	Typical Time Versus Temperature Curves for 3 Experimental Temperatures used in Study of Release from Solids	26
11	Variations in Fractional Release from Solids as a Function of Air Sweep Velocity	30
12	Size Distribution of Particles Released During Run N5a	32

LIST OF TABLES

<u>Table No.</u>		<u>Page</u>
I	Fractional Release During Air Drying of Concentrated Plu- tonium Nitrate Solutions	10
II	Fractional Release During the Heating of Pools of Plutonium Nitrate Solutions	15
III	Variation in Releases Due to Concentration During Heating Pools of Dilute Plutonium Nitrate Solutions	19
IV	Fractional Release from Solids Remaining after Evaporation of Plutonium Nitrate Solutions	29
V	Plutonium Aerosol Airborne under Stated Conditions	34

PLUTONIUM RELEASE STUDIESIV. FRACTIONAL RELEASE FROM HEATING PLUTONIUM NITRATE
SOLUTIONS IN A FLOWING AIR STREAM

INTRODUCTION

With the growth of nuclear power and increased availability of plutonium, a greater variety of accident situations can be postulated involving plutonium. To realistically evaluate the hazards involved requires information on the behavior of plutonium under the conditions of the postulated accident. A common condition in many postulated accidents is elevated temperature.

We are conducting a series of studies on the behavior of plutonium at elevated temperatures under a variety of conditions which might exist in accident situations. Since any accident conditions postulated are off-standard and atypical, they are unique and difficult to duplicate. Any study concerned with duplicating the behavior of plutonium under all postulated accident conditions would include an extensive array of experiments. We have somewhat arbitrarily chosen conditions which may represent heating stresses which could be present in a variety of accidents. The data are not meant to be used as absolute values under any one set of circumstances but are indicative of the nature and amount of plutonium aerosol released under these conditions. Since the principal biological hazard to man is inhalation^[1], we have focused our attention on plutonium aerosols.

Four parts of this study have been completed and the findings published. Initially published information germane to the release of plutonium aerosols was reviewed to properly orient our investigations.^[2] We found very little information on the behavior of physical forms of plutonium other than the metal and further study of the behavior of plutonium and its compounds in overheating situations was needed. The behavior of the material and the amount and character of the aerosol generated during heating in a stream of air has been reported for:

1. Small, ignited, unalloyed metal rods in low air flows.^[3]
2. Large, ignited, metal pieces in moderate air flows.^[4]
3. Plutonium compounds as powders in low air flows.^[5]

Aqueous solutions of soluble plutonium salts are another type of material

commonly used. Plutonium nitrate solutions are the most frequently used material of this type^[6] and was chosen for these experiments. The solid salt, plutonium nitrate pentahydrate, $\text{Pu}(\text{NO}_3)_4 \cdot 5\text{H}_2\text{O}$, is readily soluble in water. It begins to decompose at 40°C and melts at 95 to 100°C . An intermediate material (believed to be the basic plutonyl nitrate) is formed at 150° to 200°C . Decomposition to the dioxide is rapid above 220°C and is essentially complete at 250°C . The dioxide produced is normally green but the color can vary.^[7] Decomposition above 300°C for 9 hours produced a fluffy, buff green oxide composed of uniform particles less than 1 micron in size.^[8] Solid, anhydrous, plutonium nitrate is difficult to obtain.

SUMMARY

The quantity and characteristics of the material airborne was determined by the air-drying (with and without heat) and heating in a flowing air stream of plutonium nitrate solutions and heating to high temperatures of the solids remaining after the evaporation. Temperatures used during the air-drying range from ambient to $\sim 130^\circ\text{C}$ using 3 air velocities -- 10, 50 and 100 cm/sec. Four heating regimes representing a cross-section of heating rates characterized by the degree of disturbance of the liquid surface with a 2.9 cm/sec air velocity were used during the second set of experiments. The solid residues were heated to 400°C , 700°C or 1000°C , in an up-sweep of air (10, 50 or 100 cm/sec).

The data indicate:

- The amount released during the low temperature air-drying of plutonium nitrate solutions is small, 2×10^{-8} to 0.001 wt.% in 1-1/2 to 24 hours. Even with partial conversion to the oxide during heating, the amount airborne was 0.003 wt.% in air with a velocity of 50 cm/sec.
- Once dried, the low temperature releases from the solids remaining after the evaporation were low, 2×10^{-8} to 3×10^{-6} wt.%.
- The fraction airborne during heating of plutonium nitrate solution can be roughly correlated to the degree of liquid surface disturbance caused by the heating rate. Values ranged from $\sim 1 \times 10^{-4}$ wt.% during "simmering" to 0.18 wt.% with vigorous surface activity. Fractional

release increased with concentration. Activity Median Diameter of the particles airborne was 4 microns.

- Heating the solids from the evaporation of plutonium nitrate solutions to high temperatures (up to 1000°C) in flowing air (up to 100 cm/sec) made airborne 0.0046 to 0.12 wt.%. The fractional release tended to correlate with temperature and air velocity but the nature of the source material appears to be a more significant factor.
- Some decomposition occurred during the air-drying in one of the runs (N-5). The largest amount of aerosol, 0.12 wt.%, was released during the heating of this material. The Median Mass Diameter of the particles airborne was found to be 20.5 microns.

DISCUSSION AND RESULTS

Three areas were studied to characterize the behavior of plutonium compounds as liquids heated in a flowing air stream and the aerosols produced. The three areas are intended to represent some common heat stresses during potential accident situations. They were:

- A. The slow heating of shallow pools of plutonium nitrate solution in low velocity air.
- B. The heating of pools of plutonium nitrate solution in low velocity air with differing heat inputs.
- C. The heating of the solids remaining after the evaporation of plutonium nitrate solutions in low velocity air.

