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NOT MEASUREMENT SENSITIVE

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DOE HANDBOOK

AIRBORNE RELEASE FRACTIONS/RATES AND RESPIRABLE FRACTIONS FOR NONREACTOR NUCLEAR FACILITIES

Volume I - Analysis of Experimental Data

U.S. Department of Energy
Washington, D.C. 20585

AREA SAFT

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FOREWORD

1. This Department of Energy (DOE) Handbook is approved for use by the DOE and its contractors as an information source.

2. Beneficial comments (recommendations, additions, deletions) and any pertinent data that may be of use in improving this document should be addressed to: Director, Office of Engineering, Operations, Security and Transition Support (DP-31, GTN), U.S. Department of Energy, Washington, D.C., 20585.

3. The issue of airborne releases of radioactive material from nonreactor nuclear facilities has been a subject of investigation for almost four decades, during which time a large number of individuals have contributed to the current knowledge base. Beginning in the 1960's, a number of experiments were conducted in the United States and other countries to develop actual data on release potentials. In the late 1970s, the United States Nuclear Regulatory Commission (NRC) sponsored a research program to develop improved methods for realistically evaluating the consequences of major accidents in nuclear fuel cycle facilities. This program culminated in the development of NUREG-1320 (5/88), "Nuclear Fuel Cycle Facility Accident Analysis Handbook."

The U.S. DOE began placing an increased emphasis on environmental, health, and safety issues in the mid- to late-1980s. In response to these efforts, the DOE Office of Defense Programs (DP) sponsored the Defense Programs Safety Survey (11/93). One of the objectives of this study was to build upon previous work to "develop consistent data and methodologies for making conservative estimates of basic consequence derivation parameters." As part of this effort, experimental data for airborne release fractions and respirable fractions were summarized and evaluated to estimate reasonably bounding values for physical stresses associated with the experiments. The unique and valuable nature of that compilation has been judged to merit further development as a handbook that can be directly used by technical analysts.

4. This handbook contains (1) a systematic compilation of airborne release and respirable fraction experimental data for nonreactor nuclear facilities, (2) assessments of the data, and (3) values derived from assessing the data that may be used in safety analyses when the data are applicable. To assist in consistent and effective use of this information, the handbook provides:

- Identification of a consequence determination methodology in which the information can be used;

- Discussion of the applicability of the information and its general technical limits;

- Identification of specific accident phenomena of interest for which the information is applicable;

- Examples of use of the consequence determination methodology and airborne release and respirable fraction information.

It is acknowledged that the data examined in this handbook is limited to that available during the preparation process. Other data may exist or be developed, and individuals are invited to submit such material for consideration for any future revisions.

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Beyond the formal comment process used for documents of this type, the following individuals provided valuable technical insights and/or specific reviews of this document in its various stages of development:

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TABLE OF CONTENTS

VOLUME 1: MAIN TEXT

List of Figures xii
List of Tables xiv
List of Acronyms xviii

1.0 INTRODUCTION
1.1 Purpose of Handbook 1-1
1.2 Source Term Formula 1-1
1.3 Applicability of Data 1-7
1.4 Accident Stresses 1-10
1.5 Handbook Organization 1-12

2.0 MATERIALS IN THE GASEOUS STATE
2.1 Noncondensible Gases 2-1
2.1.1 Summary of Analysis of Data 2-1
2.1.2 Discussion 2-1
2.2 Vapors (Condensible Gases) 2-2
2.2.1 Summary of Analysis of Data 2-2
2.2.2 Discussion 2-2

3.0 LIQUIDS
3.1 Summary of Analysis of Data 3-1
3.2 Aqueous Solutions 3-7
3.2.1 Thermal Stress: Evaporation and Boiling 3-7
3.2.1.1 Heating Of Shallow Pools 3-10
3.2.1.2 Heating of Pools 3-13
3.2.1.3 Additional Evaporation and Bubbling Release Studies . . 3-15
3.2.2 Explosive Stress: Shock, Blast, and Venting 3-18
3.2.2.1 Shock Effects 3-18
3.2.2.2 Blast Effects 3-19
3.2.2.3 Venting of Pressurized Liquids 3-19
3.2.2.3.1 Venting Below the Liquid Level 3-20
3.2.2.3.2 Venting Above the Liquid Level or Overall Containment Failure 3-22
3.2.2.3.3 Flashing Spray 3-26
3.2.3 Free-Fall Spill 3-33

3.2.3.1 Solutions 3-33
3.2.3.2 Slurries 3-35
3.2.3.3 Viscous Solutions 3-36
3.2.4 Aerodynamic Entrainment and Resuspension 3-37
3.2.4.1 Spray Release From Large Outdoor Pond 3-37
3.2.4.2 Suspension of Liquids From Shallow Pools of Concentrated Heavy Metal Solutions on Stainless Steel 3-40
3.2.4.3 Estimate of the Resuspension of Liquids From Soil 3-40
3.2.4.4 Suspension From Soil at Higher Windspeeds 3-41
3.2.4.5 Bounding Assessments 3-41
3.3 Organic, Combustible Liquids 3-42
3.3.1 Burning of Small Volume/Surface Area 30% TBP-Kerosine Solutions, No Vigorous Boiloff 3-43
3.3.2 Pool Fires of 30% TBP-Kerosine 3-44
3.3.3 Combustion of TBP-Kerosine Solutions Over Pools of Acid, Vigorous Boiloff 3-44
3.3.4 Airborne Release of Uranium During the Burning of Process Solvent 3-46
3.3.5 Airborne Release During Combustion of TBP-Kerosine 3-46
3.3.6 UNH and Air-Dried UNH on Various Surfaces During a Shallow Pool Gasoline Fire 3-48
3.3.7 Thermal Stress Bounding Recommendations 3-49

4.0 SOLIDS
4.1 Summary of Analysis of Data 4-1
4.2 Metals 4-11
4.2.1 Thermal Stress 4-11
4.2.1.1 Plutonium 4-11
4.2.1.1.1 Room Temperature Oxidation/Corrosion . . . 4-12
4.2.1.1.2 Oxidation at Elevated Temperatures Below Ignition Temperature 4-19
4.2.1.1.3 Self-sustained Oxidation Above the Ignition Temperature 4-21
4.2.1.1.4 Disturbed Molten Metal Surface With High Turbulence 4-25
4.2.1.1.5 Small Molten Metal Drops Hurled Through Air or Explosion of Entire Metal Mass 4-31
4.2.1.2 Uranium 4-33

4.2.1.2.1 Oxidation at Elevated Temperatures 4-37
4.2.1.2.2 Disturbed Molten Metal Surface With High Turbulence 4-42
4.2.1.2.3 Small Molten Metal Drops Hurled Through Air or Explosion of Entire Metal Mass 4-44
4.2.2 Explosive Stress: Shock, Blast, and Venting 4-44
4.2.2.1 Shock Effects 4-45
4.2.2.2 Blast Effects 4-45
4.2.2.3 Venting of Pressurized Gases Over Metals 4-45
4.2.3 Free-Fall Spill and Impaction Stress 4-45
4.2.4 Aerodynamic Entrainment and Resuspension 4-46
4.3 Nonmetallic or Composite Solids 4-46
4.3.1 Thermal Stress 4-46
4.3.1.1 Vitrified Waste 4-47
4.3.1.2 Aggregate 4-48
4.3.1.3 Encased Nuclear Material 4-48
4.3.1.3.1 Spent Nuclear Fuel 4-48
4.3.1.3.2 Metal Targets 4-50
4.3.1.3.3 Metal Alloy and Cermet Targets 4-50
4.3.2 Explosive Stress: Shock, Blast, and Venting 4-51
4.3.2.1 Shock Effects 4-51
4.3.2.2 Blast Effects 4-51
4.3.2.3 Venting of Pressurized Gases Over Solids 4-52
4.3.3 Free-Fall Spill and Impaction Stress 4-52
4.3.4 Aerodynamic Entrainment and Resuspension 4-52
4.4 Powders 4-55
4.4.1 Thermal Stress 4-55
4.4.1.1 Chemically Nonreactive Compounds 4-56
4.4.1.2 Chemically Reactive Compounds 4-57
4.4.2 Explosive Stress: Shock, Blast, and Venting 4-61
4.4.2.1 Shock Effects 4-61
4.4.2.2 Blast Effects 4-63
4.4.2.2.1 Unshielded Blast Effects From Detonations and Large Volume, Confined Deflagrations 4-63
4.4.2.2.2 Shielded Blast Effects From Detonations and Large Volume, Confined Deflagrations 4-65
4.4.2.3 Venting of Pressurized Powder 4-69

4.4.2.3.1 Venting of Pressurized Powders or Pressurized Gases Through a Powder, Pressure > 0.17 MPa_g 4-70
4.4.2.3.2 Venting of Pressurized Powders or Pressurized Gases Through a Powders, Pressure ≤ 0.17 MPa_g 4-73
4.4.3 Free-Fall Spill and Impaction Stress 4-74
4.4.3.1 Free-Fall Spill of Powder with Air Velocity Normal to the Direction of Fall 4-74
4.4.3.1.1 Factors That Affect Dust Generation 4-74
4.4.3.1.2 Free-Fall Spill Experiments 4-77
4.4.3.1.3 Free-Fall Spill of Powder Model 4-81
4.4.3.2 Free-Fall Spill with Enhanced Velocity Effects Normal to the Direction of Fall 4-82
4.4.3.3 Impact 4-83
4.4.3.3.1 Vibration Shock 4-84
4.4.3.3.2 Large Falling Object Impact or Induced Air Turbulence 4-85
4.4.4 Aerodynamic Entrainment and Resuspension 4-88
4.4.4.1 Entrainment From the Surface of a Homogenous Powder Layer 4-88
4.4.4.1.1 Review Of Literature on Resuspension Phenomena, Factors, and Rates 4-88
4.4.4.1.2 Experimentally Measured Resuspension Rates 4-96
4.4.4.2 Suspension of Material by Vehicular Traffic 4-101

5.0 SURFACE CONTAMINATION
5.1 Summary of Analysis of Data 5-1
5.2 Contaminated, Combustible Solids 5-9
5.2.1 Thermal Stress 5-9
5.2.1.1 Packaged Waste 5-10
5.2.1.2 Uncontained, Cellulosic Waste or Largely Cellulosic Mixed Waste 5-13
5.2.1.3 Dispersed Ash Dropped into Airstream or Exposed to Forced Draft Air 5-15
5.2.1.4 Uncontained Plastics/Elastomers 5-16
5.2.1.4.1 Polychloropene (Elastomer, Rubber) 5-17
5.2.1.4.2 Polymethylmethacrylate (Lucite/Perspex, Windows) 5-17

5.2.1.4.3 Polystyrene (Molded Plastic, Containers, Ion Exchange Resin) 5-18
5.2.2 Explosive Stress: Shock, Blast, and Venting 5-18
5.2.2.1 Shock Effects 5-19
5.2.2.2 Blast Effects 5-19
5.2.2.3 Venting of Pressurized Gases Over Solids 5-20
5.2.3 Free-Fall Spill and Impaction Stress 5-20
5.2.3.1 Free-Fall Spill 5-20
5.2.3.2 Impact 5-20
5.2.4 Aerodynamic Entrainment and Resuspension 5-21
5.3 Solid, Noncombustible Unyielding Surfaces 5-21
5.3.1 Thermal Stress 5-21
5.3.2 Explosive Stress: Shock, Blast, and Venting 5-21
5.3.2.1 Shock Effects 5-21
5.3.2.1.1 Contaminated Solids as Monoliths 5-21
5.3.2.1.2 Contaminated Soil 5-22
5.3.2.2 Blast Effects 5-22
5.3.2.3 Venting of Pressurized Gases Over Solids 5-22
5.3.3 Free-Fall Spill and Impaction Stress 5-23
5.3.3.1 Free-Fall Spill 5-23
5.3.3.2 Impact 5-23
5.3.3.2.1 Solids That Undergo Brittle Fracture 5-23
5.3.3.2.2 Solids That Do Not Brittle Fracture 5-23
5.3.4 Aerodynamic Entrainment and Resuspension 5-24
5.4 HEPA Filters 5-30
5.4.1 Thermal Stress 5-30
5.4.2 Explosive Stresses: Shock, Blast, and Venting 5-31
5.4.2.1 Shock Effects 5-31
5.4.2.2 Blast Effects 5-31
5.4.2.3 Venting of Pressurized Gases Through Filters 5-31
5.4.4 Free-Fall Spill and Impaction Stress 5-32
5.4.4.1 Enclosed Filter Media 5-33
5.4.4.2 Unenclosed Filter Media 5-35

6.0 INADVERTENT NUCLEAR CRITICALITY
6.1 Summary of Bounding Release Estimates 6-1
6.2 Total Fission Yield 6-5
6.2.1 Historical Excursions 6-5
6.2.2 Analytical Models for Solution Criticalities 6-5
6.2.3 Assessment of Fission Yields 6-12
6.2.3.1 Solutions 6-13

6.2.3.2 Fully Moderated and Reflected Solids 6-15
6.2.3.3 Dry Powder and Metal 6-15
6.2.3.4 Large Storage Arrays 6-16
6.3 Material Release in Criticality Excursions 6-16
6.3.1 Solutions 6-17
6.3.2 Fully Moderated and Reflected Solids 6-21
6.3.3 Dry Powder and Metal 6-24
6.3.4 Large Storage Arrays 6-24

7.0 APPLICATION EXAMPLES
7.1 Application Methodology 7-1
7.2 Example Background 7-2
7.3 Mock Operation Examples 7-5
7.3.1 Feed Preparation 7-5
7.3.1.1 Hazard Summary 7-5
7.3.1.2 Release Estimation 7-6
7.3.1.3 Feed Preparation Example Assessment 7-12
7.3.2 Oxide Dissolution 7-14
7.3.2.1 Hazard Summary 7-14
7.3.2.2 Release Estimation 7-14
7.3.2.3 Oxide Dissolution Example Assessment 7-18
7.3.3 Residue Dissolution 7-19
7.3.3.1 Hazard Summary 7-19
7.3.3.2 Release Estimation 7-19
7.3.3.3 Residue Dissolution Example Assessment 7-20
7.3.4 Metal Dissolution 7-20
7.3.4.1 Hazard Summary 7-20
7.3.4.2 Release Estimation 7-21
7.3.4.3 Metal Dissolution Example Assessment 7-26
7.3.5 Liquid Sampling and Tank Farm 7-28
7.3.5.1 Hazard Summary 7-28
7.3.5.2 Release Estimation 7-29
7.3.5.3 Liquid Sampling and Tank Farm Example Assessment 7-32
7.3.6 Ion Exchange 7-34
7.3.6.1 Hazard Summary 7-34
7.3.6.2 Release Estimation 7-36
7.3.6.3 Ion Exchange Example Assessment 7-45
7.3.7 Calcination and Hydrofluorination 7-48
7.3.7.1 Hazard Summary 7-48
7.3.7.2 Release Estimation 7-49

7.3.7.3 Calcination and Hydrofluorination Example Assessment 7-51
7.3.8 Reduction Line 7-53
7.3.8.1 Hazard Summary 7-53
7.3.8.2 Release Estimation 7-55
7.3.8.3 Reduction Line Example Assessment 7-59
7.3.9 Solid Waste Handling 7-60
7.3.9.1 Hazard Summary 7-60
7.3.9.2 Release Estimation 7-61
7.3.9.3 Solid Waste Handling Example Assessment 7-66
7.3.10 Seismic Release 7-66
7.3.10.1 Hazard Summary 7-66
7.3.10.2 Release Estimation 7-67
7.3.10.3 Seismic Release Example Assessment 7-73
7.3.11 Production Support Lab 7-74
7.3.11.1 Hazard Summary 7-74
7.3.11.2 Release Estimation 7-75
7.3.11.3 Production Support Lab Example Assessment 7-81
7.3.12 Conclusion 7-81

8.0 REFERENCES

LIST OF FIGURES

Figure 3-1	Size Distribution of Bubble Induced Droplets	3-11
Figure 3-2	Entrainment Data Obtained at Small Gas Velocity	3-16
Figure 3-3	Weight Percent Airborne as a Function of Mole Fraction of Pressurizing Gas	3-17
Figure 3-4	Mass Fraction vs. Droplet Diameters for Sprays as a Function of Orifice Diameter and Upstream Pressure	3-21
Figure 3-5	Average Weight Percent of Liquid Airborne as a Function of Pressure	3-24
Figure 3-6	Measured and Predicted Temperatures for Flashing-Spray Releases	3-30
Figure 3-7	Weight Percent Airborne Versus Viscosity for Sucrose, Slurry, and Low Surface Tension Spills	3-38
Figure 3-8	Mass Release Rate vs. Fetch, 200 by 200 Meter Pond	3-39
Figure 4-1	What Affects the Aerosolization Fraction in Plutonium Burning?	4-13
Figure 4-2	Dependence of Rate of Removal of Oxide Particulate from the Metal Surface on Humidity	4-14
Figure 4-3	Particle Size Distributions Produced by the Oxidation of Plutonium Under a Variety of Conditions	4-16
Figure 4-4	Release of Particulate Oxide Less Than 10 µm Diameter from Plutonium into Saturated Air	4-17
Figure 4-5	Release of Particulate Oxide Less Than 10 µm Diameter from Plutonium into Dry Air	4-18
Figure 4-6	Representative Particle Size Distributions Obtained with Cascade Impactors for PuO_2 Aerosols Formed in Static and Dynamic Conditions	4-22
Figure 4-7	Mass-Size Distribution of Sodium-Plutonium Aerosol	4-34
Figure 4-8	Dependence of Uranium Ignition on Specific Area	4-36
Figure 4-9	Aerosol Mass of Uranium Airborne during Oxidation as a Function of Temperature	4-39
Figure 4-10	Respirable Mass of Uranium Airborne during Oxidation as a Function of Temperature	4-41
Figure 4-11	Uranium Metal Burning in Air; The Distributions of the Measurements of the Respirable Aerosol Fractions in the Static and Dynamic Experiments	4-43
Figure 4-12	Particle Size Distribution Resulting from UO_2-2 Pellet Impact Test Data for UO_2 Specimen #2 Including Mean Grain Size of Original Crystalline UO_2 Particles. (Size Distribution 3 pellets 13.7-mm diameter x 13.6-mm long; drop-weight 1.2 J/cm^3)	4-53

Figure 4-13 Variation of Respirable Size for Impact Tests of Pyrex and SRL-131 Glass Specimens as a Function of Impact Test Energy Density 4-54
Figure 4-14 Equivalent Diameter PuO_2 Particle Lifted by Various Air Velocities 4-60
Figure 4-15 Removal of ThO_2 from Stainless Steel 4-64
Figure 4-16 Aerodynamic Entrainment of Uranium Dioxide Powder from a Stainless Steel Surface at an Air Velocity of 20 mph 4-68
Figure 4-17 Average Weight Percent of Powder Airborne as a Function of Pressure 4-72
Figure 4-18 Measured Size-Specific Dust Generation Rate Versus Predicted Size-Specific Generation Rate from Equations 8-11 4-78
Figure 4-19 Titanium Dioxide and Uranium Dioxide Particle-Size Distribution 4-79
Figure 4-20 Percent of Soil Airborne as a Function of Wind Speed 4-84
Figure 4-21 Particulate Threshold Friction Speed 4-90
Figure 4-22 Resuspension Factor Ranges from Mechanical and Wind Resuspension Stresses 4-99
Figure 4-23 Particle Resuspension Rates from an Asphalt Road Caused by Vehicle Passage 4-103
Figure 4-24 Particle Resuspension Rates from an Asphalt Road Caused by Vehicular Traffic Passage Four Days After Particle Deposition 4-104
Figure 4-25 Particle Resuspension Rates from an Asphalt Road as a Function of Weathering Time (Car Driven Through Tracer) 4-105

Figure 5-1 Results from Burning Contaminated Combustibles 5-11
Figure 5-2 Variation in Adhesive Forces Observed for Various Size Particles 5-25
Figure 5-3 Resuspension Flux F_r Air Flow Velocity for Particles Smaller Than 10 Micrometer Diameter 5-26
Figure 5-4 Schematic Diagram of Turbulent Burst in the Wall Region 5-27

LIST OF TABLES

Table 3-1 Fractional Airborne Releases During the Heating with Flowing Air of Concentrated Plutonium Nitrate Solution 3-12
Table 3-2 Fractional Airborne Release During Heating of Pools of Dilute Plutonium Nitrate Solution - 90% Volume Reduction 3-14
Table 3-3 Measured ARFs from Venting Pressurized Aqueous Solutions 3-23
Table 3-4 Average Weight Percent Airborne from Pressurized Liquid Release 3-23
Table 3-5 Measured ARFs and RFs During the Venting of Superheated Aqueous Solutions 3-29
Table 3-6 Measured ARFs and RFs From the Free-Fall Spill of Aqueous Solutions 3-34
Table 3-7 Statistical Summary of Drop Size Parameters for Lognormal Distributions 3-35
Table 3-8 Measured ARFs and RFs for Free-Fall Spill of Slurries (1 liter source, 3 meter fall distance) 3-36
Table 3-9 Measured ARFs and RFs for Free-Fall Spill of Viscous Solutions (1 liter source volume, 3 meter fall height) 3-36
Table 3-10 Measured ARFs from Shallow Pools of Concentrated Heavy Metal Salt Solution (0.72 g to 0.82 g Pu, 24-hr sampling period) 3-40
Table 3-11 Measured ARFs and RFs of Uranium (UNH) From Soil at 2.5 mph and ~ 20 mph 3-41
Table 3-12 Measured ARFs From Burning Small Volumes of 30% TBP-Kerosine Traced with Selected Radionuclides 3-44
Table 3-13 Measured Uranium ARFs During the Burning of TBP-Kerosine Over Aqueous Phase 3-45
Table 3-14 Measured Transfer Coefficients and Decontamination Factors During the Burning of TBP-Kerosine Solvent 3-47
Table 3-15 Measured ARFs and RFs for Uranium Airborne During Gasoline Fires on Various Surfaces Involving UNH and Air-Dried UNH 3-48
Table 4-1 Measured ARFs During Oxidation of Unalloyed and Delta-Phase Plutonium at Elevated Temperatures in Air 4-19
Table 4-2 Summary of Oxidation Rates for Plutonium at Temperatures Below 100°C 4-20
Table 4-3 Measured ARFs During Self-Sustained Oxidation of Unalloyed and Delta-Phase Plutonium 4-21
Table 4-4 Measured ARFs During Self-Sustained Oxidation of Unalloyed Plutonium Metal in Flowing Air 4-23

Table 4-5	Measured ARFs During the Self-Sustained Air Oxidation of Large Specimens of Unalloyed and Delta Plutonium Metal	4-24
Table 4-6	Measured ARFs for Heating Small Specimens of Delta Phase Pu Metal in Various Atmospheres	4-28
Table 4-7	ARFs From Heating Small Pieces Delta-Phase Plutonium Metal Under Specific Conditions	4-29
Table 4-8	Airborne Release of Pu From Pu-Na Mixtures During Self-Sustained Oxidation	4-30
Table 4-9	Possible Factors Influencing Uranium Oxidation	4-35
Table 4-10	Airborne Release from Nonreactive Powder During Heating in Flowing Air	4-56
Table 4-11	ARFs and RFs During the Heating of Reactive Compounds in Flowing Air	4-58
Table 4-12	ARFs and RFs from the Venting of Pressurized Powders	4-73
Table 4-13	Measured ARFs and RFs from the Free-Fall Spill of Powders	4-80
Table 4-14	Airborne Soil Particle Size Distribution	4-83
Table 4-15	Measured ARFs and RFs from the Impact of Structural Debris on Powders	4-86
Table 4-16	Resuspension Factors from Wind Resuspension Stresses	4-91
Table 4-17	Resuspension Factors from Mechanical Resuspension Stresses	4-92
Table 4-18	Resuspension Rates	4-95
Table 4-19	Variables Which Influence Resuspension	4-97
Table 5-1	ARFs from Burning of Packaged, Contaminated, Mixed, Combustible Wastes	5-12
Table 5-2	Measured ARFs and RFs from the Burning of Contaminated Cellulosic Materials	5-14
Table 5-3	Measured ARFs from Burning Contaminated PC	5-17
Table 5-4	Measured ARFs and RFs from Burning of Contaminated PMMA	5-18
Table 5-5	Measured ARFs and RF from Burning of Contaminated PS	5-18
Table 5-6	Fractions of Media Mass as Particles 10 μm and Less Generated by HEPA Filters Under Shock/Vibration Stresses	5-34
Table 6-1	Summary of Known Accidental Criticality Excursions (1945 to 1974): (a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems	6-6
Table 6-2	Destructive Power Excursion Summary	6-9
Table 6-3	Inhomogeneous Water-Moderated Systems	6-10
Table 6-4	Miscellaneous Systems	6-11
Table 6-5	Theoretical Criticality Accident Fission Yields	6-13
Table 6-6	Accidents in Processing Plants	6-14

Table 6-7 Curies of Important Nuclides Released During Nuclear Excursion Involving Spent Fuel Solution 6-18
Table 6-8 Curies of Important Nuclides Released During Nuclear Excursion Involving Unirradiated, Unenriched Uranium Solution (400 g U/liter) 6-19
Table 6-9 Curies of Important Nuclides Released During Nuclear Excursion Involving Plutonium Solutions 6-20
Table 6-10 Release Fraction for Various Chemical Classes from Heated Spent Fuel 6-23

Table 7-1 Site Boundary Doses Associated with Release 7-4
Table 7-2 Feed Preparation Example Topics 7-5
Table 7-3 Important Nuclides Released from Powder Plutonium Criticality 7-12
Table 7-4 Oxide Dissolution Example Topics 7-14
Table 7-5 Important Nuclides Released from Plutonium Solution Criticality 7-17
Table 7-6 Residue Dissolution Example Topics 7-19
Table 7-7 Metal Dissolution Example Topics 7-20
Table 7-8 Liquid Sampling and Tank Farm Example Topics 7-28
Table 7-9 Ion Exchange Example Topics 7-34
Table 7-10 Calcination and Hydrofluorination Example Topics 7-48
Table 7-11 Reduction Line Example Topics 7-53
Table 7-12 Solid Waste Handling Example Topics 7-60
Table 7-13 Seismic Release Example Topics 7-66
Table 7-14 Production Support Lab Example Topics 7-74

VOLUME II: APPENDICES

APPENDIX A Tables and Figures from Source Documents with Data Used in this Study

APPENDIX B Example Facilities

LIST OF ACRONYMS

The terminology used in this handbook is commonly used in DOE management or technical communities, and is therefore not defined. A number of acronyms are used constantly throughout this large document, however, and brief definitions are provided for important acronyms to assist the reader.

AED	Aerodynamic Equivalent Diameter: the diameter of a sphere of density 1 g/cm^3 that exhibits the same terminal velocity as the particle in question.
AMAD	Activity Median Aerodynamic Diameter: the diameter of the particle for which half the activity is associated with particles larger than and half the activity associated with particles smaller than this size particle.
AMMD	Aerodynamic Mass Median Diameter: the aerodynamic diameter of the particle for which half the mass is associated with particles greater than and half the mass the mass is associated with particles less than the stated size.
ARF	Airborne Release Fraction: the coefficient used to estimate the amount of a radioactive material that can be suspended in air and made available for airborne transport under a specific set of induced physical stresses. Applicable to events and situations that are completed during the course of the event.
ARR	Airborne Release Rate: the coefficient used to estimate the amount of material that can be suspended in air and made available for airborne transport under a specific set of induced physical stresses as a function of time. The rates are often longer-term averages due to the non-discrete nature of the release. Applicable to events that are relatively continuous over some time span.
CRAC	Consequences Radiologiques d'un Accident de Criticite: experimental criticality study.
DOE	U.S. Department of Energy.
DR	Damage Ratio: the fraction of MAR impacted by the actual accident-generated conditions under evaluation.
FP	Fission Products.

GSD — Geometric Standard Deviation: standard deviation of the logarithms. For any normal distribution, one standard deviation represents the difference between the size associated with the cumulative mass of 84.1% and the median (50% cumulative mass) size (or between the 50% cumulative size and the 15.9% cumulative size). For lognormal distributions, ratios between sizes are as follows:

$$GSD = sigma_g = d_{84\%}/d_{50\%} = d_{50\%}/d_{16\%} = [d_{84\%}/d_{16\%}]^{1/2}$$

HEPA — High Efficiency Particulate Air.

IHE — Insensitive High Explosives: high explosives that are less sensitive to ordinary initiators such as shock, heat, etc. than conventional high explosives such as TNT, dynamite.

LLD — Least Linear Diameter: the size distributions determined by sieving. The fractions are categorized by the particles that can pass through the openings of a sieve and, if not spherical, represent the small dimension of that particle.

LPF — LeakPath Factor: the fraction of airborne materials transported from containment or confinement deposition or filtration mechanism (e.g., fraction of airborne material in a glovebox leaving the glovebox under static conditions, fraction of material passing through a HEPA filter.)

MAR — Material-at-Risk: the amount of radioactive materials (in grams or curies of activity for each radionuclide) available to be acted on by a given physical stress.

MMD — Mass Median Diameter: the geometric diameter of the particle for which half the mass is associated with particles greater and half the mass associated with particles less than the stated size.

MR — Mass Ratio: the ratio mass of inert material impacted to mass of TNT equivalent of an explosion.

NRC — U.S. Nuclear Regulatory Commission.

PC — Polychloropene.

PMMA — Polymethylmethacrylate.

PS	Polystyrene.
PSPILL	Powder Spill computer code: See subsection 4.4.3.1.3.
PULF	Pulverization Fragments calculation: See section 4.3.3.
RF	Respirable Fraction: the fraction of airborne radionuclides as particles that can be transported through air and inhaled into the human respiratory system and is commonly assumed to include particles 10-µm Aerodynamic Equivalent Diameter (AED) and less.
SAR	Safety Analysis Report.
TBP	Tri-normal Butyl Phosphate.
UNH	Uranyl Nitrate Hexahydrate: The uranium solution form generally associated with recovered uranium from the PUREX (plutonium uranium extraction, a liquid-liquid extraction) process for reprocessing spent nuclear fuel or irradiated product targets.

1.0 INTRODUCTION

1.1 PURPOSE OF HANDBOOK

The purpose of this document is to provide a compendium and analysis of experimental data from which airborne release fractions (ARFs) and respirable fractions (RFs) may be derived. Such values are needed to determine quantities of radioactive material driven airborne for the purpose of estimating the scope of the potential release spectrum and potential downwind consequences from a given facility or activity. The information provided in this handbook aids in making such estimates. This introduction discusses the following major topics:

- **Source term formula** - Provides a computational formula for using this information.
- **Applicability of data** - Distinguishes proper use of information.
- **Accident stresses** - Identifies the types of accident conditions for which this information is applicable.
- **Handbook organization** - Explains presentation of information and use of examples.

The data in this handbook can be used in a variety of applications, such as safety and environmental analyses, and to provide information relevant to system and experiment design. However, these data and the analyses of the data contained herein need to be critically evaluated for applicability in each situation in which they are used, and represent only one source of information in a complete safety analysis or design process.

1.2 SOURCE TERM FORMULA

The source term is the amount of radioactive material, in grams or curies, released to the air. The initial source term is the amount of radioactive material driven airborne at the accident source. The initial respirable source term, a subset of the initial source term, is the amount of radioactive material driven airborne at the accident source that is effectively inhalable. Lesser source terms are determined by applying filtration or deposition factors to the initial source term.

The airborne pathway is of primary interest for nonreactor nuclear facilities. DOE-STD-1027-92 quotes observations of the NRC to the effect that "for all materials of greatest interest for fuel cycle and other radioactive material licenses, the dose from the inhalation

pathway will dominate the (overall) dose" (NUREG-1140). The airborne source term is typically estimated by the following five-component linear equation:

$$\text{Source Term} = \text{MAR} \times \text{DR} \times \text{ARF} \times \text{RF} \times \text{LPF} \quad (1\text{-}1)$$

where:

MAR = Material-at-Risk (curies or grams),
DR = Damage Ratio,
ARF = Airborne Release Fraction (or Airborne Release Rate for continuous release),
RF = Respirable Fraction, and
LPF = Leakpath Factor.

The initial source term and initial respirable source term are products of the first three factors and first four factors respectively. A depleted source term after a subsequent stage of deposition or filtration is a product of the initial source term multiplied by the leakpath factor of the specific stage.

This handbook assesses ARF and RF values separately for sources of airborne material generated from accidents involving gases, liquids, solids, and surface contamination. All of the above factors may need to be determined for particulate releases. Some of them, however, will collapse to values of one for special cases (e.g., gaseous releases).

Material-at-Risk (MAR)

The material-at-risk is the amount of radionuclides (in grams or curies of activity for each radionuclide) available to be acted on by a given physical stress. For facilities, processes, and activities, the MAR is a value representing some maximum quantity of radionuclide present or reasonably anticipated for the process or structure being analyzed. Different MARs may be assigned for different accidents as it is only necessary to define the material in those discrete physical locations that are exposed to a given stress. For example, a spill may involve only the contents of a tank in one glovebox. Conversely, a seismic event may involve all of the material in a building.

Damage Ratio (DR)

The damage ratio is the fraction of the MAR actually impacted by the accident-generated conditions. A degree of interdependence exists between the definitions of MAR and DR. If it is predetermined that certain types of material would not be affected by a given accident, some analysts will exclude this material from the MAR.

As an example, assume 600 g from a total of 1000 g of material X are in a form that would not be affected by an explosion. Of the remaining 400 g, 200 g have a high release fraction and 200 g have a low release fraction. If the 600 g is included in the MAR, specific DRs for the MAR of 1000 g are 0 for the unaffected material, 0.2 for the high release fraction material, and 0.2 for the low release fraction material. If the 600 g is not included, specific DRs for the MAR of 400 grams are 0.5 for the high release fraction material and 0.5 for the low release fraction material. The basic distinction is whether or not DRs of 0 are officially designated. Neither convention cited in the example is necessarily correct. What is important is that one convention is used consistently to avoid an obvious potential for assigning incorrect DR values.

The DR is estimated based upon engineering analysis of the response of structural materials and materials-of-construction for containment to the type and level of stress/force generated by the event. Standard engineering approximations are typically used. These approximations often include a degree of conservatism due to simplification of phenomena to obtain a useable model, but the purpose of the approximation is to obtain, to the degree possible, a realistic understanding of potential effects.

Airborne Release Fraction (ARF)

The ARF is the coefficient used to estimate the amount of a radioactive material suspended in air as an aerosol and thus available for transport due to a physical stresses from a specific accident. For discrete events, the ARF is a fraction of the material affected. For mechanisms that continuously act to suspend radionuclides (e.g., aerodynamic entrainment/resuspension), a release rate is required to estimate the potential airborne release from postulated accident conditions. Generally, accident release rates (ARRs) are based upon measurements over some extended period to encompass most release situations for a particular mechanism. The rates are average rates for the broad spectrum of situations and, as such, the most typically meaningful time unit to reflect average conditions is 1 hour. There is evidence (discussed later in the subsection on the aerodynamic entrainment of surface contamination) that in some situations (e.g., aerodynamic entrainment of sparse powder deposits on a heterogeneous surface), the rate of release is not uniform with time. Even in the situations where the rates are relatively uniform, the source is depleted by the removal of particles from the surface by aerodynamic forces, and the amount of material airborne decreases with time unless the source is continuously replenished.

This handbook specifically deals with ARFs and ARRs, although ARF will connote both concepts in generic discussions for the sake of simplicity. The ARFs are based primarily upon experimentally measured values for the specific material (e.g., plutonium, uranium, mixed fission products) or surrogates subjected to the particular type of stress under

controlled conditions. Attention is given to the parameters, if known, that may have a significant influence upon suspension by the specific mechanism and the uncertainty in the measurement as indicated by the variability of the results. Those applying the data must be aware of the range of stress represented by the measured ARFs, and seek to define the accident conditions to determine, in a gross sense, whether or not the stresses induced by the postulated events are bounded by the experimental parameters as evaluated in this document.

It is important to note that the experiments discussed evaluate release phenomena holistically. No attempt is made to precisely characterize total airborne material in terms of individual mechanisms acting within an overall given release. To obtain useful data outside the immediate physical chaos of the stress-inducing event itself, the experimental apparatus cited almost uniformly relied upon designs to channel air to some contamination collection mechanism a short distance from the point of generation. The need to keep this distance small to avoid introducing new distortion in the form of aerosol deposition effects is one of the reasons for the relatively small scale of many experiments.

In keeping with this experimental design, the interpretation of experimental data does not consider material momentarily airborne from substrate mass ejection due to physical stresses acting on the substrate mass as an aerosol suspended in air. For example, in fire and boiling experiments, fuel mass and volumes of solution were observed to splatter or launch from the experimental substrate and land on surfaces in the local vicinity. The radioactive contamination carried with this material that deposits and is measured on the adjacent surfaces is not an aerosol suspended in air, and does not travel on air currents. It represents a source of highly localized migration that is not amenable to meaningful prediction and is not relevant to the issue of how much material might be expected to escape the immediate area and disperse in air.

Respirable Fraction (RF)

The RF is the fraction of airborne radionuclides as particles that can be transported through air and inhaled into the human respiratory system and is commonly assumed to include particles 10-µm Aerodynamic Equivalent Diameter (AED) and less. Other definitions of "respirable particles" have been presented by various groups at different times. The British Medical Research Council adopted a definition in 1952 classifying particles with a terminal velocity equal to that of a 5-µm diameter as "respirable dust." The U. S. Atomic Energy Commission defined "respirable dust" as insoluble particles that are part of inhaled dust which penetrate to the non-ciliated portions of the gas exchange region, and with a 50% respirable cut-size of 3.5-µm AED. The American Conference of Governmental Industrial Hygienists has adopted a definition that is almost identical, differing only in the 2 µm fraction allowed. The U. S. Environmental Protection Agency defines "inhalable dust"

(particles penetrating the upper respiratory airway and entering the thorax) with a 50% cut-off at 15-μm AED. The International Standards Organization - Europe defines "inhalable dust" as particles entering the nasal or oral passages with a 50% cut-size of 10-μm AED. Accordingly, use of a 10-μm AED cut-size for respirable particles is considered conservative, and may even be overly conservative since the mass is a cube function of particle diameter.

The size of a particle is a function of the measurement technique used. If the method used is optical/electron microscopy or spectrometry, particle size is a projected diameter measured by the plane that intercepts the light/electron beam or reflection from light scattered by the particle. The size represents the two-dimensional area intercepting the beam and, as with all projections of three dimensions into two, can result in considerable distortion. Projected diameter approximates the Geometric Diameter (D_g). D_g is also approximated by sieving where the size measurement is termed geometric/linear/least linear diameter. The measurement represents the smallest dimension of the particle that will pass through the openings in the sieve.

Liquid and air sedimentation techniques of inertial impaction by a cascade impactor measure the settling velocity of a particle and report size as an aerodynamic characteristic. Size is reported as an equivalent sphere with an equivalent settling velocity, or Stoke Diameter (D_{Stk}). The Aerodynamic Diameter (D_{AED}) specifically refers to an equivalent sphere with a density of 1 g/cm^3. D_{AED} is the parameter of interest for defining respirable particles (i.e., ≤ 10-μm AED) as it normalizes materials of differing density. Other size units include Ferret's diameter (the relationship between the maximum and minimum diameters) or Sauter's mean diameter (the surface to volume ratio most representative of the distribution of a group of liquid drops), but these are of little use for the purposes of this document. D_g is related to D_{AED} by the equation:

$$D_{AED} = (D_g[\rho_p]^{0.5}[C_{C,e}/C_{C,a}]^{0.5})/\alpha \qquad (1\text{-}2)$$

where:

ρ_p = Particle density (g/cm^3),
$C_{C,e}$ = Cunningham slip factor corresponding to the volume equivalent diameter,
$C_{C,a}$ = Cunningham slip factor corresponding to the aerodynamic equivalent diameter, and
α = Aerodynamic shape factor.

The Cunningham slip factor is related to the potential for particle impact with the mean free path of air molecules. Above the sub-micron size range, all particles impact with air

molecules, and the ratio of Cunningham factors can be ignored. The aerodynamic shape factor is not typically known and is assumed to be 1. Therefore, D_{AED} may be estimated from D_g by simply multiplying D_g by the square root of the particle density. More discussion of this subject can be found in available references (e.g., Fry, T.M., 1980).

Although the principal emphasis in this document is directed toward the potential downwind hazard to the populations at some distance from the point of source term generation, airborne particles larger than 10-μm AED released from the facility may constitute an onsite hazard (direct radiation) and may (if the larger particles are agglomerates that deagglomerate with time or can be subdivided by local conditions) be subject to re-dispersal. If direct shine can be a significant contributor to doses (e.g., fission product release from a criticality excursion), the respirable factor should not be accounted for in evaluating that pathway contributor.

This handbook specifically addresses RFs. RFs for particles made airborne under accident-induced stresses are dependent upon a variety of factors, such as the bulk density (i.e., how well the powder at rest compacts), the presence of moisture, how effectively the type and level of stress deagglomerates the powder or subdivides the solid/liquid, the efficiency with which the stress suspends the powder/fragments of solid over varying size ranges, and the degree of immediate proximity of surfaces on which airborne particles may impact/settle. Data to evaluate these factors individually for all cases are not found in the literature. Measured RF data from the experimental studies are applied where available.

Measured experimental data for RFs are much more limited but are from the same general sources used for the ARFs. To keep RFs at a reasonable bounding rather than an ultraconservative level, the RF associated with the measured bounding ARF is generally selected rather than the highest RF value measured. The highest RF values are often associated with the smallest ARFs, and when used in conjunction with the bounding ARF, result in ultraconservative estimates of the respirable fraction released. When no measured RF is associated with the maximum measured ARF, but other measured RFs are available for the experimental set, the greatest RFs are generally used. In some cases where significant uncertainty may exist, RFs are arbitrarily set to a value of 1.0 for conservatism.

Leakpath Factor (LPF)

The LPF is the fraction of the radionuclides in the aerosol transported through some confinement deposition of filtration mechanism. There can be many LPFs for some accident conditions (e.g., the fraction transported from the package, such as a shipping container, to the cell or enclosure; the fraction leaked from the enclosure, cell, or glovebox to the operating area around the enclosure or room; the fraction leaked from the room to the

building-atmosphere interface). Where multiple leakpaths are involved, their cumulative effect is often expressed as one value that is the product of all leakpath multiples. The LPF is a calculated or standard value based upon (1) established relationships between size of the particulate material, airborne transport mechanisms, and losses by deposition mechanisms, or (2) specified filtration efficiencies.

1.3 APPLICABILITY OF DATA

In most cases, the ARFs and RFs for conditions bounded by the experimental parameters can be defined to one significant digit. The parameter definition process has focused on estimating reasonable bounding values because of the limited quantity and variability of the data. The use of the word "reasonable" is an acknowledgement that the only definitive bounds are ARFs and RFs of 1.0, which can always be postulated if enough synchronous, extreme localized conditions are assumed. Such extreme synchronicity is neither an expected condition nor a practically useful model of reality. The NRC has commented on this subject in a survey of nonreactor nuclear facility release potential and historical experience (NUREG-1140) as follows:

> Operating experience may be more relevant for these [fuel cycle] licensees than for nuclear power plants because of the nature of the accident driving force. In nuclear power plants the driving force is the enormous amount of heat in the reactor. The available energy is so large that some unique occurrences are conceivable, such as molten cores, large-scale metal-water reactions, and rupturing of the containment by overpressurization. Because these events have never happened, they can only be studied theoretically. The dominant driving force for accidents at nonreactor licensees are common industrial accidents--fires, chemical explosions, leaks, and the like. A great deal of industrial accident experience can be drawn upon in analyzing these potential accidents.

The experiments used as a basis for this document were specifically focusing on general characterization of suspension phenomena in an industrial environment. A key element in defining bounding values from the data is understanding the physical entrainment mechanisms at work, their potential variability, and the inherent limits of such mechanisms. Accordingly, this document provides detailed discussion of entrainment mechanisms wherever possible.

Median and average values are estimated for some data. These estimates are made solely for the purpose of providing perspective on potential conservatism and should not be used as a basis for an ARF statistical distribution. It is generally not productive to attempt to use the experimental data cited in this handbook to develop assumed statistical distributions of values

for probabilistic assessments. The overall collection of data available for a wide variety of stresses will not support fine statistical resolution as a technically meaningful activity, and this handbook specifically rejects citation as a defensible basis for such attempts.

The generation and suspension of particles is the result of the interaction of multiple physiochemical variables that have not been completely characterized as the majority of the experiments performed were designed in an attempt to reflect reasonably bounding conditions for specific industrial situations of concern. Accordingly, the data obtained are more accurately characterized as selected points from multiple distributions against multiple parameters than as different values from a common distribution. Even if this point is neglected, there are still practically intractable problems in attempting to generate statistical distributions. While the data are presumed to be bounding for the purpose intended, it is largely unknown whether the data values are truly 90th percentile, 99th percentile, 99.9th percentile, etc. Further, in many cases it is considered likely that accident specific ARFs are actually distributed in a highly irregular manner (i.e., multi-modal or truncated distributions). Assuming a typical distribution (i.e., log-normal, Poisson) using standard deviations will produce seriously distorted values that may have little or nothing to do with reality.

The available data do, however, cover a range of conditions that typify the energy sources associated with nonreactor nuclear facilities. The data cover a more complete range of phenomenological concerns than the data upon which nuclear reactor source terms have been estimated. They are at least the equal of reactor source term data in overall quality, and a number of the experiments performed were very close to actual scale for the type of operations conducted at nonreactor nuclear facilities. In general, scaling effects, while not to be trivialized, are less of an issue with this data than with comparable reactor source term estimation data. The NRC has already accepted a considerable amount of this information as a basis for source term estimation in NUREG-1320, "Nuclear Fuel Cycle Facility Accident Analysis Handbook." DOE-HDBK-3010-94 serves a similar function for DOE.

The values for ARFs and RFs taken from experimental measurements are reasonably well defined. It is noted, however, that they are dependent upon the types and levels of stress imposed, the initial state (physical form, chemical composition, particle size distribution, degree of dispersion of the material-of-concern), and the response of the material-of-concern and other materials present. In most cases, the materials chosen for the experiments were selected to bound the behavior of materials under accident conditions for a specific location or process. The applicability of the experimental conditions to the complete spectrum of processes and potential accident conditions is, however, uncertain. For this reason, the discussions of the data have tried to indicate where typical industrial accident phenomena (e.g., fires, explosions, spills) are considered covered by the data.

1.0 Introduction

The estimates of ARFs and RFs applicable to various accident-generated mechanisms for the suspension of radioactive materials are based upon experimental data for specific types and levels of stresses/force. Care must be used in applying the ARFs/ARRs to ensure that the values chosen truly reflect the type and level of stress/force postulated for the event. For instance, the suspension of powder from a surface (commonly termed resuspension) is not applicable to situations where the powder is dropped into flowing gas in a dispersed fashion.

Before the ARFs and RFs presented can be properly applied, the conditions imposed and the response of critical items must be evaluated. The calculational methods to perform the engineering analysis are not part of the scope of this document. Many standard methods are applicable (e.g., the rupture pressure of tanks and piping based upon the material of construction, the thickness, the temperature and pressure). In other cases (e.g., the blast energy from the deflagration of flammable gas and oxidant mixtures in the free volume above the materials), however, standard engineering calculational methods are not available and interpretation of information and data (e.g., the fraction of the heat of combustion of reactants that translates into the shock wave) is required.

Once the forces and conditions imposed upon the material for dispersion/fragmentation and suspension are identified, the applicable ARF and RF can be selected. In most cases, precise correspondence between the event conditions and experimental conditions during the measurement of the ARFs and RFs is not found. For conservative analysis, the data are applicable if the measurement conditions exceed those calculated for the event (e.g., if the fall distances for spilled powders or liquids with characteristics like the materials used in the experiments are equal to or less than the experimental distance of 3 m). In most cases, extrapolation beyond the experimental data is valid for a limited range beyond the maximum (a factor of 2 to 5 dependent on the slope of the experimental data and the range of conditions covered in the experimental study) imposed in the experimental study. Models are available for the calculation of ARFs and RFs for some phenomena (e.g., free-fall spill of powders and liquids - Ballinger et. al., 1988; PULF formula for fragmentation by brittle fracture - Sandia, 1987). Care should be used in any extrapolation, however, to avoid producing obviously inappropriate answers. This caution is particularly apt if calculations are being used to influence facility or process design.

A final emphasis is necessary regarding application of this data. As developed for the NRC and DOE, it has never been intended to be used as absolute proof of anything. Special attention has been given to understanding suspension phenomena, ranges of relevant parameters covered in experimental studies, artifacts or limitations of the data that may have been induced by experimental conditions, and possible effects of relevant parameters that may not have been controlled or monitored. As noted, this has resulted in development of

bounding ARFs and RFs. The purpose of developing these values was (1) to better understand the potential bounding hazards presented by nonreactor nuclear facilities, and (2) to provide information to support general bases of decisionmaking. The first purpose has been fulfilled by use of this information in the Defense Programs Safety Survey Report (Pinkston, et al., 1993). This use has supported previous NRC estimates that the bounding consequence potential for nonreactor nuclear facilities is significantly less than commercial nuclear power plants, or large commercial chemical plants as well. In domino fashion, this conclusion has reemphasized the use of the term "general bases" in the second purpose. The information in this handbook can be used to indicate relative significance of unmitigated releases and to verify the effectiveness of mitigative measures, such as HEPA filtration. It is a misuse of information in this handbook to focus on ARF "pencil-sharpening" at the expense of objective, performance based evaluation, particularly to attempt subtle judgements of consequence potential to support fine distinctions such as testing HEPA filters to only 90% efficiency, or to claim that meeting a dose guideline alone using this data constitutes a complete safety basis.

1.4 ACCIDENT STRESSES

In developing this handbook, literature and historical experience on the major types of accidents in nonreactor nuclear facilities were reviewed. The evaluation of experimental data was then focused on identifying applicability for those accidents. High-energy insertion-type events that are of concern for nuclear reactors are not covered by this document, nor are some accident conditions peculiar to high-level waste tanks (e.g., response of salt or moist salts to accident-generated conditions). Some responses of materials found in high-level waste vitrification plants to accident stresses are directly covered (e.g., brittle fracture of glasses due to crush-impact, free-fall spill of liquids and slurries), but others (e.g., behavior of molten salts and glass) are not. In some cases, attempts can be made to bound such materials by using data for more limiting materials. As with extrapolation, care should be used in any such attempt.

The main types of accidents of common concern in nonreactor nuclear facilities are:

- **Spill**: Material experiences instability/shear stress at the surface of the mass resulting in sub-division of the overall mass. Airflow patterns around and through the material mass, including induced turbulence, accelerate overall sub-division. Mass breakup is further enhanced by impact with ground surface. The material sub-division can generate particles sufficiently small that they remain airborne for a significant period of time.

- **Fire**: Generates heat and combustion gases that may destroy/stress the radioactive material and/or the substrate upon which radioactive materials may be deposited, compromise barriers, and/or pressurize containers/enclosure that may lead to the airborne release of contained radioactive materials. Mass flux of vapors from the reacting surfaces suspend material in air. This material is then entrained in general convective currents that provide transport for particulate materials.

- **Explosion**: Generates shock and blast effects with potential for gas flow subsequent to the explosive event that may subdivide/deagglomerate and entrain material. Explosive reactions may result from chemical (e.g., oxidations involving branch-chain products, oxidations of gas-oxidant mixtures) or physical (overpressurization to failure of tanks or vessel, vapor explosions) reactions. Shock waves are supersonic pressure waves (pulses) that can transmit an impulse to materials and the surrounding structures resulting in shattering of solid items. Shock waves are a true wave phenomenon and involve little gross motion of propagating medium. The potential for damage from shock waves has been extensively characterized.

  Blast effects are typically subsonic and involve material entrained in the gas flow. Blast effects are often more damaging. Blast effects are not subject to the same reflection/amplification phenomena as shock waves because they have significant momentum and inertia. The gas expanding from the explosion zone carries material from the explosion site. If the explosion is adjacent to the MARs, then blast effects can cause damage above and beyond the initial impulse loading. Some explosive reactions may be followed by chemical reactions, material vaporization, or fires that lead to substantial gas flows following the explosive event. These gas flows may also entrain material. Deflagrations do not involve shock, but can simulate blast effects. Under proper conditions (e.g., confinement, structural features that enhance turbulence), deflagrations can transition to detonations and produce shock waves.

- **Criticality**: Major hazard is unshielded radiation produced. Generates fission products that may become airborne as well. Fission product gases are released from liquid criticalities and from solid criticalities to the extent the underlying critical mass is degraded. Solid fission products typically have small release fractions determined by the degree of physical stress placed on the critical mass itself. At large fission yields, solid critical masses may experience some degree of melting or oxidation.

- **Earthquake**: Typically bounds other natural phenomena. Generates severe lateral and vertical stresses upon the structure and equipment that may result in confinement failure, breach, or collapse. The response of the materials-of-construction may dislodge materials-of-concern by vibration, impact of debris, and fragmentation. Seismic forces may cause material spills but do not generate gas flow to transport particulate materials, although flows are generated by falling debris or any fires/explosion caused by the seismic event.

The information presented in this document is directed toward evaluation of the radiological consequences of these events and the suspension phenomena that they generate. Unless otherwise noted, the release from material affected by multiple phenomena can be calculated individually and summed to obtain the overall release.

1.5 HANDBOOK ORGANIZATION

The evaluation of data is given by the physical form of the material affected (e.g., gas, liquid, solid, surface contamination) and suspension stresses (e.g., spill, thermal stress, shock wave, and blast stress) in chapters 2 through 5. Because of its unique nature, criticality is treated as a phenomenon for all materials in chapter 6. Examples of application of the release fraction recommendations are given in chapter 7.

Each chapter begins with a summary of the results of data analyses, which are followed by discussions of specific data analyses in the remainder of the chapter. Bounding values for parameters and, in some cases, median values derived from the data analyses are presented in these summaries. As previously noted, where median values are presented, they are for the purpose of providing perspective on the potential conservatism of the bounding values as the available data do not generally support the derivation of data distributions. When using these data, care must be taken to assure the experimental data are applicable to the situation being analyzed. A summary table of the individual chapter summaries has not been provided in this chapter due to the sheer amount and variety of information in this document. A summary table is not considered an efficient vehicle for communication in a document of this nature as it would be either too large and unwieldy to satisfy the purpose of a table or too superficial to adequately represent the document.

Data tables in the body of individual chapters are summations from raw data, which is typically provided in Appendix A. Original graphs are presented both in the chapters and in Appendix A, while figures of experimental apparatus, where available, are presented exclusively in Appendix A. Metric units are used in most instances, but some old documents and figures cited are in English units. It is noted that many of the source reports used are difficult to obtain, and copies available are old and fading. This significantly reduces the

legibility of reproductions. Accordingly, the notation "reproduced from" indicates where raw data and figures from original source reports have been retyped or traced to enhance legibility.

Chapter 7 is designed to assist users of this document. It contains examples of application of the information for accidents that have occurred or are considered physically credible at DOE nonreactor nuclear facilities. These examples are based on example facilities detailed in Appendix B. Simple analyses of a defined facility were considered to be particularly helpful in demonstrating the use of ARFs and RFs in relation to the overall analysis process. This example also underscores previous cautions by indicating how excessive reliance on mathematical models using this data can lead to overlooking obviously inappropriate design or operational practices.

2.0 MATERIALS IN THE GASEOUS STATE

2.1 NONCONDENSIBLE GASES

2.1.1 Summary of Analysis of Data

- Loss of Physical Containment. For noncondensible gases, the recommended ARF is 1.0. All materials in the gaseous state can be transported and inhaled; therefore, a value for RF of 1.0 is assumed for the purposes of these analyses.

2.1.2 Discussion

In DOE nonreactor nuclear facilities, radionuclides in the form of noncondensible gases are only found under a few circumstances: as stored tritium, encased in a stored spent fuel matrix, generated by physical or chemical reaction, and generated by inadvertent nuclear criticalities (see Chapter 6). The radiological impact on the human body varies greatly between radionuclides. The noble gases (krypton, xenon) primarily expose the individual to an immersion dose during the passage of the cloud of gases. Their impact, in terms of total effective dose equivalent (TEDE), is from 5 to 10 orders of magnitude less than for the same level of activity of ^{239}Pu or other actinides as particles in the respirable size fraction. However, the noble gas dose will be prompt as opposed to the fifty-year cumulative dose associated with alpha-emitting actinides. Accordingly, such gases may represent a greater threat for acute health effects than the higher specific activity alpha-emitting actinides.

The most significant gaseous radionuclide handled outside of cladding matrices is tritium. Tritium is a special case where factors in addition to ARFs and RFs are especially significant. These other factors are the form of the tritium (i.e., elemental tritium or tritium oxidized to a molecular form), which significantly affects the potential dose from exposure, and the exposure modes, which include transpiration. These other factors relevant to tritium are not addressed in this document.

2.2 VAPORS (CONDENSIBLE GASES)

2.2.1 Summary of Analysis of Data

- For generation of vapors plus release from physical containment, the recommended ARF is 1.0. All materials in the gaseous state can be transported and inhaled; therefore, an RF of 1.0 is assumed for the purposes of these analyses.

2.2.2 Discussion

Vapors (materials in gaseous form due to local conditions) may result from two phenomena: chemical reaction and heating. Some vapors result from chemical reactions that generate a volatile compound (e.g., halogens in an oxidizing, acidic environment). Other vapors can be generated when the local temperature exceeds the boiling point of the element or compound (e.g., evaporation of water). Under most conditions, the ARF (the fraction of vapor formed initially airborne) assumed for vapors is 1.0. If the local conditions are not adequate for quantitative vaporization of all the material (e.g., inadequate chemical reactants, inadequate temperature), the ARF is the fraction of the material converted to vapor form. Release of vapors generated during inadvertent nuclear criticalities is covered in Chapter 6.

Loss for chemically reactive materials is difficult to quantify due to the uncertainty of the materials encountered along the pathway, the kinetics of these reactions, and the transport of the vapors to the surfaces. A conservative value is to assume all the material released is transported to the facility/environment interface without loss unless engineered emission control devices (e.g., for radioiodine - impregnated charcoal filters, silver substituted zeolite filters, silver nitrate coated ceramic saddles; HEPA or other filtration devices for condensed vapors or vapors adsorbed onto pre-existing particles) are present for removal of the specific material. In many cases, an assumption of complete transport of the airborne material without significant loss is very conservative but transport losses must be substantiated for the specific configurations associated with an event.

Many chemically volatile compounds are reactive and can be lost in transit by reaction with materials encountered along their path to the facility/environment interface or adsorption on pre-existing airborne particles. Temperature sensitive materials can condense homogeneously (particles formed directly from the vapor have been observed to be in the sub-micron range) or on pre-existing particles. Aerosols form rapidly since entrainment of cooler air invariably accompanies the formation process. Various natural processes act to attenuate transport of particles (e.g., agglomeration, gravitational settling, turbulent diffusion) and filtration or other engineered devices such as water sprays have varying removal efficiencies for particles.

For temperature-sensitive vapors (e.g., metal vapors generated at high temperatures, tritiated water vapors), the amount of material volatilized can be estimated by the amount of heat energy present and/or generated by the event. Similarly, condensation may also be calculated by heat transfer at the surfaces or by homo- or heterogenous condensation in air. The mass flux of vapors to a cool surface (diffusiophoresis) can be an effective mechanism to sweep small diameter (i.e., submicron) particles from the air.

3.0 LIQUIDS

In order for a liquid to be made airborne, in most realistic situations, the bulk liquid must be subdivided into particles/droplets small enough to be entrained in the local airflow. In some cases, it may be possible for the activity coefficient of the solute to be adequately reduced so that some material may be made airborne by vaporization.

This section describes mechanisms by which two types of liquids (aqueous solutions and organic, combustible solvents) become airborne; the descriptions are based on experiments. The mechanisms discussed for aqueous solutions include thermal stress, explosive release (i.e., shock, blast, and pressurized venting effects), free-fall spills, and aerodynamic entrainment (resuspension). Organic liquids are specifically discussed in relation to thermal effects.

3.1 SUMMARY OF ANALYSIS OF DATA

Aqueous Solutions

Thermal Stress

- Heating of aqueous solution in flowing air without surface rupture of bubbles. For the airborne release of bulk liquid during heating of aqueous solutions in flowing air without noticeable bubbles breaking on the surface of the bulk liquid, conservative values are based upon the experimental data available.

Median	ARF 6E-7/RF 1.0
Bounding	ARF 3E-5/RF 1.0

- Boiling (bubbles continuously breaking the surface of the bulk liquid with <30% of the volume of the liquid as bubbles) of aqueous solutions in flowing air. A bounding ARF for the airborne release from the bubble-burst at the surface for aqueous solutions exceeds all measured values. In the absence of a measured size distribution for the airborne droplets, a conservative value of 1.0 is assumed for the RF bound.

Median	ARF 1E-3/RF 1.0
Bounding	ARF 2E-3/RF 1.0

Explosive Stress

Releases are discussed for detonation shock effects, detonation or deflagration blast effects, deflagration pressurized venting effects, and general pressurized venting. The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock Effects. For detonations in or immediately contiguous to a pool of liquid, a bounding respirable release is assessed to be the mass of inert material equal to the calculated TNT equivalent. At low mass ratios, the respirable release is comparable to the total material release. As mass ratios increase, the respirable fraction becomes significantly less than the total amount of material released, which decreases with increasing mass ratio as well.

- Blast Effects. For detonations and deflagration at a distance where the pressure impulse is essentially equal to a flow parallel to the surface of the liquid, an ARF of 4E-3/hour (1E-6/second) for the time the pressure pulse is over the liquid and an RF of 1.0 are conservatively assumed.

- Venting of Pressurized Liquids. There are three main regimes of pressurized venting of liquids: (1) venting below liquid level, (2) venting above liquid level, and (3) venting of superheated liquid (i.e., flashing spray). This phenomena covers general pressurized venting, including deflagration induced pressurized venting effects.

  — Depressurization of liquid via a failure under the liquid surface level. Liquids covered are those at or below their boiling points. Bounding ARF and RF are estimated using the mass fraction of droplets 10-μm and less in diameter formed by commercial spray nozzles (device designed to produce small drops) under conditions that will exceed those anticipated for most accident situation (3.25-mm diameter orifice at 200 psig upstream pressure).

  Bounding ARF 1E-4/RF 1.0

  — Depressurization of containment via a failure above the liquid level or overall containment failure. Values are determined for elevated levels of dissolved gases and gas pressure pulses above liquids. Liquids covered are those at or below their boiling points.

3.0 Liquids; Summary

The ARF and RF values depend on the maximum liquid or vessel failure pressure and the density of solution. Two regimes are defined for liquid venting or vessel failure pressure. The first regime is pressure equal to or less than 0.35 MPa_g (~ 50 psig). This value is independent of solution density. The second regime is for pressures greater than 0.35 MPa_g up to 3.45 MPa_g (~ 500 psig). In this regime a distinction is made between general aqueous solutions and concentrated heavy metal nitrate solutions such as uranium nitrate hexahydrate (UNH) or plutonium nitrate. The distinction is gross, with any solution having a density $\geq$ ~ 1.2 g/cm^3 being considered a concentrated heavy metal solution. The highest release fraction is for aqueous solutions.

<u>Low Pressure (0.35 MPa_g or less)</u>	
Bounding (all solutions)	ARF 5E-5/RF 0.8
<u>High Pressure (> 0.35 MPa_g)</u>	
Median (aqueous solution, density ~ 1 g/cm^3)	ARF 3E-4/RF 0.9
Median (conc. heavy metal solution, density > ~ 1.2 g/cm^3)	ARF 2E-4/RF 0.3
Bounding (aqueous solution)	ARF 2E-3/RF 1.0
Bounding (conc. heavy metal solution)	ARF 1E-3/RF 0.4

If the containment failure is located above the critical freeboard height, allowing rapid pressurization and depressurization, the ARF and RF values may be significantly less than those noted above. For failures below the critical freeboard height, a potential method for estimating release fractions is discussed in subsection 3.2.2.3.2.B. However, difficulty in defining a critical parameter, density of gas released, precludes clear use of the method at this time. The values noted above will be conservative for such cases.

— Depressurization of liquid above boiling point. Three different values are defined for flashing spray release based on the degree of superheat: liquids with less than 50 °C superheat above the boiling point of the liquid; liquids with superheats between 50 and 100 °C above the boiling point of the liquid; and liquids with greater than 100 °C superheat. The ARF appears to increase with decreasing source size and volume. The values used in the experiments for these parameters are much below

those anticipated under most accident situation (100 ml), and therefore may be very conservative for many applications.

≤ 50 °C Superheat	
Bounding	ARF 1E-2/RF 0.6

50 to 100 °C Superheat	
Median	ARF 2E-2/RF 0.7
Bounding	ARF 1E-1/RF 0.7

For superheats greater than 100 °C above the boiling point of the liquid, the ARF value is 0.33 $(MF_g)^{0.91}$ (where MF_g is the mole fraction of pressurizing gas/water vapor flashed) with an RF of 0.3 if the calculated respirable release exceeds the 50 to 100 °C superheat value.

Free-Fall Spill

- Free-fall spill of aqueous solutions, 3-m fall distance. A distinction is made between general aqueous solutions and concentrated heavy metal nitrate solutions such as uranium nitrate hexahydrate (UNH) or plutonium nitrate. The distinction is gross, with any solution having a density ≥ ~ 1.2 g/cm^3 being considered a concentrated heavy metal solution. The highest release fraction is for aqueous solutions.

Median (aqueous solution, density ~ 1.0 g/cm^3)	ARF 4E-5/RF 0.7
Median (conc. heavy metal solution, density ≥ ~1.2 g/cm^3)	ARF 1E-6/RF 0.3
Bounding (aqueous solution)	ARF 2E-4/RF 0.5
Bounding (conc. heavy metal solution)	ARF 2E-5/RF 1.0

- Free-fall spills of slurries, 3-m fall distance, <40% solids.

Median	ARF 2E-5/RF 0.7
Bounding	ARF 5E-5/RF 0.8

- Free-fall spills of viscous solutions, viscosity >8 centipoise.

Median	ARF 3E-6/RF 0.8
Bounding	ARF 7E-6/RF 0.8

- Free-fall spills of aqueous solutions, slurries and viscous solutions, fall distances >3 m. The empirical correlations for ARF and drop size distribution parameter presented by Ballinger et al. (January 1988) are assessed to be adequate provided ARF x RF value exceeds bounding 3-m values.

Aerodynamic Entrainment and Resuspension

There appear to be very large differences in suspension rates under experimental test conditions as well as an order of magnitude uncertainty in measurements for individual conditions. On this basis, conservative values are applied.

- Indoors, on heterogeneous surface (stainless steel, concrete), low airspeeds up to normal facility ventilation flow; Outdoors, pool for low windspeeds.

  Bounding ARR 4E-7/hr; RF 1.0

- Indoors, on heterogeneous surfaces, covered with debris or under static conditions.

  Bounding ARR 4E-8/hr; RF 1.0

- Outdoors, from large pools/ponds, higher windspeeds to 30 mph.

  Bounding ARR 4E-6/hr; RF 1.0

- Outdoors, absorbed on soil, no lengthy pooling, windspeeds to 50 mph.

  Bounding ARR 9E-5/hr; RF 1.0

Use of the above values for short time frames (<100 hours) would not introduce serious error due to the severe depletion of the source. For time periods exceeding 100 hours, the reduction of the source can be accounted for from the entrainment of material. In general, these values are intended for immediate post-accident conditions and freshly deposited material. They would overestimate cumulative releases from long-term contamination of surfaces (i.e., months to years) and are not appropriate for such use.

Organic Combustible Liquids

This section specifically addresses potential releases of radioactive contaminant due to thermal stress for organic solutions or organic and aqueous solutions present together.

Thermal Stress

- Volatiles (i.e., iodine) under all conditions.

Bounding	ARF 1.0/RF 1.0

- Quiescent burning, small surface area pools, or small solvent layer over large aqueous layer burning to self-extinguishment.

Median	ARF 6E-3/RF 1.0
Bounding	ARF 1E-2/RF 1.0

- Vigorous burning large pools, or solvent layer burning over limited aqueous layer with sufficient turbulence to disrupt bulk of aqueous layer.

Bounding	ARF 3E-2/RF 1.0

- Large, vigorously burning organic fire that burns to complete dryness or burning solvent over aqueous phase burning to complete dryness for both phases (typically requires external heat source).

Median	ARF 1E-2/RF 1.0
Bounding	ARF 1E-1/RF 1.0

- Aqueous solution or air-dried salts under gasoline fire on a porous or otherwise absorbing (i.e, cracks, depressions) surface.

Bounding	ARF 5E-3/RF 0.4

- Aqueous solution or air-dried salt under gasoline fire on heat conducting surface (i.e., metal).

Bounding	ARF 2E-1/RF 0.3

No experimental data on the behavior of organic, combustible liquids in response to explosive release, venting of pressurized liquid, free-fall spills, or aerodynamic entrainment were found. In general, the values provided for aqueous solutions can be used for the types of organic solvents used in material extraction operation. However, extreme thermal effects, such as explosions, require subsequent consideration of solvent fires.

3.2 AQUEOUS SOLUTIONS

3.2.1 Thermal Stress: Evaporation and Boiling

Under most realistic scenarios involving the heating of aqueous solutions during postulated accidents in nonreactor fuel cycle facilities, the relative vapor pressures of the solvent (water) and the solute (various compounds of radionuclides, generally acidic nitrate) preclude evaporation of the solute as a viable mechanism for the airborne release of the solute. Instead, the airborne release is postulated to result from the entrainment of minute drops of the bulk liquid formed by the mechanical disintegration of the surface of the bulk liquid. Mechanical disintegration mechanisms include bubble breakup during boiling, jet drops formed from the collapse of the crater remaining after bubble breakup, and secondary drops from the reentry of jet drops. Drops are carried to the bulk flow by convective and vapor flow away from the heated liquid. An increase in surface disruption would increase the airborne release, although capture of secondary drops by the large number of primary particles may place a limit on the release.

Kataoka and Ishii (April 1983) reviewed the literature and data on the entrainment of liquid droplets from the surface of a bubbling or boiling pool. Droplets are generated by bubble bursting, splashing or foaming. Some of the entrained droplets fall back into the pool and some are carried away by the streaming gas. Entrainment, E_{fg}, is defined as:

$$E_{fg} = \text{droplet upward mass flux/ the gas mass flux} = (\rho_f\, j_{fe})/(\rho_g\, j_g) \qquad (3\text{-}1)$$

where:
ρ_f = fluid density
j_{fe} = superficial velocity of liquid flowing upward as droplets
ρ_g = gas density
j_g = superficial gas velocity.

Two levels of the gas flow through the liquid upon the surface were noted:

1. bubbly flow (condition postulated for nonreactor facility accidents): small gas flow (<0.1 m/s); droplets generated by discrete bubbles rising to surface of pool and collapsing; initial velocity of entrained droplets is a function of bubble burst time, bubble diameter, density of liquid and pressure around bubble; transition to next level at ~0.1 m/s and liquid void fraction 0.3.

2. churn turbulent flow: may be dominant mechanism for post-Loss of Coolant accident in light water reactor accident conditions; initial velocity of droplets determined by momentum exchange mechanism (during breakup of liquid

ligaments formed from surface disruption); droplets generated by all three droplet generation mechanisms (i.e., bubble bursting, splashing, or foaming).

Three regions as a function of axial distance from the pool surface were identified:

1. near-surface region: immediate vicinity of surface; entrainment dependent on height and gas velocity; entrainment consists of all droplets entrained.

2. momentum-controlled region: intermediate axial distances above pool surface; entrainment consists of droplets with initial momentum to reach height and droplets whose terminal velocity is equal to or less than the superficial gas velocity; three regimes as a function of superficial gas velocity in region:

   - low gas flux: entrainment small and consists of very fine droplets; E_{fg} approximately proportional to gas flux.

   - intermediate gas flux: larger drops ejected from pool; E_{fg} increases with the 3rd or 4th power of the superficial gas velocity.

   - high gas flux: large gas slugs form and pool surface highly agitated; considerable droplets formed by splashing; E_{fg} increases with the 7th to 20th power of the superficial gas velocity.

3. deposition-controlled region: entrained droplets of size whose terminal velocity is equal to or less than the superficial gas velocity.

A simple mechanistic model was developed based on the concepts presented above. Due to the enormous number of droplets generated, the motion of individual droplets could not be followed individually, and so droplet motion was handled statistically. Important physical parameters and distribution functions essential to the modeling and calculations were developed or assumed. Correlations for the height criteria and entrainment in each region were developed as a function of:

dimensionless gas velocity, j_g^* = $\sim j_g/[\sigma_g \Delta_\rho/\rho_g^2]^{1/4}$ (3-2)

dimensionless height above surface, h^* = $h/[\sigma/g \Delta_\rho]^{1/2}$ (3-3)

gas viscosity number, $N_{\mu g}$ = $\mu_g/[\rho_g \sigma (\sigma/g \Delta_\rho)^{1/2}]^{1/2}$ (3-4)

dimensionless vessel diameter, D^*_H = $D_H/ [\sigma/g \Delta_\rho]^{1/2}$ (3-5)

where:

σ	=	liquid surface tension
j_g	=	superficial gas velocity
g	=	acceleration due to gravity
σ_g	=	surface tension of gas
ρ_g	=	gas density
h	=	height above pool surface
μ_g	=	viscosity of gas
Δ_ρ	=	density difference between gas and liquid
D_H	=	diameter of vessel.

All correlations agreed well with the published data and could be applied to estimate ARF for specific scenarios. The correlations are relatively straightforward although the values depend upon parameters that are not readily quantifiable for many practical situations and vary with temperature. Methods to determine the temperature or the values for the parameters as a function of temperature were not presented. Use of the correlations for this study would require the definition of a range of accident scenarios (not done as of this time) to determine a bounding ARF and RF. The results indicate that large variations may be observed in measured data dependent upon the location and configuration of the sampling system.

Four important observations arise from review of literature on entrainment of liquid droplets from bubbling or boiling pools performed by Borkowski, Bunz and Schoeck (May 1986):

1. the influence of surface effects on the amount and composition of the generated aerosols;

2. the possibility of chemical enrichment and depletion of substances in aerosols;

3. the existence of two groups of droplets with different mean sizes and amounts of airborne mass;

4. the limited range of ejected jet droplets due to initial velocity;

Droplet formation during boiling is dependent upon conditions of boiling and bubble characteristics. There appear to be at least two and possibly three boiling regimes that affect bubble and droplet formation. The first regime occurs at lower rates where the volume fraction of the bubbles is less than 30%, when discrete bubbles rise through the liquid and grow due to decreasing hydrostatic head. Bubbles may coalesce or divide during ascent. Droplets are formed from three mechanisms (bubble film disintegration, jet drops from crater collapse, and secondary droplets from jet drop reentry into bulk liquid). This regime is the predominant concern for nonreactor facility accident scenarios. A second regime occurs at

higher boiling rates; the liquid is turbulently mixed and progressively disintegrates at the surface forming drops from both mechanisms. A possible third regime occurs at very high boiling rates when splashing and foaming dominate the surface (Borkowski, Bunz and Schoeck, May 1986).

Gas flow conditions and material characteristics are important parameters in bubble-induced droplet formation. Bubble size determines the number and size of the droplets formed. Bubble size is determined by the volume of vapor, surface characteristics such as surface tension, and bubble contact angle. Contact angle changes due to local turbulence during bubble formation resulting in a distribution of bubble sizes. Many bubbles are unstable and coalesce and break up during ascent. Steam bubbles are in the range of 0.5 to 5 cm diameter at low pressure and nucleate boiling (presence of rough surface, suspended particles). The formation and detachment of macro-bubbles is a function of contact angle of the liquid and the degree of superheat.

Bubble shape at the surface may range from spherical to hemispherical depending on size. The liquid in the dome of the bubble runs down the sides and thins the film. The bubble bursts when the internal pressure exceeds the external pressure and surface tension of the film. Droplets are formed by the film breakup. The crater remaining from the bubble rupture itself collapses forming an ascending liquid jet that decays into droplets after some critical length. Jets ascend up to 20 cm from the surface of the bulk liquid. Jet drops are only formed from bubbles <5 to 6 mm in diameter. Droplets from film breakup are only formed for bubbles >0.2 mm in diameter. Therefore, by inference, only jet drops are formed from bubbles <0.2 mm in diameter and only film breakup droplets are formed from bubbles >6 mm in diameter. The number and size distribution of droplets formed from film breakup correlates with the size of the bubble and may number into the hundreds for the upper limit of bubble diameter. Figure 3-1, reproduced from the reference document, shows a number distribution from the burst of two bubbles of 0.1% NaCl in water. Only one jet drop ejected from collapse of a bubble ~2-mm in diameter with up to 6 ejected from very small diameter bubbles (high internal pressure). The diameter of the drop is ~20% of the bubble diameter (100- to 1000-μm for the conditions covered here) (Borkowski, Bunz and Schoeck, May 1986).

3.2.1.1 Heating of Shallow Pools

The airborne release during heating of aqueous solution was measured and reported by Mishima, Schwendiman and Radasch (November 1968). This study involved the collection and measurement of airborne Pu during drying of shallow pools of concentrated acidic plutonium nitrate solution at three air velocities and the evaporation of 90% of the volume of a dilute acidic plutonium nitrate solution. Table 3-1 displays measurements extracted from

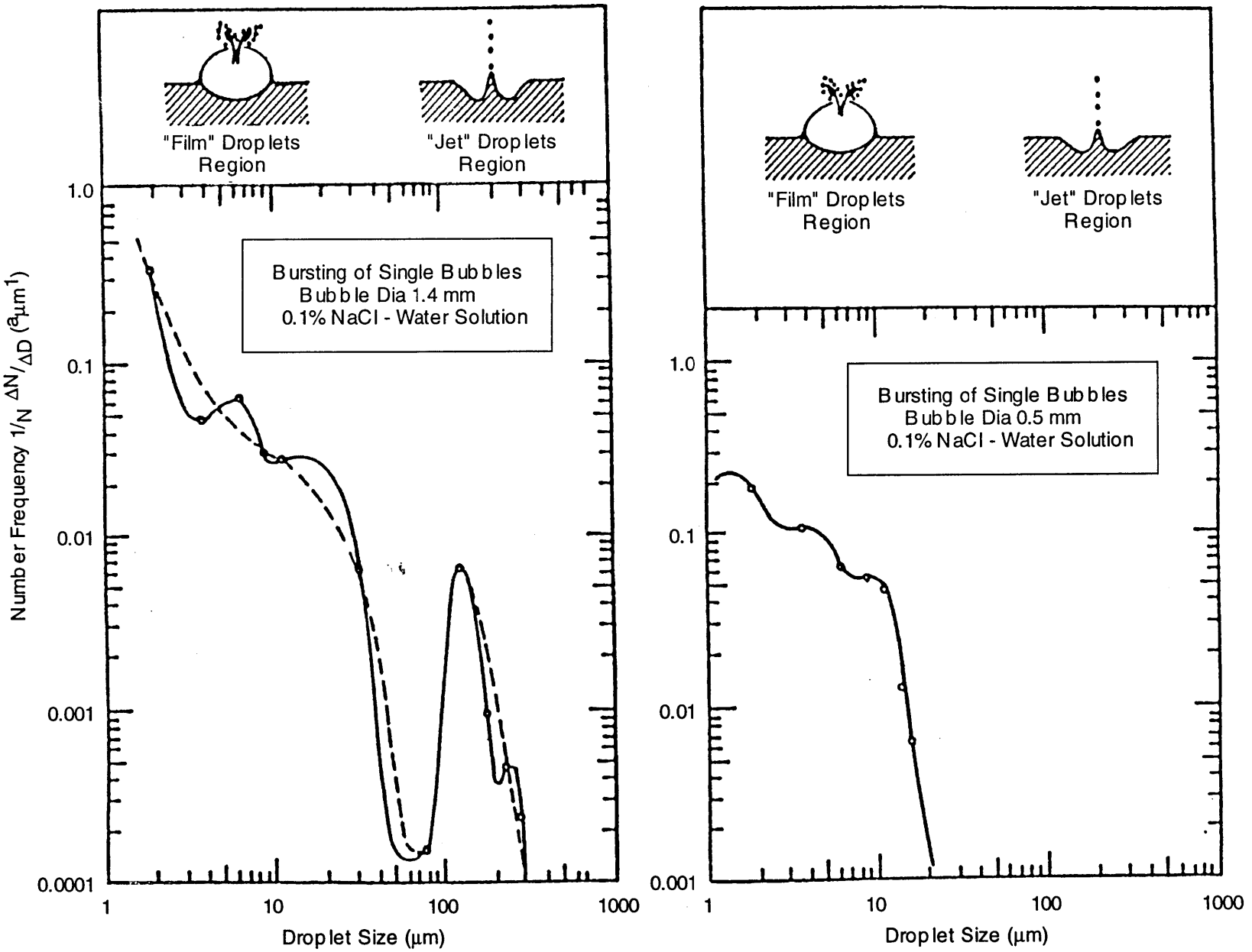

Figure 3-1. Size Distribution of Bubble Induced Droplets
(Borkowski, Bunz, and Schoeck 1986)

Table 3-1. Fractional Airborne Releases During the Heating with Flowing Air of Concentrated Plutonium Nitrate Solution (Table 1 from Mishima, Schwendiman, and Radasch, November 1968)

Temp, ° C	Air Velocity, m/s	Sampling Time, hr	ARF
50	1.0	2	1.3E-7
75	0.5	5	<1.0E-8
90	0.5	2	5.3E-7
90	1.0	1.5	5.7E-7
100	0.1	2	1.0E-5
100	0.5	2	3.0E-5

the study reference document (reproduced as Table A.1 in Appendix A) and shows the ARFs from evaporation of concentrated plutonium nitrate solutions under three air velocities (0.1, 0.5, and 1.0 m/s).

