A REVIEW OF RESEARCH ON PLUTONIUM RELEASES
DURING OVERHEATING AND FIRES

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August 1964

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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ABSTRACT

Articles and documents are reviewed which are concerned with oxidation and ignition temperatures of metallic and delta-phase stabilized plutonium, particle size distribution of oxide produced by heating plutonium and some of its compounds, deposition from turbulent streams, resuspension of plutonium dioxide particulate material from various surfaces, or release of plutonium during experimental and accidental fires.

In most instances, plutonium oxidation rates are found to increase with increasing temperature and humidity. A minimum rate is found in the delta-phase region due to changes in the oxide layer. Ignition can occur at temperatures above 100 C depending upon the conditions of the metal and heating. The rate of release of plutonium dioxide aerosol increases with increasing temperature and gas velocity. Conditions in and around fires are variable and forces affecting the deposition of particles, complex. Once deposited in the soil, particulate material shows little tendency to resuspend. Several plutonium resuspension factors embracing many types of circumstances are listed.

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INTRODUCTION

This report is a review of the literature on plutonium release due to fires or overheating. The review was undertaken to consolidate the research results available and to disclose those areas where research is needed.

The relevance of such a review becomes apparent when one considers that the world's supply of plutonium will soon be measured in tons,[1] and that many facilities will be storing and fabricating plutonium fuels. This increased usage and handling will increase the possibility of plutonium release with subsequent inhalation exposure hazards.

The information collected is concerned with the fractional release from different sources under varying conditions, deposition, and resuspension. Also, the physical and chemical properties of the released radioactive materials must be known if the potential exposure hazard is to be assessed.

BIOLOGICAL HAZARDS

Plutonium-239 is one of man's most toxic substances. The International Commission on Radiological Protection (ICRP) recommends a maximum permissible body burden of 0.04 μCi* or 0.6 μg, and maximum permissible concentrations in water and air for the general population of 1.7 x 10^{-6} and 2 x 10^{-14} μCi/cm^3, respectively. These values are taken to be 1/30 of the "continuous" occupational recommended maximum permissible concentration.[2]

The primary route of plutonium deposition in man is via inhalation. Except in a few situations in which large masses of the metal are handled, the exposure to external radiations from plutonium is small. Uptake in man following ingestion of plutonium is relatively low. Experimental data from animal exposures under a variety of conditions indicate that 0.002 to

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*   The abbreviation μCi will be used for microcurie in this report, as recommended by the International Union of Pure and Applied Physics.

to 0.007%[3,4] is taken up in the gastro-intestinal tract. Additionally, plutonium does not exhibit any tendency towards concentration at any stage of man's food cycle. Field and laboratory studies[5] have shown that the ratio of plant to soil concentration ranges from about 10^{-4} to 10^{-3}. Plutonium dioxide, the most likely compound present in accidental releases, is difficult to dissolve under laboratory conditions and may be solubilized only to a slight degree in body fluids. There is evidence, however, that fine particles are transferred through the alveolar membrane.[6]

The ICRP formula for the retention of particulate material in the lungs assumes that 12.5% of all insoluble materials taken into the lungs is retained and taken up in the body fluids with a half-life of 120 days.[2] Langham's modification assumes 10% will be solubilized and taken into the body fluids.[7] By either standard, the principal mode of uptake by man is inhalation of finely divided particulate matter.

OXIDATION RATES AND IGNITION TEMPERATURE

Both volatilization and release of particulate material may contribute to the total airborne concentration of plutonium. Hilliard, in a short review of the data available in 1961, concluded that true volatilization of plutonium or its dioxide is likely not a significant factor in the airborne hazard.[8] The amount of particulate material suspended in air as a result of the oxidation of plutonium depends on many factors--the amount and particle size distribution of the oxide produced, the deposition of particles along the path to the point of interest, and any resuspension of the deposited material.

The amount and particle size distribution of the oxide depend upon the amount and the physical division of the starting material and prior treatment. The oxide produced from a well dried oxalate heated slowly to 300 to 600 C was in the 2 to 7 µ size range.[9] Continuously precipitated oxalate that was calcined at temperatures from 420 to 720 C produced plutonium dioxide with median mass diameters from 5.2 to 12.0 µ.[10-12] Oxide produced by French experimenters[13] from oxalate for use in reactor fuel preparation was 0.4% less than 325 mesh (~44 µ) when calcined at 550 C. Oxides produced by calcination of fluorides at 550 C were 2.1% less than 325 mesh, but virtually no particles were less than 44 µ when fluorides were calcined at 700 to 800 C.[14]

The oxidation of metallic plutonium has been pictured as a rapid initial attack of the effective surface area producing an adherent, protective layer of cubic and hexagonal Pu_2O_3.[15] This protective layer has been reported to range from ~ 3 μ[16] to 50 μ[17] in depth from room temperature through the gamma phase region in air and oxygen. Once formed, the protective layer limits oxidation and oxidation rates are linear (diffusion controlled) unless the protective layer is disturbed.