Plutonium nitrate was used as the source material since it is the soluble compound most frequently utilized in plutonium technology.^[6] The starting solution was a dark brown liquid containing 288 g of plutonium per liter. All solutions were in dilute nitric acid to retard polymerization. Activity in the liquid was determined to be 4.47×10^{10} dis./min (dpm) per ml.

A. AEROSOL RELEASED DURING THE AIR DRYING OF SHALLOW POOLS OF CONCENTRATED PLUTONIUM NITRATE SOLUTION

1. Equipment and Procedure

Small volumes of concentrated plutonium nitrate solution were air dried with and without heating by passing air at low velocities across the surface of the liquid. The apparatus used is shown in Figures 1 and 2, and a schematic diagram outlines the system (Figure 3). The principal components of the system are:

- A containment vessel (CV) fabricated by modifying the tubing portion of a Pyrex 75/50 ball-socket joint.
- A stainless steel dish with a 1-1/2" diameter by 3/32" deep depression used to hold the liquid during drying.
- A teflon holder for the stainless steel dish which positioned the dish in the CV and provided a smooth air flow pattern across the face of the liquid.
- A water-cooled condenser and condensate trap.
- A 47 mm membrane filter held in a glass holder.
- A calibrated flowmeter.
- A stainless steel-clad, chromel-alumel thermocouple, inserted into a well in the stainless steel cap and an appropriate recorder to monitor liquid temperatures during the experiment.
- Two variable transformer-controlled, infrared heating lamps positioned over the stainless steel dish.
- A protective transite enclosure.
- An aluminum cradle to support the CV and a movable platform to position the apparatus in the enclosure.
- Two high efficiency particulate filters on the inlet line to prevent the release of aerosol during off-standard conditions.

The equipment was assembled and 2-1/2 or 3 milliliters of the concentrated plutonium nitrate solution (0.72 or 0.86 g plutonium) were pipetted onto the stainless steel dish through the liquid addition port. Air at one of three velocities -- 10, 50 or 100 cm/sec -- was drawn across the liquid and the

FIGURE 1

PLUTONIUM NITRATE SLOW DRYING SYSTEM

Neg. No. PNL 0673413-2

FIGURE 2

CLOSE UP OF SAMPLE HOLDER IN PLUTONIUM
NITRATE SLOW DRYING SYSTEM

Neg. No. PNL 0673413-3

FIGURE 3

DIAGRAM OF APPARATUS USED IN AIR DRYING OF SHALLOW POOLS OF CONCENTRATED PLUTONIUM NITRATE SOLUTION

liquid heated with the infrared lamps. Excess moisture entrained in the air was removed by the water-cooled condenser prior to filtering. The condensate was collected in a graduated cylinder and analyzed for plutonium at the end of the run. The particles airborne were trapped on the membrane filter. Separate filters were used to collect the material airborne during and after evaporation. After the heating-cooling cycle, the filters were examined microscopically and filters, condensate and CV wash analyzed for plutonium content. The amount of plutonium airborne during these runs was insufficient for determination of a size distribution.

2. Experimental

Run N1 -- Ambient temperature, air velocity 10 cm/sec. Three ml of concentrated $\text{Pu}(\text{NO}_3)_4$ solution used. Material appeared dry after 24 hours. Solids remaining on dish were a dark brown, with an irregular, glazed surface. Subsequent heating (run N-1a) indicated some moisture may have been trapped under surface.

Run N2 -- Three ml of concentrated plutonium nitrate solution heated to 75°C in 10 cm/sec air velocity. Maximum temperature indicated during run was 79°C . The material in the dish appeared dry after 5 hours. Filter was changed and sampling continued for an additional 20 hours. Solid remaining after evaporation was dark green in color with an irregular glazed surface. Surface was cracked and pitted.

Run N3 -- Volume of liquid used was reduced to 2-1/2 ml. This appeared to be maximum which could be retained on the dish using heat. Average temperature during the run was 110°C with a maximum of 115°C . Evaporation completed in approximately 2 hours and filter changed. Continued sampling for an additional 4 hours using same temperature and velocity. Solid residue was dark brown in color with a very irregular, glazed surface. Used as source for a long term storage test.

Run N4 -- This was an ambient temperature run using an air velocity of 50 cm/sec, and 2-1/2 ml of concentrated plutonium nitrate as the source. Specimen appeared dry after 24 hours under these conditions. The filter was changed and sampling continued for an additional 24 hours. Solids were dark greenish-brown in color with an irregular, glazed surface. After cooling, some liquid was observed under glazed surface.

Run N5 -- Two and a half ml of concentrated plutonium nitrate solution was heated in 50 cm/sec air. Initial heating rate was too high and liquid began to bubble after approximately 7.5 minutes of heating. Temperature was approximately 115°C. Heating rate was reduced but temperature increased for another 2.5 minutes reaching 125°C. The liquid bubbled violently during this period giving off brown fumes. After another 15 minutes the temperature decreased to 100°C which was maintained for remainder of the run (approximately 45 minutes). During this period, the liquid continued to bubble and give off brown fumes. The surface of the material remaining was very irregular and crusty. Heating was continued for an additional 15 minutes and then the filter was changed. After cooling, the solids had the appearance of dried mud. The solids were very unevenly distributed over the surface of the dish with many areas of the dish uncovered.

