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FINAL REPORT

on

**RECOMMENDED PLUTONIUM RELEASE
FRACTIONS FROM POSTULATED FIRES**

for

**EG&G Rocky Flats, Inc
Rocky Flats Plant**

December 1993

Prepared by

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EXECUTIVE SUMMARY

This report was written at the request of EG&G Rocky Flats, Inc. in support of joint emergency planning for the Rocky Flats Plant (RFP) by EG&G and the State of Colorado. The intent of the report is to provide the State of Colorado with an independent assessment of any respirable plutonium releases that might occur in the event of a severe fire at the plant. Fire releases of plutonium are of interest because they have been used by EG&G to determine the RFP emergency planning zones. These zones are based on the maximum credible accident (MCA) described in the RFP Final Environmental Impact Statement (FEIS) of 1980. That MCA is assumed to be a large airplane crashing into a RFP plutonium building.

The objective of this report was first, to perform a worldwide literature review of relevant release experiments from 1960 to the present and to summarize those findings, and second, to provide recommendations for application of the experimental data to fire release analyses at Rocky Flats. The latter step requires translation between experimental and expected RFP accident parameters, or "scaling". The parameters of particular concern are: quantities of material, environmental parameters such as the intensity of a fire, and the physico-chemical forms of the plutonium. The latter include plutonium metal, bulk plutonium oxide powder, combustible and noncombustible wastes contaminated with plutonium oxide powder, and residues from plutonium extraction processes.

Scaling requires an understanding of the functional dependencies of plutonium releases on system parameters. Where enough experimental data exist, these dependencies can be expressed as correlations, and these correlations can, in turn, be used to interpolate and extrapolate to expected accident conditions. Often, the data are insufficient to allow such a procedure, and bounding judgement must be used. In any case, it is important to define the expected accident scenario in sufficient detail to allow application of any correlations and to limit the conservatism of plutonium release estimates. For example, fire release correlations that depend on convection velocity are useless unless that velocity is predicted, and so on.

The development of accident scenarios for the Rocky Flats Plant was not part of the scope of the work reported here. Nor have such scenarios been available to us in enough detail to permit recommendation of single value plutonium releases. In the absence of such detailed scenarios, the recommended releases in this report are restricted to the form of correlations and prescriptions for the analyst, for use once such scenario descriptions are, in fact, available.

It is customary to express plutonium aerosol releases in terms of the airborne release fraction (ARF), which is defined as the total fraction of the affected plutonium that is released in the accident, or the airborne release rate (ARR), which is the average rate at which the ARF is released. The respirable fraction (RF) of the mass of plutonium released in the accident is conditionally assumed to include particles having aerodynamic diameters smaller than 10 μm . The aerodynamic diameter of a particle is defined as the diameter of a unit-density sphere having the same settling velocity as the particle under consideration.

The work presented in this report was intended to complement currently available material, to improve current knowledge in this technical area by providing an independent critical analysis of all available literature sources, and to provide justifiable recommendations on plutonium release parameters using a comparative analysis of the main findings presented in the literature. To meet the objectives of this project, this study was conducted in several stages, including a literature search, information review and classification, data base analysis, and development of recommendations on plutonium release parameters.

Open literature and technical reports covering airborne releases of plutonium and plutonium-simulating contaminants during combustion of radioactive metals themselves and during fires in processing and waste storage facilities were reviewed. Relevant material published after 1960 was identified through a computerized literature search using the NTIS and DOE Energy Data Base, as well as numerous technical references and several private communications.

Examination of the literature has identified three major sources of plutonium release during a severe accident at nuclear facilities:

- 1) Release by oxidation of plutonium metal,
- 2) Release by combustion of contaminated solid waste materials stored in the facility, and
- 3) Release by combustion of contaminated flammable process liquids, or evaporation of contaminated non-flammable liquids.

Our recommendations for these three sources are presented below.

Based on the information gathered in this study, airborne releases from oxidation of metallic plutonium were grouped into three general categories. These categories are:

- plutonium oxidation at a range of temperatures not exceeding the plutonium ignition point,

- exothermic combustion of plutonium leading to its melting, and
- an explosive-like dissemination of plutonium resulting in formation of airborne plutonium particles and their subsequent violent combustion.

The main results obtained for these mechanisms of plutonium release are summarized in Table I.

Table I. Plutonium Oxidation Release Parameters

Pu release conditions		Oxidation below melting point		Combustion			
				$T^{\circ}_{\text{melt}} < T^{\circ} < T^{\circ}_{\text{boil}}$		$T^{\circ} > T^{\circ}_{\text{boil}}$	
		ARR $\mu\text{Ci}/\text{cm}^2\text{hr}$	RF*	ARF (of total)	RF	ARF (of total)	RF
Static**	Pure Pu	Eq.A	Eq.C	2.e-4	0.5	---	---
	δ -Pu alloy	Eq.B	Eq.C				
Dynamic***		---	---	1.0	1.e-4	1.0	0.5

* RF is given as a fraction ($d_p < 10 \mu\text{m}$) of airborne mass

** Static conditions of plutonium release are those limited to low-velocity air flows and no external mechanical stresses to assist dispersion of the contaminant particles.

*** A complete dispersion of plutonium (ARF = 1.0) is assumed for the dynamic conditions of release, such as burning plutonium metal in a high air flow resulting in the aerosolization of the oxide formed, or bursting molten plutonium under boiling conditions or explosions.

ARR:

Pure plutonium

$$\log (ARR) = (-4.36 + 0.071 AH) \left(0.23 - \frac{350.6}{T^{\circ}K} + \frac{1.91e5}{(T^{\circ}K)^2} \right) \quad (A)$$

δ -Pu alloy

$$\log (ARR) = (-7.47 + 0.101 AH) \left(0.60 - \frac{661.3}{T^{\circ}K} + \frac{2.16e5}{(T^{\circ}K)^2} \right) \quad (B)$$

where AH = absolute humidity (mg H₂O/l air)

RF:

$$RF = 1.07 - 0.00353 \cdot T^{\circ}C + 3.82 \cdot 10^{-6} \cdot (T^{\circ}C)^2 \quad (C)$$

Combustion of contaminated solid waste materials appears to present a significant contamination hazard, because of the quantity of contaminated wastes stored in nuclear facilities, and because of the large fraction of contamination that can be released during a solid waste fire. Experiments have shown that up to 50 percent of the radioactive contamination can become airborne during combustion of a typical waste material if the material is burning in the convective plume of a fire. A large fire may also cause bursting of waste storage drums exposed to direct flames.

Airborne release parameters are presented in this report for the following contaminated combustible solid materials involved in an accidental fire:

- Rubber (polychloroprene or latex)
- Polystyrene resin
- Polymethylmethacrylate

- Paper (cellulose) and mixed waste
- Mixed waste in 55-gal drums

The recommended plutonium aerosol release parameters are shown in Table II.

Table II. Plutonium Release During Combustion of Contaminated Solid Materials

Combustible Material	ARF		RF
	Dry Powder	Solution or Salt	
Rubber	0.010	0.040	1.0
Polystyrene	NA	0.008	1.0
Polymethylmethacrylate (PMMA)	0.050	0.020	1.0
Cellulose and mixed waste materials (u = air velocity [m/s])	$0.2754 \cdot 3.254^{\ln(u)}$ (max 0.5)	$0.01202 \cdot 2.075^{\ln(u)}$ (max 0.5)	1.0

For the plastic materials, such as PMMA, polychloroprene, and polystyrene, which produce bubbles and become soft before they ignite, the contamination ARF under static burning conditions (low air flow) are higher than releases from cellulose burning under similar conditions. This is probably related to the bursting of bubbles at the surface of plastic materials. When the contaminated material is located in the convective plume of a fire, approximately half of the contamination on the combustible material is transported by the plume.

Release from burning cellulose is largely controlled by the flow of gases around contamination particles. Two different mechanisms of air flow development caused by the fire were considered: evolution of combustible gases by pyrolysis, and development of updraft air flow by natural convection. Computational procedures needed to evaluate flow velocities are suggested in this work. These procedures can be used to estimate an ARF for different scales of a fire. However, this methodology should be validated.

Releases derived for cellulose combustion are also recommended for the prediction of an ARF for mixed wastes.

Based on the very limited information available, it is recommended that unlined 55-gal drums containing combustible material should be considered to have released all of their contents if drums are exposed to flames from a large fire. Unlined drums not exposed to direct flames will maintain their seals. The evidence suggests that lined drums may maintain their seals. However, no thermodynamic considerations involving the temperature and pressure evolution in the drums exposed to the fire were found in the literature.

All releases from burning contaminated solid materials are conservatively assumed to be in the respirable size range. However, since plutonium oxide contaminant is a refractory material unaffected by fire, its original RF, if known, should be used.

Potential releases from flammable liquids are less significant, because of the lower releases expected, and because work is currently in progress at Rocky Flats to convert liquid wastes into solid form. Nevertheless, the risk of contaminant releases associated with the combustion of kerosene-like flammable liquids, such as during a jet-fuel fire caused by a crash of an airplane, will continue to be of significant concern. Tables III and IV summarize recommendations developed in this work for plutonium releases from noncombustible and combustible liquids, respectively.

Table III. Plutonium Release From Heating or Boiling Contaminated Liquids and Liquid Residues

Heating Conditions	ARF	RF
Simmering (Quiescent surface)	2.0×10^{-4}	0.5 ^(a)
Boiling (Disturbed surface)	Equation (D) (max 0.02)	0.5 ^(a)
Heating dried liquid residue	7.0×10^{-4}	1.0×10^{-5}

Note: (a) This RF value is estimated based on a single and unsubstantiated result given in Mishima et al. (1968-b)

$$\log(ARF) = -13.38 + 22070 \cdot B - 1.043 \cdot 10^7 \cdot B^2 \quad (D)$$

where B = boil-off rate (fraction/cm²/min).

Table IV. Plutonium Release From Combustion of Contaminated Organic Liquids

Contaminated material	ARF	ARR	RF
TBP-kerosene (Dissolved contaminant)	0.10	0.001/minute	1.0
Kerosene (Powder contaminant)	0.02	0.001/minute	1.0 ^(a)

Note: (a) Use the RF of the contaminant powder, if known.

For extreme conditions, some of the above cited ARFs have large maximum values. Because of this, it is necessary to stress that the potential for an unexpectedly high release fraction suggests that much more attention needs to be paid to postulating accident scenarios, and to the material inventory involved in the fire. If the release fractions expected from any accident were low, such a careful analysis of the progression of the accident would be less important.

Also, with the potential for higher release fractions, uncertainties associated with any release parameter become extremely important. As shown in this work, major uncertainties are associated with the considerable amount of scatter among the reported ARFs and RFs. Much of this scatter is due to the experimenters' desire to investigate a variety of test conditions in a limited-scope study. These variables include use of plutonium or a simulant, chemical form of the contaminant, its size distribution, its amount and application procedure, type of combustible material, flow conditions, and many others. Even when experimenters took great care to achieve reproducible fire conditions, order of magnitude variations in contaminant release were observed.

The physical arrangement of the contamination and combustible material can also affect the observed ARF. Contamination on the underside of combustible material is released at a lower rate than contamination on top. This is consistent with the observed effect of gas velocity on contaminant release. In an actual accident, many contaminant-fuel arrangements are expected to occur. Some of these arrangements will reduce the amount of airborne contaminant, while other arrangements will cause increased contaminant release.

The behavior of waste materials stored in 55-gallon drums is very uncertain. Both the location of a drum during an accident and the presence of a liner inside the drum can affect the release expected for that drum. Drums that remain sealed are expected to release negligible contamination,

but a drum that explodes has a potential for releasing half of the contained contamination if its content is blasted into the flames.

The health effects of the released contamination are strongly dependent on the size distribution and composition of the aerosol. Respirable contaminated particles present the greatest hazard to the surrounding population. Aerosol generated from solutions tends to be mostly respirable, and a conservative estimate requires a relatively large respirable fraction for this contamination form. In many experiments involving combustible materials, the mass distribution of the overall aerosol consisting of contaminant and soot particles was measured. This may not provide correct information about the size distribution of the contaminant aerosol. If the soot particles produced by the fire are large, as they were for the polychloroprene material in the study by Halverson et al. (1987), where an aerodynamic mass median diameter of $19.9\text{ }\mu\text{m}$ was observed, the mass distribution is dominated by the uncontaminated portion of the aerosol. If the contamination aerosol exists as a separate population of particles, as indicated by Buijs et al. (1988), the size distribution of the contamination may be respirable, but may be hidden by the combustion aerosol in measurements. Of course, if the total mass distribution is respirable, the contamination aerosol is respirable as well.

The conclusion of this report is a set of procedures for estimating release fractions for plutonium in the form prevalent at the Rocky Flats Plant, which requires a detailed description of the accident to be analyzed. The development of such a description is beyond the scope of this work.

INTRODUCTION

Background Information

Plutonium is a fabricated metal once produced in large amounts to be used in the production of nuclear energy and nuclear weapons. It is a potent emitter of high-energy α -particles capable of breaking chemical bonds in the molecular structures of human cells, resulting in genetic changes and mutation of affected cells. Plutonium is one of the most toxic materials known to humans.

Nuclear fuel-cycle facilities where plutonium is processed cause notable public concern regarding the safety of operating the facilities and regarding the potential adverse effect on public health of airborne releases of plutonium to the environment during accidents. These nuclear facilities are therefore required to perform safety analyses of various hypothetical accident scenarios considered relevant and credible for a plant and to present results of these studies in Environmental Impact Statements (EIS) and Safety Analysis Reports (SAR). One of the final goals of the safety analysis is to provide a scientifically justifiable estimate of the amount of aerosolized plutonium expected to be released to the environment from the postulated accident. Since plutonium has a high potential for adversely affecting human health due to inhalation exposure, safety analyses focus on aerosolized plutonium particles in the respirable size range. An overall approach to making the calculations for performing safety analyses for nuclear facilities is described elsewhere (e.g., Ayer et al., 1988) and is not discussed in this report.

One component of information implicitly required for safety analyses is the source term, or fraction of plutonium material that can be released as aerosol during an accident. This work will review current understanding of plutonium source terms and is limited only to postulated accidents involving fires.

Several definitions related to the quantification of plutonium releases are needed to clarify the terminology adopted in this work. Airborne release of material is defined as the process by which a fraction of the material becomes dispersed in the air regardless of how long it stays airborne. Thus, if a powder is dispersed into the air by some release mechanism, it is considered airborne, even though the particles settle down in a short amount of time. This definition appears to be appropriate due to its objectivity. The release fraction thus defined is independent of the boundaries of the experimental apparatus in which it was measured. Since it is just a definition, it cannot change the

results of the safety analysis, which can, however, become more accurate and consistent with other relevant studies.

Air dispersion of plutonium can be expressed in terms of the airborne release fraction (ARF) defined as the total fraction of initial material released in the accident, or the airborne release rate (ARR), which is the average rate at which ARF is released for the duration of the accident. Both these definitions are equally pertinent and should be used as appropriate in calculations involving accident duration, amount of plutonium affected by the accident, material combustion rates, and other relevant parameters.