The oxidation rate of plutonium increases with increasing temperature until the delta phase region (315 to 452 C) is reached. At room temperature Waber[18] reports only 0.96 mg/cm^2 weight gain in 900 hr in dry air. Stewart[16] reports little change in specimens after 1 wk in dry air. At 100 C in dry air, Sackman[19] observed the oxidation follows a succession of parabolic curves. Dempsey and Kay[20] reported two-stage linear oxidation at 205 C, linear oxidation at 303 C, parabolic at 416 C, and initially parabolic accelerating to ignition at 487 C. A minimum oxidation rate has been reported by Notley et al.[15] in the delta phase region and was attributed to an increased thickness in the adherent layers.

Recently, Schnizlein and Fisher[21] in their oxidation studies using air and oxygen proposed a two-step oxidation mechanism from 120 to 560 C. The initial parabolic curve in the low temperature region is shown to have an activation energy of 32 kcal/mole and the subsequent linear curve an activation energy of 16 kcal/mole. Values of 32 and 20 kcal/mole are assigned to the parabolic and linear rates above 420 C.[17]

The acceleration of plutonium oxidation by moisture has been recognized since early studies at Los Alamos.[22] Dempsey and Kay[20] showed the strong dependence of room temperature oxidation on moisture. Waber[18] reported a 65% increase in oxidation rates from dry air to 50% relative humidity. Sackman[19] found moisture can cause the breakup of the oxide layer before completion of oxidation, indicating trans- or inter-granular corrosion. In his most recent report with Raynor,[23] Sackman finds higher corrosion rates in humid atmospheres in the absence of oxygen, which, if present, would tend to seal the oxide layer from further corrosion

by moisture. Stewart[16] reports a visible increase of oxidation at 100% relative humidity. Schnizlein and Fisher[24] indicate an oxidation rate increase by a factor of 6 when the moisture content is raised from <5 to 510 ppm at 190 C in oxygen. No difference in rates was observed in oxygen and air at 240 C under the same moisture conditions. This agrees well with the findings of Dempsey and Kay[20] at 55% relative humidity in air at 205 C, and indicates moisture loses its effect around the beta-gamma transition temperature (210 C).

The change in oxidation rate with crystalline phase of plutonium is significant. Oxidation rates increase with increasing temperature until a maximum is reached near 416 C.[15] Notley et al. attribute the lowering of oxidation rates in the delta phase region to a balance between the increased thickness of the protective Pu_2O_3 layer and the decreased oxygen diffusion. Stewart[16] proposes the similarity of crystalline forms of the delta phase metal and the dioxide provides a more adherent and partially crystalline oxide layer. Schnizlein and Fisher[17] attribute the lowering of rates in this region to changes in oxide properties in this region. Waber;[18] Waber, Olsen, and Roof;[25] Sackman;[19] Notley, Hodkins, and Davison;[15] Carter, Foy, and Stewart;[26] Stewart;[16] and Schnizlein and Fisher[24] have all reported decreases in oxidation rates in orders of magnitude by the addition of various metals stabilizing the delta phase of plutonium. Stewart[16] and Schnizlein and Fisher[24,27,28] have also reported increases in the ignition temperature of various plutonium delta phase binary alloys. Sackman[29] assigns an activation energy of 21 kcal/mole to the linear oxidation of the delta alloy at 95% relative humidity in air.

As metallic plutonium or its alloys are heated, the material can ignite. If the reaction is self-sustaining, the entire bulk of the material will oxidize and considerably more material becomes available for suspension. Frank-Kamenetskii[30] suggests that the ignition temperature is the gas temperature at which the heat generated by the reaction exceeds the heat transfer capacity of the protective oxide film. At this point, the

temperature at the point of reaction (and throughout the metal with a material of the thermal conductivity of plutonium) builds up rapidly. The reaction rate is quickly transported to a higher regime until the reaction rate is limited and the heat transfer characteristics exceed the heat generated. At this time the temperature returns to near the original point and the cycle is repeated. Hilliard[31] and Wanklyn[32] have reported that uranium exhibits this type of ignition behavior. Schnizlein and Fisher[17] have reported similar curves for plutonium. Ignition temperatures have been reported ranging from 110 C[16] for a delta phase swarf in air with an area/weight ratio of 13.3 cm^2/g to 820 C[21] for a 30% plutonium, 6% zirconium, 64% uranium tertiary alloy in oxygen.