Run N6 -- Two and a half ml of concentrated plutonium nitrate solution was heated at 90°C in air with a velocity of 50 cm/sec. A maximum temperature of 99°C was attained. Sample appeared dry at the end of two hours at which time the filter was changed. Sampling continued for an additional 4.5 hours. After cooling, the solids on the dish were a dark, glazed mass with many ridges on the surface which had developed during cooling.

Run N7 -- Two and a half ml of concentrated plutonium nitrate solution dried at ambient temperature in air with a velocity of 100 cm/sec. Drying time was 24 hours and sampling continued for an additional 24 hours. The dried material was a dark, thin, crystalline crust with some liquid trapped under the crust.

Run N8 -- Air dried 2-1/2 ml of concentrated plutonium nitrate solution at 50°C (maximum temperature 57°C) in two hours using a velocity of 100 cm/sec. The filter was changed and sampling continued for 4 more hours. When first removed, the residue appeared to be a dark, solid mass but was found to be a viscous liquid on cooling. Material formed long, thin crystals upon an additional 24 hours drying in the glovebox (dewpoint of air is less than a minus 70°F).

Run N9 -- Two and a half ml of concentrated plutonium nitrate solution was air dried at 90°C (maximum temperature 97°C) using 100 cm/sec air. Surface of material was crystalline in 90 minutes, at which time the filter was changed. Sampling continued for 4 more hours. Solids remaining were dark with an irregular, glazed surface.

3. Results

Very little plutonium became airborne during the air drying of concentrated plutonium nitrate solutions. At temperature less than boiling, the largest amount released was 5.7×10^{-5} wt.% over a 2-hour sampling period (run N9). At ambient temperature, less than 2.5×10^{-7} wt.% of the plutonium was airborne in a 24-hour period. Even at temperatures high enough to cause violent bubbling (run N5), the amount airborne was only 0.003 wt.% in a 2-hour period. The amount of plutonium carried down with the condensate or depositing on the walls was in the same range as the airborne release value for a comparable sampling period. The amounts entrained from the solids remaining after the evaporation was minimal -- the maximum value found was 3×10^{-6} wt.% for a 4 hour period at 90°C in air with a velocity of 100 cm/sec. A tabulation of all results is given in Table I.

B. AEROSOL RELEASED DURING THE HEATING OF POOLS OF PLUTONIUM NITRATE IN A FLOWING AIR STREAM

1. Equipment & Procedure

Dilute aqueous solutions of plutonium nitrate were heated in one of four boiling regimes in an upsweep of air. The regimes were selected to represent a cross-section of heating rates and degrees of disturbance of the liquid surface to which a plutonium solution might be subjected during off-standard conditions. The regimes chosen were:

- a. "Simmering" - no surface disturbance and a liquid boil-off rate of approximately 0.4 ml/min (the exposed area was approximately 11.5 cm²).
- b. "First Breaking of the Surface" -- spasmodic breaking of the surface by bubbles with a liquid boil-off rate of ~ 0.6 ml/min.
- c. "Steady Breaking of the Surface" -- regular breaking of the liquid surface by bubbles with a liquid boil-off rate of ~ 0.8 ml/min.
- d. "Vigorous Rolling Boil" -- a smooth, steady stream of bubbles disturbing the liquid surface and a liquid boil-off rate in excess of 1.4 ml/min.

Although we attempted to approximate these regimes during our experimental work, the conditions are difficult to maintain. The degree of surface dis-

TABLE I

FRACTIONAL RELEASE DURING AIR DRYING OF CONCENTRATED PLUTONIUM NITRATE SOLUTIONS

(Using 0.72g Plutonium as a source)

Run No.	Temp. (°C)	Air Velocity (cm/sec)	Sampling Time		Weight Percent Plutonium Found In			
			Evap. (hr)	Residue (hr)	Containment Vessel Wash	Condensate + Wash	Sweep Air During Evaporation	Sweep Air Following Evaporation
N1*	Ambient	10	24		0.0033	8.7×10^{-3}	$<10^{-7}$	--
N2*	75	10	5	20	0.00027	9.5×10^{-8}	$<10^{-6}$	$<10^{-6}$
N3	100	10	2	4	0.0046	1.7×10^{-6}	0.001	3×10^{-7}
N4	Ambient	50	24	24	0.00035	4.5×10^{-7}	2.5×10^{-7}	1×10^{-7}
N5	100	50	1-1/2	3	0.027	1.4×10^{-4}	0.003	6×10^{-7}
N6	90	50	2	4-1/2	0.00051	5.4×10^{-6}	5.3×10^{-5}	1×10^{-6}
N7	Ambient	100	24	24	0.020	7.5×10^{-8}	$<2 \times 10^{-8}$	$<2 \times 10^{-8}$
N8	50	100	2	4	0.00045	9.4×10^{-6}	1.3×10^{-5}	$<2 \times 10^{-8}$
N9	90	100	1-1/2	4	0.00013	9.4×10^{-5}	5.7×10^{-5}	3×10^{-6}

* 0.86 g plutonium used during these runs.

turbance is a subjective criteria. The degree of surface disturbance to approximate a simmering or vigorous rolling boil were relatively easy to maintain by adjusting the heat input after attaining the desired conditions. For the other two regimes, the specified degree of surface activity was approached from both ends. Thus, the surface activity did not reach or exceeded the stated conditions during some portions of these runs. Therefore the runs are labelled simmering, disturbed surface and boiling which approximate more closely the conditions present.

An attempt was made to determine the convective air velocities created over the liquid during each of these regimes without the containment vessel in place. Velocities were below the sensitivity of the vane anemometer used. A value of 2.9 cm/sec through the annular filter was selected somewhat arbitrarily but is believed to be within the range of convective flows achieved.

The apparatus used is pictured in Figures 4 and 5.