Respirable fraction (RF) of the mass of plutonium dispersed in the air during an accident is used in safety analyses to eliminate relatively coarse airborne particles from consideration. Since these coarse particles are not considered respirable, they do not immediately contribute to public risk from the safety analysis point of view, even if the particles are capable of staying airborne along their leak paths from the release point. For the purposes of safety analyses, it is conditionally accepted that particles having aerodynamic diameters smaller than 10 μm constitute RF. The aerodynamic diameter of a particle is defined as the diameter of a unit-density sphere having the same settling velocity as the particle under consideration.

Project Objectives and Approach

The objectives of this project are to provide an independent review of information from the available literature on plutonium release parameters obtained in worldwide studies on accidental fires that might occur in nuclear facilities and to generate technically justifiable recommendations on plutonium releases based on this review. To meet these objectives, this study was conducted in several stages, including literature search, information review and classification, data base analysis, and development of recommendations on plutonium release parameters.

As mentioned above, this work was limited to the accidental fires in nuclear facilities involving plutonium metal (which is a chemically active material that can be stored in one place in large amounts) or some other materials containing plutonium. The other materials containing plutonium considered here include a number of contaminated waste materials that can be in either solid or liquid form and various process liquids that could contain relatively large amounts of plutonium. Accordingly, this report is structured in three main sections that discuss plutonium releases from

metal oxidation processes, from combustion of contaminated solid wastes, and from combustion of contaminated liquids involved in an accidental fire.

A substantial amount of work has been conducted on accidental releases of plutonium in nuclear facilities. Since many process variables strongly affect plutonium aerosolization mechanisms and since the purpose of these studies was a concrete practical application, most of the experiments were performed on an integrated basis in which plutonium releases were measured under a specific set of conditions, without separation of the effects of the variables. Also, many of the experiments were not replicated, resulting in serious scatter and inconclusiveness of some experimental data and uncertainties associated with them. In several published review papers, researchers attempted to interpret available experimental data and to develop recommendations on plutonium release fractions to be used in SARs. The purpose of this work is to complement these studies and to improve current knowledge in this technical area by providing an independent critical analysis of all available literature sources and to provide justifiable recommendations on plutonium release parameters using comparative analysis of the main findings presented in the literature.

OXIDATION OF PLUTONIUM METAL

Introduction

In a recent and most extensive draft publication on accidental releases of plutonium, Mishima (1993-a) comprehensively described the experimental source-term data base available. Based on this analysis, he constructed a table of recommendations, a part of which related to the oxidation of plutonium metal is shown in Table 1*.

* The apparent errors in Mishima's publication were corrected as shown in Table 1 (Mishima, 1993-b).

Table 1. Mishima (1993-a) Recommended Release Parameters
for Oxidation of Plutonium Metal

No.	Process Conditions	Release Rate or Fraction		Respirable Fraction	
		α -Pu	δ -Pu	α -Pu	δ -Pu
1	Room temperature oxidation in dry air ($\mu\text{g}/\text{cm}^2\text{hr}$)	2.0e-6	7.2e-8	0.7	0.7
2	Room temperature oxidation at 100% RH ($\mu\text{g}/\text{cm}^2\text{hr}$)	7.0e-3	6.0e-4	0.7	0.7
3	Oxidation at elevated temperatures (ARF)	3.0e-5 x fraction oxidized		0.04	
4	Self-sustained oxidation (ARF)	5.0e-4		0.5	
5	Disturbed surface, high turbulence, violent airborne reaction (ARF)	0.01		1.0	
6	Explosive reaction, molten metal, rapid vapor formation (ARF)	1.0		0.5	

One purpose of the present study is to accurately evaluate plutonium source terms for accidental fires that could be used in SARs. In addition to this goal, source-term information will be generalized to minimize the amount of guess work needed to perform the analysis, and yet maintain its accuracy and usefulness. Although Table 1 provides important information on plutonium releases for several process conditions, the table does not appear to be easy to use. For example, entry 3 of the table intends to cover a wide range of conditions from room temperature to ignition, within which the ARF is assumed to be constant and equal to 3.0×10^{-5} of the oxide formed. This assumption, however, is not supported by experimental observations, resulting in significant overestimation of release fractions in most cases. Also, determination of the oxidation rate of plutonium under different conditions needed for Mishima's analysis is not straightforward, which adds more uncertainty to the analysis.

Experimental Data Base

This work will first summarize the currently available experimental data base and briefly present their main findings, separately from other review papers which will be discussed further in this report. This section of the report summarizes the most notable experiments performed for studying plutonium releases from oxidation processes. The literature sources are presented in chronological order to provide a better perspective on the development on this subject.

One of the earlier systematic experimental programs involving metallic plutonium was conducted by Stewart (1963). In one part of his work, Stewart studied rusting of both pure plutonium metal and its δ -stabilized alloy (density of which is $\rho = 15.92 \text{ g/cm}^3$, Leonard, 1967) under various test conditions, without burning the specimens. At room temperature, ARR of α -phase plutonium ($\rho = 19.86 \text{ g/cm}^3$) showed strong dependence on the relative humidity of the air surrounding the plutonium pieces and was consistently different for the two allotropic phases of the metal. Stewart also performed a series of plutonium oxidation tests at a temperature of 123°C in which plutonium is stable in β -phase, $\rho = 17.7 \text{ g/cm}^3$. Under these conditions, neither the effect of air humidity nor that of the plutonium phase were well pronounced. His observations are represented in Figure 1 using open symbols. This figure also displays corresponding results obtained by Chatfield (1968) in a work described later in this report. The lines interpolate average values obtained for both Stewart's and Chatfield's datasets for each of the test temperatures. As shown in this figure, airborne release rates span several orders of magnitude in these measurements.

In another part of the study, Stewart investigated plutonium release behavior in combustion processes. He burned bulk plutonium specimens and metal shavings under different conditions varying oxygen content in the atmosphere, rate of heat transfer to the specimens prior to their ignition point, plutonium phase and physical dimensions, and other parameters. ARFs ranged from $\sim 10^{-5}$ to $\sim 10^{-2}$ (pure O_2) in these experiments.

Mishima (1965) performed a series of six tests in which $\sim 10\text{-g}$ unalloyed plutonium samples were burned in a horizontal-flow apparatus. The ARFs measured were as low as 2.8×10^{-8} to 5.2×10^{-7} . No particles were detected by microscopic examination of the filters used to collect the releases, indicating (according to the author) that the particles were smaller than $\sim 0.1\text{-}\mu\text{m}$ in diameter. However, it appears reasonable to suppose that a substantial amount of airborne particles was lost in the apparatus due to settling and other deposition mechanisms.

Figure 1. Plutonium Oxidation Below Ignition Point

In another series of experiments, Mishima (1966) performed three tests in which large plutonium specimens weighing up to almost 1.8 kg were burned to completion. These experiments were conducted in a vertical-flow apparatus where an upward air velocity of 525 cm/s was maintained. ARFs measured in this work ranged from 3.4×10^{-5} to 4.9×10^{-4} . One sample of airborne particles of plutonium oxide collected on a filter was analyzed for size distribution, resulting in a geometric mass median diameter (geometric MMD, based on the projected diameters of the particles) of $4.2 \mu\text{m}$.

Ettinger et al. (1967) presented aerosol characterization results obtained from burning samples of sodium and plutonium. In this report, we consider only those experiments in which plutonium was used as test material.

Ettinger et al. addressed two issues in their work. First, they measured particle-size distributions of airborne releases collected on electron microscope grids using an electrostatic precipitation technique. The grids were positioned approximately 20 cm above the burner where samples of α - or δ -phase plutonium or a Pu-Co-Ce alloy containing 57.7% Pu were burned. The tests were conducted varying oxygen content of the air. Based on their microscopic examination, the aerosol collected on the grids was found to have geometric MMD ranging from 0.03 to $0.14 \mu\text{m}$ and a geometric standard deviation (σ_g) ranging from 1.24 to 1.76 for all test materials. These aerosol parameters were not explicitly affected by the oxygen concentration of the burning atmosphere. In general, airborne particles collected in this study were almost 2 orders of magnitude smaller than the particles investigated by Mishima (1966). A possible explanation of this discrepancy is that Ettinger et al., used a lower upward air velocity of approximately 15 cm/s (our estimate), as compared to the 525 cm/s air velocity used in Mishima's experiments.

Also, in experiments using δ -phase plutonium, Ettinger et al. (1967) observed relatively large chain-structured agglomerates of fine primary particles, having more than $1 \mu\text{m}$ in the longer dimension. These agglomerates visually appeared to be similar to the agglomerate particles produced by burning pure metallic gallium, which was the δ -plutonium stabilization component used in their work. Based on this similarity, the agglomerates were assumed to be made of gallium and were not included in the δ -phase plutonium particle-size distribution analysis.

The other task addressed by Ettinger et al. (1967) was to measure ARFs from burning metallic plutonium. They used only the Pu-Co-Ce alloy containing 57.7% plutonium in these experiments and a horizontal-flow apparatus aspirated at an air velocity of $\sim 150 \text{ cm/s}$. Air filters were used to collect aerosol particles. The ARFs were measured for oxygen-deficient atmospheres at two burn

temperatures and ranged from 1.8×10^{-7} to 3.9×10^{-6} . Size distribution was not analyzed in these experiments.

Chatfield (1968) investigated airborne releases from burning pure plutonium and its δ -stabilized alloy in both saturated air (100% RH at room temperature) and dry air. His experimental setup was designed to sample airborne particles smaller than $\sim 20 \mu\text{m}$ in so-called Stokes' diameter (which is defined as the diameter of a spherical particle having the same density and settling velocity as the particle in question). Chatfield measured the ARRs and aerosol particle-size distributions for plutonium oxidation temperatures ranging from room temperature to almost 1000°C .

Chatfield's airborne release rates obtained for the below-ignition temperatures (converted by the authors of this report to mass terms, as before, using $0.062 \mu\text{Ci}/\mu\text{g}$ specific activity of Pu^{239}) are shown in Figure 1 above with blackened symbols. A reasonably good agreement between his results and the results of Stewart (1963) is clearly seen.

For plutonium "combustion experiments", Chatfield did not specify the time required for the plutonium specimens to burn up in the test fires. If we assume, however, that the specimens burned to completion in approximately 10 min, which was roughly the time that the similarly sized plutonium samples burned in Stewart's (1963) experiments, an average ARF of $\sim 0.2 \times 10^{-4}$ is obtained.

Chatfield also measured particle-size distributions for the ARFs from plutonium oxidation processes. According to his observations, the RF of the released aerosol noticeably decreased with higher oxidation temperatures.

In another related study, Chatfield (1969-a) investigated the combustion behavior of plutonium in the presence of sodium. The tests were conducted using a horizontal apparatus having an air velocity of about 80 cm/s. In two of his experiments considered in this report, no sodium was present. Using as before $0.062 \text{ Ci/g Pu}^{239}$ specific activity, we calculated the average ARF as 3.0×10^{-5} and the RF as 0.34 (the maximum RF measured was 0.41). Combustion temperatures were not recorded for these experiments.

Chatfield also conducted a series of tests in which thin plutonium wires were vaporized at approximately $50,000^\circ\text{K}$, using an electric discharge. The whole amount of plutonium was released in these experiments. Linear aggregates of primary particles having diameters of approximately $0.02 \mu\text{m}$ were formed by vapor condensation and agglomeration mechanisms. Most of the aggregates visually appeared to be in the respirable size range.

Carter and Stewart (1971) studied essentially two plutonium release mechanisms from burning δ -phase specimens under "static" conditions and burning molten plutonium droplets falling in the air.

In the static oxidation experiments, they burned metal fragments at the top of a 75-cm column without disturbing the sample. The residue of these burned fragments then disintegrated on cooling and fell down the column. An upward air flow was maintained such that particles with aerodynamic diameters smaller than $\sim 30 \mu\text{m}$ were collected for analyses. A maximum value of the respirable fraction related to the whole source material burned (i.e., a product of their ARF and RF according to our convention) was found to be approximately 1.0×10^{-4} .

In the second task of the study, Carter and Stewart (1970) heated small plutonium specimens to 660°C and 2000°C in argon (or air), after which the metal droplets fell into the air and burned. If a droplet that was still burning impacted on a surface, it produced bright sparks, emitting vapor from the interior, and relatively large hollow spheroids of plutonium oxide were then found. The ARF x RF product was found to be as high as 0.01 in this case. However, the authors estimated that a fraction (0.5) of the vapor produced from the spheroids will appear in the respirable size range.

Raabe et al. (1978) undertook an experimental task in support of the Plutonium Aerosol Generation Experiments (PAGE) program, in which they studied aerosol generation by burning in air droplets of δ -phase plutonium, having 50- to 500- μm diameters, ignited with a laser. In general terms, this work was conducted in a manner similar to that used in the droplet-combustion experiments of Carter and Stewart (1971) presented above, and qualitatively, similar observations were made. The authors concluded that the vaporized fraction of molten plutonium droplets increases for smaller specimens from approximately 0.1 for burning 0.4-mm droplets to 0.4 for 0.1-mm droplets. The aerosolized fraction sampled was analyzed for particle-size distributions, using an impactor and a spiral-duct centrifuge. The impactor samples appeared to produce more conservative results, perhaps due to the partial fragmentation of particles on impact, resulting in an average aerodynamic activity median diameter of $1.15 \mu\text{m}$ and $\sigma_g = 1.53$. Assuming a log-normal distribution for this aerosol, its respirable fraction was calculated by the authors of this report as approximately 0.4.

The last experimental study presented in this section of the report is a work by Eidson et al. (1988) who performed detailed experiments on δ -phase plutonium release fractions from reactions in reducing/oxidizing atmospheres. (No significant difference between respirable aerosol releases from these reactions was noticed.) The authors conducted a total of 14 experiments in which they determined ARFs using sums of all aerosol particles collected in several sampling devices and those calculated as lost on the test chamber surfaces. Although their estimates suggested to them that these sums should be used to represent respirable releases (meaning $\text{RF} \approx 1$), the authors of this report are

inclined to consider their assessment of the RF as being excessively conservative. Eidson et al. (1988) performed a statistical analysis of the experimental results obtained in their work. They determined an average ARF value as 2.1×10^{-4} , with a worst-case value of 4.6×10^{-3} .

Review Papers

A brief review of the available experimental data base presented above indicates that plutonium always releases airborne particles when it is exposed to an atmosphere that is not inert. Even at room temperature oxidation, plutonium releases aerosol particles.

During the oxidation process, a film of plutonium oxide rapidly forms on clean surfaces of metallic plutonium. The rate of oxidation reaction is controlled at this stage by the rate of oxygen diffusion through the film and therefore decreases as the thickness of the film increases over time. During this stage of the process, a buildup of mechanical stress develops in the oxide layer due to the lower density of plutonium oxide (molar volume = $23.67 \text{ cm}^3/\text{mol}$, Haschke, 1992) as compared to the density of metal plutonium (molar volume = $12.20 \text{ cm}^3/\text{mol}$), resulting in cracking and fragmentation of the oxide layer. At this point, oxidation of plutonium metal transits to the second kinetics stage, the rate of which stabilizes due to enhanced diffusion of oxygen to newly exposed metallic surfaces. Release of Pu particles is observed at this stage of the process. After approximately a day of exposure to oxygen, this process stabilizes at a somewhat higher reaction rate, indicating the beginning of the third oxidation stage. According to Haschke (1992), the origin of this behavior has yet to be explained. A comprehensive characterization of the source-term parameters appeared to be a difficult task due to their dependence on many other process parameters controlling plutonium aerosolization from chemical reactions.