As has already been noted, plutonium oxidizes exothermally (the reaction between plutonium and oxygen has a heat of reaction of 4.2 Btu/g plutonium). The amount of heat generated is a function of the area available for oxidation, the rate at which oxygen diffuses to the oxide-metal boundary, the thermal conductivity of the oxide layer, and the heat capacity of the system; because of this, both the gas temperature surrounding the specimen and the area/weight ratio of the specimen influence ignition. Schnizlein and Fisher[17] have divided ignition behavior into two regimes. Metallic pieces with an area/weight ratio of >5 cm^2/g ignite at approximately 300 C. Ignition temperatures of approximately 500 C are found for specimens with an area/weight ratio of <1 cm^2/g. Discrepancies in ignition behavior of foils with area/weight ratios of 5.3 cm^2/g are attributed to impurities or delta phase stabilization during heat treatment. Other deviations are cited in later experiments but were not explained. Local experience has shown that ignition of 2 kg buttons can occur at approximately 300 C. Stewart[16] also reports ignition of a metal rod with an area/weight ratio of 0.31 cm^2/g at 350 C and a delta phase alloy with an area/weight ratio of 0.84 cm^2/g at 340 C. Therefore, it appears temperature and area/weight ratios alone do not unequivocally define ignition temperatures.

Thus, the gas temperature at which the reaction rate jumps from a lower to a higher regime depends upon the equilibrium between heat generated (internal and external) and heat dissipated. If the heat can be

removed at a sufficient rate, ignition does not occur. It is the experience in production areas where scraps of metallic plutonium are intentionally ignited prior to storage that, even after ignition has taken place, the reaction can be terminated if the oxidizing piece is brought into contact with an object, such as a metal spatula that can conduct away the heat. If the reaction rate is limited by an adherent rate-controlling film beneath the thermal conductivity capacity of the oxide film, ignition does not occur. This type of mechanism explains the ability of some metal specimens to oxidize completely without ignition in one set of conditions and ignite under others.[21] It could also explain the similarity in oxidation rates in air and oxygen reported by Schnizlein and Fisher[24] and the dissimilar results reported by Carter, Foy, and Stewart.[26]

PLUTONIUM AIRBORNE RELEASE

The only article to date that comprehensively discusses plutonium aerosol release is K. Stewart's chapter in Vol. 5 of "Progress in Nuclear Energy," Series IV.[16] In this chapter he covers plutonium oxidation rates, particle size distribution, and release rates in roughly four temperature categories.

Stewart finds the particle size distribution of the plutonium dioxide produced depends on the temperature of oxidation, the crystalline form of the metal and the humidity in the atmosphere. At room temperature in dry air, the pure metal yields particles with a median mass diameter (MMD) of 0.5 to 2 μ. As the temperature and moisture content increase, the particles produced become coarser and, due to the increased oxidation rate, the release rate increases. The delta alloys produce material with a particle size distribution similar to that of the pure metal at room temperatures, but due to the lower oxidation rate, its release rate is an order of magnitude lower. The rate of release of the pure metal at room temperature ranges from 1.5×10^{-7} μCi/cm^2-hr in dry air to 5×10^{-3} μCi/cm^2-hr at 100% relative humidity. The delta alloys over the same range have rates from 5×10^{-9} μCi/cm^2-hr to 4.5×10^{-4} μCi/cm^2-hr.

At temperatures above the alpha-beta transformation, the behavior of the metal and delta alloy are significantly different. In this temperature

range, the release of oxide from pure metal to the air was small and continuous. The particle size distribution of the oxide produced was similar to that found at room temperature. The rate of release varied from 9×10^{-4} to about 6×10^{-3} $\mu Ci/cm^2$-hr, and was independent of temperature and humidity. It was noted that the oxide produced was friable and readily fragmented into particles <3 μ in diameter.

The material released from oxidation of the delta stabilized alloy was coarser having a MMD of 10 μ. The release of oxide was sporadic and corresponded to the stepwise oxidation of this material indicative of the successive cracking of the oxide layer. The rate of release ranged from 1×10^{-2} $\mu Ci/cm^2$-hr (an order of magnitude greater than the pure metal) to 9×10^{-5} $\mu Ci/cm^2$-hr (an order of magnitude lower than the pure metal) with increasing humidity.

Above the beta-gamma phase transformation, the particles produced are hard and amorphous with a MMD of 350 to 1000 μ. Although the amount of oxide produced increases greatly, the percentage of oxide in the respirable size range is much lower than that produced at lower temperatures.

Stewart detected the existence of two groups of particles based on their geometric standard deviations (σ) which Kottler has shown to be related to the rate constant of the formation processes in a log-normal distribution. The basic particles separated from the basic mass due to internal stress had a σ in the region of 2 to 4 while particles formed during breakaway by attrition, etc., had a σ of 10 to 15.