The system consisted of:

- A 180 ml pyrex beaker to hold the liquid. The beaker was supported on the transite ring by a fine-mesh, stainless steel screen in which a hole matching the beaker diameter was punched. The screen also supported a matching annular glass fiber filter used to collect plutonium particles escaping from the beaker but too large to remain airborne.
- A slotted aluminum heating-block around the beaker. The aluminum heating-block sat on a hot plate. Both were supported on a movable platform which facilitated rapid heating and cooling during the run.
- A containment vessel made from a 4-liter pyrex jug. The closed end of the jug was removed and a condensate collection trough was formed integrally with the inner bottom rim. The bottom rim was then ground flat to assure good sealing with the "fall-out" filter. Particles entrained in the upsweep air were caught on a glass fiber filter held in a glass holder which formed the "neck" of the jug.
- A water-cooled condenser. The glass filter holder was connected to the outlet by a ball-socket joint. The connector was wrapped with heating tape to prevent condensation of moisture in the connector. Moisture escaping from the containment vessel was removed by the water-

FIGURE 4
APPARATUS USED TO DETERMINE FRACTIONAL RELEASES DURING
THE HEATING OF POOLS OF PLUTONIUM NITRATE SOLUTIONS

FIGURE 5

DIAGRAM OF APPARATUS USED TO DETERMINE FRACTIONAL RELEASES DURING THE HEATING OF POOLS OF PLUTONIUM NITRATE SOLUTIONS

cooled condenser and collected in a graduated cylinder connected to the condenser by a standard tapered joint. The air was again filtered before passing through a calibrated flowmeter.

Plutonium nitrate solution containing the equivalent of 700 μg of plutonium was pipetted into dilute nitric acid in the beaker. The beaker and contents were subjected to a selected heating regime and the amount airborne determined. All the equipment was first assembled except the containment vessel. Two and a half ml of a dilute plutonium nitrate solution was mixed with 97.5 ml of 0.25 M nitric acid held in the 180 ml beaker. The containment vessel was put in place and 1 cfm of air drawn through the system. The solution was quickly heated to the desired temperature by raising the pre-heated aluminum block into place. Only 90% of the liquid was boiled off. During run E and J, air was not drawn through the system and the amount airborne was determined by analysis of the condensate. Only 70 μg plutonium was used in these runs. To determine the effects of concentration, the filter was changed after 30 ml was boiled off during the last 5 runs (F through J). Particle size analysis by microscopy was not possible due to the small amounts of plutonium on the filter.

2. Experimental

Some of the operating conditions during these experiments are listed in Table II.

Run A -- First signs of condensate were detected after ~ 17 minutes of heating. After 27 minutes the liquid boiled steadily. After a few minutes more, bubble size increased and the solution began "bumping". The annular fallout inlet filter became water logged after ~ 35 minutes of operation and caused the beaker to rise due to pressure drop. The heating block was raised to maintain contact. The run was terminated after 82 minutes with 90 ml of liquid boiled off. The nitric acid in the vapor was found to have destroyed the membrane filter used. (Prior tests indicated this would not occur.) All plutonium found on the filter and in condensate plus washes of entire system downstream of filter holder was considered as the amount airborne.

Run B -- First signs of condensate in 14 minutes with "first breaking of the liquid surface" (bubbles) in 18 minutes. Liquid was boiling steadily after 25 minutes with some tendency to "bump". Maintained conditions till end of run (95 minutes). Maximum temperature at hot plate of $\sim 212^\circ\text{C}$.

TABLE II

FRACTIONAL RELEASE DURING THE HEATING OF POOLS OF PLUTONIUM NITRATE SOLUTIONS

(90% solution boiled off, 700 μg plutonium as source,
2.9 cm/sec air up sweep during evaporation.)

Run No.	Average Hot Plate Temp. (°C)	Average Boil-Off Rate (ml/min)	Period Time Heated (min)	Plutonium Recovered				Visual Appearance of Solution During
				Exhausted by Sweep Air		Fall-Out		
				(dpm)	(wt.%)	(dpm)	(Wt.%)	
A	218	1.4	63	2×10^5	.18**	2.3×10^4	.02	Boiling
B	190	0.9	80	9.4×10^4	.084	3×10^5	.27	Disturbed Surface
C	175	0.73	124	2.7×10^4	.024	1.3×10^4	.012	Disturbed Surface
D	150	0.6	151	500	4.5×10^{-4}	500	4.5×10^{-4}	Simmering
E*	220	2.1	42	$\sim 5^*$	$\sim 4.5 \times 10^{-5}^\dagger$	~ 23	$\sim 2 \times 10^{-4}$	Boiling
F	150	0.5	150	150	1.3×10^{-4}	~ 3	$\sim 3 \times 10^{-6}$	Simmering
G	164	0.66	121	6.5×10^3	.0058	1.9×10^4	.016	Disturbed Surface
H	188	1.2	64	1×10^4	.008	8×10^5	.71	Disturbed Surface
I	218	1.4	59	3×10^4	.03	2.3×10^4	.02	Boiling
J*	200	1.4	66	1.2×10^4 *	.11 [†]	1.9×10^5	1.7	Boiling

* No air flow through containment vessel, 70 μg plutonium as source. ("Airborne" is that found on containment vessel wall following this run.)

** Filter ruptured. All plutonium found in acid washes of equipment downstream of filter position.

† Plutonium in condensate.

Run C -- First condensate in 18 minutes and "first breaking" in 26 minutes. Liquid began "bumping" after 28 minutes and heat was reduced. Hot plate temperature was 160°C at 57 minutes and increased heat. Solution began "bumping" again at 75 minutes with a temperature of 185°C and again reduced heat. Liquid simmered for next 40 minutes. Run terminated after 148 minutes with 10 ml remaining.