Several previously published review papers interpreted the data presented above, including (among others) work by Stewart (1963 and 1964), Mishima (1964), Felt (1967), Chatfield (1969-b), Mishima and Schwendiman (1970), Sutter et al. (1984), Ballereau (1987, translated in 1991), Mahoney and Mishima (1990), Haschke (1992), and Mishima (1993). Several of these reviews of the experimental results obtained for the oxidation of metallic plutonium are considered in this work since they reflect other attempts to organize plutonium release fractions, using different approaches and experimental data.

Stewart (1963) based his recommendations on airborne plutonium release parameters mainly on his own observations. These recommendations are reiterated below in Table 2. In brief, Stewart

summarized his findings on the plutonium release rate into the atmosphere for two categories: (1) oxidation of plutonium without ignition at different temperatures up to 500°C and (2) combustion of plutonium with temperatures of the metal exceeding its melting point of 640°C. For these two categories, he separately considered radioactive releases for either static or dynamic conditions, which are defined as corresponding to a gas flow of up to 1 m/sec around stationary material (static conditions) and as conditions under which all of the plutonium oxide formed may be released into the atmosphere (dynamic conditions). Also, for temperatures of up to 120°C, different release parameters are given for pure plutonium metal and for a δ -stabilized plutonium alloy.

To evaluate the health hazards identified with accidental releases of plutonium oxide particles into the atmosphere, respirable fractions associated with radioactive aerosols need to be determined as well. For these purposes, Stewart (1963) also measured particle-size distributions in his tests. In attempting to generalize his recommendations, he constructed one generic particle-size distribution for airborne plutonium oxide particles released in most of the plutonium-oxidation processes that he considered, except for three cases in which particle-size distributions are given for the bulk oxide powders formed.

One of these distributions was built for the entire mass of oxide powder obtained by burning plutonium under dynamic conditions. Since the airborne release fraction specified for this case was 1.0, this bulk-oxide distribution function can be used for evaluating respirable release fractions as well.

Two other bulk oxide powder distributions were also obtained for dynamic oxidation of plutonium below the ignition point. In these two cases, however, plutonium releases are given in terms of rates (per unit time), meaning that these "dynamic release rates" are actually the rates at which plutonium oxidizes without ignition under some dynamic conditions, causing oxide particles to detach from the specimen as soon as they are formed.

Respirable release fractions of airborne plutonium oxide particles are also given in Table 2. These values were calculated using Stewart's experimental observations and his recommended values for the releases.

Table 2. Stewart (1963) Recommended Release Parameters
for Plutonium Metal

No.	Process Conditions	Release Rate $\mu\text{g}/\text{cm}^2\text{hr}$		Respirable Fraction	
		α -Pu	δ -Pu	α -Pu	δ -Pu
1	Oxidation at room temperature Static ($\mu\text{g}/\text{cm}^2\text{hr}$) ¹ (fraction per hour) ²	8.1e-3 <u>3.0e-9</u>	8.1e-4	0.70	0.70
	Dynamic	80.6	8.06	0.70	0.70
2	Oxidation at ~ 120°C Static	8.1e-2	0.806	0.70	0.70
	Dynamic	8.1e+4	806.	0.70	(0.04) ³
3	Oxidation at 400-500°C (no ignition) Static	161.3	161.3	0.70	0.70
	Dynamic	6.45e+5	6.45e+5	(0.02)	(0.02)
4	Combustion Static (release fraction) (fraction per hour) ⁴	2.0e-4 <u>1.2e-3</u>	2.0e-4	0.90	0.90
	Dynamic (release fraction)	1.0	1.0	(0.001)	(0.001)

Mahoney and Mishima (1990) presented another approach to evaluating plutonium release fractions from oxidation processes. After they reviewed the available literature, they compiled an experimental data base on plutonium releases. Based on this data, two methods were proposed for the evaluation of release fractions. Neither of these two methods incorporated results from several experiments considered by the authors not applicable.

¹ Release masses of plutonium are calculated using 0.062 $\mu\text{Ci}/\mu\text{g}$ specific activity.

² Fractions (underlined values) are calculated using specimen's cylindrical geometry and α -phase plutonium density $\rho = 19.84 \text{ g}/\text{cm}^3$ (Hammond, 1984-85).

³ Values in parentheses are based on particle-size distribution functions obtained for bulk oxide powders.

⁴ Based on ~ 10 min combustion time reported for the 10-15 g Pu specimens.

First source term evaluation method considers maximum values for plutonium releases obtained from the experiments. These values are given in terms of either the total release fractions of initial material or the fractional release rates. The three highest release fractions identified ranged from approximately 2×10^{-4} to 2×10^{-2} according to Stewart's experiments. However, two of these experiments were performed using oxygen-enriched atmospheres, and one experiment was performed with pure metal under repeatedly fluctuating temperature of the specimen, which probably resulted in high releases due to fluctuating mechanical stresses. None of these experiments are likely to represent a credible accident scenario. The highest release rate specified by the authors for burning plutonium was $3.2 \times 10^{-4} \text{ hr}^{-1}$ as measured for pure metal by Mishima (1966).

Next, Mahoney and Mishima (1990) excluded "extraordinary" conditions of oxygen-enriched atmospheres from consideration and tabulated their findings separately for pure metal and for δ -alloy. Both the release fractions and the release rates were used in the table. Mahoney and Mishima noticed a certain inconsistency between several experimental results since some of the release rates, if assumed constant for the duration of sampling, could not result in the release fractions measured. This observation could probably be explained by recognizing that the table they used still contained some extraordinary experiments, resulting in the comparison of different release mechanisms. For example, during complete oxidation of pure metal and δ -stabilized alloy in two well-defined experiments in which ~ 10 -g plutonium cylinders steadily burned to completion in approximately 10 minutes, Stewart (1963) measured a 1.5×10^{-5} fraction of initial material that was airborne, resulting in approximately $9.0 \times 10^{-5} \text{ hr}^{-1}$ release rates for both cases. This agrees very well with Mishima's (1966) measurements using large plutonium specimens, where airborne release rates were found to range from 9.0×10^{-5} to $3.2 \times 10^{-4} \text{ hr}^{-1}$. Also, these experiments indicated that there was no significant difference in airborne releases from burning plutonium in either of its allotropic forms, which was to be expected since the metal loses its crystal structure as it melts under the high combustion temperatures of the experiments.

The other model developed by Mahoney and Mishima was aimed at finding empirical correlations between aerosol release rates and several process parameters assumed to affect plutonium release mechanisms. Mahoney and Mishima limited this analysis to considering only the rates of plutonium releases because the total fractional releases were thought to be less applicable in some accident scenarios in which fire is postulated to be extinguished before plutonium metal involved in the accident is completely burned.

As a result of statistical analyses performed on this more limited data base, Mahoney and Mishima arrived at several empirical correlations establishing the effects of relative humidity (RH) of the air and peak temperature of either pure plutonium or its δ -alloy on airborne releases from an accidental fire. Since these equations represent some average values of release parameters, they were adjusted by adding respective standard errors provided by the statistics, resulting in more conservative predictions. Several of these equations are reproduced below as an illustration.

Pure Pu

$$\log(ARR) = -3.483 + 0.01292 \cdot RH - \frac{2123}{T^{\circ}K} \quad (1)$$

$$\log(ARR \cdot RF) = -3.829 + 0.01232 \cdot RH - \frac{2681}{T^{\circ}K} \quad (2)$$

δ - Pu

$$\log(ARR) = -2.131 + 0.01169 \cdot RH - \frac{2823}{T^{\circ}K} \quad (3)$$

$$\log(ARR \cdot RF) = -2.384 + 0.00701 \cdot RH - \frac{2779}{T^{\circ}K} \quad (4)$$

Figure 2 reproduced from the reference for pure metal oxidation at 100% relative humidity illustrates this methodology. In this figure, blackened symbols were used for those data points that we have identified in the data base considered.

As shown in Equations 1 through 4, ARR increases by a factor of more than 15 as RH increases from 0% to 100%. This, however, is not that obvious from the available data base. First of all, in the few experiments described in the literature in which air humidity was controlled, 100% RH was measured at room temperature, which corresponds to approximately 16 mg/l absolute humidity. This was shown to significantly affect ARR for plutonium oxidation at room temperature

**Figure 2. Statistical Correlation for Pure PU Release Rate at 100% RH
(Mahoney and Mishima, 1990)**

only (Stewart, 1963; Chatfield, 1968), resulting in increases in airborne release fractions of more than 4 orders of magnitude; however, these release fractions were still extremely low. At oxidation temperatures exceeding approximately 100°C, this effect diminishes, if not reverses, as illustrated above in Figure 1. Generally speaking, air containing 16 mg/l moisture is still very dry at elevated temperatures produced during self-sustained combustion of plutonium (16 mg/l moisture content in the air corresponds to less than 3% RH at 100°C), and it seems reasonable to expect very little effect of moisture content in room air on the oxidation behavior of plutonium at temperatures above the ignition point.

The effects of plutonium combustion peak temperature on airborne release parameters are also not apparent, based on the data available in the literature. According to Figure 2, ARR of plutonium monotonically increases with the peak temperature of oxidation, including combustion regimes as well. However, the data shown in Figure 3, which are based on the plutonium combustion results of Stewart (1963) do not support Correlation (1). The figure shows airborne release fractions obtained in experiments in which combustion time of plutonium specimens weighing about 10 g was approximately 10 minutes.

Ballereau (1991) also reviewed published information on plutonium oxidation and release parameters under various temperatures and combustion conditions. Based on his analysis of the data base, Table 3 shows Ballereau's recommended release fractions for plutonium metal involved in an accidental fire.

Haschke (1992) has recently performed another review of experimental data attempting to develop a method for estimating plutonium ARR from oxidation processes based on analysis of a complete size distribution of oxide particles formed from oxidation of bulk plutonium specimens. This attempt, however, was not straightforward because (1) complete particle size distribution of a bulk oxide material was not available from experimental data presented in the literature and (2) particle size distributions available for either airborne release fractions or plutonium oxide residue particles remaining from the oxidation process were obtained using different measurement techniques. These different techniques were also based on different definitions of particle size, which therefore complicated Haschke's derivation of the total particle size distribution function. Haschke found a certain analogy between size distributions of plutonium powders formed by oxidation of plutonium metal in air (Stewart, 1963; Mishima, 1965) and plutonium hydride particles, a complete distribution of which was obtained by Stakebake (1981). Using this analogy, he suggested a complete size distribution for plutonium oxide powder, which is reproduced in Figure 4.

Figure 3. Airborne Release Fraction for Burning Plutonium Metal (Stewart, 1963)

Figure 4. Mass Distribution of PuO₂ Bulk Powder (Haschke, 1992)

Table 3. Ballereau (1987) Recommended Release Parameters
for Oxidation of Plutonium Metal

No.	Combustion Conditions	Release Fraction	Respirable Fraction
1	Static (air velocity < 150 cm/s)	1.0e-4	---
2	Dynamic (air velocity > 150 cm/s) temperature 800-1000°C	5.0e-4	1.00
3	Vigorous (projection of droplets & sparks) temperature 1500-2000°C	0.1	0.01
4	Combustion at very high temperature (T > 3000°C)	0.1	0.046

Figure 4 uses the geometric diameters of the particles as obtained from a microscope examination. These actual particles have the PuO_2 density of 11.45 g/cm^3 . The aerodynamic diameters of the particles appear to be approximately 3.4 times their geometric diameters, so that a $10 \text{ }\mu\text{m}$ aerodynamic size corresponds to a $3 \text{ }\mu\text{m}$ actual PuO_2 particle. Therefore, according to the distribution shown in Figure 4, the mass fraction of particles smaller than $\sim 3 \text{ }\mu\text{m}$ (geometric diameter) constitutes the respirable fraction of the bulk oxide powder. This fraction is found from Figure 4 to be somewhat less than 0.0001, which thus represents the maximum possible release of respirable plutonium oxide particles in a fire.

Haschke also addressed the kinetics of the plutonium oxidation process by combustion, suggesting an oxidation rate for burning plutonium of $0.2 \text{ g PuO}_2/\text{cm}^2 \text{ min}$.

Technical Discussion

An attempt to organize the available information on oxidation releases for plutonium metal in more general and technically justifiable terms usable for safety analysis is presented in this section. It appears that plutonium oxidation release mechanisms fall into three general categories: (1) oxidation

below ignition point, (2) combustion below boiling point, and (3) plutonium aerosolization from highly energetic events in which plutonium vaporization is expected. These categories are considered next.

Plutonium Oxidation Below Ignition Point

Several technical publications were reviewed in this work considering the kinetics of plutonium oxidation under various conditions. As discussed above, several methods were suggested for calculating ARFs/ARRs and RFs based on the oxidation kinetics considerations and the total particle size distribution parameters obtained for the whole oxide powder. There are however many unresolved issues associated with such a mechanistic analysis, making it more uncertain and difficult to implement.

In other studies, concrete values obtained under some specific sets of conditions are recommended for covering a large range of process parameters in which radioactivity release parameters vary to a great extent. This results in serious under- or overestimated predictions that are sometimes difficult to justify.

Our approach presented here for the below-ignition oxidation releases is to a degree after work of Mahoney and Mishima (1990) in that empirical correlations developed for actual experimental data be used for calculating the release parameters. As opposed to their work, however, we consider a more narrow range of model applicability, limiting it to only one category of oxidation conditions addressed here.

The experimental data base applicable to this analysis appeared to be reduced to only two literature sources, namely Stewart (1963) and Chatfield (1968). Experimental data extracted from their graphs were shown in Figure 1. The data presented indicate that despite the noticeable scatter and unexplained tendencies shown by the average-value lines, two variables control ARR of plutonium at low temperatures: metal allotropic phase and air humidity. At higher temperatures, these differences diminish, while ARR increases. These trends were correlated using the following equations:

Pure plutonium

$$\log (ARR) = (- 4.36 + 0.071 AH) \left(0.23 - \frac{350.6}{T (^{\circ}K)} + \frac{1.91e5}{T^2 (^{\circ}K)} \right) \quad (5)$$

where AH is the absolute humidity of the air in mg/l

δ -Pu alloy

$$\log (ARR) = (- 7.47 + 0.101 AH) \left(0.60 - \frac{661.3}{T (^{\circ}K)} + \frac{2.16e5}{T^2 (^{\circ}K)} \right) \quad (6)$$

Each of these two equations defines a two-dimensional surface of plutonium ARR as a function of air humidity and temperature for either pure plutonium or a δ -phase stabilized alloy, respectively. One of these surfaces obtained for pure plutonium (Equation 5) is shown for illustration purposes in Figure 5. Figure 6 compares experimental data with model predictions made using Equations 5 and 6 for corresponding air temperatures, the predictions are shown with dotted lines.

A constant value of 0.7 was recommended by Stewart (1963) as the respirable fraction, RF, of plutonium releases under all the conditions considered in this section. From a more careful analysis of his experimental results, however, and results obtained by Chatfield (1968), a more accurate estimate of RF is possible. The experimental data considered are shown in Figure 7. These data were obtained from oxidation of plutonium in dry air, resulting in higher and therefore more conservative values of RF. From Figure 7, the following empirical correlation is developed for predicting RF for the range of oxidation temperatures:

$$RF = 1.07 - 0.00353 \cdot T (^{\circ}C) + 3.82 \cdot 10^{-6} \cdot T^2 (^{\circ}C) \quad (7)$$

Equation (7) is recommended to evaluate respirable fractions of airborne plutonium released from oxidation processes for a range of temperatures up to the point of ignition.