A schematic diagram of the experimental apparatus is shown in Figure A.1 (Appendix A). Approximately 2.5 to 3 ml of a concentrated $Pu(NO_3)_4$ solution containing from 0.72 to 0.86 g Pu were placed in a shallow depression (~25.4-mm diameter x ~2.4-mm deep) in a 31.8-mm diameter x 6.35-mm deep stainless steel dish. The dish was placed in a teflon retainer that filled half of the diameter of a 38.1-mm diameter borosilicate glass tube. Filtered room air was drawn through the tube at three nominal velocities (0.1, 0.5, and 1.0 m/s) over the solution and through a water-cooled condenser to remove excess moisture; the airborne particles were then collected on an in-line glass fiber filter. The liquids were heated to various temperatures by heat lamps positioned over the liquid. The evaporation times ranged from 1.5 to 24 hours. None of the solutions were observed to boil during any of the experiments and the airborne release is most probably due to the aerodynamic breakup of the surface, with release increasing as temperature increases due to reduced surface tension.

The airborne fractional releases measured are shown in Table 3-1; ARFs range from <1E-8 to 3E-5. The highest ARFs were measured at the highest temperature (1E-5 and 3E-5 at 100 °C). The limited data also tend to indicate some increase in airborne release with increasing air velocity. The surfaces during the drying were relatively undisturbed (no visible surface disturbance). The upper bound release is 3E-5, and, in as much as the size distribution of the airborne materials was not measured, a conservative value of 1.0 is selected for the RF. The median value is 6E-7 (5.5E-7 rounded upward) with an average value of 7E-6.

The data are limited but do appear to consistently indicate a gradual increase in airborne release with temperature until boiling or near boiling temperatures. The air velocity range is very limited although the air velocity probably represents a much greater aerodynamic stress on the surface than the nominal velocity indicates. Air velocity measurements are usually at much greater distances from the surface than in the experimental apparatus, and, air being a fluid, the velocity decreases with distance from the surface due to frictional forces. The extremely small liquid sample sizes would also enhance effective stress as well. The concentrated plutonium nitrate solution used represents a very important class of liquids found in DOE facilities but its fluid characteristics (higher density, surface tension) may not make it bounding for other aqueous solutions.

3.2.1.2 Heating of Pools

ARFs were also measured during the evaporation of 90% of the volume of aqueous solutions at three surface disturbance levels: simmering, disturbed surface, and boiling (Mishima, Schwendiman and Radasch, November 1968). The results from the source document are reproduced in Table A.2, and the experimental apparatus is shown in Figure A.2, both in Appendix A. In the experiments, 100 ml of a dilute $Pu(NO_3)_4$ solution (0.25 $\underline{M}$ HNO_3) containing 0.7 mg Pu was placed in a 180 ml borosilicate beaker. The surface area of the liquid was 11.5 cm^2. The beaker was held in the center of a transite support ring that positioned the beaker in a aluminum plate set upon a hot plate. A screen supporting a glass fiber filter filled the annular area between the support ring and beaker and allowed air to be drawn through the 4-liter borosilicate glass bell jar to entrain particulate material escaping from the beaker. The velocity through the annular filter was estimated to be 3 cm/s. The air was drawn out of the top of the bell jar via a water-cooled condenser to remove moisture. The condensate was collected. Airborne material was collected on an in-line glass fiber filter.

The pertinent results are shown in Table 3-2. The ARFs for the four runs at boiling ranged from 4.5E-7 to 1.8E-3. Three of the four values ranged from 3E-4 to 1.8E-3. The two highest values (1.1E-3 and 1.8E-3) were estimated by the sum of the filter acid leach and from acid washes of the equipment downstream of the filter position due to loss of the filter. These values include solution splattered from the vessel onto the glassware. The values are almost certainly high, but it is not known how high. Two of the runs (ARFs 4.5E-7 and 1.1E-3) used a 0.70 mg Pu source without an airflow. The two runs performed at simmering (no surface breaking) resulted in ARFs of 1.3E-6 and 4.5E-6 that are bounded by the evaporation value of 3E-5 quoted in subsection 3.2.1.1. The four experiments with heating rates resulting in disturbed surfaces generated ARFs ranging from 5.8E-5 to 8.4E-4: an order of magnitude variation in estimates. A bounding value for boiling of aqueous solutions of 2E-3 is selected, and, in the absence of a measured particle size distribution, a

Table 3-2. Fractional Airborne Release During Heating of Pools of Dilute Plutonium Nitrate Solution - 90% Volume Reduction (Table II, Mishima, Schwendiman, and Radasch, November 1968)

Average Hot Plate Temperature, ° F	Average Boil-Off Rate, ml/min	Minutes Heated	Appearance Surface	ARF
150	0.6	151	Simmering	4.5E-6
150	0.5	150	Simmering	1.3E-6
164	0.66	121	Disturbed	5.8E-5
175	0.73	124	Disturbed	2.4E-4
188	1.2	64	Disturbed	8.0E-5
190	0.9	80	Disturbed	8.4E-4
200	1.4	66	Boiling	1.1E-3[a,b]
218	1.4	63	Boiling	1.8E-3[a]
218	1.4	59	Boiling	3.0E-4
220	2.1	42	Boiling	4.5E-7[b]

[a] Filter ruptured, estimate based on activity collected in acid washes of equipment downstream of filter position.

[b] Only 0.07 mg Pu used as source. No air sweep used during these experiments. ARF estimate from activity collected in condensate.

conservative value for an RF of 1.0 is selected. The median value for the range of surface disturbance levels is 6E-5 (6.4E-5 rounded off) with an average value of 7E-4.

The fraction of source material exiting the container and depositing nearby (fallout) ranged from ~3E-8 during simmering to 1.7E-2 during boiling. This material is considered generally indicative of the liquid ejected from the container but not airborne during such event. As with the airborne materials, the fraction ejected will increase with the increase in surface disturbance.

The data are limited for each type of heated liquid. The loss of the filters coupled with not using air sweeps in two of the boiling experiments makes the data for that type of heating especially uncertain. The surface of the heated liquid is recessed from the airflow and may reduce the airborne material due to losses to the sides of the beaker prior to escape. The configuration may be indicative of airborne release from heated liquids from the tops of vessels. These types of considerations are why the higher values obtained are selected, even though they represent overestimates to some degree due to equipment failure and are considered excessive for the experimental configuration. The liquids are very dilute aqueous solutions and should bound other more viscous liquids or those with greater surface tension.

3.2.1.3 Additional Evaporation and Bubbling Release Studies

Borkowski, Bunz, and Schoeck (May 1986) reviewed 12 experimental studies that examined the airborne release of dissolved matter from bubble-burst at the surface of aqueous solutions. The slope of the rate change for fraction entrained as a function of gas velocity changes at a gas velocity of 15 cm/s. The experimental data reported in this region are reproduced in Figure 3-2. Data generated by the venting of pressurized liquids is also presented as Figure 3-3 to assist in overall phenomena assessment.

The data by Mishima, Schwendiman, and Radasch (November 1968) covered in subsection 3.2.1.2 are plotted along with data from six other studies under reasonably comparable conditions. Manowitz et al. (1955) measured the DF (decontamination factor, the ratio between the radioactivity retained in the liquid in the vessel to that boiled off) during evaporation of waste solution using a de-entrainment device (not specified). The DFs ranged from 1E-4 to 1E-5 depending upon the boiling rate and contents suspended in solution. Garner et al. (1959) performed experiments at reduced pressures and equilibrium conditions to identify the main parameters for liquid entrainment during evaporation. Entrainment increased with evaporation rate and decreasing solute concentration. Entrainment rates ranged from 1E-5 to 1E-4. Garner et al. (1954) measured the drop size distribution and total entrainment during evaporation in vessels of various diameters (4-in. and 12-in. diameter tubes). Entrainment rates were in the 1E-5 range. Although ~95% of the drops were in the < 20-μm diameter range, the total mass entrained was primarily due to the drops > 100-μm in diameter. Shor et al. (1957) measured the radioactivity carried over in boilers by continuous monitoring of trace ^{137}CsCl driven airborne at elevated pressures (0.93 to 1.0 Mpa). Entrainment (1E-6 to 1E-4) correlated with boiling rate and had an initial high burst of activity released. Heger et al. (1982, 1983) conducted bubbling experiments to simulate reprocessing plant components. Stirring air flow velocity was ~10 m/h (2.8 cm/s). Entrainment values ranged from 1E-7 to 1E-4. The presence of tributyl phosphate (TBP) reduced the surface tension and increased entrainment by a factor of 5 to 10. The drops airborne were bimodally distributed with maxima at 0.3 and 0.8-μm diameter. Addition of the TBP increased the generation of larger diameter drops.

It is evident that the ARF of 2E-3 measured by Mishima, Schwendiman and Radasch (November 1968) bounds the value measured by the other reported studies by approximately an order of magnitude. Data generated by the venting of pressurized liquids shown in Figure 3-3 indicate that short of flashing spray conditions (superheating of the liquid by pressurization), a release value in the range of 1E-3 will bound the airborne release of liquids during boiling at normal atmospheric pressures. Thus, a bounding ARF of 2E-3 with an RF of 1.0 is considered a very conservative bound for the airborne release of respirable size drops during accident condition resulting in the boiling of aqueous solutions.

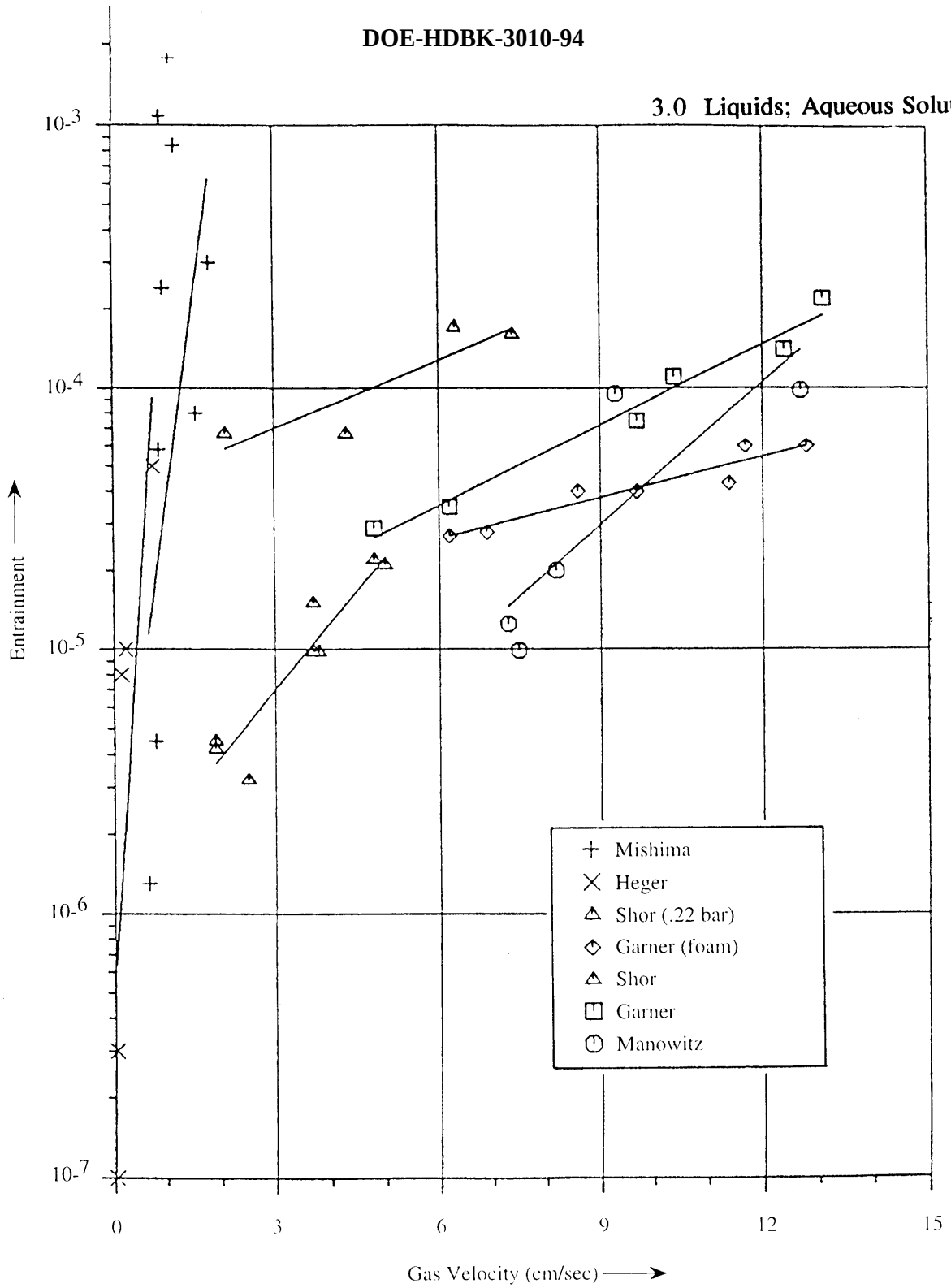

Figure 3-2. Entrainment Data Obtained at Small Gas Velocity (Borkowski, Bunz, and Schoeck May 1986)

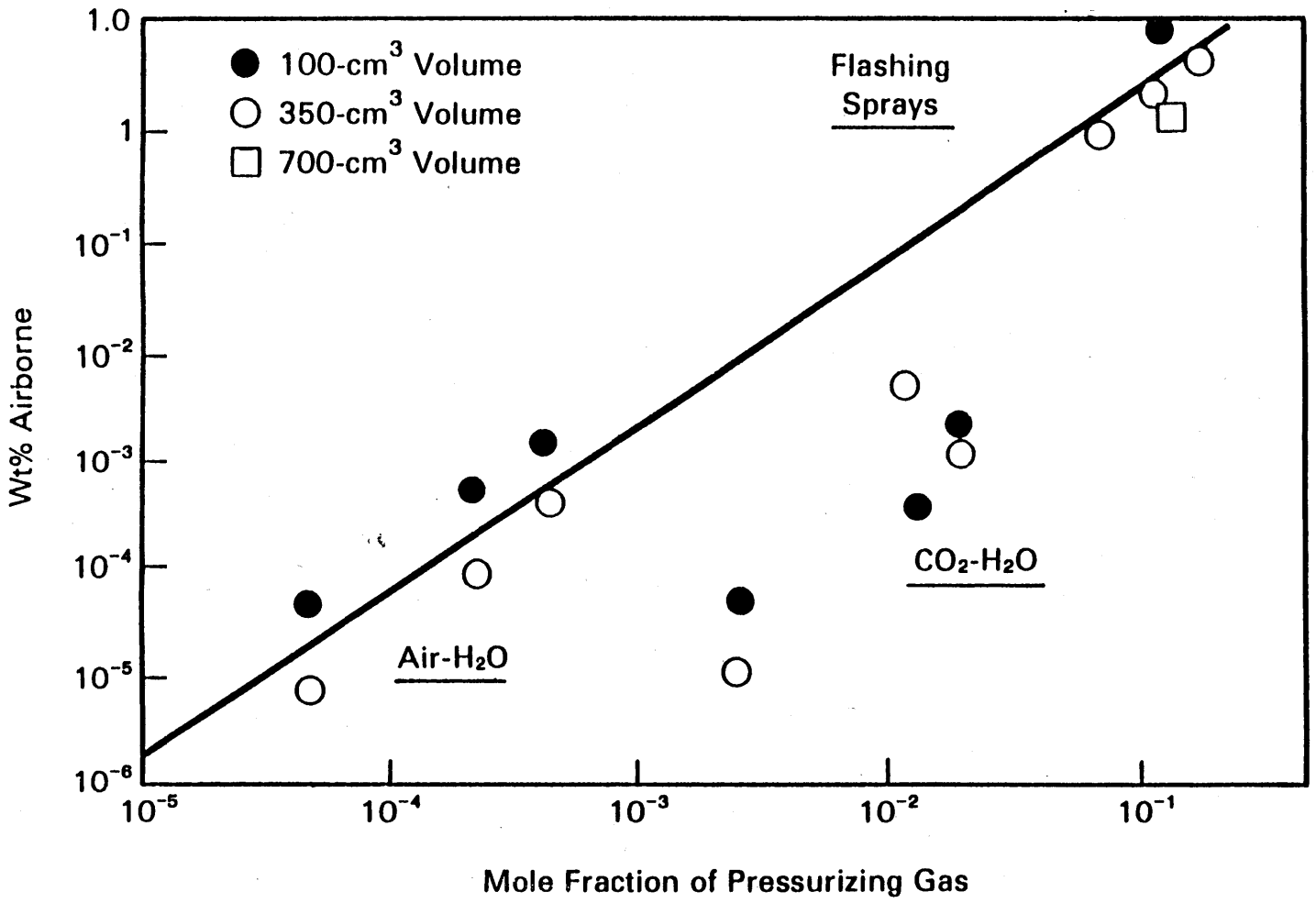

Figure 3-3. Weight Percent Airborne as a Function of Mole Fraction of Pressurizing Gas (Ballinger, Sutter, and Hodgson May 1987)

3.2.2 Explosive Stress: Shock, Blast, and Venting

Liquids may be subdivided by the shock generated by detonation-like reactions or by shear stress at the surface generated by the accelerated airflow from the blast. Venting of pressurized liquid phenomena is related to explosions as well.

3.2.2.1 Shock Effects

Steindler and Seefeldt (1980) provide an empirical correlation to experimental data on the fragmentation of metals and aqueous solution by detonations [energy releases in microseconds with brisance (shattering effect)] (Ayer, et al., May 1988). The experiments performed by others were used to relate releases to mass ratios (i.e., ratio mass of inert material to TNT equivalent) of 1 to 15. The experiments were conducted with the condensed phase explosive embedded or contiguous to the material affected. Estimates of the ARF and size distribution for various mass ratios up to 1000 are provided in Appendix C of Ayer, et al. (May 1988) for a GSD (Geometric Standard Deviation, the slope of the line on log probability plot) of 8. The GSD is much greater than normally assumed (GSD 2) due to potential uncertainties regarding actual energy distribution. The fragmentation of only a portion of the inert material occurs when MRs are large, and the fraction of shock energy absorbed by this unknown portion of material is likewise unknown.

All inert material is driven airborne as particulates in the respirable size range for an MR of one. The volume of material exposed to the shock effects of the explosion is essentially constant for an explosive charge of a given size as the surface area in proximity to the charge is fixed. However, as the amount of inert material increases, the fraction of the total mass exposed to significant shock effects in that volume decreases. Given the potential uncertainties of the data, this simple relationship will be used to extrapolate respirable releases from the point of maximum release (i.e., MR = 1).

A respirable release of inert mass equal to the TNT equivalent for the detonation (to a maximum of 100% release) is considered to bound the Steindler and Seefeldt (1980) correlation. This assumption is supported by numerical comparison. For MRs of 5, 10, and 15, the combined ARF x RF values predicted by the correlation for a GSD of 2 are 4E-3, 6E-4, and 3E-4 respectively. For MRs of 5, 10, and 15, the combined ARF x RF values predicted by the correlation for a limiting GSD of 8 are 2E-1, 9E-2, and 7E-2 respectively. The simple method used by this handbook would predict combined ARF x RF values of 2E-1, 1E-1, and 7E-2, which correlate with the values for a GSD of 8 that are believed to be very conservative.

3.2.2.2 Blast Effects

The blast effect of interest is accelerated gas velocities passing over the surface of the liquid. Mishima and Schwendiman (August 1973) reported the results of measurements of the airborne release of uranium from various surfaces (soil, vegetated soil, stainless steel, asphalt with UO_2 powder or UNH solution) before, during and after gasoline fires in a wind tunnel at air velocities of ~1 m/s and ~10 m/s. The flame speed in flammable vapor mixtures is also on the order of 10 m/s, although flame speed may propagate to sonic velocities under turbulent conditions. The results are reproduced in Table A.3 and the experimental apparatus is shown in Figure A.3 in Appendix A. The only experiments involving UNH solution were performed on a substrate of loose, sandy soil at air velocities of~1 & 10 m/s. The ARF measured at 10 m/s from soil during a 28-hour sampling period was 3.9E-4 with an RF of 0.68. The value is comparable to other experiments involving UNH residues from the fire except for one result from stainless steel at 10 m/s (ARF 2.6E-2 in 6 hours/RF 0.3; linear rate 4E-3/hr). Therefore, the rapid passage of air at an accelerated velocity from the deflagration of a flammable vapor mixture does not appear to have the potential to release large amounts of material from aqueous liquids. The ARF is assessed to be 4E-3/hr for the time duration of impulse passage over the liquid (generally on the order of 1 second).

3.2.2.3 Venting of Pressurized Liquids

Liquids, contained in a vessel or containment, can be pressurized by external sources (e.g., deflagration in a free volume above the liquid, pressurized gases from an external source) or by vapor generated by the heating of liquid. If the pressure exceeds the strength of the vessel/containment, the vessel/containment will fail and, under the proper circumstances, can release the liquid to the atmosphere as liquid drops.

Droplets of an aqueous solution under pressure can be generated by spray if the pressure is relieved by venting a cold liquid through a small opening in the wall of the vessel, by the bubbling action on the surface of the liquid resulting from the release of gases absorbed/trapped in the cold liquid, and by fragmentation of the liquid by bulk vaporization when the pressure over a superheated solution is relieved (i.e., flashing spray). The characteristic of the liquid, the source of the pressure, and the location of the failure all have an effect on drop formation and release.

Pressure driving the release is estimated by the strength of the external source, temperature/vapor pressure of the liquid, the temperature/volume expansion of the gases, etc., depending on the scenario described. The point of venting is determined by the design, construction and strength of material of the vessel/containment. If the vent is located below the level of the liquid, spray formation is the mechanism for drop formation. If the vent is

above the liquid level, whether or not the liquid can release gases or vapors upon depressurization influences the drop formation mechanism. If the liquid has come to equilibrium with the free volume atmosphere, liquids under pressure will generate drops by bubble breakup at the surface from release of trapped/dissolved gases. For heated liquids, drops are generated by bubble breakup at the surface of the liquid from vapor generation.

For liquids that have not come to equilibrium with a pressurized free volume atmosphere, the bubbling action from release of absorbed/trapped gases can be minimal. Drop formation is more accurately described as a function of shear stress on the liquid surface and stratified two-phase flow. If the vent is greater than the Critical Freeboard Height above the surface of the liquid, the flow pattern tends to be vertical and does not exert significant shear stress across the liquid surface to create drops, and the relatively great distance from the point of origin of the drops to the vent does not create conditions favorable to release. For vents that are less than the Critical Freeboard Height from the liquid surface, drop formation is by shear stress exerted due to the induced, high velocity flow parallel to the liquid surface. While this release mechanism can result in significantly smaller release fractions than the three mechanisms cited in the preceding paragraph, it requires data that is not practically attainable in most cases. As a result, this mechanism, while discussed, will be represented by the more conservative case of release of absorbed/trapped gases.

3.2.2.3.1 Venting Below the Liquid Level. If the container or pipe holding an ambient-temperature liquid under pressure is breached, the liquid can escape in a variety of ways. Breaches venting pressurized liquids can range from pinhole leaks in pipes (generating a mist) to drips from very slow leaks to large jets of liquids that may gush from large holes. The amount and aerodynamic size distribution of the spray generated are a function of the size and characteristics of the breach, the upstream pressure, and the liquid characteristics (e.g., viscosity, density, volatility).

For the purposes of airborne suspension, a conservative assumption would be the pressurized release of the liquid via a very fine hole as occurs in a commercial spray nozzle. The size distribution of some commercial spray nozzles as a function of orifice diameter and upstream pressure were shown by Mishima, Schwendiman and Ayer (October 1978). The size distribution of the liquid drops decreases with orifice diameter and increasing upstream pressure. It is not anticipated that drops formed from breaches, cracks, leaks would generate finer drop size distributions than equipment specifically designed for that purpose. Therefore, the respirable fraction of the coarsest distribution generated by commercial spray nozzles shown in Figure 3-4 is selected as the bounding ARF, 1E-4, with a RF of 1.0. For other size fractions, the values can be inferred from the 0.128-inch (3.25-mm) diameter spray nozzle values at 200 psig (1.38 MPa_g) upstream pressure.

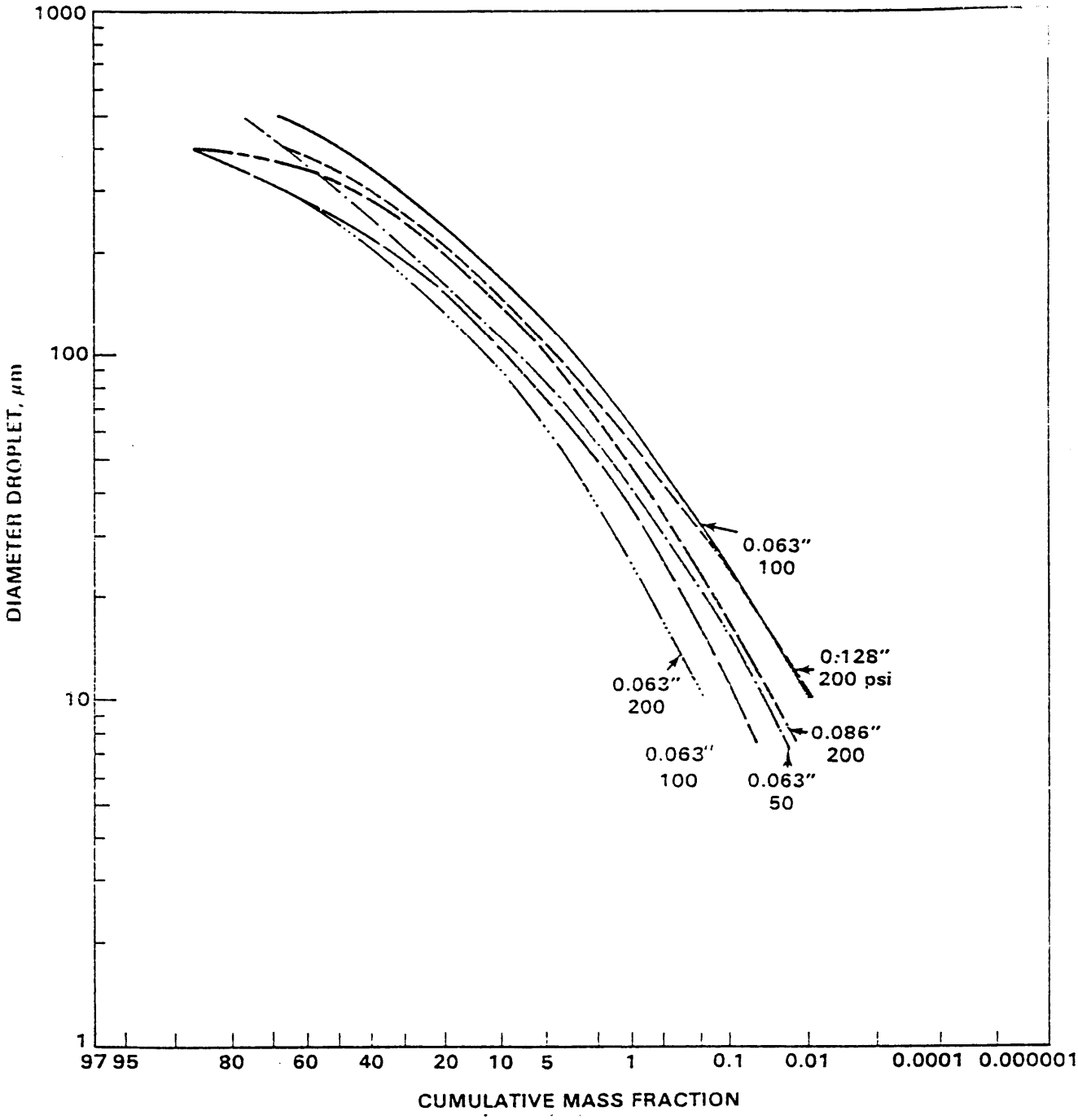

Figure 3-4. Mass Fraction vs. Droplet Diameters for Sprays as a Function of Orifice Diameter and Upstream Pressure (Mishima, Schwendiman, and Ayer October 1978)

Other recent investigations (Leach, 1993; Gieseke, Kogan and Shaw, September 1993) using an analytical model suggest that, under some conditions, the fraction of drops in the finer size fractions (i.e., 10-μm and less) are greater for fine orifices (and possibly slot-type breaches) at high pressures and that the evaporation of the liquid prior to deposition may reduce the size of the larger diameter drops to some extent. There is considerable uncertainty as to the value to assign the critical factor (Q, a drop size fitting parameter) and the analytical model, though useful in understanding the phenomenon, cannot presently be used to predict the size distribution of sprays.

3.2.2.3.2 Venting Above the Liquid Level or Overall Containment Failure. The amount of liquid entrained as droplets in depressurization flow depends on several factors. The effect of dissolved gases, surface turbulence and stratified two-phase flow are considered below.

A. Elevated Levels of Dissolved Gases. Sudden depressurization of a liquid allows the release of dissolved/trapped gases. This sudden release of gases may result in bubble formation that can create very small drops upon collapse. The drops formed can be carried with the venting gases. Figure 3-3 previously presented illustrated the relationship between the amount of gas/vapor released and the fraction airborne. Experiments were performed to measure the ARF due to venting pressurized volumes of aqueous solutions in quasi-equilibrium with their pressurizing gases (Sutter, August 1983). Data from the referenced document (reproduced as Tables A.4 through A.7 in Appendix A) describing airborne release of aqueous solutions are tabulated in Table 3-3. The experimental apparatus is shown in Figures A.4 & A.5 of Appendix A.

The average ARF as a function of pressure, solution density and source size are shown in Table 3-4 (tabulated as weight percent, 0.05 wt/o = 5E-4 fraction) from the reference and plotted in Figure 3-5. ARF increases with pressure and decreases with density and source size. Due to the pressures employed, there appears to be a significant difference between bounding ARF and RF for pressure less than 0.345 MPa_g (50 psig) and between 0.352 MPa_g (51 psig) and 3.45 MPa_g (500 psig). Bounding values for each pressure range are given. For the venting of liquids with elevated levels of dissolved gases or overall vessel failure/blowout to 0.345 MPa_g, the ARF and RF values range from 3E-6/0.8 to 5E-5/0.8. The bounding ARF and RF are assessed to be 5E-5 and 0.8.

For the venting of liquids with elevated levels of dissolved gases or overall vessel failure/blowout in a range of 0.352 MPa_g to 3.45 MPa_g, the data appears to show some dependency on experimental parameters. For the uranine solution (lower density liquid, ~1 g/cm^3), a bounding ARF of 2E-3 with a fraction of airborne

Table 3-3. Measured ARFs from Venting Pressurized Aqueous Solutions (Tables A.4, A.5, B.4 and B.5 - Sutter, August 1983)

Source Volume, ml	Pressure, psig	Material	ARF	RF	ARF x RF
350	500	uranine	6.0E-4	0.86	5.0E-4
	500	uranine	4.0E-4	0.85	3.0E-4
	500	UNH	2.0E-4	0.34	7.0E-4
	500	UNH	3.0E-4	0.37	1.0E-4
	250	uranine	7.0E-5	0.98	7.0E-5
	250	uranine	1.0E-4	0.84	1.0E-4
	250	UNH	1.0E-4	0.20	2.0E-5
	250	UNH	9.0E-5	0.45	4.0E-5
	50	uranine	4.0E-6	0.83	3.0E-6
	50	uranine	1.0E-5	0.87	1.0E-5
	50	UNH	3.0E-6	0.76	2.0E-6
	50	UNH	4.0E-6	0.70	3.0E-6
100	500	uranine	2.0E-3	0.90	2.0E-3
	500	uranine	1.0E-3	0.70	8.0E-4
	500	UNH	7.0E-4	0.46	3.0E-4
	500	UNH	1.0E-3	0.38	4.0E-4
	250	uranine	4.0E-4	0.78	3.0E-4
	250	uranine	7.0E-4	0.77	5.0E-4
	250	UNH	4.0E-4	0.36	2.0E-4
	250	UNH	6.0E-4	0.45	3.0E-4
	50	uranine	4.0E-5	NM	
	50	uranine	5.0E-5	0.80	4.0E-5
	50	UNH	2.0E-5	0.61	1.0E-5
	50	UNH	2.0E-5	0.60	1.0E-5

Table 3-4. Average Weight Percent Airborne from Pressurized Liquid Release

Pressure, psig	350 cm^3 Source		100 cm^3 Source	
	Uranine	UNH	Uranine	UNH
500	0.05	0.025	0.15	0.08
250	0.01	0.01	0.06	0.05
50	0.0008	0.0004	0.005	0.002

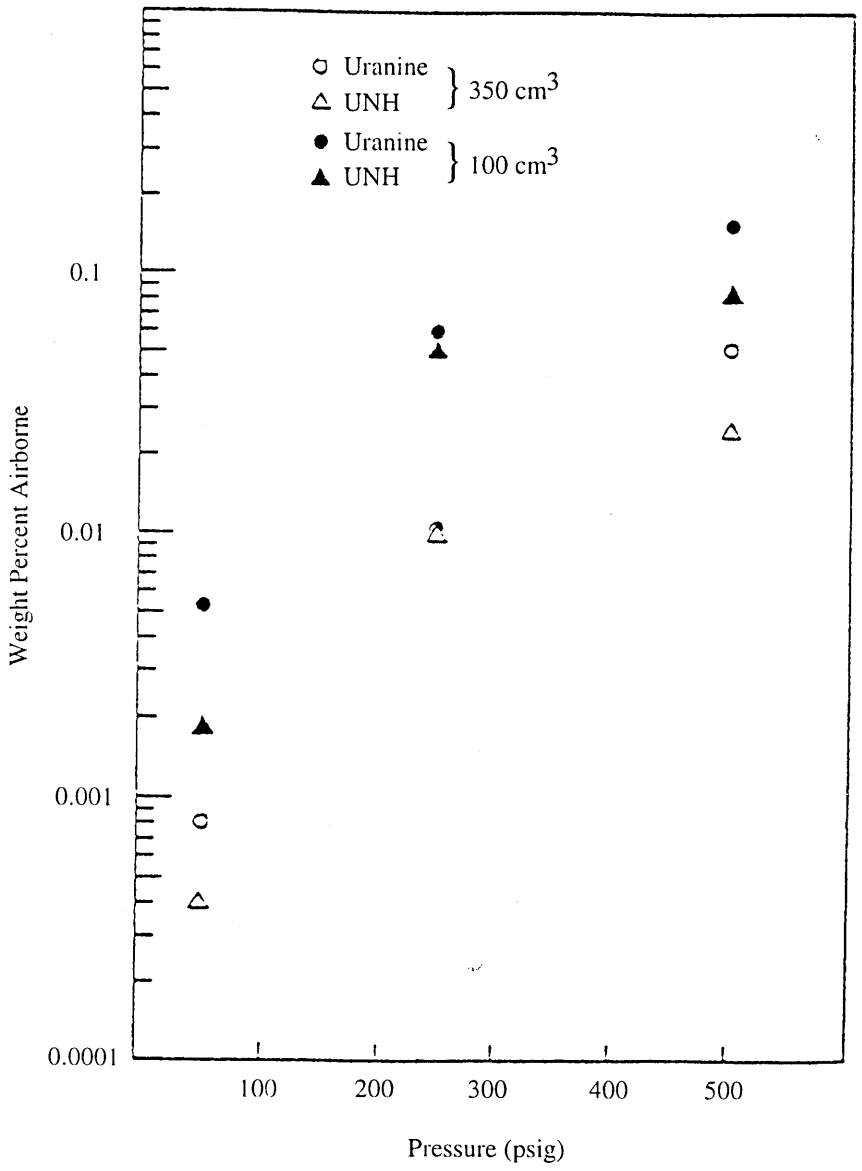

Figure 3-5. Average Weight Percent of Liquid Airborne as a Function of Pressure

material in the respirable size range of 1.0 appears conservative. For the UNH (higher density liquid, ~1.3 g/cm^3), a bounding ARF of 1E-3 (100 ml UNH at 500 psi) with a fraction of the airborne material in the respirable size fraction of 0.4 appears conservative. The "median" values for the uranine ARF and RF are 3E-4 and 0.9 and 2E-4 and 0.3 for UNH. The average values for uranine are ARF and RF of 5E-4 and 0.9 and 3E-4 and 0.5 for UNH.

For the sake of simplicity, a gross density distinction is made for determining which ARF and RF values to use. Any solution containing heavy metal salts where the liquid alone has a density in excess of ~ 1.2 g/cm^3 is considered a "concentrated heavy metal solution" for assigning ARF and RF values (i.e., 1E-3 and 0.4). Any solution containing heavy metal salts where the solution alone has a density less than ~ 1.2 g/cm^3 is considered an "aqueous solution" for assigning ARF and RF values (i.e, 2E-3 and 1.0).

B. Rapid Pressurization That Does Not Allow Gas Absorption by Liquid.
If a pressurized vessel vents, liquid contents can be suspended as droplets formed by turbulent shear stress on the liquid surface. If flow across the liquid is perpendicular to the liquid surface (i.e., entire crown of vessel is lost), the gases pass directly to the atmosphere and has little impact on the surface. Some small fraction of liquid may be suspended by the negative phase of the impulse due to the release of normally dissolved gases in the liquid.

If vent diameter is small and flow is low, little stress is created on the liquid surface. If the vent size is adequate to generate flows resulting in turbulence on the liquid surface, liquid can be suspended by drop formation. In the experimental study of stratified two-phase flow reported by Shrock et al. (August 1987), a determinant parameter for the release of airborne droplets was Critical Freeboard Height. This is the height from the gas-liquid interface to the break center where gas or liquid pull through begins. Equation 5.7 in the study report is:

$$[V/gD][(d_g/\Delta_p)^{0.5}] = 0.395(h_b/D)^{2.5} \quad (3\text{-}6)$$

where:
- V = gas velocity
- g = gravitational acceleration, 980 cm/s^2
- D = diameter of vent
- d_g = density of gas at saturation
- Δ_p = pressure differential
- h_b = Critical Freeboard Height.

For liquids that only contain dissolved/trapped gases at normal pressure and have a surface level greater than the critical freeboard height from the venting point, an undefined but insignificant fraction may be released depending on the size of the drops, the velocity of the gas to carry the drops (most of the gas has been vented prior to the formation of the drops), and the distance to the vent. Greater releases would be expected if the liquid surface level was less than the critical freeboard height from the venting point. A possible technique to estimate the fraction of these liquids released is as follows:

- Estimate the pressure based on increase in volume of the combustion product and air components raise to a temperature based on the heat generated by the deflagration.

- Estimate the volume of the combustion products and air components at standard and temperature vented.

- Estimate fraction of gas in volume vented using Equation 5.11 in Schrock et al. (August 1987).

- Estimate the mass of liquid in the gas estimated above by subtraction.

Unfortunately, the equation for estimating the Critical Freeboard Height requires estimating the gas density at the moment of pressurized release. There is no simple or recognized means other than experimentation to estimate gas density at release. The only gas density known at this point is the initial gas density at saturation, which is less than the gas density with the liquid droplets. Accordingly, the more conservative bounding estimates for the release of absorbed/trapped gases are assessed as most applicable for general use.

3.2.2.3.3 Flashing Spray. Liquids heated above the boiling temperature of the liquid/solvent/diluent "flash" upon release; that is, the excess heat above the boiling point of the liquid is expended in the bulk vaporization of the liquid and the remaining liquid is fragmented into fine droplets. The phenomenon has been investigated in experimental studies and through empirical correlations and models, as summarized below.

A. Experimental Studies. Brockman (February 1985) reviewed the literature on the possible flashing of condensed moisture during the depressurization of a LWR post-accident containment vessel. The event postulated was the vigorous boiling of water during depressurization with droplets entrained in the vapor generated. A simplified model was used to calculate the amount of water entrained. Entrainment is defined as

the ratio between mass of liquid entrained/mass of vapor generated and was calculated by the correlation developed by Rozen et al. (1970). The correlation as shown in Kataoka and Ishii (April 1983) is limited to the deposition controlled region with different correlations for the low and high superficial gas velocity regimes. The correlation used here is the general correlation for the entire region. The size distribution of droplets formed is based upon the suspension velocity and Weber breakup of liquid masses. Initial conditions for scoping calculations were as follows:

- Vessel volume: 50,000 m^3
- Pool area: 500 m^2
- Initial pressures: 0.3 MPa (43.5 psi), 0.5 MPa (72.5 psi), and 0.7 MPa (101.5 psi)
- Vent hole sizes: 1, 10, 100 and 1000 m^2
- Total water inventory: 2.71 X 10^5 kg.

The following assumptions were made:

- System instantly comes to equilibrium.
- Fluid temperature instantly comes to saturation temperature.
- Vapor generation in bulk liquid instantly produces vapor flux at surface.
- Liquid vaporization does not contribute to the containment pressure. Blowdown is calculated without vapor source.
- Blowdown is calculated by choked flow through orifice while containment pressure is above 0.18 MPa (26.1 psi). Below 0.18 MPa, blowdown is calculated by an orifice-pressure-drop/flow-rate relationship.
- Containment temperature at failure is at saturation temperature at the initial pressure and does not change throughout the calculation.
- Values of liquid specific heat and heat of vaporization are constant.

- The liquid water at the start of the calculation is the total water inventory less the amount of water necessary to pressurize the containment to the initial pressure.

- The liquid water is depleted throughout the calculation by vaporization and entrainment. The removed water is not returned to the pool.

- The liquid water is assumed to reside in a pool of constant surface in a large single volume.

Kataoka and Ishii tabulated (April 1983; Table G-11, "Entrained Water and Droplet Size") the mass of water entrained, mass geometric mean diameter and GSD of the droplet size distribution as a function of initial pressure and vent hole area. The largest mass entrained at each pressure was associated with the largest vent hole area due to the rate of release. The ARF and RF values for the largest sized vent hole for each pressure listed are:

0.3 MPa: 2.4E-1 and 3E-3 to 1.0E+0 and 6E-7

0.5 MPa: 3.4E-1 and 2E-3 to 1.0E+0 and 4E-7

0.7 MPa: 3.9E-1 and 6E-7 to 7.4E-1 and 4E-7.

Not all the water was entrained at the highest pressure, because the liquid volume in the pool reduces by vaporization, thus reducing the superficial gas velocity under these conditions. The possible effect of secondary flashing by the droplets was assessed and found not to be a serious concern but the possible reduction of the droplets due to evaporation of the solvent after release was not evaluated. Thus, although the author stated that the model tends to overestimate the entrainment, the possible increase in the fraction of dissolved FPs entrained due to the reduction of droplet size could result in significant underestimation. Nonetheless, the values for droplets in the respirable fraction from flashing sprays under these conditions do not appear to result in a significant fraction of the material as droplets in the respirable fraction (range of 7E-4 to 3E-7). The values for the many of the variable parameters are temperature dependent and methods for determining temperature or change in temperature and values for parameters as a function of temperature are not provided.

Experiments have been performed to measure the airborne release and size distribution of aqueous solution over a limited range of conditions. Table 3-5 lists the experimental results obtained (Ballinger, Sutter and Hodgson, May 1987). Aqueous

Table 3-5. Measured ARFs and RFs During the Venting of Superheated Aqueous Solutions (Tables A.3 and A.4 - Ballinger, Sutter and Hodgson, May 1987)

Source Volume, ml	Pressure, psig	ARF	RF	ARF x RF
700	125	1.0E-2	0.78	1.0E-2
350	240	5.0E-2	0.73	4.0E-2
	125	2.0E-2	0.66	1.0E-2
	60	9.0E-3	0.62	6.0E-3
100	125	9.0E-2	0.69	6.0E-2

solutions were heated to pressures of 0.39 MPa (57 psig at ~134 °C), 0.85 MPa (124 psig at ~161 °C) and 1.65 MPa (240 psig at ~202 °C) using source volumes of 700 cm^3, 350 cm^3 and 100 cm^3. Figure 3-6 reproduced from Ballinger, Sutter and Hodgson (May 1987) indicates the temperature of the aqueous solution at the stated pressures although the extrapolation to the lower temperature/pressure is questionable given datum point #5. The uncertainty at these lower values, however, does not pose any significant concern. The liquid was released from the open top of the ~4-in. diameter apparatus via a double rupture disk arrangement. The experimental apparatus is shown schematically in Figures A.5 and A.6 in Appendix A. The fraction airborne increased with initial pressure and decreasing source size. The highest ARF, 9E-2, was measured at 0.85 MPa using a 100 cm^3 source and 69% of the airborne material was in the respirable size fraction. The ARF and RF depend upon the amount of heat (sensible heat in the liquid plus in the container) available and the heat needed to vaporize the solvent. The greater the fraction of solvent that can be flashed, the larger the ARF and RF. The ARFs and RFs measured are tabulated in Tables A.8 and A.9 in Appendix A.

Two bounding values are selected. For relatively low energy liquids with temperatures greater than the boiling point but less than 50 °C superheat, bounding ARF and RF values are assessed to be 1E-2 and 0.6 based on the 60 psig experimental run in Table 3-5. For more severe conditions between 50 and 100 °C superheat, bounding values for ARF and RF are assessed to be 1E-1 and 0.7. Median values for these conditions are 2E-2 and 0.7 with average values of 4E-2 and 0.7.

B. Empirical Correlations/Models. For situations where the temperature of the liquid released exceeds 100 °C superheat, an empirical correlation was presented by Ayer et al. (May 1988) for the evaporation/settling corrected data for the flashing spray experiments performed by Ballinger, Sutter and Hodgson (May 1987):

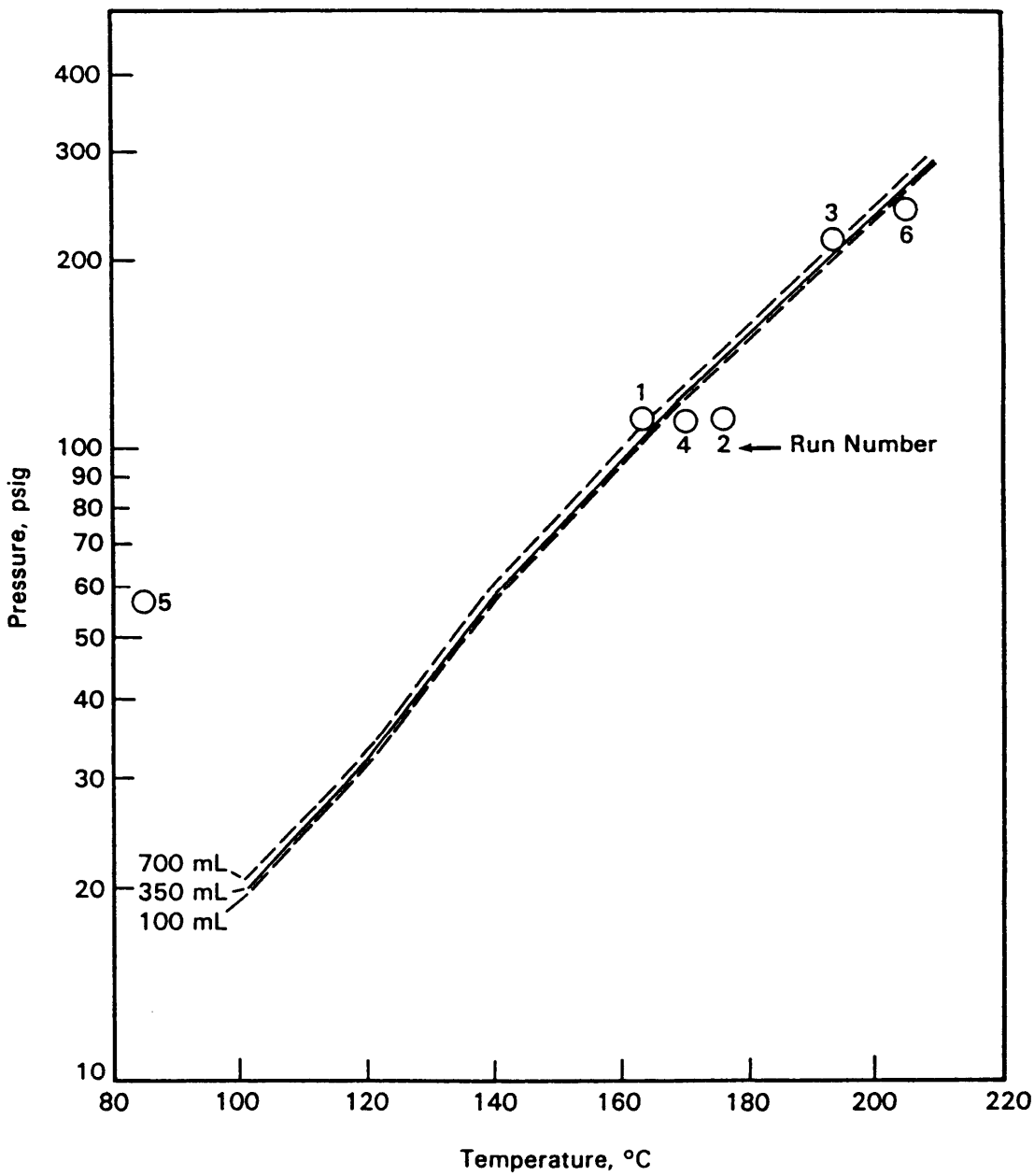

Figure 3-6. Measured and Predicted Temperatures for Flashing-Spray Releases (Ballinger, Sutter, and Hodgson May 1987)

$$ARF = 0.33\ (MF_g)^{0.91} \quad (3\text{-}7)$$

where: MF_g = mole fraction of pressurizing gas or water vapor flashed.

The mole fraction of pressurizing gas is computed using the method described in Appendix B of Ballinger, Sutter and Hodgson (May 1987) and is equal to the fraction of liquid flashed at release. The enthalpy and volume of vapor and liquid before and directly at release are used to calculate the fraction flashed.

The mass of vapor "flashed" at release is determined by an energy balance for the two phases after the initial pressure is vented. As an initial step, nomenclature must be defined. Temperature T_1 and pressure P_1 are the liquid temperature and pressure before release (i.e., superheated, pressurized liquid). Temperature T_2 is the saturation temperature of the remaining liquid and generated vapor immediately after release to a lower pressure (P_2, typically atmospheric pressure).

A simple, conservative energy balance can equate the enthalpies of the saturated vapor and liquid directly after the release to the enthalpy of the superheated liquid just before release. This energy balance, where the total mass, volume of container, and initial time are known, is as follows:

$$mass_{sh}(H_{sh}) = mass_v(H_v) + mass_l(H_l) \quad (3\text{-}8)$$

where:
- $mass_{sh}$ = mass of superheated liquid present (g),
- H_{sh} = enthalpy of superheated liquid at T_1 and P_1 (cal/g),
- $mass_v$ = mass of saturated vapor generated by flashing during depressurization (g),
- H_v = enthalpy of saturated vapor (e.g., vapor at saturation temperature T_2 for pressure P_2) (cal/g),
- $mass_l$ = mass of saturated liquid remaining after depressurization (g), and
- H_l = enthalpy of saturated liquid (e.g., liquid at saturation temperature T_2 for pressure P_2) (cal/g).

The simple mass balance for the flashing system would be:

$$mass_{sh} = mass_v + mass_l \quad (3\text{-}9)$$

The mass balance can be rearranged to

$$mass_l = mass_{sh} - mass_v \quad (3\text{-}10)$$

and substituted into the energy balance to develop an equation for the flashing fraction ($mass_v/mass_{sh}$) as follows:

$$mass_v/mass_{sh} = (H_{sh} - H_l)/(H_v - H_l) \quad (3\text{-}11)$$

The RF for the airborne material is assessed to be 0.3 based on an aerodynamic mass median diameter (AMMD) of 21 μm and a GSD of 3 recommended by the original authors for the liquid droplets. If the combined ARF x RF calculated using the correlation is less than the 7E-2 bounding value from experimental data for less than 100 °C superheat, the 7E-2 value is considered bounding.

Models also exist for the size distribution of the droplets formed (Gido and Koestel, November 1978, Brown and York, 1962). Gido and Koestel (November 1978) base their model upon the fact that drops with center-to-surface temperature differences of <5° K do not fragment. Their model requires evaluation of many parameters such as drop density, drop surface tension, vapor density, thermal diffusion, or residence time. Brown and York (1962) present a much simpler model:

$$D_{10} = (1840 - 5.18)\ T/N_{We} \quad (3\text{-}12)$$

where:

D_{10} = linear mean diameter of the droplet, μm (for water or aqueous solutions of low solute concentration, the linear diameter is roughly equivalent to the AED)

T = temperature of the jet, ° F

N_{We} = Weber number

= $density_{gas}$ X velocity of jet X diameter of jet/2 surface tension of liquid.

A conservative assumption is to assume all the excess heat is used to evaporate as much liquid as is required to reduce the temperature to less than boiling. The non-volatile radionuclides are assumed to remain in the liquid and the fraction of droplets in the respirable range (droplets 10 μm AED or less) determined by Brown and York's (1962) formula is the ARF with the RF set to a value of 1.0. In as much as the liquid temperature is just at boiling, any additional heat could reduce the size of the liquid droplet (although it is more difficult to evaporate water from concentrated solutions) or even generate solid salt particles with the addition of sufficient heat. This correlation is not assessed to be suitable for most uses due to the complexity of parameter determination.

3.2.3 Free-Fall Spill

Aqueous solution, slurries, and viscous liquids (non-Newtonian fluids) spilled onto a hard, unyielding surface can be subdivided into drops by the instability/shear stress at the surface of the liquid during the fall and by impact upon striking the surface (splashing). The passage of the falling material through the air space creates airflow patterns and turbulence that aids in suspension.

3.2.3.1 Solutions

Experiments have been performed to determine the airborne release from the free-fall spill of aqueous solutions with densities of ~ 1.0 (uranine) and ~ 1.3 g/cc (UNH). Materials that may represent airborne material deposited on the walls were measured in some experiments. The fall distance was limited, less than 3 m, and the initial dispersion of the material was uncontrolled; material was released by inverting a glass beaker holding the liquid. The experimental apparatus is shown schematically in Figure A.7 and the measured results reproduced in Tables A.10 through A.13 in Appendix A. Measured ARFs and RFs are tabulated in Table 3-6.

The ARFs for the uranine solution under these conditions ranged from 4E-6 to 2E-4 and 1E-6 to 2E-5 for the UNH. The fraction of the source airborne as particles 10 µm AED and less ranged from 2E-6 to 1E-4 for the uranine solution and from 2E-7 to 1E-5 for UNH. Both the fraction airborne and the fraction in the respirable size range appear to vary with fall distance and source size. A conservative bounding value for the ARF for aqueous solution with a density near 1 would be 2E-4 with an RF of 0.5. For TRU solutions with greater densities, a bounding ARF of 2E-5 with an RF of 1.0 (based on a high value that may be anomalous) is considered conservative. The median ARF and RF for aqueous solutions are 4E-5 and 0.7 and 1E-6 and 0.3 for the concentrated heavy metal solutions.

As previously noted in subsection 3.2.2.3.2, for the sake of simplicity a gross density distinction is made for determining which ARF and RF values to use for solutions. Any solution containing heavy metal salts where the liquid alone has a density in excess of ~ 1.2 g/cm^3 is considered a "concentrated heavy metal solution" for assigning ARF and RF values. Any solution containing heavy metal salts where the solution alone has a density less than ~ 1.2 g/cm^3 is considered an "aqueous solution" for assigning ARF and RF values.

An empirical model of ARF and droplet size distribution from free-fall spills of liquids beyond the fall distance range encompassed in the experiments has been developed by Ballinger, et al. (January 1988). In this model, the ARF value is defined by the following equation:

Table 3-6. Measured ARFs and RFs From the Free-Fall Spill of Aqueous Solutions (Tables A.2, A.4, B.2 and B.4 - Sutter, Johnston, and Mishima, December 1981)

Source Volume, ml	uranine or uranium, g	Spill Height, m	ARF	RF	ARF x RF
Uranine Solution					
1000	10	3	1.0E-4	0.45	6.0E-5
			8.0E-5	0.50	4.0E-5
500	5	3	4.0E-5	0.56	2.0E-5
			3.0E-5	0.44	1.0E-5
			3.0E-5	0.80	3.0E-5
			6.0E-6	0.82	5.0E-5
			5.0E-5	0.74	4.0E-5
			3.0E-5	0.70	2.0E-5
			4.0E-5	0.59	2.0E-5
			4.0E-5	0.64	2.0E-5
125	1.25	3	2.0E-4	0.52	8.0E-5
			3.0E-5	0.63	2.0E-5
1000	10	1	3.0E-5	0.53	2.0E-5
500	5	1	4.0E-6	0.62	2.0E-6
125	1.25	1	6.0E-6	0.72	4.0E-6
UNH Solution					
1000	208.7 U	3	1.0E-5	0.23	2.0E-6
			1.0E-5	0.19	3.0E-6
500	104.4 U	3	1.0E-5	1.0	1.0E-5
			2.0E-5	0.30	6.0E-6
			1.0E-5	0.16	2.0E-6
125	26.1 U	3	1.0E-5	0.36	5.0E-6
			2.0E-5	0.26	5.0E-6
1000	208.7 U	1	1.0E-6	0.51	5.0E-7
			1.0E-6	0.24	2.0E-7
500	104.U	1	1.0E-6	0.85	9.0E-7
125	26.1 U	1	4.0E-6	0.61	2.0E-6
			5.0E-6	0.62	3.0E-6

$$ARF = 8.9E\text{-}10\ Arch^{0.55} \quad (3\text{-}13)$$

where: $Arch$ = Archimedes Number
= $(density_{air})^2 * (spill\ height)^3 * g/(solution\ viscosity)^2$.

$Density_{air}$ is in g/cc, spill height is in cm, solution viscosity is in poise, and g is a gravitational constant, 981 cm/s^2.

This correlation covers all of the spill data, including slurries and viscous solutions. This results in it being potentially nonconservative for low-density aqueous solutions modelled on the uranine data. In order to determine a bounding ARF for this subset of solutions, the ARF value calculated from the model are multiplied by a factor of 3 (empirical observation). This factor does not apply to the other types of solutions.

No regression analysis produced a satisfactory correlation with AMMD and a simple statistical correlation representation of lognormal drop size parameters is substituted. They are presented in Table 3-7.

Table 3-7. Statistical Summary of Drop Size Parameters for Lognormal Distributions (Table 3.4 - Ballinger, et al., January 1988)

	AMMD, μm	GSD[a]	RF
All data	21.5	7.3	0.4
UNH spill	27.2	6.0	0.3
Uranine Spill	27.1	3.0	0.2
Sucrose spill	12.5	12.3	0.5
Slurry spill	15.8	10.1	0.4

[a] GSD = Geometric Standard Deviation.

The values listed in Table 3-7 should be applied to the appropriate liquids assumed involved in the event. For example, if the liquid can not be specified, the values assumed for all data would apply. The UNH represents heavy metal, aqueous solutions with densities 1.2 g/cm^3 or greater. For other aqueous solutions, the uranine values apply. The sucrose values are applicable to liquid with viscosities greater than 50 centipoise (the ARF decreases with viscosity but the AMAD appears to remain relatively constant for the limited data set available). Slurry values are applied to aqueous slurries.

3.2.3.2 Slurries

Experiments have been performed to measure the ARF and RF from the free-fall spill of slurries (Ballinger and Hodgson, December 1986). The apparatus (see Figure A.7, Appendix A) and procedures were as used in the free-fall spill experiments involving aqueous solutions. The pertinent data extracted from the reference document (Ballinger and Hodgson, December 1986) are shown in Table 3-8 (original data tables reproduced as Tables A.14 and A.15 in Appendix A). The bounding ARF and RF values are 5E-5 and 0.8 with median and average values of 2E-5 and 0.7 and 2E-5 and 0.8, respectively. The empirical model for calculation of ARFs and drop size characteristics was discussed in subsection 3.2.3.1.

Table 3-8. Measured ARFs and Rfs for Free-Fall Spill of Slurries (1 liter source, 3 meter fall distance) (Tables B.3 and B.4 - Ballinger and Hodgson, December 1986)

TiO_2, g/l	Glass Frit, g/l	Uranine, g/l	Sucrose, g/l	Viscosity, centipoise	Surface Tension, dyne/cm	SpG	ARF	RF	ARF x RF
10	0	20	250	3.2	58.2	1.12	9.0E-7	0.73	7.0E-6
100	0	20	200	NM	54.5	1.16	1.0E-5	0.64	7.0E-6
40	60	20	335	4.9	64.6	1.19	9.0E-6	0.77	7.0E-6
40	60	20	335	3.1	68.6	1.20	2.0E-5	0.76	1.0E-5
200	300	20	0	1.3	63.1	1.33	5.0E-5	0.78	4.0E-5
200	300	20	0	1.3	63.4	1.35	3.0E-5	0.81	2.0E-5
200	300	20	100	1.3	64.9	1.29	3.0E-5	0.78	2.0E-5
200	300	20	100	2.9	62.8	1.41	2.0E-5	0.72	1.0E-5

3.2.3.3 Viscous Solutions

Experiments were also performed to measure the ARFs and Rfs from the free-fall spill of viscous solutions (Ballinger and Hodgson, December 1986). The experimental apparatus is essentially the same as used for the free-fall spill experiments involving aqueous solution and slurries and is shown in Figure A.7 in Appendix A. Table 3-9 is a tabulation of pertinent data taken from the reference source (original data tables reproduced as Tables A.16 and A.17 in Appendix A).

Table 3-9. Measured ARFs and Rfs for Free-Fall Spill of Viscous Solutions (1 liter source volume, 3 meter fall height) (Tables B.1 and B.2 - Ballinger and Hodgson, December 1986)

Viscosity, centipoise	Surface Tension, dyne/cm	SpG	ARF	RF	ARF x RF
1.3	65.2	1.01	3.0E-5	0.59	2.0E-5
			3.0E-5	0.74	2.0E-5
2.6	68.9	1.10	7.0E-6	0.70	5.0E-6
			1.0E-5	0.76	8.0E-6
7.9	70.9	1.19	5.0E-6	0.80	3.0E-6
			7.0E-6	0.83	5.0E-6
17.5	77.4	1.23	3.0E-6	0.78	2.0E-6
			3.0E-6	0.90	2.0E-6
46.0	74.5	1.28	2.0E-6	0.84	1.0E-6
			3.0E-6	0.89	2.0E-6

Figure 3-7 reproduced from that reference shows the ARF tends to decrease with viscosity. For solutions that have a viscosity >8 cp, the ARFs are less than 7E-6 with a maximum RF of 0.9. For the range of viscosity >8 cp (surface tension >65 dyne/cm, specific gravity >1.2) the ARF and RF are bounded by 7E-6 and 0.8 with median values of 3E-6 and 0.8. The average ARF and RF values were 4E-6 and 0.8. Solutions with viscosities less than 8 cp are considered bounded by the values in subsection 3.2.3.1.

3.2.4 Aerodynamic Entrainment and Resuspension

Liquid can be made airborne by the passage of air over its surfaces through either parallel airflow or airflow directed into the surface, i.e., via whitecaps, spume, and film breakup due to capillary action of the liquid up the sides of its container. The latter effect may only be important for situations where the ratio of perimeter distance is a significant fraction of the surface area as in small pools used in experimental studies. Only a thin layer at the surface of the liquid can be involved in droplet formation since the droplets are formed by the film fragments that would not suspend if the film were too thick or the fragments too large. The airborne release fraction for this type of situation has been studied theoretically and measured under two sets of conditions. Calculations indicate that particles held to heterogeneous surfaces by a layer of water greater than 5 molecules thick cannot be resuspended at superficial gas velocities <5000 m/s (greater than sonic velocities) (Brockman, February 1985). Other calculations performed in the paper indicate that the aerodynamic flow profile at the surface may not be properly estimated; particles 10-μm in diameter were entrained at the lowest superficial velocity, 1.8 m/s, although most calculations indicate that the minimum velocities are required for particles an order of magnitude larger. Nonetheless, the calculations indicate the force necessary to suspend shallow pools of liquid probably requires substantial superficial velocities for suspension and that release of liquid droplets under most ordinary conditions are very low.

3.2.4.1 Spray Release From Large Outdoor Pond

A model, SPRAYMASS, was developed from empirical formulas representing ocean sprays (Roblyer and Owczarski, April 1992). Correlations between wind velocity and fetch (distance from the lee shore where turbulence begins) were developed from sea-salt aerosols (principally during surface breakup of bubbles formed in wave action) in the open sea, finite ponds and diffusion in atmosphere-surface boundary layers. The concentration of aerosol above ocean waves with finite fetch as a function of windspeed has been measured and is represented by:

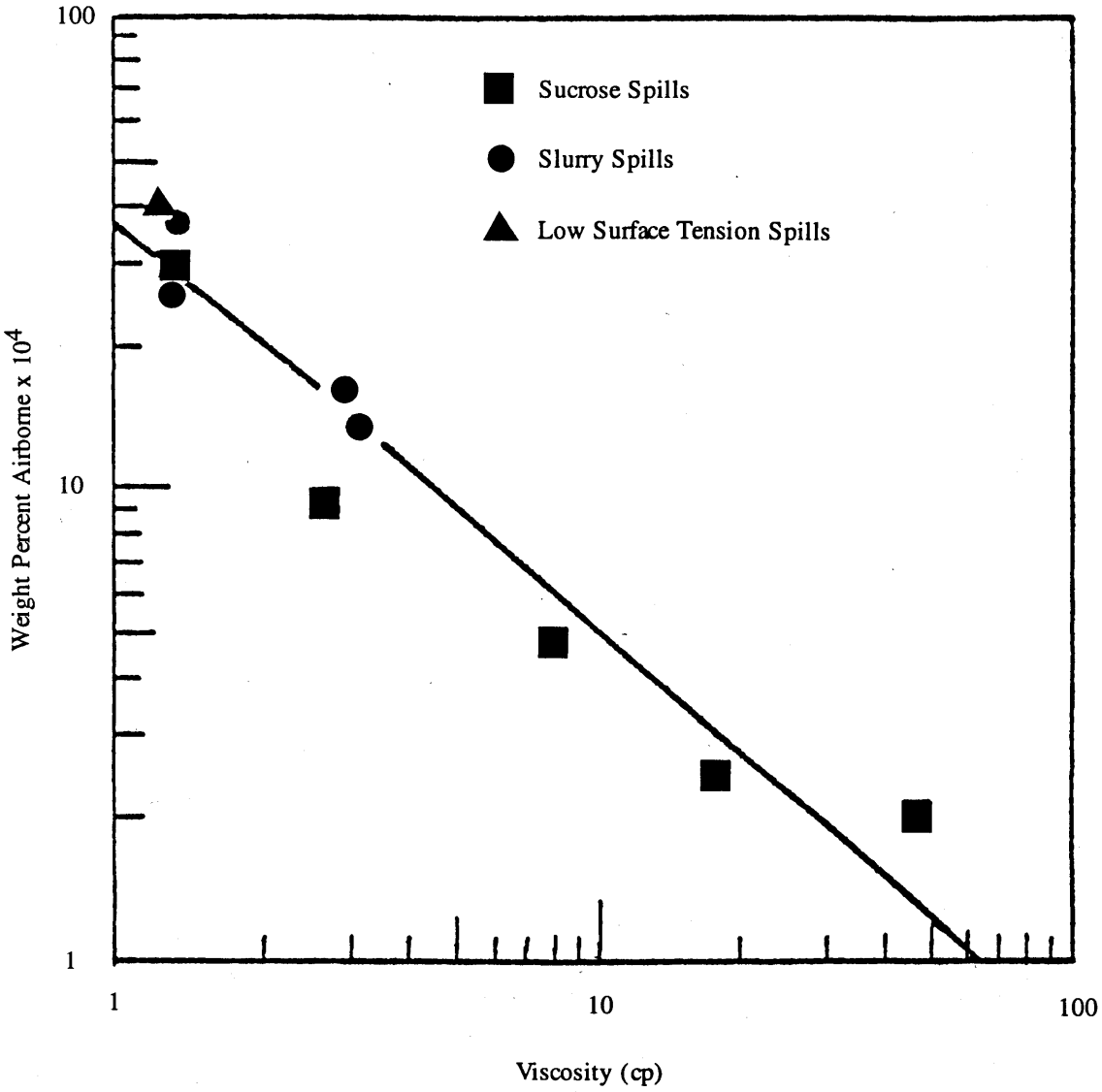

Figure 3-7. Weight Percent Airborne Versus Viscosity for Sucrose, Slurry, and Low Surface Tension Spills (Ballinger and Hodgson December 1986)

$$C_{air} = C_{water}(3.0E\text{-}11)(10^{0.098039v}) \quad (3\text{-}13)$$

where:

C_{air} = concentration of solute in air, g/m^3
C_{water} = concentration of solute in water, g/m^3
v = windspeed, m/s.

With considerations of boundary layer meteorology, an upward escape velocity (mass release rate) for a 200-m x 200-m pond as a function of windspeed and fetch can be calculated and is shown in Figure 3-8 taken from Roblyer and Owczarski (Figure 4 - April 1992). With a fetch of 10 m (~33 ft), a windspeed of 15 m/s (~33.6 mph), and an effective active layer (depth of liquid actually involved in drop generation) under these conditions of 1 mm to 1 cm, the airborne suspension rate would range from 4E-6/hr to 4E-8/hr.

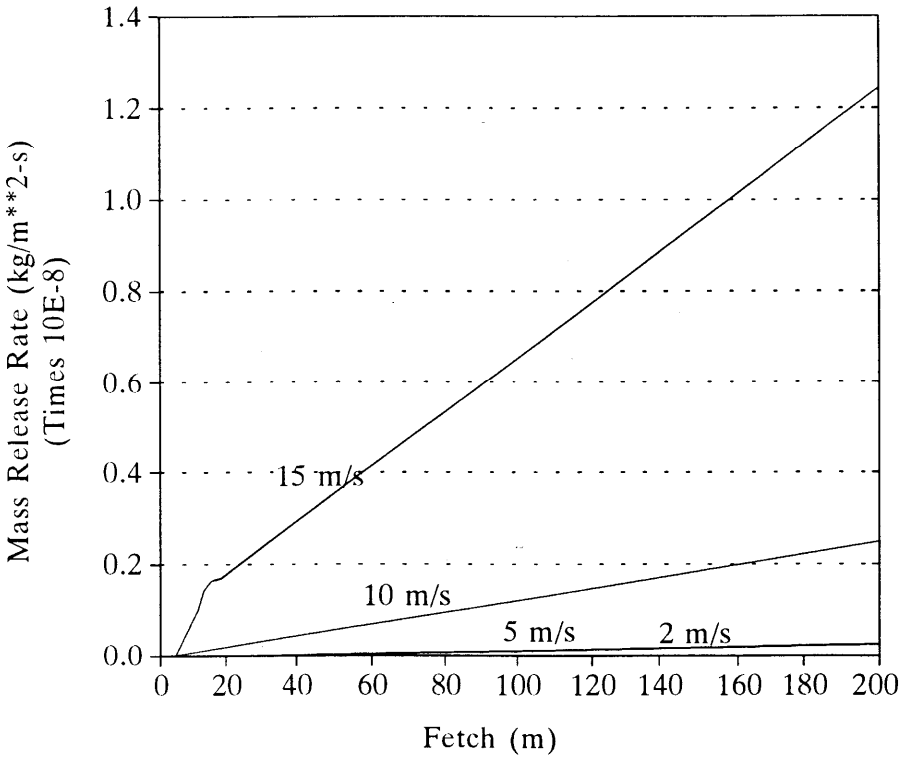

Figure 3-8. Mass Release Rate vs. Fetch, 200 by 200 Meter Pond (Roblyer and Owcsarski 1992)

3.2.4.2 Suspension of Liquids From Shallow Pools of Concentrated Heavy Metal Solutions on Stainless Steel

An indication of the ARF for this type of condition at low velocities can be gained from the entrainment of plutonium solution in air at velocities from 10 to 100 cm/s passing over the surface (Mishima, Schwendiman and Radasch, November 1968). The entrainment was from a very shallow pool (~2 to 4 mm) of limited diameter (~2.5 cm) using a dense solution. Evaporation periods lasted from 2 h to 24 h with temperatures from ambient (~21 °C) to 100 °C. For entrainment at ambient temperature with an evaporation period of 24 h, the ARF ranged from <2E-10 to 2.5E-9. The ARFs measured are listed in Table 3-10 taken from Table I in the reference document and found in Appendix A (Table A.1). The experimental apparatus is shown in Figure A.1.

Table 3-10. Measured ARFs from Shallow Pools of Concentrated Heavy Metal Salt Solution (0.72 g to 0.82 g Pu, 24-hr sampling period) (Table I - Mishima, Schwendiman and Radasch, November 1968)

Temperature, ° C	Air Velocity, m/s	ARF	Rate
ambient	0.1	<1.0E-9	<1.0E-14/s
ambient	0.5	2.5E-9	3.0E-14/s
ambient	1.0	<2.0E-10	<2.0E-15/s

Although the nominal velocities used in these experiments appear to be much lower than those quoted in subsection 3.2.4.1, the values represent velocities much closer to the surface (i.e., within cm) than the usual height for meteorological windspeed measurements of 10 m. For turbulent to laminar flow, the nominal 10 meter windspeed would be a factor of 2 to 10 higher than the values quoted in Table 3-10. The ARRs measured range from 7E-12/hr to 1E-10/hr.

3.2.4.3 Estimate of the Resuspension of Liquids From Soil

As will be discussed in Chapter 4, Sehmel and Lloyd (1976a,b) measured resuspension rates of a powder deposited on a soil surface and deduced that a reasonable value for resuspension rate was 1E-8/s to 1E-10/s. These values correspond to 4E-5/hr and 4E-7/hr. Liquids are significantly less susceptible to entrainment than powders.

3.2.4.4 Suspension From Soil at Higher Windspeeds

Aerodynamic entrainment (resuspension) at higher air velocities for UNH solution from various surfaces was measured in wind tunnel experiments (Mishima and Schwendiman, August 1973). Measurements were made at velocities of 2.5 mph and 20 to 23 mph at 1 ft above the surface of smooth, sandy soil. The experimental apparatus and measured values are shown in Figure A.3 and Table A.3 in Appendix A. The pertinent data are listed in Table 3-11.

Table 3-11. Measured ARFs and Rfs of Uranium (UNH) From Soil at 2.5 mph and ~20 mph (Table III - Mishima and Schwendiman, August 1973)

Substrate	Windspeed, mph	Sampling Time, hr	ARF	RF	Rate Fraction/hr
Soil	2.5	6	5.0E-4	0.76	9.0E-5
		24	1.0E-4	0.84	4.0E-6
	20	28	4.0E-4	0.63	1.0E-5

The ARF for 1 m/s (5E-4 in 6-hr, airborne suspension rate of 9E-5/hr) bounds the measured ARFs. The suspension rates from soil at both windspeeds for the remaining two datapoints appear to be approximately the same value (4E-6/hr vice 1E-5/hr). The two measurements at 1.0 m/s show more variation than the measurements for the two different windspeeds. In keeping with typical resuspension assumptions, the RF of the airborne material is conservatively assumed to be 1.0.

3.2.4.5 Bounding Assessments

For liquids inside buildings or other confinements, the effective airflows experienced are not considered comparable to extremes such as high external wind. The experiment involving suspension of liquids from shallow pools of concentrated heavy metal salt solutions on stainless steel (Mishima, Schwendiman, and Radasch, November 1986) produced extremely low estimates of aerodynamic entrainment in the range of 1E-11/hr to 1E-10/hr. Even the spray release from large outdoor pond values would not exceed a value of 4E-7/hr for 5m/s windspeed, and would be expected to be considerably less. Examining the results of Sehmal and Lloyd (1976 a,b), this study would expect a resuspension rate for liquids to be 4E-7/hr or less.

Based on the above considerations, a conservative ARR of 4E-7/hr is assigned for:
(1) liquids indoors on heterogenous surfaces (e.g., stainless steel, concrete) exposed to forced

building ventilation; or (2) liquids indoors exposed to other significant overall structural airflow, such as intrusion of external winds due to localized damage in various areas of the structure.

No experimental data for the effects of large debris over deposited powder on aerodynamic entrainment were found. Due to the decrease in aerodynamic stress if liquid is shielded by remnants and debris of a structure or confinement, an order of magnitude decrease in ARR is assigned. The bounding ARR for liquid covered with debris or exposed to largely static conditions in a structure that has lost forced airflow is 4E-8/hr.

For liquids spilled outside, three values are provided. For large pools (even if depth is shallow) and relatively high windspeeds (~ 30 mph), the maximum ARR estimate from the SPRAYMASS correlation of 4E-6/hr is assessed to be bounding. If only small external windspeeds are involved, the 4E-7/hr value for forced ventilation flow is more appropriate. For liquid spilled on and being absorbed by soil (i.e., rapidly depleting small puddles), however, special circumstances are involved. The contaminant is being deposited on the soil in a manner that makes it more conducive to airborne release. A bounding ARR of 9E-5/hr is assumed based on the experimental work of Mishima and Schwendiman (August, 1973). This same phenomena would not apply to very large spills with pooling as the ground would remain damp for a significant period of time and the surface would be less susceptible to aerodynamic stress.

The values assessed in this section apply only to freshly deposited contaminant that is not heavily intermingled with the overall soil or waste matrix. They are to be used for estimating releases for the short term only. It is incorrect to multiply the values cited by large time periods and assume the resulting large fractions represent real release potential from long term environmental contamination sites.

3.3 ORGANIC, COMBUSTIBLE LIQUIDS

Radionuclides are present in combustible liquids during liquid-liquid extraction processes and during decontamination procedures. In some cases, the radionuclides can be in an aqueous solution under a burning organic layer (e.g., process liquids, solvents, fuels).

The combustion of a liquid is a heterogenous reaction - heat from the flame radiates back to the liquid surface resulting in the evaporation of more fuel vapor that entrains air until a combustible mixture is attained and is ignited. The suspension of non-volatile materials appears to result from formation of drops of the bulk liquid. If the conditions are such that the surface film of the liquid is minimally disturbed, very little of the non-volatile components will be suspended from the bulk liquid. As the surface of the liquid is disturbed

by turbulence of the vapor generation, the capillary action at the edge of the liquid, and the evolution of water vapors from the aqueous solution trapped beneath the organic layer, the suspension of non-volatile components increases. If the evolution of water vapors is very rapid, a large volume of the aqueous layer may be ejected and quench the fire. Flaming combustion (smoldering combustion is observed in some solid fuels such as cellulosics) may also be quenched when the oxygen concentration diminishes to the range of 11% to 17.5% (generally flaming combustion ceases at ~ 16%) (Malet et al., 1983, Jordan and Lindner, 1983, 1985).

3.3.1. Burning of Small Volume/Surface Area 30% TBP-Kerosine Solutions, No Vigorous Boiloff

For quiescent fire (relatively undisturbed liquid surfaces), the ARFs measured by Mishima and Schwendiman (June 1973) for the combustion of 30% TBP in a kerosine-type diluent traced with various radionuclides (U, Cs, Ce, Zr, I) are applicable. The measured values are reproduced in Table A.18 and the experimental apparatus is shown in Figures A.8a and b in Appendix A. Twenty-five ml of 30% TBP-kerosine were placed in a 50-ml borosilicate beaker. Air (1- and 2-cfm) was drawn through a 2.7-in.-diameter stainless steel chimney around and over the beaker. Iodine (during the experiments using iodine tracer) was collected in a charcoal trap at the top of the chimney and airborne particulates in a glass fiber filter. The liquid was ignited and the liquid gently heated by a hand-held propane torch. Experiments were performed to self-extinguishment (no heating after initiation of flaming combustion) and supplemental heating to complete dryness. No aqueous phase in contact with the combustible organic was used in these experiments. The pertinent data are tabulated in Table 3-12.

Under the experimental conditions, the ARFs for all non-volatile materials appear to be less than 1E-2. Uranium ARFs range from 2E-4 to 3E-3, an uncertainty of approximately an order of magnitude. Cesium ARFs also show an order of magnitude uncertainty ranging from 2E-3 to 1E-2. ARFs for both cerium and zirconium are more consistent for the limited number of measurements made. The ARFs for iodine range from 7E-1 to 8E-1 and are assumed to be essentially 1E+0. In the absence of any measured airborne particle size distribution, all the airborne material is conservatively assumed to be in the respirable fraction. The volatile materials are considered to remain in the gaseous state although the volatile materials (generally iodine but may include other halogens and possibly some cesium compounds) may condense on various surfaces contacted or on pre-existing airborne particles and behave like the host particle thereafter. The effect cannot be readily characterized and the conservative assumption is that all the material is respirable. For the more industrial types of stresses encountered in nonreactor nuclear facilities, as opposed to LWR and BWR core melt conditions, semi-volatiles such as cesium and ruthenium are not expected to behave

Table 3-12. Measured ARFs From Burning Small Volumes of 30% TBP-Kerosine Traced with Selected Radionuclides (Table I - Mishima and Schwendiman, June 1973)

	ARFs Self-Extinguishment		ARFs Complete Dryness	
	1-cfm	2-cfm	1-cfm	2-cfm
Uranium	2.7E-4	2.3E-4	-----	3.0E-3
Cesium	2.2E-3	2.5E-3	1.9E-3	1.0E-2
Cerium	7.4E-3	5.6E-3	7.7E-3	7.1E-3
Zirconium	6.5E-3	ND	5.5E-3	2.4E-3
Iodine	6.57E-1	6.53E-1	8.17E-1	8.43E-1 8.28E-1 8.33E-1

as volatiles. Volatiles of interest for phenomenological stresses in nonreactor nuclear facilities are typically limited to iodine (NUREG-1320, NUREG-1140).

3.3.2 Pool Fires of 30% TBP-Kerosine

The ARFs for strontium from a large-scale 30% TBP-kerosine burn were reported by Sutter, Mishima and Schwendiman (June 1974). One hundred and fifty liters (150 l) traced with 25 g of strontium nitrate were burned in ten 17-in. x 23-in. x 3-in. deep stainless steel pans placed on concrete block above an 8-in. pool of water on the floor of a 12-ft x 12-ft cell of insulating board held in a sheet steel silo. The combustible organic phase was not in contact with an aqueous phase. Kerosine was floated on the surface of the water pool to aid in the burning of the 30% TBP-kerosine PUREX-type solvent. The organic liquids were ignited and the airborne materials carried to the exhaust gas treatment/sampling train apparatus shown in Figure A.9 in Appendix A taken from the reference document (Figure 1 - Sutter, Mishima and Schwendiman, June 1974). Two of the three burns generated usable data with ARFs of 2.2E-3 and 1.9E-3. The values are generally consistent with those generated in the small volume/surface area experiments in subsection 3.3.1.1.