The particle size distribution of the oxide formed from the pure metal and delta alloy in the four temperature ranges mentioned fell into four approximate size distributions. The effects of gas velocity upon release rates were divided into two groups: "static", which involves no physical disturbance and a velocity of <1 m/sec; and "dynamic", which assumes complete dispersal of all oxide produced. Stewart correlated the particle size distribution and release rate found for the metal and delta alloy under the stated conditions of temperature and gas velocities

with a lung retention model, and arrived at an "effective source strength."
The downwind inhalation hazard from any of the stated conditions can then
be calculated by use of one of the existing transport formulae. To calculate
the potential lung deposition in an individual exposed to the aerosol through-
out the time of emission, Stewart uses the formula:

$$\text{Dose } (\mu Ci) = (ESS)(A)(R)[f(W)]$$

where (ESS) is the effective source strength, A is the area of the metal
oxidized, R is the breathing rate and f(W) is the aerosol dispersion function
according to Sutton or Pasquill.

British workers have conducted a large scale, outdoor experiment
to determine plutonium dispersion during a fire.[33] In these tests, 200-g
rods of plutonium were suspended in a 4 ft square chimney, 11 ft high. The
rods were heated by a gasoline fire at 850 C for 30 min and by slow, pro-
grammed increases to 600 C in 1 hr. The PuO_2 release was measured by
weight loss, integration of the deposition curve and integration of the down-
wind air concentration. Weight loss gave the highest release rate but diffi-
culties in full recovery of the residue casts doubts on the validity of the
results. All release rates obtained from the lower temperature oxidation
were higher. In reporting the data Stewart concludes that:
- No significant inhalation hazard would exist at greater than 200 yards
  from burning several kilograms of plutonium.
- A release value of 0.05% is a satisfactory, safe value for estimating
  the airborne hazard downwind.

Other corroborative information is obtained from accident reports
involving plutonium. A recent incident at Hanford[34] involved a fire in a
plutonium purification area. Some explosive mechanism ruptured an ion
exchange column and ignited other materials in the vicinity. The building
filters remained intact and only 225 μCi were released via the stack.
Although smoke was observed leaking from the stairwell structure and one
door was blown open, no significant contamination was detected on the ground
beyond 20 yards. The presence of large amounts of carbon and organic vapors

in the smoke may have played a significant role in limiting the spread of contamination and increasing the deposition of airborne plutonium dioxide particles. Air monitors at the perimeter of the area recorded only slightly higher than detectable levels.

Earlier incidents occurring at Oak Ridge National Laboratory and Rocky Flats Plant[35] also indicated low airborne release of plutonium from fires and explosion even with loss of building filters. Table I summarizes the data found in literature on the behavior of plutonium at various temperatures.

DEPOSITION

Very little has been written on the deposition of plutonium or its oxide. Stewart[16] lists deposition velocities* of 7.3 cm/sec for a size distribution with a MMD of 0.3 μ, to 3.2 cm/sec for a size distribution with MMD of 16 and 29 μ for the particles airborne at or above the ignition temperature. The unusually high deposition velocity noted for the 0.3 μ particles is for the material dispersed into the glove box from the violent reaction between oxygen and a thin metallic disk and is likely not representative of normally formed particles of this size. Cheevers[37] in his exploding wire experiments on plutonium has reported only 0.00055% of the resultant oxide reaches the filter at a gas velocity of 5 fpm. In outdoor tests with large plutonium rods at 600 to 800 C, most of the airborne particles settled out within 200 yards of the release point.[38]

The deposition of particles during a fire is a complex process. The thermal updrafts created by the heat would counteract the gravitational effects near the fire. Diffusion due to temperature and concentration gradients cause some deposition of submicron particles near the source while turbulent deposition and impaction would also take place. These latter effects are especially important for particles larger than about 2 to 5 μ.

Away from the source of heat, particles will be carried and dispersed with the air currents and deposit due to settling, impingement, and turbulent impaction with objects. Friedlander and Johnstone[38] hypothesize

* The deposition velocity is defined[36] as the number of particles deposited per cm^2/sec divided by the number of particles per cm^3 in the gas above the surface.