Figure 5. Pure Plutonium Release Rate as a Function of Oxidation Temperature and Air Humidity (Eq. 5)

Figure 6. Plutonium Release Rates for Oxidation Below Ignition Point (Predictions are shown with dotted lines.)

Figure 7. Respirable Fraction of Airborne Releases from Plutonium Oxidation Below Ignition Temperatures

Combustion of Plutonium Below Boiling Point

From the review of plutonium aerosolization behavior studies presented above, it appears impractical to develop any conclusive views regarding the effects of combustion peak temperature or any other process variable on airborne plutonium release parameters. This is illustrated in Figure 8 where a number of experimentally obtained ARFs are plotted versus their respective peak temperatures.

The combustion behavior of plutonium is affected by many natural processes including heat generation and transfer by various mechanisms. During an accidental fire, heat balance will most likely be changing in time, resulting in various temperature regimes and, accordingly, varying instantaneous release rates. Thus, peak temperature might not be an important variable in quantifying plutonium releases. From reviewing Figure 8, it appears to the authors that a single ARF value of 2.0×10^{-4} will encompass most accident scenarios in which plutonium is burned under "static conditions" featuring no significant mechanical disturbance.

The respirable fractions of aerosolized plutonium measured by different researchers in combustion experiments are found to vary as well. According to the total particle size distribution graph of Haschke (1992), which was shown above in Figure 4 using aerosol geometric diameters, fractional content of respirable particles (particles smaller than $\sim 3 \mu\text{m}$ in geometric diameter) in plutonium oxide powder is on the order of 10^{-4} . Assuming that all respirable particles formed during combustion of plutonium are released as a part of the total airborne releases, a bounding value of RF in the aerosol can be estimated as a ratio $\sim 1.0 \times 10^{-4}$ over the ARF. Thus, assuming $\text{ARF} = 2.0 \times 10^{-4}$, a value of $\text{RF} = 0.5$ is obtained.

According to both Stewart (1963) recommendations and Chatfield (1968) measurements, a value of $\text{RF} = 0.7$ is recommended. Chatfield (1969-a) measured RF's in two of the experiments performed in that study and found their values of approximately 0.3 and 0.4, and, also according to Mishima (1966), a value of less than 0.5 can be used to represent the RF.

It should be recalled, however, that from the safety analysis standpoint, a product of $\text{ARF} \times \text{RF}$ is used in calculations. As discussed above, uncertainties associated with airborne releases frequently result in order-of-magnitude estimates of ARF, meaning that not much more accuracy can be gained in final results from precisely measuring RFs of the releases. A somewhat average value of 0.5 is therefore recommended for RF in this analysis for burning plutonium under static conditions.

Figure 8. Airborne Release Fractions from Combustion of Plutonium Metal

The dynamic conditions of plutonium releases from a fire are defined in this work as some conditions resulting in a complete dispersion of the oxide formed. Thus, ARF is apparently equal to 1. in this case. In terms of release rates, a value of $0.2 \text{ g PuO}_2/\text{cm}^2 \text{ min}$ according to Haschke(1992) can be used.

As discussed above, a value of 1.0×10^{-4} is envisioned for RF provided that all the oxide powder has been aerosolized (Haschke, 1992). The same value was measured by Carter and Stewart (1971) in their "static experiments" where they burned plutonium under some stationary conditions, after which the residue fell into the test column dispersing all the oxide particles (which is the dynamic conditions as defined in this work). They measured the respirable airborne fraction of the powder dispersed in those tests, and also found a maximum value of 1.0×10^{-4} , which is therefore recommended in this work for burning plutonium under dynamic conditions.

Plutonium Aerosolization From Highly Energetic Events

Certain accident scenarios postulated for safety analyses involve highly energetic events in which high thermal and mechanical stresses are envisioned, such as severe nuclear accidents involving explosions. As shown in several experimental studies where plutonium releases from these events were investigated, oxidation temperatures in excess of the plutonium boiling point of 3235°C were observed, resulting in generation of sparks and vapor.

It is not straightforward to define ARF for these scenarios. As was measured in various experiments dealing with burning airborne droplets, a fraction of approximately 0.5 of initial mass of plutonium was released as vapor, resulting in formation of approximately all-respirable aerosol. Being mostly interested in the product of ARF and RF as mentioned above, we arbitrarily assign values of 1.0 and 0.5 for these release parameters, respectively.

The following Table 4 summarizes our recommendations developed for the evaluation of plutonium releases from oxidation processes.

Table 4. Plutonium Oxidation Release Parameters

Pu Release Conditions		Oxidation Below Melting Point		Combustion			
				$T^{\circ}_{\text{melt}} < T^{\circ} < T^{\circ}_{\text{boil}}$		$T^{\circ} > T^{\circ}_{\text{boil}}$	
		ARR $\mu\text{Ci}/\text{cm}^2\text{hr}$	RF ¹	ARF (of total)	RF	ARF (of total)	RF
Static	Pure Pu	Eq.8	Eq.10	2.e-4	0.5	---	---
	δ -Pu alloy	Eq.9	Eq.10				
Dynamic		---	---	1.0	0.0001	1.0	0.5

ARR:

Pure plutonium

$$\log (ARR) = (- 4.36 + 0.071 AH) \left(0.23 - \frac{350.6}{T^{\circ}K} + \frac{1.91e5}{T^{\circ}K^2} \right) \quad (8)$$

where AH is the absolute humidity of the air (mg H₂O / l)

δ -Pu alloy

$$\log (ARR) = (- 7.47 + 0.101 AH) \left(0.60 - \frac{661.3}{T^{\circ}K} + \frac{2.16e5}{T^{\circ}K^2} \right) \quad (9)$$

RF:

$$RF = 1.07 - 0.00353 \cdot T (^{\circ}C) + 3.82 \cdot 10^{-6} \cdot T^2 (^{\circ}C) \quad (10)$$

¹ RF is given as a fraction ($d_a < 10 \mu\text{m}$) of airborne mass

PLUTONIUM RELEASE DURING SOLID FUEL COMBUSTION

Introduction

The importance of storage-related accidents will probably increase with time as processing operations are scaled back, and contaminated wastes emerge among primary plutonium-containing materials at the processing facilities. Since a large fraction of typical waste consists of a small quantity of radioactive contamination mixed into a much greater quantity of nonradioactive, but usually flammable, solid material, the properties and behavior of the flammable material control the release of the contaminant to a large degree. However, the chemical form of the contamination itself can also introduce significant variations in the amount and particle-size distribution of airborne radioactive materials. Because many contaminated waste materials (such as glovebox windows, gloves, ion-exchange resin, paper and rags) are flammable, several experimental studies have been performed to determine the characteristics of the contamination released during combustion of these waste materials. In early studies, actual plutonium was sometimes used as an oxide or salt contaminant compound, whereas in more recent studies, chemically similar materials have been used as simulated contaminants. In general, the results obtained from using the simulants are consistent with the results obtained from actual plutonium contamination.

Literature Review

Available literature references dealing with plutonium release during combustion of solid fuels were evaluated for this program. The references were published between 1970 and 1992, and represent a number of subject areas, in addition to papers directly related to the release of radioactive aerosols during fires. Fire and combustion literature (Tewarson and Pion, 1976; Heskestad, 1981; Chan and Mishima, 1983; and Pitts, 1991) provided valuable insight into a number of factors that influence the behavior of fire.

In one of the earliest studies on the subject of radioactive contaminant release during combustion of flammable waste materials, Mishima and Schwendiman (1970) measured the fractional release of plutonium from burning contaminated wood shavings, cardboard, cheesecloth, and surgeon's gloves. The plutonium was applied to the combustible material as a plutonium nitrate

solution. An upward air flow of 0.056 m/s was maintained during combustion. Under these conditions, the maximum observed aerosol release fraction (ARF) was 0.01 of the initial contamination present prior to combustion. The collection method for the airborne contaminant was not identified in the study.

In Mishima and Schwendiman's study, additional experiments were conducted using uranium compounds as simulants for plutonium. Finely divided uranium oxide (UO_2) powder (with a mass median diameter of 6 μm) was used to simulate a dry plutonium contaminant, whereas an uranyl nitrate hexahydrate (UNH) solution was used to simulate plutonium-containing solutions. Each type of simulant was applied to three cellulose-based combustible materials (i.e., tissue paper, cardboard, and cheesecloth) and to tape-sealed cardboard boxes containing contaminated tissue paper and cheesecloth in plastic bags. These materials were burned on a grate with air flow velocities of either 0.46 or 1.04 m/s.

The ARFs from these tests were the highest reported in the literature. The maximum ARF (0.55) occurred for UO_2 on tissue paper at an air flow rate of 1.04 m/s. For the UNH solution, the maximum ARF was 0.14, again on tissue paper at 1.04 m/s. The lowest releases for both simulants occurred for cardboard, but the effect of air velocity was less pronounced. The combined waste, sealed in the boxes, released similar amounts of the contaminants as the cardboard.

As shown later in this report, the results obtained in Mishima and Schwendiman's 1970 study are consistent with the results presented in other references. When a high air flow rate was used in these experiments, the ARFs during combustion of cellulose materials were much higher than those observed in experiments in which low air velocities were used. As discussed below, the updraft velocities can exceed the velocities used in these tests during a large fire, and releases of this magnitude are possible if contaminated cellulose is swept into a large fire's plume.

The variability of the release data is also similar to that from other reports dealing with contaminant release during combustion and is up to 1 order of magnitude for similar test conditions. The variability appears due to the desire of the authors to examine many factors, such as air velocity, combustible material, contaminant concentration, and contaminant type. This approach was reasonable, since little, if any, prior information existed on the subject. A bar chart showing the average ARF for each contaminant and combustible is presented in Figure 9. Results of individual experiments appear as filled circles.

Figure 9. Contaminant Release Fractions During Solid Fuel Combustion Experiments (Mishima & Schwendiman, 1970)

Several uncertainties are associated with that work besides data variability. The experimental conditions used in the tests are not fully described. Since the particle size of the released contaminant was not measured, the respirable fraction is not known.

In a series of follow-on experiments, Mishima and Schwendiman (1973-a) investigated aerosol releases during combustion of cardboard boxes containing simulated waste. These experiments were conducted in the Radioactive Aerosol Release Tank (RART) at the Pacific Northwest Laboratory (PNL). The RART was fitted with fallout trays on the vessel floor and aluminum foil strips on the vessel walls so that the quantity of aerosol on the floor and walls could be measured. The airborne contamination was collected with a filter. The material collected during the tests was dissolved in nitric acid, and the solutions were analyzed for uranium concentration by fluorimetry. The uranium detection limit was 5×10^{-8} g/ml of uranium. Cascade impactors were used to measure the aerosol size distribution.

The combustible material used in the tests was a mixture of cellulose, rubber, and plastics in ratios similar to those found in typical waste cartons at that time. Table 5 shows both the average composition of the test cartons and the range of compositions for each type of waste. When these tests were conducted, the cartons were considered acceptable as waste containers. Although the waste is now stored in 55-gal steel drums, the results of these experiments are still used for estimating the contaminant release during combustion of mixed waste. Since no plutonium was used in these tests, UO_2 powder and UNH solution were used as simulated contaminants. Approximately 90 percent of the UO_2 powder had an aerodynamic equivalent diameter of $< 10 \mu\text{m}$.

Table 5. Contents of Test Waste Cartons (Mishima and Schwendiman, 1973-a)

Material	Average (%)	Range (%)
Cardboard	17.5	15.7 - 20.9
Paper	41.1	31.7 - 43.9
Plastic	9.7	8.0 - 14.4
Rubber	2.4	1.8 - 4.2
Miscellaneous (rags, oil, tape, etc.)	29.3	23.8 - 38.2

The tests conducted by Mishima and Schwedinman (1973-a) were designed to provide an integral estimate of the contaminant released during combustion of a typical waste container, rather than estimating releases from mixed waste, based on releases measured for each waste component. The following three contaminant dispersal scenarios were included in the tests:

1. Half of the contamination was dispersed on loose waste materials sealed in plastic bags placed randomly in the carton. The remaining contamination was dispersed on additional loose waste materials within the carton. The cartons were sealed with tape.
2. All of the contamination was dispersed on loose waste materials within the carton. The cartons were sealed with tape.
3. All of the contamination was dispersed on loose waste materials within the carton. The cartons were not sealed.

The ARFs calculated for these experiments from the measured airborne concentration ranged from 3×10^{-5} to 5.3×10^{-4} , with the highest release occurring for UO_2 powder in scenario 1 above, and the lowest release occurring for air-dried UNH solution also in scenario 1. When the material collected on the walls was included as airborne contamination, the ARFs ranged between 3×10^{-5} to 2.3×10^{-3} . The ARFs observed in these tests were lower than the releases that occurred in the laboratory-scale tests described earlier. However, the results are consistent because combustion in this series of tests occurred without additional airflow. Airflow in these tests was produced by the natural convective flow generated by the fire.

Size distributions for each test burn were not given; however, in all of the tests, over 50 percent of the aerosol had an aerodynamic equivalent diameter of less than $10 \mu\text{m}$. In 8 of the 11 tests, over 80 percent of the aerosol recovered was below that size.

Bhanti et al. (1988) performed experiments to measure ARFs during combustion of polystyrene ion-exchange resins used in nuclear fuel-reprocessing plants. Three scaled down experimental chambers, representing a plutonium process cell, were used to allow estimates of the effect of chamber size on release parameters. The largest chamber was approximately 1:5 scale, whereas the medium chamber was 1:10 scale, and the smallest chamber was 1:20 scale. Thorium was used as a substitute for plutonium in these experiments. The thorium concentration in each sample was determined using either gamma spectrometry or neutron activation analysis.

In a typical experiment, the contaminated resin, wet with 8 M nitric acid, was dropped into an electrically preheated crucible. Ignition occurred within approximately 30 seconds, and the fire

continued for about 3 minutes. Aerosol samples were collected for an additional hour after the fire burned out. In the study, filters were used to measure total airborne concentration, and impactors were used to measure the aerosol size distribution. For some experiments, the resin was dried in a desiccator for 24 hours before the resin was used. The measured "ARFs", as defined by Bhanti et al., are shown in Table 6.

In the Bhanti et al., (1988) study, the ARF was defined to include only contamination that had been transported to the collection filters. Contamination deposited on the interior surfaces of the test chamber was not considered to be airborne contamination. Although the ARF defined by Bhanti et al., may be appropriate in some instances (such as when the amount of contamination entering the ventilation system needs to be evaluated), the contamination on the interior surfaces of the chamber was transported as airborne particles and should be included as part of the total release.

Table 6. Thorium Contamination Release During Combustion of Ion-Exchange Resin (Bhanti et al., 1987)

Test	Aerosol Release Fraction		
	Small-Scale Chamber	Medium-Scale Chamber	Large-Scale Chamber
1 ^(a)	0.00024	0.00093	0.00012
2 ^(a)	0.00021	0.00066	0.00010
3 ^(a)	0.00024	0.00041	0.00008
Mean	0.00023	0.00067	0.00010
4 ^(b)	0.00012	0.00025	—

^(a) Wet resin.