3.3.3 Combustion of TBP-Kerosine Solutions Over Pools of Acid, Vigorous Boiloff

Halverson, Ballinger, and Dennis (February 1987) reported measurements of airborne uranium during the burning of combustible organic liquid over aqueous solutions. Small volumes of liquid were placed in metal beakers (except in one case where a borosilicate glass beaker was used to minimize the heat transfer through the beaker) on a load cell as shown in

Figure A.10 of Appendix A. The liquids were heated by heating tapes wrapped around the metal beaker except in the single case where a borosilicate glass beaker was used. In this case, radiant heat panels were used. The organic liquid ignited and air (27.5-cfm) was drawn up the 25.4-cm diameter chimney. Airborne particles were collected on glass fiber filters as a function of time. The pertinent data taken from Table A.6 in the reference document (original data tables reproduces as Table A.19 in Appendix A) are tabulated in Table 3-13.

Table 3-13. Measured Uranium ARFs During the Burning of TBP-Kerosine Over Aqueous Phase (Table A.6 - Halverson, Ballinger and Dennis, February 1987)

Solutions Tested	Burn Duration, min	ARF	Remarks
100 ml 30% TBP-kerosine (U) + 100 ml acid	27.5 34.8 53.3	4.04E-3 5.57E-3 4.34E-3	Aqueous boiled over and quenched fire, 40% to 60% organic unburned
100 ml 30% TBP-kerosine (U) + 100 ml acid (FP)	24.8 34.0	2.52E-2 2.70E-2	Aqueous boiled over and quenched fire, 40% to 60% organic unburned
100 ml 40% TBP-kerosine + 100 ml acid (U + FP)	61.3 65.0	5.98E-2 7.09E-2	Burned to dry residue Burned to dry residue
50 ml 40% TBP-kerosine + 150 ml acid (U + FP)	40.0	1.7E-3	~40% organic unburned
50 ml 30% TBP-kerosine (U) + 150 ml acid (U + FP)	57.3	1.56E-2	Unburned organic residue
50 ml 30% TBP-kerosine (U) + 100 ml acid (U + FP)	51.0	8.09E-3	Unburned organic residue

The measured values for ARF appear to have two orders of magnitude variation. The conservative upper bound ARF is 1E-1. The RF was only measured for one experiment [50 ml 30% TBP-kerosine (U) + 150 ml acid (U + FP)] with a value of 0.99. In most cases, heat transferred through the metal solution holder resulted in a boilover that terminated the burning. Use of glass holders or no external heat addition after ignition delayed boilover. Only in experiments #52 and #53 (40% TBP in normal paraffin hydrocarbon) using heating tape to heat the liquid did the burning proceed to complete dryness. It appears that burning the liquids to dryness increases the ARF; the two highest measured ARFs are from this configuration (6.0E-2 and 7.1E-2). The variation found for the other experimental configurations may be partially due to the vigor in boiloff and composition of the aqueous phase. The presence of salts in the aqueous phase may result in a slightly greater heat

capacity for the aqueous phase resulting in a more violent eruption upon boiling. Violent eruption of the aqueous phase could cause the generation and entrainment of organic droplets. Based on the results of this experiment, it is considered appropriate to make distinctions between fires that burn to a completely dried residue and fires where, for example, the presence of a large amount of aqueous solution precludes such an outcome.

3.3.4 Airborne Release of Uranium During the Burning of Process Solvent

Jordan and Lindner (October 1983) performed small-scale burning experiments using TBP-kerosine mixtures without an aqueous phase. The experimental apparatus is shown in Figure A.13 and the uranium release as a function of the uranium concentration in the solvent is reproduced as Figure A.14 in Appendix A. The decomposition of nitric acid or nitrates extracted into the solvent resulted in additional surface disturbance during burning. The ARF for uranium dissolved in the combustible liquid increased with uranium concentration and appears to range from 2E-3 to 2E-2. A bounding value of 1.5E-2 was selected by the authors and is generally consistent with the range of uranium releases reported above in the previous section.

3.3.5 Airborne Release During Combustion of TBP-Kerosine

The airborne release of cesium, thorium and cerium was measured in tests using both small (78.5 cm^2) and large (0.4 to 5 m^2) surface areas for combustion of TBP-kerosine process solvent (Malet et al. April 1983). The experimental apparatus is shown in Figures A.11 and A.12 in Appendix A. In both cases, the solvent traced with materials to represent the behavior of heavy metal and fission product elements was held in metal trays and heated by electric heaters. Air was drawn through the test vessel to exhaust systems that collected the airborne materials. The transfer coefficient in air was determined by:

[initial mass element] - [final mass element]/initial mass element.

The decontamination factor was determined by:

[initial concentration element] x [volume]/mass collected on filter.

The pertinent results taken from the referenced documents (original data tables reproduced as Tables A.20 and A.21 in Appendix A) are presented in Table 3-14.

Table 3-14. Measured Transfer Coefficients and Decontamination Factors During the Burning of TBP-Kerosine Solvent (Table I - Malet et al., April 1983)

Trace Element	Transfer Coefficient	Decontamination Factor	Fraction Reaching Filter
(Small Scale Experiments)			
Cs	0.47	1310	3.6E-4
	0.32	>5270	<6.0E-5
	0.27	>4890	<5.5E-5
	0.49	>3760	<1.3E-4
Th	0.45	112	4.0E-3
	0.23	159	1.4E-3
	0.12	137	8.8E-4
(Large Scale Experiments)			
Ce	0.23	4260	5.4E-5
	0.42	1110	3.8E-4
	0.92	-----	-----
	0.89	-----	-----
	0.47	5620	5.4E-5

The transfer coefficient appears to be the fraction of the trace element not recovered from the test apparatus after the run. Others have experienced difficulties with recovering/detecting various elements in the residues from the combustion of TBP-kerosine solvents. The decontamination factor is the fraction of airborne material carried to the collection filters, although it appears that only the initial airborne concentration was used for the estimates.

The actual concentration may have varied during the run. The ARFs reported from other studies are generally the fraction of the material used in the experiment that is carried to the collection filters that are within a few feet and the exhaust gases relatively contained from the point of origin to the collector. The fraction reaching filter is the transfer coefficient multiplied by the reciprocal of the decontamination factor. Thus, ARFs reported from other studies should be smaller than the transfer coefficient but greater than the fraction reaching filter. The values are within an order of magnitude or two for the cesium and cerium used for the small scale and large scale experiments and are considered to corroborate those values.

3.3.6 UNH and Air-Dried UNH on Various Surfaces During a Shallow Pool Gasoline Fire

Experiments were performed by Mishima and Schwendiman (1973) in a wind tunnel to measure the ARF and RF of uranium from various surfaces (sandy soil, sandy soil with vegetation cover, stainless steel and asphalt) at windspeeds of 1.0 m/s and 10 m/s. Windspeed measurements were made at the centerline of the 2-ft x 2-ft wind tunnel and therefore represent much higher windspeeds (50 mph to 60 mph) at a standard 10-m height. UNH was placed upon the surface held in a 22-inch diameter flange in the floor of the wind tunnel. In some experiments with sandy soil, the UNH was allowed to air-dry for several days prior to testing. One gallon of gasoline was poured over the surface, ignited and the air drawn over the burning surface. In some cases, the airborne particles were only sampled during the actual burning; in other cases, the airborne particles were collected for much longer periods. The ARFs and Rfs measured are reproduced in Table A.3 and the apparatus shown in Figure A.3 in Appendix A. The relevant data are listed in Table 3-15.

Table 3-15. Measured ARFs and Rfs for Uranium Airborne During Gasoline Fires on Various Surfaces Involving UNH and Air-Dried UNH (Table III - Mishima and Schwendiman, August 1973)

Substrate	Uranium Form	Air Velocity, m/s	ARF	Rate Fraction/hr	RF
Sandy soil	liq. UNH	1.0	1.7E-5	3.0E-6	0.75
		10	5.4E-3	4.0E-2*	0.40
Vegetation on sandy soil	air-dried UNH	1.0	9.6E-4	2.0E-4	0.78
		10	2.0E-3	4.0E-4	0.86
		1.0	7.4E-5	1.0E-5	0.64
			4.0E-4	6.0E-5	0.18
Asphalt		10	1.2E-3	2.0E-4	0.32
	liq. UNH	1.0	4.1E-4	1.0E-3*	0.70
Stainless steel		10	2.4E-3	6.0E-3	0.68
	liq. UNH	1.0	1.4E-2	6.0E-2	0.40
		10	1.14E-1	6.0E-1	0.34

* Calculated based on actual burn time for gasoline fire.

The ARFs range from 1.7E-5 (soil, 1.0 m/s) to 1.14E-1 (stainless steel, 10.0 m/s). The data are extremely limited with single values at some sets of conditions (surface, windspeed). The type of surface may have some effect in that the liquid can be absorbed into the substrate or the substrate can conduct heat. If the substrate conducts heat well, the liquid can be boiled rapidly generating conditions much more favorable to generation of airborne liquid droplets. In extreme cases for any surface, a possible mechanism for suspension of liquids

and salts is explosive release of moisture trapped under the salt during rapid heating of the material. The ARFs for soil and vegetation covered soil are lower than the ARFs for stainless steel under comparable conditions but are consistent with the ARFs for asphalt, which does not conduct heat but may become plastic due to the heat and retain salts from the evaporation of solution.

3.3.7 Thermal Stress Bounding Assessments

Based on the experimental results summarized in the preceding sections, a distinction will be made between volatiles (i.e., iodine) and other radionuclides and between various burning environments. For volatiles, the bounding ARF and RF are 1.0 and 1.0. For other radionuclides, the ARF and RF are a function of the burning characteristics.

The experiments summarized in sections 3.3.1 and 3.3.2 (with additional insight obtained from section 3.3.3) represent a reasonable approximation of quiescent burning, small-scale pool burning, or the burning of a relatively thin layer, as compared to the underlying layer of aqueous solution, of organic solvent. A bounding ARF and RF of 1E-2 and 1.0 are believed to appropriately characterize these conditions. The median ARF and RF are estimated to be 6E-3 and 1.0.

The experiments summarized in sections 3.3.3 and 3.3.4 represent a reasonable approximation of vigorously burning large pools or solvent burning over aqueous layers, where the relative sizes of the two layers support significant turbulent disruption of the aqueous layer. The main distinction is whether the burning proceeds to complete dryness. When it does not, a bounding ARF and RF are assessed to be 3E-2 and 1.0. When complete dryness results, a conservative bounding value of ARF is assessed to be 1E-1 (7E-2 rounded upward), with an RF, in the absence of a measured value, of 1.0. The median ARF is 1E-2 with an average value of 2E-2. The ARF (1.5E-2) reported by Jordan and Lindner (February 1985) agrees with the median ARF for the other experimental conditions.

Two other conditions relating to large organic fires over specific types of base surfaces are discussed based on section 3.3.6. The first condition is aqueous solutions or air-dried salts on a surface that absorbs or holds liquid. Candidate surfaces include those that are porous or are significantly cracked or pitted. The bounding ARF and RF are 5E-3 and 0.4 respectively (liquid UNH on sandy soil, 10 m/s) for the actual combustion period. The median values of ARF and RF are 1E-3 and 0.8 (liquid UNH on vegetation covered sandy soil at 1 m/s) with average ARF and RF values of 1E-3 and 0.5. The second condition is aqueous solutions or air-dried salts on a surface that is a strong conductor of heat (i.e., metal). The bounding ARF and RF are 2E-1 and 0.3 (stainless steel, 10 m/s) for the actual combustion period.

4.0 SOLIDS

4.1 SUMMARY OF ANALYSIS OF DATA

In assessing release fractions for solid materials, the materials cannot be treated in the more generic sense applicable to most liquids of interest. Specific distinctions must be made in terms of types of solid material. This chapter provides information[1] for three categories of solids: (1) metals (Note: release from energetic hydride reactions also covered under topic of metals in subsection 4.2.1.1.4); (2) nonmetallic or composite solids; and (3) powders.

Metal

Thermal Stress: Plutonium

- Airborne Release of particulates formed by room temperature oxidation (corrosion). Based upon the experimental measured values, the bounding ARRs and RFs for the four situations covered are:

Bounding (unalloyed Pu)	ARR (dry air) 2 x 10^{-6} μg Pu/cm^2-hr; RF 0.7
	ARR (100% RH) 7 x 10^{-3} μg Pu/cm^2-hr; RF 0.7
(delta-phase metal)	ARR (dry air) 7 x 10^{-8} μg Pu/cm^2-hr; RF 0.7
	ARR (100% RH) 6 x 10^{-4} μg Pu/cm^2-hr; RF 0.7

  Use of the above values are generally intended for short timeframes (i.e., < 100 hours). They are not appropriate for long-term estimation (i.e., months to years) as rate is controlled by issues such as oxide coat coherence.

- Airborne release of particulates formed by oxidation at elevated temperature, greater than room temperature but less than self-sustained oxidation (ignition). The bounding values apply to static oxidation at elevated temperatures less than ignition temperatures. The MAR is the amount of oxide present under specific conditions. If oxidation is not complete, experimental data cited can provide a basis for such estimations if desired.

  Bounding ARF 3E-5/RF 0.04

[1] It is noted that there are some special issues associated with Pu-238 in compacted forms used in thermoelectric power devices as material behavior of Pu-238 is different from that of Pu-239 and power devices can be subjected to unique high-energy stresses. Specific information on this subject can be obtained from the DOE radioisotope thermal generator program.

- Airborne release of particulates formed by self-sustained oxidation (molten metal with oxide coat), self-induced convection. The phenomena applies to the self-sustained oxidation in air of metal pieces under self-induced convection. Mishima's (1965, 1966) values exceed the combined value (ARF x RF) for all other measurements and are assessed to be bounding for this situation.

  Bounding ARF 5E-4/RF 0.5

- Airborne release of particulates from disturbed molten metal surfaces (i.e., flowing metal, actions resulting in continual surface renewal), high turbulence at surface. Without continuous surface renewal, maximum release phenomena is self-sustained oxidation as described previously. The bounding value applies to situations where ignited-molten plutonium is disturbed by direct impact of high air velocities such as during free-fall, induced high turbulence on molten surface, etc. The 95% confidence limit ARF and RF values recommended by Carter and Stewart (September 1970) are assessed to bound the experimentally measured values reported.

  Bounding ARF 1E-2/RF 1.0

  This value is also considered applicable to highly energetic surface reaction as, for example, the conversion of a significant mass of fine hydride to oxide. It is not, however, applicable to oxidation of trace quantities of hydride as metal or powder contamination exposed to air.

- Airborne release of particulates from oxidation of small (hundreds of μm in diameter) molten metal drops passing through air or explosive reaction of entire metal mass. For the violent ejection of molten metal and vapor formation from droplets, Raabe et al. measured an ARF of 1.0 with an RF of 0.4 (estimated from MMD reported) from exploding wire experiments and Carter and Stewart (September 1970) measured an ARF of 0.5 with an RF of 1.0 for small molten metal drops falling through air.

  Bounding ARF 1E+0/RF 0.5

Thermal Stress: Uranium

- Airborne release of particulates during complete oxidation of metal mass, >500 ^{o}C, gas flow 0 - 2 m/s. Based upon the experiments performed by Carter and Stewart (September 1970) heating uranium in an upflow of air with oxide generated allowed to sluff away during the oxidation process, the mean

value as designated by the authors is chosen. The measured values were only for airborne particles <10 µm AED. The values for solubility class in simulated lung fluids are assumed to be >95% "Y" class with remainder in "D" class, based on the solubilities of sintered oxides recovered from the burn tests performed as part of the hazard classification tests on armor-defeating munitions.

The 95% confidence level airborne release value for oxidation of uranium metal at flow velocities <100 cm/s reported by Carter and Stewart (September 1970) is exceeded by the value reported by Elder and Tinkle (December 1980) during the oxidation of staballoy penetrators in laboratory experiments. The ARF x RF values obtained for experiments at temperatures less than 900 ^{o}C were less than 1E-3. ARF and RF values of 1E-3 and 1.0 are assessed to be bounding for this category with the solubility classification for the oxides formed given above.

Median	ARF 1E-4/RF 1.0
Bounding	ARF 1E-3/RF 1.0

- Airborne release during free-fall of molten metal drops. The median ARF x RF values for free-fall of molten uranium metal droplets in air are as given by Carter and Stewart (1970). The ARF x RF value assessed to be bounding is an arbitrary increase of the 95% confidence level value assigned by Carter and Stewart (to 1E-2) to be consistent with the comparable value for plutonium. Since the airborne material is cooled rapidly after formation, the solubility of the airborne oxides formed from plastic deformation and ignition of the thin film of metal generated by the impact of penetrators against hard targets is assessed to be 50% "Y" class and 50% "D" class.

Median	ARF 2E-3/RF 1.0
Bounding	ARF 1E-2/RF 1.0

- Airborne release from explosive dispersal of molten uranium. The values for explosive release of molten uranium indicate that, if the uranium is molten and subdivided in very small drops (as by the exploding wire technique) and ejected into air at sonic velocities (as by the electrodynamic thruster technique described), all the uranium could be made airborne as a very fine particulate material with all particles or aggregates 10 µm AED and less (Rader and Benson, June 1988). The solubility class of the airborne material is anticipated to be like the airborne material formed during impact of staballoy

penetrators against hard targets such as armor (i.e., thin film formed by plastic deformation of the metal in passage through armor is ignited by frictional heat and rapidly cooled in the air). The maximum solubility values reported for the type of situation are 50% "D" class + 50% "Y" class.

Bounding ARF 1E+0/RF 1.0

Explosive Stress

The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock Effects. For detonations in or contiguous to solid metal, a respirable release of a mass of inert material equal to the TNT equivalent is assessed to be bounding. At low mass ratios (mass inert material/mass TNT equivalent), the respirable release is comparable to the total material release. As mass ratios increase, the respirable fraction becomes significantly less than the total amount of material released, which decreases with increasing mass ratio as well[2].

- Blast Effects. No significant airborne release is postulated. For effects on metal surface contamination, see Chapter 5.

- Venting of Pressurized Gases Over Metal. No significant airborne release is postulated. For effects on metal surface contamination, see Chapter 5.

Free-Fall Spill and Impaction Stress

No significant airborne release is postulated for this accident configuration. For effects on surface contamination, see Chapter 5.

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[2] The release estimation methodology presented here is not intended to cover high explosive detonations involving nuclear weapons or associated assemblies, which can involve mass ratios much less than 1. Extensive investigation into such events has been conducted and actual releases depend on highly sensitive configuration information. An estimate of 20% of weapon metal released as respirable particles has precedent as a general upper bound for such events. However, specific configuration studies also support respirable releases in the range of 1% to 10% of the metal. If the 20% value is not used, thorough knowledge of the literature and consultation with knowledgeable individuals at relevant national laboratories is necessary to finalize values assumed for a given purpose.

Aerodynamic Entrainment and Resuspension

No significant release is expected for this effect. Based upon the assumption that the airborne release during corrosion of plutonium metal is equal to the aerodynamic entrainment (resuspension) of the corrosion product from the metal surface, bounding ARR and RF values for this configuration are assessed for both unalloyed and delta phase metals:

Bounding (unalloyed metal):	ARR (dry air) 2 x 10^{-6} μg Pu/cm^2-hr;RF 0.7
	ARR (100% RH) 7 x 10^{-3} μg Pu/cm^2-hr;RF 0.7
(delta-phase metal):	ARR (dry air) 7 x 10^{-8} μg Pu/cm^2-hr;RF 0.7
	ARR (100% RH) 6 x 10^{-4} μg Pu/cm^2-hr;RF 0.7

For effects on metal surface contamination, see Chapter 5.

Nonmetallic or Composite Solids

Thermal Stress

- Vitrified Waste. No significant release of radionuclides is postulated for thermal conditions generated by industrial-type fires.

- Aggregate (e.g., concrete, cement). No significant release is postulated at ambient conditions. For large fires that can heat the aggregate to temperatures in excess of 100 °C, material incorporated in the aggregate may be released at rates increasing with time and temperature. Water bound with the aggregate is lost equally in two main temperature stages; up to 200 °C (i.e., adsorbed water) and 200 to 600 °C (i.e., chemically bound water). Portions of solid aggregate will be broken down into a powder-like form at higher temperatures and thus be available for suspension at the maximum value for heating of nonreactive powders in subsection 4.4.1.1.

Bounding (Tritium, if present as H_2O, to 200 °C)	ARF 5E-1/RF 1.0
Bounding (Tritium, if present as H_2O, to 600 °C)	ARF 1E+0/RF 1.0
Bounding (Suspendible powder)	ARF 6E-3/RF 0.01

- Spent Commercial Nuclear Fuel. Values are estimated for spent nuclear fuel for commercial power generation, metal targets, and metal alloy and cermets targets. Refer to the text for explanations on these materials.

Explosive Stress

The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock Effects. The value used for metal is considered adequate for use with aggregates such as concrete as well. For detonations in or contiguous to solid material, a respirable release of the mass of inert material equal to the calculated TNT equivalent is assessed to be bounding.

- Blast Effects. If aggregate is hurled with considerable velocity to impact, the crush-impact correlation of Subsection 4.3.3 may be used to characterize the response provided an impact velocity can be estimated. If the aggregate is not hurled with significant velocity, no significant airborne release is postulated. For effects on surface contamination, see Chapter 5.

- Venting of Pressurized Gases over Solid. No significant airborne release is postulated. For effects on surface contamination, see Chapter 5.

Free-Fall Spill and Impaction Stress

The ARF x RF for the fragmentation of an aggregate solid that can undergo brittle fracture can be estimated by the equation:

$$ARF \times RF = (A)(P)(g)(h) \qquad (4\text{-}1)$$

where:

ARF x RF	=	(Airborne Release Fraction) (Respirable Fraction)
A	=	empirical correlation, 2E-11 cm^3 per g-cm^2/s^2
P	=	specimen density, g/cm^3
g	=	gravitational acceleration, 980 cm/s^2 at sea level
h	=	fall height, cm.

Aerodynamic Entrainment and Resuspension

No significant airborne release is postulated for this accident configuration. For effects on surface contamination, see Chapter 5.

Powders

For low-energy stresses, powders do not tend to significantly fragment. For high-energy stresses considered in this document, the size fraction of powders of concern have relaxation times on the order of fractions of a millisecond, and thus do not tend to present themselves for fragmentation. Therefore, the amount of respirable material assumed airborne using this handbook should not exceed the amount of respirable material originally present in the source MAR, if that value is known. If the value is not known, and the bounding value specifically uses as the RF the fraction of original MAR less than 10 μm AED, an RF of 0.1 can be assumed based on Ayer, et al. (May 1988).

Thermal Stress

The ARFs and RFs assessed to be bounding for various plutonium compounds subjected to thermal stress (temperature <1000 °C, natural convection) are given below. For the chemically non-reactive compounds in powder form, suspension is from the entrainment of pre-formed particles by the flow upwards from the heated surface. The mass of particles in any size range (respirable size) released cannot exceed the total mass of particles in the source powder since no new particles are formed. For the reactive compounds in powder form, the release is dependent on the particles formed by the reaction (oxidation in the experiments presented) and the phenomena that would carry the particles formed into the local flow field. Thus, some variation is anticipated.

Bounding	(non-reactive compounds)	ARF 6E-3/RF 1E-2
	(reactive compounds except PuF_4)	ARF 1E-2/RF 1E-3
	(PuF_4)	ARF 1E-3/RF 1E-3

Explosive Stress

The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock effects. Based upon data for cratering and associated airborne release of soil a respirable release of the mass of inert material equal to 20% of the calculated TNT equivalent is assessed to be bounding for detonations in or contiguous to powder in open air where shock and blast effects will quickly disperse. The mass of the material-of-concern is dependent upon the concentration of the material-of-concern in the powder.

- Blast effects. Two cases are noted for both detonations and deflagrations.

  — For detonations in confined areas where immediate dissipation of blast effects is not anticipated, blast effects may produce larger releases than shock effects. The largest of the blast and shock numbers is used. A large blast release is expected for deflagrations of large volumes (>25% of free confinement volume) of flammable mixtures above powder with high confinement failure pressure (~ > 0.17 MPa_g [25 psig]). For these cases, an ARF of 1.0 with an RF equal to the fraction of the original source powder < 10 μm is selected.

  — Accelerated airflow parallel to surface. This condition is representative of powders shielded from the effects of a detonation or strong deflagration by standard containers such as a glovebox or can that are damaged or open. It is noted that small sources of confinement such as a can should not be in the immediate proximity of a detonation when using this value.

    Bounding ARF 5E-3/RF 0.3

- Venting of Pressurized Powders. This phenomena applies to deflagrations (i.e., subsonic release of pressure from confinement) as well as pressurization by other means. Deflagrations involving flammable gases occupying less than 25% of the confinement volume use venting data to estimate ARF and RF values. Larger volume deflagrations (i.e., >25% of available free volume) can use these values depending on estimated confinement failure pressure.

  — For the venting of powders or confinement failure at pressures to 0.17 MPa_g (~ 25 psig) or less. Large volume deflagrations (> 25% of confinement volume) where confinement, such as a glovebox, fails at or less than 0.17 MPa_g can also use this value for evaluating explosive phenomena.

    Bounding ARF 5E-3/RF 0.4

  — For the venting of powders or confinement failure at pressures in the range of 0.18 to 3.5 MPa_g (~ 500 psig) or the venting of pressurized

gas through the powder. Large volume deflagrations (flammable gas volume > 25% of confinement volume) use the value for large deflagrations given under blast effects.

Median	ARF 5E-2/RF 0.4
Bounding	ARF 1E-1/RF 0.7

Free-Fall Spill and Impaction Stress

For the free-fall spill of cohesionless powders:

- Fall distance <3 m, air velocity normal to powder flow, general forced enclosure ventilation or general low-wind outside conditions.

Median	ARF 3E-4/RF 0.5
Bounding	ARF 2E-3/RF 0.3

- Fall distance >3 m, air velocity normal to powder flow. Apply the calculations outlined in subsection 4.4.3.1.3 provided ARF and RF values exceed 2E-3 and 0.3.

- Suspended solid dispersed into flowing air for enhanced air velocities normal to direction powder flow. The mass airborne in the RF is limited by the total mass of material in this size fraction in the source material.

$$ARF = 0.0134\ U + 0.00543 \qquad (4\text{-}2)$$

where U = windspeed, m/s.

- Suspension of bulk powder due to vibration of substrate from shock-impact to powder confinement (e.g., gloveboxes, cans) due to falling debris or external energy (e.g., seismic vibrations).

Bounding	ARF 1E-3/RF 0.1

- Suspension of bulk powder by debris impact and air turbulence from falling objects.

Median	ARF 4E-4/RF 0.2
Bounding	ARF 1E-2/RF 0.2

Aerodynamic Entrainment and Resuspension

Based upon the information on resuspension factors and rates found in the articles reviewed and the outdoor rates, the long-term ARR suggested by Sehmel and Lloyd (1976a,b) of 4E-5/hr appears to be a reasonable bound. Although ARRs appear to decrease with time, use of the initial ARR measured is conservative. Not including MAR depletion with time tends to overestimate the release. The resuspension tends to fluctuate as the level of stress fluctuates and the surface conditions respond to the previous stresses. The bounding ARF and RF for aerodynamic entrainment is due to the passage of vehicular traffic.

Use of the values given above for short time frames (<100 hours) would not introduce serious error due to the severe depletion of the source. For time periods exceeding 100 hours, the reduction of the source can be accounted for from the entrainment of material. In general, these values are intended for immediate post-accident conditions and freshly deposited material. They would overestimate releases from long-term contamination (i.e., months to years) and are not appropriate for such use.

- Homogeneous bed of powder exposed to ambient conditions (normal process facility ventilation flow or less, or nominal atmospheric windspeeds <2 m/s with gusts up to 20 m/sec) following an event.

Bounding	ARR 4E-5/hr, RF 1.0

- Homogeneous bed of powder buried under structural debris exposed to ambient conditions or under static conditions within structure following an event.

Bounding	ARR 4E-6/hr, RF 1.0

- Entrainment of powders from road surface by passage of vehicular traffic.

Bounding	ARR 1E-2/passage, RF 1.0

4.2 METALS

4.2.1 Thermal Stress

Most metals react with oxygen in some fashion, with some reactions occurring at room temperatures. At low temperatures, the reaction rates may be so slow that the oxidation is not readily recognized, or a protective oxide film may form that limits/reduces additional oxidation. Many metals generate heat from the oxidation reaction. Metals that attain a self-sustaining reaction at ambient temperature are called pyrophoric. Some nuclear metals (uranium, plutonium) can achieve a self-sustaining reaction at elevated temperature dependent upon surface-to-volume ratio and heat transfer conditions.

Initial oxidation rate is a function of the temperature; when heat is externally supplied and generated by the oxidation reaction, the kinetic controlled regime exists. At some temperature (a function of the balance between the heat available and the heat loss), the reaction becomes self-sustaining (plutonium ignition temperature ~ 500 ^{o}C), and the reaction rate and the temperature become limited by the diffusion of the oxygen to the reaction interface and the overall heat transfer characteristics of the system. Under these sustained conditions, known as the diffusion-controlled regime, temperatures range from 900 to 1100 ^{o}C and plutonium (mp 641 ^{o}C) is molten but uranium (mp 1132 ^{o}C) is not. Both metals may form protective suboxide films at the interface that are adherent, but more stable oxides are formed as the depth of the oxide layer increases. The matrix spacing for some oxides is sufficiently different from the metal phase spacing that the oxide is non-adherent and can be made airborne by sloughing of the oxide from the oxidizing mass and entrainment in the convective currents generated by the heated metal. Other heat sources such as fires may also generate convective currents that may carry the airborne materials once ejected from the oxidizing mass. High temperatures (>1000 ^{o}C) may coarsen the size distribution of the residual powder or reduce suspension by sintering the powder oxide.

4.2.1.1 Plutonium

Haschke (July 1992) reviewed and evaluated the data on the oxidation of plutonium. "The oxidation is a 'paralinear' process involving three stages. ... has the functionality characteristic of a diffusion-controlled process. As the thickness of the adherent oxide layer increases on the metal surface, the rate of oxygen diffusion through the layer decreases according to a parabolic curve like observed for stage I ... formation of oxide particles begins during stage II ... characterized by linear rate ... at onset of stage II, the thickness of the inherent product layer attains a critical value determined by buildup of stress induced by forming of low-density oxide (molar volume = 23.67 cm^3/mol) on the high-density metal (molar volume 12.10 cm^3/mol). This stress is relieved by cracking and spalling of oxide

layer ... stage III is a similar linear process entered after a transition period, but origin of this change is unknown.... observed increase in particle size with increasing reaction temperature interpreted as combination of two factors ... metal hardness and malleability with temperature ... hardness decreases with temperature ... increasing malleability reduces stress generated at metal-oxide interface and promotes formation of thicker product layer before spallation ... indicates formation of centimeter sized hydride particles if reaction temperature equal or exceeds half the melting temperature of metal in degree centigrade ... second factor that may alter particle size is kinetic in nature ... spallation involves nucleation of crack at surface and propagation of those cracks through the stresses material with ultimate coalescence and formation of free particles. Nucleation and propagation are both time dependent ... growth rate of oxide layer is large compared to spallation rate at high temperature and formation large particles are favored, at room temperature the oxide growth rate is extremely slow and the longer time available for spallation favors extensive crack formation and small particle size ... largest diameter observed for low-temperature oxide is ~ 5 µm in geometric diameter ..."

Condit (October 1986) reviewed the airborne release of plutonium metal under fire and explosion conditions. The effects of various factors on the airborne release of plutonium are listed in Figure 4-1 from that document. Five configurations are covered below based upon the temperature of oxidation and the airflow (i.e., turbulence) around the oxidizing material or oxide. Other experimental data covering the airborne release of plutonium during highly energetic accident situations involving assembled nuclear weapons (e.g., aircraft crashes and rocket/jet fuel fires with potentially concurrent falling molten material) indicate larger release fractions for the plutonium metal under these circumstances (ARF = 1E-2) and would correspond to the configuration covered here of disturbed molten metal surface (e.g., flowing molten metal where surface is renewed allowing greater surface turbulence), high surface turbulence, and violent reactions.

4.2.1.1.1 Room Temperature Oxidation/Corrosion. Stewart (1963) observed no detectible change in appearance of a plutonium metal surface in 7 days at room temperature and dry air. In moist air, a loose coating of powder was evident. Only 10% of the airborne particles formed in 100% relative humidity air were in the respirable size range. The oxide removal rate (µCi/cm^2-hr) as a function of metal phase and humidity is shown in Figure 4-2 reproduced from the reference document. The ordinate is expressed in terms of µCi of activity/cm^2-s. Assuming the specific activity (7.4E-2 Ci/g) quoted for weapons grade plutonium by Raabe (November 1978) and Eidson and Kanapilly (February 1983), the author calculates the maximum rates for unalloyed and delta-stabilized metal in dry air and 100% humidity to be:

Little effect:

- **Mass of the sample**
- **Area to volume ratio**
- **Gas flow rates**
- **Gas chemistry by itself (complex alloy interactions)**

Significant effect:

- **Tumbling of reaction products through the gas stream**
- **Temperature**
  - **Fraction decreases up to around 1500°C (oxide sintering)**
  - **Fraction increases at higher temperatures (Pu vaporization)**
- **Alloying of the Pu**
- **Chemical state of the Pu**

Unknowns:

- **Processes at high temperature**
- **Broader experience with alloying**
- **Age of Pu**
  - **Helium**
  - **Americium**
- **Optimum firefighting and cleanup techniques**

Figure 4-1. What Affects the Aerosolization Fraction in Plutonium Burning? (Condit October 1986)

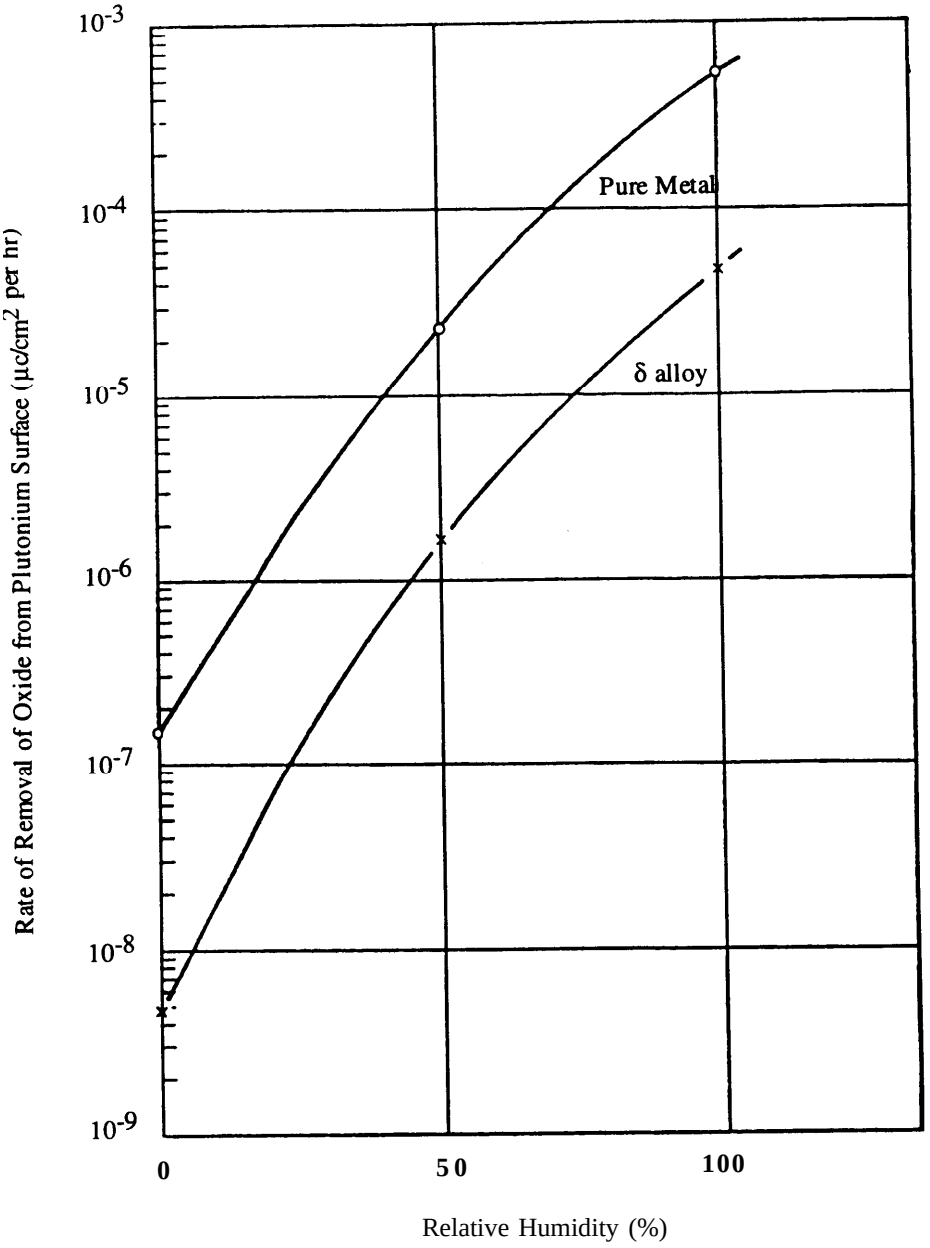

Figure 4-2. Dependence of Rate of Removal of Oxide Particulate from the Metal Surface on Humidity. Flow rates were:

Dry Air	24 cm/s
Medium Humidity	18 cm/s
Saturated Air	14 cm/s

(Stewart 1963)

unalloyed	(dry air) 2E-6 µg/cm^2-hr
	(100% RH) 7E-3 µg/cm^2-hr
delta stabilized	(dry air) 7E-8 µg/cm^2-hr
	(100% RH) 6E-4 µg/cm^2-hr.

Figure 4-3, also reproduced from Stewart (1963), shows the size distribution of the particles obtained at various temperatures (only the metal type is specified for one distribution). The diameter of the particles is given as equivalent spheres indicating that the dimensions are Geometric Diameter and must be multiplied by the square root of the density of PuO_2 (11.46 g/cm^3) to approximate the AED. Using distribution A from the figure (i.e., material airborne under static conditions at all temperatures in air), the RF is between 0.6 and 0.7.

Chatfield (1968) conducted a series of experiments to measure the airborne release in air flowing at velocities from 10 to 80 cm/s involving both delta-alloy and unalloyed plutonium. Experiments were performed using both horizontal and vertical glass tubes with exterior resistance heating. The experimental configuration was such that particles >20 µm were lost prior to sampling. Size distributions of the airborne materials were by Casella cascade impactors that yield AED. Figures 4-4 and 4-5 reproduce, from the reference, plots of the rate of release of activity (µCi/cm^2-s) as particles 10 µm AED and less for both delta-alloy and unalloyed plutonium in dry and saturated air as a function of the reciprocal of the absolute temperature. The data are plotted in this fashion to ascertain if the release rate is linearly proportional to the oxidation rate. Assuming the specific activity for weapons grade plutonium given by Raabe (November 1978) and Eidson and Kanapilly (February 1983) of 7.4E-2 Ci/g, the author calculates airborne releases to be:

unalloyed	(dry air) 0.1 -- 4E-6 µg Pu/cm^2-hr
	(100% RH) 2 -- 4E-3 µg Pu/cm^2-hr
delta-alloy	(dry air) 0.1 -- 1E-7 µg Pu/cm^2-hr
	(100% RH) 0.1 -- 4E-3 µg Pu/cm^2-hr.

The values are close to those from Stewart (1963) above. However, the data from Stewart were judged to be more consistent. Therefore, the ARRs and RFs assessed to be bounding for the low release phenomena of room temperature oxidation (corrosion) of plutonium metal for a short time duration are based on the Stewart data.

unalloyed metal:	2E-6 µg/cm^2-hr/0.7 (dry air)
	7E-3 µg/cm^2-h/0.7 (100% RH)
delta-alloy	7E-8 µg/cm^2-hr/0.7 (dry air)
	6E-4 µg/cm^2-hr/0.7 (100% RH).

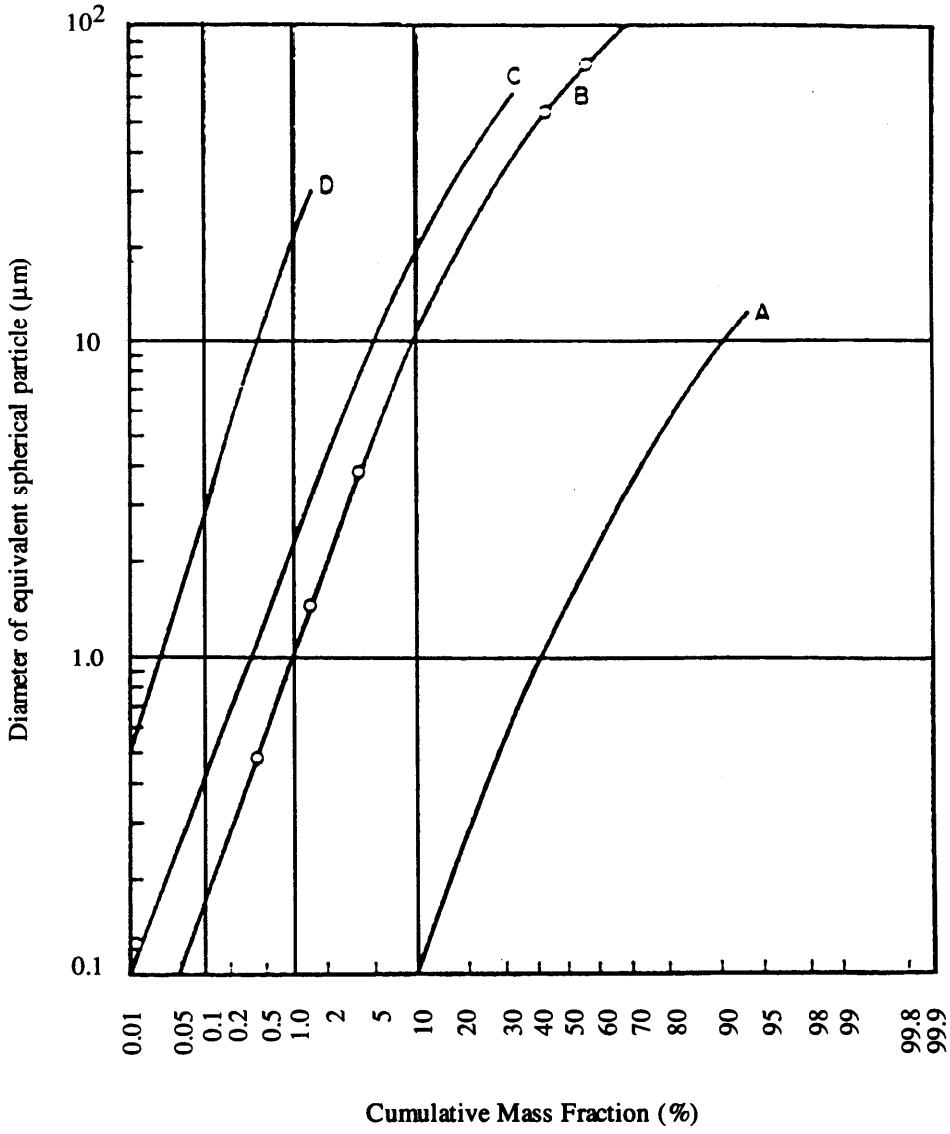

Figure 4-3. Particle Size Distributions Produced by the Oxidation of Plutonium Under a Variety of Conditions

Curve A Material airborne under static conditions at all temperatures in air
Curve B Oxide formed at 123°C from δ stabilized alloy
Curve C Oxide formed at 400°C to 500°C
Curve D Oxide formed above Ignition Point
(Stewart 1963)

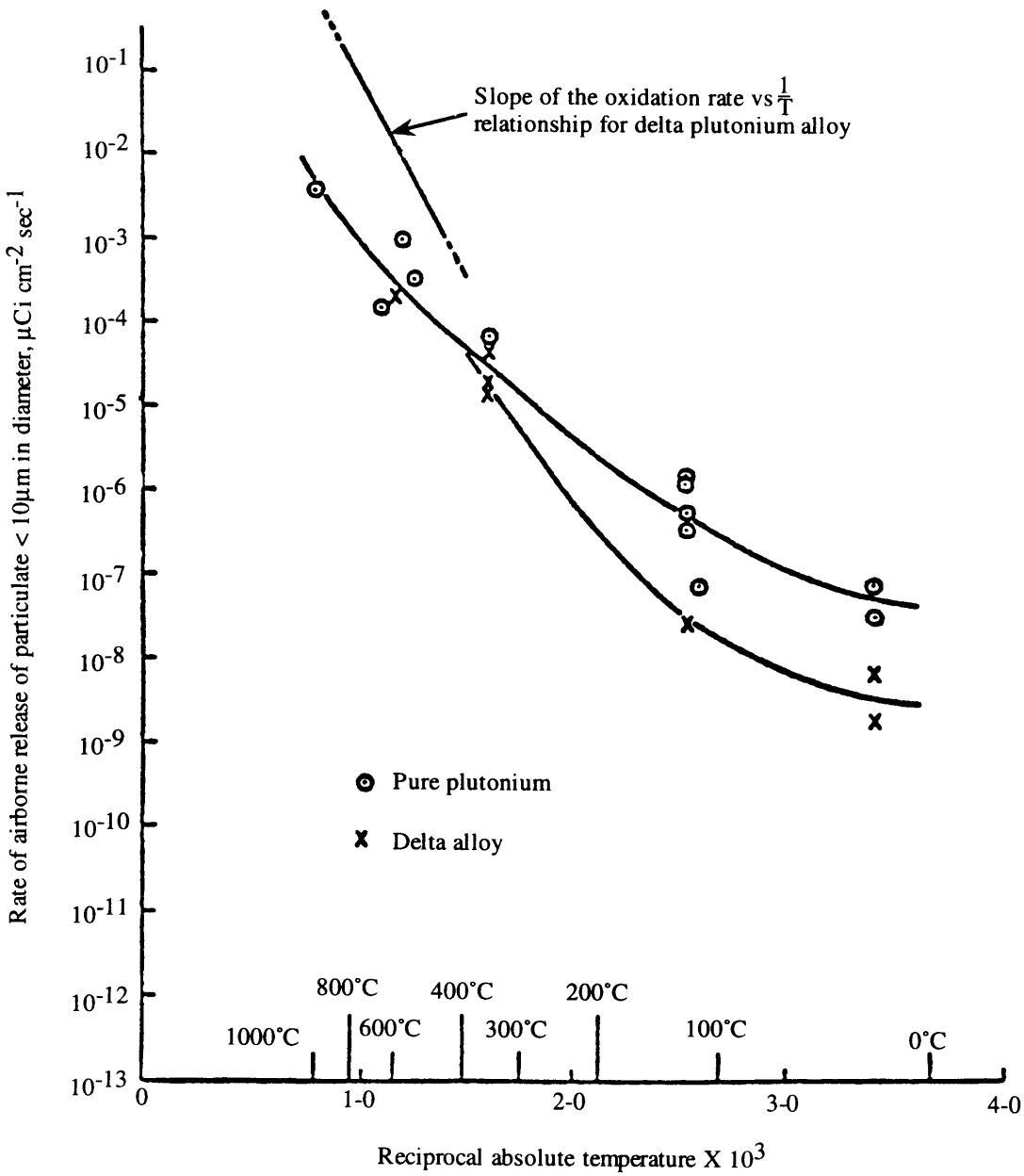

Figure 4-4. Release of Particulate Oxide Less Than 10 µm Diameter from Plutonium into Saturated Air

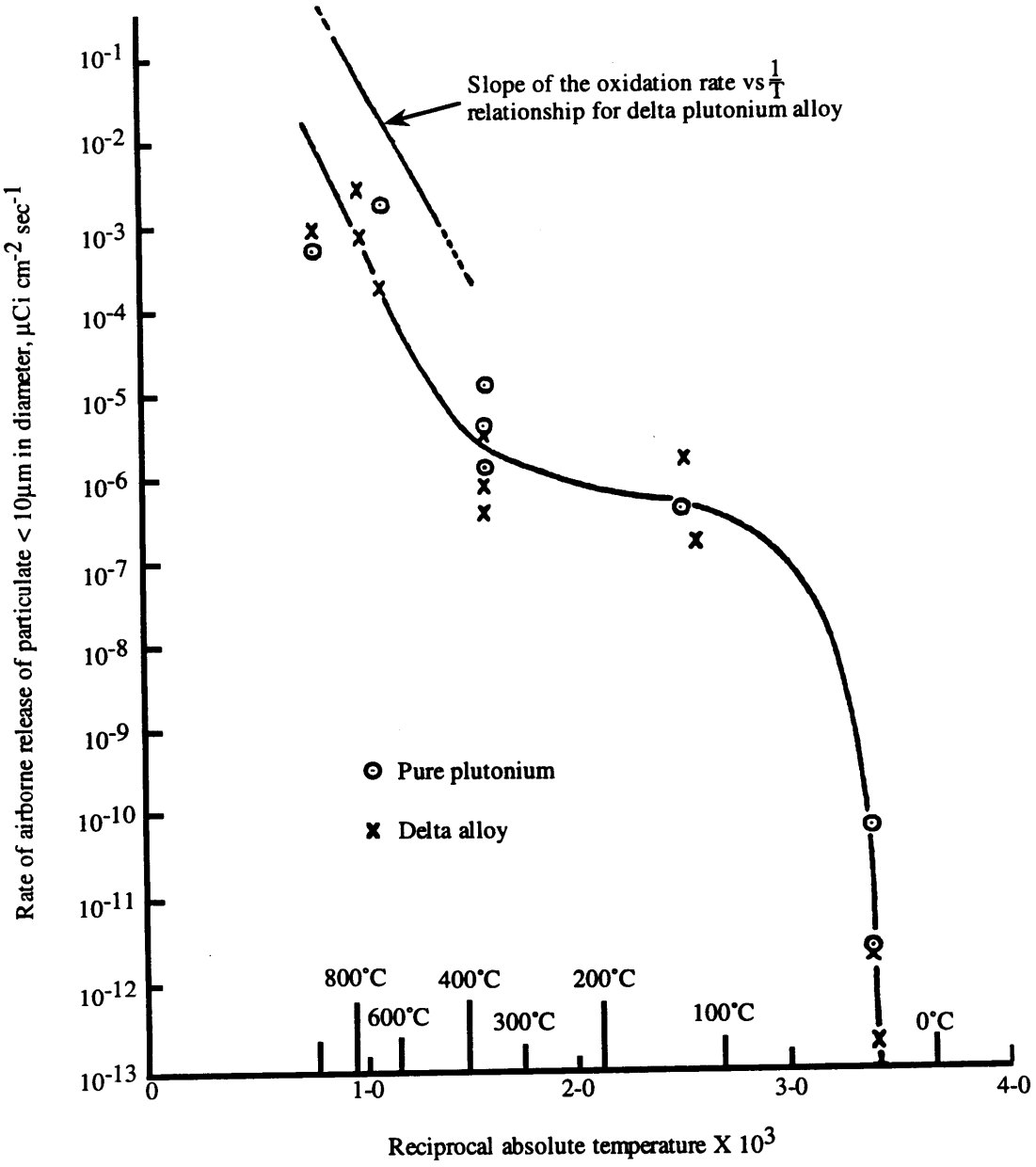

Figure 4-5. Release of Particulate Oxide Less Than 10 µm Diameter from Plutonium into Dry Air

4.2.1.1.2 Oxidation at Elevated Temperatures Below Ignition Temperature. Stewart (1963) reported on oxides from heating small pieces (less than 13 g) of unalloyed and delta-alloy plutonium metal to various temperatures in various atmospheres. Two shapes were used: billets (cylinder 0.7 cm diameter by 1.0 cm long) and swarf (turnings). Various experimental configurations were needed for different experimental conditions. The pertinent data are tabulated in Table 4-1 with source data from the referenced document reproduced in Tables A.28, A.29a, A.29b, and A.30 in Appendix A. The size distributions of airborne materials are shown in Figures A.23 and A.24 in Appendix A.

Table 4-1. Measured ARFs During Oxidation of Unalloyed and Delta-Phase Plutonium at Elevated Temperatures in Air (Tables 3, 5 and 7 - Stewart 1963)

Metal Phase	Temperature	Mass Specimen (g)	Humidity mg H_2O/l air	ARF
delta	123 °C	7.483	0.03	1.5E-3 (0.033 oxidized)
delta	113 °C	7.344	1.50	3.2E-5 (0.054 oxidized)
delta	123 °C	8.602	16.0	4.8E-6 (0.035 oxidized)
beta		11.021	0.03	1.4E-6 (0.57 oxidized)
beta		10.802	8.0	1.1E-6 (1.0 oxidized)
beta		7.191	8.0	3.3E-6 (0.76 oxidized)
alpha	113 °C	9.397	16.0	1.3E-6 (0.21 oxidized)
beta	123 °C	11.265	16.0	5.6E-7 (0.17 oxidized)
beta		8.154	16.0	1.0E-6 (0.65 oxidized)

* Mass Median Diameter in µm Aerodynamic Equivalent Diameter.

At temperatures below the ignition temperature (experiments performed at 113 and 123 °C), the unalloyed and alloyed metal behave differently. The oxidation rate for the delta-phase alloy is stepwise and two orders of magnitude less than for the unalloyed metal. The difference is attributed to the formation of a protective oxide film (the crystalline matrix spacing for delta-phase metal and the dioxide are very similar and the dioxide adheres to the metal surface). The film must crack and fall away before additional oxidation can occur. The particle size distribution of the bulk oxide generated is very wide (0.1 to 300 µm D_g).

The oxidation and release rate experimentally observed were continuous during the oxidation of unalloyed metal. Except for a single anomalous high release value measured for delta-phase metal at 123 °C in low humidity air with a very low fraction oxidized (1.5E-3), the ARFs for both types of metal phases are bounded by a release of 3E-5 (also very low fraction oxidized) and ranging to a value of 6E-7. The measured size distribution as

Equivalent Sphere for oxidation at this temperature is shown as slope B in Figure 4-3 for the delta-phase and the mass fraction for the RF can be approximated by using the mass fraction for particles 3 μm (i.e., 10 μm AED). Under this assumption, an RF between 0.02 and 0.04 is indicated. If the metal is not completely oxidized in the event, the ARF and RF must be applied to the fraction of material oxidized. Some indication of the oxidation rate can be obtained from measured rates (as mg of PuO_2/cm^2-hr formed) shown in Table 4-2 from early data taken from the referenced source. Haschke (July 1992) reported "the observed (oxidation) rate of 0.2 g PuO_2/cm^2-min is independent of temperature."

Table 4-2. Summary of Oxidation Rates for Plutonium at Temperatures Below 100 °C[1] (Stewart 1963)

Metal Phase	Temperature °C	Relative Humidity %	Oxidation Rate mg PuO_2/cm^2hr	Source
α	30	95	0.01	Sackman 1960
	90	55	15 during 10-60 hr period	
	90	95	200 after initially slower rate[2]	
	100	0	0.04	
α	35	20	0.01	Waber et al. 1960
	75	0	6E-4	
	75	50	0.4	
α	50	0	0.025[3]	Dempsey and Kay 1958
	50	100	0.04	
γ	30	95	0.01	Sackman 1960
	90	55	0.2	
	90	95	0.4	
	100	0	0.006	

[1] Data extracted from graphs by the authors quoted and are intended to give a general trend only.

[2] The oxidation rates during the initial phase were much smaller; about 2 and 4 mg PuO_2/cm^2hr at 55 and 95 per cent relative humidity, respectively. More recent studies indicate that onset of such rapid corrosion is exceptional and may be due to impurities in the metal and its pretreatment (Sackman, private communication).

[3] This rate applied up to 20 hr and thereafter the weight of the sample remained constant.

The airborne release rates of respirable particles can also be estimated from Figure 4-4 and 4-5 reproduced from Chatfield (1968) by converting the rates expressed in activity/cm^2-hr to mass/cm^2-hr as shown subsection 4.2.1.1.1. An ARF of 3E-5 is considered to bound the experimentally measured ARFs at elevated temperatures less than ignition. This value also bounds most of the data for measured release during the self-sustained oxidation of plutonium

under natural convection covered in subsection 4.2.1.1.3. Therefore, bounding values of ARF and RF are assessed to be 3E-5 and 0.04.

4.2.1.1.3 Self-sustained Oxidation Above the Ignition Temperature. Stewart (1963) reported on the oxides formed by heating to various temperatures small pieces (less than 13 g) of unalloyed and delta-alloy plutonium metal in various atmospheres. Two shapes were used: billets (cylinders 0.7 cm diameter by 1.0 cm long); and swarf (turnings). Various experimental configurations were needed for different experimental conditions. The data for higher temperature oxidation (in the region of temperatures found during self-sustained oxidation) are tabulated in Table 4-3, and the source data from the referenced document are reproduced in Tables A.28, A.29a, A.29b, and A.30 in Appendix A. The size distributions of the airborne materials are shown in Figures A.23 and A.24 in Appendix A.

Table 4-3. Measured ARFs During Self-Sustained Oxidation of Unalloyed and Delta-Phase Plutonium (Tables 3, 5 and 7 - Stewart 1963)

Phase	Temperature	Mass Specimen (grams)	Atmosphere	Humidity mg H_2O/l air	ARF
delta	900 °C	12.7	air	0.03	1.5E-5 (MMD 8 µm[1])
unalloyed	950 °C	10.3		0.03	1.5E-5 (MMD 8 µm[1])
	630 °C	15.06		0.03	2.4E-3 (MMD 11 µm[1])
delta	800 °C	12.2		16.0	1.3E-4 (MMD 4.5 µm[1])
	895 °C	1.95			6.0E-5 (MMD 2.1 µm[1])
delta	780 °C	2.18			1.0E-4 (MMD 2.1 µm[1])
delta	820 °C	1.15	carbon dioxide		(not self-sustaining)
delta		1.34	nitrogen		(no reaction)
delta	>1000 °C	5.34	oxygen		2.1E-2 (violent reaction, MMD 0.3 µm[1])
delta	970 °C	5.89	30% O_2: 70% N_2		1.8E-4 (0.25 oxidized MMD 29 µm[1])
delta	~1200 °C	7.41	40% O_2: 60% N_2		1.9E-4 (MMD 16 µm[1])
delta	690 °C	5.476	0 to 0.2 atm O_2		1.4E-4 (0.53 oxidized)
delta	625 °C	5.476	0 to 0.099 atm O_2		5.4E-7 (0.17 oxidized)
unalloyed	620 °C	7.216	0 to 0.167 atm O_2		1.2E-4 (0.12 oxidized)
	560 °C	7.352	0 to 0.167 atm O_2		4.7E-5 (0.056 oxidized)

[1] Mass Median Diameter in µm Aerodynamic Equivalent Diameter.

For oxidation of both delta-alloy and unalloyed metal at or above ignition temperature in air, the ARFs range from 1.5E-5 to 2.4E-3. Particle size distribution as a function of oxidation atmosphere conditions is shown in Figure 4-6 reproduced from the reference document. For

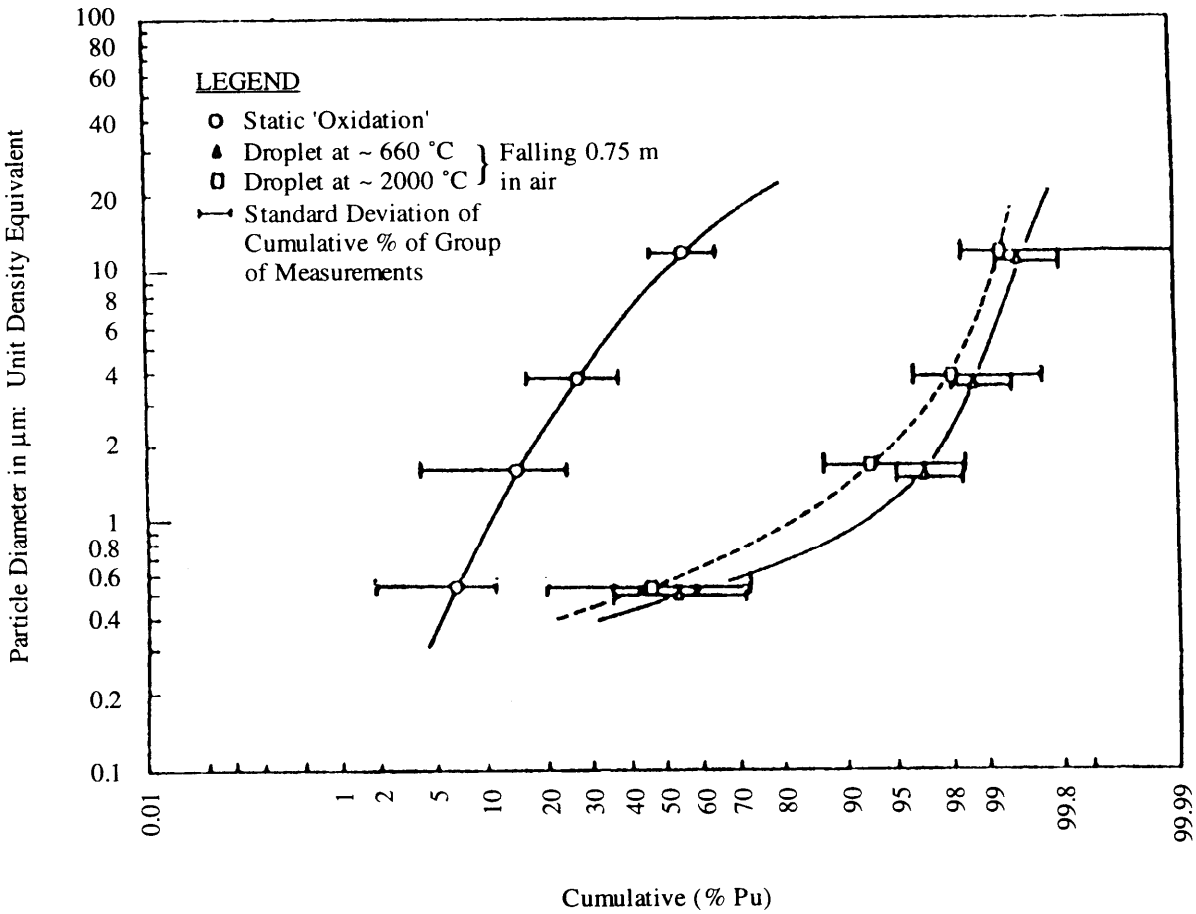

Figure 4-6. Representative Particle Size Distributions Obtained with Cascade Impactors for PuO_2 Aerosols Formed in Static and Dynamic Conditions

the highest ARF (2E-3 at 950 °C in air) with the RF provided by the authors of 0.001 (in Figure A.22, Appendix A), the respirable source term (ARF x RF = 2E-6) is less than that measured by Mishima (November 1966) of ~ 3E-4 (5E-4 x 0.5 = 2.5E-4).

Mishima (December 1965) measured the airborne release during the oxidation of unalloyed plutonium metal at elevated temperatures in flowing air and the size distribution of the residue. Right cylinders of unalloyed plutonium metal (0.594 cm to 0.625 cm in diameter by 1.73 cm to 1.89 cm long, weighing 9.89 g to 11.34 g), were heated to temperatures exceeding the ignition temperature of the metal. Ignition temperatures ranged from 490 °C to 500 °C with temperatures (measured above the oxidizing specimen by thermocouple) during the complete oxidation of the specimens ranging from 410 °C to 900 °C. Air, at predetermined velocities ranging from 3.3 to 50 cm/s, was passed over the oxidizing specimen and particles entrained from the oxidizing mass were collected on a membrane filter. The size distribution of the powder residue was determined by a combination of sieving and air elutriation. The experimental apparatus is shown schematically in Figure A.15 and the measured results are reproduced in Table A.24 in Appendix A. The pertinent data are shown in Table 4-4.

Table 4-4. Measured ARFs During Self-Sustained Oxidation of Unalloyed Plutonium Metal in Flowing Air (Table III - Mishima December 1965)

Sampling Duration (min)	Temperature Range °C	Air Velocity cm/s	ARF
155	Amb to 900 °C	3.3	2.8E-8
74	Amb to 560 °C	13.5	3.1E-7
75	Amb to 650 °C	50.0	5.3E-7
146	Amb to 650 °C	3.3	4.1E-8
153	Amb to 560 °C	3.3	2.6E-7
117	Amb to 560 °C	20.0	3.1E-8

The values appear to be lower than the ARFs reported by Carter and Stewart (1970) for the airborne release during oxidation after ignition. The highest ARF is ~ 5E-7, an order of magnitude less than the geometric mean value specified by Carter and Stewart (1970). The measurements are limited and do not appear to be strongly influenced by any measured parameter (temperature, air velocity). A possible factor is the limited convective flow to entrain any particles ejected from the oxidizing mass due to the limited size of the specimen and the presence of a boat that may partially shield the oxide mass from the airflow. If the measurements represent almost comparable conditions, experimental

variations for the measurements would be a factor of ~ 20. The fraction of the residual powder 20 µm AED and less was <3.1E-4. The material was friable and the value may represent some fraction of material fragmented during sieving.

Mishima (November 1966) reported the ARFs and RFs from the self-sustained air oxidation of four large specimens of delta-phase and unalloyed plutonium metal. The three unalloyed pieces ranged in mass from 455.5 g to 1770 g. The single delta-stabilized specimen was 997 g. The metal specimen was placed within a ring made of insulating material set upon a sheet of insulating material (the metal would not sustain ignition in the absence of the insulating material due to heat transfer to the metal enclosure). The top surface of the metal specimen was ignited with a heliarc torch. Air was drawn into a quartz chimney placed within one inch of the metal surface at a nominal velocity of 525 cm/s through the 2.685-in. (68-mm) i.d. chimney. Airborne material was collected on a glass fiber filter sealing the end of the chimney. Air was drawn through a side arm at 550 to 800 cm/s to collect airborne materials on a membrane filter for size distribution analysis by transmission electron microscopy. The experimental apparatus is shown in Figure A.16 and the results reproduced in Table A.25 in Appendix A. The size distribution measured for material airborne during oxidation of the largest metal specimen, considered representative of all measurements, is reproduced as Figure A.17 in Appendix A. Pertinent data are listed in Table 4-5.

Table 4-5. Measured ARFs During the Self-Sustained Air Oxidation of Large Specimens of Unalloyed and Delta Plutonium Metal (Table II - Mishima November 1966)

Total Sampling Time (min)	Weight Metal (g)	Form	ARF
90	569.8	Unalloyed	4.9E-4
45	1770	Unalloyed	1.4E-4
22	997	Delta	3.4E-5
60	455.5	Unalloyed	3.9E-6

With the limited data base, the ARFs do not appear to correlate with any measured parameter (mass, form) but Haschke (July 1992) reports that the airborne release is time-dependent. The largest ARF, ~ 5E-4, is in the range of but exceeds the 95% confidence level value (1E-4) specified by Carter and Stewart (1970). Although the masses are larger, the duration required for complete oxidation to self-extinguishment appear to be much less than required for much smaller specimens used by Mishima (December 1965) and the ARFs are two to four orders of magnitude larger. The principal differences between the conditions under which these measurements and the previous measurements were made are the specimen masses (larger) and air flow patterns (up and around at higher nominal velocities).

The increase in ARFs are attributed to the higher convective flows and increased turbulence. The Mass Median Diameter shown in Figure A.17 is 4.2 µm Geometric Diameter and would approximate a 14 µm AED particle. Thus, an RF of 0.5 is a conservative assumption.

Carter and Stewart (1970) surveyed the data available primarily in the United Kingdom on the airborne release of plutonium during self-sustained oxidation. Based on the large body of data available, they recommended the following values for ignition and burning in air (melting), with air velocity less than 100 cm/s (static):

geometric mean	ARF 7E-6/RF 1.0
95% confidence limit	ARF 1E-4/RF 1.0.

Haschke (July 1992) reported in his conclusions "If the largest diameter of releasable materials is assumed to be 10 µm, the maximum releasable mass fraction for plutonium+O_2 is 0.0007 ...". Tables II through V that list the distributions state "Particle diameters are geometric, not aerodynamic." The fact that all estimates are geometric diameters, not AED, was confirmed in conversations with the author. If the GSD is assumed to be 2.0, the mass fraction of particles 10 µm AED and less would be two or more orders of magnitude less. Thus, the ARF and RF of 5E-4 and 0.5 from Mishima's data above that results in a mass fraction of 10 µm AED and less particles of 2.5E-4 certainly would bound Haschke's results. Based on the results reported for this configuration, bounding ARF and RF values are assessed to be 5E-4 and 0.5.

4.2.1.1.4 Disturbed Molten Metal Surface With High Turbulence. This subsection examines conditions involving flowing metal or actions involving continual exposed surface renewal of molten metal. Very energetic reactions of plutonium compounds, such as bulk hydrides, are representative of this phenomenological range as well.

Stewart (1963) reported on the oxides formed by heating small pieces (less than 13 g) of unalloyed and delta-alloy plutonium metal to various temperatures in various atmospheres. Two shapes were used: billets (cylinders 7 mm diameter by 10 mm long) and swarf (turnings). Various experimental configuration were needed for different experimental conditions. The data for higher temperature oxidation (in the region of temperatures found during self-sustained oxidation) and other pertinent information are covered in subsection 4.2.1.1.3. Results of one experiment in pure oxygen, which resulted in a violent reaction, are shown below:

> delta-phase plutonium, >1000 ºC, 5.34 g, oxygen atmosphere, ARF 2.1E-2, violent reaction, MMD 0.3 µm (AED) ...

Carter and Stewart (1970) reported the results of experiments to determine the ARF and RF for plutonium under fast reactor processing conditions. Two types of experiments to measure airborne release and particle characteristics were performed: free fall of ignited metal droplets and exploding wires. The ignited metal drop experiments were conducted in a 0.14-m diameter by 0.75-m tall vertical cylinder with a resistance furnace on top. Taller tubes were used for the U experiments conducted. The metal was heated in the resistance furnace to the predetermined temperature. An upflow of air adequate to entrain particles <30 µm AED was passed through the cylinder. For static experiments (air velocities <1.0 m/s), the Pu was heated in air and the residue crumbled/disintegrated into the cylinder. For the 660 ºC experiments, the Pu was heated in argon to the desired temperature and fell through the upflow of air in the cylinder (ignited and may have attained temperature equivalent to the 2000 ºC case discussed next). For the 2000 ºC (estimated from the temperature of ignited Pu in previous experiments) experiments, Pu metal was heated in air until ignited and allowed to fall through the upflow of air in the cylinder.

The morphology of the airborne particle from the free-fall drop of ignited metal appear to be very similar to that from the exploding wire experiments (see Table A.22, Comparison Between Exploding Wire Aerosol and Droplet Fume in Liquid Suspension, in Appendix A). Both show a wide size range of spherical particles with a significant number >1 µm AED. Sparking (incandescent airborne material) of the metal during the fall or on impact indicated the presence of large (0.1 to 1.0-mm) spherical particles in the residual powder and the presence of airborne vapor.

The total metal dispersed as particles or aggregates 10 µm unit density spheres (AED) or less was determined for each experiment. Figure 4-6 reproduced from Carter and Stewart (1970) shows representative distribution for the three major experimental regimes. The geometric mean and 95% confidence total Pu airborne dispersion as a function of accident stress are reproduced as Table A.23 in Appendix A. The ARF and RF values for the various conditions are:

geometric mean	ARF 3.5E-3/RF 1.0
95% confidence limit	ARF 1E-2/RF 1.0.

Both Mishima (August 1964) and Kanapilly (March 1982) reported on the measured ARF from Pu metal suspended over a gasoline fire. Two 100-gram Pu rods were suspended in a metal basket over a ignited pool of gasoline in a 4-ft square by 11-ft tall chimney. Estimates of the airborne release based on weight loss gave the highest values but difficulties in recovery of the residual materials created substantial uncertainty in the estimates. The authors quote 5E-4 as a bounding ARF value. Luna (February 1994) reevaluated the data for airborne release from the Vixen A trials (outdoor burning of Pu metal suspended above a

gasoline fire in a chimney). The ARF for the two trials (the plutonium metal was incompletely oxidized in one of the two experiments and the results are normalized for the fraction oxidized) were 1E-2 and 3E-2 with RFs of 0.01 and 0.008 respectively. The resultant ARF x RF values would be 1E-4 and 3E-4.

Eidson and Kanapilly (February 1983) measured the airborne release from heating small specimens of delta-stabilized Pu metal in various gas mixtures. The atmospheres were air, inert (argon), reducing (hydrogen or hydrogen-nitrogen in argon) and gas mixture from the decomposition of fuel or Insensitive High Explosives. Pu metal pieces 0.5 g and 1.0 g were placed in a tantalum crucible with a boat underneath to collect any powder residue that fell out of the crucible. A heated reaction atmosphere was introduced into the stainless steel foil-lined quartz tube surrounding the crucible-boat and passed into a foil-lined aerosol chamber. The particulate materials passing through the apparatus were collected on filters sealing the apparatus and exhaust. Airborne release was the summation of the activity collected on the filters and estimates of deposition based upon the activity collected on the foil samples extrapolated to the entire surface area of the tube and aerosol chamber. In later experiments, the entire tube and chamber were lined and provided estimates of deposition that appeared to be lower than estimates used in the earlier experiment. The size distribution and morphology of the airborne material are based upon transmission electron microscopy of material collected in the aerosol chamber. A schematic drawing of the system is reproduced as Figure A.18 and the experimental results are reproduced in Table A.26 in Appendix A. The pertinent experimental data and measured ARFs reported are shown in Table 4-6.

The ARFs appear to be in reasonable agreement (order of magnitude) with the ARFs specified by Carter and Stewart (1970) for Pu metal burning in air or melting. The maximum measured ARF is ~ 5E-3 exceeding Carter and Stewart's 95% confidence level value (1E-4), and that measured by Mishima (November 1966) (5E-4). The bounding ARF measured is from the values determined during the initial experiments and, according to the authors, may well be overestimated due to accounting for material that may not have been related to airborne release. The median ARF is 3E-5 with an average value of ~ 3E-4.

The size distribution of the residual powder was determined by a combination of 320 mesh sieve (~ 62 µm LLD - the sieves are vibrated during use to cause the particle to bounce on the sieve screen and the material passing through the screen indicates that the non-spherical particles have at least one dimension of the size of the opening) and sedimentation measurements. The range of residual powder in the less than 62 µm LLD was 18% to 41% with MMDs from 5.7 to 7.5 µm geometric diameter (D_g). The material was primarily PuO_2 (x-ray diffraction) with some unburned metal particles. The authors conclusions were that 1) atmosphere and temperature affected the formation of the powder residue but did not affect aerosol formation, and 2) mass-to-surface ratio did not have a discernible affect on

Table 4-6. Measured ARFs for Heating Small Specimens of Delta Phase Pu Metal in Various Atmospheres (Table A-2 - Eidson and Kanapilly February 1983)

Reaction Atmosphere	Mass of Specimen (g)	Temperature (°C)	ARF
argon (air)	0.1	1000	1.7E-4[1]
argon	0.5	1000	1.2E-5[1]
argon + IHE ash	1.0	1000	1.6E-5
air	0.5	23	2.4E-5[1]
	0.5	450	1.6E-4[1]
	0.5	450	2.0E-5[1]
	1.0	1000	9.0E-6
3% H_2 + argon	0.5	100	5.7E-6[1]
	0.5	200	1.5E-3[1]
	1.0	200	1.0E-5
	0.5	450	2.8E-5[1]
	0.5	450	4.1E-5[1]
	0.5	450	9.6E-5[1]
	1.0	450	3.7E-5
	0.5	650	1.4E-5[1]
	1.0	650	5.1E-5
	0.5	200 - 1000	8.8E-5
	1.0	100 - 450	1.3E-4
3% H_2 + 5% N_2 + argon	0.5	100	1.8E-5[1]
	0.5	450	1.5E-5[1]
	0.5	450	5.2E-3[1]
	1.0	450	2.3E-5
	0.5	650 - 750	5.2E-6[1]
Kel-F + argon	0.5	450 - 650	1.0E-5
Kel-F #800 + argon	0.5	450	2.5E-4
	1.0	450	3.2E-5
Kel-F #800 + air	1.0	650	2.6E-5
complex (1)[2]	0.5	450	7.6E-6[1]
complex (2)[2]	0.5	450	2.8E-5
	0.5	650	2.4E-4
complex (2)[2] + NO + H_20	1.0	200	1.3E-5
	1.0	450	1.2E-5
	1.0	1000	1.3E-5

[1] Values determined during initial experiments. May be overestimated.
[2] Complex = IHE combustion product surrogate . . . (1) + 100 ppm O_z. . . (2) < 5 ppm O_2

aerosol formation under the experimental conditions. The size distribution measured for the airborne particles under various conditions of atmosphere and temperature are shown in Figures A.19 and A.20 taken from the reference document. In all cases, the RFs range from 0.05 to 0.5.

Eidson, Yeh, and Kanapilly (1988) reported the airborne release and size distribution from heating 1-gram or 10-gram pellets or foil to 450 °C in a reducing atmosphere followed in some cases by heating the powder produced in air. Gas velocities over the reacting materials ranged from 0.4- to 10-m/s in the 24-mm i.d. quartz reaction tube. The airborne materials were collected on a glass fiber filter or sized in a 7-stage cascade impactor. Point-to-plane electrostatic precipitator samples were taken for transmission electron microscopy to characterize the airborne materials. The pertinent data are shown in Table 4-7 with the data from the source document reproduced in Table A.27 in Appendix A.

Table 4-7. ARFs From Heating Small Pieces Delta-Phase Plutonium Metal Under Specific Conditions (Table 1 - Eidson, Yeh and Kanapilly 1988)

Atmosphere	Linear Flow Velocity cm/s	Pu Mass grams	Specimen Shape	ARF[1]
3% H_2 + Ar then air	500	0.94	pellet	1.1E-4
	1000	1.05	pellet	1.1E-3
	1000	0.92	pellet	7.4E-4
3% H_2 + Ar	500	1.08	pellet	3.7E-3
	1000	1.12	pellet	4.6E-3
3% H_2 + 5% N_2 + Ar	500	1.05	pellet	3.0E-4
	1000	1.13	pellet	6.6E-5
	1000	1.05	pellet	4.5E-4
	40	1.39	foil	9.2E-5
	40	9.63	pellet	1.2E-5
	40	10.22	foil	8.5E-6
3% H_2 + Ar	40	1.40	foil	2.8E-4
	40	9.62	pellet	1.8E-5
	40	9.64	foil	4.2E-4

[1] All in the respirable fraction defined in this study as particles 5 µm AED and less.

The bounding ARF and RF are 5E-3 and 1.0 with median values of 2E-4 and 1.0. There does appear to be a detectible effect by the atmosphere present or surface to mass ratio. The MMD of the residual powders ranged from 4 to 350 µm Stokes diameter (equivalent sphere). The Activity Median Aerodynamic Diameter (AMAD, in which half the activity in the sample is associated with particles less than and half the activity is associated with particles

greater than the stated size), for the airborne materials measured by cascade impactor ranged from 4 to 10 µm AED.

Chatfield (1969) measured the Pu airborne during the combustion (oxidation after ignition) or explosive release of unalloyed Pu foil encased in sodium. The oxidation experiments were performed in a horizontal glass tube externally heated by a resistance furnace. Air or gas, at a velocity of 0.8 m/s, was drawn through the tube over the Pu foil encased in a rectangular block of Na held in a nickel or tantalum boat to sampling equipment (Casella Mk 3 cascade impactor or membrane filter). The Pu-Na was heated in an argon flow to a temperature of 300 °C and the flow switched to air. The results from these experiments are shown in Table A.31 (Tables 1 & 2 from the source document) in Appendix A. The ARF are presented as µCi of activity released but no specific activity for the source material was given. Under the assumption that the Pu is "weapons grade" material similar to that specified in Eidson and Kanapilly (February 1983) and Raabe et al. (November 1978) with a specific activity of 7.4E-2 µCi/g, the release fractions shown in Table 4-8 were estimated.

Table 4-8. Airborne Release of Pu From Pu-Na Mixtures During Self-Sustained Oxidation (Table 1 - Chatfield 1969)

Weight Pu (grams)	Activity in Source (µCi)	Fraction Activity Airborne (µCi)	Mass Fraction Pu Airborne	RF[1]
0.0549	4.06E+3	1.7E-3	4.2E-7	0.88
0.0570	4.22E+3	1.5E-3	3.6E-7	0.80
0.0644	4.77E+3	1.8E-3	3.8E-7	0.72
0.0830	6.14E+3	1.5E-2	2.4E-6	0.40
0.1021	7.56E+3	5.0E-2	6.6E-6	
0.0899	6.65E+3	2.8E+0	4.2E-4	0.98
0.0946	7.00E+3	8.0E-2	1.1E-5	
0.1069	7.91E+3	5.6E-2	7.1E-6	
0.0732	5.42E+3	4.0E-3	7.4E-7	
0.0710	5.25E+3	5.0E-2	9.5E-5	
0.0981	7.26E+3	4.7E+1	6.5E-3	0.28
0.0900[2]	6.66E+3	1.8E-1	2.7E-5	0.11
0.0763[3]	5.65E+3	5.5E-2	9.7E-6	0.27
0.0651[3]	4.82E+3	2.1E-1	4.4E-5	0.41
0.0915[4]	6.79E+3	3.5E-2	5.3E-6	0.12

[1] Pu size data presented as particles 11 µm and less AED (unit density spheres). Conservative assumption to include all material in respirable fraction.
[2] Pu as dioxide powder dispersed on sodium wire.
[3] No sodium used in this experiment.
[4] No ignition, Pu heated in argon only.