TABLE I

SUMMARY OF OBSERVATIONS OF PLUTONIUM BEHAVIOR AT VARIOUS TEMPERATURES

Temperature	Property, Behavior	Values for Relatively Pure Metal	Values for Stabilized δ Phase (Binary)
To 120 C (Pure metal in α phase)	Oxidation Rate (mg PuO_2/cm^2-hr)	0.009 to 10	Not detectable to ignition[*]
	Particle Sizes	85% <6 μ	85% <6 μ
	Release Rate (μCi/cm^2-hr) (at about 14 to 24 cm/sec) Dry Air 100% Relative Humidity Air	~1.5 x 10^{-7} ~5 x 10^{-4}	~5 x 10^{-9} ~5 x 10^{-5}
	Range of Release Rate as a Function of Velocity over Specimen: Low Flow: <1 m/sec High Flow: Sufficient to make airborne all oxide particles	Low: 0.0005 to High: 5	Low: 5 x 10^{-5} to High: 0.5
120 to 210 C (Pure metal in β phase)	Oxidation Rate (mg PuO_2/cm^2-hr)	0.2 to 9	0.5 to ignition[**]
	Particle Sizes	85% <6 μ	85% <6 μ to 5% <6 μ
	Release Rate (μCi/cm^2-hr) (in ambient air)	0.005	0.0005
	Range of Release Rate as a Function of Velocity over Specimen: Low Flow: <1 m/sec High Flow: Sufficient to make airborne all oxide particles	Low: 0.005 to High: 5000	Low: 0.0005 to High: 50
210 to 315 C (Pure metal in γ phase)	Oxidation Rate (mg PuO_2/cm^2-hr)	4.5 to ignition[†]	0.35 to ignition[††]
	Particle Sizes	Not available	Not available
	Release Rate	Not available	Not available
	Range of Release Rate, etc.	Not available	Not available
315 to 452 C (Pure metal in δ phase)	Oxidation Rate (mg PuO_2/cm^2-hr)	11 to ignition[§]	4 to ignition[§§]
	Particle Sizes	85% <6 μ to 3% <6 μ	85% <6 μ to 3% <6 μ
	Release Rate (effect of humidity)	Difference in rates not measurable	
	Range of Release Rate as a Function of Velocity over Specimen: Low Flow: <1 m/sec High Flow: Sufficient to make airborne all oxide particles	Low: 10 to High: 40,000	Low: 10 to High: 40,000
At Ignition	Oxidation Rate (mg PuO_2/cm^2-hr)	Highly variable	Highly variable
	Particle Sizes	MMD 350 to 1000 μ ~0.2% <6 μ	MMD 350 to 1000 μ ~0.2% <6 μ
	Integrated Release for 100% Conversion to Oxide	0.02% to 100% (Highly variable, depending upon mass and geometry of specimen)	0.02% to 100%

[*] δ metal turnings: area/weight = 13.3 cm^2/g
[†] 2 kg metal buttons
[§] Metal rod: area/weight = 0.31 cm^2/g

[**] δ metal turnings: area/weight = 8.7 to 13.8 cm^2/g
[††] δ metal rod: area/weight = 1.4 cm^2/g
[§§] δ metal rod: area/weight = 0.84 cm^2/g

that particles in a turbulent stream are projected through the last portion
of the boundary layer near a surface by the transverse eddy velocities in
the turbulent layer. The thickness of the boundary layer is related to
the stopping distance, S, of the particle. Mass transfer rates can be
deduced with the assumption that particles need to diffuse to within one
stopping distance of the surface. For particles in the 1 to 10 μ size range
and at low gas velocities, deposition varies with the square of the terminal
velocity.

Chamberlain[39] found that a roughness element extending 10 to
100 μ into the laminar sublayer may be significant in the deposition of
small particles. In his work with dry deposition on grass, he deduces a
minimum in deposition velocities for particles with a diameter of 0.1 μ.
He proposes that for particles of high density and >10 μ in diameter, the
deposition velocity is equal to the terminal velocity. For submicron parti-
cles, deposition velocities near 0.1 to 0.3 cm/sec are indicated.

Tanner[40] has calculated the disproportionate disappearance of
particles from an aerosol containing a spectrum of particle sizes due to
gravitational settling. For a turbulent aerosol (one having random eddies
of velocities less than 1 mile/h) at room temperature, the ratio of the
number remaining suspended to the initial number is:

$$-\ln\left(\frac{n_t}{n_o}\right) = \frac{(\rho_p - \rho_g)gd^2Ct}{18\eta h} \tag{1}$$

where n_t = number of particles at time t

n_o = number of particles at time zero

ρ_p = density of particles (g/cm^3)

ρ_g = density of gas (g/cm^3)

g = acceleration due to gravity (cm/sec^2)

d = diameter of particle (cm)

t = time (sec)

η = viscosity of gas (poise, g/sec-cm)

h = height of chamber (cm)

C = Cunningham's correction factor (dimensionless).

Table II shows the fraction of various size PuO_2 spherical particles (density 11.46) remaining airborne in a 10 ft high chamber after release. Under these conditions greater than 99.9% of all particles aerodynamically equivalent to a sphere of PuO_2 larger than 10 μ would have settled out within 10 min. Within approximately 1 hr, greater than 99.9% of all particles larger than 4 μ would have settled out. More than 10 hr would be required to reduce the airborne concentration of 1 μ particles to this level.