^(b) Dry resin.

The location of the deposition in the chamber was monitored by analyzing material collected on aluminum foil placed in the test chambers. Table 7 shows the deposition distribution in these experiments. Airborne releases, according to Bhanti et al., were highest for the medium-scale test chamber. This is probably due to the surface deposition effects which were more pronounced in the tests using the small-scale chamber, and higher experimental errors expected in the large-scale test chamber. In the small chamber, the nearness of the walls allowed rapid deposition of the aerosol on

the surfaces, thus lowering the airborne concentration. The total release, however, was greatest in the small-scale tests, consistently decreasing as the scale of the tests increased. Based upon these results, a large fire may not generate higher fractional releases than smaller fires involving smaller amounts of contamination.

Table 7. Distribution of Thorium Contamination During Combustion of Exchange Resin
(Bhanti et al., 1988)

Location	Fraction of Initial Thorium Recovered		
	Small-Scale Chamber	Medium-Scale Chamber	Large-Scale Chamber
Ceiling	0.00002 ± 0.00005	0.00024 ± 0.00003	0.00005 ± 0.00001
Walls	0.01013 ± 0.00010	0.00085 ± 0.00008	0.00029 ± 0.00002
Floor	0.03540 ± 0.00005	0.01710 ± 0.00009	0.00074 ± 0.00008
Total surface	0.04555 ± 0.00012	0.01819 ± 0.00012	0.00108 ± 0.00008
Airborne (ARF)	0.00023 ± 0.00004	0.00066 ± 0.00002	0.00010 ± 0.00002
Total release	0.04578 ± 0.00013	0.01885 ± 0.00013	0.00118 ± 0.00008
In crucible	0.952	0.981	0.999

Aerosol size distributions were measured for the medium-scale and large-scale test chambers, using 1-hr sampling with a cascade impactor. Size distributions were determined for total mass and thorium activity. The aerosols generated in both tests were in the respirable size range. For the medium-scale chamber model, the aerodynamic mass median diameter (AMMD) was $1.35 \mu\text{m}$. The corresponding diameter for the large-scale model was $1.45 \mu\text{m}$. The thorium size distribution (based on radioactivity) was somewhat different from the mass size distribution. Thorium had a nearly log-normal distribution. The aerodynamic activity median diameters (AAMD) were 1.65 and $2.15 \mu\text{m}$ for the medium-scale and large-scale models, respectively, with a σ_g of 2.1 for both. The large chamber contained more large particles, probably because the sedimentation removal mechanism becomes less effective whenever the particles must fall a greater distance to reach the floor of the chamber.

Bhanti et al. (1988) concluded that two types of aerosols were produced during combustion of the resin: stable and unstable. Unstable aerosol consists of the very large and very small particles

that are most affected by sedimentation and diffusion removal mechanisms, respectively. Unstable aerosol is only airborne when the fire is generating new particles. When combustion stops, the unstable particles are removed rapidly. Stable aerosol consists of the intermediate-sized particles, which are less affected by the removal mechanisms and remain airborne for much longer periods of time.

Based on their results, Bhanti et al. (1988) recommended an ARF of 0.001 for ion-exchange resin fires in full-size cells. These results assume that the facility remains intact and that the ventilation system in the plant remains in operation. For a severe accident that causes extensive damage to the structure of the building, the fraction of contamination that escapes can be significantly greater.

Halverson et al. (1987) conducted a series of 40 experiments using four solid fuels and three contaminant species. The fuels tested were polychloroprene rubber, polymethylmethacrylate (PMMA), polystyrene, and cellulose. A fine depleted UO_2 powder ($\text{MMD} = 1 \mu\text{m}$, $\sigma_g = 2$), uranyl hexahydrate (UNH) solution and UNH salt cake, from dried UNH solution, were used as simulated contamination. External heat sources were used to provide additional radiant heat flux to the burning fuel. This additional heat was used to simulate the heat input from a fire larger than the actual test fire. The radiant heat flux used was at 0, 13, and 24 kW/m^2 . An external air flow of 8 l/min was drawn through the experimental apparatus. At the dimensions of the apparatus, this flow was equivalent to a flow velocity of approximately 0.5 cm/s.

Halverson et al. measured releases from contaminated materials as functions of time. From their measurements, two distinct types of contamination release behavior can be noticed. As shown in Figure 10-a, plastic materials produce high releases during the initial stage of the tests, after which the releases effectively cease. Release profiles for two typical tests involving cellulose, which are shown in Figure 10-b, indicate that more uniform releases of the contaminants take place during combustion of cellulosic materials.

The release from burning polychloroprene rubber produced the greatest release in the tests using UNH solution. Prior to actual ignition, the rubber smoldered for several minutes. Release from the polystyrene also occurred prior to ignition. While the contaminant was emitted, the polystyrene beads collapsed into a viscous mass. After the mass stabilized, release of contaminant was very low. Timing of the burning PMMA release was similar to that of the polychloroprene and polystyrene - the bulk of the release occurred when the fuel was softening prior to flaming combustion. Halverson et al., (1987) noted that the PMMA and polystyrene bubbled during the high

Figure 10. Contaminant Release Profiles for Burning Solid Materials (Halverson et al., 1987)

release period. This behavior suggests that contaminant release is caused by outgassing of the fuel. When the gas source was exhausted, the release ceased.

Combustion of cellulose produced the lowest releases observed in these tests. One cellulose test burn was run at twice the normal air flow rate, and the observed release was much higher in that case as well. This dependence on air flow is consistent with the observations of Mishima and Schwendiman (1970). Interestingly, when cellulose was burned without application of external heat, the material burned more strongly, and the release fraction increased compared to burns with external heat. This also suggests that fuel outgassing could be an important cause of contaminant release.

The variability of the contamination releases in the study of Halverson et al. is shown in Figure 11. Three contamination forms were tested, with four different solid fuels. Only UNH contamination was used with the polystyrene resin.

The particle-size distribution of the released aerosol was presented for several tests (shown in Table 8). The study did not indicate whether the size distribution was for total aerosol or for contamination aerosol. As shown in the Table, the respirable fraction represents a significant portion of the aerosol material.

Table 8. Size Distributions of Released Aerosol Particles
(Halverson et al., 1987)

Run Number	Fuel	AMMD, μm	σ_g	Fraction Respirable
10	PMMA	4.3	2.3	0.84
11	Polychloroprene	19.9	4.6	0.16
12	Cellulose	< 0.1	—	0.97
14	PMMA	4.9	4.6	0.63
23	Cellulose	10.5	5.7	0.40
28	Cellulose	7.8	5.2	0.47
30	PMMA	3.9	2.5	0.84
38	Polystyrene	1.7	3.8	0.90
51	PMMA	1.4	4.3	0.95

Figure 11. Contaminant Release During Combustion of Solid Fuel (Halverson et al., 1987)

Buijs and Chavane de Dalmasey (1983, 1988) conducted an extensive series of tests to determine radioactive ARFs resulting from combustion of contaminated PMMA. PMMA is commonly used as the window material in gloveboxes. Since old gloveboxes comprise a significant portion of the waste material currently in storage, their combustion characteristics are important in evaluating the behavior of fires in plutonium-processing facilities.

The experiments conducted by Buijs and Chavane de Dalmasey (1983 and 1988) were performed in a 170 m³ fire room designed to handle complete gloveboxes or parts of gloveboxes. A mixture of cerium oxide and europium oxide was used as simulated contaminants in two series of tests.

The initial tests conducted in 1983 were designed as scoping experiments intended to develop the procedures needed for the full test series. In the initial report, two fire tests were described: a 1-kg burn with 0.5 g of contamination and a 6.6-kg burn with 5 g of contamination. The range of temperatures generated in the fires for both tests was similar – 400°C to 500°C.

The mean combustion rate was defined in these tests as the amount of PMMA, in grams, consumed per one minute in the fire. The airborne contaminant concentration was ~0.03 mg/m³ for a small fire with a mean combustion rate of 60 g/min and ~1 mg/m³ for a large fire with a mean combustion rate of 300 g/min. ARFs were not given for these tests, and no particle-size distributions were measured. However, the particles were examined by scanning electron microscopy (SEM). The simulated oxide contaminant appeared to exist as a distinct aerosol population rather than as particles coagglomerated with soot.

A series of simulated glovebox fires was described in detail in the second report (Buijs et al., 1988). Here, the intent was to perform a number of experiments under reproducible fire conditions. Reproducibility of the fires themselves was needed so that the effects of different contaminant loadings and placement procedures in the burning material could be studied. Approximately 5 kg of PMMA was burned in each experiment, yielding a 0.6-MW heat source with a reproducibility better than 10 percent. The room ventilation rate was 2000 m³/hr for these tests. The fuel stack of five 0.32- x 0.32-m PMMA plates was constructed for the standard fires. Each 8-mm-thick plate had a 60-mm hole bored in the center to improve air flow through the burning stack. For fires involving PMMA only, the contamination was applied to all five plates. The mixed oxide Ce_{0.84}Eu_{0.16}O_x used as a simulated contaminant was prepared from a mixture of solutions. The hydroxides were coprecipitated with ammonia, filtered, and calcined at 900°C to convert hydroxide to oxide, then were ground and sieved through a 40-μm screen. The fraction that passed the 40-μm screen

(AMMD = $10.5\ \mu\text{m}$, $\sigma_g = 2.2$) was used in the tests. During combustion, the weight of the fuel was monitored continuously with an electronic balance that supported the burn pan. Mean combustion rates were calculated from the weight data. Analyses of the aerosol were performed by neutron activation analysis of both components of the mixed oxide. No alteration of the rare earth mixture composition was detected, and the 4:1 Ce to Eu atomic ratio remained constant throughout the tests. The morphology of the airborne oxide particles, as determined by SEM examination, was not affected by the fire.

During the fire, 60 to 70 percent of the contaminant became airborne, as indicated by the quantity of contaminant found on the walls, floor, and ceiling and in the residue of the burn pan. Only 1 to 2 percent of the contaminant was collected with the combustion gases in the ventilation duct. These findings appear consistent with the observations reported by Bhanti et al. (1988) in that most of the released contamination remains near the fire site. The number mean aerodynamic diameter of the particles collected in the ventilation duct was $1.05\ \mu\text{m}$, with a σ_g of 2.2.

The report by Buijs et al. (1988) also presents the distribution of contamination collected on the walls, floor, and ceiling of the fire room; however, the data are reported as mg/m^2 and are not converted to release fractions. This report provided valuable insight into the amount of aerosol released during fires. In addition to aerosol concentration measurements, Buijs et al. described their experimental procedures in detail and provided measurements of temperature and combustion gas composition.

In a supplemental report, Buijs et al. (1992) described contamination releases resulting from other contaminated materials exposed to PMMA fires. These materials included stainless steel (painted and unpainted), aluminum (painted and unpainted), polycarbonate, polyethylene, neoprene, hypalon, plywood, PVC, and Kleenex. Contamination was applied to a sample of the test material placed above the standard PMMA fuel stack. No contamination was added to the PMMA itself. These experiments were conducted in the same fire room used for the work described above and involved using the standard 0.6 MW PMMA fire. In addition to the $\text{Ce}_{0.84}\text{Eu}_{0.16}\text{O}_x$ oxide powder used in those tests, a nitric acid solution of the rare earths was used as a contaminant for some of the tests. The releases observed from the solution contamination experiments were generally lower than those obtained from the oxide-powder contamination experiments. This is consistent with the results obtained by other workers.

The aerosol release results obtained for the contamination simulated by the rare earth oxides are summarized in Figure 12 for both noncombustible materials and combustible materials. The

Figure 12. Release of Contaminant Oxide Powder During Exposure of Test Materials to PMMA Fires

results are given in terms of the mass fractions of the original contaminant that were recovered on duct filters, fell into the fire residue, or remained on surfaces of the test materials, as measured by the researchers. According to our definition of ARF, the fraction of material that was not recovered in fire residue or on the surface of test material is considered as the airborne release. For the combustible materials, in nearly all cases, the majority of the contamination was dispersed in the fire room.

Aerosol particles produced from the solution-contaminated fires were much smaller than the aerosol particles from the fires contaminated with the oxide powder contamination: 76 percent of the contaminant particle mass produced in the solution-contaminated fire was smaller than 8 μm aerodynamic diameter, whereas approximately 37 percent of the particle mass produced in the powder-contaminated fires was smaller than 8 μm .

In a series of large-scale fire tests performed at Sandia National Laboratory (1979), fire-resistant plywood (FRP) boxes and 55-gal steel drums filled with simulated waste material were exposed to diesel fuel fires. The purpose of these tests was only to qualitatively observe the behavior of the containers exposed to the fires under several specific sets of conditions. No measurements of contaminant release fractions were made in this work.

In the first burn, 13 FRP boxes were stacked in an horizontal "L" configuration in a 6- x 6-ft square burn pan with a 2-in salt layer. Five 55-gal drums with polyethylene liners, but without close rings, were placed in the corner formed by the boxes. Fifteen gallons of diesel fuel were poured onto the salt bed and ignited. After 6 minutes, mild detonations were heard in the drums, and the lids lifted. The most intense combustion occurred at the corner where the boxes ignited.

In the second fire, 16 drums were stacked in four layers of 4 drums/layer. The relative positions of the drums and boxes were not well described for this test. Twenty gallons of diesel fuel was burned for 50 min. Again 5 to 8 min after ignition of the fuel, detonations were heard in the bottom two layers of drums. The lids were held in place by the upper drums. A hole was burned in one of the boxes. A peak temperature of 1375°F was recorded in the chimney-like gap between the drums and boxes. A similar temperature was observed in the first test at a similar location. The peak temperature rise in one box located in the bottom layer was 65°F.

In the third test, 12 drums were placed in a 3 x 4 arrangement on a 3-inch thick salt bed in a 10-ft-diameter stock tank. Fifty gallons of diesel fuel were poured onto the salt bed and ignited. Wind conditions during the burn confined the flames to only four of the drums. All four of these drums leaked. The lid of the unlined vented drum was blown off, and burning debris was scattered.

The lids of all three of the unlined drums exposed to the flames were blown off or lost their seal. Two additional unlined drums retained their seals, but these drums were not exposed to strong flames, since they were in the second row of drums. The lined drum exposed to the flames experienced a failure of the bottom seal, but retained its lid. The fire was extinguished after 45 minutes.

Several additional tests were performed to compare the behavior of lined and unlined drums. These tests were flawed, because the wind conditions always favored the lined drums (i.e., the wind blew the flames toward the unlined drums); however, the unlined drums did not retain their seals, and the lid of one drum was blown into the air.

Technical Discussion

The literature described above provides some information required for estimating the quantity of radioactive contaminant released during severe nuclear facility accidents that involve fires. The experiments were conducted using either actual plutonium or, more commonly, another substance as a surrogate for plutonium. Based on the overall consistency of the results, the selected surrogates appear to simulate plutonium releases reasonably well. In test series where several materials were burned, the highest release fractions generally occurred during combustion of PMMA or combustion of rubber materials.