The authors concluded that 1) total airborne release was unaffected by the presence of sodium (the ARFs measured for the two experiments in which only Pu was present were well within the range of ARFs for the remaining experiments), 2) the fraction of airborne particles in the <11 µm AED fraction was higher when Na was present (0.28 to 0.98 vice 0.27 and 0.40), and 3) the fraction <3.7 µm AED is also higher. The fact that the presence of Na did not result in greater total airborne releases with the procedures used (Pu-Na heated in argon with air suddenly introduced at temperature) is surprising in that the reaction as described appears to be very energetic. Perhaps the fact that the Na surrounded the Pu and reacted first may have limited oxygen availability and thus the Pu oxidation rate. Thus, Pu oxide may not have been generated until after the greater part of the turbulence from the sodium reaction was terminated. The ARF measured for the experiment in which Pu was only heated resulted was in the range for the other experiments but the RF was low (0.12). Review of the data does not indicate any specific cause to which the wide variability in the data can be attributed.

The ARFs measured range from 3.6E-7 to 6.5E-3. The maximum ARF measured was 6.5E-3, which is in the range of ARFs reported by Eidson and Kanapilly (February 1983) and Stewart (1963) of 1E-2. The median ARF measured is 7E-6 with an average of 5E-4. The ARFs for the 2 experiments involving only Pu were 9.6E-6 and 4.4E-5 with an average value of ~ 3E-5. The RFs are 0.27 and 0.41 with an average of 0.34. The average RF for the 5 experiments involving Pu with sodium was 0.76; twice as high as for Pu only. Since the value exceeds the measured values for 5 experiments, the reported ARFs may be an order of magnitude less.

The data from Eidson and Kanapilly (February 1983) and Eidson, Yeh and Kanapilly (1988) were included in this configuration primarily because the intent was to determine airborne release under more severe accident conditions and other atmospheres. With a few exceptions (5 ARFs ranging from 1.1E-3 to 5.2E-3) the results would be bounded by the ARF and RF values assessed as bounding for airborne release during self-sustained oxidation. These results are for specimen weights ~ 1 gram or less and in some cases (heating in hydrogen and argon atmospheres) would appear to represent ARFs for hydrides. However, the Carter and Stewart data and other studies bound these results. Accordingly, bounding ARF and RF values are assessed to be 1E-2 and 1.0.

4.2.1.1.5 Small Molten Metal Drops Hurled Through Air or Explosion of Entire Metal Mass. The term "small molten metal drops" refers to drops with a maximum diameter in the hundreds of µm range. It is also noted that the term "explosion of entire metal mass" refers to a phenomena internal to the metal matrix itself, not external explosive effects.

Carter and Stewart (1970) reported the results of experiments to determine the ARF and RF for plutonium under fast reactor processing conditions. Two types of experiments to measure airborne release and particle characteristics were performed: free fall of ignited metal droplets and exploding wires. The ignited metal drop experiments were conducted in a 14-cm diameter by 75-cm tall vertical cylinder with a resistance furnace on top. The dimensions were limited by the size of the glovebox for delta-alloy Pu experiments. Taller tubes were used for the U experiments. The metal was heated in the resistance furnace to the predetermined temperature. An upflow of air adequate to entrain particles <30 µm AED was passed through the cylinder. For static experiments, the Pu was heated in air and the residue crumbled/disintegrated into the cylinder. For the 660 °C experiments, the Pu was heated in argon to the desired temperature and fell through the upflow of air in the cylinder (ignited and may have attained temperature equivalent to the 2000 °C case discussed next). For the 2000 °C (estimated from the temperature of ignited Pu in previous experiments) experiments, Pu metal was heated in air until ignited and allowed to fall through the upflow of air in the cylinder.

For the exploding wire experiments, 50 to 400 mg of metal were violently dispersed by a large electrical charge discharge (4000 J) through the metal in a 3.5-liter chamber. The aerosol within the chamber was discharged ~ 1 min after formation via a cascade impactor (size distribution of airborne particles) and a membrane filter (transmission electron microscopy for morphology). Based on these results, bounding ARF and RF values (at a 95% confidence limit) for vapor formation from droplets (exploding wire, violent ejection molten droplets) are assessed to be 5E-1 and 1.0.

Raabe, et al. (November 1978) reported the ARF and size distribution of the fume made airborne during the free-fall of ignited drops of delta-phase plutonium metal. Small discs were cut from 50-µm-thick foil in the size range believed representative of fragments that could result from explosive damage. The discs were positioned on a very thin film of combustible material on top of a 0.15-m wide by 0.18-m deep by 3-m tall stainless steel chamber with a glass viewing window down the front panel. The discs were ignited by a laser (500 °C, adequate to ignite but not to vaporize metal) and formed drops 50 to 500 µm in diameter. The ignited material fell down the chamber. Air was drawn through a perforated plate down through the chamber. Large particles were collected in a aluminum foil lined cup at the bottom of the chamber with airborne materials carried to aerosol samplers (seven-stage cascade impactor, spiral centrifuge, point-to-plane precipitator + glass fiber filters). The experimental apparatus is shown in Figure A.21

Essentially all of the plutonium available was driven airborne with ~ 40% of the source in the respirable size range. The airborne material was primarily a web-like chain of crystalline, cubic particles 0.004 to 0.1 µm on a side with a few discrete spherical particles

up to 0.5 μm in diameter. The AMAD of the aggregates was 1 to 2 μm with geometric standard deviations of ~ 1.5. The size distribution of the airborne material from free-fall of a 400-μm drop as determined by the spiral centrifuge is reproduced as Figure A.22 (Appendix A).

Chatfield (1969) reported results of exploding wire experiments primarily to determine the morphology and solubility of the particulate materials generated. Plutonium wire, 0.75-mm, or plutonium wire encased in 2-mm sodium metal tubing were placed in heavy current electrodes in the side of a 2.5-liter vessel. Energy of 4000 J accumulated in a capacitor bank was discharged at 10 kV vaporizing the wire at a very high temperature (peak temperature ~ 50,000 ^{o}K). The fume formed by the condensation was collected directly on carbon-coated electron microscope grids with the remainder exhausted through a membrane filter. All the Pu or Pu-Na involved was made airborne. The Pu fume was composed of linear aggregates of generally spherical particles <0.2 μm in diameter. It was observed that in some cases only particles of similar size appeared to form aggregates. The MMD of the aggregate was 1.4 μm with a very narrow distribution. The size distribution of the aerosol is plotted in Figure 4-7 reproduced from the referenced source and, since the diameters plotted are Geometric Diameters, indicates that essentially all of the airborne material is in the respirable fraction.

On the basis of the experimental data presented, the ARF and RF is either 5E-1 and 1.0 or 1E+0 and 0.4. Based on a combination of values from these experiments, the bounding ARF and RF values assessed for this handbook are 1E+0 and 0.5.

4.2.1.2 Uranium

Mishima, et al. (March 1985) reviewed the published literature on uranium behavior under fire conditions. For natural or depleted uranium or uranium with ^{235}U enrichment <10%, the toxic hazard of uranium as a heavy metal is of greater concern than the radiological hazard. The toxicological hazard from uranium results from transport of inhaled, soluble uranium compounds to the kidneys. For non-volatile (soluble and non-soluble) materials to be an inhalation hazard, the size of the particles/aggregates must be 10 μm AED (more probably 3 μm AED) or less. For normal and depleted uranium, the materials must be soluble. For uranium with enrichments >10%, the radiological hazard is of concern and the solubility of the uranium in interstitial lung fluids determines the critical organ. Fire is a phenomenon that could subdivide uranium metal by conversion to the oxides.

Due to the similarity in matrix spacing, hyperstoichiometric uranium dioxide formed at the metal-atmosphere interface is adhering and limits oxygen availability. At temperatures

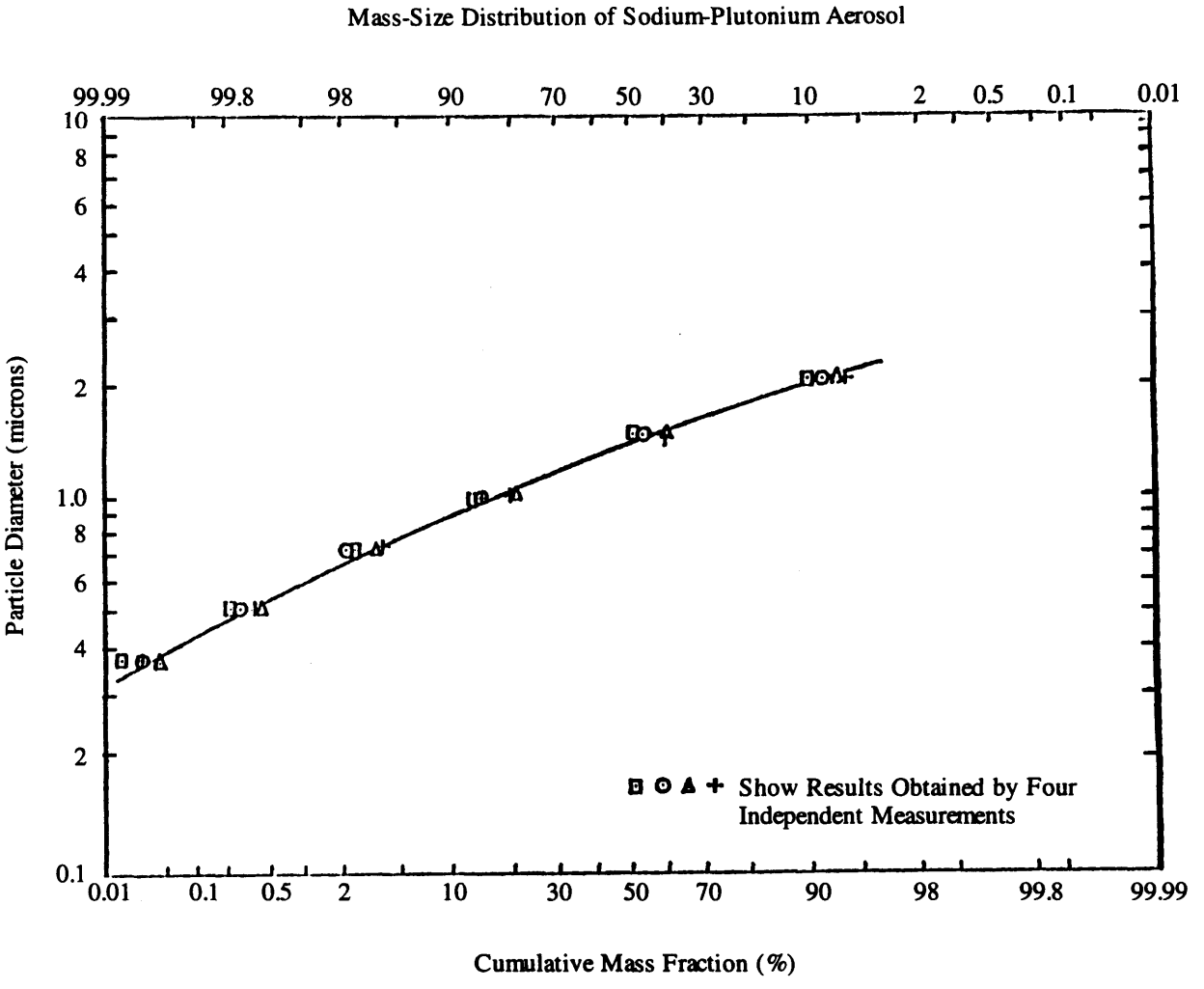

Figure 4-7. Mass-Size Distribution of Sodium-Plutonium Aerosol (Figure 8 - Chatfield 1969)

<200 °C, the hyperstoichiometric dioxide, UO_{2+x}, is the principal product. At slightly higher temperatures, a mixture of various suboxides (e.g. U_3O_7, U_3O_8, etc.) are found. At temperatures >275 °C, UO_2 and predominantly U_3O_8 are produced. In the temperature range of 350 °C to 600 °C, the UO_2 formed rapidly oxidizes to U_3O_8 that falls away as a black, fine powder. In the temperature range of 650 °C to 850 °C, the UO_2 forms a protective layer that at some point breaks away. At temperature >900 °C, the UO_2 is adherent and protective. The presence of water vapor accelerates oxidation in air at temperature <300 °C and in carbon dioxide at temperatures <350 °C to 500 °C. Uranium reacts with hydrogen, nitrogen and carbon at elevated temperatures and the presence of surface inclusions accelerates oxidation.

The presence of some additive used to phase-stabilize uranium (e.g., aluminum, titanium) may change the first- or second-stage oxidation rates or the break weight (plateau) or prevent transition to protective oxide formation that may result in a single, accelerated oxidation rate. Some of the factors that affect oxidation rates are listed in Table 4-9 taken from the reference document. Measured oxidation rates in air, carbon dioxide and oxygen are available in the reference document, but the oxidation rate during a fire will be the sum of a variety of rates dependent upon local conditions at many sites on the metal surface and is difficult to predict.

Table 4-9. Possible Factors Influencing Uranium Oxidation (Table 4.1 - Mishima et al. March 1985)

Step	Possible Factors Influencing Rate
Metal surface of unit area oxidizing in air	1) Metal purity 2) Metallurgical condition (grain size, strains, etc.) 3) Temperature 4) Time 5) Gas composition 6) Type of oxide film formed (protective or not)
Loss of heat of reaction by conduction to the surroundings	1) Thermal conductivity of metal 2) Thermal conductivity of oxide coating 3) Cross-sectional area at right angles to direction of heat flow 4) Temperature gradient

Unlike plutonium, uranium is difficult to ignite. The presence of an adherent, protective layer of hyperstoichiometric dioxide at the interface limits oxygen availability. Also, the heats of reaction are lower. Figure 4-8 reproduced from the reference document shows the ignition temperature for uranium as a function of surface area/mass ratio. At surface to mass ratios <1.0 cm^2/g, the ignition temperature exceeds 500 °C and is increasing rapidly

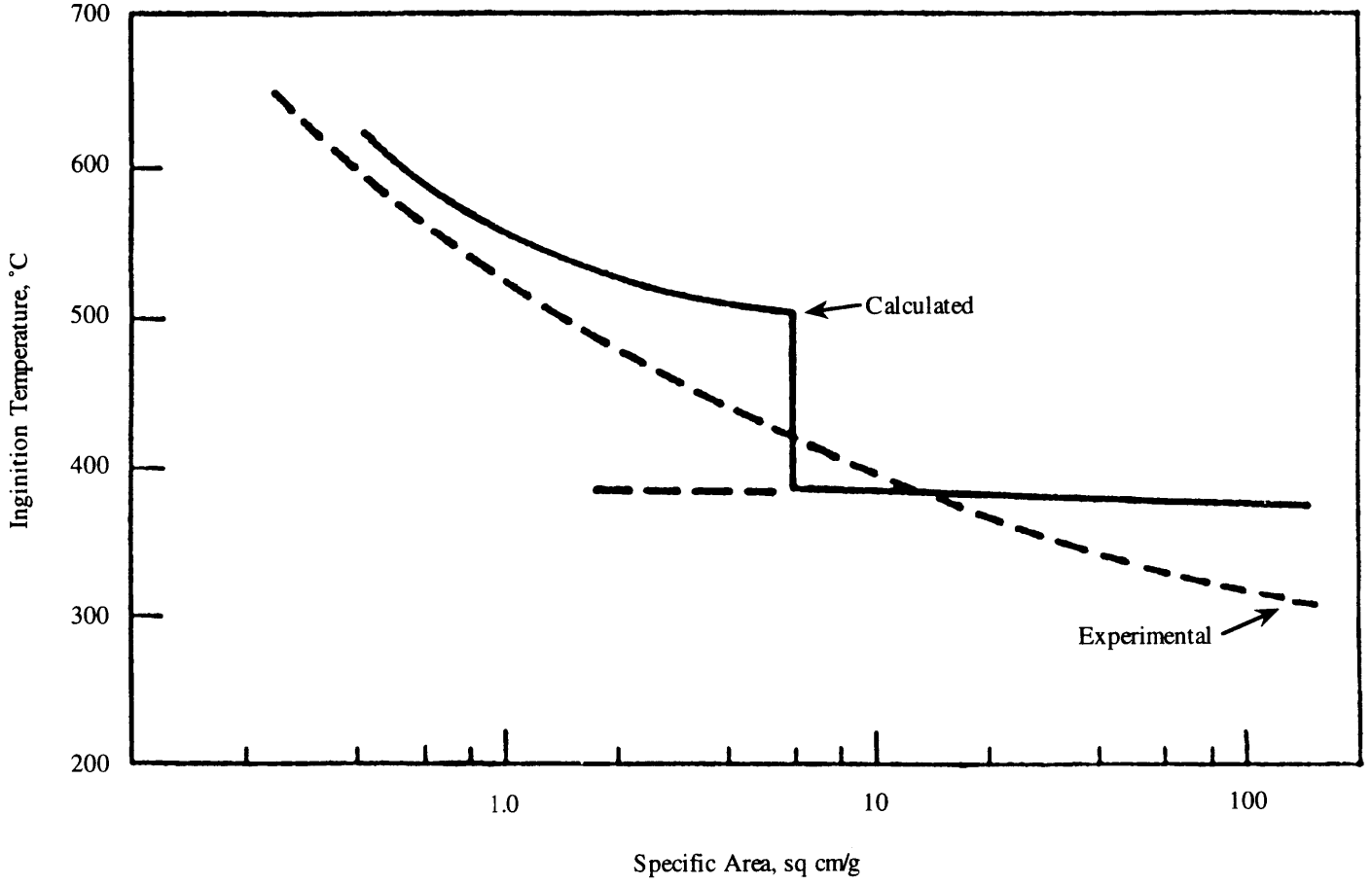

Figure 4-8. Dependence of Uranium Ignition on Specific Area
(Figure 4.1 - Mishima, et al. March 1985)

indicating that large pieces of uranium are very difficult to ignite as large amounts of external heat must be supplied and serious heat loss prevented.

The particle size distributions of residual oxides produced under a variety of conditions have also been measured and are shown in the reference document (see Figures 4.8, 4.10 and 4.11). The distribution becomes coarser and the solubility in simulated lung fluid decreases as the temperature increases. Oxidation of the metal at <450 °C generated a fine, black non-adherent powder. At temperatures around 535 °C, the oxide was a fine, black powder sintered into lumps. At temperatures >700 °C, the oxide appeared to be a hard, black scale.

The ARF and RF for three potential accident configurations for thermal stress (airborne release during the oxidation of uranium at elevated temperatures, airborne release from disturbed molten uranium surfaces, and airborne release during explosive release of fine molten metal drops) are covered below.

4.2.1.2.1 Oxidation at Elevated Temperatures. Mishima et al. (March 1985) characterized the oxide generated by the April 1983 burn test involving munitions containing depleted uranium (DU) penetrators and reviewed the literature on airborne release. Tests subjecting munitions to rigorous fire conditions are performed prior to deployment to ascertain the thermal and blast hazards during transport and storage. Twelve 120-mm rounds containing 48 kg of DU as rods ~ 1 in. in diameter by 30" long were subjected to a wood and diesel fuel fire. The rounds cooked-off (i.e., the propellant used flared) and the DU rods were retained in the burning mass at temperatures from 800 to 1100 °C range for ~ 3 hours. No detectible airborne DU was collected by air samplers surrounding the burn at distances <100 m. Samples of the oxides generated were collected and the particle size distribution, morphology and solubility in simulated interstitial lung fluid were measured. The fraction of the oxide generated by the burn <10 µm AED ranged from 0.2 to 0.65 wt/o. The fraction of the residual oxide <10 µm AED were predominantly U_3O_8 and all in the "Y" class (dissolution halftimes in simulated interstitial lung fluids of >100 days).

The ARF x RF values for uranium during oxidation at elevated temperatures found in the literature were:

Elder and Tinkle (December 1980):	
Air, up to 3.2 m/s, fire	5E-3
Air/Air-CO_2, 3.2 m/s, 500 °C	1E-7
900 °C	4E-6
Carter and Stewart (September 1970)	
Air, static, molten metal	4E-4
Free-fall molten drops	6E-3

Carter and Stewart performed experiments to measure characteristics of airborne uranium from molten metal under static (no metal movement) and dynamic (free-fall drop) conditions. The experimental apparatus and procedures were covered in Subsection 4.2.1.1.4. The mean and 95% confidence level ARF x RFs from the oxidation of static molten metal with airflow around and over the metal were 1.1E-4 and 3.6E-4 respectively.

A. Oxidation of Depleted Uranium (DU) Rods at Elevated Temperatures in a Fire. Elder and Tinkle (December 1980) performed a series of experiments on DU rods used as penetrators in armor-defeating weapons. The rods were made of staballoy (beta-stabilized uranium, 99.25% uranium + 0.75% titanium) with a nominal diameter of 25.9-mm (~ 1 inch), a length of 0.345 m (13.6 inches), and weighing 3355 +/- 3 grams. The rods were subjected to oxidizing conditions (heat + air or air-carbon dioxide atmospheres) with a test configuration for each set of conditions. The various testing configurations are shown in Figure A.25 and A.26 in Appendix A. The rods were heated in a rack in an upflow of air. In the first three tests, the heat was generated from either ignited uranium turnings or munitions propellant. In the fourth test (burn 4), heat was supplied by 10 batches of packing materials (wood and paper). Observations indicated that the rods in the first three tests under went very little if any oxidation. From 42% to 47% of the three rods tested in Burn 4 were oxidized greatly exceeding the largest value for fraction oxidized in the laboratory study (30%) and was attributed to the spalling of the oxide coat from the temperature fluctuations resulting from introduction of the 10 batches of fuel. Individual 5-min air samples taken during portions of four fuel additions show apparent airborne concentrations ranging from 4.2 to 783 mg U/m^3 with fractions <10 μm AED from 20% to 62%. The times during the oxidation process when the samples were extracted are not reported and the great variation in the mass airborne concentrations makes determining the ARF difficult.

The oxidation rate and airborne release were measured during thirteen laboratory experiments in air or 50% air - 50% carbon dioxide at temperatures from 500 to 1000 ^oC. In twelve of the tests, a gas velocity of 2.23 m/s (5 mph) was passed around the oxidizing rod. In one experiment (air at 700 ^oC), the test was performed under static conditions (no gas flow). No self-sustained reaction was observed under any of the test conditions. The fractions oxidized under these conditions ranged from 6.0% to 30.2% (original data tables reproduced as Table A.32 in Appendix A). The total aerosol mass (summation of the high volume filter sample, precutter and cascade impactor stages + back-up filter) are shown in Figure 4-9 reproduced from the reference document. It is difficult to ascertain if the presence of carbon dioxide has any discernable effect upon the airborne release due to experimental scatter and

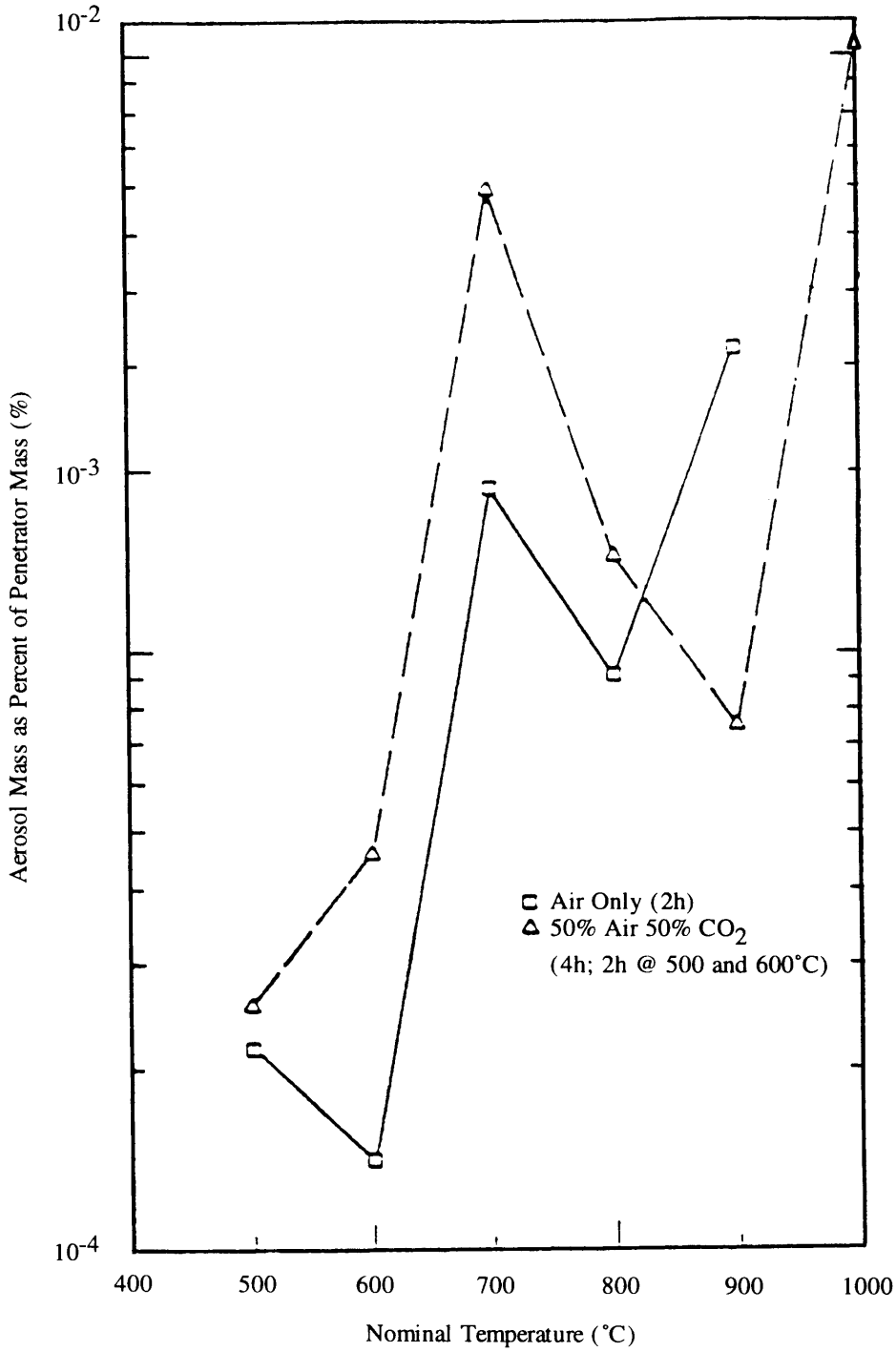

Figure 4-9. Aerosol Mass of Uranium Airborne during Oxidation as a Function of Temperature
(Figure 22 - Elder and Tinkle December 1980)

different oxidation periods used. The mass of particles 10 µm AED and less as a function of temperature is shown in Figure 4-10 reproduced from the reference document. The values range from ~ 1E-7 at 500 ºC to 8E-6 at 700 ºC and 900 ºC for the fraction of the total mass oxidized. Adjusted for the time to completely oxidize the metal would increase the apparent ARFs to 1E-4 to 4E-2 with RFs from 0.006 to 0.17. The ARF x RFs range from 5E-5 to 4E-3 and are comparable to the values reported by Carter and Stewart (1970). The authors listed the following conclusions:

1. Uranium particles in the respirable size range (10 µm AED and less) were made airborne when the rods were exposed to temperature exceeding 500 ºC for time greater than ~ 0.5 hours.

2. Production of oxide and airborne materials were enhanced by forced-draft and temperature cycling during oxidation.

3. Metal rods with the test configuration did not exhibit any tendency towards self-sustained oxidation, although complete oxidation would no doubt be achieved if adequate fuel and time (longer than 4 hours) were provided.

B. Review of Experimental Studies on Airborne Release From Depleted Uranium Munitions. Jette et al. (August 1989) reviewed the published information available on the characteristics of the DU particles suspended during testing (firing of kinetic energy rounds against hard targets, burn tests during hazard classification of rounds prior to deployment) of the munitions and one study on the characteristics of the aerosols from the explosive ejection of molten metal droplets. Many studies have been performed on the DU particles formed by the impact of penetrators against hard targets (Gilchrist and Nicola, January 1979; Glissmeyer and Mishima, November 1979; Chambers et al., October 1982; Sutter et al., January 1985; Wilsey and Bloore, May 1989; Parkhurst et al., April 1990; Jette, Mishima and Hadlock, August 1990). Generally, a substantial portion of the mass of DU in the penetrator becomes airborne by the impact against hard targets (armor) of sufficient thickness to expend most of the energy of the kinetic round (up to 80%). The size of the airborne material is very fine with fractions in the 10 µm AED and less range of 0.34 to 1.0. The airborne materials are predominantly U_3O_8. Up to 50% of the particles in the respirable fraction may be "D" class (dissolution halftime <10 days).

The other large group of studies providing information on the potential behavior of uranium under accident conditions are the hazard classification test conducted on

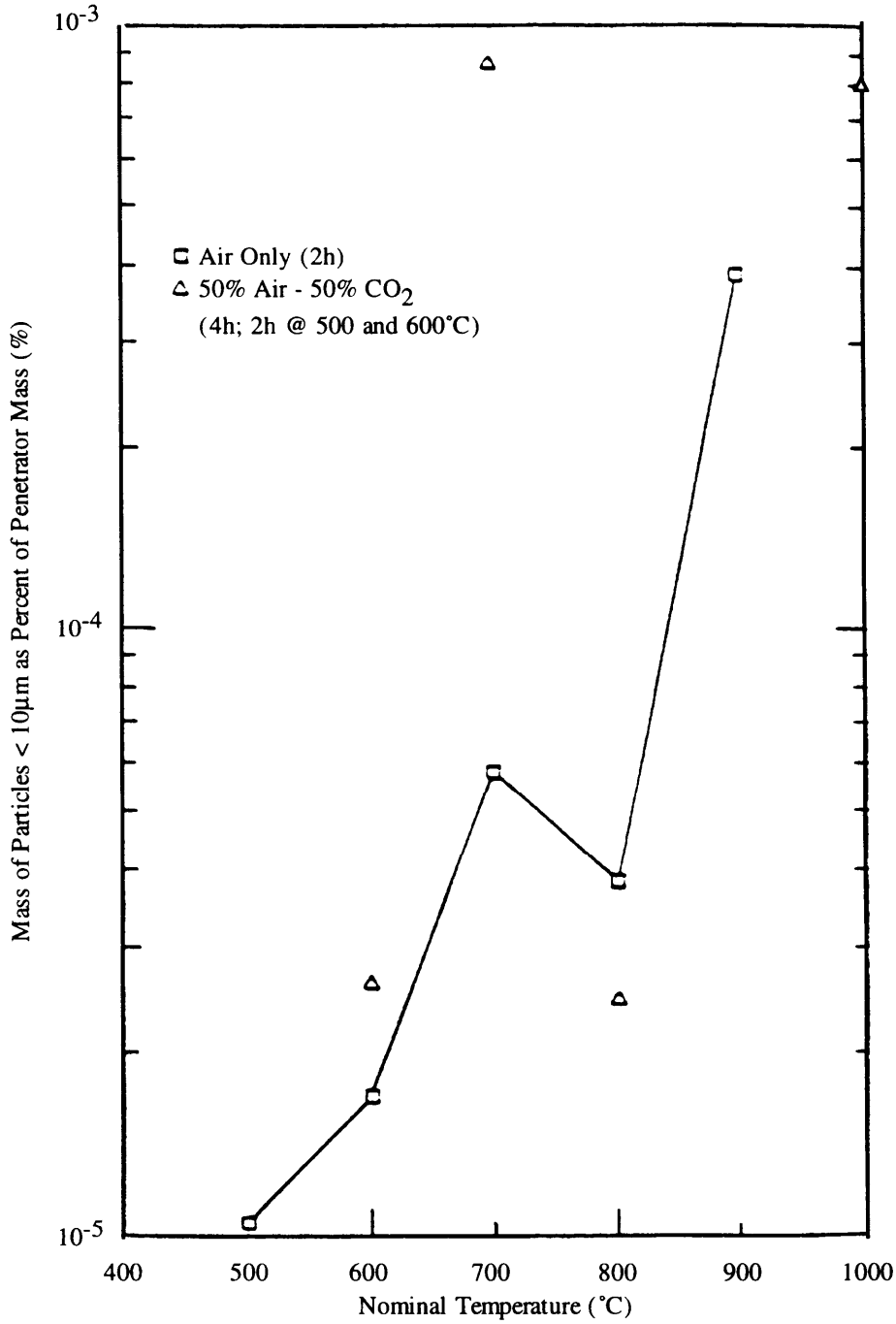

Figure 4-10. Respirable Mass of Uranium Airborne during Oxidation as a Function of Temperature
(Figure 23 - Elder and Tinkle December 1980)

munitions prior to deployment (Gilchrist, Parker and Mishima, March 1978; Hooker et al., March 1985; Haggard et al., July 1986; Parkhurst et al., March 1990). A pre-determined number of boxes of munitions are subjected to an intense wood-fuel oil fire. The distance large fragments (pieces of munitions components and cases, packing) are ejected and the thermal and blast levels are determined to establish the exclusion area requirements in the event of accidents in transport and storage. In all cases, no airborne DU was collected in the air samplers set downwind at various distances downwind of the fire. Size distributions of the residual oxide powders (predominantly U_3O_8) were determined and estimates of the respirable fraction are based on the presence of particles of 10 μm and less AED in the residual oxide. The size distributions measured show less than 0.01 of the residual oxides are in the respirable size range. The material in the respirable fraction is much less soluble than the airborne oxide from impact tests ranging from 96% to 100% in the "Y" class.

From the data reproduced in Figure 4-11 from Carter and Stewart (1970), a geometric mean ARF x RF of 1E-4 was reported. The 95% confidence level ARF x RF of 4E-4 reported by Carter and Stewart (1970) is exceeded by the ARF and RF estimated for tests performed by Elder and Tinkle (December 1980), where the gas flow and temperatures used exceeded those used by Carter and Stewart (1970). Six of nine ARF x RF values obtained are less than 1E-3, with the greater values at the higher temperatures (900 ^{o}C). The ARF and RF values of 1E-3 and 1.0 are assessed to be bounding for this thermal stress configuration. The value for the lung solubility class is assumed to be that determined for the sintered oxides collected from wood-oil fires involving DU rods in munitions (i.e., >95% "Y" class with the remainder being "D" class uranium). This value is assessed to be bounding for the issue of lung solubility class.

4.2.1.1.2.2 Disturbed Molten Metal Surface With High Turbulence. Carter and Stewart performed a series of experiments to measure the characteristics of airborne uranium from molten metal under static (no metal movement) and dynamic (free-fall drop) conditions. The experimental apparatus and procedures were covered in subsection 4.2.1.1.4. The size distributions of the airborne materials are shown in Figure 4-11 reproduced from that document (the velocity of the updraft through the vertical tube was set at a velocity to carry particles <30 μm AED). The distribution of airborne material from the experiments under static conditions appears to be a single mode. The airborne material from experiments under dynamic conditions (2 to 4 m free-fall) are bimodal. The distribution from the experiments in which "obvious sparking" was observed appears to fit the coarser (upper) curve with the finer (lower) curve represented by the airborne material under static conditions.

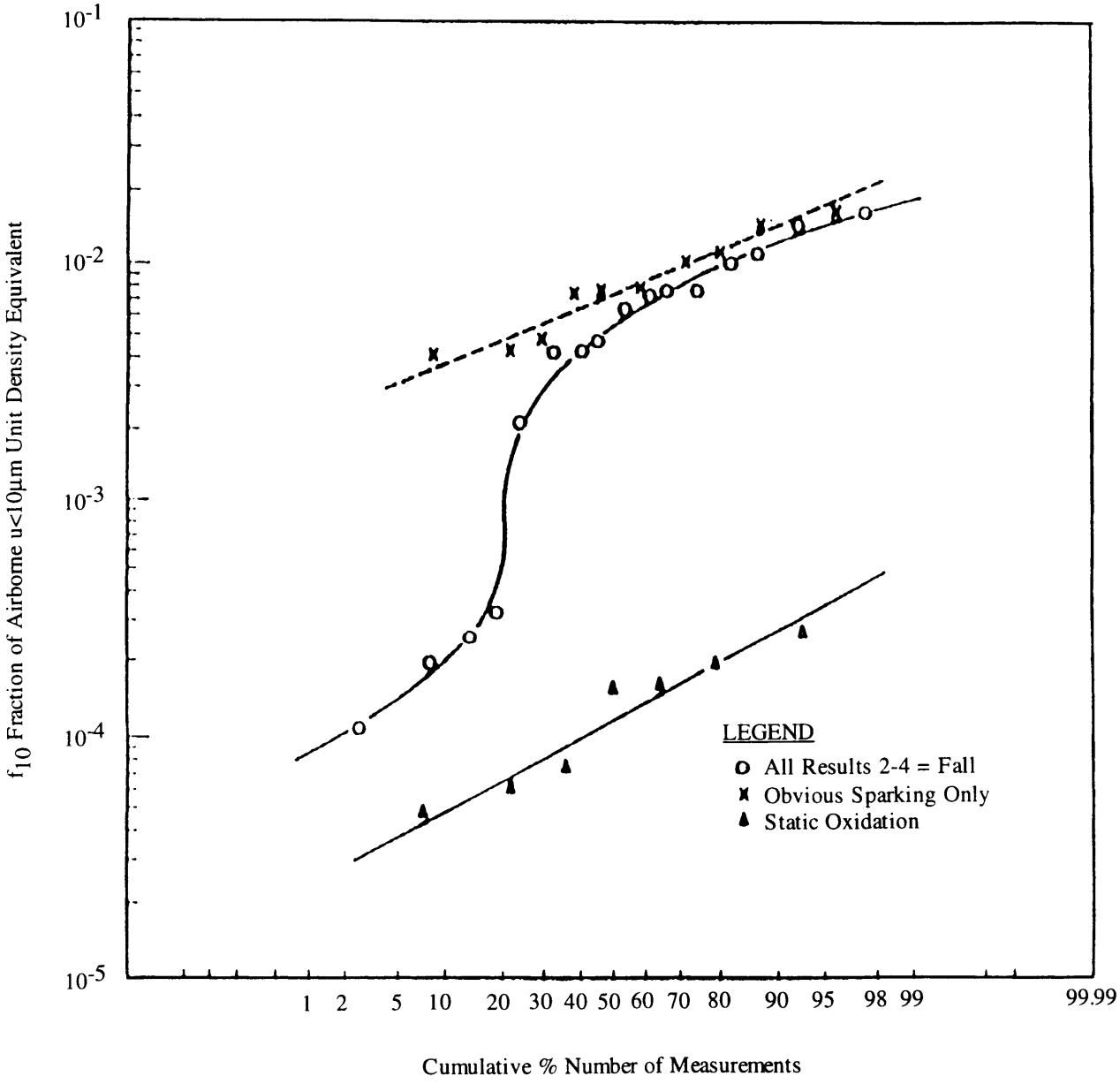

Figure 4-11. Uranium Metal Burning in Air; The Distributions of the Measurements of the Respirable Aerosol Fractions in the Static and Dynamic Experiments (Carter and Stewart September 1970)

Fractional airborne release values of particles 10 µm AED or less for these conditions are reproduced in Table A.23 in Appendix A and are:

Dynamic, Geometric mean	1.9E-3
95% confidence level	6E-3

On the basis of the available experimental data, the "median" ARF x RF value is assumed to be the geometric mean value, 2E-3, reported by Carter and Stewart (1970). The bounding value is assumed to be the 95% confidence ARF x RF value, 6E-3, reported by Carter and Stewart rounded upwards to be the same as the comparable value for Pu, 1E-2. Since the airborne material cooled rapidly after formation and would be comparable to the fine particulate material from the plastic deformation and rapid oxidation of a thin film of metal generated during the impact of DU rods against hard targets (armor), the measured value for this situation, 50% "Y" class + 50% "D" class, is assessed to be bounding.

4.2.1.2.3 Small Molten Metal Drops Hurled Through Air or Explosion of Entire Metal Mass. "Small molten metal drops" refers to drops with a maximum diameter in the hundreds of µm range. It is also noted that the term "explosion of entire metal mass" refers to a phenomena internal to the metal matrix itself, not external explosive effects.

Rader and Benson (June 1988) generated molten uranium by the exploding wire technique and accelerated the molten drops downrange by an electrostatic device. Approximately 36% of the molten material had been aerosolized by the time the drops had traveled 6-ft and would probably all aerosolize within the next 10 to 12-ft. The size of the airborne particles was <1 µm AED with a geometric standard deviation of ~ 2. The majority of the airborne material was in the form of web-like aggregates. Experiments performed in inert gas (argon) aerosolized orders of magnitude less material although the size distribution/geometric standard deviation of the airborne material remained unchanged demonstrating the importance of oxidation to the airborne release phenomena.

On the basis of the available experimentally measured value, the bounding ARF and RF values for this accident configuration are assessed to be 1.0 and 1.0. As in the previous subsection, the bounding solubility class for this type of airborne material (rapid cooling of fine particulate material/fume) is assessed to be 50% "Y" class and 50% "D" class.

4.2.2 Explosive Stress: Shock, Blast, and Venting

Materials can react explosively from internally generated stress (e.g. rapid heating, chemical reactions in composite/aggregate materials) or from external stresses. Only shock effects appear to have the characteristics to result in a significant airborne release from metals.

4.2.2.1 Shock Effects

Steindler and Seefeldt (1980) provide an empirical correlation to experimental data on the fragmentation of metals and aqueous solution by detonations [energy releases in microseconds with brisance (shattering effect)] (Ayer, et al. May 1988). The experiments covered the work performed for TNT related to the mass ratios (MRs, ratio of mass of inert to TNT equivalent) of 1 to 10. The experiments were conducted with the condensed phase explosive embedded or contiguous to the material affected. Estimates of the ARF and size distribution for various mass ratios up to 1000 are provided in Appendix C of Ayer, et al. (May 1988) for a GSD of 8. The GSD is much greater than normally assumed (GSD 2) and provides greater fractions in the larger size ranges (a nonconservative assumption for the assessment of radiological impacts). Due to the rapid change in size distribution, the maximum mass of inert material airborne in the respirable fraction is for an MR of 1. Therefore, a bounding ARF/RF of inert material equal in mass to the TNT Equivalent for the detonation is assumed. [Note: The explosive dispersal of molten Pu and U was covered in previous subsections.]

4.2.2.2 Blast Effects

No experimentally measured values for ARF and RF are available. No significant airborne release is postulated. Potential releases for loose surface contamination on metal are covered in Chapter 5.

4.2.2.3 Venting of Pressurized Gases Over Metals

No experimentally measured values for ARF and RF are available. No significant airborne release is postulated. Potential releases for loose surface contamination on metal are covered in Chapter 5.

4.2.3 Free-Fall Spill and Impaction Stress

No experimentally measured values for ARF and RF are available. No significant airborne release is postulated. Potential releases for loose surface contamination on metal are covered in Chapter 5.

Airborne release from free-fall of molten metal is covered under Thermal Stresses in subsections 4.2.1.1.4 and 4.2.1.2.2.

4.2.4 Aerodynamic Entrainment and Resuspension

Aerodynamic entrainment from a coherent, bulk solid would be limited to the surface dust unless the solid is eroded by the airflow. The phenomenon would be similar to suspension of a powder from a hard, unyielding surface. For large pieces that project beyond the boundary layer, entrainment due to "wake effect" may be a significant process. The only experimental data for the resuspension/aerodynamic entrainment from a coherent monolith of material that have been reported are found in Stewart (1963). It is assumed that the airborne release of Pu during oxidation at room temperature is the aerodynamic entrainment (resuspension) of the corrosion products (oxides) from the metal surface and that the bounding values measured for that configuration would bound the aerodynamic entrainment from the metal (see subsection 4.2.1.1.1). It would be conservative in that it is assumed that sufficient corrosion product exists for material that is normal stored under conditions to minimize corrosion and is contained. The ARR and RF values previously quoted for this configuration are assessed to be bounding:

unalloyed metal	ARR (dry air) 2E-6 µg Pu/cm^2-hr; RF 0.7
	ARR (100% RH) 7E-3 µg Pu/cm^2-hr; RF 0.7
delta-phase metal	ARR (dry air) 7E-8 µg Pu/cm^2-hr; RF 0.7
	ARR(100% RH) 6E-4 µg Pu/cm^2-hr; RF 0.7.

No experimental data for the aerodynamic entrainment of uranium metal were uncovered. The data reported for the oxidation of uranium in air (Mishima March 1985) did not show rates for temperatures less than ~ 400 ^{o}C. Since the oxidation of uranium is inhibited by the formation of an adherent oxide layer at the metal-atmosphere interface, the values reported above for plutonium are conservative for uranium.

4.3 NONMETALLIC OR COMPOSITE SOLIDS

Examples of the types of solids in nonreactor facilities other than simple metals or powders are aggregates such as concrete/cement, limestone/sandstone; glasses such as vitrified high-level waste and products of slagging pyrolosis; and spent nuclear fuel, including clad, irradiated, and compacted ceramic oxide.

4.3.1 Thermal Stress

The types of materials discussed in this section behave differently under thermal stress and are discussed individually below. All the materials are normally contained (overburden, container, equipment or cladding) when holding/incorporating radionuclides and this barrier must be breached before the material is directly exposed to the ambient environment. The

levels of thermal stress considered here are those commonly associated with industrial-type fires. More rigorous conditions that may be generated in nuclear reactors or other more severe events are not considered. Although some spent nuclear fuel within the DOE complex are composed of other materials (metals and alloys), the overall material composite is covered in this subsection because it deals with the airborne release of all radionuclides rather than the base material.

4.3.1.1 Vitrified Waste

Data from the experimentally measured airborne release from heating this form were not uncovered. The stresses that are generated by subjecting the canister to the conditions generated by an industrial-type fire do not appear to be adequate to breach the heavy-wall stainless steel canister enclosing vitrified high level (HL) waste. The impact of fire-generated conditions on the behavior of container holding other types of vitrified waste (e.g. products of slagging pyrolysis) should be considered and the consequences of any breach mechanism evaluated. If the vitrified HL waste canister is breached by large pressurization from the heating of the gases in the free-volume, a fraction (ARF 1E-1, RF 0.7) of the fines present in cooled, vitrified HL waste (estimates as high as 0.035%, WHC, 1993) on the surface of the vitrified HLW waste held in the canister could be expelled.

Monolithic borosilicate glasses incorporating high-level waste do not appear to have the potential to release any significant amount of non-volatile radionuclides. These materials would have been heated to temperatures exceeding those anticipated for most fire situation during formation and are not anticipated to undergo any chemical change under fire conditions. Borosilicate glasses are more resistant to thermal shock when cool but could be affected by very rapid cooling rates (molten glass poured into large bodies of cool water). Molten glass does not appear to possess the characteristics to generate a physical (vapor) explosion; it does not remain plastic over the required temperature range to generate fine (100 μm diameter) drops of molten glass adequate to result in the rapid heat transfer necessary for vapor explosions. Molten glass poured in pools of water would lead to vigorous boiling that could suspended solid particles carried in the liquid (see Borkowski, Bunz and Schoeck, May 1986). A possible method to estimate the fragmentation of glass by rapid cooling is to calculate the energy from cooling the glass and use that value in the crush-impact correlation outlined in section 4.3.3. Notwithstanding, a bounding ARF and RF cannot be assessed at this time due to the lack of applicable data. From the information available, however, any release under industrial-type fire conditions appears to be negligible.

4.3.1.2 Aggregate

If heated for an adequate period of time, aggregate such as cement/concrete will undergo chemical change to carbon dioxide, water (release of tritiated water incorporated in the concrete) and CaO that may carry any non-volatile radionuclides as contamination on the particles generated. Up to 30% of the material may be volatilized over a long period of time-at-temperature depending on the composition of the concrete (Chan, Ballinger and Owczarski, February 1989).

The materials loss over various temperature ranges were (Chan, Ballinger and Owczarski February 1989):

20 to 200 °C -	loss of evaporable water, equal to or greater than 5% of concrete mass;
200 to 600 °C -	loss of chemically bound water, equal to or greater than 5% of concrete mass;
>650 °C -	loss of carbon dioxide, ~ 22% of concrete mass may be degraded to a suspendible powder

Thus, if tritiated water was used in concrete formation, approximately half the tritium activity would be released from 20 to 200 °C and half the tritium activity at 200 to 600 °C. Up to 22% of the non-volatile activities would be at risk if adequate time at temperature is postulated at a temperature of 650 °C or greater. Since a vapor flux away from the surface is present, the bounding ARF and RF assessed in section 4.4.1 for heating of powders of 6E-3 and 0.01 are applicable. The MAR is the fraction of material actually present as a powder.

4.3.1.3 Encased Nuclear Material

4.3.1.3.1 Spent Nuclear Fuel. Various types of spent nuclear fuel (SNF) are found within the DOE complex - SNF from commercial nuclear power, SNF from research reactors, SNF from naval reactors, spent targets, etc. Various fuel materials may be used such as: sintered, compacted, ceramic oxide; uranium metal; uranium alloys; and uranium cermets. Various types of cladding may be used to contain the fuel during use and storage: zircaloy metal; stainless steel; and various compositions of aluminum. Some small fraction of the SNF may have cladding failures exposing the base material to the ambient environment during storage (generally water). SNF from commercial nuclear power generation that is not breached can be under high pressures. The SNF considered here is

sufficiently cooled so that it will no longer melt simply from the absence of water without a fire.

Releases from commercial power generation SNF has been extensively studied (Restrepo, 1991; Soffer, July 1993; Mendel et al., December 1988; Nourbakihsh, Khnatib-Rahbar and Davis, March 1988). Although the thermal stress in severe reactor accidents is well beyond the range normally encountered in industrial-type fires, the release fractions are certainly a "bound". Current NRC proposed releases are for PWRs [Pressurized Water Reactors, the more limiting case (Soffer July 1992)]:

	"Gap" Activity Fraction	Fuel Melting/Slumping Fraction
Noble Gases	0.05	0.95
Iodine	0.05	0.22
Cesium	0.05	0.15
Tellurium	0	0.11
Strontium	0	0.03
Barium	0	0.03
Ruthenium	0	0.007
Cerium	0	0.009
Lanthanum	0	0.002

Other radionuclides not specifically covered would be assumed to behave in the manner as their chemical analogues (e.g., Pu = lanthanum).

The conditions represented here are extreme and represent fuel melting and slumping, behavior not anticipated for the long-cooled SNF found in DOE facilities. Melting of long-cooled SNF from the temperatures typically encountered in even large industrial fires is not a major concern. The principal concern is the loss of the "gap activity" on loss of integrity of the cladding (e.g. corrosion, puncture, impact) and even that is significantly diminished by the long cooling times.

If the base material (sintered, compacted, uranium dioxide) is exposed to the ambient atmosphere, further oxidation may occur. Iwasaki et al. (December 1968) presented the results of a study to recycle off-spec fuel pellets. Production pellets were exposed to air at temperature from 400 ^{o}C for 3-hr to 500 ^{o}C, 600 ^{o}C and 700 ^{o}C for 1-hr and size distribution of the resultant powder measured. All the UO_2 was converted to higher oxides under the test conditions. The highest fraction of particles measured in the respirable size range was 0.18

at 400 °C. Under these conditions, it is assumed that all the noble gases and volatile materials (if any) are released and the fraction of non-volatile materials suspended as particulate materials due to thermal stress is as assessed in section 4.4.1. This results in an ARF and RF of 6E-3 and 0.01.

4.3.1.3.2 Metal Targets. Production targets are normally clad in aluminum metal. The cladding material is relatively soft and breaches do occur. For small breaches under water storage conditions, the base metal near the breach is converted to oxide and hydride. Eventually, substantial losses may occur.

If the cladding integrity is intact at elevated temperatures in air, the metal is not exposed to the air and oxidation does not proceed until cladding failure. Uranium metal does undergo phase changes at elevated temperatures with the attendant volume expansion but the cladding is ductile. If the uranium metal oxidizes, the noble gases and volatile radionuclides in the fraction oxidized are released as well as a fraction of the base material containing the non-volatile radionuclides as defined in subsection 4.2.1.2.1.

4.3.1.3.3 Metal Alloy and Cermet Targets. The experimental effort to define the release of radionuclides during the heating of irradiated uranium-aluminum alloys and U_3O_8 particulate dispersed in aluminum are discussed in Woodley (June 1986, March 1987), Taleyarkhan (January-March 1992) and Ellison et al. (1994).

The metal alloys used for targets melts at a relatively low temperature (~ 650 °C) and is molten at cladding failure a few tens of degrees C higher (Ellison et al. 1994). At high heat input rates, material changes proceed in three phases:

1. 100 °C below the melting temperature of cladding - blistering of the target due to the gases incorporated during manufacturing and volume expansion during heating;

2. at slightly higher temperature - cladding cracking and molten fuel flows through gaps in the cladding; and

3. for high-burnup metal - molten foam is extruded.

Taleyarkhan (January-March 1992) tabulated experimental release results from various sources as a function of time and temperature. The lesser temperatures in this tabulation (800 °C and 900 °C) are more in line with the maximum temperatures anticipated under industrial fire conditions and the data for the higher temperatures (1000 °C and 1100 °C) are not included. Under the worst response, close to 100% of the noble gases are released

(Figure 1, Taleyarkhan January-March 1992 for U-Al alloy, 62% burnup). Initial release of noble gases was associated with blistering of the fuel at ~ 600 °C. For the other elements listed, the measured releases are:

	2-min collection time	60-min collection time
cesium	0.06	0.092
iodine	0.79	0.85
tellurium	0.00	0.007

These releases are for a rapid temperature rise associated with reactor events and would not be representative of most industrial fire scenarios. The release fractions listed would represent losses after fuel cladding failure (>660 °C). No data was presented for the other categories of radionuclides (e.g. other nonvolatiles) but their release must be bounded by the release of tellurium (7E-3).

4.3.2 Explosive Stress: Shock, Blast, and Venting

4.3.2.1 Shock Effects

The response of materials that undergo brittle fracture such as aggregates and glass may not be adequately described by the Steindler and Seefeldt empirical correlation based on materials that undergo plastic deformation such as metals and aqueous solutions. Although the elastic response of materials can play an important part for the instantaneous stress generated by detonations, the presence of solids of varying strengths (e.g. concrete) would indicate some subdivision that is a function of the initial particle size of the solids. However, the use made of the Steindler-Seefelt correlation in this handbook is considered sufficiently conservative that it can be accepted as a bound for aggregate materials. Accordingly, for detonations in or contiguous to aggregate materials, a respirable release of the mass of inert material equal to the calculated TNT equivalent is assessed to be bounding.

4.3.2.2 Blast Effects

For the pressure impulses generated by explosive events that may entrain and hurl aggregate materials, the crush-impact correlation presented in section 4.3.3 may be used to characterize the response provided an impact velocity can be estimated. No experimentally measured values for ARF and RF are available. If aggregate materials are not hurled at considerable velocity, no significant airborne release is postulated. The potential releases for loose surface contamination on the solid are covered in Chapter 5.

4.3.2.3 Venting of Pressurized Gases Over Solids

No significant airborne release is postulated. The potential releases for loose surface contamination on the solid are covered in Chapter 5.

4.3.3 Free-Fall Spill and Impaction Stress

Brittle materials (e.g. glass, aggregate such as mechanically-compacted UO_2, concrete, limestone) can be fragmented when impacted or crushed. Jardine et al. (1982) performed experiments to measure the fraction and size distribution generated by the impact of various materials resting on a unyielding surface. Figure 4-12 reproduced from the reference documents illustrates that the size distribution of UO_2 pellets to an impact energy density of 1.2 J/cm^3 is linear. Note that both the sieve and Coulter Counter data shown are physical diameters and must be corrected by the square of the material density (10.96 g/cm^3). Thus the average grain size shown corresponds to a particle ~ 29 µm AED. The degree of fragmentation and the size distribution are a function of the material, the strength/age of the material, and the energy input per volume (Mecham et al., October 1981). The fraction in the size range 10 µm AED and less is relatively uniform as shown in Figure 4-13 reproduced from the reference document and was empirically correlated with the energy input (J/cu-cm) in subsection 5.1.3, Appendix F in Vol. 4 of SAND (September 1987) as:

$$ARF \times RF = (A)(P)(g)(h) \qquad (4\text{-}1)$$

where: ARF X RF = (Airborne Release Fraction)(Respirable Fraction)
A = empirical correlation, 2E-11 cm^3 per g-cm^2/s^2
P = specimen density, g/cm^3
g = gravitational acceleration, 980 cm/s^2 at sea level
h = fall height, cm.

Use of this correlation to estimate the ARF x RF value is considered very conservative, and may be excessively so if large debris from substantial heights is considered.

4.3.4 Aerodynamic Entrainment and Resuspension

No significant airborne release is postulated. Potential releases for loose surface contamination on metal are covered in Chapter 5.

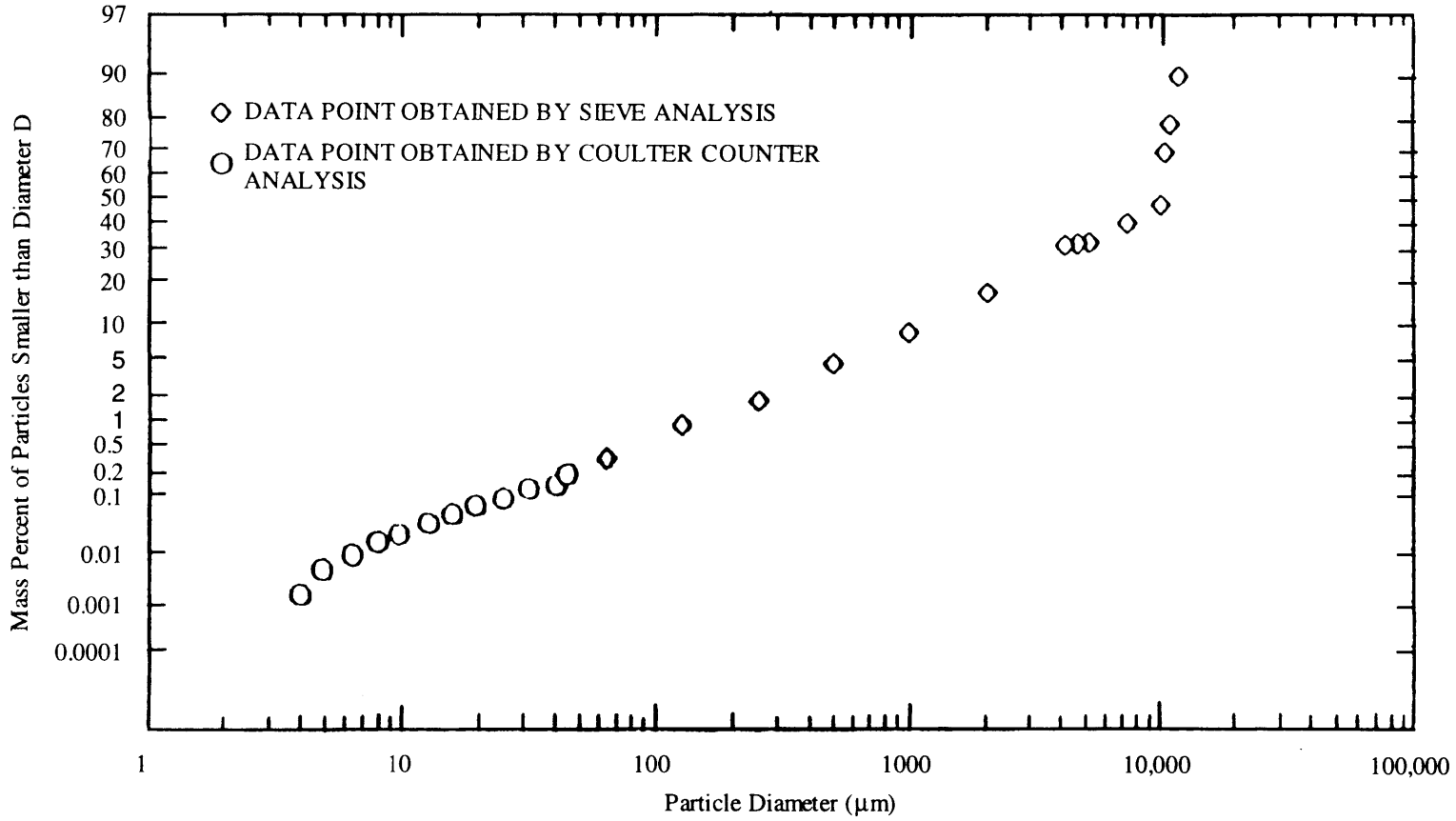

Figure 4-12. Particle Size Distribution Resulting for UO_2-2 Pellet Impact Test Data for UO_2 Specimen #2 Including Mean Grain Size of Original Crystalline UO_2 Particles. (Size Distribution 3 Pellets 13.7-mm diameter x 13.6-mm long; drop-weight 1.2 J.cm^3.)
(Jardine, et. al. 1982)

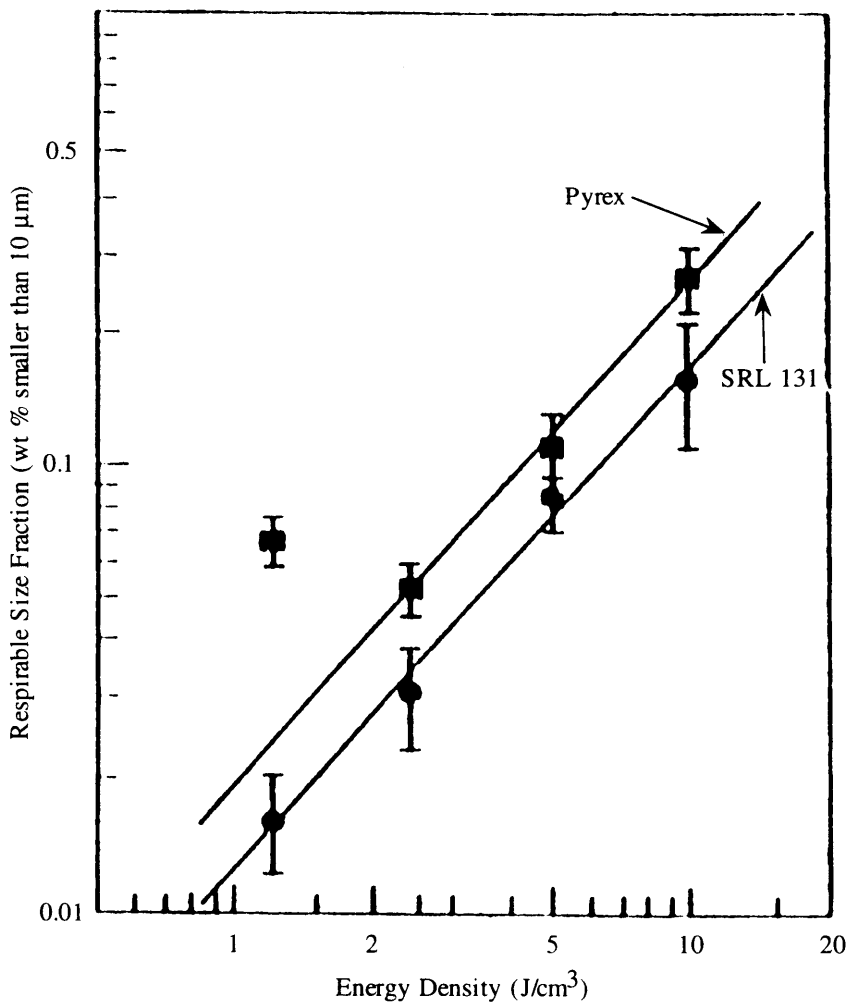

Figure 4-13. Variation of Respirable Size for Impact Tests of Pyrex and SRL-131 Glass Specimens as a Function of Impact Test Energy Density (Jardine, et al. 1982)

4.4 POWDERS

For low-energy stresses, powders do not tend to significantly fragment. For high-energy stresses considered in this document, the size fraction of powders of concern have relaxation times on the order of fractions of a millisecond, and thus do not tend to present themselves for fragmentation. Therefore, the amount of respirable material assumed airborne using this handbook should not exceed the amount of respirable material originally present in the source MAR, if that value is known. If the value is not known, and the values assessed to be bounding specifically use as the RF the fraction of original MAR less than 10 μm AED, an RF of 0.1 can be assumed based on Ayer, et al. (May 1988).

4.4.1 Thermal Stress

Experiments are reported by Mishima, Schwendiman and Radasch (July 1968, November 1968) covering the airborne suspension of particulate materials during the heating of various compounds. Two types of situations are covered - the suspension of non-reactive powders during heating in a flowing airstream and the suspension during oxidation of reactive compounds in a flowing airstream.

Plutonium fluoride, oxalate, and air-dried oxalate powders withdrawn from production, air-dried Pu nitrate from the low temperature drying of concentrated Pu nitrate solutions, or the 15 to ~ 150 μm AED fraction from the air oxidation of Pu metal were placed in the shallow depression of a stainless steel planchet on top of a graphite core used for induction heating. A thermocouple in a 3.2-mm diameter well drilled into the side of the planchet measured the temperature during the experiment. A 75-mm quartz bell that formed the bottom of a 42.2-mm i.d. quartz chimney surrounded the planchet. Air at a pre-determined flow was drawn up and around the planchet, through the apparatus, and the entrained material was collected on a glass fiber filter sealing the upper end of the chimney. At the flows used, the nominal velocities through the chimney were adequate to carry particles from up to 17 μm in diameter at 0.1 m/s to ~ 300 μm in diameter at a nominal velocity of 1.0 m/s. A 0.076-mm thick mild steel liner covering the interior surface of the chimney was used to collect any material lost to the wall during heating but analysis indicated no significant loss in any experiment. Results from these analyses were not reported due to the uncertainty in results created by the difficulty in the analytical separation of the high iron content from lining dissolution solutions. The experimental apparatus is shown in Figures A.27a and A.27b in Appendix A. The measured suspension rates during the heating of the plutonium solid compounds are reproduced in Tables A.33, A.34, A.35, A.36 and A.37 also in Appendix A.

All sampling periods were one hour for all tests and, therefore, the ARFs are the same as the suspension rates converted to fraction of source. The measured ARFs are the fraction of the

initial activity collected on the glass fiber filter during the experiment. In some experiments, portions of the material passing through the chimney were collected on a membrane filter and sized by optical microscopy using a graticule to determine the size distribution of the airborne materials. Particles were grouped into seven categories - <5, 5-8, 8-12.5, 12.5-20, 20-32, 32-50 and >50 µm equivalent spheres. The size quoted in the text must be multiplied by the square root of the density of Pu oxide (11.46 g/cm^3) to approximate the AED. These measurements are the basis for the RFs quoted in the tables. In the case of compounds that were oxidized, the time required to convert all the powder to oxide is not known. This introduces an additional source of uncertainty into the use of these measurements for ARFs during the heating and oxidation of the powders. In most cases, microscopic examination of the residual materials after the heating indicated that for the conditions under which the higher releases were measured (higher temperatures and air velocities), the oxidation was relatively complete. Furthermore, the airborne materials were entrained in an induced flow that probably exceeds that anticipated for convective flow and bounds the entrainment for convective flow. The pertinent data from these tables for the two situations are listed in Tables 4-10 and 4-11.

4.4.1.1 Chemically Nonreactive Compounds

The measured ARFs for heating nonreactive PuO_2 particles are tabulated in Table 4-10.

Table 4-10. Airborne Release from Nonreactive Powder During Heating in Flowing Air (Table V - Mishima, Schwendiman and Radasch, July 1968)

Temperature, °C	Airflow, m/s	ARF
Ambient	0.1	6.1E-6
	1.17	5.6E-3
800-900	0.1	5.3E-6
	1.17	2.5E-4

The source material for these experiments was oxide nominally in the size range of 15 to 150 µm AED (the upper value is given as 44 µm but is the fraction passing through a 325 mesh screen and is LLD rather than AED for the lower value), although some respirable particles may have been present. The two values at each separate air velocity appear to be relatively consistent with the ARF for the higher temperature lower than that at ambient temperature. Since the lower air velocity (0.1 m/s) is calculated to carry particles as large as 17 µm that is barely above the lower size of the powder used (although it was noted during the oxidation experiments that the oxide formed was friable), the low ARF value would be anticipated. Particles as large as 300 µm could be carried by the higher velocity (1.17 m/s) and the ARF

values here probably represent the material that could be entrained. It would appear that for nonreactive powders, temperature has little effect on entrainment. Therefore, a value, based on entrainment at high velocities compared to convective flow, of 6E-3 is assessed to be bounding for the material.

The size distribution of the airborne material was not measured during these experiments. The entrainment appears to be entirely due to the airflow with temperature playing a minor or negligible role. In the other experimental studies, special efforts were required to obtain initial source powders with RFs in the range of 0.01 to 0.1. Even so, powders at rest are difficult to deagglomerate (see Figure A.41). The RF for reactive powders discussed below generated very small RFs (<0.00001). Since entrainment for nonreactive powders depends solely on airflow and the same airflow was applied in the experiments with reactive powders, there is no compelling reason for the RFs to be orders-of-magnitude apart. The oxides formed from these reactive compounds can be very fine under the proper circumstances although, under the experimental conditions, large amounts of fine oxide were not generated. Under fuel cycle facility accident conditions, much of the oxides present would be from the oxidation of reactive compounds and metal that would have characteristics similar to those generated by heating reactive compounds used in these experiments. On these bases, an RF value of 0.01 was selected as a reasonable, conservative value consistent with other measured RFs discussed below.

4.4.1.2 Chemically Reactive Compounds

The measured ARFs during heating of reactive plutonium compounds in flowing air are listed in Table 4-11. The median and bounding ARFs for the four reactive compounds tested are:

	Median	Bounding
partially oxidized Pu oxalate	4.4E-3	8.8E-3
Pu oxalate	1.0E-4	9.5E-3
Pu fluoride	<8.0E-5	7.0E-4
air dried Pu nitrate	1.5E-4	1.5E-3

The ARFs for reactive plutonium compounds appear to fall into 2 groups. The ARFs measured for the two oxalate forms have maximum values near 1E-2. The value for nitrate is much lower but the median value for this compound is similar to the value for oxalate. Due to the uncertainty of the completeness of the oxidation, it is assumed that the nitrates behave as oxalates and an ARF of 1E-2 is assessed to be bounding for these compounds.

Table 4-11. ARFs and RFs During the Heating of Reactive Compounds in Flowing Air (Tables VI, VIII & X - Mishima, Schwendiman and Radasch, July 1968 and Table IV - Mishima, Schwendiman and Radasch, November 1968)

Air Velocity, cm/s	Temperature, °C	ARF[1]
Partially Oxidized Plutonium Oxalate		
10	ambient	5.6E-4
	1000	2.7E-3
50	ambient	<8.0E-5
	1000	6.2E-3
100	ambient	6.6E-3
	400	6.5E-4
	700	7.8E-3[2]
	1000	8.8E-3
Plutonium Oxalate		
10	ambient	<8.03-5
		<8.0E-5
	700	9.0E-5
	1000	8.0E-5
50	ambient	<8.0E-5
		1.4E-4
	700	<8.0E-5
	1000	1.2E-4
100	ambient	1.0E-4
		4.0E-4
		9.8E-4
		5.8E-3
	400	4.9E-3
	700	9.5E-3
	1000	3.3E-3
Plutonium Fluoride		
10	ambient	<8.0E-5
	1000	<8.0E-5
50	ambient	<8.0E-5
	1000	<8.0E-5
100	ambient	<8.0E-5
		<8.0E-5[2]
	400	1.1E-4
	700	7.0E-4[2]
	1000	7.0E-4[2]
		2.0E-4

Table 4-11. ARFs and RFs During the Heating of Reactive Compounds in Flowing Air (Tables VI, VIII & X - Mishima, Schwendiman and Radasch, July 1968 and Table IV - Mishima, Schwendiman and Radasch, November 1968)

Air Velocity, cm/s	Temperature, °C	ARF[1]
Air-Dried Plutonium Nitrate		
10	1000	6.2E-5
50	400	1.9E-5
	700	2.3E-4
	1000	1.5E-3[2]
100	400	1.7E-4
	700	1.3E-4
	1000	1.9E-4
		1.3E-4

[1] Sum of material collected on foil chimney liner and filter sealing chimney.

[2] Size distributions determined by optical microscopy indicates a RF <1E-5 to <1E-8. May indicate that oxides formed at high temperatures sinter together during oxidation forming aggregates that are larger in size.

The maximum and median ARFs for fluoride appear to be an order of magnitude less with less variability for individual measurements. Therefore, a bounding ARF value is assessed to be 1E-3 for this compound.

The upsweep air velocities used during the tests appears to be adequate to entrain all significant particle sizes as shown in Figure 4-14 reproduced from Mishima, Schwendiman and Radasch (November 1968). The size distributions of the airborne materials during the heating of some compounds are shown in Figures A.28, A.29a, A.29b, A.29c and A.30 in Appendix A. The sizes designated on the graphs are equivalent spheres (the linear diameter of the particle) and, since the theoretical density of PuO_2 is 11.46 g/cm^3, the sizes must be multiplied by 3.4 (the square root of the density) to arrive at the approximate diameters in AED. The RF estimated from the size distribution plots indicate a value ~ 1E-5 to 1E-8 associated with ARFs ranging from <7E-4 to 8E-3. Although the RFs measured appear relatively consistent associated with ARFs spanning the range of ARFs found for all materials, the values are very small and a RF of 1E-3 is assessed to be bounding for all these materials under these conditions to provide a high degree of conservatism.

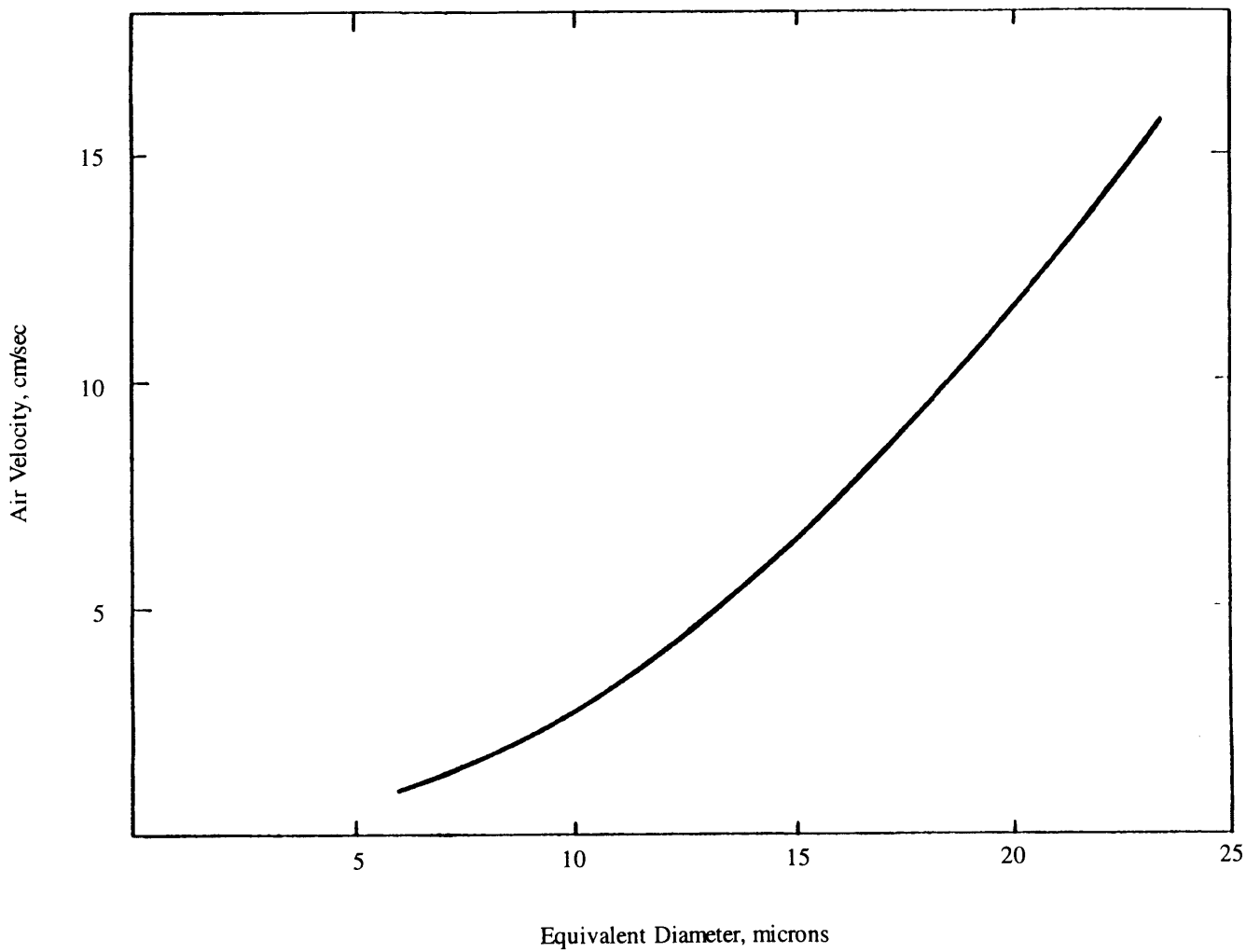

Figure 4-14. Equivalent Diameter PuO_2 Particle Lifted by Various Air Velocities
(Figure 9 - Mishima, Schwendiman and Radash November 1968)

The bounding ARFs and RFs for the response of the three categories of powders established to thermal stress are:

nonreactive compound	6E-3/1E-2
reactive compounds (except fluorides)	1E-2/1E-3
plutonium fluoride	1E-3/1E-3

4.4.2 Explosive Stress: Shock, Blast, and Venting

The effects of shock upon powders of interest in the nuclear industry (dry ceramic oxides, chemically reactive compounds) are not well defined. Blast effects from both detonations and deflagrations are assumed to result in the entrainment of powders without substantial subdivision of the finer fractions (the relaxation time of a particle 10 µm in diameter is 3E-4 second and the particle would most probably be entrained rather than fragmented) that are the primary concern for inhalation. Fragmentation of the coarser fraction with adequate momentum upon impact or chemical reactions for reactive compounds are possible. The primary entrainment mechanism is assumed to be the accelerated gas velocity resulting from the blast effects; that is, the suspension of powders by the impact of air at velocities greater than those normally associated with aerodynamic entrainment under non-accident conditions (e.g., suspension by air velocities used for ventilation and exhaust and by ambient outdoor winds). Two types of aerodynamic entrainment of powders are found: aerodynamic entrainment from homogenous beds (beds of powder greater than two particle diameters deep) and aerodynamic entrainment of sparse particles contamination from a heterogeneous surface (i.e., a hard, unyielding surface). The former is covered here and in section 4.4.4 and the latter is discussed in section 5.3.4.

4.4.2.1 Shock Effects

Gerrard (1963) reported the detection of a velocity component towards the surface from detonation that appeared to pass through powder on the surface and was reflected by the surface. If such is the case, it would be anticipated that powder could be suspended by the reflected wave. The effects of explosion on battlefield dust generation have been reported (Long, Mason and Durst, September 1984; Strange and Rooke, November 1988). The latter study was an extensive survey covering some 550 tests involving both uncased (bare) charges and munitions on or near the surface with TNT equivalent up to 1×10^6 kg but primarily focused on methods to estimate crater characteristics (Strange and Rooke, November 1988). Estimates of the mass of soil lofted (dislodged) from the crater averaged 150 kg/(kg TNT equivalent)$^{0.84}$ and are reasonably consistent with the amount of soil dislodged in Long, Mason and Durst (September 1984).

Long, Mason and Durst (September 1984) conducted a series of tests using spherical charges of C-4 explosive on the soil surface with TNT equivalent ranging from 3.4 kg to 11.3 kg. The tests were performed at two sites with sandy clay and sandy clay-sandy soil. Moisture content ranged for one site from 6.7% to 26.7% (tests performed after a light rain) and from 7.2% to 14.9% at the second site. The soil size distributions ranged to 7.5 mm and 1.2 mm for the sites (sieve analysis) but were not designated by site. The bulk density of the soils ranged 1.23 to 1.94. If a common theoretic density of 3 g/cm^3 is assumed for the soil, the fraction <10 µm AED would range from 0.24 to 0.26 and the fraction <100 micrometer AED would range from 0.42 to 0.50. High volume samplers at various heights were used to collect airborne dust samples in one series and high volume samplers at various heights plus tethered balloons were used for the second series. Sampling distances ranged from 19 m to 75 m from the Point-of-Burst. The mass of soil dislodged (based on the apparent crater volume) ranged from 65 to 232 TNT equivalent in kg (from 65 kg to 235 kg of soil was dislodged for each kg of TNT equivalent of the explosive).

The ARF values (based on the mass of soil in the clouds generated, estimated to range from 1.6 kg to 7.1 kg) were estimated to range from 0.184 to 0.84 kg TNT equivalent. The ARFs estimated for the explosions occurring in the sandy clay (moisture 6.7% to 26.7%) were estimated to range from 0.184 to 0.568. The ARFs for the explosions occurring in the sandy clay-sandy soil (moisture 7.2% to 14.9%) ranged from 0.544 to 0.84. The principal parameters influencing dust generation were: in-situ soil particle size distribution; moisture content of the soil; chemical composition of the soil; the presence/absence of vegetation; and meteorological conditions. Based on this limited set of data, it appears that some parameter or combination of parameters is affecting the ARFs but the specific parameter or combination of parameters cannot be identified. Based on the difference between the mass of soil dislodged and carried downwind and the size distribution of the soil, the soil is not significantly deagglomerated. If the soil were completely deagglomerated, from 32 to 115 TNT equivalent (assuming 50% of the soil is in the <100 µm AED size fraction) would be anticipated downwind. Many of the powders of concern in the DOE complex are dry, ceramic, metallic oxides that would more readily agglomerate than soils. A further consideration would be the chemical reactivity of the powder compound and the characteristics of the products of such a reaction.

For shock waves in open air where blast effects will quickly dissipate, an ARF of 0.8 x TNT equivalent (in kg) is assessed to be a bounding value and, based on the soil composition and the fact that the soil does not appear to have been significantly deagglomerated, an RF of 0.25 is assessed to be bounding. The ARF x RF of 0.2 x TNT equivalent is not dissimilar to the ARF of 1 x TNT equivalent for the respirable size fraction based on a conservative interpretation of the Steindler and Seefeldt (1980) correlation. That correlation was not developed for powders, and the data reported here is considered a more

appropriate source for evaluating powders. The lesser values obtained from this data are considered to confirm that the bounding ARF x RF values assigned for liquids and solids, based on an assumed GSD of 8, are very conservative.