Run D -- Condensate in 18 minutes and liquid simmering in 25 minutes. Heat was reduced after 30 minutes. Conditions remained steady till end of run (169 minutes). Maximum temperature hot plate during run was 165°C. No detectable amounts of activity anywhere in system with alpha survey instrument: only filter samples submitted for analyses.

Run E -- Condensate drain and filter opening on containment vessel sealed off during run. First appearance of condensate in 14 minutes. After 24 minutes heating, liquid began bumping violently and continued to do so till end of run (61 minutes).

Run F -- Condensate first appeared after 15 minutes of heating and "first breaking" detected after 39 minutes. Heat was cut back and liquid simmered at 42 minutes. This condition was maintained till end of run. Initial filter changed at 70 minutes. About 7 ml of liquid evaporated during filter change.

Run G -- First condensate detected after 17 minutes. "First breaking of liquid surface" reached in 26 minutes. After 28 minutes the liquid was at a slow, steady boil and heat reduced. Solution simmering and began reheating at 39 minutes. Reestablished "first breaking" conditions at 54 minutes. Continued adjusting heat to maintain conditions till end of run (138 minutes). Temperature of heating block ranged from 142°C to 208°C during this period. Initial filter change after 68 minutes. About 7 ml evaporated during the filter change. Lip of beaker was reading $\sim 10^3$ dpm by survey alpha meter during filter change and $\sim 4 \times 10^4$ dpm at end of run.

Run H -- Polyvinyl filters with 6 μ pore size were used. First sign of condensate appeared in 19 minutes. Solution began "bumping" at 24 minutes. (Some liquid splattered on containment vessel wall.) Heat was reduced and the heating block was lowered. Reheating started at 37 minutes. Conditions maintained till end of run (83 minutes). Initial filter changed at 45 minutes. Liquid level decreased 10 ml during filter change. Particle size analysis made on

a portion of polyvinyl chloride filter by alpha track counting of NTA nuclear track film.

Run I -- First indications of condensate after 19 minutes. Liquid began "bumping" and boiling at 22 minutes. Conditions were same till end of run (78 minutes). Initial filter changed at 37 minutes.

Run J -- Condensate drain and filter outlets were sealed. Only 70 μ g plutonium was used as a source. Condensate appeared after 18 minutes heating. Liquid began to "bump" violently at 27 minutes and continued to do so for remainder of run.

3. Results

The fractional release obtained during the heating of pools of dilute plutonium nitrate solutions are listed in Table II. There appears to be a rough correlation between boil-off rate and amount swept out of the containment vessel by the exhaust air. At low boil-off rates (runs D and F), very little (approximately 5×10^{-4} wt.%) is carried out. As the boil-off rate increases, the amount swept out increases. The highest values for aerosol airborne (0.18 and 0.11) are at the highest boil-off rates (runs A and J).

These high values are not directly comparable to the values given for the lower boil-off rates. The membrane filter used during Run A was punctured by the acid fumes and the value given for the amount airborne is the total amount of plutonium collected downstream of the containment vessel outlet plus condensate from the containment vessel. Thus, this value (0.18 wt.%) is a composite of the material normally captured by the exhaust filter and the particles which are deposited on the walls of the containment vessel. It is believed to be at least 15 to 25% high. The value of run J (0.11 wt.%) is a similar situation. Air was not drawn through the vessel during this run. The value is taken wholly from the condensate and again is undoubtedly high.

There are some apparent inconsistencies. Run E at the highest boil-off rate has the lowest release value. Both the airborne and the fallout filter values indicate little if any plutonium and it appears probable that the plutonium nitrate was inadvertently omitted during this run. It is also noted that Run I at the highest boil-off rate shows a considerably lower release value than similar runs A, and J. The release values for runs A and J represent the activity which was deposited on the walls as well as that airborne and show

reasonable agreement with each other. Boil-off rate is a rough approximation but many factors contribute to the behavior of the surface. We had considerable trouble maintaining uniform conditions during the course of these runs (see preceeding section B.2 Experimental) and the boil-off rates are average values. The total amount leaving the beaker (airborne plus fall-out) shows even a better correlation with boil-off rate indicating the part liquid film breakup plays in the amount airborne.

As the liquid becomes more concentrated, the amount released increases. Table III lists the amounts of plutonium found on the filters used during the boil-off of the first 30 ml of liquid compared to that found on the filter used during the boil-off of the last 40 ml. (Usually 7 to 10 ml of the liquid boiled off during the filter change.) Time required to obtain these samples generally bear the same ratio, 3:4, indicating approximately the same boil-off rate. Even correcting for this difference in sampling time, the second filters are significantly higher than the initial filters used. Again, the obvious explanation for the inconsistency (run I) are the local conditions at the liquid surface during each portion may have created a more disturbed surface at one portion of the run than another.

Thus, we deduced that under ordinary conditions of liquid heating, the amount of plutonium airborne is small. The amount released is a function of the degree to which the surface is disturbed and has a rough correlation with "boil-off" rate. Local conditions at the surface increasing the breaking of the surface film such as "bumping" can increase the amount released. The amount airborne also depends upon other conditions surrounding the liquid film break-up. Under the most vigorous conditions imposed during these experiments, as much as 0.18 wt.% was airborne at a short distance ($\sim 3''$) from the source. The amount which would be swept from the vessel was undoubtedly less.