Two distinct types of contamination release behavior were noticed in the experiments that measured release profiles during the tests. Materials that softened prior to ignition released the majority of the contamination prior to ignition. When a molten mass of fuel was formed by softening of the combustible material, release of both powder and solution contamination nearly ceased. This probably occurred because the soft fuel flows around the contaminant particles on the surface and adheres to them, thus holding them in place. Some experimenters noted that the plastic materials bubbled before they ignited (Halverson et al., 1987). This physical activity may be the cause of the high release fractions observed for these materials, i.e., ejection of contaminant particles by bursting bubbles.

Releases from cellulose-based material, such as wood and paper, were lower than releases from polymers under static (minimal additional air flow) burning conditions, and were more uniform in time. However, in tests that included strong upward air flows (Mishima and Schwendiman, 1970), releases from cellulose combustion exceeded the releases from all other fuels. Since cellulose materials represent the majority of typical waste mass (Mishima and Schwendiman, 1973-a), releases

from burning cellulose are expected to dominate contaminant release. This conclusion is supported by the observation that releases from mixed waste that included paper were similar to releases from combustion of cellulose alone (Halverson et al., 1987).

One possible explanation for this conclusion is that paper, the predominant form of cellulose waste, represents most of the waste volume. The other combustible materials are effectively embedded in a paper matrix. Any releases from the embedded combustibles are trapped by the paper until combustion of the paper occurs. Some contamination release from the paper occurs during charring of paper prior to flaming combustion, but the magnitude of release at this time is much lower than releases for polymers or flaming combustion of cellulose.

Based on data from the literature, the air flow velocity appeared to be an important parameter controlling the release of contaminant from burning cellulose (Mishima and Schwendiman, 1970; Halverson, et al. 1987). Figure 13 shows a plot of contaminant release fractions from cellulose materials versus air velocity constructed using available experimental data. Three linear regression lines were calculated using the log-log scale shown in Figure 13: one of them was obtained for all the contamination types combined, and two others were derived for the powder and solution contamination types individually. The releases shown in Figure 13 are much higher than the values commonly used to estimate plutonium releases during fires, even under moderate air velocities. For example, the value recommended by Mishima (1993) is 0.00053, which was derived from Mishima and Schwendiman (1973-a) for undisturbed burning of a simulated waste carton.

The regression formulas in Figure 13 are as follows:

$$ARF_{\text{powder}} = 0.2754 * 3.254^{\ln(u)} \quad (11)$$

and

$$ARF_{\text{salt}} = 0.01202 * 2.075^{\ln(u)} \quad (12)$$

where:

ARF_{powder} = Aerosol release fraction for powder contamination,
 ARF_{salt} = Aerosol release fraction for liquid or dried salt contamination, and
 u = Air velocity in m/s.

Equations (11) and (12) are purely empirical correlations, and are valid therefore only within the range of experimental data used to derive them. According to various experiments described

Figure 13. Effect of Air Velocity on Contaminant Release During Cellulose Combustion

above, a maximum value of $ARF = 0.5$ can be assumed for contaminant releases from burning cellulose materials under high updraft air flow rates. This value is therefore recommended as the upper limit of model applicability.

Equations (11) and (12) need to be combined with estimates of the air velocity developed near burning cellulose to predict the ARFs for the contaminated waste materials involved in the fire. This is addressed in the following discussion.

Release of a noncombustible contamination material from burning cellulose is largely controlled by the flow of gases around contamination particles. Two different mechanisms of air flow development caused by the fire are considered in this study: evolution of combustible gases by pyrolysis of fuel material (such as cellulose), and development of updraft air flow by natural convection. Although both these mechanisms appear to concurrently contribute to the releases of noncombustible contaminants, each of them can dominate the other depending upon a number of combustion conditions.

Pyrolysis of Cellulose Fuel

A probable mechanism for release of contamination particles during combustion of a contaminated solid fuel is transport of the particles by vapors released from the pyrolyzing solid exposed to the radiant heat of the fire. This radiant heating and attendant pyrolysis of the remaining fuel produces gaseous decomposition products that rise into the main reaction zone of the fire and are burned, generating additional heat. This cycle of heating, decomposition, and combustion, which generates more heat is a characteristic of fires.

To estimate the importance of fuel vaporization to contamination release from burning cellulose, an estimate of the vapor generation rate per unit area of the fire needs to be made. In this work, cellulose material is assumed to decompose at 150°C , which is roughly the boiling point of glucose (146°C). Chemically, cellulose resembles a polymeric material made from chains of glucose molecules (Morrison and Boyd, 1966). During combustion, the cellulose decomposes to produce combustible vapors, and we assume that glucose-like molecules are formed as a result of cellulose pyrolysis processes.

Based on ideal gas considerations, the flow velocity of cellulose decomposition vapors can be expressed as the following:

$$V = \frac{m''}{M} \cdot \frac{R T}{P} \quad (13)$$

where V is the gas velocity, m/s,
 m'' is the mass-burning rate of cellulose, kg/m²s,
 M is the molecular weight ($M=180$ atomic mass units for glucose),
 $R = 82.0567 \times 10^{-3}$ (atm · m³)/(kg-mole · °K) is the universal gas constant,
 T is the temperature, °K, and
 P is the pressure, atm.

In order to calculate the mass-burning rate of a combustible material, m'' , required in Equation 13, we address the following considerations in this work. Tewarson and Pion (1976) concluded, based on their experiments on flammability of plastics, that the burn rate of a combustible material can be represented as a sum of an "ideal" combustion rate, m''_{ideal} , and an additional term (either positive or negative) that accounts for heat exchange between the fuel and fire surroundings. They defined an "ideal" combustion condition where any additional heat flux transferred to the burning surface from external sources (Q''_E) is equal to the heat flux lost by the surface (Q''_L).

When a combustible substance burns in the presence of an external heat source (i.e., cellulose waste burning at the base of a large fire), the mass-burning rate of the substance is increased by external radiant heating. Radiant heating accelerates the pyrolysis of the substance, causing it to decompose more rapidly into combustible vapors. Tewarson and Pion (1976) give the following relationship between actual mass burning rate and fire conditions:

$$m'' = m''_{ideal} + \frac{(Q''_E - Q''_L)}{L_G} \quad (14)$$

where: m'' is the actual mass-burning rate (kg/m²/s),
 m''_{ideal} is the ideal mass-burning rate (kg/m²/s),
 Q''_E is the external heat flux (W/m²),
 Q''_L is the heat flux lost by the burning surface (W/m²), and
 L_G is the heat of gasification (J/kg).

Using an ideal mass-burning rate of a representative cellulosic material (Douglas fir) as 0.013 kg/m² · s (Tewarson and Pion, 1976), Equation 14 can be used to calculate the actual rate of

cellulose combustion in a fire. In order to use Equation 14, however, the net external heat flux also needs to be estimated.

The main component of heat transfer from flame to combustion surfaces is by radiation. The Stefan-Boltzmann law can be used to estimate the radiant heat exchange between two blackbody surfaces having respective temperatures of T_1 and T_2 :

$$Q_E'' - Q_L'' = \sigma \cdot (T_1^4 - T_2^4) \quad (15)$$

where $\sigma = 5.67 \times 10^{-8} \text{ W}/(\text{m}^2 \cdot (\text{°K})^4)$ is the Stefan-Boltzmann constant.

Then, assuming flame temperature above the combustion surface of approximately 1000°K, and using cellulose decomposition temperature of 423°K (150°C), a radiant heat flux of approximately $55 \times 10^3 \text{ W}/\text{m}^2$ is calculated for burning cellulose.

The heat of cellulose (Douglas fir wood) gasification, L_G , is taken as 435 cal/g ($1.82 \times 10^6 \text{ J}/\text{kg}$) according to Tewarson and Pion (1976). The actual mass-burning rate of cellulose is then calculated using Equation 14 as $0.04322 \text{ kg}/(\text{m}^2 \cdot \text{s})$.

This burn rate is next used in Equation 13 to calculate the velocity V of the combustible vapors evolved from cellulose by pyrolysis as $8.334 \times 10^{-3} \text{ m}/\text{s}$. At this velocity of combustible vapors released by the fuel, the ARF of plutonium oxide contamination is predicted using Equation 11 as $\text{ARF}_{\text{powder}} = 9.7 \times 10^{-4}$.

Mishima and Schwendiman (1973-a) measured release fractions from burning contaminated waste cartons ranging up to 5.3×10^{-4} , which was based on their measurements of airborne concentrations of the contaminant. However, as discussed above in this report, according to our definition of airborne releases, the ARFs reached in Mishima and Schwendiman's experiments a value of 2.3×10^{-3} if both the airborne concentration measurements and the material deposited on the RART walls were to be accounted for. Therefore, our predicted result for $\text{ARF}_{\text{powder}}$ being approximately 1.0×10^{-3} is generally consistent with the experimental results of Mishima and Schwendiman (1973-a), as well as experimental results of Halverson et al. (1987), both conducted under "static" burning conditions using no forced air flow.

Based on this analysis, it appears that a somewhat conservative value of 1.0×10^{-3} can be recommended as a release fraction of plutonium oxide contamination from cellulose and mixed-waste materials burning in an accidental fire. In some accident scenarios, however, combustion of these waste materials can be postulated to occur in the flames of a large fire, such as a jet-fuel fire resulted from crash of a commercial airplane. Under these conditions, high updraft air velocities will develop

around various contaminated materials involved in the plume, far exceeding the vapor velocity of decomposing combustible materials discussed above. Different considerations need to be addressed for predicting airborne releases under these conditions.

Combustion of Solid Wastes in Fire Plumes

As the temperature of combustion gases rises in a fire above ambient, causing density of the gases to decrease, an upward air flow develops due to natural convection. Above flames, the heat of combustion raises the temperature of the surrounding air, causing it to rise in a plume above the fire. In very large fires, such as forest fires and urban fire storms, inward surface winds of 10 to 40 m/s can occur (Pitts, 1991) as the surrounding air flows inward to replace the air rising in the plume. If a contaminated material burns in such a fire plume, high releases of the contaminant can be expected. The purpose of this analysis is to estimate the rate of convective air flow in a fire plume, so that ARFs of the contaminant could be predicted using Correlations 10 and 11 shown above.

Heskestad (1981) studied the updraft air velocity developed in fires under various combustion conditions. He derived a single parameter, ξ , combining several combustion variables, which appeared to remain constant in different locations throughout the plume. Based on his extensive analysis, Heskestad provided the following formula that can be used to estimate a local plume-rise velocity when the air-heating rate of the fire and temperature rise above ambient are known:

$$\xi = \frac{T_0^{0.4} (c_p \rho_0)^{0.2} u}{g^{0.4} (\Delta T Q_c)^{0.2}} \quad (16)$$

where: ξ is the dimensionless constant suggested by Heskestad as 2.2,
 T_0 is the ambient temperature ($^{\circ}\text{K}$),
 c_p is the heat capacity of air ($\text{J/kg}^{\circ}\text{K}$),
 ρ_0 is the density of air (kg/m^3),
 u is the plume rise velocity (m/s),
 g is the acceleration of gravity (m/s^2),
 ΔT is the temperature rise above ambient ($^{\circ}\text{K}$), and
 Q_c is the air-heating rate (W).

Considering the standard PMMA fire produced by Buijs et al. (1988), the fuel mass-burning rate was estimated as $0.042 \text{ kg}/(\text{m}^2 \cdot \text{s})$, resulting in the total energy release of $0.6 \times 10^6 \text{ W}$. If one fourth of this total energy release ($0.15 \times 10^6 \text{ W}$) is arbitrarily assumed to be convective energy from

the lower half of the fire, and the temperature rise in the center of the fuel stack is assumed to be 300°K, the plume velocity predicted by Equation (15) is 4 m/s. A velocity of this magnitude should be sufficient to cause emission of a large fraction of the contamination in the upper half of the fuel stack. Buijs et al. (1988 and 1992) reported that approximately half of the contamination remained in the fire residue, and the other half of the contamination was transported to the internal surfaces of the fire room. The calculations presented here appear to be consistent with the experimental observations. In two tests, in which Kleenex in cardboard containers was placed on top the PMMA fuel stack, 42 and 64 percent of the contamination was transported by the fire. In these tests, the top plate of the PMMA stack was 0.1 m below the Kleenex. The results of Buijs et al. (1992) for incombustible steel plates also demonstrate the fire's ability to remove contaminated particles from surfaces located in the turbulent gas flow above the fire.

Airborne Releases from Contaminated Incombustible Surfaces

Mishima et al. (1968-a) conducted a series of small-scale experiments the purpose of which was to measure aerosol formation from heating four dry plutonium compounds in bulk powder form: plutonium oxide, plutonium oxalate, partially oxidized oxalate, and plutonium fluoride. In their tests, a sample of test powder was heated by conduction in a shallow dish to a temperature of up to 1000°C. An upward air flow was provided around the dish at up to 100 cm/s velocity. Airborne particles were collected on a membrane filter and the walls of the test apparatus used. The amount of airborne releases was presented in terms of airborne release rates - ARR (fraction/hour). The highest release rate observed was 9.0×10^{-3} 1/hr for a plutonium oxalate sample heated to 700°C at 100 cm/s air velocity, and the highest ARR for plutonium oxide powder was 5.6×10^{-3} 1/hr measured at ambient temperature under 110 cm/s air velocity.

Based on their experiments, Mishima et al. concluded that the temperature of test powders had little effect on ARRs, if any, and the release rates measured were generally consistent with the mechanism of powder resuspension from solid surfaces.

A review of Mishima et al. (1968-a) experimental setup suggests that air velocity over the surface of bulk powder, although not known, was probably low. The re-entrainment of particles from bulk powder was likely caused by a vortex flow developed over the test dish as the air streamed around it. Therefore, the results obtained in their experiments are applicable to a specific accident

scenario, such as exposing an open container containing a powder of plutonium salt or oxide to a relatively low wind.

Buijs et al. (1992), as discussed above in this report, used their standard 0.6 MW PMMA fire to study airborne releases from incombustible surfaces contaminated with $\text{Ce}_{0.84}\text{Eu}_{0.16}\text{O}_x$ oxide powder. They used stainless steel and aluminum specimens, both metallic and painted, in the study. The amount of the simulated contaminant remained on the surfaces of test materials or fell into combustion residue after the fire ranged from 0.23 to 0.90, meaning that up to 77 percent of the contaminant was once aerosolized during the fire. Results of Buijs et al. (1992) work were shown above in Figure 12 together with the results obtained for burning combustible materials in their standard test fire.

According to our analysis of the PMMA fire that Buijs et al. used in their experiments, an upward air velocity of at least 3 m/s developed in the fire plume where the samples were exposed directly to the flames. In large fires, such as in the MCA scenario, much more intensive air currents are expected. It appears reasonable therefore to use an approximate ARF value of 0.5 for both combustible and noncombustible solid materials contaminated with plutonium compounds, directly involved in an accidental fire.