4.4.2.2 Blast Effects

The impact of gas flow upon powder deposited upon a surface is dependent upon the characteristics of the gas flow, powder and surface. The experiments performed by Royster and Fish (1967) illustrates the importance of the angle the air flow impacts the surface on the efficiency of suspension. Experiments were performed where air was drawn into an apparatus at a 30° angle and directed upon sparse populations of particles freshly deposited on various surfaces. The apparatus is shown in Figure A.31 in Appendix A. The material entrained was collected on a filter sealing the inlet to the air blower. The fraction of activity removed by the apparatus compared to other estimation techniques as shown in Table A.38 in Appendix A. This illustrates the fact that, under ordinary circumstances of parallel flow to surface, aerodynamic forces are not very effective for removal of particles deposited upon surfaces. Figure 4-15 reproduced from the referenced article shows the effect of velocity impacting at an angle to surface upon the fraction of 5 and 0.5 μm diameter particles entrained from a stainless steel surface. Removal is relatively complete at fairly low velocities impacting the surface.

Entrainment from blast effects is divided into two categories:

1. Unshielded blast effects from detonations and large volume, confined deflagrations. The dominant effect in either case is accelerated air impacting powder and supporting surfaces from many angles.

2. Shielded blast effects from detonations and large volume, confined deflagrations. The dominant effect in either case is accelerated air flow parallel to powder and supporting surfaces.

4.4.2.2.1 Unshielded Blast Effects From Detonations and Large Volume, Confined Deflagrations. Deflagration of a system filled with a flammable mixture may accelerate to a detonation if the system configuration induces high turbulence during the burning. Typically, this can occur in piping lines sized incorrectly with regard to National Fire Protection Association Standards, inadequate use of flame arrestors in piping systems, or confinement of the flammable mixture. In a study of flammable gas detonations in plutonium storage facilities (Fry, 1991), it was concluded that an actual detonation, as opposed to a deflagration, was highly unlikely. That study reiterated common assumptions that effective confinement of vapor cloud explosions requires a flammable gas volume at least half the size

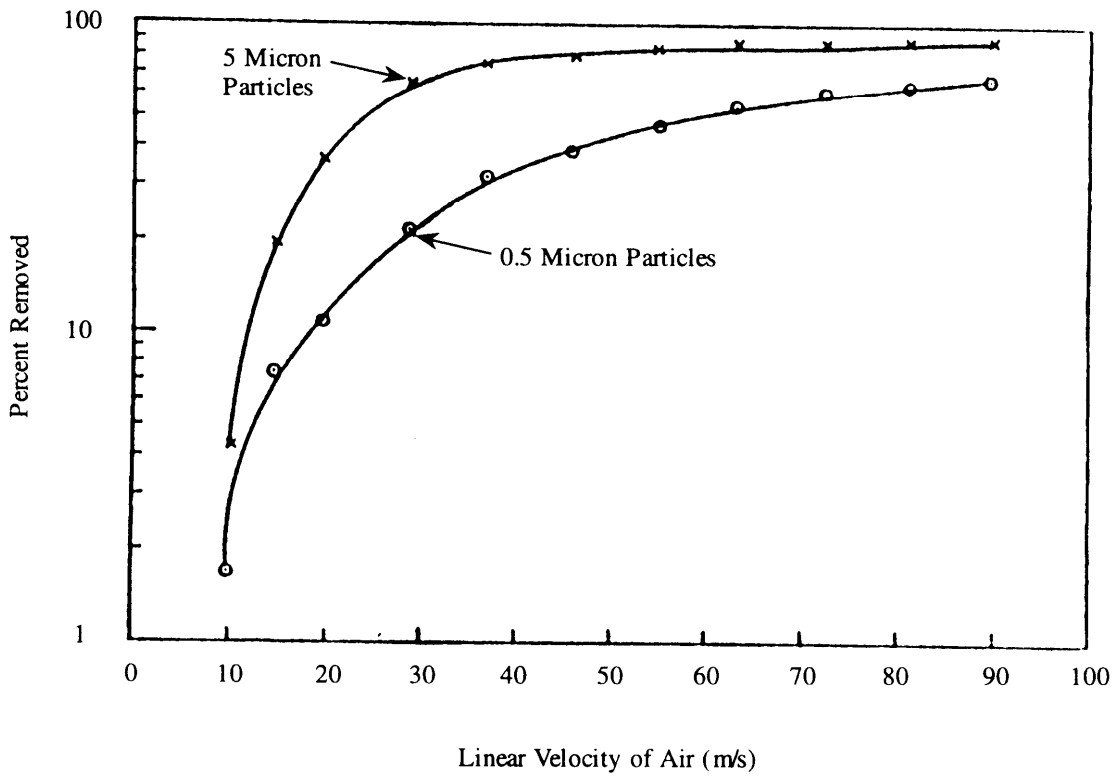

Figure 4-15. Removal of ThO_2 from Stainless Steel
(Figure 7 - Royster and Fish 1967)

of the overall room, and more likely the same size as the overall room. To insure reasonable conservatism, this document is assuming significant enhancement of deflagration wind effects if (1) the flammable gas volume is 25% of the containing volume, and (2) the confinement is sufficiently strong to allow pressures > 0.17 MPa_g to be sustained in the primary confinement (e.g., glovebox, vessel). The latter condition is not met if the primary confinement fails at pressures less than or equal to 0.17 MPa_g.

The two phenomena being equated are: (1) a detonation occurring within some confining structure and in the vicinity of unshielded material; and (2) deflagration of a flammable gas mixture with a reactive component cloud volume exceeding one-fourth the volume of a strong enclosure (rupture pressure > 0.17 MPa_g) over powder lying on hard, unyielding surfaces. In either case, the gas currents generated act upon the powder as gas flow directed at the surface from various angles of attack (an efficient mechanism to suspend particles from surfaces). Braaten, Shaw and Paw U (1986) did not observe any substantial entrainment during the velocity increase to 20 m/s in a wind tunnel. John, Fritter and Winklmayr (1991) observed some suspension of deposited powder during the increase to 40 m/s in a wind tunnel. Wright (1984) reported the suspension of ~ 95% of the deposited powder from the floor of a wind tunnel during the few seconds necessary to raise the velocity to 60 m/s (~ 135 mph). A few seconds is, however, a significantly longer momentum period than would typically be experienced in an explosion environment. If confinement or blowout ports fail at relatively low pressures, the interaction of the blast wave with powder will be mitigated. Events where significant interaction is considered feasible are detonations in other than relatively open-air conditions (i.e., small volume relative to magnitude of explosion) and deflagration of large volumes of flammable gas mixtures above the powder where confinement failure exceeds ~ 0.17 MPa_g. For these cases, an ARF of 1E+0 is assumed with the RF equal to the RF of the source powder. This value is directly applicable for the deflagration case, and is applicable for detonations if it yields a larger release than the ARF x RF recommended for shock effects.

4.4.2.2.2 Shielded Blast Effects From Detonations and Large Volume, Confined Deflagrations. In a survey of published literature on accident generated particulate material (Sutter May 1982), no experimental study was found that followed the release history of the aerosol or dust cloud immediately after the event. Many of the experimental studies centered around the suspension of coal dust following explosions but the models developed are for large piles of coal or ore that are not applicable here even though the materials are adhesionless powders similar to the ceramic oxide powders found in many fuel cycle facility accident situations. The amount of powder involved in accidents in fuel cycles under most circumstances is orders of magnitude less and parameters used in the analysis, such as velocity at half height, are not relevant. Two types of powders were addressed: cohesive

(6 to 80 μm diameters) and free-flowing (100 to 150 μm diameters). Different entrainment mechanisms govern the types of materials. A variety of observations on the sequences or phenomena governing entrainment are reported.

For velocities following weak or marginal explosions, Singer, Cook and Grumer (1972) reported entrainment resulting from the erratic rupture and removal of large clumps from the surface of cohesive dust ridges and their dispersal in the midstream. They attributed the entrainment to a five step process: 1) detachment of single particles from the loose material on the surface; 2) detachment of small clumps and particles; 3) partial fracturing and subsequent entrainment of large clumps; 4) ridge breakup and complete breakup of large clumps; and, 5) continued breakup and dispersal of clumps in midstream. Steps 1 through 4 required from 0.1 to 0.5 seconds to entrain 2 grams of material from cohesive dust ridges. Fresh deposits were more readily dispersed and deposits became cohesive when slightly compacted (such as after spill of material).

Singer, Harris and Grumer (1976) observed that explosions generated oscillatory flow (increased flow followed by flow reversal) in wind tunnel experiments. Entrainment appeared to be a weak function of the instantaneous air speed over the bed. Wetted or wetted-dried layers of coal and rock dust dispersed faster due to the selective lifting of relatively large briquetted fragments. Entrainment proceeded simultaneously by longitudinal regression of the leading edge of a cohesive bed and the lifting of material from the surface layer. The threshold instantaneous air velocity to entrain bulk quantities from cohesive beds ranged from 5 to 30 m/s (11 to 67 mph). If the threshold velocity is exceeded for one component of a mixed layer but not the other, the components can be entrained individually.

Chepil (1945) observed that the entrainment process begins with a rolling or sliding particle motion as drag forces exceed friction forces. Punjrath and Heldman (1937) attributed entrainment to the sudden increase in aerodynamic shear stress from the transition from laminar to turbulent flow. Einstein (1942) observed that entrainment depended on the fluctuation of the air velocity at the surface rather than of critical fluid properties (e.g., mean velocity of bulk flow). Kalinske (1947) and Graf and Acaroglu (1968) reported entrainment was due to fluctuating pressure and velocity components. Parthenaides and Passwell (1970) presented the entrainment rate equations for the erosion of cohesive beds based on the fluctuating flow components inducing instantaneous tensile stresses within the bed that exceed the weakest bond holding particles in the bed.

No theory for entrainment due to the pressure waves generated by explosion appears to be generally accepted. Various equations were uncovered that estimated the entrainment rate under these conditions but all required either experimentally derived/empirical factors or

parameters not readily determined for accident conditions. Estimates of entrainment rates still require estimates of the duration of the pressure wave/accelerated flow over the deposit to arrive at estimates for ARFs. Singer, Cook and Grumer (1972) reported entrainment rates of ~ 10 g/m^3 at 5 m/s to ~ 90 g/m^3 at 40 m/s (89 mph) for velocities at the mid-height of the deposit. Freeman (1972) reported concentrations of 700 to 1100 g/m^3 in 1 second at an explosion equivalent to 1E+8 g TNT but did not indicate if this was entrainment or due to blast effects.

In the absence of any directly applicable predictive models or release data, other related experimental values are applied. From the information quoted above, it appears the entrainment process results from the creation of surface flaws by the detachment of particles or clumps. Once created, the surface flaws allow more general lifting of the surface until the entire surface is disrupted. The process requires some period to initiate as shown by the 0.1 to 0.5 seconds necessary to entrain 2 grams from a cohesive surface (Singer, Cook and Grumer 1972).

Mishima and Schwendiman (August 1973) reported the entrainment of UO_2 powder and air-dried UNH from various surfaces at two air velocities (~ 1.1 and 8.9 m/s 1-ft above the surface) at ambient temperatures. The experimental apparatus is shown in Figure A.3 and the experimental results reproduced in Table A.3 in Appendix A. An ARF and RF of 7.6E-2 and 0.14 were measured for the higher of two values for the suspension of UO_2 powder from a stainless steel surface for a wind velocity of 8.9 m/s 1-ft above the surface (comparable to a 22 m/s under normal wind speed measurements) in a 24-hour period. The velocity of the bulk fluid at distances above the surface may not be a meaningful measurement and the velocity profile near the surface may be more relevant. In the case of explosion-generated air velocities, the duration of the peak velocity is in terms of milliseconds. Approximately 60% of the resuspended powder (4.56E-2) was made airborne during the first hour of the wind as shown in Figure 4-16 taken from the referenced document. If resuspension is assumed to be linear over the initial hour the ARF would be 7.6E-4 per minute. It is noted that recent studies indicate resuspension of sparse particle contamination is not linear, especially at the beginning and end of the suspension period and that the suspension from the bed of a cohesive material appears to be initiated at surface imperfections. However, for the flow to be parallel over the surface, the center of the explosion must be some distance from the deposited material. The entrainment due to the pressure wave over the surface will be limited as the wave will only be over the deposit for a short period of time (fractions of a second).

Fry (July 1991) specifically evaluated the effects of a flammable gas detonation outside a typical glovebox on clump powder sitting on the floor of the glovebox. The simulation used the HULL hydrocode to model detailed interactions of blast waves loading onto structures

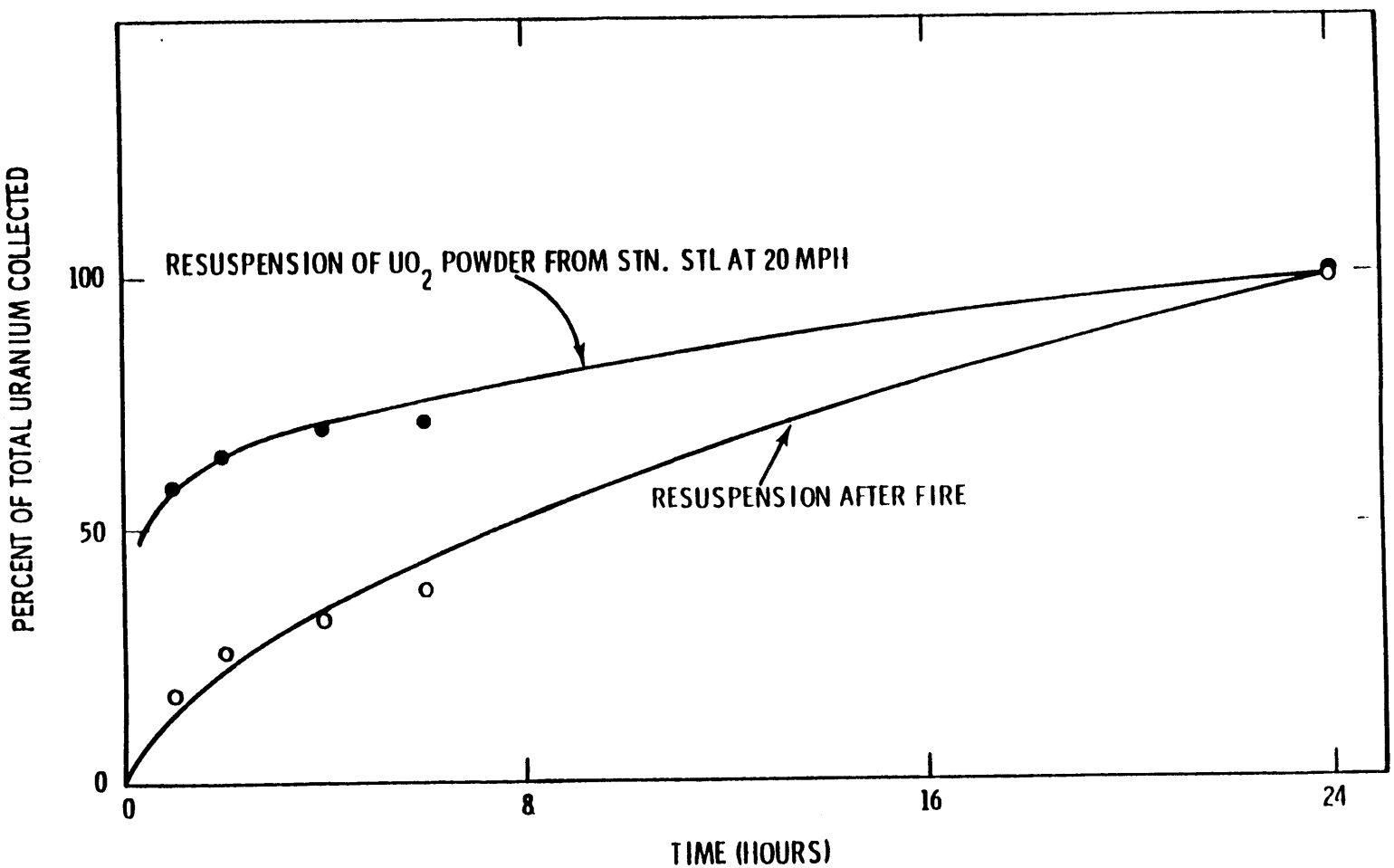

Figure 4-16. Aerodynamic Entrainment of Uranium Dioxide Powder from a Stainless Steel Surface at an Air Velocity of 20 mph
(Figure 16 - Mishima and Schwendiman August 1973)

and components. Shock and blast waves and associated TNT equivalents were estimated for 1.8 and 4 meter diameter flammable gas clouds. The point of detonation was assumed to be 0.75 m from the front of the glovebox.

The modelling indicated that even relatively weak shielding such as the lexan windows or gloves of the glovebox provided significant shielding from shock waves. This was due to the speed of the shock wave (total glovebox envelopment in 2.5 milliseconds), which would almost completely pass over the structure and initiate reflection waves in the time it took for shielding material to fail: "the shock wave moving inside the glovebox is approximately spherical in shape and much weaker than the outside shock." Peak overpressures in the glovebox ranged from ~ 8 to 28 psig at the glovebox floor and from ~ 5 to 15 psig at 0.3 m above the glovebox floor. As would be expected, the higher pressures were on the side of the glovebox facing the explosion.

The peak velocity and density of the shock and blast wave moving across the bottom of the glovebox were 300 m/sec and 0.004 g/cm^3 respectively. Kinetic energy density was computed from these values. Halverson and Mishima (1986) had developed an empirical equation for wt% of powder airborne as a function of energy density. In this calculation, powder mass was minimized (~ 30 g) to maximize energy absorbed per gram. The fraction of material driven airborne was estimated to be 5E-3. The main uncertainty associated with this calculation is the unaccounted potential for localized, high energy density regions that would be expected in a non-uniform distribution. To attempt to determine the relative severity of conditions inside the glovebox, massless tracer particles were inserted into the model to follow flow with no drag. Particle motion indicated an absence of strong shear forces or turning forces that might enhance breakup. Most particle movement was uniformly to the rear of the glovebox.

The explosion study is considered to support the basic interpretation of phenomena in studies by Mishima and Schwendiman. Based on those studies, values for ARF and RF of 5E-3 and 0.3 appear to be conservative for the suspension of a powder from a smooth, unyielding surface from the pressure impulse generated (i.e., gas flow parallel to surface) by an explosion. The release phenomena is considered to cover powders shielded from the direct impact of the blast as well. Examples of such situations include powder buried under debris, in a can/container that is uncapped by the blast, or in a glovebox with blast external to the glovebox.

4.4.2.3 Venting of Pressurized Powder

For the entrainment due to the rapid burning of a limited volume of combustible mixture (equal to an unconfined vapor explosion - cloud volume, <0.25 volume of container) over

the deposited material, ARFs and RFs may exceed the entrainment from accelerated parallel flow (i.e., shielded material) but be less than a value of 1E+0. If the expansion wave from the deflagration incident on the surface is essentially planar, gases may be pushed through the powder and be reflected from the surface resulting in suspension of the powder under pressure. In order to generate a pressure wave that will have an essentially planar impact upon the surface, it would be necessary to have an ignition source that is far from the surface, distributed parallel to the surface, or that is confined in a direction perpendicular to surface. The latter circumstance also requires that the radial distance to reach the radial constraint is less than the distance to the surface. Even with these restrictive geometry considerations, this may not be equivalent to the passage of a wave front since the wave can travel in the absence of bulk flow. Timing of the wave reflection, and its speed through the powder, are relevant to the extent the powder will pressurize. The maximum pressure in the powder would be inside the expanding wave front at a distance from the ignition point equal to that at the bottom of the powder bed (surface). The total volume of gas pushed into the bed and then expelled is probably a relevant parameter. Use of experimental data from the venting of pressurized gases through a powder bed is probably conservative since the experiments released from 2.7 to 27.2 liter of air through 100 g and 300 g of powder.

4.4.2.3.1 Venting of Pressurized Powders or Pressurized Gases Through a Powder, Pressure > 0.17 MPa_g. If the gases in and around a powder are compressed during pressurization, the gases expand rapidly during venting and result in airborne dispersal of the powder. Sutter (May 1982) reported ARF estimates for two reported accidental overpressurizations in nuclear fuel cycle facilities. An external building release of 1 mCi of 604 Ci of activity occurred after catastrophic rupture of an ion exchange column at 0.69 MPa (estimated pressure of column failure), yielding an overall estimated ARF of 2E-6 (includes building leakpath factor). An estimated 1.2 to 1.3 mCi of Pu was reported released during the depressurization at 60 psi of a slip-fit container (may also have been wrapped in multiple layers of plastic) holding 12,168 Ci as PuO_2 powder. The ARF estimated for this incident is 1E-7.

Experimental data for the airborne release of 2 powders with varying densities (TiO_2 with a material density of 4.2 g/cm^3 and UO_2 with a material density of 10.96 g/cm^3) for two masses-at-risk (100-g and 350-g) have been reported by Sutter (August 1983), and Ballinger, Sutter, and Hodgson (May 1987). Both airborne releases from the venting of pressurized powders and from venting pressurized gases through powder were measured. The apparatus for venting pressurized powders is shown in Figures A.32, A.33a, and A.33b. The arrangement to collect the airborne materials in the 10-ft diameter stainless steel tank used for confinement is the same as used for liquid releases and is shown in Figure A.4 in Appendix A.

Initially, tests for the venting of pressurized powders were performed using pressure to 6.9 MPa but significant masses of powder were impacted and adhered to the ceiling of the 10-ft tall containment vessel compromising the measurement of the fraction airborne. Although powder is probably lost by adhesion to structural features in actual accident situations involving high pressures, the effect is indeterminate and the ARFs measured by such test would not bound the ARFs from unimpeded aerosolization. Figure 4-17 reproduced from the referenced document shows the weight percent airborne as a function of pressure. Although some powder may have been lost by impaction/adherence to the ceiling in tests performed at 3.5 MPa, the effect at this and lower pressure did not appear to be significant. Either some material is loss by adhesion to the ceiling or entrainment is not a linear function of pressure and the reduction is due to some characteristic of the tests or release mechanism. All subsequent test for venting of pressurized powders or the venting of pressurized gases through powders were limited to pressures of 3.4 MPa or less.

Another factor that affected airborne release during the venting of pressurized gas through a powder was the potential increased dispersal action by the remnants of the rupture disks (thin metal foils) used to initiate the venting at the correct pressures, which would have enhanced airborne release. For the same pressures, the airborne releases measured for both venting configuration using the small mass-at-risk (100 g) were approximately twice those measured for the larger mass-at-risk (350 g) and may indicate that the depth of material may influence the release fraction. The masses used in the experiment are not representative of the powder masses normally associated with processes, which are much less, and the stress configuration is much more conservative than realistically expected. On this basis, only the ARFs and RFs from experiments using the larger powder masses are considered. The data are reproduced in Tables A.39a, A.39b, A.40a, A.40b, A.40c, and A.40d (Sutter, August 1983), and in Tables A.40e and A.40f (Ballinger, Sutter and Hodgson, May 1987) in Appendix A. The pertinent data are tabulated in Table 4-12.

The ARFs for the larger source mass only ranged from 5E-5 to 1E-1. The three greatest measured ARF values are for the venting of pressurized gas (3.4 and 1.7 MPa) through powder beds and the venting of pressurized TiO_2 at 3.4 MPa. The two values for the venting of pressurized UO_2 at 3.4 MPa are at essentially the same value. As mentioned above, the values for the venting of pressurized gas through a powder bed may be enhanced by the dispersal of powder resulting from the passage of the rupture disk remnants through the powder. The median value is 5E-2 with an average of 5E-2. The RFs ranged from 0.29 to 0.88 with a median value of 0.44 and an average of 0.47. The RFs associated with the bounding ARF values range from 0.31 to 0.72 with all but a single value at or less than 0.54. On these bases, the ARF and RF of 1E-1 and 0.7 are assessed to be bounding.

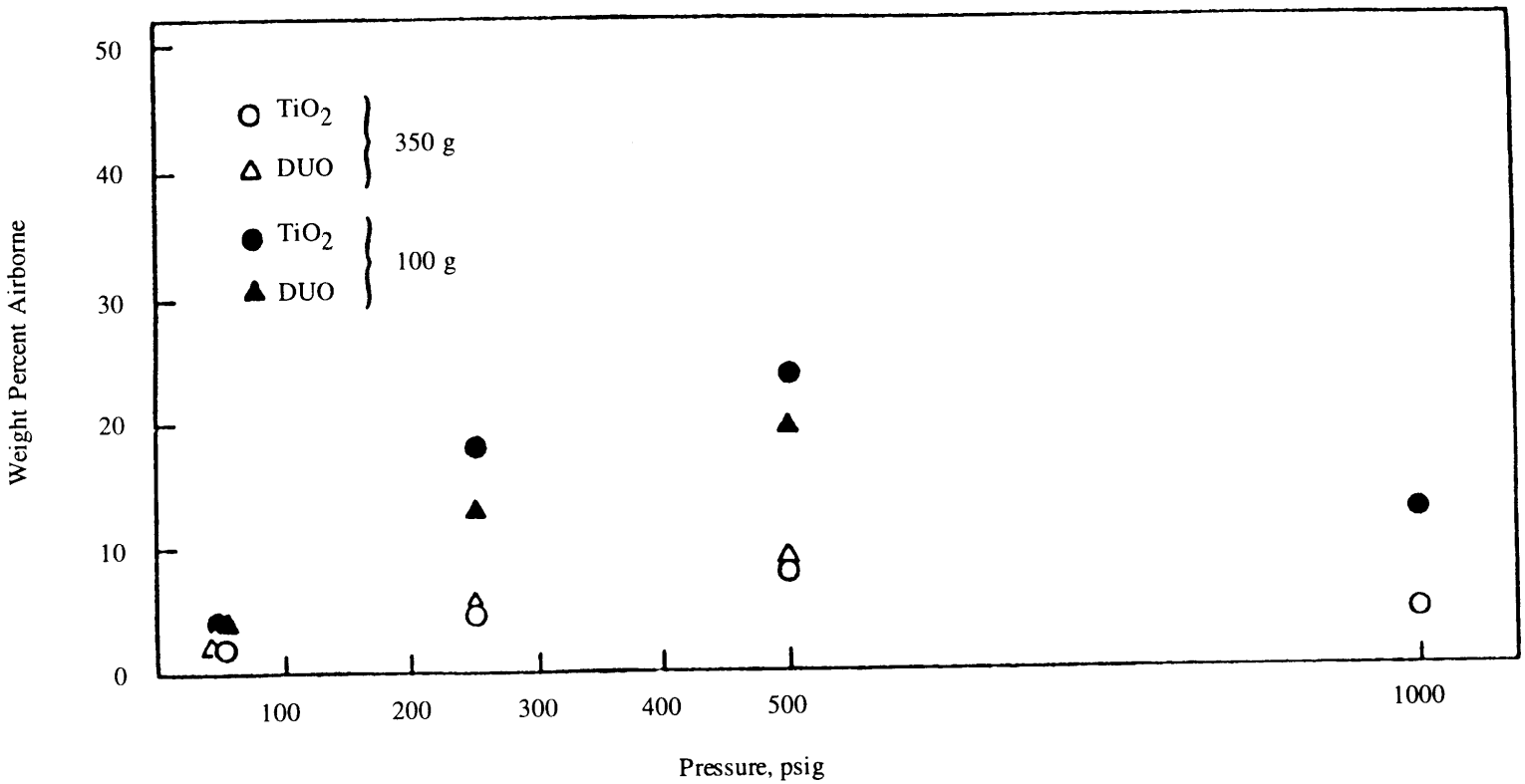

Figure 4-17. Average Weight Percent of Powder Airborne as a Function of Pressure
(Figure 7 - Sugger August 1983)

Table 4-12. ARFs and RFs from the Venting of Pressurized Powders

(Tables A.1, A.2, A.3, B.1, B.2 and B.3 - Sutter August 1983 and Tables A.5 and A.6 - Ballinger, Sutter and Hodgson May 1987)

Material	Pressure, MPa_g	ARF	RF
TiO_2	6.9	4.5E-2	0.64, Material adhered to ceiling
	3.4	6.1E-2	No RF, cascade impactor overloaded
		9.8E-2	0.40/0.44
		1.09E-1	0.54/0.40, Pressurized gas release
UO_2		9.0E-2	0.34
		9.0E-2	0.31
TiO_2	1.7	2.89E-2	No RF, cascade impactor overloaded
		7.6E-2	0.44
		1.05E-1	0.48/0.72, Pressurized gas release
UO_2		6.0E-2	0.42
		6.0E-2	0.29
TiO_2	0.69	9.4E-3	No RF, cascade impactor overloaded
	0.34	6.2E-3	No RF, cascade impactor overloaded
		3.6E-2	0.38/0.48
		4.5E-2	0.46/0.88, Pressurized gas release
UO_2		2.0E-2	0.33
		2.0E-2	0.31
	0.17	1.73E-3	0.35
	0.12	1.11E-3	0.60
	0.06	5.0E-5	0.61

4.4.2.3.2 Venting of Pressurized Powders or Pressurized Gases Through Powders, Pressure $\leq$ 0.17 MPa_g. The values for the airborne release during the venting of pressurized powders at lower pressures, 0.17 MPA_g (25 psig) or less, reported in Ballinger, Sutter and Hodgson (May 1987) are substantially lower than those covered above. With the exception of a single value (6E-3), all ARFs for the venting of pressurized powders at these pressures were 3E-3 or less. The three values listed in Table 4-12 for source masses of powder of 350 g are less than 2E-3. Since the data is very limited (only six results at 3 pressures 0.17 MPA_g and less reported with only three listed in Table 4-12), a conservative ARF of 5E-3 is assessed to be bounding for these situations. This value is commensurate

with the value for accelerated airflow parallel to surface, which is expected to be the dominant effect for low pressure venting above powder. The limiting RF for the 350 g source mass of 0.4 is considered appropriate.

4.4.3 Free-Fall Spill and Impaction Stress

The following subsections discuss powder dispersal due to (1) acceleration by gravity and impact with unyielding surfaces, and (2) impaction by falling objects of powder lying at rest.

4.4.3.1 Free-Fall Spill of Powder with Air Velocity Normal to the Direction of Fall

4.4.3.1.1 Factors that Affect Dust Generation. Plinke et al. (1991) surveyed the literature on dust generation and performed tests to ascertain the factors that may affect dust generation. Background literature attributed dust generation rates to some function of the ratio between the separation forces generated by the operation/event versus the binding forces present in the powder. Some parameters that affect the separation forces are bulk density, fall height and sample mass. Factors affecting binding forces identified were particle size distribution and moisture content. Bulk density is an indication of initial dispersion. Fall distance and sample mass along with bulk density are indices of the speed and momentum of the falling material and an index of turbulence upon impact. The amount of air that can be entrained in the particle mass increases dispersion and would enhance aerosolization. Confinement of the falling material in a manner like these experiments would increase turbulence at impact enhancing aerosolization of the powder.) Particle shape was also identified as a possible critical parameter in some materials. The dustiness of powders [the authors define "dustiness" as the ability of a material (powder or solid) to generate particles by mechanical or aerodynamic stresses normally encountered in routine use] was found to be independent of test methods. An early model indicated that:

$$L = 16.6\ (W)^{-0.75}(S_g)^{3.9}(D)^{-1.2}(M_g)^{-0.45} \qquad (4\text{-}3)$$

where:
- L = fractional mass loss, mg of dust/kg of material dropped
- W = moisture content, %
- S_g = geometric standard deviation for material size distribution
- D = material bulk density, g/cm^3
- M_g = mass median diameter for material size distribution, µm.

The experimental apparatus used is shown in Figure A.34 in Appendix A. The individual data points were not reported but the results of the tests are shown graphically in Figures A.35a, A.35b, A.35c, A.35d, and A.35e in Appendix A.

Four readily available materials in powder form were allowed to free-fall spill known distances and the material airborne was collected as a function of aerodynamic particle diameter in a cascade impactor. Two powders were nonporous and nonreactive to water, inorganic, crystalline materials (sand and limestone). One porous, reactive, inorganic material (cement) and one porous, reactive, organic material (flour) were also tested. Sand and limestone are similar to some process generated wastes (e.g., slag and crucible) and coarse fractions of process materials (e.g., heavy metal oxides). The finer cohesive heavy metal oxides are similar in characteristics to cement but a nonreactive with water. The solid salts of other heavy metal compounds (e.g., fluoride, chloride, oxalate, hydride) were not represented here.

The powder fell in the test apparatus from a funnel with an interchangeable tube through a hole in the center of the lid of the receiving hopper. The interchangeable diameter tubes (24-, 37- and 49-mm) adjusted the material flow rates. The total mass dropped ranged from 2 to 10 kg. Flow rates (ranging from 0.1 to 10 kg/s) were determined by the time required to release a known mass of material. The fall distance was measured from the bottom of the tube to the top of the pile of material. The stream of falling powder impacted a natural pile of powder formed of the same material under the same conditions. Air was entrained in the falling powder stream.

The dusty air generated was drawn into the second section (the aerosol collection section) of the apparatus by a fan that was turned off after all the powder had been released. A second fan circulates the air through the elutriation column (flow velocity designed to prevent particles >25 μm AED from being carried to the cascade impactor) and air return channel. The mass collected per impactor stage or filter was used to determine the size specific dust generation rate, G_i (fraction of dust particles generated with an aerodynamic diameter i). The summation of all fraction collected was the total dust generation rate, G. (The total dust generation rate, G, is expressed as mg airborne/mg source and is equivalent to the ARF.) The size distribution and moisture content for the source powders were determined.

At low Fs (material flow rates), the falling stream of material stops abruptly on top the pile and slides down the sides. At higher Fs, the individual materials exhibit different behavior:

- sand: the falling stream penetrates the pile and displaces material radially.

- limestone: the pile is compressed by the falling stream and forms a crater, 70- to 150-mm in diameter. The diameter of the crater increases with fall distance. The falling powder stream strikes the center of the crater and bounces back in all directions.

- cement: behavior is between the responses of the previous two materials. The penetration of the pile by the falling stream of powder is not as deep as with sand and does not compress the pile as much as limestone. The impact does not result in bouncing of the powder. Most of the material slides down the sides of the pile.

- flour: the only material tested that formed piles with a peaked top. Material slid down sides of pile without penetrating.

Dust appears to be generated by two mechanisms: impact of the falling stream creates separation forces; and, the change in flow direction of the entrained air in the impaction area due to its inability to enter in the solid powder results in radial flow that transport airborne particles away. The total dust generation rates are substantially different for the four materials tested and:

- increased as fall distance increased. Greater fall distance appears to increase the normalized entrained air, V, increasing total dust generation. The slopes of dust generation rates for sand, cement and flour are almost identical indicating similar responses to energy input.

- decreased with increased F (material flow rate). The material in the center of a falling stream of powder is less exposed to the surrounding air and the material in the center of the falling stream increases with F. As a result, less air is entrained, V, in the falling powder stream and reduces the radial flow that is a factor in the transport of suspended powder from the impact area. For sand and cement, the decrease in G may also be due to the reduced impact forces imposed on these materials by their penetration into the pile. The decrease in G for flour appears to be due to the reduction in entrained air. The increase in G with F observed for limestone is attributed to the proportionately greater increase in separation forces resulting from the formation of a crater by the material in the impact area with the reduced shock absorption from the reduced layer of powder.

- G decreased as W (moisture content) increased for all materials tested. Cement formed agglomerates with the addition of moisture altering the particle size distribution of the source material and was not tested. The rates varied substantially for the other 3 materials. For the crystalline, nonreactive-to-water materials (which also appear to be noncohesive), additions of small amounts of water increased the liquid film of the surface of the individual particles and appear to increase the capillary interparticle binding forces. For

the non-porous, reactive to water materials (flour), the effect was less pronounced due to the absorption of the water.

The coefficients derived from analysis of variance on the data from the tests performed on the various materials were compiled in the equation:

$$G_i = \text{const}(H)^A(F)^B(W)^C(\text{Frac}_i)^E[D_i \ln(D_i/25)] \quad (4\text{-}4)$$

where:
- H = fall distance, cm
- F = material flow, kg/s
- W = moisture content, %
- Frac_i = fraction of particles in i size range in source material
- Di = average diameter of particles collected on cascade impactor stages

A, B, C, D & E are coefficients calculated by analysis of variants and varied with material.

The formulae accounted for 72% to 93% of the experimental variation for the four materials and indicates that the parameters chosen are appropriate for the purpose. The correlation between measured and predicted size specific dust generation rates is shown in Figure 4-18. In all cases, for drop heights ranging from 0.25- to 1.5-m, values for G were less than 2E-4 to 3E-4.

4.4.3.1.2 Free-Fall Spill Experiments. Sutter, Johnston and Mishima (December 1981) reported on experiments performed to measure the fractional airborne release of powders (TiO_2, density 4.2 g/cm^3; depleted uranium oxide, density 10.96 g/cm^3) during free-fall spill. The size distribution of the two source powders was measured by liquid sedimentation using suspensions of ultrasonically dispersed powder in water containing a surfactant is shown in Figure 4-19 reproduced from the referenced document. Masses ranging from 25 to 1000 g were spilled from a beaker at the ceiling of a 3.05-m (10-ft) diameter by 3.05-m (10-ft) tall stainless steel vessel. Airborne powder was collected by high volume total particulate and cascade impactor samplers. Material deposited on walls of the vessel was estimated from the amount collected on aluminum foil strips on the vessel walls at various locations. The apparatus is shown in Figure A.5 in Appendix A. The pertinent data are tabulated in Table 4-13, with measured data reproduced in Tables A.41a, A.41b, A.41c and A.41d in Appendix A.

The bounding ARF and RF values for the largest spill height are 2E-3 and 0.3. The median ARF and RF values are 9E-4 and 0.4 with average values of 9E-4 and 0.5. The maximum RF measured for spill from the greater height is 0.9 associated with the smallest ARF

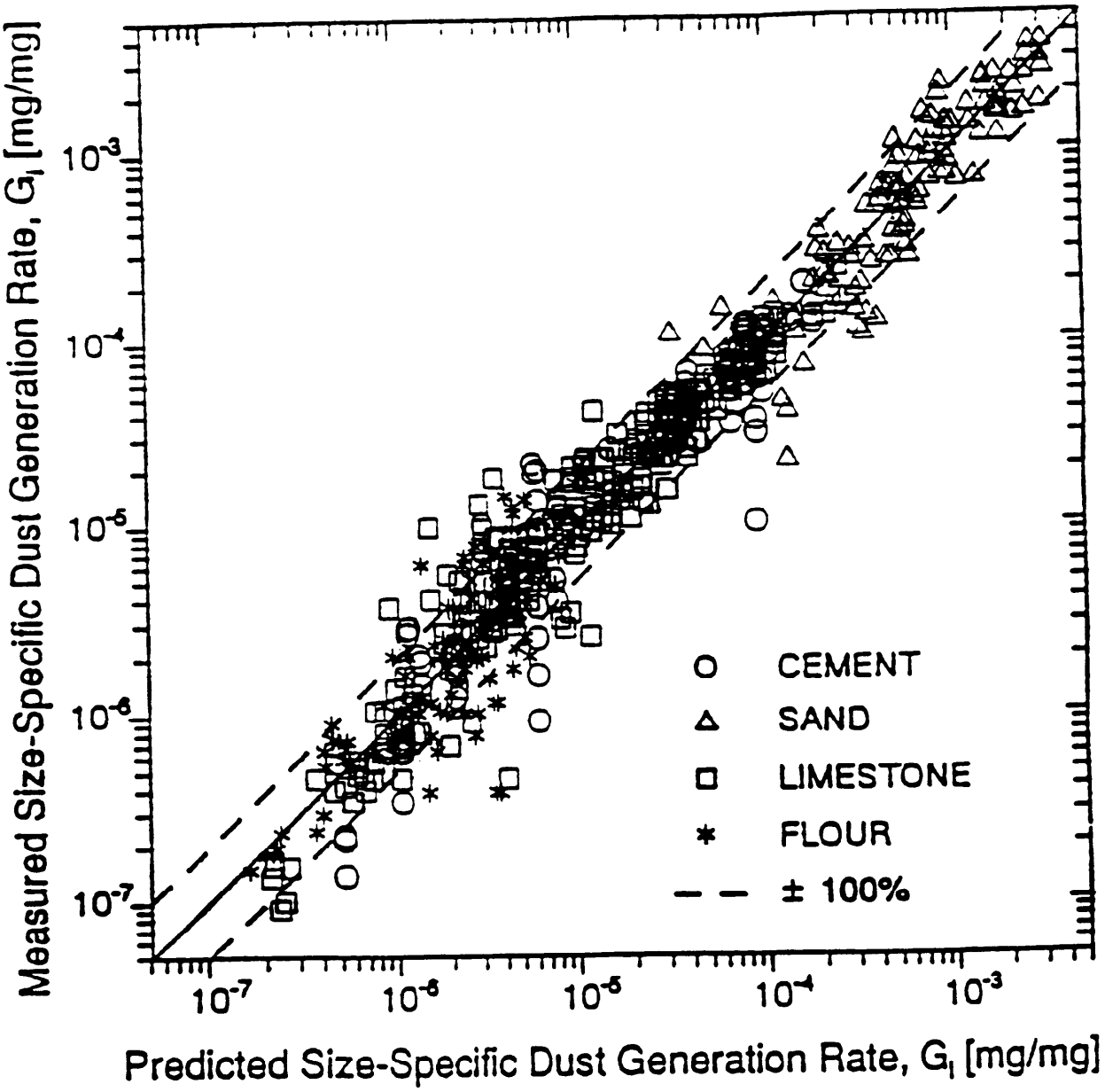

Figure 4-18. Measured Size-Specific Dust Generation Rate Versus Predicted Size-Specific Generation Rate from Equations 8-11 (Figure 7 - Plinke, et al. 1991)

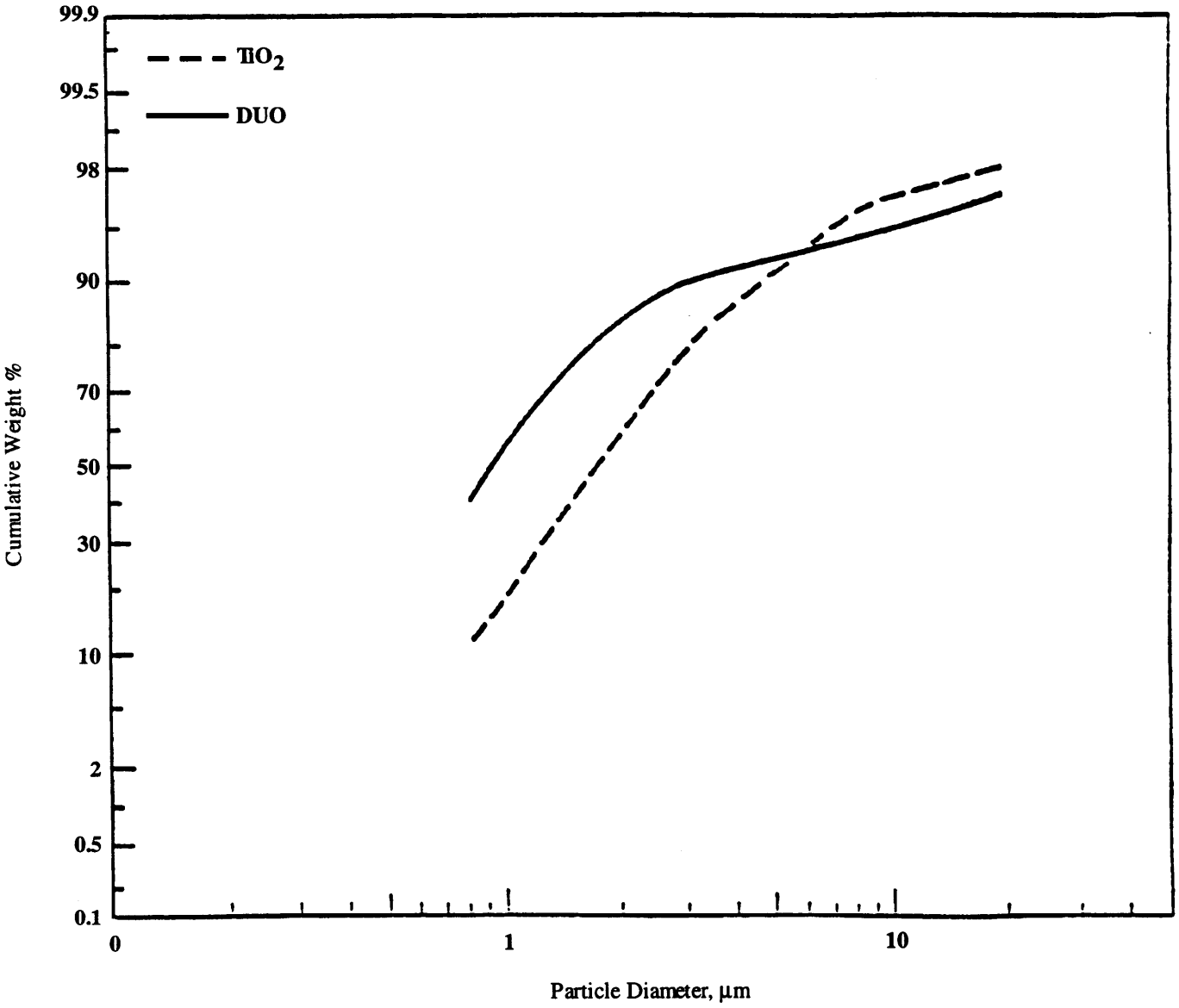

Figure 4-19. Titanium Dioxide and Uranium Dioxide Particle-Size Distribution (Figure C.1 - Sutter, Johnston and Mishima December 1981)

Table 4-13. Measured ARFs and RFs from the Free-Fall Spill of Powders (Tables A.1, A.3, B.1 and B.3 - Sutter, Johnston and Mishima December 1981)

Fall Height, m	Material	Source Mass, g	ARF	RF
3	TiO_2	1000	1.5E-3	0.25
			9.6E-4	0.40
	UO_2		2.3E-4	0.25
			1.2E-3	0.49
		500	4.0E-4	0.70
			1.1E-3	0.38
	TiO_2	471.9	1.9E-3	0.34
		460.0	9.0E-4	0.40
		450.4	3.3E-3	0.62
		431.1	2.0E-4	0.40
		100	1.2E-3	0.31
			9.9E-4	0.46
	UO_2		4.0E-5	0.91
			4.0E-4	0.44
	TiO_2	25	7.0E-4	0.50
			8.0E-5	0.50
1	TiO_2	1000	5.0E-4	0.53
			1.7E-5	0.40
	UO_2		6.0E-5	0.46
			8.0E-5	0.50
		500	4.0E-5	0.83
			8.0E-5	0.52
	TiO_2	451	8.0E-5	0.42
		441.1	8.0E-5	0.41
	UO_2	100	2.0E-5	0.93
			7.0E-5	0.51
	TiO_2	25	1.0E-4	0.62
			2.0E-4	0.58

measured for these conditions. The bounding ARF and RF values for the lesser spill height are 5E-4 and 0.5. The median ARF and RF are 8E-5 and 0.5 with average values of 1E-4 and 0.6. The largest RF measured was 0.9 for the next to smallest ARF measured under these conditions. The measured ARFs are consistent with the values measured in Plinke et al. (1991) at approximately the same fall height.

The variation in the measured ARF values appears to be somewhat dependent on the level of the values. For ARFs at 1E-3 to 1E-4, the values appear to reflect variability of a factor of

5 to 10. At an ARF level of 1E-4 to 1E-5, the variability appears to be in the range of an order of magnitude or greater.

Figure 4-19 indicates that the RF of the source material is 0.95+ for both powders. The measured RFs indicate that the airborne particles have not been completely deagglomerated by the stresses imposed by the event. Powder this fine is extremely atypical of powder produced by process operations at nonreactor nuclear facilities.

4.4.3.1.3 Free-Fall Spill of Powder Model. Ballinger et al. (January 1988) proposes a model using the assumption that the powder disperses at a constant angle during falling and the diameter and velocity of the powder front can be calculated from the angle of dispersion and properties of the powder. Particles are sheared off during the descent and remained suspended. The model does not account for the suspension of particles upon impact. The powder spill model is based upon the following assumptions that appear to be somewhat inconsistent with observations in subsection 4.4.3.1.1:

- the growth rate of the powder front is constant and can be characterized by an angle of dispersion;
- amount of powder airborne is proportional to drag force on the powder; and
- the diameter of the powder front at the start of the spill is equal to the diameter of the container from which it was spilled.

A computer code, PSPILL, was developed to model powder spills. The model was run for varying values of M_o (mass of powder spilled, kg). An algorithm was developed based upon the statistical analysis of the results of the computer runs. The algorithm may be used to predict the ARF if the air density and viscosity are 1.18 kg/m^3 and 1.85E-5 Pa-sec, respectively. The fraction airborne release is:

$$ARF = 0.1064\ (M_o^{0.125})(H^{2.37})/\rho_{BP}^{1.02} \qquad (4\text{-}5)$$

where:
- ARF = airborne release fraction
- M_o = mass of powder spilled, kg
- H = spill height, m
- ρ_{BP} = bulk density of powder, kg/m^3.

In order to determine the bounding ARF, the value calculated from the model must by multiplied by a factor of 2 (the difference between the average and maximum value for the

3-m fall distance; the difference appears to diminish with fall distance probably due to the greater consistency for the impact dispersion as the force increases).

The best available correlation for particle size of the airborne material is:

$$AMMD = 12.1 - 3.29(\rho_{BP}) + 7540(F) \quad (4\text{-}6)$$

where: AMMD = aerodynamic equivalent mass median diameter, μm
F = fraction airborne
ρ_{BP} = bulk density powder, kg/m^3.

The equation only has a 46% correlation coefficient due to the variability in the data. The GSD of the size distribution for all powders (based on the powders used in the experiments), TiO_2 and UO_2 are 4.82, 3.73 and 5.60, respectively.

The median and bounding ARFs and RFs are based on the experimental measurements uncovered. For fall distances less than 3 m, the measured combination of ARF and RF values that yield the highest fraction of material in the respirable fraction, 2E-3 and 0.3, are assessed to be the bounding values. The median values are those determined considering both powders, 3E-4 and 0.5. For fall distance greater than 3 m, the bounding value can be estimated using the average airborne release fraction calculated by the model discussed in this subsection multiplied by a factor of 2 provided the value calculated exceeds that obtained with the ARF and RF combination of 2E-3 and 0.3.

4.4.3.2 Free-Fall Spill with Enhanced Velocity Effects Normal to the Direction of Fall

The conditions represented here are extreme for the normal range of stresses normally encountered in industrial-type accidents. The conditions may be found for powders undergoing pneumatic transfers or for minute quantities of soil fines that sift/spill during some earth moving operations such as digging with a backhoe or steam shovel in extremely windy conditions. The factors derived are not meant to be applied unless there is reasonable evidence that the conditions in the event or for the fraction of inventory indeed mimic the experimental conditions. For example, this data is not intended for fine judgements about low quantity releases from contaminated soil or for evaluating simple bulk powder spills in an atmosphere of overall room/glovebox ventilation or typical ambient conditions.

Sutter (August 1980) reported results of experiments to measure the entrainment of dispersed soil spilled into flowing air. Contaminated soil was collected, mixed and dried and pumped into a 0.61-m by 0.61-m wind tunnel. The experimental setup is shown schematically in

Figure A.39 in Appendix A. The size distribution of the soil is shown in Figure A.40 and tabulated in Table A.44. The soil was deagglomerated and pumped into the wind tunnel at speeds of 1.4-, 4.6-, 6.8- and 8.9-m/s (3.1, 11, 15 and 20 mph). The results are plotted in Figure 4-20, taken from the referenced source. The fraction airborne, ARF, is:

$$ARF = 0.0134\ U + 0.00543 \qquad (4\text{-}2)$$

where U = windspeed, m/s.

The RF measured at various windspeeds, shown in Table 4-14, ranges from 0.44 at 1.4 m/s to 0.90 at 8.9 m/s. The ARF at 8.9 m/s is 0.125 and, using the measured RF, 0.90, the fraction of soil less than 10 µm diameter is ~ 0.113. The fraction of the soil <10 µm diameter listed in Table A.43 is 0.00088. It would appear that a large mass of smaller particles was shed by the larger soil particles or some fragmentation has occurred.

The equation would only apply to soil or powder with a similar size distribution and characteristics. Process powders tend to be finer and more coherent and the observation here may not be applicable. On the basis of the experimental measurements, a bounding ARF and RF are assessed to be 0.0134 U + 0.00543 and 1.0 (limited by the total mass of material in this size fraction in the source material) are recommended.

Table 4-14. Airborne Soil Particle Size Distribution (Table 6 - Sutter, August 1980)

Wind Speed, mph	Aerodynamic Mass Median Diameter, µm	%10 µm and less
3.2	>10	44
10.4	6.7	63
15.2	9.8	50
20.0	5.3	90

4.4.3.3 Impact

Under some circumstances, powder at rest could be ejected into the air by the response of the solid substrate on which the powder rests by the vibration/jolting induced by the impact of falling debris or strong seismic vibrations. The effect could be minor for very solid substrates such as structural members. The flexing of thin, metal sheets could be substantial especially if the flexing is repetitive. Also the cohesiveness of the powder could inhibit

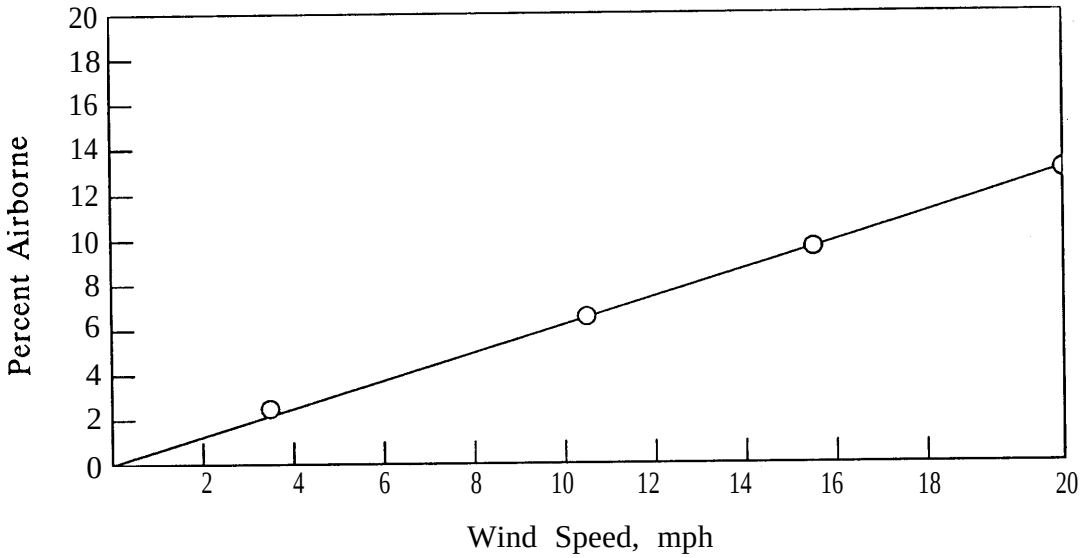

Figure 4-20. Percent of Soil Airborne as a Function of Wind Speed
(Figure 6 - Sutter August 1980)

suspension. Powders at rest are difficult to deagglomerate and disperse often requiring substantial mixing in suspensions to attain their original size distribution.

4.4.3.3.1 Vibration Shock

No study directly applicable to this phenomenon was found. It appears that the value for ARF should exceed that for resuspension alone (see section 4.4.4) but be less than for a free-fall spill of powder (ARF and RF of 2E-3 and 0.3 in subsection 4.4.3.1.2). The powder undergoing vibration shock (e. g., seismic vibration) is bounced into the air while subject to the same airspeeds as would impact the material for aerodynamic entrainment. The effect is less than free-fall of the same powder as it is not subject to the same intensity of phenomena.

Based on experience and judgement, Mishima, Schwendiman and Ayer (October 1978) selected a bounding ARF and RF of lE-3 and 1.0 for the suspension of powder-like surface contamination by shock-vibration. The experiments performed by Langer (1987) discussed in Subsection 4.4.3.3.2 below involving the suspension into flowing air of powders on plywood by impact of large pieces of debris also indicated a maximum ARF value of lE-3. Particles comprising surface contamination are assumed to be more widely dispersed and not as agglomerated as "thick" layers of particles that represent powders. Figure A.41 indicates the forces necessary to deagglomerate/disperse powders. Therefore, for clumps/piles of

powder, the same value for the ARF, 1E-3, is recommended but the RF is reduced to 0.1 due to the difficulty of deagglomerating powders.

4.4.3.3.2 Large Falling Object Impact or Induced Air Turbulence. Under some circumstances (e.g., seismic events) substantial portions of structural features and equipment may fall into radionuclide-bearing-powders released from confinement. If the fall of the objects generates a substantial air movement, the powder impacted may be suspended by the aerodynamic stress imposed.

Langer (1987) dropped three rocks (1.29 kg, 1.17 kg, and 1.82 kg) 3.7-m onto powder on a plywood sheet (called the "impact area") or held in a can in a vented metal box placed on the impact area. The experimental apparatus is shown schematically in Figure A.36 and the results are reproduced in Table A.42 in Appendix A. Air (430 cfm) was drawn into the box via a filter that removed particles >5 µm in diameter and passed through the impact area at a velocity of 0.8 mph (0.36 m/s). Most of the air (91%, 390 cfm) was drawn through a 8-in. diameter cyclone with a 10 µm AED cutoff. The particles penetrating the cyclone were collected on a special high volume filter paper. The remainder of the air (~ 9%, 40 cfm) was passed through a cyclone with a 5 µm AED cutoff, a one-stage impactor that removed all particles >0.5 µm AED, and the particles penetrating the system were collected on a glass fiber filter. Two optical spectrometers (1 l/min. instrument that classified particles 5 to 100 µm into 4 classes and a 3 l/min. instrument that divided particles 0.2 to 12 µm into 16 classes) provided real-time aerosol particle size distributions and number concentrations.

Four powders were tested - sand (<2000 µm and <500 µm), sand plus Al_2O_3, Al_2O_3, and nickel metal. An indication of the size of the powders is reproduced in Table A.42. The intent of the experiments was to determine the release of plutonium powders impacted by building debris. For typical plutonium dioxide powder formed in the foundry, ~ 0.01% is in the respirable size range (defined by the author as particle <3 µm AED), 0.3% in the inhalable range (defined by the author as particles <10 µm AED) and ~ 2.2% was <25 µm AED (based on data from optical sizing of the powder). All three materials used as surrogates were free-flowing (non-cohesive) powders unlike fine PuO_2. The size distribution of both Al_2O_3 (gritty, free-flowing, large proportions of fines) and nickel (hard, nearly spherical metal, free-flowing) were finer than that for foundry PuO_2. Airborne releases for non-cohesive powders should be greater than for cohesive powders, with Al_2O_3 characteristics most closely paralleling the PuO_2. The density of Al_2O_3 is 3.965 g/cm^{3,} approximately one-third that of PuO_2, and the density of nickel is 11.5 g/cm^3. The relevant data from the experiments are tabulated in Table 4-15.

The highest measured ARF for all materials is 1E-3 for the Al_2O_3 uncontained on the pad. The RF associated with this configuration is 0.3, a value near the middle of the RFs

Table 4-15. Measured ARFs and RFs from the Impact of Structural Debris on Powders (Table 1 - Langer, November 1987)

Material	Configuration	<10 µm AED	>10 µm AED	ARF	RF
Sand, <2000 µm	qt. can	2.5E-6	2.6E-4	2.6E-4	0.01
Sand, <500 µm 1.8% <25 µm	qt. can	2.0E-5	2.8E-4	3.0E-4	0.07
Sand, <500 µm 1.8% <25 µm	on pad	3.1E-4	5.6E-4	8.7E-4	0.36
Sand, <500 µm plus 2.6% Al_2O_3	on pad	7.2E-5	4.3E-4	5.0E-4	0.14
Al_2O_3 <300 µm 24% <25 µm	on pad	3.3E-4	8.9E-4	1.2E-3	0.27
Nickel 0.2% <25 µm	on pad	1.0E-4	7.5E-5	1.8E-4	0.57

measured. The median ARF and RF for all materials were 4E-4 and 0.2 with average values of 6E-4 and 0.2. The range of ARFs is a factor of 5, from 2E-4 to 1E-3. Without multiple measurements under the same test conditions, it is difficult to estimate a level of uncertainty. The limited range of test conditions and the difficulty in reproducing the test conditions described introduce considerable uncertainty. The analytical measurement instruments used, however, are not a source of high uncertainty. Other measurements under similar circumstances at this level of ARF resulted in an order of magnitude uncertainty in the values. This level of uncertainty appears appropriate for these values.

There are only single experiments on each material and configuration. It is assuring that the ARF for nickel metal powder, which has approximately the same density as PuO_2 and a finer distribution of particles like PuO_2, is less than the ARFs measured for sand. As would be expected, the ARF for Al_2O_3, a low density fine powder, is high. Accordingly, the data do not appear to involve unforeseen response characteristics that would make the limited data set essentially indeterminate.

The size and weight of the debris used and the fall heights appear to bound a number of phenomena in nonreactor nuclear facilities, including seismic vibration and impacts on large confinement structures such as gloveboxes. However, the size and weight of debris and the fall heights also appear to be unrealistically low for severe conditions in facilities such as a large building collapse, where large-sized debris from multiple levels may impact the released materials. In as much as the release mechanism appears to be air turbulence and shock-vibration, factors that can potentially increase with mass and size of debris and fall height. On the other hand, as the debris size increases, the impact effect is less likely to be fully concentrated in one area, and debris will provide cover for material that could limit releases.

To be conservative, however, it must be considered that the data may not be bounding. The "median" values of ARF and RF for all experimental configurations tested were 4E-4 and 0.2 (ARF x RF = 8E-5) and the highest ARF x RF value was 4E-4. Due to the uncertainty in the test conditions, a conservative bounding value for the ARF is assessed to be 1E-2 with a RF of 0.2. This yields an ARF x RF value of 2E-3, which is a factor of 5 greater than the largest measured value, a factor of 25 greater than the median, and of the same order as values assessed for accelerated airflow parallel to powder surface and deflagration over relatively unconfined powder.

The bound noted above applies to loose powders directly impacted by debris and associated air currents. There are, however, other material configurations, with some degree of material protection, of interest in nonreactor nuclear facilities. Powders may be contained in cans, cans in gloveboxes, cans in cans, etc. Damage to cans from debris impact is anticipated, ranging from cracks due to plastic deformation to displacing lids or jagged shearing. As this level of damage requires impact force, it will occur when the can comes to rest on some solid surface. In a total building collapse, this would typically be at the debris-strewn ground/basement floor, where can remains would be buried under the falling debris.

The Langer data presented in Table 4-15 include two cases where the sand aggregate was placed in a typical steel quart can without a lid. The rocks were dropped from the same distance onto this can. Case #1 involved powder screened to be < 2 mm (2000 µm) in diameter, while case #2 involved powder screened to be < 0.5 mm (500 µm) in diameter with 1.8% < 0.025 mm (25 µm). An experiment was also performed with the same powder used in case #2 uncontained. The ARF x RF for the various experiments are:

	ARF	RF	ARFxRF
< 2 mm sand in can	3E-4	0.01	3E-6
< 0.5 mm sand in can	3E-4	0.07	2E-5
< 0.5 mm sand uncontained	9E-4	0.4	4E-4

There appears to be a significant decrease in the overall respirable release, due most likely to some combination of shielding of the powder and interaction between the powder and confining surfaces. As in the estimate for loose powder, there is considerable uncertainty associated with this data. If the highest ARF from the data set (1E-3 for uncontained Al_2O_3 powder) is used in conjunction with the largest RF from the contained experiments (rounded up to 0.1), the bounding values would be the same as that assessed for vibration shock of loose, clump powders, and the overall ARF x RF would be a factor of 5 greater than that measured in the experiment (1E-4 vice 2E-5). Accordingly, for powder held in cans failed by debris, an ARF of 1E-3 with an RF of 0.1 is assessed to be bounding.

4.4.4 Aerodynamic Entrainment and Resuspension

Particles can be entrained from a surface in two configurations: 1) entrainment of particles from the surface of a homogeneous bed (e.g., relatively flat pile of powder, soil), and 2) entrainment of particles from sparse deposits on a heterogeneous surface (hard, unyielding). Since the entrainment of particles is a function of the characteristics of the flow, particles and surface, the entrainment of particles from the surfaces can be substantially different. Entrainment from homogeneous beds is discussed in this subsection covering powders exposed to normal air flow characteristics. The entrainment from sparse deposits on hard, unyielding surface will be covered in the subsection discussing aerodynamic entrainment of surface contamination (section 5.2.4).

The suspension of particles from the surface of a homogeneous bed (includes powders and contamination on soil) under routine process (stable ventilation flow velocities and patterns indoors) and meteorological (steady windspeeds less than 5 m/s) conditions appears to be dependent upon the interaction of various factors such as source, surface, upwind topographical and flow characteristics.

4.4.4.1 Entrainment From the Surface of a Homogeneous Powder Layer

4.4.4.1.1 Review of Literature on Resuspension Phenomenon, Factors, and Rates. Sutter (May 1982) reviewed much of the known information and data for resuspension factors and rates and fractional releases. Both suspension by aerodynamic and mechanical stresses are covered.

Resuspension factors are defined as the ratio between the airborne concentration of a pollutant per cubic meter directly over a contaminated surface and the areal pollutant surface contamination. The units are meters^{-1}. In concept, the factor represents the uniform concentration above a contaminated surface at whatever height. As measured, the factor represents the airborne concentration of a pollutant measured at some height above the surface collected over some period of time versus the surface contamination without knowledge of what is the true inventory-at-risk. The relationship may have some relevance for indoor (static volumes and relatively reproducible conditions) but does not appear to reflect the physics of the situations outdoors.

For outdoor situations where the contaminated surface is predominantly soil (although contamination can be resuspended from vegetation, asphalt roadways, rocks, buildings surfaces, etc.) and the aerodynamic or mechanical stress can be imposed from many directions and levels, the situation is much more complex. Particulate contamination is assumed to agglomerate (become attached) with the soil particles. The removal and

displacement of material from soil surfaces as a function of surface stress show three types of behavior - saltation, surface creep and suspension.

Saltation is defined as a mode of soil/sand particle movement where particles have alternate contact with air and ground in a layer close to the surface and affects particles in the size range of approximately 100 to 500 μm in diameter. The size range of particles suspended is shown in Figure 4-21 reproduced from Martin et al. (October 1983) indicating that particles in this size range are the most readily suspended (depends upon local wind conditions and particle morphology). It is noted that the graph plots the square root of particle diameter vice friction velocity and is not a direct indication of the air speeds or particle diameters. This is the approximate size range for saltating particles. Saltating particles can initiate surface creep and suspension upon impact and/or continue saltation.

Larger particles move by surface creep where particles always maintain contact with the surface. Surface creep affects particles in the size range of 500 to 1000 μm (0.5 to 1.0 mm) in diameter and cover the behavior of particles that slide or roll across the surface pushed by wind stresses and momentum exchange resulting from the impact of saltating particles.

Suspension is the mode of movement where particles do not come in contact with the surface locally. Suspension affects particles in the size range less than 100 μm in diameters and cover the behavior of particles that tend to follow the air motions. The fraction of soil eroded by the three modes varies greatly - 50 to 75 wt/o by saltation, 5 to 25 wt/o by surface creep, and 3 to 40 wt/o by suspension.

Three tables (Tables 4-16, -17, and -18) reproduced from the referenced source are included. Table 4-16 tabulates resuspension factors (mostly from soil or vegetated soil but unspecified city surfaces are included) from wind stresses. The values range from ~ 1E-10/m to 5E-5/m. Table 4-17 tabulates the resuspension factors measured for mechanical stresses and are almost completely for indoors situations. The values range from 1E-10/m to 2E-2/m. Table 4-18 shows the resuspension rates from the published literature and all represent outdoors conditions (a mix of soil erosion and experimental data). The values ranges from 4E-9/hr to 4E-1/hr. The latter value, 4E-1/hr, is for corn pollen and thus is extreme. Pollen has been naturally selected over millennia to be readily driven airborne (low density, large surface area) and is not representative of the particles of interest in accidents at nonreactor nuclear facilities. Not all the relevant factors (e.g., windspeeds, particle size distribution of soil or pollutant, local surface conditions) are given in the data nor is it always clear whether the relevant factors are known.

Sehmel (1980) provided a comprehensive review of literature on the resuspension/suspension (erosion) of soil. Resuspension is defined as re-entrainment of material deposited on the

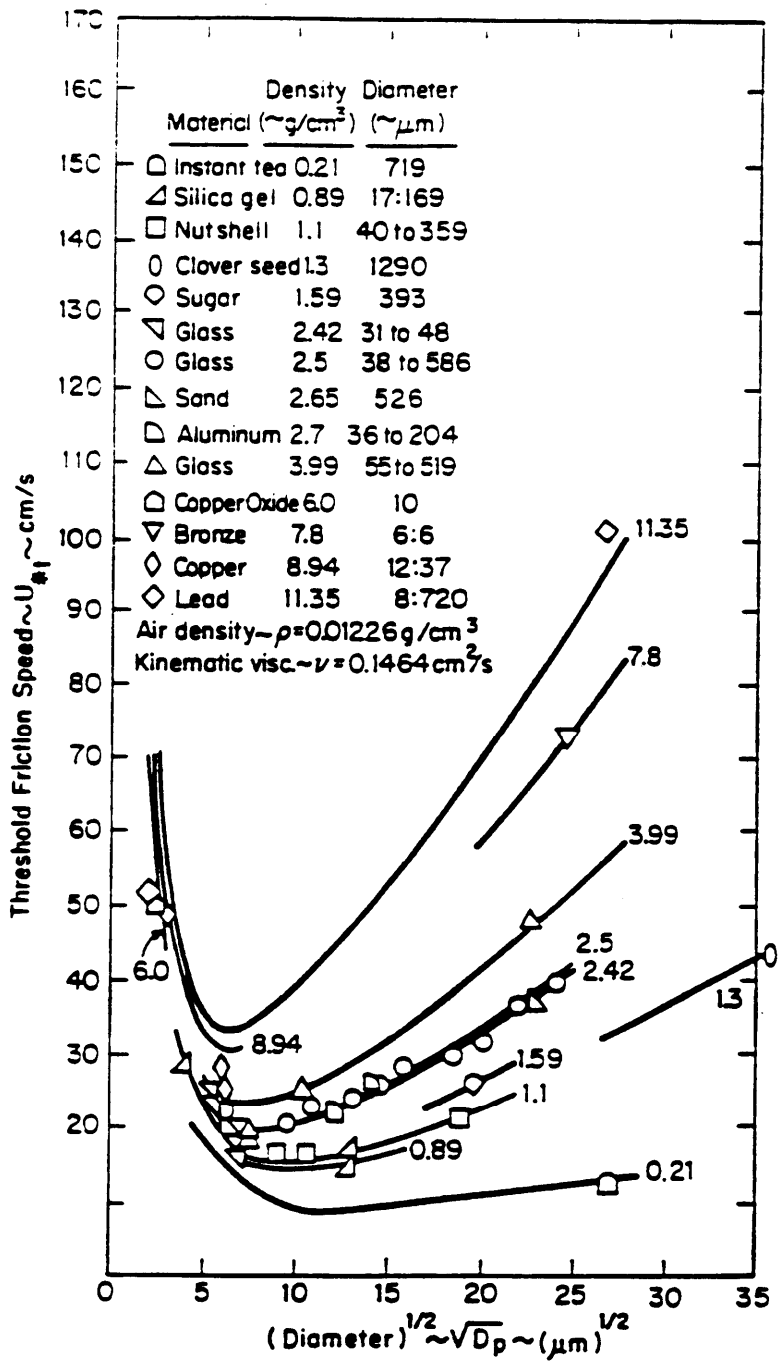

Figure 4-21. Particulate Threshold Friction Speed (Figure 2 - Martin, et al. October 1983)

Table 4-16. Resuspension Factors from Wind Resuspension Stresses (Table 2.2-1 - Sutter, May 1982)

Location	Source Material	Wind Speed Dependency	Resuspension Factor Range, m^{-1}	Reference
Nevada Test Site, Schooner	^{131}I		10^{-7} to 6 x 10^{-5}	Anspaugh et al. 1970
Nevada Test Site, GMX near center GMX near edge	Pu		 3 x 10^{-10} 3 x 10^{-9}	Anspaugh, Shinn, and Phelps 1975
New York--average air concentrations and surface depositions	Fallout		5 x 10^{-9} to 2 x 10^{-6}	Calculated from Bennett 1976
New York	^{238}Pu		5 x 10^{-8}	Bennett 1976
United Kingdom	^{238}Pu		5 x 10^{-9}	Bennett 1976
Palmares, Spain	Pu 10 day samples	U <9.7 m/sec	1.4 x 10^{-9} to 7.8 x 10^{-6}	Calculated from Icanzo and Salvador 1970
Nevada Test Site, dusty rural air	Pu		7 x 10^{-6}	Langham 1971
Hanford, Washington	Inert tracer, 1 m	Increases	1.7 x 10^{-10} to 2.7 x 10^{-7}	Sehmel and Lloyd 1975
Rocky Flats, Kentucky	Pu		10^{-9} to 10^{-5}	Sehmel and Orgill 1973
Maralinga trials	U 0.3 m ht 0.6 m ht	U <5 m/sec	3 x 10^{-4}	Stewart 1967
Paving stones	U_3O_8, 2 m^2 source	U <5 m/sec	8 x 10^{-9} to 6 x 10^{-6}	Stewart 1967
Sandy desert grass	U_3O_8, 9 m^2 source		1 x 10^{-9} to 5 x 10^{-5}	
Sandy soil with charred debris	^{210}Po, 1 m^2 source ^{210}Po, 1300 m^2 source	U <5 m/sec U <5 m/sec	2 x 10^{-9} to 9 x 10^{-6} 7 x 10^{-8} to 1 x 10^{-5}	Stewart 1967
Monte Bello Islands, hurricane trials	Fallout: 16 m source, lightly vegetated sand and rock near tower. Shot near road, no disturbance.	U <5 m/sec	1 x 10^{-6} to 1 x 10^{-5} 1 x 10^{-8} to 2 x 10^{-7} 1 x 10^{-8} to 1.5 x 10^{-6}	Stewart 1967
Cleared sandy soil	YCl_3, aqueous, 1 m^2 source YCl_3, aqueous, 9 m^2 source	U <5 m/sec	9 x 10^{-11} to 5 x 10^{-8} 5 x 10^{-10} to 6.2 x 10^{-7}	Stewart 1967

Table 4-17. Resuspension Factors from Mechanical Resuspension Stresses (Table 2.2-2 - Sutter, May 1982)

Location	Source Material	Wind Speed Dependency	Resuspension Factor Range, m^{-1}	Reference
Unventilated room	Be	Vigorous sweeping	1×10^{-2} to 4×10^{-2}	Mitchell and Sutsler 1967
Small unventilated room	Alpha floor contamination	People walking	3×10^{-4} to 2×10^{-2}	Calculated from Brunskill 1967
Change room with no ventilation	Alpha on coveralls	2 to 4 people changing coveralls	2×10^{-4} to 1.7×10^{-2}	Calculated from Brunskill 1967
Change room, concrete floor, 9 air changes/hr	"Loose" alpha floor contamination	4 to 6 people moving in room	2×10^{-4} to 3×10^{-3}	Brunskill 1967
Simulated work	Asbestos contaminated coat	Machining operations Stacking sheets	1.2×10^{-3} to 5.3×10^{-3} 2.0×10^{-3} to 4.2×10^{-3}	Carter 1970
Room--concrete floor	Pu facility	No circulation Fan air stress Fan and dolly movement No circulation after tests	1×10^{-3} to 2×10^{-4} 3×10^{-4} to 3×10^{-3} 4×10^{-3} to 1.5×10^{-2} 5×10^{-4} to 1×10^{-3}	Glauberman, Bootmann, and Breslin 1967
Room--concrete floor	U facility	No circulation Fan air stress Dolly movement Fan and dolly movement	7×10^{-5} to 4×10^{-4} 3×10^{-5} to 2×10^{-4} 1×10^{-4} to 2×10^{-4} 2×10^{-4} to 1×10^{-3}	
Maralinga trials	Fallout U Pu	Road survey, at 1-2 days Cab Land Rover, 5th hr 8th hr Dust stirred, 0.3 m ht Vehicle dust, 0.3 m ht Pedestrian dust, 0.3 m ht	1×10^{-8} to 2×10^{-6} 6.4×10^{-5} 2.5×10^{-5} 1×10^{-3} 3×10^{-7} to 7×10^{-4} 1.5×10^{-6} to 3×10^{-4}	Stewart 1967

Table 4-17. Resuspension Factors from Mechanical Resuspension Stresses (cont.)
(Table 2.2-2 - Sutter, May 1982)

Location	Source Material	Wind Speed Dependency	Resuspension Factor Range, m^{-1}	Reference
Particles settled in room	ZnS MMD = 3.1 μm σg = 1.9 CuO MMD = 2.0 μm, σg = 2.3, alpha particulate	Vigorous work-sweeping Walking Light work Light sweeping Pedestrian and equipment	1.9×10^{-4} 3.9×10^{-5} 9.4×10^{-6} 7.1×10^{-4} 4.6×10^{-3} to 5×10^{-5}	Fish et al. 1967 Walker 1978
Civil defense trial	^{131}I	Bomb recovery operation with brick/plaster dust Enclosed space Open area	4.6×10^{-3} to 5×10^{-5} 4×10^{-5} to 2×10^{-4} 2×10^{-6}	Stewart 1976
Nevada Test Site	Pu	Extensive vehicular traffic	7×10^{-5}	Langham 1971
Laboratory room	PuO_2	No movement 14 steps/min 36 steps/min	2×10^{-8} 2×10^{-6} 5×10^{-6}	Jones and Pond 1967
Monte Bello Islands, hurricane trails	Fallout	Road survey from back Land Rover, 4th day 7th day At tailboard, 7th day	8×10^{-7} to 3×10^{-5} 7×10^{-7} to 4×10^{-6} 1.6×10^{-5} to 3.1×10^{-5}	Stewart 1967
Australian desert "Totem" 1953	Fallout gamma	Walking survey Vehicle survey at tailboard Inside vehicle	3×10^{-7} 2×10^{-6} 2×10^{-5}	Stewart 1967

Table 4-17. Resuspension Factors from Mechanical Resuspension Stresses (cont.)
(Table 2.2-2 - Sutter, May 1982)

Location	Source Material	Wind Speed Dependency	Resuspension Factor Range, m^{-1}	Reference
Contaminated field	Pu	Downwind tractor in tractor cab Fertilizing Subsoiling Planting, light discing Subsoiling Planting, light discing Mowing	5×10^{-8} to 1.0×10^{-6} 2.4×10^{-7} 7.9×10^{-9} 5×10^{-6} 1.8×10^{-8} to 7.9×10^{-6} 5×10^{-6} 1.8×10^{-8} to 7.9 to 10^{-6} 1.8×10^{-5}	Calculated from Milham et al. 1976
Tractor cab in contaminated field	Pu	Tractor operations	4×10^{-9} to 2×10^{-6}	Milham et al. 1976
New York	Fallout Pu	Average air concentration/ surface concentration	5×10^{-9} to 2×10^{-6}	Calculated from Bennett 1976
Palmares, Spain	Pu	Yearly average farming	1.2×10^{-10} to 8.8×10^{-7}	Calculated from Iranzo and Salvador 1970
Contaminated sludge	Pu	Rototilling	5.6×10^{-8}	Calculated from Myers et al. 1976

Table 4-18. Resuspension Rates
(Table 2.2-3 - Sutter, May 1982)

Location	Resuspension, sec^{-1}, Determined from:			Reference
	Source Material	Calculated Using Transport Mode and Concentration at Only One Height	Integration of Measured Concentrations	
Corn leaves	Pollen		10^{-6} to 10^{-4}	Calculated from Aylor 1976
Nevada test site	Pu	2.7×10^{-12} to 4.8 to 10^{-10}		Anspaugh, Shinn, and Phelps 1975
Nevada test site, GMX	Pu	10^{-12} to 10^{-10}		Healy 1974
Nevada test site	Pu	1×10^{-10} to 5×10^{-8}		Healy 1974, calculated from Wilson, Thomas, and Stannard 1960
Hanford site	ZnS			Healy 1974, recalculated from Healy 1955 and Healy and Fuquay 1958
Eroding field 0.5 tons/acre/mo 40 tons/acre/mo 310 tons/acre/mo	Soil	 2×10^{-9} 2×10^{-7} 1×10^{-6}		Calculated from Skidmore and Woodruff 1968
Prairie terrain	Calcium molybdate	10^{-8} to 10^{-10}		Sehmel and Lloyd 1975
Hanford site	Respirable particulates	10^{-11} to 10^{-7}		Sehmel 1979

surface from the atmosphere. Suspension is defined as the entrainment of particles on the surface from nonatmospheric processes. The term resuspension is used to cover both processes since, once the material is entrained, it is not possible to distinguish the behavior of material generated by either process. For soil, the pollutant particles are attached to the host particles. The data are insufficient to validate deposition, resuspension, diffusion and transport deposition and airborne plume models.

Soil transport was extensively covered. Various equations are available to estimate soil loss predominantly for agricultural purposes and are not particularly relevant here. Models based upon wind stresses on individual soil particles are subject to formidable uncertainties when required to integrate over the entire surface area. Due to the uncertainty as to the applicability of these models, direct measurements of resuspension are preferred.

Table 4-19 lists some factors that may influence outdoor resuspension from soil. Not all these factors are operative in any given situation and some factors may dominate in certain regimes. The range of published resuspension factors for various aerodynamic and mechanical stresses are shown in Figure 4-22 from Sehmel (1980) and are the same as shown in Tables 4-16 and 4-17. Although shown together, it is difficult to compare the listed values due to the differences in conditions and measurement techniques.