TABLE II

GRAVITATIONAL SETTLING OF PARTICLES

Time (in minutes) to reduce airborne PuO_2 concentration to a given fraction
of the original concentration in a 10 ft tall enclosure [as predicted by Equation (1)]

Diam., μ:	1	2	3	4	5	6	7	8	9	10	12	15	20
η/n_o													
0.0	13.7	3.7	1.7	0.95	0.62	0.43	0.32	0.24	0.19	0.16	0.11	0.07	0.04
0.8	29.1	7.8	3.56	2.03	1.31	0.92	0.68	0.52	0.42	0.33	0.23	0.15	0.08
0.7	46.5	12.5	5.69	3.24	2.10	1.46	1.08	0.83	0.65	0.53	0.37	0.24	0.13
0.6	66.6	17.9	8.16	4.65	2.98	2.10	1.54	1.17	0.94	0.76	0.53	0.34	0.19
0.5	90.3	24.3	11.1	6.30	4.07	2.82	2.10	1.61	1.27	1.03	0.72	0.46	0.26
0.4	119.4	32.1	14.6	8.32	5.37	3.75	2.77	2.13	1.68	1.37	0.95	0.61	0.37
0.368	130.3	35.1	16.0	9.09	5.85	4.09	3.00	2.32	1.84	1.49	1.04	0.66	0.34
0.3	156.9	42.2	19.2	11.0	7.07	4.92	2.77	2.79	2.21	1.79	1.25	0.80	0.45
0.2	209.8	56.5	25.7	14.7	9.44	6.59	4.87	3.72	2.95	2.40	1.67	1.07	0.60
0.1	300.1	80.8	36.8	21.0	13.5	9.44	6.97	5.34	4.23	3.43	2.39	1.53	0.86
0.01	600.3	161.6	73.6	41.9	27.0	18.9	13.9	10.7	8.46	6.86	4.76	3.06	1.73
0.001	900.4	242.4	110.4	62.9	40.6	28.3	20.9	16.0	12.7	10.3	7.15	4.60	2.59

Outdoors, many other factors such as meterological and geographical factors would influence transport and deposition. Stewart[41] reports that half the fine airborne particulate material from Operation Plumbob settled out in approximately 37 days. This is indicative of the small size of the particles remaining airborne.

RESUSPENSION

After the plutonium particulate material is deposited, the principal hazard is inhalation of resuspended material. The amount resuspended, under stated conditions, is generally given as a resuspension factor:

$$K \ (m^{-1}) = \frac{\text{Airborne concentration (units/m}^3)}{\text{Surface contamination (units/m}^2)}$$

Table III is a compilation of resuspension factors that have been reported in the literature.