Recommendations

Airborne Release Fractions

Recommended ARFs and RFs are presented below for the following contaminated combustible solid materials involved in an accidental fire:

- (1) Polymethylmethacrylate
- (2) Polystyrene resin
- (3) Rubber gloves (polychloroprene or latex)
- (4) Paper (cellulose) waste
- (5) Mixed waste

The gas generation velocity calculated for cellulose under static burning conditions, combined with the contaminant release versus air velocity relations, Equations (11) and (12), appear to yield reasonable estimates for contamination release during cellulose combustion. For the plastic materials, such as PMMA, polychloroprene and polystyrene, which produce bubbles and become soft before

they ignite, the contamination release observed in experimental fires under static burning conditions are higher than ARFs observed for cellulose burning under similar conditions. In the combustion of these materials, an additional release mechanism, possibly related to bursting of bubbles at the material surface, may cause increased contamination release. When the contaminated material is located in the convective plume of a fire below the combustible material, approximately half of the contamination on the combustible material is transported by the plume. The recommended aerosol release fractions are shown in Table 9.

Table 9. Plutonium Release During Combustion of Contaminated Solid Materials

Combustible Material	ARF		RF
	Dry Powder	Solution or Salt	
Rubber	0.010	0.040	1.0
Polystyrene	NA	0.008	1.0
Polymethylmethacrylate	0.050	0.020	1.0
Cellulose and mixed waste materials (u = air velocity [m/s])	$0.2754 * 3.254^{\ln(u)}$ (max 0.5)	$0.01202 * 2.075^{\ln(u)}$ (max 0.5)	1.0
Mixed waste in unlined 55-gal drums	0.500	0.500	1.0

Releases derived for cellulose combustion are also used for mixed waste combustion, since the experiments show that mixed waste behaves similarly to cellulose during combustion. Using the results of the Sandia fire experiments, it is recommended that contamination in unlined 55-gal drums that also contain combustible material should be considered to have released all of their contents if drums are exposed to flames from spilled fuel or other combustible materials. Unlined drums not exposed to the flames and lined drums are expected to maintain their seal.

Respirable Fractions

The work of Buijs et al. indicates that dry oxide powder contamination is released as a separate aerosol component during PMMA fires, rather than attached to soot particles. The particle-size distribution of the aerosolized contamination is thus expected to resemble the size distribution of the contaminant powder before combustion. Since plutonium oxide powders are typically of respirable

size, the recommended respirable fraction is 1.0 for cases in which the powder-size distribution is not known. Halverson et al. (1987) obtained an RF value of 0.16 for powder contaminated UO_2 on polychloroprene; however, the size distribution was determined for the total aerosol mass, and not for the contaminant particles alone. The particle-size distribution data for liquid or salt-contaminated solid combustible materials indicate that 70 to > 90 percent of the released contamination is of respirable size. The recommended respirable fraction for all solution or salt-contaminated combustible solids is 1.0, because the experimental data demonstrate that very small contaminant particles are generated during combustion of liquid or salt-contaminated combustible solids.

PLUTONIUM RELEASE DURING LIQUID FUEL COMBUSTION

Introduction

The third category of contamination release considered in this report is associated with heating or combustion of contaminated liquids. These liquids include process solutions of nitric acid containing plutonium, and plutonium contaminated organic liquids, such as kerosene, generated during liquid-liquid extraction processes. Chemical processes are used to separate plutonium from uranium and fission products produced in the nuclear reactor. During the course of processing, many liquids become contaminated with plutonium. Some of these liquids are non-flammable, such as nitric acid, while other liquids are flammable, such as kerosene or mixtures of kerosene and tributyl phosphate (TBP). After use these process liquids must be stored at the facility until they can be disposed of. Contaminated liquids are a potential source of airborne radioactive contamination. If a fire occurs in the liquid waste storage area, contamination may be released to the environment through evaporation or combustion of contaminated liquid wastes.

Contaminated liquid wastes are currently being converted to solid wastes at Rocky Flats. As elimination of the liquid wastes proceeds, potential for airborne releases from burning liquid wastes will become less significant. However, radioactive releases associated with combustion of kerosene-like flammable liquids, such as during a jet-fuel fire caused by a crash of an airplane, will continue to be of significant concern.

Literature Review

Releases of radioactive contamination produced by evaporation of plutonium nitrate solutions were studied in three series of experiments by Mishima et al. (1968-b). In the first series of their study, plutonium nitrate solutions were evaporated in a sealed apparatus producing aerosol particles. An adjustable air flow swept the particles through a condenser and a filter. The quantity of plutonium in the apparatus, in the condensate and on the filter was measured. In one series of experiments, air passed horizontally over a pool of plutonium nitrate solution. The fraction of contamination collected for three air flow rates, 0.1, 0.5 and 1.0 m/s, is shown in Figure 14. Air temperatures ranged from ambient to 100°C. The liquid surface remained calm during these experiments; therefore, the releases observed were due to evaporation of the liquid rather than due to bubble burst mechanisms. The fraction of contamination collected in the condensate and filter increased with increasing air temperature; however, the fraction of contamination in the apparatus wash did not correlate with the air temperature.

In the second series of experiments, Mishima et al. (1968-b) determined the fraction of contamination released from boiling plutonium nitrate solutions. In these tests, the air flowed vertically upward past a heated beaker containing the boiling solution. As in the previous experiments, the condensate and backup filter were analyzed for plutonium. Boiling continued until 10% of the initial solution volume remained in the beaker. The fraction of plutonium released in these experiments was a function of the boil-off rate of the solution. The boil-off rate was expressed as a fraction of the solution volatilized per unit surface area (cm^2) per minute. The release fraction peaked at 0.02 for a boil-off rate of approximately $0.0011 \text{ l}/(\text{cm}^2 \cdot \text{min})$. The airborne release fractions obtained in these experiments are shown in Figure 15.

The plutonium detected on the filter was termed the airborne contamination. The total of the airborne contamination from the filter and the contamination washed from the interior of the apparatus was designated as the recovered contamination. Much of the contamination inside the apparatus was released by bursting of vapor bubbles during boiling. An empirical correlation, developed using the boil-off rates of the experiments, is given in Equation (17):

Figure 14. Pu Release During Air Drying of Pu Nitrate Solutions (Mishima, et al., 1968)

Figure 15. Pu Nitrate Evaporation at 100 - 220°C

$$\log(ARF) = -13.38 + 22070 \cdot B - 1.043 \cdot 10^7 \cdot B^2 \quad (17)$$

where

ARF is the airborne release fraction of the contaminant, and

B is the boil-off rate of the solution (fraction/cm²min).

Equation (17) can be used to estimate the ARF at boil-off rates between 0.0004 and 0.0011. According to these tests, a constant ARF value of 0.02 can be recommended for boil-off rates above 0.0011 1/cm²min.

In the third series of experiments, Mishima et al. (1968-b) determined the fraction of contamination released from the dry residue remaining after evaporation of plutonium nitrate solutions. In these experiments, dry plutonium nitrate decomposed forming plutonium oxide particles and releasing brown fumes of nitrogen oxides. Residues from air-dried plutonium nitrate solutions were heated to high temperatures (400°C to 1000°C) in the same apparatus they used to study airborne releases from heated plutonium bearing powders (Mishima et al., 1968-a). The residue was placed in an alundum thimble and heated to the test temperature with an induction heater. A vertical air flow having velocities ranging from 10 cm/s to 100 cm/s swept particles from the residue to the collection filter. Mishima et al. (1968-b) noted some correlation between plutonium ARF and the upsweep air velocity, which was done, as before, by considering only the fraction of airborne releases that was collected on the filters.

In this report, according to our definition of ARF, plutonium nitrate fractions collected on the filter and on the interior surfaces of the apparatus were combined to yield the airborne release fraction. The ARFs thus obtained are shown in Figure 16, from which no reliable correlation between ARF and the two process variables studied in Mishima et al. (1968-b) work can be identified. Therefore, a constant value ARF = 0.0007, which is equal to the upper 95% confidence limit of their data is recommended for the releases from heated residues from solidified plutonium salts.

The particle size distribution of airborne releases was obtained in Mishima et al. (1968-b) study for only one test, where the highest release fraction (ARF = 0.00152) was observed using plutonium nitrate residue. The particle size distribution measured by microscopy examination is shown in

Figure 16. Fractional Release from Solids Remaining After Evaporation of Plutonium Nitrate Solutions (Mishima, et al., 1968)

Figure 17. According to this Figure, a conservative value of $RF = 1.0 \times 10^{-5}$ can be recommended. Mishima, et al. noted however that the residue used in this test had a different appearance than the residues used in the other experiments. Based on the color of the residue, they suggested that the residue used in the high release test was plutonium oxide of small particle size rather than plutonium nitrate. This test therefore may not be representative of releases from heating nitrate residues. In other tests performed in this series, aerosol particles detected on the filters ranged from approximately $20 \mu\text{m}$ to up to $125 \mu\text{m}$ in their geometric sizes.

Mishima and Schwendiman (1973-b) described results of 21 laboratory scale experiments performed to measure the fractional airborne release of uranium during combustion of 30% n-tributyl phosphate in kerosene. The test solutions used also contained small quantities of cesium, cerium, zirconium and iodine to simulate a PUREX process solution. A 25 ml aliquot of n-TBP-kerosene mixture was heated to $\sim 90^\circ\text{C}$, then ignited. Combustion products were collected in a 2.7 inch diameter chimney assembly placed over the burning solution. Air was drawn through the chimney at 1 and 2 cubic feet per minute to filters located at the top of the chimney. The quantity of uranium collected on the filters and deposited on the chimney walls was measured. The mixture burned with a smoky flame until self-extinguishing after 42 to 56 minutes, leaving 4 to 9 ml of syrupy residue in the beaker. For some experiments the residue was heated with a propane torch until the residue was charred. No particle size measurements were performed in this work.

Releases of uranium appeared to be low in these tests. In average, the highest release, 2.7×10^{-4} , occurred for uranium at a 1 cfm flow rate. Air flow rate did not significantly affect contaminant releases during combustion. Charring of the combustion residue, however, produced an approximately 10 fold increase in the uranium releases, resulting in an average ARF of 3.0×10^{-3} .

Sutter et al. (1974) reported results for two experimental tributyl phosphate - kerosene fires. Strontium nitrate was used as the contaminant. An almost saturated solution of strontium nitrate in 100 ml of water was mixed with 150 liters of the TBP - kerosene mixture. Experiments were conducted in the ARHCO Fire Test Facility where combustion occurred in a 12' x 12' cell. Combustion gases flowed through a 1.5' x 2' exhaust duct, through a water spray cooling system, and to a filter test section. Particulate samples were obtained upstream and downstream from the water spray cooling system. The strontium ARF obtained in the first test was 0.0022. The strontium ARF in the second test was 0.0019. These results were obtained from the samplers located upstream from the water spray cooling system. The downstream samples contained much less contamination,

Figure 17. Size Distribution of Particles Released from Pu Nitrate Residue (Highest-Release Test) (Mishima et al., 1968-b)

because the water spray removed the contamination from the air stream. The particle size distribution was not measured in these experiments.

In an extensive series of experiments, Seehars and Hochrainer (1983) determined ARFs from plutonium oxide and cerium oxide powders and plutonium nitrate solutions exposed to kerosene fires contained in a glovebox. Three experimental configurations were used in the cerium oxide powder tests:

- 1) Dry test powder in a 5 cm metal dish placed 10 to 12 cm above the fire,
- 2) Powder as above, but wet with 1 ml of kerosene, and
- 3) Powder immersed in burning kerosene.

The respirable aerosol release fractions having particles with the aerodynamic diameter less than 10 μm were measured. An elutriator system designed to separate particles greater than 10 μm was used to remove the large particles from the air stream. To determine the respirable fraction of the dust released, the respirable particles that passed through the elutriator were collected on a filter and analyzed for contaminant content by gamma spectroscopy. In two of the tests, the total airborne release fractions were measured as well. The ARFs and their respective RFs calculated for these experiments are shown in Table 10.

The results of two series of TBP - kerosene combustion tests were reported by Malet et al. (1983). These experiments were conducted in both a small scale (316 liter) and a large scale enclosure (400 m^3). Either cerium or thorium was used as the heavy metal contamination. In these tests, the amount of liquid fuel consumed in the fire was limited by the quantity of oxygen in the test chamber. Combustion stopped when the O_2 concentration dropped to between 12 and 15%. Within each test series, the amount of fuel consumed was similar – 18 ± 4 ml in the small scale tests and 12.5 ± 2.5 kg in the large scale tests. The results presented by Malet et al. were expressed in terms of a transfer coefficient, TC, defined as follows:

$$TC = \frac{C_i V_i - C_f V_f}{C_i (V_i - V_f)} \quad (18)$$

where C is the mass concentration of the contaminant,
V is the volume of the test solution, and
indices i and f symbolize the initial and final test parameters, respectively.

Table 10. Contaminant Release Parameters (Seehars and Hochrainer, 1983)

Contaminant	Total Aerosol Release Fraction		Respirable Aerosol Release Fraction		Respirable Fraction
	15 min Release	25 min Release	5 min Release	25 min ^(a) Release	25 min Release
Dry powder ^(b)	--	--	5.2×10^{-5}	--	--
Wet powder ^(b)	0.00188	0.00314	4.4×10^{-5}	2.2×10^{-4}	0.07
Immersed ^(b) powder	0.01217	0.02025	1.6×10^{-3}	8.0×10^{-3}	0.40
Pu(NO ₃) ₄ in nitric acid	--	--	2.6×10^{-4}	--	--
Dry powder ^(c)	--	--	1.96×10^{-4}	--	--
Wet powder ^(c)	--	--	2.12×10^{-4}	--	--
Immersed ^(c) powder	--	--	2.73×10^{-4}	--	--

Note: (a) The 25 minute release is estimated to be 5 times the 5 minute release.
 (b) Cerium oxide contaminant
 (c) Plutonium oxide contaminant

Since kerosene was more volatile than the dissolved contamination, the concentration of the contamination increased in the liquid pool during the fire. The final contaminant amount was thus greater than it would have been if the kerosene and contamination were consumed at the same rate. For the small scale tests, the transfer coefficients were converted to ARFs using the information presented in Malet et al. (1983) report. The transfer coefficients and ARFs calculated on their basis are given in Table 11. The initial quantity of combustible liquid was not given for the large scale tests, so ARFs could not be derived. Aerosol size distributions were not given for the small scale tests. The aerosol particles produced in the large scale experiments were all respirable.

Table 11. Aerosol Release Parameters from Combustion of Contaminated TBP - Kerosene Mixtures (Malet et al., 1983)

Contamination	Initial Combustible Volume (ml)	Volume Consumed (ml)	Transfer Coefficient	Calculated Aerosol Release Fraction
Ce	157	21	0.47	0.063
Ce	78.5	20.5	0.32	0.084
Ce	39	19	0.27	0.132
Ce	20	14.6	0.49	0.358
Th	157	16	0.45	0.046
Th	79	22	0.23	0.064
Th	39	20	0.12	0.062

Halverson et al. (1987) performed a series of experiments to measure contamination releases from pools of organic liquids floating on nitric acid. They examined several combinations of acid and organic liquids and varied the type and location of the contamination. The experimental configurations are shown in Table 12.

Table 12. Aqueous - Organic Liquid Experimental Configurations (Halverson et al., 1987)

Test Configuration	Acid Composition	Organic Liquid Composition
1	Pure nitric acid	30% TBP in kerosene + uranium
2	Nitric acid + fission products	30% TBP in kerosene + uranium
3	Nitric acid + fission products + uranium	Organic ^(a) + uranium
4	Nitric acid + fission products + uranium	Pure 40% TBP in kerosene

Note: (a) Composition not specified.