4.4.4.1.2 Experimentally Measured Resuspension Rates. Sehmel and Lloyd (1976) reported the results of a 4-month study of the resuspension of a submicrom diameter tracer from a lightly vegetated soil surface due to ambient wind stresses. An aqueous slurry of submicron sized calcium molybdate stabilized with a surfactant was sprayed on the surface of a lightly vegetated area in a circle with a 22.9-m radius on the Hanford site. The surface roughness height was 34 mm. The average concentration over the area was 0.63 g molybdenum/m^2. Material resuspended were sampled by the arrangement shown schematically in Figure A.37a using sampler as shown in Figure A.37b that aligned the sampler inlet with flow. During the first two test periods (10/2/73 to 11/4/73 and 11/16/73 to 12/16/73) only one sampler was used at each height and all samplers operated continuously. During the third test period (1/16/74 to 2/8/74), the sampling arrangement was as shown in Figure A.34 and a single sampler at each height turned on automatically for three velocity ranges (1.3 to 3.6 m/s, 3.6 to 5.8 m/s, and >5.8 m/s). The maximum recorded gust during these periods was 20.1 m/s (45 mph). Samples were not taken during periods of precipitation. The sampler inlet were isokinetic for a windspeed of 0.52 m/s. Corrections for anisokinetic sampling are difficult to estimate for the continuous sampling and are at least an order of magnitude less than the measured levels.

Resuspension rates for the test conditions were of the order of 4E-5/hr to 4E-7/hr. The proper application required an equation that continuously depletes the source since there are

Table 4-19. Variables Which Influence Resuspension
(Table 1 - Sehmel 1980)

Pollutant Particle Properties	Soil Properties	Particle-Soil Interaction	Surface Properties	Topography	Meteorological Variables
Chemical Reactivity	**Moisture content**	**Attraction forces**	**Areas of erodible surface**	**Agricultural**	**Air density, affected by:**
Density	Ratio of erodible to non-erodible fractions	• Molecular forces: Van-der-Waals forces	Cohesiveness of particles	Broken	• Humidity
Hydrophobic		• Chemical binding forces (Valence forces)	• Binding action of materials	Desert	• Pressure
Hydrophilic Particle Diameter	Soil size frequency distribution	• Electrostatic forces	• Surface moisture	Diurnal	• Temperature
• Respirable • Nonrespirable	Specific gravity structure	• Magnetic forces Capillary forces at the surface of aggregates filled with liquid	Cover Obstructions	Flat Forested	Soil burden carried • Influence on air velocity
• Frequency Distribution	• Organic matter	Chemical reaction	Roughness, large scale	Hills	• Avalanching
Shape	• Lime content	Contact area effect on adhesion	• Mechanical turbulence	Humidity	• Saltation
Surface Roughness	Texture Temperature	• Particle shape • Particle size • Nature of particle and surface • Surface roughness • Adhesion of agglomerates vs. individual particles	• Overall sheltering Roughness, small scale • Orientation of obstacles • Percent of area covered by non-erodible aggregates or obstacles	Seasonal Temperature Undulating Uniform Urban	Surface moisture content • Dew • Frost upheaval Relative humidity Temperature

Table 4-19. Variables Which Influence Resuspension (cont.)
(Table 1 - Sehmel 1980)

Pollutant Particle Properties	Soil Properties	Particle-Soil Interaction	Surface Properties	Topography	Meteorological Variables
Chemical Reactivity	**Moisture content**	**Attraction forces**	**Areas of erodible surface**	**Agricultural**	**Air density, affected by:**
		Crystallization of dissolved substances	• Sheltering of individual particles	Valley	• Surface dry
		Deposition of suspended colloidal particle	• Stability of aggregates against abrasion and disintegration by moisture and watering	Water	• Surface drying • Surface frozen
		Hardening binders			• Wind speed Wind removal forces
		Liquid bridges Mineral bridges Relative humidity	Temperature Topographic features • Broken		• Mean wind speed • Frequency, period, and intensity of gusts penetrating to the surface
		Temperature	• Uniform		• Transient and steady drag
		Time of contact	Vegetative cover		• Vertical wind profile
		Viscous surface coating	• Live vegetation		• Stability • Vertical turbulent exchange
			• Plant residue		• Surface roughness • Temperature stratification • Wind direction

Reference	Location	Stress
Mitchell and Eutsler, 1967	Room	Sweeping Vigorously
Calculated from Brunskill, 1967	Room	Walking
Calculated from Brunskill, 1967	Change Room	Changing
Glauberman, et. al., 1967	Room	Fan, Dolly
Carter, 1970	Room	Machining
Carter, 1970	Work	Stacking Sheets
Glauberman, et. al., 1967	Room	Fan
Brunskill, 1967	Change Room	Walking
Stewart, 1967	Maralinga Trials	Stirred Dust
Glauberman, et. al., 1967	Room	After Tests
Glauberman, et. al., 1967	Room	Fan, Dolly
Fish, et. al., 1967	Room	Sweeping, Light
Stewart, 1967	Maralinga Trials	Vehicle, 0.3m.
Glauberman, et. al., 1967	Uranium Facility	No Circulation
Stewart, 1967	Maralinga Trials	Walking
Glauberman, et. al., 1967	Room	Dolly
Stewart, 1967	C.D. Trials	Work, Enclosed
Stewart, 1967	Room	Fan
Glauberman, et. al., 1967	Pu Facility	No Circulation
Fish, et. al., 1967	Room	Sweeping Vigorously
Langham, 1971	Nevada Test Site	Vehicular
Stewart, 1967	Maralinga Trials	Cab, Landrover
Jones and Pond, 1967	Room	36 Steps/Minute
Fish, et. al., 1967	Room	Walking
Stewart, 1967	Monte Bello Islands	Vehicle, 7th Day
Stewart, 1967	Monte Bello Islands	Vehicle, 4th Day
Stewart, 1967	Australian Desert	Inside Vehicle
Calculated From Milham, et. al., 1976	Field	Mowing
Jones and Pond, 1967	Room	14 Steps/Minute
Fish, et. al., 1967	Room	Work, Light
Calculated From Milham, et. al., 1976	Field	Planting, Disking
Jones and Pond, 1967	Room	36 Steps/Minute
Calculated From Milham, et. al., 1976	Field	Subsoiling
Stewart, 1967	Monte Bello Islands	Vehicle, 7th Day
Stewart, 1967	C.D. Trials	Work, Open
Stewart, 1967	Australian Desert	Vehicle, Trail Board
Stewart, 1967	Maralinga Trials	Vehicle, 1-2 Days
Calculated From Bennett, 1976	New York	Fallout Concentrations
Calculated From Milham, et. al., 1976	Field	Tractor, Downwind
Jones and Pond, 1967	Room	14 Steps/Room
Calculated From Iranzo and Salvador, 1970	Palmares, Spain	Yearly Farming
Stewart, 1967	Australian Desert	Walking
Calculated From Milham, et. al., 1976	Field	Tractor Cab
Milham, et. al., 1976	Field	Tractor
Calculated From Myers, et. al., 1976	Sludge	Rototilling
Jones and Pond, 1967	Laboratory	No Movement
Calculated From Milham, et. al., 1976	Field	Fertilizing
Stewart, 1967	Maralinga Trials	Wind
Anspaugh, et. al., 1970	Nevada Test Site	
Stewart, 1967	Sandy-Grass	
Stewart, 1967	Sandy-Debris	
Stewart, 1967	Monte Bello Islands	
Sehmel and Orgill, 1973	Rocky Flats	
Calculated From Iranzo and Salvador, 1970	Palmares, Spain	
Langham, 1971	Nevada Test Site	
Stewart, 1967	Paving Stones	
Calculated From Bennett, 1976	New York - Fallout	
Stewart, 1967	Sandy-Cleared	
Sehmel and Lloyd, 1975	Hanford	
Calculated From Bennett, 1976	New York - Ur	
Hamilton Calculated by Bennet, 1976	United Kingdom - Ur	
Krey, et. al., 1976	Rocky Flats	
Anspaugh, et. al., 1970	Nevada Test Sit	

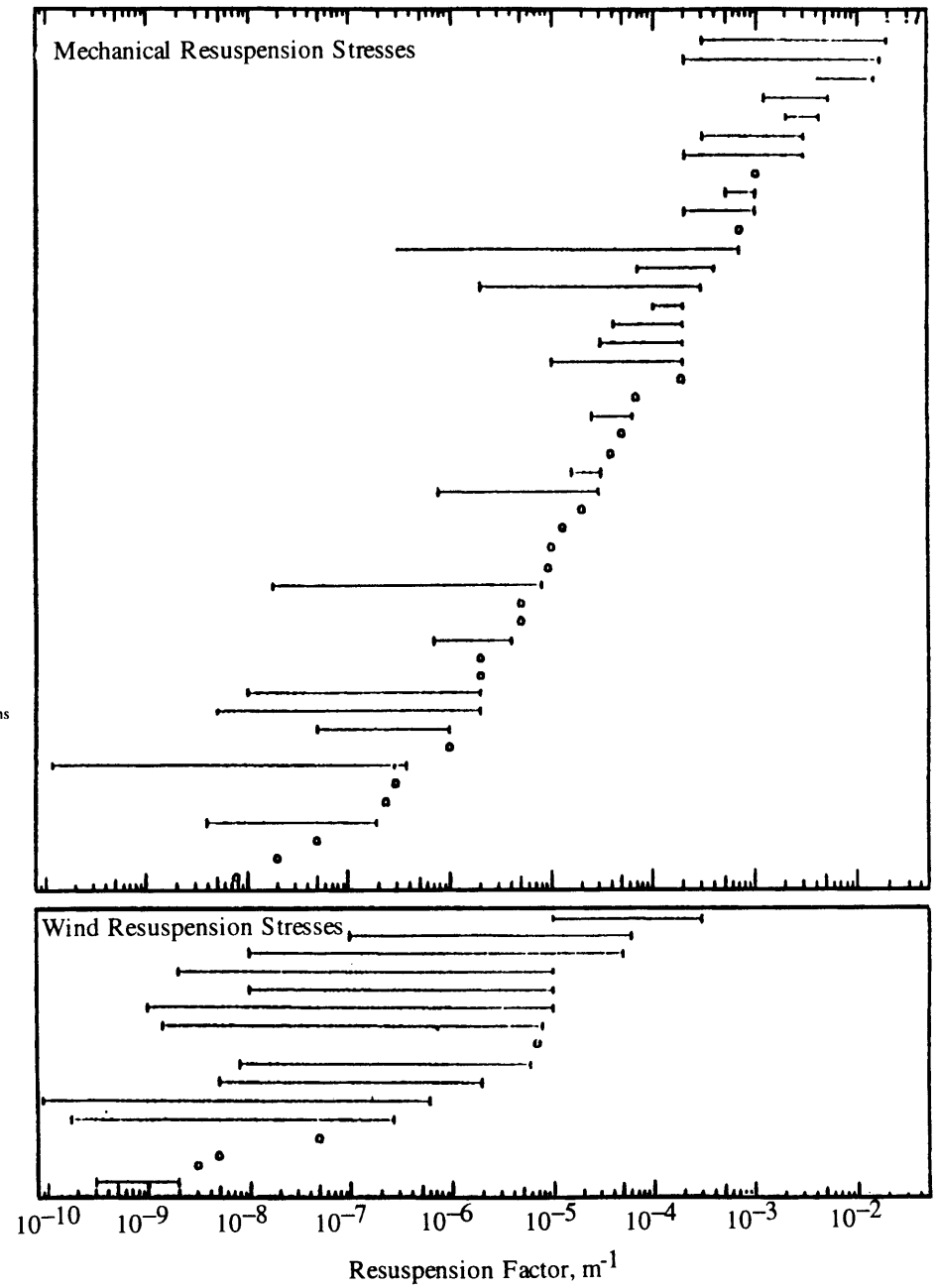

Figure 4-22. Resuspension Factor Ranges from Mechanical and Wind Resuspension Stresses (Figure 2 - Sehmel 1980)

8760 hr/yr and from 0.35% to 35% of the source is depleted per year. The error would not be significant for short periods of time (e.g., 4E-5 of the source is depleted in one hour at a resuspension rate of 4E-5/hr). An uncertainty is the possible loss of resuspended materials if the plume height exceeded the sampling height. Furthermore, it is uncertain which surface areas are being sampled by the samplers. The material plume spreads as it rises and travels downwind. The material sample is some cumulative fraction of some unknown upwind area.

Most recently, several researchers have investigated the long-term effects of the radionuclides deposited from the Chernobyl Nuclear Power Generating Station incident. Garland and Pigford (1992) investigated the resuspension of the material under a variety of circumstances. Resuspension factors varied inversely with the amount deposited. Some seasonal variation was observed and some increase due to traffic. Generally, results support the use of the lowest resuspension factors. Typical and mean resuspension factors from site with highest deposition during the initial periods were 5E-9 and 2E-8 and decreased for next periods. The sampling configurations nor periods were given but they most certainly exceed a second and are more likely in days. Therefore, even the initial resuspension factors would be less than the rates measured by Sehmel and Lloyd (1976).

Garger, Gavrilov and Zhukov (1992) modeled the transfer and fallout of radionuclides deposited from the Chernobyl accident. The airborne concentration and deposition rate of radionuclides due to resuspension and activities were modeled for normal and unfavorable steady-state meteorological conditions. Resuspension rates of 1E-9/s at 5 m/s and 2E-7/s at 15 m/s were calculated based on air sampling.

Based upon the information on resuspension factors and rates found in the two reviews, the experimentally measured rates tend to indicate that the long-term resuspension rate bound determined by Sehmel and Lloyd (1976), 4E-5/hr, is reasonable. Although the rates appear to decrease with time (and even higher rates are probably possible for short periods of time during the initial phases of the aerodynamic entrainment), using the initial, higher rates would be conservative. The value is derived for the continuous phase of the aerodynamic entrainment and does not represent the instantaneous airborne release from puffs. Caution is advised in evaluating results that are treated as instantaneous release values for nonreactor conditions. Furthermore, in an operational facility with a effluent exhaust system, the airflow was present during the release and materials deposited have already experienced the flow during transit. The fraction released with time is for MAR that may be difficult to define and decreases with depletion by the entrainment and other phenomena such as burial, cover by debris deposited on the surface, etc. The resuspension tends to fluctuate as the level of stress fluctuates and the surface conditions respond to the previous stresses. After an event, the powder released may be exposed to primarily aerodynamic stresses within the facility or remnants of the facility until remedial action can be taken. The time interval of

exposure would be hours rather than seconds. Thus, a bounding ARR of 4E-5/hr with a RF of 1.0 is recommended. However, as previously noted, the estimated respirable release should not exceed the total respirable material in the source if known.

No experimental data on the effects of large debris over the deposited powder on aerodynamic entrainment were found. Schmitt (May 1975) reported an approximate order of magnitude reduction in particulate emissions from carbon microsphere used to extinguish a fire. Due to the decrease in aerodynamic stress if the powder is shielded by remnants and debris of the structure or exposed to static conditions within the structure, an ARR and RF for powder under debris of 4E-6/hr and 1.0 are recommended.

- Bounding ARR and RF, homogeneous bed of powder exposed to ambient conditions (normal process facility ventilation flow or less, or nominal atmospheric conditions <2 m/s with gusts up to 20 m/sec) following an event: 4E-5/hr, 1.0

- Bounding ARR and RF, homogenous bed of powder buried under structural debris exposed to ambient conditions or under static conditions within the structure following an event: 4E-6/hr, 1.0

It is noted that these values are for freshly deposited material in the immediate aftermath of release. It would be inappropriate to use these values for cumulative summing of hourly releases for long-term contamination (i.e., months to years).

4.4.4.2 Suspension of Material by Vehicular Traffic

In the event that radioactive materials in powder form are spilled onto a roadway during transport or deposited by airborne transport during the accident, the deposited material may be impacted by vehicular traffic unaware of the presence of the material or by evacuation of personnel. Sehmel (1973) reported the results of experiments performed to measure the suspension of material on an roadway from the passage of a vehicle (passenger car and 3/4-ton truck) through the deposited material and in an adjacent lane. Particles of ZnS <25 μm in diameter (MMD <5 μm in diameter) were deposited on the asphalt surface of a 3-m by 30-m section of roadway (one lane of a two-lane road). The ZnS particle size distribution during deposition is shown in Figure A.38a in Appendix A. Sampling towers were located at three distances (3, 6, and 9 m) from the road with filter samplers at 0.3-m, 1-m, 2-m and 2.4-m heights. Realtime ZnS monitors were located at the 0.6-m level of each tower. Deposition samplers were located between the towers on each row and at 1-m, 18-m and 30-m. The arrangement is shown schematically in Figure A.38b in Appendix A.

The experimental results are tabulated in Table A.43a (resuspension by a passenger car) and Table A.43b (resuspension by a 3/4-ton truck). The integrated average resuspension per passage by or through the deposited material was calculated by mass balance using an average source of 0.5 g ZnS/ft^2. The resuspension rate for a passenger car driven past the unaged deposited materials ranged from 4.8E-5 per passage to 1.1E-3 per passage and increased with vehicles speed. The resuspension rates for a passenger car and 3/4-ton truck driven through unaged deposited material ranged from 1.9E-4 per passage to 1.09E-2 per passage and 2.5E-3 per passage and 6.7E-3 per passage, respectively. The measured rates are plotted against vehicle speed on Figure 4-23 reproduced from the referenced source.

The resuspension rate decreased rapidly with time. The resuspension rates after four days of aging are shown in Figure 4-24 reproduced from the referenced source and show an order of magnitude decrease. The calculated effects of ageing are shown in Figure 4-25 reproduced from the referenced source. In general, this correlation is of use only for the immediate aftermath of an accident. The highest resuspension rate for unaged deposited material is 1E-2/passage for a passenger car driven through the material and is recommended as the bounding value. In the absence of any measured RF values, a conservative value of 1.0 is recommended as the bounding RF. However, the amount of material released as a respirable aerosol should not exceed the total respirable material in the source, if known.

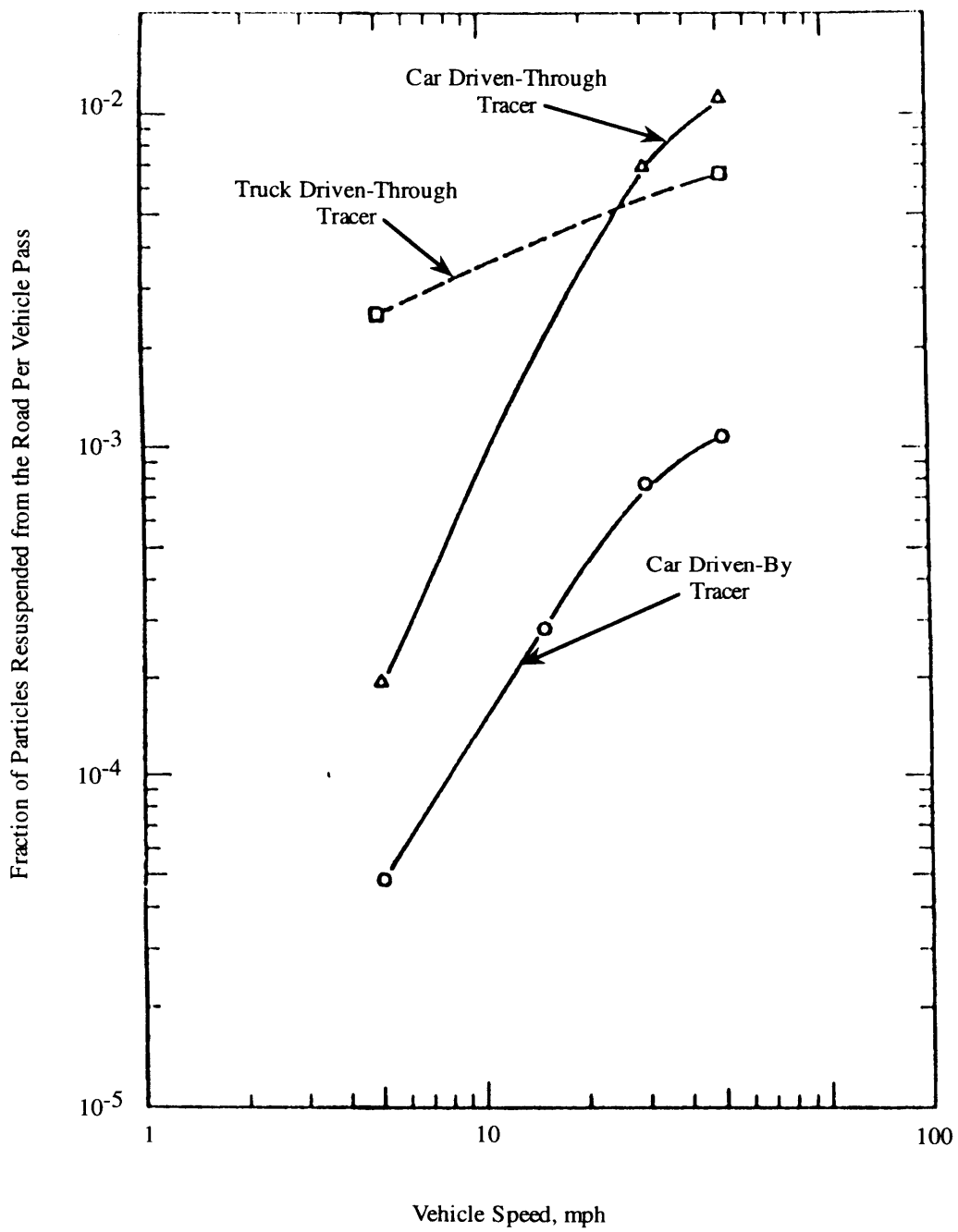

Figure 4-23. Particle Resuspension Rates from an Asphalt Road Caused by Vehicle Passage (Figure 3 - Sehmel 1976)

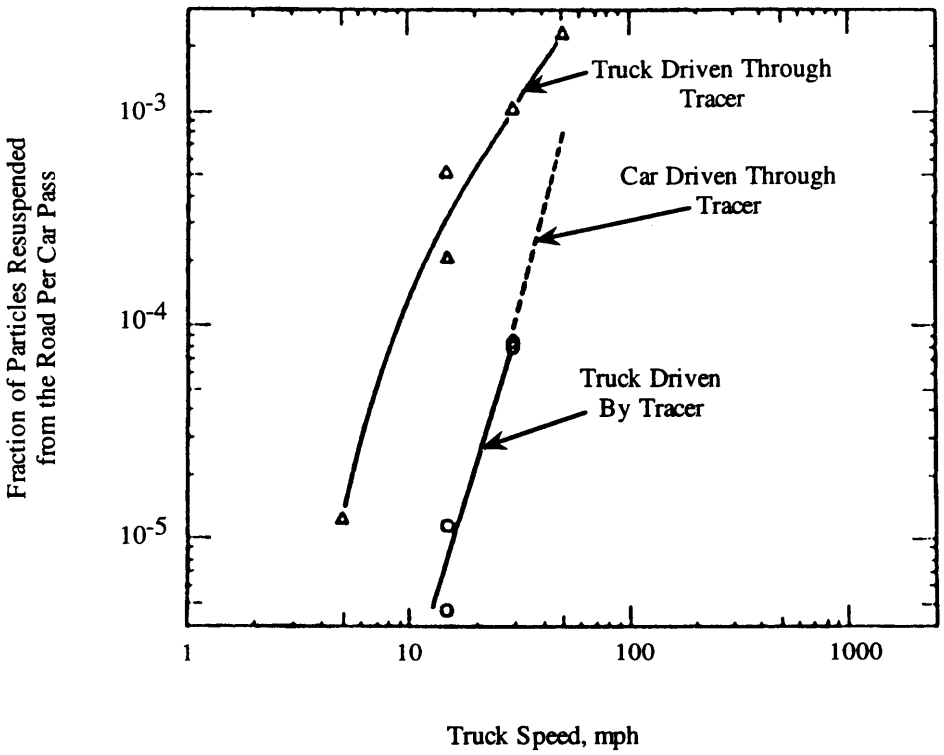

Figure 4-24. Particle Resuspension Rates from an Asphalt Road Caused by Vehicular Traffic Passage Four Days After Particle Deposition
(Figure 5 - Sehmel 1986)

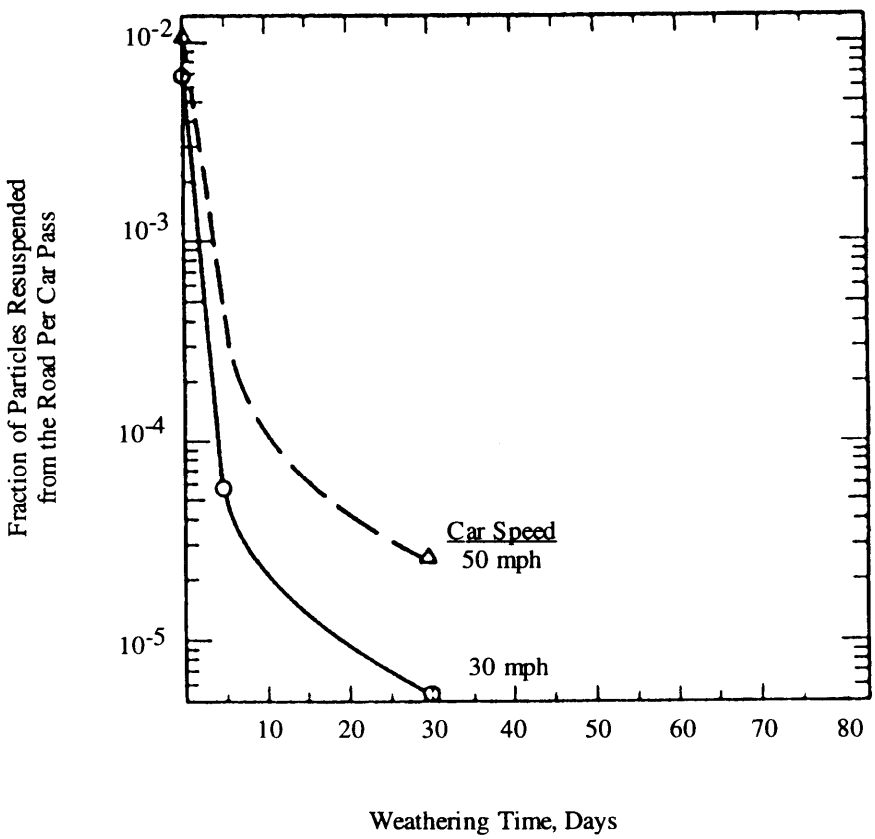

Figure 4-25. Particle Resuspension Rates from an Asphalt Road as a Function of Weathering Time (Car Driven Through Tracer) (Figure 4 - Sehmel 1976)

4.0 Solids; Powders

5.0 SURFACE CONTAMINATION

Release fractions are assessed for combustible and noncombustible materials on which surface contamination may be found. Combustible materials include solids such as trash, wood, and wall board. Trash may be made up of paper, rags, cardboard, and plastic (wrapping sheets, bags, containers, glovebox windows, tools, casings, and ion exchange resins). Where possible, distinctions are made between loose combustibles and combustibles in containers (e.g., pail packages, drums). Noncombustible materials are generally large pieces associated with the facility structure, enclosure, or containers. Such pieces or equipment may be concrete, metal, glass, etc. This material may include hard, unyielding surfaces, deformable surfaces, or even brittle surfaces. The contamination ARF and RF values, with the noted exceptions of burning and explosive shocks fragmenting base surfaces, are intended to be applied to loose surface contamination only.

An RF of 1.0 is assigned for the majority of these materials. To be technically correct, however, in all cases the total quantity of airborne respirable material should not exceed the total mass of respirable material in the original source MAR. Unfortunately, it is very difficult to characterize the size distribution of deposited radioactive contamination, which may come from a large number of different sources and is intermingled with non-radioactive particles that are not of interest. Accordingly, it is not expected that defensible bases exist for assuming an original source RF in most cases. Given that, it is important not to treat significant powder accumulations as being a surface contamination phenomena.

5.1 SUMMARY OF ANALYSIS OF DATA

Contaminated, Combustible Solids

Thermal Stress

- Packaged Mixed Waste. For contaminated combustible materials heated/burned in packages with largely non-contaminated exterior surfaces (e.g., packaged in bags, compact piles, pails, drums), the following values are assessed to be bounding. These values are based on the maximum experimentally determined ARF and conservative assumption of RF = 1.0.

Median	ARF 8E-5/RF 1.0
Bounding	ARF 5E-4/RF 1.0

- Uncontained Cellulosics or Largely Cellulosic Mixed Waste. For burning of unpackaged, loosely strewn cellulosic materials, such as paper, cardboard, rags, and wood shavings, the following values are assessed to be bounding.

These values are based on the maximum experimentally determined ARF and RF for a variety of cellulosic materials.

Median	ARF 5E-4/RF 1.0
Bounding	ARF 1E-2/RF 1.0

- Uncontained Plastics. The following values apply to burning of unpackaged contaminated combustible plastics.

  - all plastic materials except polystyrene: Based upon maximum experimentally determined ARF and RF for polychloroprene and polymethylmethacrylate:

Bounding	ARF 5E-2/RF 1.0

  - polystyrene: Based upon maximum experimentally determined ARF and RF for a limited set of experiments involving polystyrene contaminated with UNH solution. The value selected is based on rounding upward the maximum value from the data set:

Bounding	ARF 1E-2/RF 1.0

- Dispersed Ash Dropped into Airstream or Forced Draft Air. This ARF and RF is not typically applied to burning masses of combustible material in even large fires. It is applicable to extremely severe conditions where loosely contaminated combustible material is driven airborne as part of an updraft fireball. The material ignites and burns to substantial completion in the air currents, with complete disintegration of the substrate, for these values to be applicable. Such conditions are normally experienced by only a fraction of material even under very severe conditions. The maximum ARF experimentally obtained came from burning very small combustible specimens with a sprinkling of fine powder contaminant. Directed airflows of 0.46 and 1.0 m/s were used to suspend the disintegrating combustible material to maximize release potential. The ARF and RF assessed to be bounding for the fraction of material exposed to these severe conditions are:

Bounding, loose powder	ARF 4E-1/RF 1.0
Bounding, air-dried solution or adherent contamination	ARF 8E-2/RF 1.0

Explosive Stress

There are no applicable data for the release of contaminant from combustible solids due to the effects of shock or blast. The values assessed to be bounding are based on reasoned judgment. The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock Effects. Materials can be fragmented by shock effects but much of the material is extremely flexible and particles are unlikely to be dislodged from porous and fibrous surfaces. This phenomena is considered bounded by the values assessed for venting of pressurized gases over material discussed below. Subsequent ignition and burning are likely to provide the most significant overall release.

- Blast Effects. This phenomena is bounded by the values assessed for venting of pressurized gases over material discussed below. Again, subsequent ignition and burning of the substrate material may provide the most significant release.

- Venting of Pressurized Gases Over Contaminated, Combustible Waste. Given the flexible nature of this waste, which does not provide a rigid surface for airflow to act upon, the ARF and RF of 1E-3 and 1.0 for shock flexing of substrate material given in subsection 5.3.3.2.2 is considered to bound the phenomena. This value is applicable only to the portion of waste surfaces that are actually exposed.

Bounding	ARF 1E-3/RF 1.0

Free-Fall Spill and Impaction Stress

There are no applicable data for free-fall spill of combustible solids. Release estimates based on other phenomena are provided for packaged and unpackaged combustible solids.

- For materials with high surface area to mass ratios, no significant suspension is expected for freefall spill from typical working heights (~ 1 - 1.5 m).

- For the situation where the combustible material is unpackaged/lightly packaged (e.g., plastic wrapping only) and strongly impacts the floor or is impacted by falling debris (shock-vibration induced by impact), the bounding ARF and RF are based on reasoned judgment that suspension under these

circumstances are bounded by suspension postulated for loose surface contamination from shock/impact stresses (see subsection 5.2.3.2):

Bounding ARF 1E-3/RF 1.0

- For the situation where the combustible material is packaged in a relatively robust container (e.g., hard pail, drum) that is opened or fails due to impact with the floor or impaction by falling debris (shock-vibration induced by impact), the bounding ARF and RF are based on reasoned judgment that suspension under these circumstances will be bounded by suspension postulated for debris impacting powders in cans (see subsection 4.4.3.3.2):

  Bounding ARF 1E-3/RF 0.1

Aerodynamic Entrainment and Resuspension

No applicable data were found. The bounding ARRs and RFs quoted are based on reasoned judgement that entrainment under these conditions would be bounded by suspension of powders under similar circumstances (see section 4.4.4).

- Indoors or outdoors exposed to ambient conditions (normal process facility ventilation flow or less, or nominal atmospheric windspeeds < 2 m/s with gusts up to 20 m/s) following an event:

  Bounding ARR 4E-5/hr; RF 1.0

- Buried under debris exposed to ambient conditions or under static conditions within the structure following an event:

  Bounding ARR 4E-6/hr; RF 1.0

Use of the values given above for short time frames (<100 hours) would not introduce serious error due to the severe depletion of the source. For time periods exceeding 100 hours, the reduction of the source can be accounted for from the entrainment of material. In general, these values are intended for immediate post-accident conditions and freshly deposited material. They would overestimate releases from long-term contamination (i.e., months to years) and are not appropriate for such use.

Contaminated, Noncombustible Solids

Thermal Stress

Bounding values were selected based on reasoned judgement that the suspension of surface contamination (most probably in the form of a sparse population of particles attached to the surface) under thermal stress is bounded by the suspension of non-reactive powders under thermal stress in a flowing airstream (see subsection 4.4.1.1).

Bounding	ARF 6E-3/RF 0.01

Explosive Stress

The values assessed to be bounding are based on reasoned judgment. No applicable data were found. The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock Effects. For detonations in or contiguous to unyielding solid material, a respirable release of the mass of inert material equal to the calculated TNT equivalent is assessed to be bounding. At low mass ratios (mass inert material/mass TNT equivalent), the respirable release is comparable to the total material release. As mass ratios increase, the respirable fraction becomes significantly less than the total amount of material released, which decreases with increasing mass ratio as well.

- Blast Effects. This phenomena is considered bounded by values assessed for venting of pressurized gases over material discussed below.

- Venting of Pressurized Gases Over Contaminated, Noncombustible Material. The entrainment of the material is a function of the characteristics of the flow over the particulate material that may be lying on the surface, the particles, and the surface. Some of the flow characteristics are dependent on the initial pressure and the size of the vent. If the solid is characterized only by loose, externally deposited surface contamination, the release is characterized as to whether the overall containment volume in which the contamination exists is pressurized or not. If it is not, then the gases from the venting source are considered bounded by the phenomena of accelerated airflow parallel to the surface of powders (subsection 4.4.2.2.2.). If the volume is pressurized, the

phenomena can be considered bounded by venting of pressurized powders (subsection 4.4.2.3).

- Accelerated gas flows in area without significant pressurization

  Bounding ARF 5E-3/RF 0.3

- Venting of pressurized volumes

  Source > 0.17 MP_g (25 psig)

  Bounding ARF 5E-3/RF 0.4

  Source < 0.17 MP_g (25 psig)

  Bounding ARF 1E-1/RF 0.7

These release values would apply only to loose surface contamination on the solid, not the solid as a whole. No bounding ARF and RF can be uniquely identified for eroded or corroded material on the solid surface that no longer adheres tightly to the bulk solid. However, the release of such material is bounded by the release parameters assumed for loose surface contamination.

Free-Fall Spill and Impaction Stress

No applicable data. Based upon assumptions that release for various mechanisms bounded by airborne release of powder under similar circumstances.

- Free-Fall Spill. Most materials will not experience free-fall spill. The release for those that do is bounded by the values given for impact and shock below.

- Impact, shock-vibration.

  - Materials that undergo brittle fracture: airborne release by brittle fracture bounded by the ARF and RF postulated for brittle fracture in section 4.3.3. The contamination present on the portion of the base material calculated to be driven airborne will be assumed to go airborne as well.

    Bounding ARF x RF = (A)(P)(g)(h) (5-1, 4-1)

- Materials that do not undergo brittle fracture: airborne release by shock-vibration.

  Bounding ARF 1E-3/RF 1.0

Aerodynamic Entrainment and Resuspension

Bounded by ARR and RF postulated for aerodynamic entrainment of powders (section 4.4.4).

- Indoors or outdoors exposed to ambient conditions (normal process facility ventilation flow or less, or nominal atmospheric windspeeds < 2 m/s with gusts up to 20 m/s) following an event:

  Bounding ARR 4E-5/hr; RF 1.0

- Buried under debris exposed to ambient conditions or under static conditions within the structure following an event:

  Bounding ARR 4E-6/hr; RF 1.0

Use of the values given above for short time frames (<100 hours) would not introduce serious error due to the severe depletion of the source. For time periods exceeding 100 hours, the reduction of the source can be accounted for from the entrainment of material. In general, these values are intended for immediate post-accident conditions and freshly deposited material. They would overestimate releases from long-term contamination (i.e., months to years) and are not appropriate for such use.

HEPA Filters

Thermal Stress

Based upon conservative extrapolation of the maximum experimental measurement of release of particles accumulated by the passage heated air through HEPA filters.

Bounding ARF 1E-4/RF 1.0

Explosive Stress

The effect most closely resembling stresses in a given explosive-type accident scenario is chosen. There is no need to assume cumulative releases for all effects cited.

- Shock Effects. Based on experimentally measured releases of accumulated particles from HEPA filters experiencing localized failure from a momentary high pressure pulse.

  Bounding ARF 2E-6/RF 1.0

- Blast Effects. Based on maximum measured release of accumulated particles by passage of high velocity air through filters to filter break pressure.

  Bounding ARF 1E-2/RF 1.0

- Venting of Pressurized Gases Through Filters. Releases bounded by ARF and RF for blast effects in section 5.4.2.

  Bounding ARF 1E-2/RF 1.0

Free-Fall Spill and Impaction Stress

No applicable experimental data for airborne release of particles during free-fall of HEPA filters were uncovered. No significant release is postulated during free-fall. Releases on impact are bounded by conservative extrapolation of maximum releases measured for contained (in packages) and uncontained HEPA filters.

- Enclosed (e.g packages, filter or plena housing) HEPA with accumulated particles upon impact with hard unyielding surface

  Bounding ARF 5E-4/RF 1.0

- Unenclosed HEPA filter media with accumulated particles upon impact with hard, unyielding surfaces

  Bounding ARF 1E-2/RF 1.0

Aerodynamic Entrainment and Resuspension

Since the filters are designed and manufactured to collect and retain particles in flowing air, no significant release of accumulated particles is postulated by passage of air across face of filter. Even if the filter medium is exposed, no significant release of accumulated particles by nominal air velocities is postulated.

5.2 CONTAMINATED, COMBUSTIBLE SOLIDS

There are a wide variety of contaminated, solid, combustible materials found in nuclear facilities. These materials are generally trash, such as paper, rags, cardboard, plastic from wrapping, sheeting, bags, containers, windows, tools, casings, and ion exchange resins. There could also be contaminated structural material such as wood and wall board. The nature of the contaminant also shows great variation, ranging from solid particles adhering to the surfaces (resulting from wiping smearable contamination) to materials absorbed in the surface or matrices of the substrate (e.g., dilute aqueous solutions that have evaporated, solutions that have been absorbed in the material). Trash is generally held in a plastic bag in a container that is sealed after filling. Other forms of contaminated solid combustibles (e.g., windows, gloves) are uncontained until retired from use. Because each combination of substrate and contaminant may react differently to the various stresses imposed and suspend materials on their surfaces or absorbed into their matrices by different mechanisms, different ARFs are defined for different types of materials.

5.2.1 Thermal Stress

Contaminants on solid, combustible materials can be suspended by the destruction of the substrate and vapor flow generated during the formation of pyrolyzates or smoldering combustion and subsequently entrained in the overall fire convective currents for airborne transport. Various factors may affect either the burning rate (such as the availability of oxygen or the surface area of the combustible material exposed) or the entrainment (such as whether air passes through a burning mass). For example, individual thin layers of cellulose such as sheets of paper or tissue will burn rapidly and release the mineral content as ash. Burning of solid materials is a heterogeneous process; volatile vapors are released and mix with the air to form a flammable mixture. This action forms the commonly observed diffusion flame. Strands of cellulose also burn by smoldering combustion, which produces the glow observed in wood fires.

Particles that comprise the contaminant are released by the destruction of the substrate within the burning mass and are contained within the waste mass, general inorganic remains, and, for the period of combustion, an increasing depth of mineral ash. The vapors that are generated below the released particles by pyrolyzation of the cellulose may carry the released particle but must pass through the waste and ash mass before release into the atmosphere. It is the total waste matrix form and mass and this flow internal to the waste matrix from pyrolysis gases that are the controlling features in particulate releases for even large fires on the industrial scale associated with nonreactor nuclear facilities. The material in the waste matrix is not subject to the overall fire convective flow external to the waste. If the external flow intrudes upon the fuel-rich zone of combustion to a significant extent, the flame is displaced and less heat is radiated to the surface lessening the burning rate. If the intrusion

is sufficient, the fuel-rich zone and flame are sufficiently displaced so that insufficient fuel is vaporized to continue flaming combustion.

If thin layers of paper rest on other loosely strewn combustible materials, the internal vapor generation provides a lifting force that can suspend small particles and inject them into the convective flow induced by the flame. If the paper is introduced into a significant external upward airflow, an increase in suspension can occur. However, if the thin layers are tightly packed, the layers cannot burn efficiently due to the lack of oxygen, the burning may only be a surface phenomenon, or only smoldering combustion may occur. The lifting effect of vapors is substantially decreased by forcing it to pass through significant waste and ash mass as well. In such cases, the suspension of contaminants and ash is significantly reduced.

Because of the range of potential burning conditions, experimental measurements of the airborne release of surface contamination were used to provide the basis for the release fractions cited below. Various experimental studies have been performed to measure the ARF during the burning of contaminated solid materials (Mishima and Schwendiman, October 1970, April 1973; Halverson, Ballinger and Dennis, March 1987). The range of ARF values for the various materials is reproduced in Figure 5-1. The experimental data for each form are limited and only span a limited range of parameters. Inspection the results for burning of contaminated, combustible wastes shows that the ARFs and RFs can be placed in several categories - packaged mixed waste, uncontained cellulosics, uncontained plastics, and extreme situations where significant pressurized airflow passes through the burning mass or ashes from the burning mass falls into an strong induced airflow.

5.2.1.1 Packaged Waste

Current requirements have led to the packaging of contaminated, combustible waste in relatively substantial packages such as metal containers and drums. The material that is placed in these containers often consists of less robust waste packages in the form of single- or double-wrapped plastic packages bagged out of glovebox lines. Even waste placed together in a pile without even bag containment forms a loosely agglomerated package of sorts. The latter two cases of less robust packaging have been the subject of experiments by Mishima and Schwendiman (April 1973). These experiments examined the ARF and RF from burning mixed waste (i.e., paper, rags, tape, plastic, cardboard, oil) contaminated with uranium dioxide powder of a size distribution shown in Figure A.41 in Appendix A, uranyl nitrate liquid, and air dried uranyl nitrate. These experiments are considered to bound the airborne release from more substantial packages. The presence of more substantial material surrounding the contaminated, combustible waste would limit the availability of oxygen and force particles generated in the interior of the mass to pass through the ash/residue formed prior to release.

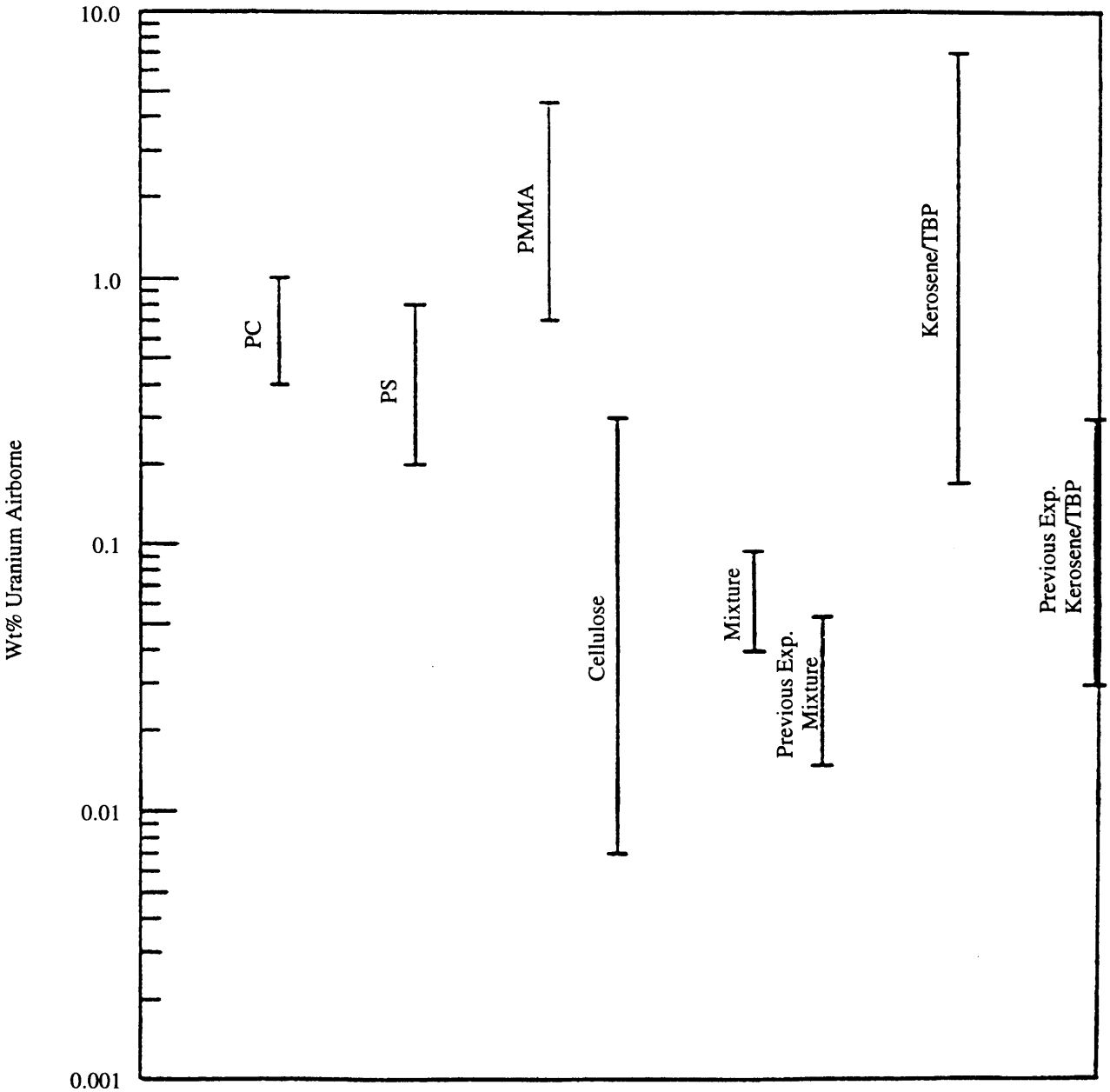

Figure 5-1. Results from Burning Contaminated Combustibles (Figure 2 - Halverson, Ballinger, and Dennis March 1987)

The pre-contaminated wastes were packaged in a plastic bags and sealed in an 18-in. x 18-in. x 24-in.-high cardboard carton. The carton was placed on a grille-like holder shown in Figure A.42 in Appendix A in a 10-ft diameter x 10-ft tall stainless steel vessel system shown in Figure A.3. The carton was ignited and allowed to burn to self extinguishment under natural convection. Fractional airborne release was determined by collecting airborne material at the vessel outlet on the ceiling. The results are reproduced in Table A.46 in Appendix A and the relevant data shown in Table 5-1.

Table 5-1. ARFs from Burning of Packaged, Contaminated, Mixed, Combustible Wastes (Table II - Mishima and Schwendiman, April 1973)

Mass U, g	U Cpd.	ARF
9.1	UO_2	5.3E-4
25.2	a.d. UNH[1]	3.0E-5
25.5	UNH[2]	8.3E-5
25.5	UNH	4.7E-5
64.1	UO_2	1.7E-4
51	UO_2/UNH	4.1E-5
25.5	UNH	3.0E-5
50.2	UO_2	5.2E-5
25.5	a.d. UNH	1.5E-4
51	UNH	1.5E-4
51.6	UO_2	1.5E-4

[1] a.d. UNH = air dried uranyl nitrate hexahydrate
[2] UNH = uranyl nitrate hexahydrate

The ARFs range from 3E-5 to 5E-4, roughly an order of magnitude. No differences for compound are discernible from the limited data set. The maximum ARF measured was 5E-4. The median value is 8E-5 with an average ARF of 1E-4.

Contamination was also measured on the floor and walls following the runs and is listed in Table A.46. As previously noted in the discussion of ARF in the Introduction to this handbook, some of the contamination was observed to come from burning fuel material directly ejected from the fire. Some of the contamination detected on the walls may also have been airborne contamination or contamination entrained in soot agglomerations that deposited during the experiments. Floor and wall deposition were not included in the ARF reported for several reasons. First, it was not possible to clearly distinguish between the remains of burning fuel ejected from the fire, which is not airborne material, and other potential sources of contamination. Secondly, any plateout in the immediate vicinity of the fire is considered a generic phenomena. Considering that many more deposition surfaces

exist around waste under most fire situations, more generic close-proximity plateout is expected for real situations. Accordingly, for all practical purposes there is no need to attempt highly uncertain phenomenological modelling to distinguish between the effects.

Based upon the above data, a bounding ARF is assessed to be 5E-4 for packaged waste. For this purpose, even waste in taped plastic bags or pails is considered packaged. In lieu of any measured size distribution for the airborne materials, a conservative RF of 1.0 is assessed to be bounding.

5.2.1.2 Uncontained, Cellulosic Waste or Largely Cellulosic Mixed Waste

The ARFs from the burning of contaminated cellulosic materials have been measured in three sets of experiments by Mishima and Schwendiman (1970) and Halverson, Ballinger, and Dennis (1987). The relevant experiments for this subsection involved very small specimens in a stainless steel shallow dish in a forced air upflow of ~ 60 mm/s, and in a shallow aluminum pan centered in a 0.18-m i.d. quartz tube. An additional set of experiments relevant to the next subsection involved an upflow of air during combustion. The results of all these experiments are reproduced in Tables A.47, A.48, A.50, and A.54 in Appendix A, and the relevant data from the three sets are summarized in Table 5-2. The experimental apparatus used by Halverson, Ballinger, and Dennis is shown in Figure A.43 in Appendix A.

Caution is necessary when interpreting the data relevant to this subsection. The airflows reported do not correspond to velocities at the actual pyrolysis gas-fuel interface where the gasflow effect is relevant. The variability in materials and contamination orientation are at least as significant. The data is neither sufficient in quantity nor sufficiently varied in parameter investigation to support characterizing results as a function of variables.

In most cases within the DOE complex, contaminated materials are multiply contained (e.g., plastic bags/wrappings inside plastic/metal/wood containers). Finding significant quantities of uncontained highly contaminated materials uncontained ex-process (storage or handling) is the rare exception rather than the rule. The experimental configuration from which these values are obtained was limited by the space available and tended to be very small (with respect to what is anticipated in facilities). Gram quantities were used rather than kilogram or multi-kilogram quantities, and these did not provide the depth of burn-residue that may attenuate airborne release from large piles of material. The radionuclides (such as plutonium solutions) were freshly applied and emphasized the goal of maximizing release. The experimental configuration did not allow the contaminant material to attain the degree of adhesion and packing expected for real stored wastes. Accordingly, the values may be very conservative for most, if not all, applications.

Table 5-2. Measured ARFs and RFs from the Burning of Contaminated Cellulosic Materials

Composition	Burn Time, min	Velocity, m/s	ARF	RF
liquid Pu nitrate contaminant (Table II - Mishima and Schwendiman October 1970)				
10-g corrugated cardboard	12.0	0.06	1.0E-2	---
10-g corrugated cardboard	6.2	0.06	5.0E-4	---
5-g cheesecloth	6.2	0.06	4.3E-3	---
5-g cheesecloth	6.7	0.06	5.2E-3	---
paper towels (Halverson, Ballinger, and Dennis March 1987)				
0.1 g DUO/g	14.0		1.88E-4	---
0.03 g DUO/g	17.0		2.39E-4	0.97
0.09 g DUO/g	33.5		3.03E-4	---
0.09 g DUO/g	11.0		4.78E-4	---
0.08 G DUO/g	10.4		5.46E-4	
0.09 G DUO/g	12.7		4.78E-4	
0.095 g DUO/g	11.4		9.03E-4	0.47
0.1 g DUO/g	7+		3.01E-3	---
0.1 g DUO/g	10.3		1.53E-3	---
0.2 g DUO/g, aero entrainment	15.8+		1.08E-4	---
a.d. UNH	9.8		2.9E-4	---
a.d. UNH	8.3		1.0E-4	---
UNH liquid	44.5		7.5E-5	---
UNH liquid	15.3		7.0E-5	---
air passes up through burning material (Mishima and Schwendiman October 1970)				
10 g tissue paper + DUO	3.7	1.04	3.4E-1	---
10 g tissue paper + DUO	6.0	0.46	3.3E-1	---
10 g tissue paper + DUO	3.5	1.04	3.8E-1	---
10 g cheesecloth + DUO	2.0	1.04	2.0E-1	---
10 g cheesecloth + DUO	2.1	1.04	3.5E-1	---
10 g cheesecloth + DUO	1.7	1.04	3.3E-1	---
10 g cheesecloth + DUO	3.2	0.46	7.0E-1	---
10 g corrugated cardboard + DUO	2.6	0.46	1.2E-1	---
	3.5	1.04	8.0E-2	---
	2.8	0.46	1.6E-2	---
	1.7	1.04	1.2E-1	---
10 g tissue paper + UNH	2.5	1.04	8.0E-2	---
10 g cheesecloth + UNH	7.7	1.04	2.0E-3	---
10 g cheesecloth + UNH	2.5	0.46	3.4E-6	---
10 g cheesecloth + UNH	3.1	1.04	5.0E-4	---
10 g cor. cardboard + UNH	13.0	1.04	1.3E-2	---
10 g cor. cardboard + a.d. UNH	7.0	1.04	2.0E-3	---
	7.0	1.04	1.0E-3	---

The ARFs from the two sets of experimental data ranged from 7E-5 to 1E-2. This represents a very significant variation by any standard. Only two values of RF are available, 0.5 and 1.0. Given that a lower value of ARF has already been assigned for burning of packaged or contained waste, which is defined to include significant compact piles of combustible waste, the value assessed for uncontained waste should be more conservative. The experimentally measured ARFs during the burning of contaminated cellulosic material are bounded by a value of 1E-2 with a median value of 5E-4 and an average value of 2E-3. The measured RFs are for the release from cellulosics contaminated by a fine, nonreactive powder. Nonetheless, the conservative measured RF value of 1.0 and an ARF of 1E-2 are assessed to be bounding. However, the potential extent of the conservatism needs to be appreciated so that the value is not cavalierly applied to the majority of burning waste circumstances inappropriately.

5.2.1.3 Dispersed Ash Dropped into Airstream or Exposed to Forced Draft Air

Air drawn through the burning material or ash falling into a flowing airstream is a more rigorous stress upon the relatively light powder formed by combustion (i.e., ash). The situation is akin to that found in a forced draft incinerator or by dropping dispersed ash into an active chimney. The experimental data shown in Table 5-2 for "air passing through burning material" show a maximum experimental ARF of 3.8E-1 with no RF measured. The maximum measured value is for the entrainment of fine, very loose UO_2 particles dropped into an accelerated airflow as fine combusting substrate lofts and disintegrates. This condition of updraft suspension is not relevant or related to the pyrolysis driven airflow phenomena of concern in the fire fuel zone examined in the previous data sets. It is also not considered representative of most expected combustion phenomena of concern. The data set examined in this subsection was intended to study maximized release phenomena. It is discrete from the other data sets discussed in subsections 5.2.1.1 and 5.2.1.2 and is treated as such.

The conditions generated in this experiment are not similar to the stress imposed by turbulent air on a pile of ash unless the velocity of the air is greater that 10 m/s at the air/ash interface, not in the general fire convective updraft. This condition is not foreseeable for even large fuel fires at nonreactor nuclear facilities. Further, the extremely small quantities of combustible material used in the experiments are atypical of combustible accumulations at nonreactor nuclear facilities, which would further minimize the chance of such extreme phenomena.

This data applies to genuinely extreme circumstances where very light combustible materials (e.g., light, individual pieces of paper) are injected into a large flame updraft where particles released by the destruction of the substrate can be immediately suspended in

convective flow. Candidates for such a release include light, individually packaged material in a non-robust container in an airplane crash fuel explosion, or possibly some small fraction of material ejected from a violent waste drum overpressurization in a waste drum fire. A critical point is that it is an airborne phenomena. Once waste lands on the ground and begins to burn in place, this phenomena is not considered applicable for any foreseeable fire conditions at DOE nonreactor nuclear facilities.

For the case of an overpressurized drum, the fraction of total material that fits this requirement is small. The material must be ejected from the container in a direction attuned with the flame, be heavy enough to cross the convective flow lines but light enough to suspend momentarily in the flames, and be of such configuration that the material will ignite and substantially combust in the time frame that it is in the flames. Even upon loss of lids due to overpressurization, sealed waste drums would not eject the material violently enough or in the correct direction to eject a large percentage of the contents in a direction attuned with the flames. Typical individual pieces of contaminated combustibles are of sufficient mass to pass through the flames with only localized ignition, let alone complete combustion. Accordingly, this release fraction is not treated as a reasonable expectation for the majority of waste in even large jet fuel fires. For more typical stresses, such as a large combustible fire or a drum with combustible material venting around or through its lid, any use of this ARF and RF is inappropriate.

For this data covering the types of phenomena discussed, an ARF of 4E-1 with a RF of 1.0 is assessed to be bounding for lightly scattered powder contamination. A bounding ARF of 8E-2 with an RF of 1.0 is applicable to air-dried solution or more adherent powders, such as those which were damp at the time of entrainment or which have been smeared onto the combustible surface. In both cases, the RF is a conservative value in the absence of measured data.

5.2.1.4 Uncontained Plastics/Elastomers

Experimental ARFs are available for three materials - polychloroprene (PC), polystyrene (PS) and polymethylmethacrylate (PMMA). The results for the individual materials is presented below but, due to the material and ARF range similarity, the ARF and RF for polychloroprene and polymethylmethacrylate are combined.

The bounding ARFs and RFs for this class of materials under thermal stress are divided into two groups - polychloroprene and polymethylmethacrylate, and polystyrene - by the measured values. Based upon the experimental data, bounding ARF and RF values for the PC + PMMA group are assessed to be 5E-2 and 1.0. The bounding ARF and RF value for polystyrene is assessed to be 1E-2 and 1.0.

5.2.1.4.1 Polychloroprene (Elastomer, Rubber). Five experiments were conducted using plutonium nitrate and rubber from surgeon's gloves (Mishima and Schwendiman, October 1970) and five experiments were performed using PC contaminated with DUO powder, UNH liquid and air dried UNH (Halverson, Ballinger and Dennis, March 1987). Experimental results are reproduced in Tables A.48, A.50, and A.51 in Appendix A and the relevant data summarized in Table 5-3. The bounding ARF for the limited data set is 4E-2 (3.5E-2 rounded upward). The single RF value, 0.16, was determined for a different set of conditions than the largest ARF value. The median value is 4E-3 with an average of 7E-3.

Table 5-3. Measured ARFs from Burning Contaminated PC (Table A.1 - Halverson, Ballinger and Dennis, March 1987, Table II - Mishima and Schwendiman, October 1970)

Burn Time, min	Composition	ARF	RF
7	5 g rubber, Pu nitrate	7.0E-4	---
9	5 g rubber, Pu nitrate	1.9E-3	---
5	5 g rubber, Pu nitrate	2.0E-4	---
7.2	5 g rubber, Pu nitrate	8.0E-4	---
6.2	5 g rubber, Pu nitrate	6.2E-3	---
7.5	DUO powder	1.0E-2	---
10.0	DUO powder	8.0E-3	---
15.7	DUO powder	3.7E-3	0.16
9.0	UNH liquid	3.5E-2	---
7.5	a.d. UNH	4.2E-3	---

5.2.1.4.2 Polymethylmethacrylate (Lucite/Perspex, Windows). Thirteen experiments were performed to measure the ARF from burning contaminated PMMA. All three forms of uranium compounds were tested using granular PMMA. One experiment measured the aerodynamic entrainment from DUO powder mixed with PMMA. Tests were performed, varying the location of the contaminant as follows: (1) powder mixed with the PMMA; (2) a thick layer of powder on the surface; and (3) a pile of powder on the surface; and (4) the powder under the PMMA. The results are reproduced in Tables A.50 and A.53 in Appendix A with the relevant data summarized in Table 5-4.

The highest measured ARF is 5E-2 (4.5E-2 rounded upward). The highest measured ARF using UNH was 2E-2. The highest RF measured, 0.95 was associated with an ARF of 1.9E-2, not far below the maximum ARF value. The single ARF measured using air dried UNH is within the range of values for UNH liquid. The median value for all ARFs is 3E-2 with an average value of 3E-2 (2.5E-2 rounded upward). The median values for DUO and UNH are 4E-2 and 1E-2, respectively. In keeping with the selection of conservative values, an ARF of 5E-2 and a RF of 1.0 are assessed to be bounding.

Table 5-4. Measured ARFs and RFs from Burning of Contaminated PMMA (Table A.3 - Halverson, Ballinger and Dennis March 1987)

Burn Time, min	Composition	ARF	RF
20.2	DUO powder, no combustion (aerodynamic entrainment)	2.5E-3	---
31.5	DUO powder	3.2E-2	---
34.6	DUO powder	3.0E-2	---
18.4	Thick layer DUO powder	3.5E-2	---
17.0	Thick layer DUO powder	3.6E-2	0.84
19.5	Pile DUO powder	4.4E-2	---
19.3	Pile DUO powder	4.5E-2	---
21.0	DUO powder under PMMA	1.3E-2	---
18.6	DUO powder	3.7E-2	---
30.3	DUO powder	1.5E-2	0.95
26.0	0.12 ml UNH liquid/g PMMA	1.9E-2	0.84
15.5	0.05 ml UNH liquid/g PMMA	2.0E-3	---
16.7	0.12 ml UNH liquid/g PMMA	2.0E-2	---
16.6	a.d. UNH	6.0E-3	0.63

5.2.1.4.3 Polystyrene (Molded Plastic, Containers, Ion Exchange Resin). Only three experiments were performed to measure the ARF during the burning of contaminated PS. Only liquid UNH was used as a contaminant. The results are listed in Tables A.50 and A.52 in Appendix A and relevant data summarized in Table 5-5.

Table 5-5. Measured ARFs and RF from Burning of Contaminated PS (Table A.2 - Halverson, Ballinger and Dennis, March 1987)

Burn Time, min	Composition	ARF	RF
18.5	0.17 ml UNH/g PS	1.6E-3	---
12.8	0.4 ml UNH/g PS	1.8E-3	---
12.8	0.5 ml UNH/g PS	7.8E-3	0.90

The highest measured ARF is 7.8E-3, with an RF of 0.9. There is no meaningful median value for such a restricted data set.

5.2.2 Explosive Stress: Shock, Blast, and Venting

Experimental data on the behavior of combustible solids, such as trash, during explosive events have not been uncovered. Based upon some general observations, the behavior of the substrate can be estimated and the associated behavior of the surface contamination inferred.

An attempt will be made to provide some train of logic for certain situations that appear to be amenable to definition.

5.2.2.1 Shock Effects

Combustible solids will undergo fragmentation by the shattering effect of detonation like other materials. Due to the extreme flexibility of some materials (thin sheets of paper, rubber, plastic), these materials may be more subject to dislodgement by shock-vibration at very short distances from the blast interface. No experimental data on the airborne release of contaminants from this phenomenon were uncovered.

The particulate contamination on the surface of the combustible material may be dislodged by the vibration of the substrate in response to the shock wave. The small mass of the particulate contamination would probably require many cycles before dislodging and the airborne release would be small. The particle attached to porous surface such as wood or paper would probably not be dislodged by fragmentation of the substrate. Accordingly, this phenomena is considered bounded by venting of pressurized gases over the material as discussed in subsection 5.2.2.3. Due to the high temperatures that may be generated and the dispersive action of the pressure impulse from the explosion, burning of the combustible material may also be a concern. This combustion would be bounded by values in subsection 5.2.1.2 for burning of uncontained waste provided containment is broken or, for very severe circumstances (i.e., light, loose waste immediately contiguous to a true detonation and entrained in shock wave), the values in subsection 5.2.1.3 for some fraction of dispersed ash dropped into a flowing airstream.

5.2.2.2 Blast Effects

Due to the extreme flexibility of most of the materials that comprise combustible wastes (e.g., sheets of tissue paper, toweling, paper, plastic, rags), the principal impact of accelerated velocity would appear to be dispersal of the material with loss by dislodgement of the contaminant by shock-vibration. Accordingly, this phenomena is considered bounded by venting of pressurized gases over the material as discussed in subsection 5.2.2.3. Due to the high temperatures that may be generated and the dispersive action of the pressure impulse from the explosion, burning of the combustible material may also be a concern. Highly pressurized waste drum releases, general area deflagrations, etc. could cause releases defined in subsection 5.2.1.2 for burning of uncontained waste provided containment is broken.

5.2.2.3 Venting of Pressurized Gases over Solids

No directly applicable data were uncovered. The entrainment of loose surface contamination is a function of the characteristics of the flow over the particulate material that may be lying on the surface, the particles, and the surface. However, as noted above, the combustible material substrate will not typically provide a rigid surface for venting gases to act upon. As noted in subsection 5.3.3.2.2 for impact/vibration releases from non-brittle materials, flexing of the substrate is the primary mechanism for aerodynamic entrainment. The ARF x RF value given for that phenomena (i.e., 1E-3) is essentially identical to the next most applicable phenomena, release from powders due to accelerated flow parallel to the surface as discussed in subsection 4.4.2.2.2 (ARF x RF = 1.5E-3). Accordingly, the ARF and RF of 1E-3 and 1.0 are assessed to be bounding.

5.2.3 Free-Fall Spill and Impaction Stress

Loss/dislodgement of surface contamination during the free-fall spill of contaminated, combustible materials would not appear to generate any significant stress upon the surface.

5.2.3.1 Free-Fall Spill

No applicable data were uncovered. In many cases the combustible materials are light and have high surface area to mass ratios (e.g. paper, cardboard, plastic sheets and wrapping) and thus would generate little force during impact with surfaces. For such materials, no significant suspension of surface contamination is postulated. In the case of material with appreciable mass, the dislodgement of surface contamination upon impact would appear unlikely but would be bounded by the bounding ARF and RF described in subsection 5.3.3.2.2 for the suspension of surface contamination by vibration-shock.

5.2.3.2 Impact

A distinction is made for impact releases as to whether the base material is packaged or unpackaged. For unpackaged materials with appreciable mass that generate significant forces upon impact with a surface (forces adequate to result in dislodgement of the surface contamination), the bounding ARF and RF for the suspension of surface contamination from shock, 1E-3 and 1.0, described in subsection 5.3.3.2.2 are assessed to be bounding.

For the situation where the combustible material is packaged in a reasonably robust container (e.g., drum) that fails due to impact, the bounding ARF and RF assigned are 1E-3 and 0.1. This value is the same as that assigned to suspension of powder in a can due to debris impact (subsection 4.4.3.3.2), which bounds this phenomena.

5.2.4 Aerodynamic Entrainment and Resuspension

The value for the aerodynamic entrainment (resuspension) of powder lying on a heterogeneous surface under nominal flow condition indoors or outdoors has been assigned a value of 4E-5/hr with a RF of 1.0 (section 4.4.4). For thin layers of powders lying on the surface of a heterogeneous surface under debris or for static conditions within facilities, the ARR is degraded to 4E-6/hr with a RF of 1.0. It is not anticipated that the resuspension of contaminants for solid, combustible wastes would exceed these values under the same conditions. On these bases, these values are also assessed as bounds for the ARR and RF for the resuspension of surface contamination from combustible solids.

5.3 SOLID, NONCOMBUSTIBLE UNYIELDING SURFACES

The surface contamination on solid, noncombustible surfaces is assumed to be a sparse population of loose (not combined with the surface matrix) particles lying on the heterogeneous surface. In actual cases, surface contamination can range from contamination mixed with a worn, abraded surface (e.g., rust, concrete) to materials chemically attached to the surface.

5.3.1 Thermal Stress

The flexing (expansion and contractions) of metal and other noncombustible surfaces may eject some particles contaminating their surfaces. The experimental data that most closely represent such situations would be the suspension of powders during heating and oxidation (see section 4.3.1). Due to the uncertainty of the composition of the contaminant (may be a chemically active compound in powder form), the higher value for heating non-reactive compounds in powder form, ARF and RF of 6E-3 and 0.01 (Mishima, Schwendiman and Radasch, July 1968), are assessed to bound these situations.

5.3.2 Explosive Stress: Shock, Blast, and Venting

5.3.2.1 Shock Effects

5.3.2.1.1 Contaminated Solids as Monoliths. Steindler and Seefeldt (1980) provide an empirical correlation to experimental data on the fragmentation of metals and aqueous solution by detonations [energy releases in microseconds with brisance (shattering effect)] (Ayer, et al., May 1988). The experiments covered the work performed by TNT related to the mass ratios (ratio mass of inert to TNT equivalent) of 1 to 10. The experiments were conducted with the condensed phase explosive embedded or contiguous to the material affected. Estimates of the ARF and size distribution for various mass ratios up to 1000 are provided in Appendix C of Ayer, et al. (May 1988) for a GSD of 8. The GSD is much

greater than normally assumed (GSD 2) and provides greater fractions in the larger size ranges (a nonconservative assumption for the assessment of radiological impacts). Due to the rapid change in size distribution, the maximum mass of inert material airborne in the respirable fraction is for an MR of 1. Therefore, a bounding ARF and RF of inert material equal in mass to the TNT Equivalent for the detonation are assumed. This correlation previously used for solids and liquids is considered bounding for concrete forms as well. The amount of surface contamination driven airborne as a respirable aerosol will be the amount of contamination associated with the mass of inert material driven airborne. This association may be on a per volume or surface area basis. The depth or area fragmented by the detonation must be estimated based on general explosive information in texts or specific data.

5.3.2.1.2 Contaminated Soil. For soil or soil-like powders (aggregated, compacted powder), the ARF and RF assessed in subsection 4.4.2.1 for suspension of powders from shock effect (ARF mass of material suspended is calculated 0.8 TNT Equivalent for the explosion, RF 0.25) may be applied. The amount of the radioactive contaminant made airborne is estimated by multiplying the mass of soil/powder airborne by the concentration of the contaminant (Ci/g).

5.3.2.2 Blast Effects

No experimentally measured values for ARF and RF are available. Potential releases for loose surface contamination on metal are bounded by the assessed values for venting pressurized gases over metal given in the following subsection.

5.3.2.3 Venting of Pressurized Gases Over Solids

Release of pressurized gases over the solid could suspend material if the surface of the solid were covered with loose oxide (corroded) and the aerodynamic profile over the surface possessed the characteristic to entrain the oxide. For loose, externally deposited surface contamination, there are two mechanisms defined for powders that can be used to assess this situation. The first is if the venting gas source does not significantly pressurize the confinement in which the contamination exists. In this case, the ARF and RF of 5E-3 and 0.3 for accelerated flow parallel to powder on a surface given in subsection 4.4.2.2.2 would be an appropriate bound.

The second case is when the venting gases pressurizes the volume in which the contamination exists. The ARF and RF values (5E-3 and 0.4 for pressure < 0.17 MPa_g, 1E-1 and 0.7 for greater pressures) determined in subsection 4.4.2.3 to bound the venting of pressurized

powders would certainly bound material in this configuration. In either case, the ARF and RF values are only applied to the loose surface contamination, not the base solid quantities.

5.3.3 Free-Fall Spill and Impaction Stress

No applicable measured data were uncovered. No significant impacts are anticipated for material that will undergo plastic deformation (e.g., metal, plastics, wood) during free-fall or upon impact with a hard, unyielding surface. Any suspension of surface contamination is considered bounded by the bounding values assessed for suspension by shock-vibration. For materials that undergo brittle fracture (e.g., aggregate, glass), no suspension is anticipated during free-fall but suspension can occur at impact.

5.3.3.1 Free-Fall Spill

No significant suspension is anticipated; no data were available. The value assessed for crush-impact will bound the phenomena.

5.3.3.2 Impact

5.3.3.2.1 Solids That Undergo Brittle Fracture. If the solid can brittle fracture (e.g., aggregate, glass) due to crush-impact forces, the activity present on the surface may be carried by the fragments. The fragmentation of solids into particles in the respirable size range was discussed in section 4.3.3. The fraction is calculated by:

$$ARF \times RF = (A)(P)(g)(h) \qquad (5\text{-}1,\ 4\text{-}1)$$

where: ARF x RF = (Airborne Release Fraction)(Release Fraction)
A = empirical correlation, 2E-11 cm^3 per g-cm^2/s^2
P = specimen density, g/cm^3
g = gravitational acceleration, 980 cm/s^2 at sea level
h = fall height, cm.

The amount of surface contamination associated with the fraction of base material driven airborne is assumed to be released.

5.3.3.2.2 Solids That Do Not Brittle Fracture. Solids that experience predominantly plastic deformation (e.g., metal, plastics) as opposed to brittle fracture, respond to vibration and shock of the substrate by flexing. Materials adhering to the surface are ejected by the movement depending on how the contaminant is attached to the surface. The only experimental data that may have any relevance are those for aerodynamic entrainment from surfaces and the dislodgement mechanisms are different. The bounding

ARF assessed for suspension from a pile of powder from a hard, unyielding surface due to accelerated flow generated by an explosion parallel to the surface is 5E-3 with an RF of 0.3 (see subsection 4.4.2.2.2). The stresses upon the surface contaminant, much of which is embedded in the surface, appear to be fewer than those described for powders on unyielding surfaces. The stresses are certainly greater than those assumed for the resuspension of materials from nominal airflow in facilities or outdoors, 4E-5/hr. An ARF of 1E-3 with a RF of 1.0 was selected for this phenomenon by Mishima, Schwendiman and Ayer (October 1978) in an analysis of severe natural phenomena effects. The ARF value is 20% of the value assigned for the suspension of loose powders by explosion generated accelerated flows parallel to the surface and more than an order of magnitude greater than values assigned for resuspension. On these bases, an ARF of 1E-3 with a RF of 1.0 is assessed to bound the suspension of surface contamination from non-brittle solid material for this phenomena.

5.3.4 Aerodynamic Entrainment and Resuspension

The parameters governing the suspension of particles from a heterogenous surface (e.g., metal, some plastic, concrete, glass) are the same as for suspension from a homogeneous bed; the parameters are the characteristics of the flow, particles and surface. The effects of the various factors that contribute to the parameters (e.g., aerodynamic lift forces, drag forces, adhesive forces) vary greatly. Figure 5-2 reproduced from Brockman (February 1985) shows the effect of particle size on various adhesive forces. Figure 5-3 reproduced from Fromentin (January 1987) illustrates the effect of surface roughness on the suspendability of small particles. Adhesion decreases with substrate surface roughness until the macroroughness becomes the same size as the particles, when it increases rapidly (Hubbe, 1984). The surface roughness of the particles, the presence of moisture, the plasticity of the surface (Johnson, Kendall and Roberts, 1971), and other factors all appear to affect the adhesion of particles to substrates or to each other.

Current consensus assumes that flow must be turbulent before significant suspension occurs (Fromentin, January 1987). This is not necessarily the case for aerodynamic entrainment in facility ventilation flow or outdoors at windspeeds less than 5 m/s. The turbulent flow is divided into three regions: 1) core; 2) transition; and 3) viscous sublayer with regions two and three comprising the wall region (Alonso, Bolado and Hontanon, July 1991). Present consensus also agrees that turbulent bursts (intermittent ejections of discrete fluid elements from wall region towards the core) play some role in suspension (Cleaver and Yates, 1973). The burst process is composed of three steps: 1) deceleration of axial fluid velocity within local region near wall, 2) progressive acceleration from approach of fluid with mean velocity, and 3) before affected region totally accelerated, ejection of fluid from region of unaccelerated fluid. The process is shown schematically in Figure 5-4.

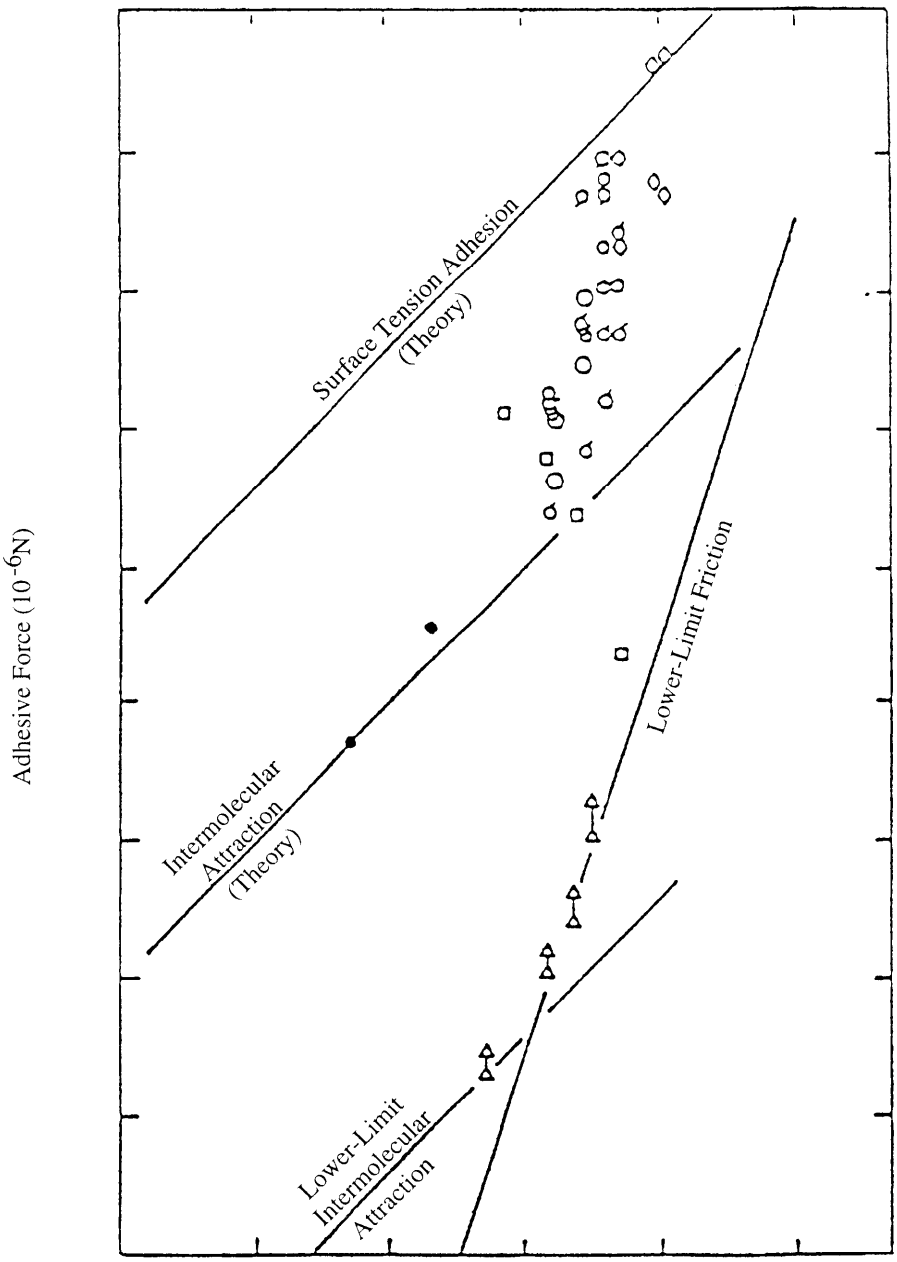

Figure 5-2. Variation in Adhesive Forces Observed for Various Size Particles (Brockman February 1985)

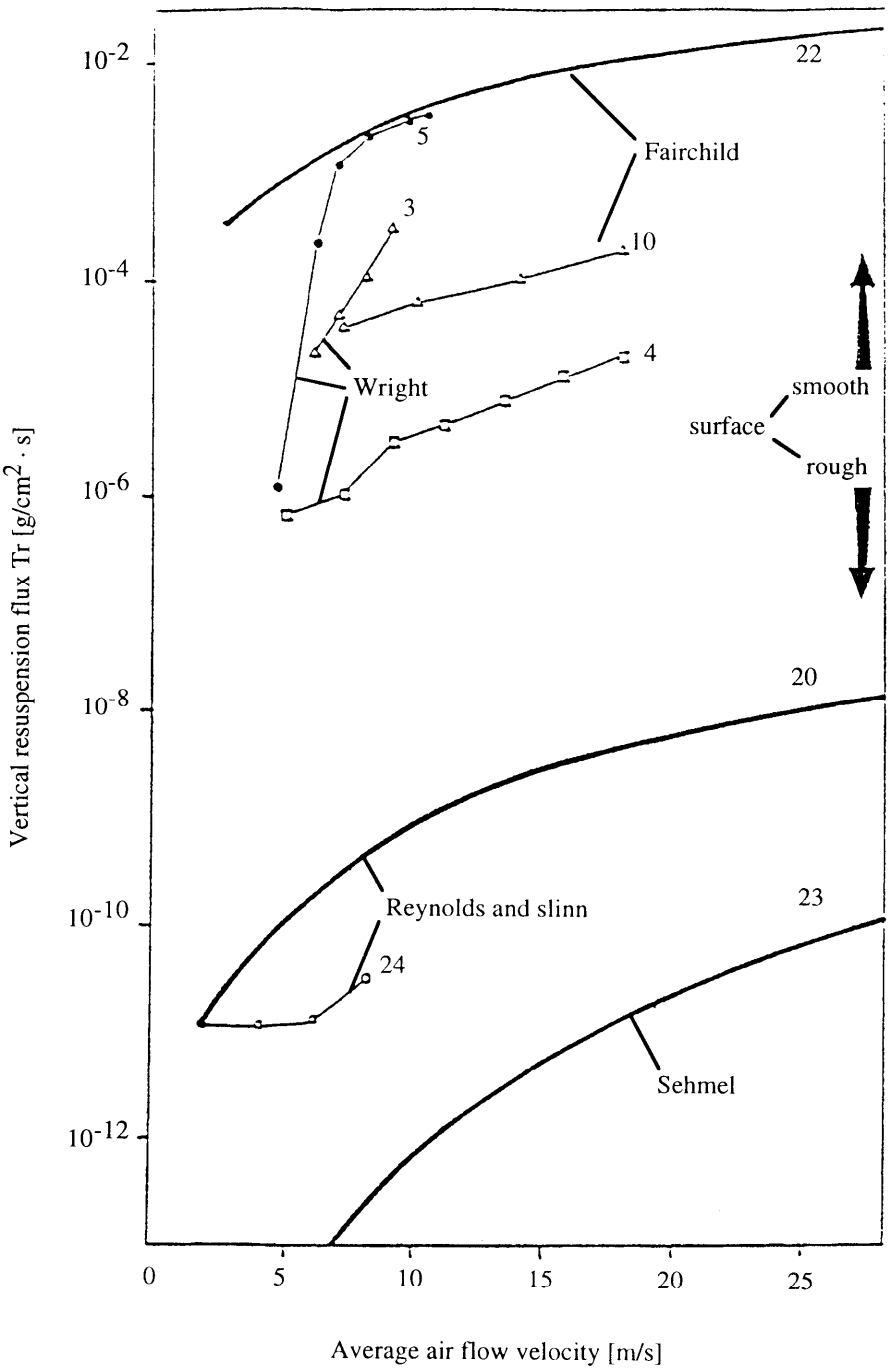

Figure 5-3. Resuspension Flux F_r Air Flow Velocity for Particles Smaller Than 10 Micrometer Diameter (Fromentin January 1987)

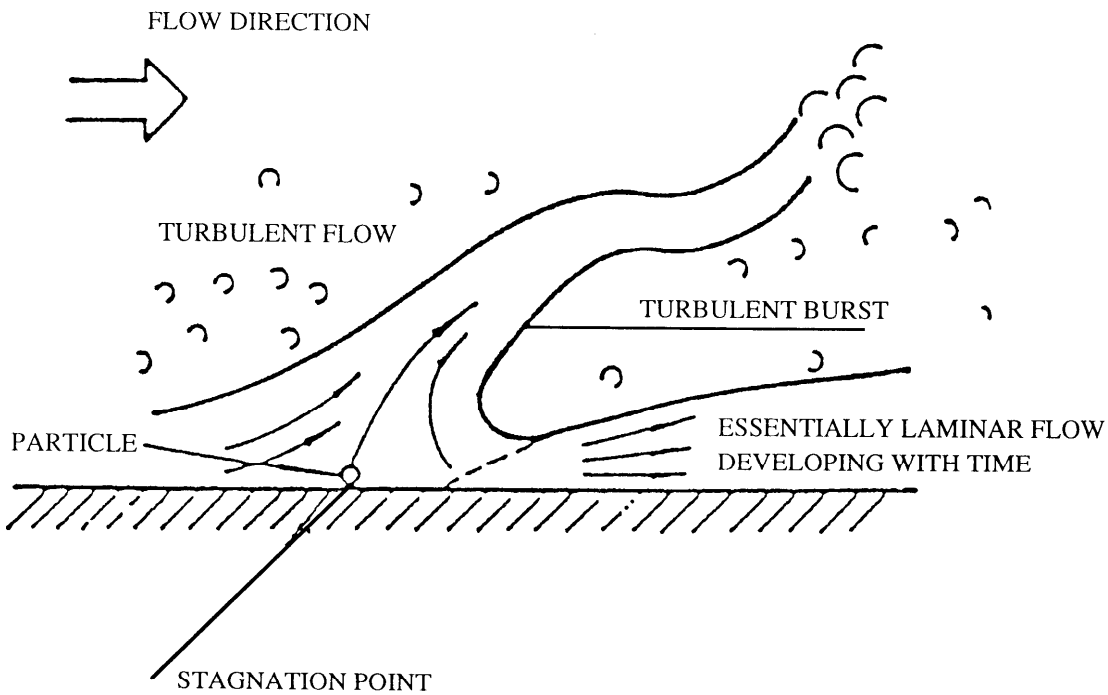

Figure 5-4. Schematic Diagram of Turbulent Burst in the Wall Region (Fromentin January 1987)

There are two general conceptions for aerodynamic entrainment - force balance and energy balance. The force balance concept is that aerodynamic lift/drag forces exceed the adhesive forces causing particles on the surface of the substrate to be suspended. Most authors have subscribed to the force balance concept. Recently, Reeks et al. (May 1985a, May 1985b, June 1985a, June 1985b, 1988) have proposed an energy balance concept that postulates resuspension of small (<100 µm diameter) particles by energy transfer from turbulent flow. The concept is similar to removal of molecules from surfaces by Brownian motion. Energy maintains the particle in motion at the surface within the surface potential well. Particles are suspended when they accumulate sufficient vibrational energy to escape the potential well.