TABLE III

RESUSPENSION FACTORS FOR AIRBORNE RADIOACTIVE AEROSOLS

Conditions of Resuspension	Resuspension Factor	Reference
Average Resuspension Factor in Accidents Involving Plutonium	$4 \times 10^{-6} \text{ m}^{-1}$	14
Vehicular Traffic (Nevada)	7×10^{-5}	13
People Working or Active in a Closed Area	4×10^{-5}	
Dirty Rural, Suburban, or Metropolitan Areas	7×10^{-6}	
People Working or Active in an Open Area	2×10^{-6}	
Isolated Area	$\sim 7 \times 10^{-7}$	
Resuspension of Aged Plutonium Deposit (0.74 to 752 μCi/m^2) from "Plumbob"	6.2×10^{-10} to 10^{-13}	5
Plutonium Oxide, No Movement	2×10^{-8}	46
Plutonium Oxide, 14 steps/min	10^{-5}	
Plutonium Oxide, 36 steps/min	5×10^{-5}	
Plutonium Nitrate, No Movement	2×10^{-8}	
Plutonium Nitrate, 14 steps/min	10^{-6}	
Plutonium Nitrate, 36 steps/min	5×10^{-6}	
Plutonium Oxide, Change Room (>3000 ft^2), 9 air changes/hr, 0.01 μCi/m^2, 4 to 6 persons active in area:		47
"Loose" Contamination (estimated by smears)	10^{-3}	
"Loose" Contamination (estimated by water-detergent wash)	2×10^{-4}	
Changing Coveralls, Static Sampler, No Ventilation	2.8×10^{-3}	
Changing Coveralls, Personal Sampler, No Ventilation	6.4×10^{-3}	
Personnel Traffic in a Small Unventilated Room	4×10^{-3}	
Proposed Resuspension Factors for Plutonium Oxide:		39
Outdoors (quiescent conditions)	10^{-6}	
Outdoors (moderate activity)	10^{-5}	
Indoors (quiescent conditions)	10^{-6}	
Indoors (moderate activity)	10^{-4} to 10^{-5}	
From Crater of Tower Shot, No Artificial Disturbance	8×10^{-8}*	39
Survey of Road, No Artificial Disturbance	8×10^{-8}*	
Survey of Road, Landrover, D-Day + 4	1.4×10^{-5}	
Survey of Road, Landrover, D-Day + 7	1.5×10^{-6}	
Survey of Road, Tailboard of Landrover, D-Day + 7	2×10^{-5}	
Survey of Road, D-Day + 1 and 2	4×10^{-7}	
Sample Collection in Cab of Landrover, H-Hour + 5	6.4×10^{-5}**	
Sample Collection in Cab of Landrover, H-Hour + 8	2.5×10^{-5}†	
Uranium Sample Downwind of Crater, Sample Height: 1 ft Above Ground	3×10^{-4}††	
Uranium Sample Downwind of Crater, Dust Stirred Up, Sample Height: 1 ft	10^{-3}††	
Uranium Sample Downwind of Crater, Sample Height: 2 ft	10^{-5}††	
Plutonium Sampled 1 ft Above Ground:		
Vehicular Dust	5×10^{-4}§	
Pedestrian Dust	1.5×10^{-6}§	
Iodine-131, Enclosed (Chamberlain & Stanbury)	2×10^{-4} to 4×10^{-5}	
Iodine-131, Open (Chamberlain & Stanbury)	2×10^{-6}	
Yttrium-91, 0-8 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:		
Ground Contamination Level 1.8 μCi/m^2	1.8×10^{-7}	
Ground Contamination Level 8.8 μCi/m^2	10^{-8}	
Ground Contamination Level 24.6 μCi/m^2	3×10^{-10}	
Polonium-210, 0-8 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:		
Ground Contamination Level 0.6 μCi/m^2	2×10^{-6}	
Ground Contamination Level 3 μCi/m^2	2×10^{-6}	
U_3O_8, 0-4 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:		
Ground Contamination Level 112 g/m^2	2.4×10^{-6}	
Grass Contamination Level 70 g/m^2	8×10^{-6}	
Concrete Contamination Level 180 g/m^2	2×10^{-6}	
Plutonium Oxide, Sampling Height: 5 ft		40
Floor Level 0.1 μCi/m^2, No Circulation	1.6×10^{-6}	
Floor Level 24.6 μCi/m^2, No Circulation	4.4×10^{-7}	
Floor Level 0.1 μCi/m^2, Fan	1.3×10^{-3}	
Floor Level 0.91 μCi/m^2, Fan	1.4×10^{-3}	
Floor Level 0.086 μCi/m^2, Fan and Dolly	1.0×10^{-2}	
Floor Level 1.3 μCi/m^2, Fan and Dolly	9.4×10^{-3}	
Floor Level 1.3 μCi/m^2, After Test	9×10^{-4}	
Floor Level 1.1 μCi/m^2, After Test	9×10^{-4}	
Uranium, Sampling Height: 5 ft		
Floor Level 0.086 μCi/m^2, No Circulation	1.8×10^{-6}	
Floor Level 0.95 μCi/m^2, No Circulation	2.2×10^{-7}	
Floor Level 0.095 μCi/m^2, Fan	1.5×10^{-4}	
Floor Level 1.1 μCi/m^2, Fan	1.1×10^{-4}	
Floor Level 0.11 μCi/m^2, Dolly	1.6×10^{-4}	
Floor Level 1.3 μCi/m^2, Dolly	1.3×10^{-4}	
Floor Level 0.91 μCi/m^2, Fan and Dolly	4.6×10^{-4}	
Floor Level 1.0 μCi/m^2, Fan and Dolly	1.9×10^{-4}	

* One high value excluded   ** Only ~10% of particles <6 μ
† Only ~20% of particles <6 μ †† Only ~5% of particles <6 μ
§ Particles primarily in 20 to 60 μ size range, <1% <6 μ

The various factors listed show considerable variations ranging from 1×10^{-2} [42] (resuspension of a finely divided material from a newly painted concrete floor due to air and mechanical motion) to 10^{-13} m^{-1} [5] (resuspension of aged plutonium particulate material from desert soil by natural turbulence).

Values ranging from 7×10^{-7} to 7×10^{-5} m^{-1} were calculated from data obtained during Operation Plumbob. [43] The U.S. Navy suggests a factor of 4×10^{-6} for use in weapons accidents calculations, [44] which agree agrees well with Stewart's [41] proposed value of 1×10^{-6} m^{-1} derived from resuspension data obtained from many sources. In his paper presented to the International Symposium on Surface Contamination, Stewart outlines some of the significant mechanisms and factors influencing resuspension of particulate material.