The experimental configuration used by Halverson et al. (1987) was similar to the configuration employed in their solid combustion tests, described elsewhere in this report. In these experiments, 100 ml each of nitric acid and organic liquid was contained in a metal or glass beaker. During combustion, heat was transferred, via the beaker walls, to the acid liquid layer below the burning organic liquid. The acid boiled out of the glass beaker within 10 to 20 minutes and extinguished the fire. Reducing the external heating or substituting a glass beaker for the metal one, delayed the boil over but did not prevent it. Boilover quenched the fire after 40 to 60 % of the organic liquid was consumed. The size distribution of the released aerosol was measured in one test in which the RF was 0.99. The release results obtained in these experiments are described below in more detail. The experimental results of Halverson et al. (1987) will be presented in a series of graphs of increasing complexity. Each of the test configurations will be considered in turn.

The ARF's for the first configuration where a uranium-contaminated TBP-kerosene mixture was burning over pure nitric acid are reproduced in Figure 18. The maximum ARF was 0.00557. All three experiments yielded similar release values.

When fission products were added to the nitric acid, the ARFs increased by a factor of 5, as shown in Figure 19. Halverson, et al. suggested that the presence of fission products in the acid changed the liquid-liquid partition coefficient to reduce the solubility of the uranium contamination in the acid. Analysis of the remaining acid after combustion, however, did not support this hypothesis, because the concentration of uranium in the acid residue was the same following the tests with pure acid and acid with fission products added. Since temperature may also alter the partition coefficient, it is possible that the presence of fission products in boiling nitric acid reduce the solubility of uranium, resulting in the increased ARF.

Aerosol releases for the third test configuration are reproduced in Figure 20. Addition of both uranium and fission products to the acid reduced the contamination releases during combustion. Halverson et al. noted the reduced release in this configuration relative to the configuration in which the acid did not contain uranium initially, but attributed the results to experimental variation.

The tests using uncontaminated organic liquids produced the most unusual results. As shown in Figure 21, combustion of pure organic liquid over nitric acid contaminated with both uranium and fission products produced the lowest ARF. In this test, the organic liquid did not burn to dryness; however, when the organic liquid was burned to the incombustible TBP residue, the highest ARFs observed in this test series were obtained. The curves for the tests that burned to dryness are added in Figure 22. A glass beaker was used to contain the liquids in the test that gave the lowest release.

Figure 18. U Releases from Burning Contaminated Liquids (Test Configuration 1)
(Halverson et al., 1987)

Figure 19. U Releases from Burning Contaminated Liquids (Test Configuration 1 & 2)
(Halverson et al., 1987)

Figure 20. U Releases from Burning Contaminated Liquids (Test Configuration 1 & 2 & 3)
(Halverson et al., 1987)

Figure 21. U Releases from Burning Contaminated Liquids (Test Configuration 1 & 2 & 3 & 4)
(Halverson et al., 1987)

Figure 22. U Releases from Burning Contaminated Liquids (All Test Configurations)
(Halverson et al., 1987)

The data in Figure 22 indicate that the rate of contamination release falls into two groups. The high release rate tests coincided with the high total release tests, but the burn durations were similar to the burn durations of tests that yielded low release rate values. The high release rate was approximately 7.3×10^{-4} 1/min. The low release rate was approximately 1.2×10^{-4} 1/min.

Technical Discussion

The information obtained from the literature sources described above will be examined in this section of the report. The following basic experimental measurements were obtained:

- 1) Aerosol releases from heating contaminated aqueous solutions
- 2) Aerosol releases from boiling contaminated aqueous solutions
- 3) Aerosol releases from burning powder-contaminated organic liquids
- 4) Aerosol releases from burning solute-contaminated organic liquids

The lowest contamination aerosol releases were observed from the heating of contaminated aqueous solutions. ARFs remained below approximately 1.0×10^{-4} for all of the experimental conditions. These results appear to be reasonable given the low volatility of the nitrate salts used for these tests. During boiling of the solutions, additional release mechanisms can become active. When vapor bubbles rise through the boiling liquid, the surface area of the bubbles increases the effective surface area of the liquid-vapor interphase. The greater surface area allows more contamination to cross from the liquid phase into the vapor phase and, when the bubbles reach the liquid surface, any vapor they contain is released into the air. Additional mechanisms can also take place when the liquid film forming the surface of the bubbles disintegrates, resulting in higher airborne releases of the contamination. The high effectiveness of the bubble burst release mechanism is demonstrated by observing ARFs from boiling aqueous liquids as compared to the releases obtained from their heating. ARFs observed for strongly bubbling aqueous liquids are up to 3 orders of magnitude greater than ARFs obtained for simmering aqueous liquids. The recommended values for ARF and RF during heating or boiling of contaminated aqueous solutions are shown in Table 13.

Figure 23. Contaminant Release Fractions from Burning Contaminated Liquids

Table 13. Recommended Contaminant Release Parameters for Heating or Boiling Contaminated Aqueous Liquids and Liquid Residues

Heating Conditions	ARF	RF
Simmering (Quiescent surface)	2.0×10^{-4}	0.5
Boiling (Disturbed surface)	Equation (19)	0.5
Heating dried Pu nitrate residue	0.0007	1.0×10^{-5}

$$\log(ARF) = -13.38 + 22070 \cdot B - 1.043 \cdot 10^7 \cdot B^2 \quad (19)$$

where B is the boil-off rate of the solution (fraction/cm²min). A constant ARF value of 0.02 is recommended for boil-off rates above 0.0011 1/cm²min.

The maximum ARFs obtained in each of the experimental studies involving combustion of contaminated liquids are presented in Figure 23. The maximum ARFs span a range of 3 orders of magnitude. Seehars and Hochrainer (1983) datum represents the release fraction for only the respirable particles rather than the total aerosol release fraction shown for the other workers. Also, Seehars and Hochrainer's value only represents the respirable releases obtained over a 5 min time interval, while the release values given by the other researchers were measured for the entire duration of the test.

The greatest releases were obtained by Halverson, et al. (1987) and by Malet, et al. (1983). The releases observed by Sutter, et al. (1974) were similar to the releases observed by Mishima and Schwendiman (1973-b) in the experiments in which the organic liquid was burned to dryness.

Based on this analysis of the available data base, the recommended ARF for contaminated combustible liquids is 0.10. This ARF bounds all of the experimental release values except two values obtained by Malet, et al. (1983) in a study where up to 30% of cerium oxide was released from burning contaminated TBP-kerosene mixtures. The cause of the high results observed by Malet, et al., is not known, but appears abnormally high when compared with the results obtained by the other researchers.

Very little information on aerosol size distributions was obtained from the literature for contaminant releases from combustible liquids. Based on the size distributions measured by

Halverson et al. (1987) and Malet et al. (1983), the recommended respirable fraction of the contaminated aerosol generated by combustion of TBP-kerosene mixtures is 1.0.

The experiments performed by Seehars and Hochrainer (1983) comprise the only measurements of release of powder contamination during combustion of kerosene. Based on their data, the recommended ARF is 0.02 which was obtained for a contaminant powder immersed in burning kerosene. For this combination of contamination and combustible liquid, the recommended RF is 1.0 as well, or the respirable fraction of the source powder if that is known. The recommended ARF and RF values during combustion of contaminated process liquids are shown in Table 14.

Table 14. Recommended Contaminant Release Parameters for Burning Contaminated Organic Liquids

Contaminated Material	ARF	ARR	RF
TBP-kerosene (Dissolved contaminant)	0.10	0.001/minute	1.0
Kerosene (Powder contaminant)	0.02	0.001/minute	1.0 ^(a)

Note: (a) Use the RF of the contaminant powder, if known.

SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS

Open literature and technical reports covering airborne releases of plutonium and plutonium-simulating contaminants during combustion of radioactive metals themselves and during fires in processing and waste storage facilities were reviewed. Relevant material published after 1960 was examined utilizing a computerized literature search using the NTIS and DOE Energy Data Base, as well as numerous technical references and several private communications.

Examination of the literature has identified three major mechanisms of contamination release during a severe accident at nuclear facilities:

- 1) Release by oxidation of plutonium metal,
- 2) Release by combustion of plutonium contaminated solid waste materials stored in the facility, and

- 3) Release by combustion of plutonium contaminated flammable process liquids, or evaporation of contaminated non-flammable liquids.

Based on the information gathered in this study, airborne releases from oxidation of metallic plutonium were grouped into three general categories. These categories include:

- Plutonium oxidation at a range of temperatures not exceeding the plutonium ignition point,
- Exothermic combustion of plutonium leading to its melting, and
- Explosive-like dissemination of plutonium resulting in formation of airborne plutonium particles and their subsequent violent combustion.

Combustion of contaminated solid waste materials appears to present a significant contamination hazard, because of the quantity of contaminated wastes stored in nuclear facilities, and because of the large fraction of contamination that can be released during a solid waste fire. Experiments have shown that up to 50 percent of the radioactive contamination can become airborne during combustion of a typical waste material, if the material is burning in the convective plume of the fire. A large fire may also cause bursting of waste storage drums exposed to direct flames.

Airborne release parameters are presented in this report for the following contaminated combustible solid materials involved in an accidental fire:

- Rubber (polychloroprene or latex)
- Polystyrene resin
- Polymethylmethacrylate (PMMA)
- Paper (cellulose) and mixed waste
- Mixed waste in 55-gal drums

For the plastic materials, such as PMMA, rubber, and polystyrene, which produce bubbles and become soft before they ignite, the contamination ARF under static burning conditions (low air flow) is higher than releases from cellulose burning under similar conditions. This is probably related to bursting of bubbles at the surface of plastic materials. When the contaminated material is located in

the convective plume of a fire, approximately half of the contamination on the combustible material may be transported by the plume.

Releases from burning cellulose is largely controlled by the flow of gases around contamination particles. Two different mechanisms of air flow development caused by the fire were considered: evolution of combustible gases by pyrolysis, and development of updraft air flow by natural convection. Computational procedures needed to evaluate flow velocities are suggested in this work. These procedures can be used to estimate the ARF for different scales of a fire. However, this methodology should be validated.

Releases derived for cellulose combustion are also recommended for predicting the ARF for mixed wastes.

Based on the very limited information available, it is recommended that unlined 55-gal drums containing combustible material should be considered to have released all of their contents if drums are exposed to flames from a large fire. Unlined drums not exposed to the flames and lined drums may maintain their seal.

All releases from burning contaminated solid materials are conservatively assumed to be in the respirable size range. However, since plutonium oxide contaminant is a refractory material unaffected by fire, its original RF, if known, should be used.

Potential releases from flammable liquids are less significant, because of the lower releases expected, and because work is currently in progress at Rocky Flats to convert liquid wastes into solid form. Nevertheless, the risk of contaminant releases associated with combustion of kerosene-like flammable liquids, such as during a jet-fuel fire caused by a crash of an airplane, is of concern.

Based on results of this study, recommendations on airborne release parameters were formulated for emergency planning for the Rocky Flats Plant. The release categories addressed include such categories as the release of airborne plutonium from burning powder-contaminated combustible liquids, and releases from contaminated solid materials located in the fire plume. For the last category, up to 50% of the contaminating plutonium may be released into the environment. With the potential for such high releases, it is essential to carefully consider the details of any postulated accident scenarios. Consideration must be given, for example, to the effect of the fire on storage containers. Use of the correlations developed in this report requires an estimate of fire convection velocities and the degree to which the plutonium inventory is exposed to these. If the release fractions expected from the accident were low, such a careful analysis of the progression of the accident would be less important.

Also, with the potential for higher release fractions, uncertainties associated with any release parameter become extremely important. As shown in this work, major uncertainties are associated with the considerable amount of scatter among the reported ARFs and RFs. Much of this scatter is due to the experimenters' desire to investigate a variety of test conditions in a limited-scope study. These variables include use of plutonium or a simulant, chemical form of the contaminant, its size distribution, its amount and application procedure, type of combustible material, flow conditions, and many others. Even when experimenters took great care to achieve reproducible fire conditions, order of magnitude variations in contaminant release were sometimes observed.

The physical arrangement of the contamination and combustible material can also affect the observed ARF. Contamination on the underside of the combustible material is released at a lower rate than contamination on top. This is consistent with the observed effect of gas velocity on contaminant release. In an actual accident, many contaminant-fuel arrangements are expected to occur. Some of these arrangements will reduce the amount of airborne contaminant, while other arrangements will cause increased contaminant release.

The behavior of waste materials stored in 55-gallon drums is very uncertain. Both the location of a drum during an accident and the presence of a liner inside the drum can affect the release expected for that drum. Drums that remain sealed are expected to release negligible contamination, but a drum that explodes has a potential for releasing half of the contained contamination.

The health effects of the released contamination are strongly dependent on the size distribution and composition of the aerosol. Respirable contaminated particles present the greatest hazard to the surrounding population. Aerosol generated from solutions tends to be mostly respirable, and a conservative estimate requires a relatively large respirable fraction for this contamination form. In many experiments involving combustible materials, the mass distribution of the released aerosol (contaminant plus soot) was measured. This may not provide correct information regarding the size distribution of the contaminant aerosol. If the soot particles produced by the fire are large, as they were for the polychloroprene material in the study by Halverson et al. (1987), where a mass median aerodynamic diameter of $19.9 \mu\text{m}$ was observed, the mass distribution is dominated by the uncontaminated portion of the aerosol. If the contamination aerosol exists as a separate population of particles, as indicated by Buijs et al. (1988), the size distribution of the contamination may be respirable, but may be hidden by the combustion aerosol in measurements.

The conclusion of this work is a set of procedures for estimating release fractions for plutonium in the form prevalent at the Rocky Flats Plant. These procedures require a detailed description of the

accident scenario under consideration. Since to the best of our knowledge, this task was not adequately addressed for RFP, and since the development of such a description was beyond the scope of this work, there exists the need to formulate a technically justifiable progression of MCA in order to be able to apply these procedures sensibly.

There also appears to be a need for resolving several uncertainties presented above in this report. The most important uncertainties are those that impart the applicability of the high ARF observed by the researchers under various test conditions. These include:

- 1) Release of plutonium during combustion of contaminated cellulose and mixed waste, and
- 2) Release behavior of 55 gallon steel drums exposed to a fire.

With the decomposition of contaminated cellulosic materials exposed to elevated temperatures, the contaminant becomes "locally airborne" due to the disappearance of the substrate. Depending upon the air flow that develops around the contaminant, ARFs have been measured to range from approximately 1.0×10^{-4} to as high as 0.5. Predictions in this report of ARFs using estimated rates of cellulose pyrolysis and fire plume flows resulted in reasonable agreement with the experimental measurements. The procedure used to make these predictions needs to be validated to be used in plutonium release analyses with adequate certainty.

The 55 gallon steel drums containing plutonium contaminated waste materials will develop high initial pressure conditions due to the evolution of material decomposition gases and adiabatic heating. As the pressure exceeds some value, the drums will lose their seals causing either continuous leak of the contaminant or an explosive-type release. The latter mechanism can result in burning the contaminated content of the drum in the flames and a potential for high release. At the same time, providing a technically justifiable credit for the storage drums can significantly reduce ARFs expected from an accident. Therefore, an improved knowledge of the release behavior of the containers involved in a fire is needed in order to more accurately and less conservatively estimate the release of plutonium from the drums.

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