C. THE AEROSOL RELEASED DURING THE HEATING OF THE SOLIDS REMAINING AFTER THE EVAPORATION OF PLUTONIUM NITRATE SOLUTIONS

1. Equipment and Procedure

The solids resulting from the air drying of concentrated plutonium nitrate solutions described earlier were heated in the apparatus used to determine the fractional release from plutonium compounds in powder form.^[5]

A photograph of the experimental system chosen is shown in Figure 6, and

TABLE III

VARIATION IN RELEASES DUE TO CONCENTRATION DURING HEATING POOLS
OF DILUTE PLUTONIUM NITRATE SOLUTIONS

Run No.	Plutonium				Filter
	Airborne		As Fall-Out		
	(dpm)	(wt.%)	(dpm)	(wt.%)	
F	55 m ~83	7×10^{-5}	~150	~0.0014	First
	113 m ~1	$\sim 10^{-6}$	1.8	2×10^{-6}	Second
G	53 m 1500	0.0013	2500	0.0022	First
	68 m 5000	0.0045	16,000	0.014	Second
H	25 m 2340	0.0021	1.1×10^5	0.098	First
	32 m 7260	0.0065	6.9×10^5	0.62	Second
I	22 m 1300	0.0012	1.38×10^5	1.23	First
	36 m 15400	0.014	6.8×10^5	0.61	Second

FIGURE 6
SOLIDS RELEASE APPARATUS

Neg. No. PNL 0673413-1

a schematic diagram in Figure 7. The principal parts of the apparatus are:

- A 15 kW, 10 kHz induction heating unit*.
- A water-cooled work coil of 1/4 inch 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-inch diameter well was drilled into each cap from its rim to the centerline to hold a thermocouple for measuring run temperatures (see Figure 8).
- A platinum-platinum 13% rhodium thermocouple.
- A 44 mm OD quartz chimney to carry the entrained particles upward to the filter.
- A pyrex 2-inch filter holder with a fritted glass support modified to minimize the pressure drop across the holder.
- A temperature recorder**.

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 8). 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 the chimney and either deposited on the 0.003-inch mild steel chimney liner or collected on a 2-inch 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 microns diameter (see Figure 9). 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.

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

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

† 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 7

APPARATUS FOR MEASUREMENT OF RELEASE FROM OVERHEATED
SOLIDS FROM PLUTONIUM NITRATE SOLUTION EVAPORATION

FIGURE 8
STAINLESS STEEL FURNACE CAP AND BOTTOM OF QUARTZ CHIMNEY

FIGURE 9
EQUIVALENT DIAMETER PuO_2 PARTICLE LIFTED BY VARIOUS AIR VELOCITIES

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.

Plutonium released was determined by:

1. Placing a specimen of the solids remaining after evaporation in the depression in the stainless steel furnace cap in the apparatus.
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-inch membrane filter in a disposable holder, a pair of tweezers, a 2-inch glass fiber filter, and a piece of 0.003 inch mild, steel shimstock chimney liner in a Pyrex carrier were loaded into the glovebox prior to each run. Each item was packaged separately in a plastic bag to minimize contamination.

The 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-inch 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 reestablished. 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°C to 250°C per minute. Figure 10 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 $\pm 2\%$) for one hour. Sampling, to

FIGURE 10
 TYPICAL TIME VERSUS TEMPERATURE CURVES FOR 3 EXPERIMENTAL
 TEMPERATURES USED IN STUDY OF RELEASE FROM SOLIDS

include material released during cooling, was continued for one hour after the heat was turned off.

The chimney filter and liner 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 analyses. Special handling procedures were used during processing to minimize the effects of cross-contamination.

2. Experimental

Run N1a -- The dark brown solids remaining after air-drying run N1 were heated for ~ 1 hour at 1000°C in an upswEEP of air with a nominal velocity of 100 cm/sec. Solids began to bubble at a temperature of ~ 500°C and solidified into large, thin flakes at ~ 800°C. The material in the dish after the 1 hour cooling period was green in color. All but the bottom 2" of the interior of the shimstock chimney liner appeared rusted by the NO₂ fumes generated during the heating cycle. A microscopic examination of the glass-fiber chimney filter revealed a few particles -- some fragmentary particles 7 to 60 microns in diameter, a few large flakes ~ 125 microns in diameter and some amorphous material 50 × 100 microns.

Run N2a -- The solids remaining from run N2 were heated for one hour in an upswEEP of 100 cm/sec air. The source material was a dark greenish-brown solid with a glazed surface. Specimen began to bubble at ~ 290°C and solidified into large, thin, green-colored flakes at ~ 700°C. A large amount of brown fumes accompanied the bubbling. The chimney liner was again rusted. A few particles were seen on the glass fiber chimney filter -- primarily black ash-like particles (fragments of graphite susceptor) with a few yellow, translucent particles ~ 50 microns in diameter.

Run N4a -- Solids resulting from air drying plutonium nitrate at ambient temperatures were heated at 400°C in air with a velocity of 100 cm/sec. The source material was dark brown in color with an irregular, glazed surface. Some of the liquid trapped under the solid surface escaped onto the surface

during handling. Solids bubbled and emitted brown fumes during heating and solidified into a few large, thin, green flakes after cooling. Many black ash-like particles were also visible on the dish which was discolored in spots. Even a greater area of the interior of the shimstock liner appeared rusted. Most of the particles observed on the chimney filter were black ash-like particles but a few yellowish, translucent particles ranging in size from 20-30 microns in diameter were also observed.