Authors (Reeks et al., May 1985a, Wright, 1984) have reported suspension of large fractions of deposited particles during the buildup to test velocities. Wright (1984) reports that the initial suspension of 95% of the deposited particles on the floor of a wind tunnel during an air velocity increase to 60 m/s (~ 135 mph). Braaten, Shaw and Paw U (1986) reported negligible loss during an increase to 20 m/s in a wind tunnel. John, Fritter and Winklmayr (1991) found some suspension of 8.6 µm sodium fluorescein particles when filtered air at 40 m/s from a nozzle was impinged upon the Tedlar surface. The point may be moot for the conditions covered here since the material is continually subjected to at least some air flow during deposition and any suspension occurring in the initial seconds would be lost with the release from the occurrence. Garland (1982), Corn and Stein (July 1965), Cleaver and Yates (1973) and Wright (1984) found resuspension fluxes inversely proportional to time (decayed with time) while Sehmel and Lloyd (1976) did not find any apparent decay over a 21-month test period.

A vast array of literature exists on various aspects of the resuspension of sparse particles from the surface of substrates (Corino and Brodkey, 1969; Corn, 1966; Dahneke, January 1975; Ettinger, 1974; Fairchild, 1982; Fish, 1967; Hall, 1988; Hubbe, 1984; Larsen, August 1958; Nelson, 1973; Punjrath, 1972; Spenser, 1976). Zimon's (1969) text book provides a good theoretical background of the topic. More recently several reviews (Brockman, February 1985; Fromentin, January 1987; Alonso, Bolado and Hontanon, July 1991) directed towards the dry resuspension of particles in light water reactor coolant systems during the depressurization of the containment following a severe core disruptive events have been published. In general, most of the experimental data have been obtained on sparse particle suspension from heterogeneous substrates with few field studies that is the converse of the situations for suspension from homogeneous beds covered in section 4.4.5. Fromentin (January 1987) concluded from his review that:

- Both theoretical and experimental studies of the suspension of solid particles from solid surfaces are still in an elementary state.

- There is still controversy over which basic resuspension concept to apply - force balance or energy balance.

- Due to an incomplete understanding of the turbulent boundary layer, and more precisely of the bursting phenomenon, uncertainty exists as to the removal forces acting on particles.

- Some very important effects such as particle agglomeration on the surface (alters the size of the particles to be suspended and raise questions as the deagglomeration in flow), saltation (role of energy transfer from impacting particles on suspension), and initial suspension have not been well studied.

- The wide range of values for adhesive forces and the lack of models to explain influence of roughness (both substrate and particle surface) results in great uncertainty.

A few field studies have reported the suspension of particles from heterogeneous substrates. Garland and Pigford (1992) reported a resuspension factor (K) of 1E-6 for fallout from Chernobyl from the Harwell parking lot. If a sampling period of 24 to 72 hours is assumed, the resuspension flux would be 1 to 4 E-8/hr. Sutter (1982) lists Ks reported for Bennett (1976) of 2E-6 to 5E-9 for New York City and the United Kingdom and 6E-6 to 8E-9 for paving stones. These are in the same range or less than reported by Garland and Pigford (1992). Sutter reported K values for the study of resuspension of plutonium as oxide powder or dried nitrate solution from various floor covering (Jones and Pond, 1967) ranging from 1E-10/hr to 1E-8/hr assuming an 16-hr sampling period for air movement only. The values appear to be consistent with the bounding value assessed for homogeneous beds of powder of 4E-5/hr under process facility ventilation flow conditions and nominal outdoor conditions. For particle deposits shielded from the ambient flow by debris or in a static (no induced flow) facility, a resuspension flux an order of magnitude less, 4E-6/hr, is assessed to be bounding.

Therefore, the ARR and RF values assessed to be bounding for aerodynamic entrainment (resuspension) of surface contamination (sparse population) from a heterogeneous (hard, unyielding) surfaces are:

- Indoors or outdoors exposed to ambient conditions (normal process facility ventilation flow or less, or nominal atmospheric windspeeds < 2 m/s with gusts up to 20 m/s) following an event: ARR 4E-5/hr; RF 1.0

- Buried under debris exposed to ambient conditions or under static conditions within the structure following an event: ARR 4E-6/hr; RF 1.0

5.4 HEPA FILTERS

5.4.1 Thermal Stress

The ARF from the heat-induced damage to a HEPA filter is estimated to be very small. HEPA filters resisted temperature as high as 825 ºC for period of tens of minutes before loss of efficiency and 500 ºC for in access of 45 min (Hackney, 1983). The filter medium is very fine diameter glass fiber that softens and melts when heated and thus, tends to retain materials adhering to the fibers. The release rate for several types of HEPA filter in flowing air at elevated temperatures less than required to induce failure (up to 400 ºC) are very low (Ammerich et al., 1989).

HEPA filters, both unused and removed from service due to high differential pressures (clogged), were tested using solid particles at a range of temperatures less than required for failure. The efficiencies of the filters prior to testing for 1.8 µm particles ranged from 99.97% to 99.9999989%. Two high flow (2000 cfm) and one 1000 cfm HEPA filters with glass fiber media and various sealant and gasket materials were tested. No releases were found at temperatures below 150 ºC (175 ºC for one of the high flow filters). For the 1000 cfm type filter, the release rates for temperatures from 175 ºC and 190 ºC started at 1E-6/min and reduced to 5E-8/min within 1 hour (the lower limit of detection was 2E-8/min). The high flow HEPAs were tested to temperatures of 200 ºC and 250 ºC with release rates starting at 2E-4/min and 2E-5/min and reducing to 3E-7/min in 30 min and 2E-8/min in 60 min. The decay in release was exponential during the initial 30-minutes approaching the 60-min rate asymptotically. There was no release of contamination from a oven-fired, mineral sealant, high flow type filter at temperatures up to 350 ºC and the release in other types of HEPA filters is associated with the emission of smoke (binder, degradation of inert dust on filter, pyrolysis of gaskets). Thus, it appears that the heat-induced release from 1000 cfm HEPA filter prior to failure may be as high as 1E-5. It is assumed that HEPA filters destroyed by flame intrusion or by the impact of air at a temperature sufficiently high to melt the glass fiber are subjected to high temperature air to result in the release given above for heat-induced release. The RF is assumed to be 1.0 without an experimental basis. ARFs for high-flowrate HEPA filter may be an order of magnitude higher (1E-4). On these bases, bounding ARF and RF values for the impact of heat upon loaded HEPA filters are assessed to be 1E-4 and 1.0.

5.4.2 Explosive Stresses: Shock, Blast, and Venting

5.4.2.1 Shock Effects

Very limited experimental data have been reported on the release of collected contamination as a result of the failure of glass fiber media HEPA filters from a dynamic pressure pulse. Gregory et al. (February 1983) reported the release of 0.341, 1E-6 and 2E-6 of the collected solid particles for high-flow HEPA filter during explosive transient. The large release from the first filter was attributed to a medium that was slightly creased prior to the test and appears to be an artifact of the test equipment. All the filters appeared to have vented by blowout of the filter media at selected locations along the rear folds of the pleated filter mat. The areas for the other two filters vented were very small. The total collection area of a HEPA filter is approximately 1.8 X 10^5 cm^2. If all the collected contaminant in the area affected by the blowout is approximately 0.1 cm^2 (a slit 1 cm long X 0.1 cm wide) and the contaminant collection efficiency in these area is the same for the rest of the filter, the ARF would be ~ 5E-7. Dislodging all the contaminant is difficult since much of it penetrates into the medium. Furthermore, the affected area would appear to be the depth of medium exposed by the rupture of the glass fibers. The test results appear to be consistent with this type of phenomenon. Thus, it would be anticipated that the amount of the deposited material subjected to the mechanical stress of the filter rupture is a minute fraction of the nearly 200 ft^2 filter mat. Only during the initial stages of use would a large fraction of deposited material be located in the rear fold. Thus, if the high value is discounted, ARF and RF value of 2E-6 and 1.0 bound the data.

5.4.2.2 Blast Effects

Gregory et al. (October 1983) also performed tests on from standard (1000 cfm) glass fiber media HEPA filters. The break pressure from static pressure loading for standard HEPA filters ranged from 9.1 to 20.0 kPa with a mean value of 16.3 kPa. High flowrate HEPA filter demonstrated lower structural capacity with break pressures ranging from 9.0 to 15.9 kPa with a mean value of 11.0 kPa. Removal efficiency of the standard HEPA filters for 0.46 µm diameter polystyrene latex beads in tornado conditions degraded from the 99.97% for filter under normal conditions to 98.9%. The release of the approximately 1 kg of 0.46 µm diameter PSL collected for one type of standard HEPA filters was 1.46% and 0.71% with a mean value of 0.935%. Bounding ARF and RF values are assessed to be 1E-2 and 1.0.

5.4.2.3 Venting of Pressurized Gases Through Filters

See subsections 5.4.2.1 and 5.4.2.2.

5.4.4 Free-Fall Spill and Impaction Stress

Material accumulated on the filter media could be dislodged by the jarring of the media/filter by a single severe shock (e.g., earthquake, impact of heavy object such as aircraft engines, forklift) or by mechanical vibration. In the case of a severe shock, it is assumed that the housing holding the filter banks would also be compromised and material made airborne both in and out of the housing are of concern. The media is a relatively fragile material with considerable flexibility and dislodging only the accumulated particles does not appear to be consistent with experience. Rather, the fragmentation of the media by the vibration/shock appears to be principal mode for particle generation as shown in Table A.55 from Arnold (1986) where filters without accumulated Pu (although other inert, airborne materials may be collected) release the same fraction of particles as loaded filters within each filter set.

Arnold (1986) reported on a series of studies conducted at the Rocky Flats Plant on the particles generated by HEPA filters (large plenum sized filters, 2-ft x 2-ft x 1-ft), and filters used to seal the exhaust outlets on gloveboxes (8-in x 8-in x 6-in and 12-in x 12-in x 6-in) subjected to repetitive slamming on a hard, unyielding surface and mechanical crushing. In one study, 30 plenum-sized filters, wrapped in two layers of plastic, were mechanically crushed and approximately 0.75 lb of material in an outer box holding the filter during transport were collected. Less than 5 wt/o of the collected material was in the size range of <200 µm (LLD, sieves used) and <0.5 wt/o was <25 µm. All the filter media disintegrated under test conditions for filter from plenum FU2B (from a recovery facility where strong acid vapors are present in the effluents) and the particulate weight listed is that for all the media. The Matrix Weight listed in that table for filters from other plena is the total weight of the filters and ranges from 13,190 g to 24,380 g. On that basis, the media represents from 5.3% to 12.2% of the total filter weight. The average weight of a plenum sized filter in this study was given as 17.613 kg (the average weight of plenum sized filters was 18.18 kg and 18.1 kg in other studies) and the total weight of 30 plenum sized filters would be 528.39 kg. The total mass of particles collected, 255 g, represents 0.0483 % of the total filter weight. If it is conservatively assumed that the total media mass is 5% of the total filter weight, the particles collected represent 0.97% of the filter media mass and the fraction <25 µm LLD is 0.005% of the filter media mass.

Thirty plenum-sized filters were crushed in a hydraulic press and a total of 34.1 g of particles collected. The total filter weight was 528.29 kg with a media mass of 26.42 kg under the assumption that the media mass is 5 wt/o of the total filter mass. Thus, the particles represent 0.13 wt/o of the total media mass. The size distribution of particles generated was found to be 8.525% <200 µm with no measurement of the 10 µm fraction. In the other portions of this study, the 10 µm fraction was no greater than 10% of the <200 µm fraction and on this basis, the fraction 10 µm and less would represent 0.0013 wt/o of the media mass.

In another study, five plenum-sized filters were tapped and crushed and a total of 349 g of particulate materials collected. The total weight of the filters was 90,900 g and the material generated represents 0.384%. Under the assumption that the media mass is 5% of the filter weight, the particles collected represent 7.68% of the media mass. The fraction 10 µm and less was 9.261 wt/o of the total collected and 0.71 wt/o of the filter media mass.

Two different glovebox-sized filters were also studied in this effort and the same test conditions. A total of 9 g was collected with a total filter weight of 41.4 kg. The particles collected represent 0.022 wt/o of the total filter mass and, under the assumptions used previously, 0.43 wt/o of the filter media mass. The size distribution of the particles generated was not characterized but in all other studies reported was <10% (in most cases much less). Under the assumption the 10 µm fraction is 10%, the particles generated represent 0.043 wt/o of the media mass.

The data reproduced in Table A.55 are from a study where plenum-sized filters from a variety of plena were wrapped in plastic and projected 3-ft repeatedly onto an unyielding concrete floor. Table 5-6 presents relevant data from Table A.55 taken from the referenced document. For the fraction 10 µm in diameter and less generated based upon the assumed total media mass, the bound is 5E-3 with a median of 2E-3 and an average of 2E-3. This is consistent with the results above that indicate releases of particles 10 µm in diameter and less ranging from 1E-5 to 7E-3.

5.4.4.1 Enclosed Filter Media

Filter media is generally contained within multiple enclosures. The filter housings themselves are enclosures, and in-use filters are contained in plenum enclosures that are substantially more robust than the plastic wrap holding filters in the data cited. Used filters are generally held in plastic wrapping inside waste drums. Even severe shock-impact for these configurations is no worse than the mechanical crushing performed on the filters examined. It is assumed that the fraction of particles 10 µm diameter and less released from the double-wrapped HEPA filters that were crushed bounds the release from enclosed HEPA filter media subjected to crush-impact stresses that compromise the enclosure.

For the first test with 30 filters, the total mass of particulate obtained <25 µm LLD was 5E-5 as a fraction of overall media weight. In the second crushing experiment with an additional 30 filters, the fraction <10 µm was estimated to be 1E-5. The majority of this material would not be expected to be filtered radioactive material. However, due to the great uncertainties associated with the test results, an additional order of magnitude increase is imposed resulting in bounding ARF and RF values of 5E-4 and 1.0 for this stress.

Table 5-6. Fractions of Media Mass as Particles 10 µm and Less Generated by HEPA Filters Under Shock/Vibration Stresses

Wt. Pu, g	Wt. Particles 10 µm and less[3], g	ARF[2]
FU2B Plenum, Pu Recovery Facility, all filter media disintegrated upon being subjected to mechanical stress, ARF[2] based upon Wt. Particles 10 µm and less divided by Wt. Particulate (g) that is the media weight.		
43	<6.0	<5.0E-3
38	<6.6	<4.0E-3
41	<6.3	<4.0E-3
39	<5.6	<4.0E-3
45	<6.2	<4.0E-3
35	<6.0	<4.0E-3
42	<5.9	<4.0E-3
46	<5.2	<4.0E-3
FU2B-2, second stage HEPA filter from FU2B Plenum		
6	<0.9	<1.0E-3
7	<0.3	<3.0E-4
13	<0.3	<3.0E-4
12	0.4	4.0E-4
Incinerator		
1	1.3	1.0E-4
0	0	2.0E-3
2	1.5	2.0E-3
1	1.6	2.0E-3
1	3.1	3.0E-3
0	0	0
2	<1.4	<1.0E-3
1	2.4	2.0E-3
202 Plenum		
0	<1.1	<1.0E-3
1	<2.4	<3.0E-3
1	<1.4	<2.0E-3
0	<0.9	<1.0E-3
0	<1.4	<2.0E-3
1	0.3	3.0E-4
1	<2.3	<3.0E-3
0	<0.2	<2.0E-4

Table 5-6. Fractions of Media Mass as Particles 10 µm and Less Generated by HEPA Filters Under Shock/Vibration Stresses (Cont.)

Wt. Pu, g	Wt. Particles 10 µm and less[3], g	ARF[2]
41 Plenum		
1	0	0
1	0	0
0	0.8	9.0E-5
0	0	0
0	0	0
0	<0.04	<5.0E-5
0	0.02	2.0E-5
0	0	0
321 Plenum		
0	<0.1	<2.0E-4
0	1.0	1.0E-3
0	0	0
0	0	0
0	0	0
0.02	0	<2.0E-5
0	0	0
0	0.1	1.0E-4

[1] Back calculated from stated fraction 10 µm and less on Table A.55.
[2] Media Airborne Release Fraction <10 µm. Based on media mass = 0.05 X total filter weight.

5.4.4.2 Unenclosed Filter Media

The fractions of particles 10 µm in diameter and smaller generated by crushing or repetitive jarring of spent, plenum-sized HEPA filter ranged from 1E-5 to 7E-3. From the data in Table A.40, it would appear that the response of the filter is dependent on previous service that appear to somewhat correlate to radioactive material loading. Under the assumption that the particles generated represent the fragmentation of the filter media and that the surface contamination remains associated with the filter media during this process, the bound value under the study conditions was 7E-3 (particles generated by tapping and crushing plenum sized filters). Conservative ARF/RF values for the phenomenon are 1E-2/1.0 and include glovebox sized filters. This value is intended to be applied to HEPA filter media with no enclosure or for which the enclosure has been totally destroyed and the filter media widely scattered and impacted. This is not a typically foreseeable condition.

6.0 INADVERTENT NUCLEAR CRITICALITY

6.1 SUMMARY OF BOUNDING RELEASE ESTIMATES

Under appropriate accident conditions, fissile and fissionable radionuclides may undergo a self-sustaining nuclear reaction (chain reaction) called an inadvertent nuclear criticality. The initial airborne release from a nuclear criticality is estimated by use of relevant factors of the five-component linear equation used to estimate the airborne release from other events covered in the previous chapters. However, because the evaluation of nuclear excursions is a complex process, some additional topics used in the equation are discussed below.

For nuclear criticalities, the MAR is determined by the fraction of fission products generated by the criticality and the fraction of the fissile/fissionable material that may be suspended by the event generated conditions (primarily heat) [Note: since fissile materials [^{233}U, ^{235}U, ^{239}Pu] are also fissionable, both will be referred to as fissionable]. The amount of fission products and actinides produced by the excursion is a function of the total fissions from the criticality and the specific fissionable radionuclide involved. The fraction that is at-risk of airborne suspension depends upon the physical form of the fissionable materials involved. Estimation of fission product quantities can be done with computer codes, such as ORIGEN2, or by simple ratios (total fissions for scenario/1E+19 fission) to values in NRC regulatory guides prepared for fuel cycle facilities.

The airborne release from nuclear excursions in various physical systems can be estimated for general purposes using the equations which follow. Unless otherwise noted, all respirable fractions are equal to 1.0 and are not specifically mentioned. The physical systems considered are: (1) solutions; (2) fully moderated/reflected solids; (3) bare, dry solids; and (4) large storage arrays. Conservative fission yields are assigned to generate maximum quantities of fission products, but physical effects to the fissionable material have not been evaluated to the same degree of conservatism to avoid implying the need for analytical calculations of severity exceeding a considerable historical record and contrary to generally accepted consensus. Meaningful enhancements in criticality safety are more appropriately accomplished through required criticality safety evaluations in accordance with guidelines such as DOE-STD-3007-93, "Guidelines for Preparing Criticality Safety Evaluations at Department of Energy Nonreactor Nuclear Facilities," than by speculative consequence modelling.

Solutions

Solutions refers to fissile materials dissolved in a liquid solvent, typically aqueous acid solutions. The release of volatile radionuclides generated by the criticality is evaluated separately from the release of nonvolatiles and fissile material originally present in the solution. The criticality is generically considered terminated by the evaporation of 100 liters of water, or some lesser amount if less than 100 liters are present.

$$\text{Airborne Source Term} = (MAR_{c1} \times DR_{c1} \times ARF_{c1}) + (MAR_{s1} \times DR_{s1} \times ARF_{s1}) \quad (6\text{-}1)$$

where:

MAR_{c1} = inventory of gas and volatile (i.e., iodine) radionuclides generated from criticality in a solution calculated by computer code such as ORIGEN2 based on 1E+19 total fissions; or, [1E+19 fissions/1E+19 fissions] X table.[1]

DR_{c1} = damage ratio for gases and volatiles (i.e., iodine) generated in criticality, 1.0.

ARF_{c1} = Airborne Release Fractions for solution criticality in NRC Reg Guides[2]: 1E+0 for noble gases,
2.5E-1 for radioiodines.

MAR_{s1} = inventory non-volatile fission products generated and radionuclides in solution prior to the nuclear excursion evaporated, curies.

DR_{s1} = Damage Ratio radionuclides in solution, 1.0.

ARF_{s1} = Airborne Release Fractions for solution from NRC Reg Guides[2]: 5E-4 for non-volatiles in 100 liters of solution, 1E-3 for radioruthenium in fuel reprocessing solutions.

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[1] Tables from Reg. Guide 3.33 (Spent Commercial Nuclear Fuel solution), 3.34 (uranium) & 3.35 (plutonium) "Radioactivity of Important Nuclides Released From the Criticality Accident ..." reproduced in this document (Tables 6-7, 6-8, and 6-9).

[2] ARF (Airborne Release Fraction) for:
fission product noble gases = 1.0
radioiodines that can be generated as vapors by accident conditions = 0.25
radioruthenium present in fuel reprocessing solutions = 1E-3
salts (i.e., non-volatile fission products and base solution) in liquid evaporated (assume material in 100 liters for large volume solution excursion or total amount of material for solutions with less than 100 liters) = 5E-4

Fully Moderated/Reflected Solids

This configuration covers reflected bulk metal and metal pieces or solid fines, such as powders, that are moderated or reflected. The equation assumes some softening and local deformation of metals to allow fission product release, but no severe molten eructations, reactions, or vaporizations are considered credible for the configuration due to the limited time of the excursion and the significant heat transfer occurring to the moderator. The moderator and/or reflector typically assumed is water. A coherent solid cannot be moderated and solids < 100 μm in diameter must be intimately mixed with the moderator to be fully moderated.

$$\text{Airborne Source Term} = MAR_{c2} \times DR_{c2} \times ARF_{c2} \qquad (6\text{-}2)$$

where: MAR_{c2} = inventory of fissionable material and radionuclides from criticality in reflected/ moderated solids calculated by computer codes such as ORIGEN2 based on 1E+18 total fissions; or, [1E+18 fissions/1E+19 fissions] X table (see footnote 1).

DR_{c2} = Damage Ratio for radionuclides generated by criticality:
metal pieces, 0.1
fines, (e.g., powders), 1.0.

ARF_{c2} = Airborne Release Fraction: fissionable material and non-volatile fission products can be neglected, 5E-1 for noble gases, 5E-2 for iodine[3]

Bare, Dry Solids

This configuration covers solids that may be reflected but have no moderation (i.e., not immersed in liquid). The equation assumes some softening and local deformation of metals to allow fission product release, but no general oxidation or molten dispersion. To achieve criticality, kg quantities of material must be involved as a minimum. Generally, single parameter limits will be significantly exceeded as sphere configurations are not common.

[3] Table 6-10 indicates quantities of radionuclide groups released from molten ceramic oxide fuel. For non-volatiles, these values would be multiplied by 5E-4 for release from the moderating solution, which will render them an insignificant contributor to dose. The values for gases and semi-volatiles are taken directly from Table 6-10 without any additional credit taken for capture in solution as these values are already less than the NRC solution values. As a potential gas, the value from Table 6-5 for ruthenium could be used without capture as well, but the quantities of ruthenium generated would typically make it a minor contributor.

$$\text{Airborne Source Term} = (MAR_{c3} \times DR_{c3} \times ARF_{c3}) \qquad (6\text{-}3)$$

where: MAR_{c3} = inventory of radionuclides from criticality in dry, solids calculated by computer codes such as ORIGEN2 based on 1E+17 total fissions; or, [1E+17 fissions/1E+19 fissions] X table (see footnote 1).

DR_{c3} = Damage Ratio for radionuclides generated by criticality:
metal pieces, 0.1
fines, (e.g., powders), 1.0.

ARF_{c3} = Airborne Release Fraction: 5E-1 for noble gases, 5E-2 for iodine[4].

Large Storage Arrays

This configuration deals with driving together by external forces large amounts of material (i.e., reactor fuel storage arrays, extremely large quantities of fissionable materials). The total fission yield of 1E+20 is based on historical reactor excursions with moderation, and was originally developed in the Defense Programs Safety Survey solely for the purpose of evaluating criticalities in spent fuel storage pools when fuel was driven together. However, issues related to criticality and potential extreme excursions in storage environments are most appropriately handled in the arena of criticality safety evaluations performed by criticality safety professionals for the purpose of minimizing chances of occurrence. Design considerations relating to these issues are not effectively driven by unrealistic dose calculations by accident analysts. If it is necessary to consider an upper bound of consequence, 50% of the noble gases and 5% of the radioiodine (based on fuel slump/melt values used for moderated solids and dry solids) considered released without attempting mechanistic modelling of base material physical effects is assessed to be adequate.

[4] Table 6-10 indicates quantities of radionuclide groups released from molten ceramic oxide fuel, which will bound this condition where minimal, if any, melting is assumed. Other non-volatile releases may be assumed in accordance with Table 6-10, but will not typically be major contributors to airborne dose outside the facility. Historical records indicate some surface oxidation and deformation may occur, but this should be bounded by the ARF and RF values of 3E-5 and 0.04 given for oxidation of metals at elevated temperatures in Chapter 4, and thus not be a significant release source.

6.2 TOTAL FISSION YIELD

The potential releases from criticality events are directly related to the total fission yield of the event. This section provides historical and analytical information relating to fission yields to assess bounding values.

6.2.1 Historical Excursions

To provide some perspective on the types of situation that have resulted in excursions in the past, lists compiled by various authors, of the excursions that have actually taken place are presented. Several incidents are listed in multiple tables due to the diverse nature of sources used.

Stratton (September 1967) reviewed information on 34 excursions that occurred between 1945 and 1965. Ten involved water solutions of ^{235}U or ^{239}Pu systems, 9 involved metal assemblies in air, and 9 involved inhomogeneous light- and heavy-water-moderated metal systems. Olsen et al. (July 1974) reviewed Stratton's excursion data and experimental data from the French study Consequences Radiologiques d'un Accident de Criticite (CRAC). The excursions are listed by system type in Table 6-1. The total fission yield of the 12 excursions listed involving aqueous solutions ranges from 1E+15 to 4E+19 fissions. All but one are bounded by a total fission yield of 1.3E+18 fissions. The 11 excursions listed for excursions involving metal systems (including some reflection) range from ~ 3E+15 to 3.8E+17 fissions. All but one are bounded by a yield of 1.5E+17 total fissions. Only 3 excursions are listed for moderated foil and powder systems with total fission yield listed for only 2 excursions (~ 6E+15 & 3E+16 total fissions). Excursions in reactor and reactor experiments are reproduced in Table 6-2, (Nyer, Bright and McWhorter, 1965) and Tables 6-3 and 6-4 (Table I - Inhomogeneous Water-Moderated and Miscellaneous Systems, Stratton, Sept 1967). These values are considered at least partially representative of excursions that may occur in an array. The total fission yields range from 3E+16 to 1.2E+20 for the 9 inhomogeneous water-moderated excursions listed in Stratton and 3.3E+17 to 4.7E+18 for the reactor excursion experiments covered in Nyer, Bright and McWhorter (1965). A value of 1E+20 (rounding the coefficient to a single digit) bounds the reported values.

6.2.2 Analytical Models for Solution Criticalities

Two empirical models to estimate the total fission generated in excursions involving solutions have been developed (Tuck August 1974, Olsen et al., July 1974). Tuck's (August 1974) empirical relationships were developed from data for experimental studies on solution criticalities in Kinetic Experiments on Water Boilers and the French study CRAC.

Table 6-1. Summary of Known Accidental Criticality Excursions (1945 to 1974): (a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems
(Olsen, et al, July 1974)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
(a) Solution Systems										
SE 1	12/49	LASL, New Mexico	Water Boiler	$U(93)O_2(NO_3)_2$ ($\approx$ 1 kg U235; 13.6 l)	Sphere, graphite-reflected	$\approx 3x10^{15}$ (barely over prompt critical)	Not known	$3\text{-}4x10^{16}$	Control rods withdrawn too fast	None
SE 2	11/16/51	Hanford works - Richland, WA	P-11	$PuO_2(NO_3)_2$ (1.15 kg Pu; 63.8 l)	Sphere, 93% full reflected	$8x10^{16}$	Single burst	$8x10^{16}$	Too high fuel addition	None
SE 3	5/26/54	ORNL, TN	Spider	UO_2F_2 (18.3 kg U235; 55.4 l)	Cylindrical annulus, unreflected	$5x10^{16}$	Not known	$1x10^{17}$	Shift of poison	None
SE 4	2/1/56	ORNL, TN	Scram blade	UO_2F_2 (27.7 kg U235; 58.9 l)	Cylinder, unreflected	$1.6x10^{17}$	Single burst	$1.6x10^{17}$	Geometry change	Warping of bottom of cylinder
SE 5	1/30/68	ORNL, TN	U-233	$UO_2(NO_3)_2$ ($\approx$ 1 kg U233; 5.8 l)	Sphere, water-reflected	$1.1x10^{16}$	Single burst	$1.1x10^{16}$	Air in line	None
P 1	6/16/58	ORNL, TN - Y-12 Processing Plant	Y-12	$UO_2(NO_3)_2$ (2.5 kg U235; 56 l)	Cylinder, concrete reflected below	$\approx 1x10^{16}$	13 min.	$1.3x10^{18}$	Valve leaked or left open	None (loss: $1000)
P 2	12/30/58	LASL, NM - Pu Processing Plant	Agitator	$PuO_2(NO_3)_2$ (3.27 kg Pu; 168 l)	Cylinder, water-reflected below	$1.5x10^{17}$	Single burst	$1.5x10^{17}$	Procedure not followed	None
P 3	10/16/59	Idaho Reactor Testing Area, Chemical Processing Plant	IF-1 (siphon)	$UO_2(NO_3)_2$ (34.5 kg U235; $\approx$ 800 l)	Cylinder, concrete reflected below	$\approx 10^{17}$	Not known	$\approx 4x10^{19}$	Sparge gauge plugged	None (loss: $62,000)
P 4	1/25/61	Idaho Reactor Testing Area, Chemical Processing Plant	IF-11 (air lift)	$UO_2(NO_3)_2$ (8 kg U235; 40 l)	Cylinder	No estimate	Not known	$6x10^{17}$	Instruction misinterpreted	None (loss: $6000)
P 5	4/7/62	Hanford works - Richland WA	Recuplex	Pu complex (1.5 kg Pu)	Cylinder, unreflected	$\approx 10^{16}$	37 hrs.	$8x10^{17}$	Valve leaked or opened	None (loss: $1000)

Table 6-1. Summary of Known Accidental Criticality Excursions (1945 to 1974): (a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems (cont.)
(Olsen, et al, July 1974)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
P 6	7/24/64	Wood River Junction, RI - scrap recovery facility	Wood River	$UO_2(NO_3)_2$ (2.64 kg U235)	Cylinder, unreflected	$1.1x10^{17}$	Not known	$1.3x10^{17}$	Procedure not followed	None
P 7	8/24/70	Windscale Works, England	Windscale	Pu complex ($\approx$ 2.5 kg Pu; $\approx$ 100 l)	Cylinder	No estimate	5-10 sec.	$1x10^{15}$	Pu accumulated in organic	None
(b) Metal Systems										
ME 1	6/4/45	LASL, NM	Metal Cubes	83% U235 Enriched U metal 1/2 in. cubes (35.4 kg)	Array of cubes; H_2O reflected	$\approx 3x10^{15}$	Not known (perhaps 3 bursts)	$\approx 3x10^{16}$	Water leaked into array	None
ME 2	8/21/45	LASL, NM	Dragon	Delta phase Pu metal (6.2 kg)	Sphere reflected by WC	$\approx 1.8x10^{15}$ (10 cents over)	< 1 sec	$\approx 1x10^{16}$	Dropped reflector block	None
ME 3	5/21/46	LASL, NM	Screw-driver	Delta phase Pu metal	Sphere reflected by Be	$\approx 1.8x10^{14}$	Not known	$\approx 3x10^{15}$	Screwdriver holding reflector away from Pu slipped	None
ME 4	2/1/51	LASL, NM	Aquarium machine	Two cylinders U(93) metal (24.4 kg and 38.5 kg)	Side by side in water tank	$\approx 6x10^{15}$	Not known (perhaps several bursts)	$1x10^{17}$	Went critical during practice scram	Slight oxidation
ME 5	4/18/52	LASL, NM	Jamima	U(93) metal (92.4 kg)	Cylinder, unreflected	$\approx 1x10^{15}$ (21 cents over)	< 1 sec	$1.5x10^{16}$	Computation error	None
ME 6	2/3/54	LASL, NM	Godiva I	U(93) metal (53 kg)	Sphere-unreflected	$5.6x10^{16}$	Single burst	$5.6x10^{16}$	Assembled too rapidly	Slight warping of pieces (loss: $600)
ME 7	2/12/57	LASL, NM	Godiva II	U(93) metal (54 kg)	Sphere-unreflected	$1.2x10^{17}$ (21 cents over)	Single burst	$1.2x10^{17}$	Graphite fell against assembly	Warping oxidation, near melting close to center (loss: $2100)

Table 6-1. Summary of Known Accidental Criticality Excursions (1945 to 1974): (a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems (cont.)
(Olsen, et al, July 1974)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
ME 8	6/17/60	LASL, NM	9-inch cylinder	U(93) metal (48 kg)	Cylinder, graphite-reflected	$\approx 1x10^{15}$	Not known	$6x10^{16}$	Error in addition estimate	Trivial
ME 9	11/10/61	ORNL, TN	U-Paraffin	U(93) metal ($\approx$ 75 kg)	Cylinder, parraffin-reflected	$\approx 1x10^{15}$	Not known	$\approx 1x10^{16}$	Error in addition estimate	None
ME10	3/26/63	LARL, CA	LRL	U(93) metal (47 kg)	Cylinder, Be-reflected	$1x10^{17}$	Not known	$3.8x10^{17}$	Ram caught reflector; lifted; fell	Metal melted and some burned, contamination (loss: $95,000)
ME11	5/28/65	WSMR, NM	U-Mo Alloy	U(93)-1.0% Mo (96 kg)	Cylinder-unreflected	$1.5x10^{17}$	Not known	$1.5x10^{17}$	Incorrect operation	Assembly bolts broken, minor damage to coating
(c) Moderated Foil and Powder Systems										
H 1	2/11/45	LASL, NM	UH_3-Styrex blocks	U(93)H_{10} pressed in Styrex (UC_4H_{10})	Assembly of blocks	$\approx 6x10^{15}$	Single burst	$\approx 6x10^{15}$	Excess reactivity addition	Cubes swollen and blistered
H 2	7/3/56	LASL, NM	Honey-comb	U(93) metal foil sandwiched with carbon	Cylinder, Be-reflected	Not known	Not known	$3.2x10^{16}$	Assembled too rapidly	None
H 3	12/11/62	LASL, NM	ZEPO	U(93) metal foils sandwiched with carbon	Cylinder, C and Be reflected	$3x10^{16}$ (12 cents prompt critical)	Single burst	$3x10^{16}$	Excess fuel addition	None

Table 6-2. Destructive Power Excursion Summary
(Nyer, Bright and McWhorter 1965)

Reactor	Reactivity addition, S	α, s^{-1}	Peak power, MW	Energy release, MWs	Maximum temp., °C	Maximum energy density, W-s/cm^3	Maximum pressure, psi	Remarks
BORAX I [28]	3.1	384	≤19,000	135	≤1,800	≤6,500	6,000 - 10,000	Destroyed core, vessel, and some associated equipment. Small fission-product release. Steam explosion proposed as cause.
SL-I [29]	3.0	280	~19,000	133	>2,075	>7,300	10,000	Destroyed core, bulged vessel, local fission-product contamination. 10% fission-product release. Steam explosion - minor contribution from metal-H_2O reaction.
SPERT-I [30] D-12/25	2.6	200	1,130	11	585	2,000	7	Melted >0.5% of core.
	2.7	218	1,270	19	680	2,300	8	Melted ~3% of core.
	3.55	313	2,250	31	1,360	4,600	≤4,000	Melted ~35% of core. Destroyed core and associated equipment, bulged tank. ~4% fission-product release. Probable steam explosion - Al_2O_3 analysis indicates ~3.5 MWs energy release from metal-H_2O reaction.
SPERT I [31] oxide core	2.6	455	17,400	155	1,800	2,200	70	Two fuel rods ruptured. Discoloration and/or deformation of 25% of fuel rods. Negligible fission-product release.
	3.3	645	35,000	155	1,800	2,200	130	Two fuel rods ruptured. Discoloration and/or deformation of 25% of fuel rods. Negligible fission-product release.
SNAPTRAN-3 [32]	3.5	1,400	~20,000	50	>2,500	7,100	~4,000	Burst pressure vessel. All fuel rods ruptured, ~half of fuel reduced to powder form. Negligible fission-product escape.

Table 6-3. Inhomogeneous Water-Moderated Systems
(Stratton September 1967)

Date	Location	Active Material	Geometry	Total fissions	Cause	Physical damage
June 4, 1945	Los Alamos, New Mexico	35.4 kg U ~83% ^{235}U 1/2 in. cubes	Pseudosphere, water-reflected	~3 x 10^{16}	Water seeping between blocks	None.
February 1, 1951	LASL, New Mexico	2 cylinders U 24.4 and 38.5 kg 93% ^{235}U	2 cylinders, water-reflected	10^{17}	Scram increased reactivity	Slight oxidation.
July 6, 1952	ANL, Illinois	6.8 kg ^{235}U oxide particles in plastic	Inhomogeneous cylinder water-reflected	1.22 x 10^{17}	Manual withdrawal of central safety rod	Plastic destroyed.
December 12, 1952	Chalk River, Canada	Normal U	Rods, D_2O-moderated, graphite-reflected	1.2 x 10^{20}	Safety circuits failed; control rod misoperation	Core ruined.
July 22, 1954	Reactor Testing Area, Idaho Falls, Idaho	U-Al plates, Al clad	Inhomogeneous cylinder, water-moderated	4.68 x 10^{18}	Estimate of expected excursion too low	Reactor destroyed.
October 15, 1958	Vinca, Yugoslavia	3996 kg Normal U	Rods D_2O-moderated, unreflected	2.5 x 10^{18}	Too much D_2O added in final step of experiment	None.
March 15, 1960	Suclay, France	2.2 tons UO_2, 1.5% enriched	Canned UO_2 rods in water	3 x 10^{18}	Control rod withdrawn	None.
January 1, 1961	Reactor Testing Area Idaho Falls, Idaho	U-Al plate, Al clad	Inhomogeneous cylinder, water-moderated	4.4 x 10^{18}	Quick manual withdrawal of control rod	Reactor destroyed, building contaminated.
December 30, 1965	Mol, Belgium	1.2 x 10^6 g UO_2 7% enriched	Canned UO_2 rods in H_2O-D_2O	4.3 x 10^{17}	Manual withdrawal of control rod	None.

Table 6-4. Miscellaneous Systems
(Stratton September 1967)

Date	Location	Active Material	Geometry	Total fissions	Cause	Physical damage
February 11, 1945	Los Alamos, New Mexico	UH_3 pressed in styrex	Cylinder	$\sim 6 \times 10^{15}$	Reflector added and/or source too large	UH_3-styrox cubes swollen and blistered.
1953	USSR	Unknown	Unknown	Unknown	Unknown	Unknown.
November	Idaho Reactor Testing Area	1/2 in. ^{235}U rods	Cylinder, rods NaK cooled	4.7×10^{17}	Incorrect scram used	Core molten.
July 3, 1956	LASL, New Mexico	58 kg U 93% ^{235}U, 2- and 5-mil foils	Cylinder	3.2×10^{16}	Change of k from previous assembly too large	None.
November 18, 1956	Reactor Testing Area, Idaho Falls, Idaho	^{235}U Ni-Cr elements, ZrH moderated	Cylinder, prototype aircraft engine	2.5×10^{19}	Incorrect wiring in ion chamber circuit	Every fuel cartridge melted.
December 11, 1962	LASL, New Mexico	^{235}U foils in graphite	Cylinder, graphite and Be-reflected	3×10^{16}	Inadequate communication between work crews	None.

The model calculates the maximum fissions during a 5-sec burst, total fissions, and specific fission rate (the duration is assumed to range from 10 min to 40 hours based upon historical excursions). Only the volume of the tank (V, in liters) is required to calculate total fission using the formula:

$$\text{Total fissions} = V \times 10^{17} \text{ fissions} \quad (6\text{-}4)$$

For an excursion involving a large volume (1000 liters, 264 gallons), the estimate of total fissions (1E+20) exceeds the estimate of total fission by Woodcock as cited in McLaughlin (1991) by an order of magnitude. See Table 6-5 for a display of criticality accident fission yields estimated by Woodcock (1966).

Olsen et al. (July 1974) also developed empirical models for solution excursions based upon the CRAC experiments. Total fissions are the sum of F_b, fissions in initial burst, and F_p, fissions during the plateau. The fissions during the initial burst are:

$$F_b = 2.95(10)^{15}(V_b)^{0.82} \quad (6\text{-}5)$$

where: V_b = volume solution at time of burst, liters.

The fission during the plateau are:

$$F_p = 3.2 \times 10^{18} (1\text{-}t^{-0.15}) \quad (6\text{-}6)$$

where t = duration of the plateau, seconds. No method to determine the duration of the plateau is identified. Currently, neither technique discussed in this subsection is commonly used and both are considered very conservative.

6.2.3 Assessment of Fission Yields

Once it is determined that a postulated scenario may physically result in a excursion, it is necessary to determine the potential fission yield for the purposed excursion. The total fission yield is defined by the type of system and the fissile material involved. This section provides reference fission yields that may be used for general SAR purposes. Care should be taken to insure that, as the NRC has noted, potentially unique situations do not exist at a given facility.

Table 6-5 provides a list of estimated criticality fission yields (Woodcock, 1966) that have been discussed within both DOE and NRC literature. Many of these values are currently considered excessively conservative in light of historic experience and current knowledge of

Table 6-5. Theoretical Criticality Accident Fission Yields
(McLauglin, 1991)

System	Initial Burst Yield (fissions)	Total Yield (fissions)
Solutions under 100 gal (0.46 m^3)	1×10^{17}	3×10^{18}
Solutions over 100 gal (0.46 m^3)	1×10^{18}	3×10^{19}
Liquid/powder[(a)]	3×10^{20}	3×10^{20}
Liquid/metal pieces[(b)]	3×10^{18}	1×10^{19}
Solid uranium	3×10^{19}	3×10^{19}
Solid plutonium	1×10^{18}	1×10^{18}
Large storage arrays[(c)] below prompt critical	None	1×10^{19}
Large storage arrays[(c)] above prompt critical	3×10^{22}	3×10^{22}

[(a)]A system where agitation of a powder layer could result in progressively higher reactivity insertion.
[(b)]A system of small pieces of fissile metal.
[(c)]Large storage arrays in which many pieces of fissile material are present and could conceivably come together.

criticality behavior. As a potential precedent, however, they will be discussed in the individual criticality categories of (1) solutions, (2) moderated, reflected solids, (3) dry solids, and (4) large arrays.

6.2.3.1 Solutions

The eight excursions that have occurred in processing plants (non-experimental facilities) are listed in Table 6-6 reproduced from Paxton (1980). Total fission yield ranges from ~ 1E+15 to 4E+19 fissions. All 9 events involve solution systems. Five involved inadvertent transfer to non-safe vessels or systems. Two involve extraction/concentration into a solvent layer with the aqueous phase providing reflection. The last event was attributed to equipment/procedural failures. The highest value (4E+19 fission) resulted from the siphoning of a large volume (~ 800 liter) containing 34.5 kg of ^{235}U (approximately 70 time the minimum critical mass) from a safe to unsafe geometry with neutrons reflected by concrete below the cylinder.

Woodcock lists two levels of solution excursion, >100 gallon and <100 gallons, with an order of magnitude difference in both initial and total fission yields. The U.S. NRC

Table 6-6. Accidents in Processing Plants
(Paxton, 1980)

Date	Plant	Total fissions	First pulse	Doses (Rads)	Notes
6/16/58	Y-12	1.3×10^{18}	$\sim 7 \times 10^{16}$	365, 339, 327, 270, 236, 69, 69, 23	^{235}U solution washed into drum
12/30/58	LASL	1.5×10^{17}	1.5×10^{17}	~4400 (fatal), 135, 35	Pu concentrated in solvent layer
10/16/59	Idaho CPP	4×10^{19}	$\sim 10^{17}$	50 R, 32 R, mostly beta	^{235}U solution siphoned into tank
1/2/61	Idaho CPP	6×10^{17}	6×10^{17}	None	^{235}U solution forced into cylinder by air
4/7/62	RECUPLEX	8.2×10^{17}	$\sim 10^{16}$	87, 33, 16	Pu solution in sump sucked into tank
7/24/64	Wood River Junction	1.3×10^{17}	$\sim 10^{17}$	10000 (fatal), two 60-100	^{235}U solution poured into tank
8/24/70	Windscale	10^{15}	$\sim 10^{15}$	Negligible	Pu concentrated in trapped solvent
10/17/78	Idaho CPP	3×10^{18}	Unknown	None	^{235}U buildup in diluted scrub solution

assumes, in its Regulatory Guides (NRC, April 1977, NRC, April 1979, NRC, July 1979), an initial burst of 1E+18 fissions followed by 47 bursts of 1.9E+17 fissions at ten minute intervals for eight hours for a total of 1E+19 fissions. The boiloff of 100 liters solution is postulated to terminate the excursion, implying a relatively large system. Since the 1E+19 total fissions has been in use for a long period, appears to bound most historical and estimated yields, and is not substantially less than the maximum historical occurrence or Woodcock estimate, it is assessed to be the bounding reference yield. The excursion is assumed terminated by the evaporation of 100 liters of solution over eight hours.

The 100 liters is a general value assumed for release fraction purposes, and is not assigned any specific technical meaning for real systems.

6.2.3.2 Fully Moderated and Reflected Solids

McLaughlin (1991) discusses the basis for the total fission yield reported by Woodcock (3E+20 fissions) that is based upon an autocatalytic phenomena ("... dry powder becomes flooded, goes prompt critical as an equivalent very rich solution, and then the mixing and dilution which accompany the excursion introduces additional reactivity since one is sliding down the critical mass versus concentration curve.") Competing negative feedback mechanisms (e.g., thermal expansion and microbubble formation) are ignored. Woodcock admits "this estimate is rather a shot in the dark." The possibility of positive feedback during dilution of rich solutions is considered by Stratton but "it is difficult to imagine an explosive reaction". A prompt total fission yield in a few hundred liters of liquid would be a significant explosion. McLaughlin (1991) cites the Woodriver Junction criticality as most closely approximating this situation with a total fission yield of 1E+17 fissions.

For water-moderated metal pieces, McLaughlin (1991) notes that Woodcock does not provide a discussion on the basis for prompt yield of 3E+18 and total yield of 1E+19. Instead of the values referring to a "system of small pieces of fissile material" as indicated in the footnotes to the Table 6-5, the values are for "1 or a small number of pieces".

The three historical excursion listed in Olsen et al. (July 1974) are bounded by a value of 3E+16 total fissions. Given the types of situation encountered in DOE facilities where it is difficult to accumulate the quantity of materials required, contain the material and moderator, and assume any shape that would be unfavorable, a reference value of 1E+18 fission in a single burst is assessed to be the bounding reference value and is believed to be very conservative.

6.2.3.3 Dry Powder and Metal

"Criticality accidents with solid metal systems (including alloys) should be readily controlled at a likelihood of occurrence that is vanishingly small. ... Only rarely are there operational requirements which necessitate working with more than the water reflected spherical critical mass (^{233}U 6.0 kg, ^{235}U 20.1 kg, ^{239}Pu 5.0 kg) ... " (McLaughlin, 1991). Eleven excursions are listed for Metal Systems in Olsen et al. (July 1974). All the historical excursions have taken place in experimental systems. Five resulted from mechanical failures that increased the reflection in the system. Three resulted from incorrect operations, and three resulted from computational errors. None are especially representative of anticipated accidental conditions, and are likely to be conservative representations of expected conditions.

The range of the eleven excursions is from 3E+15 to 4E+17 total fissions. All the excursions are bounded by 4 yields at or around 1E+17 (1E+17 to 4E+17 fissions) and involved large masses of metal (47 to 96 kg U) well beyond the water-moderated spherical critical mass. The highest yield is from a mechanical failure. The remaining 3 yields are 1E+17, 1.2E+17 and 1.5E+17 fissions. It appears adequately conservative to assess 1E+17 fissions as the bounding reference total fission yield for dry solid systems.

6.2.3.4 Large Storage Arrays

As McLaughlin (1991) states "... common sense storage practices and case-specific analysis should lead to the conclusion that either the critical state cannot be credibly reached or, if the upset condition is so severe that criticality cannot be precluded, then the consequences of the criticality accident are minor compared to the total accident consequences. Under no circumstances can an accident scenario be envisioned which would incorporate the simultaneity, speed, and neutron source requirements which would lead to anything approaching the '3 X 10^{22} fissions' and 'serious explosion' Woodcock proposes...The fundamental storage practice for unmoderated fissile material should be a maximum effective density, i.e. the fissile mass divided by the outer container volume, which does not exceed about 1 kg/liter."

Excursions in reactor and reactor experiments listed in Table 6-2 (Nyer, Bright and McWhorter, 1965) and Tables 6-3 & 6-4 (Table I - Inhomogeneous Water-Moderated and Miscellaneous Systems, Stratton, Sept 1967) are considered partially representative of potential excursions in an array. The total fission yields range from 3E+16 to 1.2E+20 for the nine inhomogeneous water-moderated excursions listed in Table 6-3 (Stratton, September 1967) and 3.3E+17 to 4.7E+18 for the reactor excursion experiments covered in Table 6-2 (Nyer, Bright and McWhorter, 1965). A value of 1E+20 (rounding the coefficient to a single digit) bounds the reported values and is assessed to be the bounding reference yield. This value is generally considered only applicable to reactor fuel arrays, typically with moderation. For analytical purposes, it is not considered representative of uranium or plutonium vault storage conditions, even under severe stress conditions such as earthquakes.

6.3 MATERIAL RELEASE IN CRITICALITY EXCURSIONS

The radionuclides generated by an excursion are a function of the fissionable material undergoing the reaction. The quantity of each fission product or actinide formed is a function of the total fission yield. Reference values for the various systems were designated in the previous section. Estimation of fission product quantities can be done with computer codes, such as ORIGEN2, or by simple ratios (total fissions for scenario/1E+19 fission) to values in NRC regulatory guides prepared for fuel cycle facilities. The regulatory guides do

not include particulate fission product generation, and are thus less precise than computer codes. However, the quantity of particulate fission products are normally small, their potential release fractions are likewise small, and, unlike noble gases and iodine, they are subject to facility filtration. Accordingly, not accounting for them is typically an adequate approximation. It is noted again that site personnel should perform whatever background work they believe is warranted to verify that unusual or unique circumstances do not exist at their facilities.

The anticipated quantities and radionuclides generated by solution criticalities with total fission yields of 1E+19 fissions involving three fissionable material compositions are:

- Table 6-7 - spent nuclear fuel (<5% ^{235}U Enrichment) (NRC, April 1977)

- Table 6-8 - unirradiated 400 g enriched uranium/l (enrichment level not specified but assumed to be <5% as in Reg. Guide 3.34) (NRC, April 1979); and,

- Table 6-9 - plutonium (of unspecified isotopic composition) (NRC, July 1979).

Distinct ARFs are assigned for different types of radionuclide fission products and, if necessary, base fissionable materials.

6.3.1 Solutions

The NRC Regulatory Guides define the airborne release of the noble gases and radioiodine generated in a solution from the facility. In almost all cases, the amount of energy generated by the excursion is not sufficient to disable the engineered gaseous effluent treatment devices on the facility exhaust, but no DOE non-reactor facility is known to have gaseous effluent treatment device to attenuate the noble gases. All the noble gases generated in a liquid are assumed to be released to the atmosphere, generally through the facility effluent treatment system. Additionally, the NRC assumes that all the radioiodine generated is released to the liquid and a fraction of 0.25 is not captured by the liquid. If iodine removal devices (e.g. impregnated activated charcoal, silver-substituted zeolite, silver-salt coated ceramic pieces) are not present in the gaseous effluent system, the LPF is assumed to be 1.0. The RF for a vapor is assumed to be 1.0.

As previously noted, the heat generated by the excursion results in heating of the liquid and the combination of density changes/microvoids rapidly terminates the reaction. Some of the liquid is assumed lost by evaporation. The system again becomes critical with sufficient cooling but the yield level is lower due to density changes not reversed. The reaction is

Table 6-7. Curies of Important Nuclides Released During Nuclear Excursion Involving Spent Fuel Solution
(NRC, April 1977)

Nuclide	Half-life[b]	Radioactivity, Ci[a]		
		0 to 0.5 hr	0.5 to 8 hr	Total
Kr-83m	1.8 h	3.7E+0	3.3E+1	3.7E+1
Kr-85m	4.5 yr	1.6E+1	1.5E+2	1.7E+2
Kr-85	1.7 yr	1.5E-4	1.4E-3	1.6E-3
Kr-87	76.3 m	1.0E+2	9.0E+2	1.0E+3
Kr-88	2.8 h	6.5E+1	5.9E+2	6.6E+2
Kr-89	3.2 m	4.1E+3	3.7E+4	4.1E+4
Xe-131m	11.9 d	3.8E-4	3.5E-3	3.9E-3
Xe-133m	2.0 d	5.5E-2	1.9E+0	2.2E+0
Xe-133	5.2 d	1.3E+0	2.3E+1	2.7E+1
Xe-135m	15.6 m	1.1E+1	2.8E+3	3.3E+3
Xe-135	9.1 h	1.6E+1	3.5E+2	4.1E+2
Xe-137	3.8 m	3.8E+3	4.2E+4	4.9E+4
Xe-138	14.2 m	1.2E+3	9.5E+3	1.1E+4
I-129	2E+7y	3.9E-10	4.3E-10	4.3E-10
I-131	8.1 d	1.8E-1	1.6E+0	1.8E+0
I-132	2.3 h	6.7E-1	6.1E+0	6.7E+0
I-133	0.8 h	3.5E+0	3.1E+1	3.5E+1
I-134	52.6 m	4.8E+1	4.3E+2	4.8E+2
I-135	6.6 h	1.2E+1	1.0E+2	1.2E+2

Table 6-8. Curies of Important Nuclides Released During Nuclear Excursion Involving Unirradiated, Unenriched Uranium Solution (400 g U/liter)
(NRC, April 1979)

Nuclide	Half-life[b]	Radioactivity, Ci[a]		
		0 to 0.5 hr	0.5 to 8 hr	Total
Kr-83m	1.8 h	2.2E+1	1.4E+2	1.6E+2
Kr-85m	4.5 yr	2.1E+1	1.3E+2	1.5E+2
Kr-85	1.7 yr	2.2E-4	1.4E-3	1.6E-3
Kr-87	76.3 m	1.4E+2	8.5E+2	9.9E+2
Kr-88	2.8 h	9.1E+1	5.6E+2	6.5E+2
Kr-89	3.2 m	5.9E+3	3.6E+4	4.2E+4
Xe-131m	11.9 d	1.1E-2	7.0E-2	8.2E-2
Xe-133m	2.0 d	2.5E-1	1.6E+0	1.8E+0
Xe-133	5.2 d	3.8E+0	2.3E+1	2.7E+1
Xe-135m	15.6 m	3.1E+2	1.9E+3	2.2E+3
Xe-135	9.1 h	5.0E+1	3.1E+2	3.6E+2
Xe-137	3.8 m	6.9E+3	4.2E+4	4.9E+4
Xe-138	14.2 m	1.8E+3	1.1E+4	1.3E+4
I-131	8.1 d	1.2E+0	7.5E+0	8.7E+0
I-132	2.3 h	1.5E+2	9.5E+2	1.1E+3
I-133	0.8 h	2.2E+1	1.4E+2	1.6E+2
I-134	52.6 m	6.3E+2	3.9E+3	4.5E+3
I-135	6.6 h	6.6E+1	4.0E+2	4.7E+2

Table 6-9. Curies of Important Nuclides Released During Nuclear Excursion Involving Plutonium Solution
(NRC, July 1979)

Nuclide	Half-life[b]	Radioactivity, Ci[a]		
		0 to 0.5 hr	0.5 to 8 hr	Total
Kr-83m	1.8 h	1.5.E+1	9.5E+1	1.1E+2
Kr-85m	4.5 yr	9.4E+0	6.1E+1	7.1E+1
Kr-85	1.7 yr	1.2E-4	7.2E-4	8.1E-3
Kr-87	76.3 m	6.0E+1	3.7E+2	4.3E+2
Kr-88	2.8 h	3.2E+1	2.0E+2	2.3E+2
Kr-89	3.2 m	1.8E+3	1.1E+4	1.3E+4
Xe-131m	11.9 d	1.4E-2	8.6E-2	1.0E-1
Xe-133m	2.0 d	3.1E-1	1.9E+0	2.2E+0
Xe-133	5.2 d	3.8E+0	2.3E+1	2.7E+1
Xe-135m	15.6 m	4.6E+2	2.8E+3	3.3E+3
Xe-135	9.1 h	5.7E+1	3.5E+2	4.1E+2
Xe-137	3.8 m	6.9E+3	4.2E+4	4.9E+4
Xe-138	14.2 m	1.5E+3	9.5E+3	1.1E+4
I-131	8.0 d	1.5E+0	9.5E+0	1.1E+1
I-132	2.3 h	1.7E+2	1.0E+3	1.2E+3
I-133	20.8 h	2.2E+1	1.4E+2	1.6E+2
I-134	52.6 m	6.0E+2	3.7E+3	4.3E+3
I-135	6.6 h	6.3E+1	3.9E+2	4.5E+2
Pu-238[c]				5.9E-4
Pu-239[c]				2.7E-5
Pu-240[c]				5.8E-5
Pu-241[c]				1.8E-2
Pu-242[c]				4.3E-7
Am-241[c]				2.4E-5

[a]Total Ci, except for Pu and Am, are based on cumulative yield for fission energy spectrum. The assumption of cumulative yield is very conservative, i.e., it does not consider appropriate decay schemes. Calculations regarding individual nuclide yields and decay schemes may be considered on an individual basis. Data in this table do not include the iodine reduction factor.

[b]y = year, h = hour, d = day, m = minute.

[c]Total radioactivity assumes the isotopic mix to be the equilibrium mix for recycled plutonium and 1 mg of PuO_2 released.

finally terminated when adequate liquid has been boiled off to result in non-critical conditions. The NRC Regulatory Guides assume that the boiloff of 100 liters of liquid is required to terminate the reaction and that 0.05% of the salt content of the 100 liters is made airborne as particulate material. The value is only a factor of 4 lower than the ARF assessed for airborne release during the vigorous, continuous boiling of solutions containing radionuclides.

The ARF of 5E-4 with an RF of 1.0 is adequate for base quantities of fissionable material in solution. The value is applied to the amount of material in 100 liters, or all of the material if less than 100 liters are involved. The 5E-4 factor can be applied to the fraction of other fission products generated by the criticality in 100 liters of solution. However, the total quantities of other fission products generated are small enough that they will not typically result in a significant dose increase. If HEPA or sand filters attenuate the particulate material carried in the gaseous effluents, particulates will not significantly contribute to doses outside the facility.

For fuel reprocessing solutions, the NRC assumes a fraction of 1E-3 of the radioruthenium in solution is released as well. For solutions that do not contain ruthenium prior to the criticality, the small quantities generated in the criticality will not typically increase potential dose consequences outside the facility.

6.3.2 Fully Moderated and Reflected Solids

The fission and activation products formed by an excursion in a solid are enclosed within the matrix of the solid fissionable material. The fissionable solids that are generally found in DOE non-reactor nuclear facilities are metal and ceramic oxides of the metals that may be clad in metal (e.g., aluminum, zircalloy, stainless steel). Because of the wide range of fissionable mixtures that may be used for fuel in the production, experimental, and test reactors at DOE sites, each generating its own spectra of irradiation products, fuel (unirradiated or spent) is not covered in this discussion.

It is postulated that the radionuclides generated by the criticality and present as fissionable material are in the solid matrix but covered by water, which acts as a moderator and reflector. Heat generated by the excursion is assumed to be dissipated in the water surrounding the fissionable material resulting in boiling of the water. The criticality is

ultimately terminated by density changes within the metal itself and the moderator due to heat generated.

The products contained in the matrix of the solid fissionable materials are not exposed to the ambient environment and would not constitute a source term hazard unless released from the matrix. Of the 11 historical excursion involving metal system listed on Table 6-1 (b), only 1 (with a total fission yield estimated at 4E+17 fissions) exhibited any melting of the metal. Warping/oxidation are listed as consequences of 3 additional events (total fission yields ranging from 6E+16 to 1E+17 fissions). Therefore, a limited amount of softening/melting of the solid would be anticipated at the reference yield level of 1E+18 fissions, although this level is likely to be conservative. This softening/melting would not, however, cause significant disruption of the solid mass itself. It would most likely produce physical distortion of the material due to almost instantaneous melting and congealing from heat transfer to and interaction with water.

It is assumed that 10% (0.01 fraction) of the metal melts/softens due to the heat generated, thus allowing noncondensible gases and volatile radionuclides (radioiodine) in that fraction to be released. For powders or fines in solutions, the surface area to volume ratio of individual particles is considered sufficiently large that all noncondensible gases and radioiodines will escape. Restrepo (April 1992) reviewed the post NUREG-0772 research and experiments on fission product release rates from heated/melted spent fuel. The specific elements involved were categorized into 12 discrete chemical groups (as shown below in Table 6-10) based upon the similarity of their responses to thermal stress. "... release fractions within each group varied by as much as two to three orders of magnitude ... the geometric mean and standard deviation of the release fractions for each of those groups were obtained." The geometric mean of the airborne release for each chemical groups, rounded off to a single digit, was selected as the release fraction for that group (except the noble gases) and is listed in Table 6-10. The upper limit estimate for release was selected for the noble gases. Also, no release fractions are reported for chemical group 12 (boron) and, since the behavior of that group's components is similar to that of the trivalent group (chemical group 9), the release rate for the trivalent elements is used for the boron elements in Table 6-10.

Thus, for metals, 5E-1 of the noble gases is assumed to be released from the solid matrix to the moderator and all of the noble gases released to the moderator are released to the ambient environment around the moderator. Likewise, 5E-2 of the halogen (iodine) is released from the matrix to the moderator. The 0.25 capture factor for iodine in solution is not assumed since the iodine release is already less than that for solutions and the water depth associated with this configuration is assumed to be limited. The 2E-3 factor assigned for radioruthenium may also be used without considering solution capture as the ruthenium may be generated as discrete gas bubbles escaping from the solid surface. As with solutions,

Table 6-10. Release Fraction for Various Chemical Classes from Heated Spent Fuel (Restrepo, 1991)

Group #	Group Name	Rep. Ele.	Elements in Group	ARF
1	Noble Gases	Xe	Xe, Kr, He, Ne, Ar, Rn, H	5E-1
2	Alkali Metals	Cs	Cs, Rb, Li, K, Fr, Na	2E-1
3	Alkali Earths	Ba	Ba, Sr, Mg, Ca, Ra, Be	3E-2
4	Halogens	I	I, F, Cl, Br, At	5E-2
5	Chalogens	Te	Te, S, Se, O, Po, N	7E-2
6	Platinoids	Ru	Ru, Rh, Pd, Os, Ir, Pt, Au, Ni	2E-3
7	Transition Metals	Mo	Mo, V, Cr, Fe, Co, Mn, Nb, Tc	3E-2
8	Tetravalent	Ce	Ce, Ti, Zr, Hf, Th, Pa, U, Np Pu	4E-4
9	Trivalent	La	La, Al, Sc, Y, Ac, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Am, Bk, Cf	6E-4
10	Main Group I	Cd	Cd, Hg, Zn, As, Sb, Pd, Tl, Bi	4E-3
11	Main Group II	Sn	Sn, Ca, In, Ag	4E-3
12	Boron	B	B, Si, P, C	6E-4

5E-4 of all non-volatile materials released to the moderator are released to the ambient environment (e.g. Group #2 2E-1 X 5E-4 = 1E-4). Since quantities will be small and the excursion does not generate sufficient energy to fail particulate filters on the exhaust system, the contribution of the airborne nonvolatile materials to ex-facility doses is generally ignored.

If the material involved is a ceramic oxide powder, no melting/softening is postulated that no significant fraction of the non-volatile fission products generated are released. Due to the size of the individual particles in a powder, it is assumed that the fraction of volatile fission products (noble gases, iodine, ruthenium) present are so close to the surface that they are released in the following fractions: 5E-1 of the noble gases; 5E-2 of the iodine; and, 2E-3 of the ruthenium.

Other groupings of radionuclides are possible. For instance, ruthenium is unique in that it can readily form volatile oxides under appropriate conditions. Ruthenium could be logically included with the Transition Metals increasing its release fraction from 2E-3 to 3E-2 (greater than an order-of-magnitude increase). The ARF for the Alkali Metals is 4 times greater than for the Halogens and is difficult to justify. Generally, the elements in the Main Group I elements appear to be more volatile than the elements included in the Main Group II category although both are assigned the same ARF. The role of cladding is critical for the Chalogens (Group V) elements especially Te for which significant interactions with the cladding are predicted in all but very oxidizing conditions. Ba releases appear to be high and such values only occur for very reducing conditions. The ARFs are those determined from the

evaluation of the most recent experiments on irradiated field heating and may not reflect response for lesser conditions.

6.3.3 Dry Powder and Metal

As discussed in the previous subsection, the products generated from a nuclear excursion are assumed to be enclosed in the matrix of the solid and the fractions listed in Table 6-10 (Release Fractions for Various Chemical Classes From Heated Spent Fuel) are assumed to be made airborne from the response of the solids to the thermal energy generated by the excursion. At the reference yield (1E+17 fissions), very little if any melting of the solid would be anticipated. It is still assumed that 10% (0.1 fraction) of the metal melts/softens due to the heat generated, thus allowing noncondensible gases and volatile radionuclides (radioiodine) in that fraction to be released. For powders or fines in solutions, the surface area to volume ratio of individual particles is sufficiently large that all noncondensible gases and radioiodines will escape.

The 10% value is supported by a simple precedent within DOE for such calculations. If it is postulated that the all the fission products in the gaseous state under heated conditions in a shallow layer (1 mm deep) on the surface of the metal or ceramic oxide exposed to air are released. The Damage Ratio would be:

$$DR_{c3} = [A_s \text{ X } 1\text{-mm}]/Vol_f \qquad (6\text{-}7)$$

where: DR_{c3} = Damage Ratio for Bare, Dry Solids during a nuclear excursion.
A_s = Area of fissionable material exposed to air, mm^2
Vol_f = Volume of fissionable material, mm^3

For 3 kg of plutonium metal considered to be in the shape of an ideal cube, this equation would indicate ~ 10% of the overall metal is softened/damaged. The same ARFs assigned for moderated, reflected solids are assigned to dry solid criticalities (see Table 6-10).

6.3.4 Large Storage Arrays

This configuration was intended to apply only to fuel storage arrays with moderation. The same ARFs assigned for moderated, reflected solids (see Table 6-10) are assigned to these fuel arrays without assuming physical damage to the fuel array material. Therefore, only the fission product noble gases and iodine generated are considered released.

7.0 APPLICATION EXAMPLES

This handbook provides a number of cautions against misapplication of the release data presented. The fundamental purpose of the handbook, however, is not to complicate the process of assigning release fractions, but to simplify it. This chapter is intended to assist the handbook user by providing examples of ARF selection within the context of specific accident models. In some cases, the examples may be trivial. In all cases, they attempt to demonstrate the fundamental point that pursuit of absoluteness in accounting for local release conditions is unwarranted. It represents a goal that often is not realistically attainable and is of little use for the overall purposes of hazard assessment. As noted in the Introduction of this document, the ARF and RF values provided are only intended to allow a better understanding of bounding consequences for accidents modelled at nonreactor nuclear facilities and to provide information to support general bases for decisionmaking.

7.1 APPLICATION METHODOLOGY

From a kilogram of material, the phenomena covered by this handbook can produce release estimates varying from micrograms to hundreds of grams. Accordingly, the most significant activity in assigning ARFs is the development of a physical model of the accident and its associated stresses. This model may range from a rudimentary statement (e.g., the material dropped two feet) to a condition requiring phenomenological engineering calculations to specify parameters (e.g., nature and strength of an explosion). Without such models, ARFs have no relationship to physical details at a given facility and are useful only for drawing gross conclusions (e.g., overall facility hazard classification as Category 2 or 3 in DOE-STD-1027-92, 100% release has consequences below a given value).

The thought process leading to development of a model, hazard identification and evaluation (i.e., hazard analysis) with potential followup by quantitative accident analysis, is described in: (1) DOE technical standards such as DOE-STD-3009-94, "Preparation Guide for U.S. Department of Energy Nonreactor Nuclear Facility Safety Analysis Reports;" (2) DOE technical reports such as DOE/EH-0340, "Example Process Hazard Analysis of a Department of Energy Water Chlorination Process;" and (3) any number of recognized textbooks such as "Guidelines for Hazard Evaluation Procedures" (American Institute of Chemical Engineers, 1992). The identification and analysis process is at least as important as any associated source term estimate as it has a broader purpose than merely defining the five numerical parameters used in such calculations. The overall process, of which identifying a model for source term and release estimation is only one component, allows for identification and evaluation of a wider variety of more subtle issues that cannot be generically captured by a release or dose construct (e.g., design or operational deficiencies not easily defined by "back-end" dose models).

For this reason, it is typically useful to limit model definition only to the level of complexity necessary to support the evaluation process, and to perform engineering analysis in source term estimation only to the level necessary to identify the appropriate release fraction category. Resolution of highly complex models is problematic, and can cause undue focus on release quantification at the expense of the overall evaluation process, which is designed to identify tangible improvements in safety based on many considerations other than source term estimates. To assist in model development at an appropriate level, this handbook has made extensive efforts to relate release data to phenomena with some generic applicability to the typical industrial accidents associated with nonreactor nuclear facilities, thus providing a straightforward reference for many circumstances. The application examples in the remainder of this chapter are designed to show both release fraction determination for an accident model and source term use within the context of the overall evaluation process.

7.2 EXAMPLE BACKGROUND

The examples are based on a mock operation described in Appendix B, which was developed by the DOE Office of Defense Programs for hazard analysis training. This makes it a reasonable vehicle for examining ARF and RF selection within the context of an overall analysis effort. The portions of the mock operation examined are a plutonium processing facility, an auxiliary lab, and a solid waste handling facility. A basic hazard identification for the mock operations is provided in Appendix B to assist in focusing discussion.

The examples follow a common format, beginning with a brief top-level summary of hazards associated with a given operation that identifies the most significant airborne release potentials. Sufficient detail is provided to understand source term model development, but this handbook is not an exhaustive treatise on accident phenomena. Bounding material-at-risk and damage ratios for given accidents are estimated as part of physical model development, ARFs and RFs are assigned in accordance with the information provided in this handbook, and an initial respirable source term at the point of release is estimated. Individual examples conclude with an assessment of results. The discussion in the examples represents the level of modelling rigor generally adequate for assignment of ARF and RF values.

When reviewing the examples provided, it is important to keep in mind that the focus is on developing a practical model, not on obtaining an absolute representation of the potential phenomena. A number of models are possible, and each might be appropriate for a given need. For example, if it is desired to demonstrate that a facility with small amounts of material is an insignificant hazard, simply assuming that 100% of the material is released could suffice. If, however, a more restrictive criteria of what is insignificant is subsequently adopted, the extreme model previously used may not satisfy the given need. It need not be attributed any special significance or meaning simply because it has a precedent, and can be abandoned in favor of a credible bounding ARF based on facility-specific information.

7.0 Application Examples; Introductory Material

The mock operation and example models are developed to specific levels. The overall processes and activities have been described at a functional level sufficient for identifying scenarios. The scenario development in examples is at a level sufficient to understand the use of operational parameters in the development of models. To demonstrate the traditional uses of such models, a simple consequence estimation sufficient to understand radiological dose implications is provided as well.

Because one of the purposes of using this handbook for source term determination is to "indicate relative significance of unmitigated releases," a basic measure of significance is provided for the mock operation. That measure is maximally exposed individual dose at the site boundary. This exposure is determined by the equation:

$$Q = \frac{\pi * Dose * \sigma_y * \sigma_z * \mu}{SA * EDE * BR} \qquad (7\text{-}1)$$

where:

Q	=	source term (grams),
Dose	=	total committed effective dose equivalent (rem),
σ_y	=	crosswind concentration standard deviation for F stability at site boundary (m),
σ_z	=	vertical concentration standard deviation for F stability at site boundary (m),
μ	=	wind speed (m/sec),
SA	=	material specific activity (Ci/gram)
EDE	=	material committed effective dose equivalent (rem/Ci)
BR	=	maximally exposed individual breathing rate (m^3/sec)

This equation is a simple algebraic rearrangement of the equation for Gaussian plume centerline dose for a ground level release to solve for source term instead of dose.

The mock operation handles plutonium, predominantly in the form of Pu-239. Therefore, doses calculated are committed effective dose equivalent has been used since, for alpha-emitters, the 50-year internal exposure due to inhalation dwarfs all other dose contributors. No attempt is made to correct for precise isotopic composition because the effect is not major unless an obvious error is made such as equating the health effects of curies of Pu-239 to curies of Pu-241. The specific activity of Pu-239 is 0.062 Ci/g, with committed effective dose equivalent values of 3.3E+8 rem/Ci for insoluble plutonium (i.e., oxides) and 5.1E+8 rem/Ci for soluble plutonium, which includes most other compounds by default. The mock operation handles plutonium metal, oxide, nitrates, and fluorides. However, the site

boundary distance for the mock operation is 2 km, and any respirable release would be anticipated to be an oxide by the time it traversed such a distance through air.

The dose measures used in the calculation are as presented in Table 7-1 below. At 2 km, the values of σ_y and σ_z are ~ 63 and 19 respectively for limiting F at 1 m/sec windspeed conditions, and a conservative breathing rate is 3.3E-4 m^3/sec. Using these values indicates, for example, that a source term of 0.9 Ci of Pu-239, or ~ 14.5 g, produces a dose of 25 rem to a maximally exposed person at the site boundary (i.e., individual there for duration of plume passage with no protection). Use of one of the Gaussian plume model computer codes currently in use in the DOE complex provided an estimate of 1 Ci release to obtain a dose of 25 rem, thus confirming the general appropriateness of the hand calculation. This code also indicated that if particulate deposition was accounted for, using a typical velocity of 1 m/sec, the release needed to achieve a 25 rem dose at the site boundary increased by a factor of 5.

Table 7-1 lists releases external to the building and their resulting site boundary doses in terms of 50-year committed effective dose equivalent, both with and without deposition. If building leakpath factors are taken into account, the initial source term producing these releases would be orders of magnitude greater if airflow passed through HEPA filtration. Even if ventilation fails and the building is in a relatively undamaged static condition, it would not be unusual for the required initial source term for a given site boundary dose to be at least an order of magnitude greater due to building deposition alone. The examples in this handbook provide initial source terms at the point of generation only, which would equate to the values in Table 7-1 for a completely unmitigated case.

Table 7-1. Site Boundary Doses Associated with Release

Case	100 rem	25 rem	5 rem	1 rem
1. No deposition	3.6 Ci (58 g)	0.9 Ci (14.5 g)	0.2 Ci (2.9 g)	0.04 Ci (0.6 g)
2. Deposition	18 Ci (290 g)	4.5 Ci (73 g)	0.9 Ci (14.5 g)	0.2 Ci (2.9 g)

As noted in the discussion of release models, neither set of values in Table 7-1 is inherently meaningful. Each merely provides a perspective that may be useful under certain circumstances. Either model is only as good as the decisions it leads to. If a model leads to obviously inappropriate conclusions or actions, there is no reason to accord the model any credence. This point is further amplified by the examples.

7.3 MOCK OPERATION EXAMPLES

This section examines major operations described in appendix B. For example purposes, aspects of operations are discussed only where potential new ARF applications or unique circumstances exist. In specific example topic tables, the entry "none" under a subject heading indicates no illustrative examples are presented for that subject, even though it may exist or be possible (i.e., criticality) in the specific example operation under consideration. The entry "N/A" means the subject is not applicable to the specific example operation under consideration. Appendix B provides detailed descriptions of the mock operations examined at a level sufficient to understand the examples, including basic flow diagrams.

7.3.1 Feed Preparation

Release topics explored in this example are listed in Table 7-2.

Table 7-2. Feed Preparation Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- N/A	- Self-sustained oxidation	- Thermal stress - Venting - Free-fall spill - Shock-impact - Aerodynamic entrainment	- Thermal stress packaged waste - Impact - HEPA pressure pulse	- Dry powder

7.3.1.1 Hazard Summary

This activity handles only solid materials in the form of plutonium oxide, plutonium residues, or plutonium metal fines in oxide feed. The only major energy sources in the glovebox are a muffle furnace for burning impure oxide feed and a small impactor that breaks up residues. A 0.21 MPa_g (30 psig) air line supplies a glovebox branch line to provide forced air to the furnace. A 0.1 MPa_g (15 psig) nitrogen supply can provide inerted ventilation for the glovebox, but it is not normally used. There are no large accumulations of combustible material in the glovebox or room, nor is there any reason for the operation to accumulate large amounts of combustible material. Likewise, there are no significant explosive energy sources in the room.

7.3.1.2 Release Estimation

The potential energetic phenomena are a powder spill, ignition of plutonium metal fines, residue impaction, an air line rupture, and a criticality. Additionally, although there is no clear mechanism for a significant fire in the room, one will be examined.