From available information, this author concludes that after particles are deposited on the ground, they tend to remain fixed in place due to mixing into the top layer of loose soil (the process of weathering). This is illustrated well by the data from Operation Plumbob trials [45] which show that greater than 90% of the plutonium deposited remained fixed in the soil. Olafsen and Larson [5] returning to the original Plumbob site found resuspension factors of 10^{-10} to 10^{-13} m^{-1} from this aged material.

The boundaries of the contaminated area tend to increase somewhat with time due to the wind erosion processes of saltation, suspension, and surface creep but losses from the area are small since only a small fraction is of the size range to become airborne. In the presence of greater turbulence, a larger proportion of small particles, or increased physical activity in the area, more material is resuspended.

In the event of an accidental release, plutonium dioxide deposited would tend to remain fixed. The amount resuspended would depend upon surface, turbulence, particle size distribution of the particulate material and activity within the area. In a controlled area, the resuspension factor would be in the range of 10^{-5} to 10^{-6} m^{-1} and the amount airborne would tend to depend upon the conditions and contamination level of the area upwind.

Some extension of the contaminated area with time is to be expected.
Resuspension rates will decrease with time due to "weathering" of the
contamination and the depletion of fine material.

Corn's[48] assertion that the most probable mechanism for redis-
persion of particles from a surface under normal conditions is a transfer
of momentum from large moving objects appears to be borne out by the
data presented to date.[41-43, 46, 47]

CONCLUSIONS

Since the conditions existing during any individual fire are almost
unique for the circumstances existing at that moment in time, it is only
possible to discuss plutonium aerosol release from fires in general terms.

Fires involving plutonium compounds in air generally result in the
formation of plutonium dioxide. The particle size distribution and release
rate of the dioxide would vary with prior treatment and fire conditions.
If the material is a well-dried, free-flowing oxalate, the dioxide produced
would be in the form of a finely divided particle, readily dispersed and
primarily in the size range of greatest inhalation hazard. If the same
material were in a wet cake or slurry and heated at a rapid rate, the amount
released would be significantly reduced.

In fires involving metallic plutonium, physical shape, prior treat-
ment, and fire conditions play significant roles. Generally below ignition,
the greater the surface area, temperature, and gas velocities, the greater
is the release. For instance, the "effective source strength" at 400 to
500 C is 0.02 $\mu Ci/cm^2$-min under static conditions and 2 $\mu Ci/cm^2$-min for
dynamic conditions.[16] At temperatures above 100 C, the equilibrium
between heat dissipated and heat input (internal and external) can result
in the ignition of the plutonium. Above ignition, the amount of material
released depends on the total weight of plutonium available and gas veloci-
ties. The "effective source strength" at and above ignition temperatures
under static conditions is 2000 $\mu Ci/kg$ and 10,000 $\mu Ci/kg$ under dynamic
conditions.[16]

Airborne aerosols in nominal plutonium fires and routine operations are controlled by high efficiency filters at each work zone with back-up filters just prior to release. In serious fires and explosions in which these barriers may be by-passed and breached, it becomes necessary to use the data outlined above to predict the consequences of the incident. Additional research is indicated to better define deposition in ducts and on surfaces, and to establish realistic release factors for typical fires.

In the event of fires in the open, even if several kilograms of plutonium were in a fire, no significant inhalation hazard will likely exist beyond several hundred yards downwind. Once deposited, plutonium dioxide tends to "weather into" the top layer of soil and become fixed at the location. Only large disturbances such as vehicular traffic, unrestricted movement of personnel, wind storms or displacement of the upper layers of soil would move the deposited material any significant distance.

In general, knowledge of the trends listed above is required in understanding the mechanisms involved in the formation and release of a plutonium dioxide aerosol, the amount of material released in quiescent conditions, the amount potentially available to be released under these conditions and the behavior of the aerosol once it has been deposited. The information available is not sufficient to make a quantitative evaluation of the amount of plutonium dioxide released during any given fire situation.

No data are given on the increase or reduction of plutonium dioxide particulate material in the respirable range from variations in fire conditions. The effects of variations in gas velocity upon the particle size distribution and amount released, and the effects of other materials such as organics or alloying metals, upon the amount released are not available. Most experimenters have been concerned with metallic plutonium. Rags and paper wipes and other debris which have been used to absorb soluble plutonium salts may yield aerosols of unique properties, which should be identified.

The penetration of high efficiency filters by particles released in plutonium overheating incidents should be measured for some incidents to

gain assurance that particles will be effectively retained. Although the
filter media are very reliable, a demonstration of high efficiency for the
airborne material in a plutonium incident would be of value.

Correlations between existing oxidation rate values and release
rates could not be made due to the variations in experimental conditions.
Since all the release values available are under one set of experimental
conditions, additional data and corroboration of existing values is
desirable.

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