Run N5a -- The source material used in this run was significantly different from those used in any other runs. It was tan in color and had the appearance of expanded mud. The solids had more bulk than the other source materials and covered only one half the surface area of the dish. Only a minor amount of bubbling and fumes were observed during the 1 hour heating cycle. After the sample was cooled, little change could be observed in the solid -- a slight volume increase may have taken place. There were a considerable number of light-brown spherical particles with a few larger brown particles up to 160 micron in diameter. A sufficient number of particles could be observed for a size distribution to be determined.

Run N6a -- The solids remaining after run N6 were heated to 1000°C in 10 cm/sec air. Source material was a dark mass covering the entire surface of the furnace cap with a glazed, deeply ridged surface. The specimen bubbled and emitted reddish-brown fumes during initial portion of 1 hour heating cycle (from ~ 100°C to 400°C). The material remaining after heating was considerably reduced in volume and had the appearance of a small charred deposit on the cap. The interior of the chimney liner was rusted except the lower 2". Particles on the filter were mostly black ash-like with a few yellow, translucent spheroids.

Run N7a -- Dark, crystalline solids from run N7 were heated at 700°C in 50 cm/sec air. A small amount of moisture was observed on the glass section behind the filter soon after run was initiated. Behavior of source materials was similar to that described in previous runs except run N5a. Material remaining in the dish after cooling was a mass of lumps ~ 1/8" in diameter. Filter contained black ash-like particles.

Run N8a -- Thin, crystalline solids from run N8 were heated at 400°C in air with a velocity of 50 cm/sec. The glass fiber insulator around the graphite core ignited during the run but was extinguished with little diffi-

culty. Material behavior was similar to other runs. Many thin, light-weight particles having an appearance like paper-ash remained after heating. The chimney liner was corroded. Particles, if any, on chimney filter were obscured by soot from the burning glass fiber binder.

Run N9a -- Crystalline solids from run N9 were heated at 1000°C in 100 cm/sec air. The solid had lightened in color during the 30-day storage in the glovebox atmosphere. Temperature was maintained with some difficulty. The material behaved as in other runs and produced a chimney liner and filter similar in appearance to those in previous runs.

3. Results

The amount of aerosol released during the heating of the solids remaining after the evaporation of plutonium nitrate solutions is roughly equivalent to heating the liquid and shows some correlation to upsweep air velocity and temperature (see Figure 11). The physical condition of the source material is also a significant factor. In Table IV is a tabulation of the fractional releases determined during the experiments.

TABLE IV
FRACTIONAL RELEASE FROM SOLIDS REMAINING AFTER EVAPORATION
OF PLUTONIUM NITRATE SOLUTIONS
(0.72g plutonium used, 2 hours sampling time.)

Run No.	Temp. (°C)	Air Velocity (cm/sec)	Plutonium				Total Evolved (wt.%)
			Deposited In Chimney		Carried Through Chimney		
			(dpm)	(wt.%)	(dmp)	(wt.%)	
N1a*	1000	100	9.2×10^6	.0067	1.7×10^7	.012	.0187
N2a*	700	100	5.3×10^6	.004	1.2×10^7	.0092	.0132
N4a	400	100	1.1×10^7	.0097	7.6×10^6	.0068	.0165
N5a	1000	50	3.6×10^7	.032	1.4×10^8	.12	.152
N6a	1000	10	1.8×10^6	.0016	5.1×10^6	.0046	.0062
N7a	700	50	1.7×10^7	.015	8.4×10^6	.0075	.0225
N8a	400	50	6.2×10^6	.0055	7.2×10^6	.0064	.0019
N9a	1000	100	1.4×10^6	.0012	7.9×10^6	.0071	.0013

* 0.864 g plutonium used.

FIGURE 11

VARIATIONS IN FRACTIONAL RELEASE FROM SOLIDS
AS A FUNCTION OF AIR SWEEP VELOCITY

The amount of material released during the heating of these nitrate solids, (excluding the value found for run N5a) ranged from 0.0046 to 0.012 wt.%. All exhibited the characteristics of nitrate decomposition to oxide.^[6] The density of the material is reported to increase with increasing calcination temperature.^[8] Increasing the density should decrease the entrainment but apparently other factors more than compensate for the density effect.

Airborne release in run N5a does not appear to be the entrainment from a decomposing solid nitrate. The nitrate behavior during the preparation of the source material (bubbling and generation of reddish-brown fume) indicated a change in chemical composition. Considerably less oxides of nitrogen (reddish-brown fumes) were given off during the heating of this specimen compared to the amount detected with the other specimens.

The solids used as a source material in run N5a were quite different in appearance than the material used for the other runs. It had the color of the oxide produced from the nitrate -- a dull yellow^[9] -- rather than the green-brown color of the rest of the solids used. The volume was greater and the material had much more surface area due to its volume and irregular surface.

Thus, the value for run N5a, 0.12 wt.% airborne, is probably the entrainment from a finely-divided oxide rather than from a decomposing nitrate. This value is higher than that previously reported for entrainment from the 15 to 44 micron fraction of the oxides from the ignited metal^[5] and probably reflects the small particle size of material present. The Median Mass Diameter of the particles airborne during this run was 20.5 microns (see Figure 12).

Another factor which appeared to have affected the airborne release during these experiments is the presence of liquid trapped beneath the solid surface. The source material used for runs N1a, N4a and N7a were all air-dried at ambient temperatures and liquid was observed under the surface. Run N1a and N9a were run using the same conditions but run N1a had twice the amount airborne found for run N9a (0.012 wt.% compared to 0.0071 wt.%). The difference in the total amount released (airborne + deposited on chimney) is even larger for these two runs. A considerably greater percentage of the total amount released during runs N4a and N7a was deposited on the chimney. The presence of moisture may have created a different size distribution during these runs or material may have been forcefully impinged on the wall by the force of escaping vapor. In either case, some of the material would remain

FIGURE 12

SIZE DISTRIBUTION OF PARTICLES RELEASED DURING RUN N-5a

airborne under slightly different conditions. Thus, the presence of moisture under the type of surface resulting from air-drying plutonium nitrate can result in more material airborne.

The amount of aerosol released during the decomposition of the solid residues from the air-drying of plutonium nitrate solutions are dependent on many factors. When subjected to low temperatures, the solids form in a continuous layer. Heating these solids releases amounts of aerosol in the same range as found for heating liquids. If the air-drying occurs at temperatures high enough to produce the oxide but does not calcine the mass, the solids are finely-divided and greater amounts of aerosol can be released. Mechanisms increasing the surface area or producing a more divided solid will increase the material airborne.

CONCLUSIONS AND FUTURE WORK

The amount of plutonium which can be carried away by passing air up and over heated aqueous plutonium nitrate solutions brackets the range found for the oxides from ignited metal, Table V lists the values determined for the plutonium materials studied thus far. The fraction airborne at low temperatures (less than 135°C) from the solid residues formed by the evaporation of aqueous plutonium nitrate solutions is equivalent to that entrained during the oxidation of ignited metal. The amount is very small -- $<3 \times 10^{-6}$ wt.%. Somewhat more was released during the air drying of the aqueous plutonium nitrate solutions -- up to 0.003 wt.%.

The amount carried away during the forced evaporation of plutonium nitrate solutions can be considerably higher. With an undisturbed liquid surface, the amount is low -- up to 4.5×10^{-4} wt.%. Disturbing the surface increases the amount -- up to 0.084 wt.%. A considerable amount of material is airborne at a short distance from the liquid during boiling of plutonium nitrate solutions (0.18 and 0.11 wt.%) and, under proper conditions, could be swept away in a stream of air.

A similar amount of aerosol is released during the decomposition and heating of the solid residues from air-drying of plutonium nitrate solutions (0.0046 to 0.12 wt.%). The physical conditions of the residue plays an important part in determining the amount airborne. The largest amount airborne,

TABLE V

PLUTONIUM AEROSOL AIRBORNE UNDER STATED CONDITIONS

Solids from evaporation of plutonium nitrate solution: Ambient to $\sim 130^{\circ}\text{C}$, 10 to 100 cm/sec air, sampled 3 to 24 hours.	10^{-8} to 2.6×10^{-6} wt.%
Oxidation of ignited metal: 560° to 900°C , 3.3 to 50 cm/sec air, sampled 74-155 min.	2.8×10^{-6} to 5.3×10^{-5} wt.%
Oxidizing ignited metal covered with MgO sand: 525 cm/sec air, sampled 60 min.	0.00030 wt.%
Air drying plutonium nitrate solutions: ambient to $\sim 130^{\circ}\text{C}$, 10 to 100 cm/sec, samples 1-1/2 to 24 hours.	10^{-7} to 0.003 wt.%
Fine oxide from ignited metal: ambient to 900°C , 10 to 100 cm/sec air.	5.3×10^{-6} to 0.025 wt.%/hour
Oxide from ignited metal: ambient, 6.9 cm/sec air, sampled 20 to 90 min.	8.6×10^{-6} to 0.0311 wt.%
Oxidizing ignited metal: 525 cm/sec air, sampled 22 to 90 min.	0.0045 to 0.032 wt.%
Fluoride powder: 400° to 1000°C , 10 to 100 cm/sec air.	0.007 to 0.05 wt.%/hour
Nitrate solution: simmer to rolling boil, 2.9 cm/sec air, sampled 59 to 151 min.	0.00045 to 0.18 wt.%
Solids from evaporation of plutonium nitrate solutions: 400° to 1000°C , 10 to 100 cm/sec air, sampled 2 hours.	0.0046 to 0.012 wt.%(0.125)
Oxalate powder: 400°C to 1000°C , 10 to 100 cm/sec air.	0.005 to 0.9 wt.%/hour
Partially oxidized oxalate powder: 400°C to 1000°C , 10 to 100 cm/sec air.	0.041 to 0.82 wt.%/hour

0.12 wt.%, was found during the heating of the bulky, oxide-like powder formed by overheating the nitrate-solid during air-drying.

The solids studied here were in thin layers. We believe this is a more probable configuration with liquid residues than thick cakes. Whether the same amount would be airborne from a thicker cake was not determined but we infer from the behavior of the powders used in the previous study that the hardened surface layer produced by heating to high temperatures would constitute a barrier to particle entrainment deeper in the mass. Thus, thin layers seem more conducive to greater fractional airborne releases.

It appears then that in overheating situations aqueous plutonium nitrate solutions constitute a somewhat greater potential inhalation hazard than the metal. Under certain conditions, greater fractional releases can be obtained from the liquids and solid residues from the evaporation of liquid than from the oxides produced by the ignited metal.

The results reported must be interpreted as being indicative of the amounts which could be made airborne under the circumstances selected. Using the data without interpretation is only valid under exactly the same circumstances described in the experiment. Since these conditions are not intended to simulate any particular set of accident conditions, the data must be interpreted for application to most practical situations.

Other accident situations involving plutonium and its compounds in other physical form not yet tested can also occur. Two of the conditions are currently being studied. Plutonium compounds mixed with combustible materials as in packaged contaminated wastes is a material commonly found in all facilities handling these compounds. The amount and size distribution of the activity airborne by the burning of the combustible materials is under study. Heating of plutonium nitrate solutions mixed with soil or sand is also a potential situation especially in the transportation of plutonium. This situation will also be investigated by heating the sand and/or using a flammable material directly on the sand.

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