A. Powder Spill. The maximum allowed transfer unit (2000 g as Pu) is larger than the individual batches prepared for dissolution in the feed preparation line (1000 g and 1322 g as Pu). This quantity could be present outside the glovebox in double containers on a transfer cart or in the single inner container in the entry airlock to the glovebox. The most likely place for a spill that could damage the can and discharge its contents would be at the airlock, where the maximum potential fall height would be ~ 1.3 m (4 ft). The DR for such an occurrence is assumed to be 1.0 (i.e., all material spills).

If the material falls inside the can to the ground and the lid of the can is dislodged by impact, the release is not really a free-fall spill. It is perhaps most appropriately modelled as vibration or impact shock. For clumps of bulk powder, the bounding ARF and RF are assessed to be 1E-3 and 0.1 (subsections 4.4.3.3.1 and 4.4.3.3.2). The resulting initial respirable source term is:

$$2000 \text{ g} * 1.0 * 1E\text{-}3 * 0.1 = 0.2 \text{ g}$$

For the purposes of example, a case where the material falls freely through the air is also considered. The ARF and RF assigned for a free-fall spill of powder of less than 3 m are 2E-3 and 0.3 (subsection 4.4.3.1.2). The initial respirable source term is:

$$2000 \text{ g} * 1.0 * 2E\text{-}3 * 0.3 = 1.2 \text{ g}$$

Alternatively, for free-fall spill the ARF may be determined based on the PSPILL equation in subsection 4.4.3.1.2. Although subsection 4.4.3.1.2 only discusses applying the PSPILL equation to fall heights greater than 3 m, it can be used for smaller distances. The equation for bounding ARF is:

$$ARF = 0.2128 \, (M_o^{0.125})(H^{2.37})/\rho_{BP}^{1.02} \qquad (7\text{-}2, 4\text{-}5)$$

where: ARF = airborne release fraction = 1.5E-4
M_o = Mass of powder spilled = 2 kg
H = spill height = 1.3 m, and
ρ_{BP} = bulk density of powder = 2,500 kg/m^3.

The equation as used in this case has already been multiplied by the factor of two necessary to obtain a bounding value. It is noted that the absolute density of plutonium oxide (11.5 g/cm^3) was not used. The equation requires the use of bulk powder density, which must take void volume into account. The value of 2.5 g/cm^3 used is representative of heavy metal oxides such as plutonium oxide.

The value of ARF obtained from this equation is not directly related to the bounding RF of 0.3 assigned. While it may be adequate to assume the 0.3 value for spill distances greater than 3 m, it is not for distances significantly less than 3 m as the small fall distance minimizes the time for particulate interaction and agglomeration. The equation provided in subsection 4.4.3.1.2 for estimating particle size is:

$$AMMD = 12.1 - 3.29(\rho_{BP}) + 7540(ARF) \qquad (7\text{-}3,\ 4\text{-}6)$$

where: AMMD = aerodynamic equivalent mass median diameter = 5 µm,
ρ_{BP} = bulk powder density = 2.5 g/cm^3, and
ARF = airborne release fraction = 1.5E-4.

An AMMD of 5 µm would indicate an RF significantly higher than 0.3, producing a combined ARF x RF of ~ 1E-4. This is a factor of 6 less than the combined ARF x RF (6E-4) assessed for fall distances less than 3 m, and approximately the same as the median ARF x RF (1.5E-4) estimate provided in the handbook for perspective on conservatism. For the types of estimates being made, there is not a major difference between the two values. Either estimate could be defended, but for the sake of conservatism and consistency, the bounding value is used.

B. Ignition of Plutonium Metal Fines. If plutonium metal fines are present in an oxide container opened in an oxygen environment, it is not uncommon to see brief sparking after opening. This is generally not a major concern due to the limited amount of fines. Even if a given small piece of metal sustains smoldering oxidation, the overall fire hazard is minimal. The metal is not in close proximity to a significant amount of combustible material with which to interact; it will generally cease combustion due to heat transfer losses through metal surfaces, and the combustion can easily be extinguished by the operator simply by closing the container.

The hazard identification states that 100 g of metal fines is the maximum amount of metal contamination anticipated in impure oxide received for processing. The absolute limit is 2000 g if an entire feed can contains nothing but metal fines, but the maximum anticipated DR is 0.05. The DR for metal fines present is never

anticipated to be 1.0, but that value can be assumed for the sake of simplicity. Other batches are not affected by the localized combustion.

The ARF and RF assigned for self-sustained oxidation of plutonium metal are 5E-4 and 0.5 (subsection 4.2.1.1.4). The maximum initial respirable source term is:

$$2000 \text{ g} * 1.0 * 5E\text{-}4 * 0.5 = 0.5 \text{ g}$$

The source term decreases to 3E-2 g if the maximum expected DR of 0.05 is used.

As opposed to the plutonium powder spill, it is anticipated that a fines fire would occur inside glovebox confinement, unless the container that spilled outside the box contained fines, a highly unlikely conjunction of events given that the number of containers with significant fines accumulation is small.

C. Residue Impaction. Residue impaction is not an accident. It is part of normal operations. Small amounts of material are driven airborne within the impactor but do not escape the device in significant quantities. The only accident that might release material is a catastrophic failure that results in a large shrapnel pattern from the unit. The design of the unit is supposed to preclude that possibility, but it is included as an example of using release fraction data for unusual circumstances.

Based on the size of a residue batch for the residue dissolver, one feed cycle of the device would not process more than 1000 g as plutonium, and would typically process much less. For the sake of example, it is also assumed that after years of operation, holdup monitors indicate 50 g of plutonium present when the unit is shutdown.

Only a small fraction of the feed material would be in the impaction zone at any one time. The rest would be loaded into the feed hopper, in the rubble output chamber, or in a can at the impactor station waiting to be cycled into the feed hopper. Very complicated models and algorithms could be developed to try and describe the situation. However, a rigorously defensible definition of the exact nature of the catastrophic failure, vis-a-vis the plutonium, is likely unattainable and, in any case, not worth seeking. It provides no new insight into a problem inherent in a mature technology that is already well understood. A bounding MAR of 1050 g with a DR of 1.0 can simply be assumed.

The shock to material when the unit flies apart is not greater than the shock from internal impaction when the unit is operating properly. If release fractions even approaching 1% were experienced in normal operations, significant accountability and

holdup issues would arise. Accordingly, the phenomena can be considered bounded by impact of contaminated non-combustible solids. The ARF and RF assigned are 1E-3 and 1.0 (subsection 5.3.3.2.2). The maximum initial respirable source term is:

1050 g * 1.0 * 1E-3 * 1.0 = 1.0 g

D. Air Line Rupture. Given that the air line is equipped with a flow-restricting orifice and the ventilation system is normally operating, the event is expected to be insignificant. For the purposes of example, it will be assumed that when the line breaks, more flow is attained and/or ventilation flow is secured for whatever reason. Even in these circumstances, the only obvious effect would be limited contamination release from glovebox-mounted HEPA filters due to reverse flow. The break would have to occur in close proximity to powder to significantly affect any in-line material.

The most powder that can be exposed at any one location is 2000 g, the maximum quantity procedurally allowed at any one station. This material can be exposed powder in the loading pan at the furnace or at the weighing and batching station, although less material is typically handled. A maximum of 1000 g of plutonium as contamination on residue can be exposed at the impactor station as well. This material is not as susceptible to release as loose powder. For example, plutonium on magnesium oxide crucible residues may be literally embedded in crucible pores. Treating it like a loose, powdery surface contamination is extremely conservative.

If the air line does not break in close proximity to exposed material, all the material in the glovebox is affected by the overall glovebox phenomena, and the DR for this phenomena is 1.0. If the break is in close proximity to one batch (assume the batch in the furnace pan), the DR is 0.4 for this material (2000/5000) and 0.6 for the different phenomenological effect on the remaining material.

For the expected case where no material is in close proximity to the break and air flow from the pipe, the phenomena is certainly bounded by aerodynamic resuspension of powder exposed to normal ventilation flow. The ARF and RF assigned are 4E-5/hr and 1.0 (subsection 4.4.4.1). The maximum initial respirable source term is:

5000 g * 1.0 * 4E-5/hr * 1.0 = 0.2 g/hr

If the break occurred in close proximity to the furnace storage station (within ~ 30 to 40 cm), the 0.21 MPa_g flow could cause an effect more vigorous than simple resuspension. The effect of any typical air line pressure (e.g., up to 0.7 MPa_g) would be to produce some small level of pressurization in the glovebox and accelerated

airflows in localized areas. The bounding ARF and RF assigned for this phenomena are either 5E-3 and 0.4 (subsection 4.4.2.3.2) for venting of pressurized volumes at pressures less than 0.17 MPa_g or 5E-3 and 0.3 (subsection 4.4.2.2.2) for accelerated airflow parallel to the surface of powders. The higher value is arbitrarily selected, and the maximum initial respirable source term, neglecting the resuspension contribution of material not in proximity to the airflow, is:

$$5000 \text{ g} * 0.4 * 5\text{E-}3 * 0.4 = 4 \text{ g}$$

The most extreme case is if the break results in: (1) the pressurized air line pointing directly at exposed powder no more than 20 cm from the pressurized release; (2) the pressurized air pointed downward at the powder so that the underlying metal surface supports maximum impact; and (3) optimal angle of impact with the powder. In such a case, larger amounts of material are driven airborne, with the total respirable source term being directly dependent on the nature of the powder (adhesiveness and size distribution), characteristics of the surface, and angle and velocity of gas impacting the powder. For all practical intents and purposes, meaningfully estimating a source term for such a highly unlikely condition is an indeterminate problem.

All of the releases discussed above are inside the glovebox and would be precluded by normal use of a flow-restricting orifice in the air line feeding the glovebox. Because the basic purpose of a flow-restricting orifice is to prevent disrupting confinement differential pressures, the application of the release data to the case of glovebox overpressurization is briefly considered as well. If the line break is not in close proximity to any material, the effect on the powder in the glovebox is bounded by the aerodynamic entrainment calculation already made. Given that a significant pressurization is not expected to last more than several seconds, the overall airborne release from process material is, at most, in the milligram range.

The other major potential release mechanism is reverse flow through glovebox-mounted HEPA filters. The reverse pressurizations typically experienced in a glovebox environment are not large enough to damage filters, and the bounding value for a high pressure pulse of gas through filters defined in this handbook is conservative for the effect. The bounding ARF and RF assigned for this phenomena are 2E-6 and 1.0 (subsection 5.4.2.1). If a typical dirty glovebox filter loading of 10 g is assumed, the maximum initial respirable source term is:

$$10 \text{ g} * 1.0 * 2\text{E-}6 * 1.0 = 20 \text{ } \mu\text{g}$$

As is expected from historical experience, the estimated releases are not large.

E. Criticality. A powder criticality in a dry operation is unlikely, but not impossible. A yield of 1E+17 fissions is assessed to be bounding for bare, dry criticalities (subsection 6.2.3.3). A simple method for estimating fission product release from the criticality is to ratio the results indicated by the NRC for a 1E+19 fissions plutonium criticality to 1E+17 fissions. The DR refers to the fraction of isotopes exposed for release, and it would be 1.0 for gases and volatiles in powder.

The ARF is 0.5 for noble gases and 0.05 for radioiodines. Table 7-3 on the following page shows a simplified release estimation based on important isotopes identified by the NRC. The potential dose associated with this release is well under 1 mrem at the site boundary, although prompt worker fatalities can occur due to radiation produced by the criticality. This criticality is a good example of the situation where material release estimates and associated potential doses have little if any relevance to the principal safety concerns for a given phenomena.

Plutonium airborne releases are possible as well. Significant pressurization is not possible with loose (i.e., uncontained powder) or loosely contained powder (i.e., non-pressure tight seal, damaged can). For loose material, however, significant heating can occur. Experimental data covers heating of plutonium oxides to sustained temperatures of 1000 ^{o}C. The ARF and RF assigned for this condition are 6E-3 and 0.01 (subsection 4.4.1.1). Assuming 10,000 g of material are brought together to initiate the criticality, the maximum initial respirable source term is:

$$10{,}000 \text{ g} * 1.0 * 6\text{E-}3 * 0.01 = 0.6 \text{ g}$$

Again, the exact value obtained is of little relevance to criticality safety management.

F. Large Room Fire. There is nothing inherent in the feed preparation glovebox or associated operating and maintenance rooms that indicates a large room fire is possible. However, one can be postulated to occur by an undefined mechanism. The potential MAR is the powder in the glovebox and, at most, one pail of waste in the maintenance room that had been bagged from the glovebox prior to the fire. The maximum amount of material allowed at one time in the glovebox is 20,000 g as plutonium, with all of this material essentially plutonium oxide. Up to 100 g of plutonium can be in a waste package, but 1 to 2 g is typical. If a genuinely large fire existed, the DR for oxide in the glovebox is 1.0, as heating is the release phenomenon of concern. The DR for the waste pail, which would burn, is 1.0 as well.

As previously discussed under the examination of criticality, the ARF and RF for heating oxide powders are 6E-3 and 0.01 (subsection 4.4.1.1). The ARF and RF for

Table 7-3. Important Nuclides Released from Powder Plutonium Criticality

Nuclide	Ci Produced	ARF	Ci Released
Kr-83m	1.1	0.5	0.55
Kr-85m	0.7	0.5	0.35
Kr-85	8E-5	0.5	4E-5
Kr-87	4.3	0.5	2.2
Kr-88	2.3	0.5	1.2
Kr-89	130	0.5	65
Xe-131m	1E-3	0.5	5E-4
Xe-133m	2E-2	0.5	1E-2
Xe-133	0.3	0.5	0.15
Xe-135m	33	0.5	17
Xe-135	4.1	0.5	2
Xe-137	490	0.5	245
Xe-138	110	0.5	55
I-131	0.1	5E-2	5E-3
I-132	12	5E-2	0.6
I-133	1.6	5E-2	8E-2
I-134	43	5E-2	2.2
I-135	4.5	5E-2	0.2

combustion of packaged mixed waste are 5E-4 and 1.0 (subsection 5.2.1.1). The maximum initial respirable source term is:

$$(20{,}000\ g * 1.0 * 6E\text{-}3 * 0.01) + (100\ g * 1.0 * 5E\text{-}4 * 1.0) = 1.3\ g$$

7.3.1.3 Feed Preparation Example Assessment

The feed preparation operation, like any materials-handling activity, has the potential to generate airborne releases. It is not, however, a potential source of large releases, and a

reasonable case can be made that none of the releases calculated are high-frequency events. Except for the highly unrealistic 4-g estimate for the air line rupture, the largest material release estimated is ~ 1 g. Using the most conservative meteorological assumptions for an unmitigated release could produce a site boundary dose of 2 rem, while accounting for deposition alone would reduce the value to well below 1 rem. The 4-g value does not represent an anomaly with regard to this conclusion for the types of estimates being made, where appreciation for actual material behavior is as important as the release data itself.

The most significant factor to consider in this example is that the design insight gained in this evaluation does not come from the release or dose estimates. It comes from recognizing the nature of the unit operations being conducted in a given environment. The need to not spill contained carcinogenic material is self-evident. If the material were being hand-carried in a single can with a partially taped lid, an effective hazard evaluation would indicate this is an unacceptable practice independent of potential release estimates. Likewise, the need to not have rotary machinery that ejects high-velocity fragments due to catastrophic failure is independent of the material being handled. The use of a flow-limiting orifice in air supply lines to a glovebox is a basic design consideration driven by the desire to prevent pressurizing the glovebox. Detailed requirements exist for precluding criticality that are wholly independent of dose considerations. The major insight that can be construed from the release estimates, if thought was not previously focused on the types of stresses causing release, is that it is a good idea to make sure loose oxide is not stationed directly adjacent to supplied air piping.

The main value of the release estimates is the indication that the unmitigated consequences from the feed preparation operation are low and, accordingly, worker safety is the principle focus of evaluation. It is clear that a release external to the building will be insignificant if the airborne material generated is directed through a HEPA filter plenum. That plenum needs to have capabilities to cope with high temperature exhaust from a fire, but the feed preparation operation is not a particularly useful basis for determining those requirements. Further, even if the ventilation system fails, particulate deposition in the static building would be expected to make consequences outside the building small. This is an important perspective on the feed preparation operation, and it may be useful in applying a graded approach to facility management.

7.3.2. Oxide Dissolution

Release topics explored in this example are listed in Table 7-4:

Table 7-4. Oxide Dissolution Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Thermal stress - Free-fall spill	- N/A	- None	- None	- Solution

7.3.2.1 Hazard Summary

This activity receives plutonium oxide powder from the feed preparation operation via a conveyor. This material is fed into a nitric acid dissolver, after which only liquid materials are handled. 64% nitric acid is fed into the dissolver by a dedicated feed tank on the second floor, and 35% nitric acid is added to subsequent vessels by a common feed tank servicing all three dissolving line gloveboxes. Process and instrument air and nitrogen are fed into the glovebox by pressurized sources ranging from 0.1 to 0.55 MPa_g (15 to 80 psig). The dissolver is heated by 0.21 MPa_g steam (35 psig). Transfers between tanks in the glovebox are by vacuum eduction. There are no highly energetic reactions associated with the dissolution process itself. Radiolysis of water can produce small amounts of hydrogen over long periods of time, but the vessels are supplied with an ever-open vent to the vessel vent system and a continuous small air purge while in use.

7.3.2.2 Release Estimation

The potential energetic phenomena are a liquid spill, boiling liquid due to dissolver temperature control malfunction, and a liquid criticality. There is no clear mechanism for a large room fire, and one is not examined because its only effect would be liquid boiling.

A. Liquid Spill. The dissolution process is a batch operation. The maximum feed allowed in any one batch is 1322 g as plutonium (1500 g PuO_2). A spill can occur from a piping or vessel leak due to corrosion, or inadvertent damage from an activity such as maintenance or an unrelated accident. If the leak in a line is small, or a leak in a vessel is above the vessel bottom, not all of the material would be spilled. For the sake of simplicity, a leak large enough and situated so as to allow all of the liquid to drain from confinement is postulated (i.e., DR = 1.0). At a vessel low point, the maximum possible fall height is ~ 0.3 m (1 ft), while a leak from a transfer line could produce a fall of ~ 1.2 m (4 ft).

7.0 Application Examples; Dissolving Operations Examples

The key question in matching phenomena is whether or not the release will act predominantly as a free-fall spill of liquid or, for a vessel leak, as a depressurization of liquid via a failure under the liquid surface level. The only pressure acting on the liquid is its own static head, since the vessel itself is under a slight vacuum. The static head of the liquid can be determined from the equation:

$$P_{sh} = H*\rho*g \qquad (7\text{-}4)$$

where: P_{sh} = static head pressure (Pa),
H = height of fluid (m),
ρ = fluid density (kg/m^3), and
g = acceleration due to gravity (9.8 m/sec^2)

The 30-liter slab vessel has a height of 0.75 m (2.5 ft), with 20 cm of freeboard during operation. This leaves a liquid height of 0.55 m (1.8 ft). The 64% nitric acid solution has a density of ~ 1400 kg/m^3. These values yield a static pressure head at the bottom of the vessel of 7550 Pa (~ 1 psi). At this low pressure, the depressurization spray effect will be immaterial.

The density of the solutions in the oxide dissolution glovebox exceeds 1.2 g/cm^3 at all points in the process, so the spill is modelled as that of a concentrated, heavy metal solution. The ARF and RF assigned for a free-fall spill of a concentrated heavy metal solution of less than 3 m are 2E-5 and 1.0 (subsection 3.2.3.1). The initial respirable source term is:

$$1322 \text{ g} * 1.0 * 2\text{E-}5 * 1.0 = 3\text{E-}2 \text{ g}$$

This source term would be generated within glovebox confinement. Although subsection 3.2.3.1 discusses applying the spill ARF equation only to fall heights greater than 3 m, it can be used for smaller distances. The equation for bounding ARF is:

$$ARF = 8.9\text{E-}10\ (\rho_{air}{}^2 * H^3 * g / v_{soln}{}^2)^{0.55} \qquad (7\text{-}5,\ 3\text{-}13)$$

where: ARF = airborne release fraction,
ρ_{air} = density of air (1.18E-3 g/cm^3),
H = spill height, (cm),
g = acceleration of gravity (981 cm/sec^2), and
v_{soln} = solution viscosity (poise).

The additional multiple of 3 discussed in relation to this equation in subsection 3.2.3.1 is not used as it applies only to solutions with a density less than 1.2 g/cm^3.

The maximum fall height is 1.2 m (4 ft), and the viscosity of 60% nitric acid at ambient temperature is 2E-2 poises. These values yield an ARF of 5E-6. After initial dissolution, however, and during initial transfer to the vacuum run tank, the solution temperature is between 90 and 100 ^{o}C, not ambient. If the spill occurs at that point, solution viscosity is decreased to 7E-3 poise. The resulting ARF would be 1.5E-5. As for the powder spill examined for the feed preparation operation, the bounding value of 2E-5 is used for the sake of conservatism and consistency.

B. Boiling Liquid. If the temperature control system on the dissolver heat exchanger malfunctions, sufficient steam flow could occur to initiate boiling. Testing of the heat exchanger at full capacity has indicated that complete boiloff of surrogate water in the vessel does not result in significant vessel pressurization as the vessel vent system can handle the rate of vapor generation. However, if the condition occurs during operation and remains undetected, the entire contents of the dissolver can eventually be boiled, with the resulting vapor flowing to the vessel vent system. In this case, the MAR would again be 1322 g with a DR of 1.0.

The ARF and RF assigned for continuous boiling of liquids are 2E-3 and 1.0 (subsection 3.2.1.2). The maximum initial respirable source term is:

$$1322\ g * 1.0 * 2E\text{-}3 * 1.0 = 2.6\ g$$

This source term is generated within glovebox and piping system confinement.

C. Criticality. A solution criticality is unlikely in the geometrically favorable dissolvers. Mechanical distortion of the vessel and/or severe overbatching of feed could, however, produce a criticality. A yield of 1E+19 fissions is assessed to be bounding for solution criticalities (subsection 6.2.3.1). A simple method for estimating fission product release from criticality is to use NRC results for a 1E+19 fissions plutonium criticality. The DR refers to the fraction of isotopes generated in the criticality exposed for release, and is 1.0 for gases and volatiles in solution.

The ARF is 1.0 for noble gases and 0.25 for radioiodines in solution based on NRC precedents. Table 7-5 shows a simplified release estimation based on important isotopes identified by the NRC. The potential dose from this release is well under 100 mrem at the site boundary, although prompt worker fatalities can occur due to direct radiation.

Table 7-5. Important Nuclides Released from Plutonium Solution Criticality

Nuclide	Ci Produced	ARF	Ci Released
Kr-83m	110	1.0	110
Kr-85m	71	1.0	71
Kr-85	8E-3	1.0	8E-3
Kr-87	430	1.0	430
Kr-88	230	1.0	230
Kr-89	13,000	1.0	13,000
Xe-131m	0.1	1.0	0.1
Xe-133m	2.2	1.0	2.2
Xe-133	27	1.0	27
Xe-135m	3300	1.0	3300
Xe-135	410	1.0	410
Xe-137	49,000	1.0	49,000
Xe-138	11,000	1.0	11,000
I-131	11	0.25	2.8
I-132	1200	0.25	300
I-133	160	0.25	40
I-134	4300	0.25	1100
I-135	450	0.25	110

Plutonium airborne releases have been estimated by the NRC as well. The generic criticality assumed for solutions was an initial burst of 1E+18 fissions followed at 10-minute intervals for 8 hours by bursts of 2E+17 fissions for a total of 1E+19 fissions. The criticality was considered terminated by the eventual evaporation of 100 liters of solution over the 8-hour period. The evaporation process over 8 hours would not be characterized by continuous, vigorous boiling. The NRC assigned a respirable release fraction of 5E-4 to the amount of plutonium in the 100 liters that evaporated. This value is between the respirable ARF of 3E-5 for heating solutions (subsection 3.2.1.1) and the ARF of 2E-3 for continuous boiling (subsection 3.2.1.2) and is considered appropriate for the purposes intended.

The dissolution vessel holds only 23 l of solution. In the larger glovebox accountability slab tank, sufficient nitric acid has been added for cooling and dilution that 125 l of solution are present. If the criticality is postulated to occur in the dissolver, the 23 l versus 100 l discrepancy can be ignored for the sake of simplicity. The DR assigned for the plutonium in solution is 1.0. If the criticality were assumed to occur in the accountability tank, a DR of 0.8 (100/125) can be used due to the 100 l evaporation basis assumed. Keeping in mind that the purpose of the release model is not to derive the "absolute" answer, it is fitting, at least initially, to use a DR of 1.0 and accept the result as conservative. If a double batching of plutonium is arbitrarily considered the source of the criticality, the respirable airborne release is:

$$(1322*2)\ g * 1.0 * 5E\text{-}4 * 1.0 = 1.3\ g$$

As in the previous powder criticality example, the exact value obtained is of little relevance to criticality safety management.

7.3.2.3 Oxide Dissolution Example Assessment

The release estimates associated with the oxide dissolution line are comparable to those associated with the feed preparation line (subsection 7.3.1.3), with the maximum estimate of 2.6 g for boiling dissolver solution being twice the expected bound for feed preparation. Using the most conservative meteorological assumptions, potential consequences at the site boundary are below 5 rem, and below 1 rem if deposition is accounted for.

Actual operational insight generated by the numbers is again small compared to the total operational considerations for the oxide dissolution line described. The vessels and piping need to be capable of withstanding temperatures and acid concentrations associated with the process, but no release estimate was needed to make that realization. Such a simple, informal verification would be a matter of course in any hazard analysis. The main insight potentially attainable is, again, a new insight only if thought was not previously focused on types of stresses causing release. It is that the dissolver heating cycle can significantly contaminate vessel vent piping and tanks due to a single failure (e.g, temperature indication low, steam flow valve sticks). Even though this process upset condition is not expected to directly produce an occupational exposure, it is worth preventing at some level of cost to minimize future decontamination and decommissioning difficulties. Accordingly, while the issue does not need high level attention, such as safety-class or safety-significant structure, system, or component designation, it is a desirable goal for the design and operation of the process to be aware of the issue and, to a reasonable degree, minimize the chance of the event and of a failure to detect it.

The main value of the release estimates is again the indication that the unmitigated consequences from the oxide dissolution are low and, accordingly, worker safety is the principle focus of concern. This is an important perspective that, again, may be useful in applying a graded approach to facility management.

7.3.3 Residue Dissolution

Release topics explored in this example are listed in Table 7-6.

Table 7-6. Residue Dissolution Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Venting of pressurized liquids	- N/A	- N/A	- None	- None

7.3.3.1 Hazard Summary

This activity receives, via a conveyor, residues heavily contaminated with plutonium that have been reduced in size in the feed preparation operation. This material is placed in a feed hopper that slowly feeds it into the nitric acid dissolver, after which only liquid materials are handled. This operation is virtually identical to the oxide dissolution operation previously described. The only new complication in residue dissolution is the need for a screw feed conveyor to prevent an excessive dissolution rate of residue impurities from causing an eructation in the dissolver vessel.

7.3.3.2 Release Estimation

The only new potential energetic phenomenon is an eructation from the dissolver vessel due to an excessive impurity feed rate.

A. Dissolver Eructation. The dissolution process is a batch operation. The maximum feed allowed in any one batch is 1000 g as plutonium. The maximum release is obtained if the eructation occurs when the majority of the plutonium has been fed to the dissolver. The eructation is a relatively low pressure phenomena, and any gas backleakage through the material-filled conveyor would have almost no effect on the feed residue. For an initial bound, a DR of 1.0 is conservatively assumed.

Small eructations are handled by the vessel vent system. A larger eructation can drive gas into the glovebox. In a worst case, the dissolver blowout plug may fail,

and momentary reverse pressurization of the glovebox can occur. However, the actual pressure generated in the dissolver is not large, and is certainly below the 0.345 MPa_g (50 psig) threshold this handbook defines for differentiating a high-pressure release from a low-pressure release. The ARF and RF for depressurization of liquid containment via a failure above the liquid level at less than 0.345 MPa_g are 5E-5 and 0.8 (subsection 3.2.2.3.2). The initial respirable source term is:

$$1000\ g * 1.0 * 5E\text{-}5 * 0.8 = 4E\text{-}2\ g$$

This source term is generated within glovebox confinement.

7.3.3.3 Residue Dissolution Example Assessment

See oxide dissolution example assessment in subsection 7.3.2.3.

7.3.4 Metal Dissolution

Release topics explored in this example are listed in Table 7-7.

Table 7-7. Metal Dissolution Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Explosion shock effect - Venting of pressurized liquids	- Explosion shock effects	- Energetic oxidation reaction	- None	- None

7.3.4.1 Hazard Summary

Recycle metal is brought into the recovery facility in a ready-to-dissolve form after receipt in the adjoining Plutonium Vault Facility. The inner can containing the metal is brought into the glovebox via an airlock. The metal is removed and placed in a small spray chamber sitting on top of a slab vessel similar to the oxide and residue dissolvers. First, 14% by weight sulfamic acid is added to the slab vessel from a dedicated feed tank. It is circulated through the spray chamber via a small centrifugal pump to dissolve the metal and generate a plutonium solution. Process and instrument air and nitrogen are fed into the glovebox by pressurized sources ranging from 0.1 to 0.45 MPa_g (15 to 65 psig). A heat exchanger using 0.21 MPa_g steam (35 psig) is used to heat the circulating acid. When the process is complete and the circulating plutonium solution is diverted to the accountability slab tank, 4% nitric

acid is added to the dissolving slab tank and circulated to insure any hydride generated has been passivated. Additionally, 35% nitric acid is also supplied to the glovebox to adjust product acid concentration.

Plutonium hydride sludge, which can become oxide in a spontaneous exothermic reaction under certain conditions, can be generated if the dissolution temperature is not a minimum of ~ 50 °C. It is desired to keep the temperature below ~ 71 °C to prevent dissolving an impurity associated with the metal. Hydrogen gas is also generated by the dissolution reaction at a rate that increases with solution temperature, presenting a potential explosive hazard. The development of an explosive concentration of hydrogen is prevented first by controlling steam flow to the heat exchanger by temperature indication from the slab vessel. High temperature in the dissolving spray chamber will activate an alarm at 64 °C and turn off the centrifugal pump circulating acid at 68 °C. Securing acid flow results in a rapid reaction rate decrease as the limited acid supply in the spray chamber depletes.

The second means to prevent developing an explosive concentration of hydrogen is a continuous air purge of the spray chamber supplied by the process air system. The purge exhaust goes to the vessel vent system and is monitored by a hydrogen detector that also turns off the centrifugal pump if hydrogen concentration in the offgas exceeds 2.5% (4.0% is lower flammable limit).

7.3.4.2 Release Estimation

The potential energetic phenomenon not previously covered in other examples are a pump or piping failure causing liquid spray, a hydrogen explosion, and plutonium hydride oxidation.

A. Liquid Spray. The use of a centrifugal pump for liquid circulation generates positive pressure. While the pressure is not high in this small process, it is sufficient to produce liquid spray and thus a different release stress than the vacuum transfer systems in other dissolution lines. A pump seal, flange failure, or even a piping leak could cause spray generation. The maximum amount of material available in solution is 1200 g of plutonium if all of the metal is dissolved. The DR will probably not be 1.0 even if no operator intervention occurs as the pump eventually shuts off from loss of net positive suction head after sufficient liquid is lost. The distinction, however, could be minor; therefore a DR of 1.0 is used for the sake of simplicity in this example.

The phenomena is bounded by the depressurization of liquid via a failure under the liquid surface level. The ARF and RF for this phenomena, which are based on liquid

spray through commercial nozzles, are 1E-4 and 1.0 (subsection 3.2.2.3.1). The initial respirable source term is:

$$1200 \text{ g} * 1.0 * 1E\text{-}4 * 1.0 = 0.1 \text{ g}$$

This source term is generated within glovebox confinement.

B. Hydrogen Explosion. The original metal feed is 5 moles of Pu-239. The stoichiometry of the dissolution reaction generates 1.5 moles of hydrogen for every mole of plutonium dissolved, allowing a maximum of 7.5 moles. At ambient glovebox conditions, this is ~ 190 l of gaseous hydrogen. The glovebox holding the process is 4.5 m long, 1.2 m wide, and 1.8 m tall. Even allowing for 25% occupancy by equipment, the total volume of the glovebox is over 7500 l. Uniformly dispersing the maximum attainable hydrogen in the glovebox produces a volumetric concentration of 2.5%. The lower flammability limit of hydrogen in air is 4.0%. Given that the 170 l of hydrogen cannot be instantaneously generated and the ventilation flowrate through the glovebox is over 10,000 lpm, a hydrogen explosion is not a major concern in the glovebox air volume.

The hydrogen generation rate peaks at the beginning of the cycle, when both metal surface area and acid concentrations are at their maximum. At that point, the rate of hydrogen generation is ~ 0.08 ml/(cm^2 sec) at the lower end of the operating temperature range and ~ 0.3 ml/(cm^2 sec) above the upper end based on experimental data obtained by the site technical laboratory. To estimate hydrogen generation rates, it is necessary to know the size of the metal piece being dissolved.

The minimum density for plutonium metal is ~ 16 g/cm^3 in the delta phase, as opposed to ~ 19.9 g/cm^3 in the alpha phase. For example purposes, the minimum density is assumed as this will yield the largest surface area. To avoid discussing details of the metal, it is assumed to be in the shape of a square wafer with a thickness of 2 cm. The 1200 g of plutonium metal would occupy a volume of 75 cm^3 using the minimum density. For the given thickness of 2 cm, the length of a square side is 6.1 cm. If the metal surface is assumed to be fully available for dissolution on all surfaces, which it is not, the total surface area available is 123 cm^2. The corresponding minimum and maximum initial hydrogen generation rates are:

Minimum: $0.08 \text{ ml/(cm}^2 \text{ sec)} * 123 \text{ cm}^2 = 10 \text{ ml/sec}$

Maximum: $0.3 \text{ ml/(cm}^2 \text{ sec)} * 123 \text{ cm}^2 = 37 \text{ ml/sec.}$

The actual reaction rate will drop off from these values fairly quickly, but if the values were sustained for a minute, the amount of hydrogen generated would range from 0.6 to 2.2 l. The dissolving chamber and the slab solution vessel have 2 l and 14 l of gas space available while operating. For the slab vessel alone, the bounding range of hydrogen estimated would yield concentrations of 4.2% and 16% for the minimum and maximum cases respectively. Therefore, the attainment of explosive conditions within the operating vessels themselves is a legitimate source of concern.

Given that an explosion in the vessels is considered feasible, the question arises as to whether the explosion should be considered a deflagration or a detonation? The vessels represent the main volume for hydrogen gas collection due to their size and the placement of flame arrestors in the vent piping. Both vessels are vented enclosures, as opposed to confined; there are no unique or highly energetic ignition sources available; and the hydrogen generation rate and potential accumulations over time are fundamentally limited. These factors indicate a detonation is not expected.

The hydraulic diameter for vessels such as the slab dissolver is obtained by multiplying the ratio of cross-sectional area to perimeter by a factor of 4. When dissolution is occurring, the controlling dimensions for hydraulic diameter are the 20 cm freeboard in the vessel and the 7.6 cm slab separation in which liquid is held. The cross-sectional area is 152 cm^2 and the perimeter is 55 cm, yielding a hydraulic radius of 11 cm for a vessel with a length of 91 cm. The corresponding length to hydraulic diameter ratio is therefore 8.3, which is below the minimum value of 10 necessary to consider transition to detonation (NFPA 68). The ratio for the dissolving chamber is even less favorable.

The appropriate course for evaluating the accident source term is to consider the event a deflagration. However, for the purposes of example, both deflagration and detonation estimates will be made. For the case of a deflagration, the significant venting area provided for the vessel in the form of an always open vent line and a large, low-pressure blow-out plug is likely to prevent extensive explosion damage to the surrounding environment. Between the vessel vent line and the blow-out plug, the total available venting area is greater than the vapor space cross section, in accordance with National Fire Protection Association guidelines (NFPA 68). Accordingly, the vessel itself would be expected to survive. If it did, the release would occur from the top of the vessel. This would correspond to the conditions of depressurization of liquid containment via a failure above the liquid level at less than 50 psig previously discussed in the residue dissolution example of section 7.3.3. In this case, however, little gas absorption in liquid would occur, and the associated release could be essentially zero depending on the critical freeboard height.

However, since this value is essentially indeterminate, the conservative bounding ARF and RF values of 5E-5 and 0.8 (subsection 3.2.2.3.2) are used. It is also assumed that dissolution is virtually complete so all of the plutonium feed is in solution in the slab vessel. The resulting initial respirable source term is:

$$1200 \text{ g} * 1.0 * 5E\text{-}5 * 0.8 = 5E\text{-}2 \text{ g}$$

If it is assumed that the vessel failed, an additional effect is liquid free-fall spill less than 3 m, for which the ARF and RF are 2E-4 and 0.5 or 2E-5 and 1.0 (subsection 3.2.3.1) depending on the density of the solution. The lesser concentration of the sulfamic acid (14%) may warrant use of the larger value, in which case an additional 0.1 g is added to the source term, making the spill phenomena dominant.

If detonation phenomena is assumed, the TNT equivalent must be estimated. The maximum explosive energy potential occurs when a stoichiometric mixture of hydrogen and air exists (i.e., 30% hydrogen by volume). If the explosion occurred in the slab dissolver, stoichiometric conditions would involve 4.2 l of hydrogen in the 14-l vapor space. At standard temperature and pressure (22.4 l/mole), 0.2 moles of hydrogen would be available. Correction for actual temperature is neglected as it would only reduce the moles of hydrogen present. The available energy is ~ 13,700 calories for a heat of combustion of 68,317 cal/mole H_2. This value corresponds to 12.5 g of TNT (1100 cal/g). The value for an explosion in the 2-liter dissolving chamber vapor space would be ~ 2 g of TNT.

A detonation can be assumed to occur in either the dissolution chamber or the slab vessel. If it is assumed to happen in the slab vessel, which would have to be considered confined, the maximum TNT equivalent for a stoichiometric mixture of air and hydrogen is calculated to be 12.5 g. Subsection 3.2.2.1 indicates that it is bounding to assume an amount of inert material equal to the mass of TNT equivalent becomes a respirable aerosol. In this case, that would be 12.5 g of solution. Assuming an aqueous solution density of 1.1 g/cm^3 yields an airborne release of ~ 12 ml of solution from the available total of 30 l. Because the plutonium is considered uniformly distributed in the solution, the resulting ARF x RF value is 4E-4 (0.012 l/30 l). If the dissolution process is considered almost complete at the time of the explosion (i.e., 1200 g Pu available in solution), the initial respirable release is:

$$1200 \text{ g} * 1.0 * 4E\text{-}4 = 0.5 \text{ g}$$

If the detonation is assumed to occur in the dissolving chamber, the release will be maximized if plutonium metal is the affected material because it is not diluted

throughout an inert volume. It is noted, however, that the effects cannot realistically be confined to one material. The TNT equivalent previously calculated for the dissolving chamber was 2 g, which would release a corresponding 2 g of plutonium as a respirable aerosol. It is emphasized again that the use of a detonation model to bound the physical circumstances described in this example is unwarranted. The maximum error of this type would be to assume that since 7.5 moles of hydrogen can theoretically be generated over the entire dissolution process, this is the amount of material involved in a detonation in proximity to the material. The resultant TNT equivalent would be ~ 470 g, for which a 470 g release of plutonium could be postulated. To use such an estimate as a meaningful estimate of potential consequences would be ridiculous.

C. Plutonium Hydride Oxidation. If the dissolution temperature is less than 50 ^{o}C, plutonium hydride sludge may be formed due to metal-hydrogen vapor interactions at relatively slow reaction rates. Experience indicates that such sludge may account for, at most, 5% of the original metal charge, or ~ 60 g. The hazard identification indicates 100 g is a generally accepted upper limit due to the fact that sludge formation must compete against the still thermodynamically favored normal dissolution reaction even at lower temperatures. This sludge will also dissolve if dissolution temperature exceeds 50 ^{o}C during the overall process.

The plutonium hydride sludge produced in the dissolver is actually a damp oxide + hydride mixture. As long as it has been passivated by dilute nitric acid and remains damp, it can be recycled into the dissolution process without risk. If, however, the material becomes dry, highly exothermic reactions may result. Finely divided hydride is pyrophoric in air at room temperature. This is due to the rapid oxidation reaction that produces PuO_2 and H_2. The reaction has been observed in materials contaminated with hydride once exposure to air occurs. The hydride is not a detonable material, nor is the hydrogen generated expected to be an explosive hazard unless unusual confinement conditions exist.

It is assumed that 100 g of hydride sludge was generated in the process and has been incorrectly handled, allowing it to dry in the glovebox atmosphere. Several phenomena could be used to model the potential airborne release from initiation of the oxidation reaction. The main concern is that the material could become a fire source, but that would not be a major problem in the metal dissolution glovebox, which contains very little combustible material. The release phenomena could be considered similar to heating of reactive powders, for which the ARF and RF are 1E-2 and 1E-3 (subsection 4.4.1.2). However, the phenomena is a combustion reaction, and the

ARF and RF for burning of plutonium metal, at 5E-4 and 0.5 (subsection 4.2.1.1.3), yield a considerably higher ARF x RF (2.5E-4 vice 1E-5).

The plutonium metal reaction phenomena will not be the same as the hydride reaction, but Table 4-8 of subsection 4.2.1.1.4 indicates that highly energetic combustion of plutonium-sodium mixtures at 300 ºC exceeded the 2.5E-4 ARF x RF value in only 2 of 12 successful ignition experiments. The two larger ARF x RF values obtained were 4E-4 and 2E-3, with the other 10 combined values being comparable to heating of reactive powders or less. Subsection 4.2.1.1.4 states that the ARF and RF of 1E-2 and 1.0 for disturbed molten metal can be applied to very energetic compound reactions. This value is accepted as very conservative and used. Such an approximation, however, is not intended to cover all situations where small amounts of hydride may be present intermingled with other materials. The initial respirable release is:

$$100 * 1.0 * 1E\text{-}2 * 1.0 = 1.0 \text{ g}$$

7.3.4.3 Metal Dissolution Example Assessment

The source term determined for the hydrogen explosion might exceed a dose of 100 mrem at the site boundary if the release were completely unmitigated. Using the unrealistic estimates of stoichiometric detonation of a limited quantity of hydrogen could possible result in exceeding a dose of 1 rem. These are not major doses for an unlikely accident. Examination of the explosion, however, would be a primary focus of any design or evaluation process because it is not necessary to even know what is being processed to know that an explosion, and especially a detonation, must be precluded if possible.

As was previously noted, the second means to prevent developing an explosive concentration of hydrogen in the example is a continuous air purge of the spray chamber supplied by the process air system. In systematically evaluating the functioning of the dissolver system, the original hazard analysis training allowed discovery of a design flaw based on actual precedent within the DOE complex. The air-operated valve on the air purge line is discovered to be an air-to-open valve. Therefore, a loss of instrument air will cause the valve to shut and the air purge to be lost with metal dissolution still occurring. Given that the instrument and process air systems are unrelated, and the supply line is equipped with a check valve to prevent reverse flow, the only functional need in this regard is for the valve to be an air-to-close valve. This is an example of a safety improvement or correction that can easily be determined without the need for a source term calculation. As presented in the example, it is not an uncovered flaw for which blame must be fixed. It is an opportunity to make a tangible improvement in safety, which is the purpose of the evaluation.

The questioning nature of an evaluation independent of source term is apparent in other questions that could be asked in this example. Is it worth considering altering the system so as to use nitrogen as the purge gas instead of air? Should a branch of the nitrogen line be attached to the purge line to provide back-up purging capability if process air is lost? The hydrogen deflagration event discussed above does not seem to warrant a large, complicated set of redundant safeguards, but it is important to at least consider such issues in an evaluation. Other obvious questions exist with regard to the heating system. If there is a heat exchanger leak, and steam flow has been shut off due to high temperature in the slab tank, pressure on the steam condensate line will be quickly lost and process solution will leak into the condensate line. The process steam condensate is collected in raschig ring tanks that are sampled for contamination twice before being returned to the site steam plant. Is that good enough? Should a change in the operation of the heat exchanger be considered? What is the proper tradeoff between worker man-rem, potential contamination, or even waste generation and increased maintenance to check for leaks? There are no "right" answers per se to these questions, and the original operation is not negligent simply because the questions can be asked.

These are the kinds of considerations other than source term estimates mentioned at the beginning of this chapter that are related to developing a simple understanding of why things are done the way they are. Such considerations are an example of why the Introduction to this document states that the source term formula alone cannot be a vehicle for determining the complete facility safety basis. It would be a misuse of the overall process in which ARF models are developed to simply note that site boundary consequences are below a given value and do no further evaluation. In this example, such an approach would have continued to overlook improper use of an air-to-open valve on the purge line that creates an obvious mechanism for obtaining explosive concentrations of hydrogen in a vessel. Likewise, it would be extremely inefficient to try to use a restrictive dose criteria that would force "backing into" the problem. The ARF and RF data of this handbook are not an effective mechanism for fine parsing of results, or for replacing process and operational knowledge.

The main value of the release estimate obtained by application of ARF values indicates that the accident consequences are not large, let alone catastrophic, which places it firmly within the basic spectrum of industrial accidents. As part of a graded approach to safety, it does not demand any unique or excessive expenditure of resources beyond industrial norms. That expenditure would rightly include a basic explosion analysis, of the type summarized in this example, indicating that detonation is not a major issue and vessel venting capability is adequate. Such a basic analysis needs to be performed if one does not already exist on file for the metal dissolution process. The analysis would not, however, be expected to require integration with a large-scale quantitative risk assessment or extensive computer modelling.

7.3.5 Liquid Sampling and Tank Farm

Release topics explored in this example are listed in Table 7-8.

Table 7-8. Liquid Sampling and Tank Farm Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Thermal stress - Venting liquid below surface level - Venting superheated liquid - Free-fall spill	- N/A	- N/A	- None	- None

7.3.5.1 Hazard Summary

This activity involves only liquid material. Plutonium process and liquid waste solutions are stored in annular tanks in three small rooms located on the west wall of the wet processing line maintenance room. These tanks receive solution and transfer it to gloveboxes via small gear pumps. The sample glovebox, located adjacent to the tank storage areas, uses small centrifugal pumps to circulate and sample tanks. The tanks are equipped with ever open vents to the vessel vent system and pressure relief lines leading to a raschig ring sump. Other than pump discharge fluid pressure of 0.14 MPa (20 psia), the major energy source in the area is a branch-line from the 0.45 MPa_g (65 psig) instrument air main for remote operation of tank inlet and outlet valves.

The tanks are geometrically favorable, not geometrically safe, so a solution criticality is possible. Specific mitigation features include the raschig ring sump for spills and, for misrouting of material, a relatively common annular design so that any one tank can hold the maximum flowsheet plutonium concentration while remaining subcritical. Leaks and spills are possible. There is no significant combustible loading in the area, but a large room fire could boil liquid and potentially pressurize tanks. The hazard evaluation noted that no documented estimation of tank venting capability had been made with regard to potential heat fluxes from a fire.

7.3.5.2 Release Estimation

The only new potential energetic phenomenon presented by this example is superheating of liquid solutions due to a fire. However, liquid spills are examined as well because this operation can be perceived to be different from previous examples and involves more material.

A. Liquid Spill. Spills for the tank farm and sampling operation can occur as small drop leaks, pressurized spray releases, or free-fall spill from piping or tank failure. The waste tanks will be ignored in this example as any given waste tank only holds less than 5 g of plutonium when full. Maximum operational tank MARs and solution concentrations by type and number of available tanks are: 1320 g for ion exchange feed at ~ 7 g/l (6 tanks); 5830 g for ion exchange eluate at ~ 30 g/l (2 tanks); and 11,650 g for precipitator feed at ~ 90 g/l (2 tanks). The ion exchange eluate and precipitator feed solutions have densities between 1.1 g/cm^3 and 1.2 g/cm^3, which will result in higher spill release fractions.

The plutonium solution piping outside gloveboxes is sheathed, annular piping with leak detection points. The first example will consider a small leak inside a glovebox. The leak could also be assumed to occur in the maintenance rooms for the wet processing or sampling lines if accumulated liquid in the annular piping region was dripping from a leak detection point. That effect would be the same as if unconfined, single piping were used.

In a number of pipetting operations, 20 drops is a standard for 1 ml. It is assumed that this is the drop size leaking from piping so that each drop contains 0.05 ml. If the leaking solution is at the maximum concentration of 90 g/l, each drop contains 4.5E-3 g, or 4.5 mg. The ARF and RF for free-fall spill of an aqueous radionuclide solution (density less than 1.2 g/cm^3) less than 3 m are 2E-4 and 0.5 (section 3.2.3). The initial respirable source term is:

$$4.5 \text{ mg} * 1.0 * 2E\text{-}4 * 0.5 = 0.45 \text{ µg}$$

If the leak rate is 1 drop/sec, the airborne release is almost 5 µg in 10 seconds, 30 µg in a minute, 1.6 mg in an hour, and 39 mg in a day.

If a tank experiences a large failure, or a pipe is sheared in half, a continuous flow of liquid would experience free-fall spill release. If the entire contents of the most heavily loaded tank is released in such a manner, the initial airborne source term is:

11,650 g * 1.0 * 2E-4 * 0.5 = 1.2 g

If all process tanks are full at their flowsheet parameters, the MAR is 42 kg. The free-fall spill of all this material produces an initial airborne source term of:

(7920 g * 1.0 * 2E-5 * 1.0) + (34,960 g * 1.0 * 2E-4 * 0.5) = 3.7 g

A pipe or tank failure that resulted in the release of a pressurized spray due to pump pressure, would produce the same release as free-fall spill for the lower density solutions (i.e. < 1.2 g/cm^3), and a greater release for the high-density solutions. This scenario would be similar to the spray leak examined for the metal dissolver in subsection 7.3.4.2. The physical phenomena is depressurization of liquid via a failure under the liquid surface level, for which the ARF and RF are 1E-4 and 1.0 (subsection 3.2.2.3.1). If the spray failure occurs for one of the high density ion exchange feed tanks, the initial airborne source term is:

1320 g * 1.0 * 1E-4 * 1.0 = 0.1 g.

The free-fall spill release for the high-density solution is only 0.03 g.

B. Large Room Fire. There is no obvious mechanism for postulating a large room fire in the maintenance side of the wet processing line, but one is postulated from an undefined source. Unlike the previous case where an undefined large fire was postulated (Feed Preparation example, section 7.3.1), it will cause a significant increase in source term for the process. The principle mechanisms for release are boiling of liquid and release of superheated liquid.

Before estimating releases, it is appropriate to consider the type of fire that would significantly affect the stored liquid. The precipitator feed tanks (13 and 14) have both the largest plutonium (11,650 g) and smallest liquid inventories (130 l). The heat required to boil and vaporize this solution is estimated assuming the thermodynamic properties of water, the density of the nitric acid solution, and an initial temperature of 30 ^oC. The sensible heat needed to raise the temperature to boiling is:

130 l * 1000 cm^3/l * 1.3 g/cm^3 * 1 cal/g ^oC * (70 ^oC) = 1.2E+7 calories

The latent heat needed to vaporize the solution is:

130 l * 1000 cm^3/l * 1.3 g/cm^3 * 539 cal/g = 9.1E+7 calories

The total energy required to boil all of the solution in one tank is 1E+8 calories. NUREG-1320 gives simple correlations for heat generated by wood and kerosine fires from which estimates of the amount of wood and kerosine that would produce 1E+8 calories can be made. Barring extreme configurations, assuming that one-tenth of the energy liberated by the burning material is absorbed by the liquid in the tanks is conservative. This assumption will be used to estimate the range of fuel quantities needed to completely vaporize the liquid.

The amount of material needed can be determined by the rearranged equation:

$$M_b = Q_a / (X_a * H_t) \qquad (7\text{-}6)$$

where: M_b = fuel burned (g),
Q_a = actual heat released (calories),
X_a = theoretical heat release efficiency, and
H_t = fuel heat of combustion (cal/g).

The general heat of combustion of wood is 4300 cal/g, and the actual heat release is 70% of the theoretical value. Designating Q_a as 1E+8 calories yields 33,000 g of wood. It is doubtful that large accumulations of hardwood, such as oak or mahogany, would be present, so a typical density of 450 kg/m^3 for cheaper wood is assigned. Taking into account the factor of 10 previously mentioned, the amount of wood fuel necessary to evaporate a precipitator feed tank is between 0.07 and 0.7 m^3 (2.5 to 25 ft^3). A linear relationship can be assumed for quantities of fuel necessary to evaporate less than the entire tank contents.

The general heat of combustion of kerosine is 11,000 cal/g, and the actual heat release is 91% of the theoretical value. These values yield 10,000 g of kerosine. At a general specific gravity of 0.75, this value corresponds to 13 l. Taking into account the factor of 10 previously discussed, the fuel range estimated is 13 to 130 l (3.4 to 34 gal.) of kerosine.

All of the plutonium storage tanks (i.e., no waste tanks) together, hold 1780 l. To boil all of this material would require 1.5E+9 calories. The fuel requirements for such a fire would be 1.1 to 11 m^3 of wood (40 to 400 ft^3) or 200 to 2000 l of kerosine (50 to 500 gal.). These numbers may seem ludicrous on initial examination. However, for the sake of example, assume the tank farm structure walls are made of Benelex for shielding purposes because the process had significant Americium contamination. The walls would be approximately 7.5 m and 4.2 m wide, and 3.3 m tall. If the walls were only 0.15 m thick (0.5"), the total volume of pressed wood

would be 11.6 m^3, albeit with a smaller density. It is not out of the question that a chimney effect could be created as well, depending on the use of floor and ceiling openings.

The ARF and RF for continuous boiling of liquid are 2 E-3 and 1.0 (subsection 3.2.1.2). For a general fire in the vicinity of the tanks, the initial respirable source terms for DRs of 0.1 and 1.0 are:

$$42{,}000\ g * 0.1 * 2E\text{-}3 * 1.0 = 8.4\ g$$

$$42{,}000\ g * 1.0 * 2E\text{-}3 * 1.0 = 84\ g$$

The range of possible release could be even higher if it is assumed the liquid becomes superheated. The liquid will become superheated if the rate of liquid boiloff exceeds the rate of vapor removal through the vessel vent line. The vessel relief valves lift at ~ 0.07 MPa_g (10 psig), thus limiting the potential level of superheat to ~ 15 ^oC. However, the lifting of the vessel relief valves presents problems of its own. Potential pressure and superheat are limited, but the release is now occurring directly into a process room as opposed to the vessel vent system. The ARF and RF for depressurization of liquid above the boiling point with $\leq$ 50 ^oC superheat are 1E-2 and 0.6 (subsection 3.2.2.3.3). The associated initial respirable source terms for DRs of 0.1 and 1.0 are:

$$42{,}000\ g * 0.1 * 1E\text{-}2 * 0.6 = 25\ g$$

$$42{,}000\ g * 1.0 * 1E\text{-}2 * 0.6 = 250\ g$$

7.3.5.3 Liquid Sampling and Tank Farm Example Assessment

The spill examples are not particularly different from the previous spill assessments, and in general, they support the obvious conclusion that it is desirable to avoid spilling confined carcinogenic material. Even the large quantities of material involved do not generate releases above the relatively small range estimated for previous operations. The large room fire, on the other hand, can be considered genuinely insightful with regard to design and operation. Up to this point in the examples, bounding initial releases have been on the order of gram quantities or less. The large room fire is potentially very unrepresentative of this spectrum, even though the releases cited for this unlikely, but not impossible, event would still not result in catastrophic consequences to the public. In terms of defense in depth, in comparison to events previously examined, this event represents a potentially significant

uncontrolled release of hazardous material. Accordingly, greater attention to detail and, potentially, more analytical documentation are warranted.

It is possible that the large release potential from the room fire may have been fully appreciated before formally estimating the source term. It is at least as likely, however, that it had not really been considered by the majority of personnel. In this example, the fact that no tank venting capacity estimate could be produced would tend to indicate the latter. Given the day-to-day difficulties of operating a facility, it would be understandable to simply think "there's nothing that burns in there and we have a vent and a relief valve, end of story." The amount of fuel estimated to be needed to affect large amounts of solution could also be used to bolster this position. This would be a natural response if the meaning of the source term was blown out of proportion and potential variability of the source term became the sole focus of extensive effort. It is important to remember that, even unmitigated, the source terms calculated do not represent consequence potentials approaching the catastrophic level of consequences from higher hazard types of operations for a given area. The source term estimation process has already provided all the useful information it has to offer.

The source term estimation indicates that the conjunction of a large amount of material in a form uniquely vulnerable to fire release makes fire prevention a particular concern in the tank farm area. A potentially serious vulnerability has been identified. The potential downside of poor combustible control in this area is significantly greater than an area such as the operating side of the residue dissolver, and allocation of limited resources may need to reflect that fact. If this information was understood during the hazard evaluation process, several obvious recommendations would be made. The first recommendation is that an assessment of the particulars of the combustible loading control program be made and any available analyses as they relate to the tank farm area be reviewed (or performed if unavailable) to determine: (1) if the potential mechanisms for and quantities of combustible loading in the tank farm area have been estimated; (2) if this estimation was based on a reasonable assessment of existing conditions as verified by a walkdown and considered nonroutine operational configurations, such as maintenance; and (3) if the significance of minimizing combustibles in the tank farm area is understood in the safety management programs. "Closing the issue," results in a hazard evaluation process of no value if such recommendations are not carried out. The issue can be closed on paper while an existing assessment is based on facility drawings and old documentation only, and the tank farm enclosures are used as overflow storage capacity for unused resin bead drums because "nobody spends much time in there and there's nothing else that burns in there."

The second recommendation is that the venting capacity of the tanks be documented by analysis or testing. It is important to know whether the vessel vent system can adequately relieve the generated vapor for the largest credible fire based on the realistic assessment

previously recommended, or whether the relief valves would be expected to lift. If they did lift, it is important to know that they would be able to control pressure buildup. Until the answers to these questions are known, it is a meaningless activity to manipulate the source term.

A number of other implications arise from the source term estimate as well. If the only fires that are postulated are large fires from undefined causes, the tank farm fire may bound all other such fires. For the largest credible fire (i.e., based on actual or expected combustible loading) in the tank farm area, it is important to know whether the ventilation filtration system would still be expected to function. If it would not, the importance of combustible loading control and fire suppression becomes even greater. Other issues can be imagined as well. If the walls of the tank farm had been constructed out of Benelex, the importance of controls to limit the flammability of that material and to make the fire suppression system available would increase. If an unavoidably large accumulation of flammable material must be present outside the tank farm enclosures, a legitimate consideration may be to install, for new design, or backfit, for existing facilities, fire-rated walls instead of bare stainless steel walls around the tanks. In any case, the importance of the source term and consequence estimation process is not that it provides definitive results, but that it serves as an identifier that triggers other actions having direct, tangible effects on facility safety.

7.3.6 Ion Exchange

Release topics explored in this example are listed in Table 7-9.

Table 7-9. Ion Exchange Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Thermal stress - Venting superheated liquid	- N/A	- N/A	- Thermal stress of contaminated polystyrene	- None

7.3.6.1 Hazard Summary

This activity involves liquid plutonium solutions and plutonium absorbed on solid resins. Plutonium-bearing solutions from the dissolving lines are transferred by a small gear pump to three ion exchange columns in series. Plutonium in the ~ 35% nitric acid feed is absorbed onto the resin in the loading cycle, and eluted as a purified plutonium nitrate solution by a 4% nitric acid elution flow. Effluent solution from the resin loading cycle as well as resin wash and reconditioning cycle effluent (also 35% nitric acid) are directed to annular waste

holding tanks in the tank farm. The 4% nitric acid elution flow (purified plutonium solution) is directed to eluate holding tanks in the tank farm that serve as feed to the evaporation line. Each ion exchange column contains ~ 15,000 g of a strong base, polystyrene anion resin. Process and instrument air are fed into the glovebox by pressurized sources ranging from 0.2 to 0.45 MPa_g (30 to 65 psig). An in-line heat exchanger heats solution by 0.2 MPa_g (35 psig) process steam. Process chilled water is supplied as well.

The presence of a large number of active sites designed to exchange ions accompanied by extensive polymer cross-linking in the overall resin matrix creates an inherent potential for instability in the type of resin used. Under the right circumstances, this instability can be expressed in a wide variety of exothermic reactions. Such reactions involving ion exchange resins have resulted in vessel ruptures, fires, and explosions throughout general industry. Significant potential for worker injury exists in such an incident, and explosions of large ion exchange columns have resulted in worker fatalities.

The extreme range of the phenomena is exemplified by an actual explosion in a Czechoslovakian sugar factory where the introduction of a hot sugar solution to an ion exchange reservoir resulted in a complex reaction involving amino acids and the glucosidic hydroxyl groups of the sugar. The experience of ion exchange column ruptures, fires, and explosions in nuclear materials purification units in the United States, France, South Africa, and Russia have typically involved the by now well-established oxidizing potential of nitric acid with ion exchange resins. A variety of reactions is possible, but once the thermal excursion reaches an autocatalytic state, an overpressurization incident of some type is inevitable.

A large number of laboratory studies and incident investigations have codified a general set of empirical precautionary measures designed to prevent exothermic reactions. A summary of major preventive measures is provided below:

1. Nitric acid should be used only with strong-base resins due to the significantly greater instability of weak-base resins;

2. When not in use, the resin column should be kept wet with very dilute nitric acid or water because dry resin has a lower ignition temperature, and repeated wetting and drying cycles can cause premature resin degradation;

3. The temperature of the resin column should be kept below 60 ^{o}C (140 ^{o}F) as studies have indicated temperatures in excess of this value can ignite resin and sustain a decomposition reaction;

4. The process should not be interrupted for a long period of time when strong nitric acid or radioactive metals are in the column, conditions that could accelerate degradation of resin;

5. The ion exchange column should be vented at all times as ignition temperature decreases in a closed system; and

6. Service life limits for resin in the column should be specified based on chemical and radiological loading to prevent use of degraded resin (e.g., total of 10^8 rad over service life).

7.3.6.2 Release Estimation

The only new potential energetic phenomenon presented by this example is the resin exotherm.

A. Resin Exotherm. The first step in assessing potential releases is to decide upon an appropriate model for the event among at least four theoretical models. The first is a propagating detonation in the condensed (resin-liquid) phase. The second is formation of highly unstable compounds, such as ammonium nitrate, which can decompose explosively. The third model is a deflagration or detonation in gases generated by the exotherm. Nitrogen oxide fumes can be detonated under the right circumstances with hydrogen or other organic vapors, and some tests have shown decomposition reactions can generate ignitable vapors of trimethyl amine and methyl nitrate. The last model is exothermic resin degradation reactions, including the recombination of plutonium with nitrate, generating heat sufficient enough that a pressure buildup leads to the onset of structural failure. Numerous references discuss actual incidents and potential phenomena.

As will be discussed in the assessment of this example, exactness in choosing a model is not as important as it may seem. For now, the history of actual incidents and investigations will be used to select a model. The level of damage that would be associated with a condensed phase detonation, which would have the same affect as a given quantity of TNT, has not been historically observed. The theory does not have general acceptance as an accurate explanation, and was specifically rejected in the investigation of the 1976 Hanford exotherm incident with the following rationale (ERDA Richland report, 1976):

A condensed phase detonation is probably further ruled out by theoretical thermochemical considerations. The energy density of a 7M HNO_3 slurry with polystyrene beads is not great enough to support a steady-state detonation.

There is at least one case on record, at a fertilizer manufacturing plant, where ammonium nitrate is reported to have been present in a cation unit that suffered an explosion upon addition of 54% nitric acid. This condition, while theoretically possible, has not been reported for other incidents such as those that have occurred at nuclear material processing plants. A rapid and violent decomposition reaction involving ammonium nitrate would be expected to produce damage on the order of the condensed phase detonation discussed above. This is not observed. In any case, the knowledge base to determine how much ammonium nitrate might be generated or involved in a reaction is not available.

The gaseous phase explosion is not directly ruled out by technical considerations. A gaseous detonation, although not as violent as a condensed phase detonation, would produce more damage and visible fireballs of a type not characteristic of historical incidents either. Further, meaningful prediction of the explosive yield of such an event is probably not attainable beyond the empirical observation that major detonation effects have not been present in the incidents in nuclear processing plants. The small diameter of the typical nuclear material ion exchange column, due to criticality concerns, lends intuitive support to that observation as well.

In most of the incidents where detailed information is available, the timeframe between initiation of a degradation reaction and vessel failure appears to have been in the range of minutes, and noticeable heat flux was often reported in the vicinity of the equipment shortly prior to failure. The explicit conclusion of the investigation of the Hanford incident of 1976 is that the dominant effect of the degradation reaction was accelerating heat and gas generation resulting in thermally induced pressure failure of the ion exchange vessel. Factory Mutual labels such an event a thermal explosion defined as "the result of an exothermic reaction occurring under conditions of confinement with inadequate cooling ... the reaction rate and heat generation accelerate until the container fails due to overpressure" (Factory Mutual, 1981). In such cases, the initial source of the resin exotherm is highly localized. This localized area may actually dry out resin and heat it above the resin autocatalytic ignition temperature, at which point the column condition can no longer be stabilized. For the purposes of example, a representative model of effects associated with the ion exchange exotherm will be assumed to be a thermal explosion as discussed above.

The appropriateness of this assumption will be considered again after a source term has been developed.

Precedents within DOE for source term estimation have used the model of a thermal explosion. The closest attempts at a specific model have generally assumed some release correlation with the TNT equivalent of the physical explosion. One example considered plutonium disassociating from resin and recombining with nitrate in solution to have the same essential effect as oxidizing reactions. The heat of reaction (~ 210 cal/g) was used to estimate a TNT equivalent by assuming some small percentage of resin reacting, as only a small amount of reaction is believed necessary to generate significant pressure. This method estimated TNT equivalents around 45 g of TNT, which is the same value estimated for the 1976 resin exotherm incident at Hanford (0.1 lb TNT). The Hanford event resulted in ductile failure of the metal resin column, blowout of both the window above the column and the window in front of the column, and numerous glove failures.

A second example simply assumed the 0.1 lb value based on the observation that the column being examined had less structural strength than the Hanford column. This example estimated the mass of plutonium airborne by use of a mass ratio (grams inert material/grams TNT) in correlations from Halverson and Mishima (1986) based on the Steindler-Seefelt data discussed in this handbook. There are two potential problems with such an approach. First, as noted in this handbook, as the mass ratio becomes large, the undefined physical configuration of material to explosive becomes the dominant factor for estimating release as only a small fraction of the material may be subject to the relevant shock effects. Secondly, any method that focuses solely on TNT equivalent scales for release is overlooking two other release mechanisms associated with a thermal explosion. Resin may burn before, during, and after the explosion, and the generation of significant pressure will result in release from flashing spray of superheated liquids at the moment of column failure.

MAR

The source term for the ion exchange exotherm is a function of MAR distribution as damage ratios are variable and there are competing release mechanisms for solid and liquid phases with no constant ratio of plutonium between the phases. At the start of a load cycle, only trace holdup plutonium is in the resin beds. Then solution at a maximum plutonium inlet concentration of ~ 7 g/l passes through the beds. At the completion of a flowsheet loading cycle, almost 6000 g of plutonium are absorbed in the beds with a maximum of 6500 g allowed. The majority of this material is absorbed on the first two of the three ion exchange columns. During the subsequent

washing cycle, the resin remains fully loaded while the clean wash solution picks up only trace quantities of plutonium. Over the course of the elution cycle, the loading process is reversed as the plutonium in the resin beds is reduced to trace holdup once again, and the clean elution solution reaches a plutonium concentration of 50 - 60 g/l as it exits the last ion exchange unit.

The maximum amount of plutonium on the solid resin is 6500 g if an exotherm has been postulated after completion of a loading cycle. The potential variability in this number is solely a function of how much material is processed and where in the loading or eluting cycle the process is. The amount of material that could be in solution is fundamentally dependent on which operation is occurring. For the wash cycle, there will be essentially no plutonium in solution. The amount of plutonium in solution must be estimated for the loading and elution cycles.

Each ion exchange unit is a 5.5-foot tall column with a 6-inch diameter, having a total volume of 30.6 liters and a resin bed volume of 23.9 l. A conservative estimate of the void volume of the resin bed is 50%, in which case a total of 19 liters of liquid may be present in one column. The liquid is distributed as 3.7 l in the top void space of the column, 11.6 l in the resin bed, and 3.7 l in the bottom void space of the column.

Loading cycle feed solution enters the top of ion exchange column 1 at a concentration of 7 g/l, and exits through the bottom of the column. The reduced concentration solution then enters the top of ion exchange column 2, and exits through the bottom as a solution normally containing only waste concentrations of plutonium. Ion exchange column 3 serves only to capture plutonium if something has gone wrong with the absorption process in the first two columns, such as depletion of resin bed capability. The process is reversed for elution. Clean 4% nitric acid flows into the top of ion exchange column 3, and exits through the bottom of the column. It then flows to the top of ion exchange column 2 and then to the top of ion exchange column 1, before exiting from the glovebox as a concentrated plutonium solution.

The maximum mathematical estimate of plutonium in solution in the three columns for either case is:

Loading: 19 l/column * 3 columns * 7 g/l = 399 g
Elution: 19 l/column * 3 columns * 60 g/l = 3420 g

These maximum mathematical estimates are, however, not physically attainable in the system described. During the loading cycle, plutonium concentration is depleted from

a starting value of 7 g/l as it passes through the columns. In the elution cycle, initial plutonium concentration is 0 and builds up to 60 g/l only at the exit of the last column. It is not nonconservative to account for this reality. If the system works like it is supposed to (i.e. plutonium absorbed on columns 1 and 2), it can be assumed that 50% of the plutonium is absorbed or eluted in each column. Accordingly, for loading, the inlet concentration to column 1 is 7 g/l, and the exit concentration to column 2 is 3.5 g/l. The exit concentration to column 3 is 0 g/l, which yields average concentrations for columns 1, 2, and 3 of 5.25 g/l, 1.75 g/l, and 0 g/l respectively. For elution (reverse flow), the inlet and outlet concentrations for column 3 are both 0 g/l, the exit concentration from column 2 is 30 g/l, and the exit concentration from column 1 is 60 g/l. The average concentrations for columns 1, 2, and 3 are therefore 45 g/l, 15 g/l, and 0 g/l respectively. The net effect of this averaging is the same as assuming one column is filled with the maximum available concentration, and the resulting estimates of plutonium in solution are:

Loading: 7 g/l * 19 l = 133 g
Elution: 60 g/l * 19 l = 1140 g

Greater plutonium loadings can be obtained if the system is being operated abnormally. The maximum case occurs if the resin in column 1 is completely depleted and columns 2 and 3 are absorbing all of the plutonium. In that case, the average loading cycle concentrations for columns 1, 2, and 3 are 7 g/l, 5.25 g/l, and 1.75 g/l respectively. The average elution cycle concentrations for columns 1, 2, and 3 are 15 g/l, 45 g/l, and 60 g/l respectively. The net effect of these changes is to double the values previously obtained as the assumption of one column having no material is replaced by one column filled with the maximum concentration. The MAR values are 266 g for loading, and 2280 g for elution.

Damage Ratio

The first key question regarding a damage ratio is how many columns will be affected, and in what way? The initial degradation reaction will start in one of the columns. There are three mechanisms by which other columns can be affected. First, if the temperature of the liquid flowing from the affected column to the next column in line is sufficiently high, it may initiate a resin exotherm in the next column. Secondly, when the affected column ruptures, historical experience and understanding of the phenomena indicate that at least some of the resin from the damaged column will continue to burn on the glovebox floor. How much will burn depends on whether the resin is grouped together to maintain local temperature above the autoignition temperature, and on the effectiveness of available means of fire

suppression. If a large amount of resin burns, the heat generated may be sufficient to initiate resin exotherm reactions in the undamaged columns. This effect is by no means certain and there are historical incidents where an exotherm in one column was followed by a fire with no subsequent exotherm in adjacent columns. In the Hanford exotherm incident, the presence of a significant amount of uncharred resin was reported after the incident. Finally, the other columns may not be undamaged in the initial explosion. They may be shattered by shrapnel from the damaged column, in which case spilled resin may burn, but pressurization of multiple columns is not possible.

Therefore, depending on how many columns are assumed to be affected by a given stress, the first potential factor of the damage ratio is 0.33, 0.67, or 1.0. A second potential factor is, at least for solids, how far the process is into a loading or elution cycle. For example, if only 2000 g are loaded at the time of an exotherm reaction, the DR is 2000/6500 = 0.3. If both potential factors are used, they must be defined together so that "double-counting" does not occur.

In the examples in this handbook, DRs are typically bounded by assuming a value of 1.0 for the sake of simplicity. The above discussion indicates how conservative such a bound can be. It is important not to lose sight of the fact that the phenomena being examined are generally unlikely to highly unlikely. By the time a maximum MAR has been assumed, the DR has been maximized as 1.0, the bounding ARFs and Rfs of this document have been applied, no leakpath is accounted for, and 95% or greater meteorology has been used for dispersion, the answer obtained is extreme. Objectivity must be retained in the evaluation process so that a rote conception does not distract available resources from areas where greater real gains in safety can be made. As previously cautioned in this handbook, answers obtained are only as good as the decisions they lead to.

Source Term

The combustion of polystyrene resin has a combined ARF x RF of 1E-2 (subsection 5.2.1.4.3). The combined ARF x RF for depressurization of superheated liquid is either 6E-3 or 7E-2 depending on whether the degree of superheat is less than or greater than 50 ^{o}C (subsection 3.2.2.3.3), although higher values can be estimated if the degree of superheat is in excess of 100 ^{o}C. Assuming the properties of water as expressed in steam tables, a superheat of 50 ^{o}C corresponds to 0.76 MPa (110 psia) and a superheat of 100 ^{o}C corresponds to 3.1 MPa (450 psia). Therefore, whether the largest release mechanism is burning resin or flashing spray depends on the failure pressure of the ion exchange column.

7.0 Application Examples; Liquid Storage and Ion Exchange Examples

For the purposes of example, five separate source terms will be calculated. The first will be a bounding initial source term for the example facility. The second calculation will provide perspective on the potential conservatism of that answer. To demonstrate use of all ARF x RF values and correlations for the flashing spray release, the remaining calculations will assume that the ion exchange columns are made of steel instead of Pyrex. The third and fourth calculations will be for normal and abnormal operations using the largest bounding flashing spray value provided in this handbook. The final calculation will use the flashing spray release empirical correlation provided in subsection 3.2.2.3.3.

The Pyrex ion exchange vessels in the example facility will fail at pressures below 0.69 MPa (100 psia). Therefore, the maximum flashing spray ARF x RF is 6E-3, which is less than the burning resin value of 1E-2. Therefore, the largest source term will occur if the exotherm happened with all plutonium absorbed on resin. The bounding source term would occur if the loading cycle was complete and the exotherm initiated during the resin wash cycle. The exotherm in one column leads to a rupture and subsequent resin fire on the glovebox floor. If the fire is not suppressed and the majority of the resin burns, the heat generated leads to subsequent failure of the other resin columns. If the DR is assumed to be 1.0, a very conservative assumption, the source term is:

$$6500\ g * 1.0 * 1E\text{-}2 * 1.0 = 65\ g.$$

The potential conservatism of the assumption that all resin burns is obvious, as the source term would decrease in direct proportion to whatever amount of resin less than 100% is assumed to burn. A different configuration would also reduce the source term. For example, assume the exotherm occurs during the loading cycle after 1000 g is loaded onto resin. It will be assumed that the columns are being operated abnormally (i.e., no loading on column 1) so that a total of 266 g of plutonium is available in solution. The exotherm occurs in column 1, leading to eventual exotherms in the remaining two columns as before. However, to maximize release, it could also be assumed that the glovebox pump remained on. If a new oxide dissolution feed tank had just been lined up to the pump before the incident, another 1322 g of plutonium solution can be pumped into the glovebox before the tank runs dry. This material will spill and boil from the heat of the fire, with a combined ARF x RF of 2E-3 (subsection 3.2.1.3) (Note: This assumption would require failure of the sump level-pump interlock). The source term for this situation is:

$$(6500\ g * 0.15 * 1E\text{-}2 * 1.0) + (6500\ g * 0.04 * 1E\text{-}2 * 0.6) + (6500\ g * 0.2 * 2E\text{-}3 * 1.0) = 13.9\ g.$$

Even using the same very conservative assumptions about accident progression, and adding an additional conservatism in the form of the boiling liquid release, the initial source term can go down by a factor of five by simply changing the time in the overall process at which the incident occurs.

The remaining source term calculations examine more extreme conditions by assuming the Pyrex ion exchange columns are constructed out of steel, thus allowing significantly higher pressures before vessel failure. If the degree of superheat is greater than 50 ^{o}C but less than 100 ^{o}C, the associated ARF x RF of 7E-2 is seven times greater than the ARF x RF for burning resin. The worst case for flashing spray release is an exotherm shortly following initiation of the elution cycle, when columns are fully loaded with plutonium in solution and significant plutonium remains on resin. For the normal case (i.e., columns 1 and 2 loading), 1140 g of plutonium is in solution in all of the columns with 5360 g absorbed on the resin. For the abnormal case (columns 2 and 3 loading) 2280 g of plutonium is in solution in the columns, with 4220 g still absorbed on the resin. The maximum initial source terms are:

Normal: (6500 g * 0.18 * 0.1 * 0.7) + (6500 g * 0.82 * 1E-2 * 1.0) = 135 g
Abnormal: (6500 g * 0.35 * 0.1 * 0.7) + (6500 g * 0.65 * 1E-2 * 1.0) = 201.5 g

Even these may not be worst-case values for steel vessels. System pressures as high as 10.3 MPa (1500 psia) have been recorded at the onset of vessel or flange failure in steel systems. The fact that the example system, if made of steel, could support such pressures is apparent from simple calculations. Such a column would essentially act like a straight pipe under internal pressure. A correlation to estimate the minimum thickness of pipewall for a given design pressure is (Perry and Chilton, 1973):

$$T = PD/[2*(SE + PY)] + C \qquad (7\text{-}7)$$

where:

- T = minimum required thickness (in) = typical vessel thickness of 0.3 in.
- P = design pressure (psi)
- D = outside diameter (in) = average of 6.6 in for 6 in inner diameter and 0.3 in thickness
- S = maximum allowable material stress (psi) = average of 15,000 for stainless steel at 400 ^{o}F.
- E = weld joint efficiency factor = assume 1.0
- Y = variable temperature coefficient to account for redistribution circumferential stress = 0.4
- C = Allowance for corrosion = assume 10% of thickness = 0.03 in

Rearranging the equation to solve for pressure gives:

$$P = [2*(T-C)*SE]/[2*(C-T)*Y + D] \quad (7\text{-}8)$$

which, upon substitution of the above values, yields a design pressure of ~ 9 MPa (1300 psi). Internal defects and flaws could cause the vessel to fail locally at substantially smaller pressure, as was suspected in the Hanford incident, and flange or gasket failure could occur at lower pressures as well. However, because no relief valve or rupture disc is installed on the system, a failure pressure of 9 MPa will be assumed.

The correlation for airborne release fraction for superheats in excess of 100 °C requires determining the fraction of liquid that flashes. Subsection 3.2.2.3.3 provides a basic thermodynamic relationship for making this determination for superheated liquid at temperature T_1 and pressure P_1 being depressurized to temperature T_2 and pressure P_2:

$$mass_v/mass_{sh} = (H_{sh} - H_l)/(H_v - H_l) \quad (7\text{-}9,\ 3\text{-}11)$$

where

- $mass_v$ = mass of saturated vapor generated by flashing during depressurization (g),
- $mass_{sh}$ = mass of superheated liquid present (g),
- H_{sh} = enthalpy of superheated liquid at T_1 and P_1 (cal/g),
- H_l = enthalpy of saturated liquid (e.g., liquid at saturation temperature T_2 for pressure P_2) (cal/g), and
- H_v = enthalpy of saturated vapor (e.g., vapor at saturation temperature T_2 for pressure P_2) (cal/g).

The 4% nitric acid solution behaves substantially like water, so the steam tables are used to estimate enthalpies. At 9 MPa and 303 °C (203 °C superheat), the enthalpy of superheated liquid is 325 cal/g. Once depressurized (0.1 MPa and 100 °C), the enthalpies for the flashed vapor and the remaining liquid are 639 cal/g and 100 cal/g, respectively. Inserting these values into the thermodynamic relationship yields a mass vapor to total original mass ratio, or MF_g, of 0.4. The ARF can then be determined from the equation

$$F = 0.33\ (MF_g)^{0.91} = 0.15. \quad (7\text{-}10,\ 3\text{-}7)$$

The RF assessed for this correlation is 0.3, which yields a combined ARF x RF of 5E-2. As this value is less than the combined ARF x RF of 7E-2 assessed for less extreme conditions, the 7E-2 value is used. Therefore, this calculation winds up estimating the same source term as the previous calculation that used the bounding ARF x RF for less than 100 ^{o}C superheat.

7.3.6.3 Ion Exchange Example Assessment

The bounding initial respirable source term estimated for the example configuration (i.e., Pyrex columns) is 65 g of plutonium, although this value could be as low as 10 g or less depending on when in the process the exotherm occurred and what percentage of resin actually burned. However, definitively specifying such parameters is not possible. It is probably best to estimate how much the source term would be reduced by the building leakpath factor before expending effort trying to reduce the source term by arguments such as the ability of fire suppression systems/practices or explosive dispersion to minimize the amount of resin that actually burns. In any case, the appropriate focus is on identifying controls to prevent and/or mitigate the accident. What is being determined by the source term calculation is the level of priority of those controls, not an absolute assurance that a release will be below a given value if preventive and mitigative systems fail.

Now that a spectrum of source terms has been developed, the potential nonconservatism of assuming a thermal explosion as opposed to a chemical detonation can be revisited. If a detonation had been assumed as the release model, burning resin release and flashing spray would not be significant factors due to the speed of the reaction and its energy, which would disperse resin widely. The guidance in this handbook would estimate that a mass of inert material equal to the TNT equivalent mass of the detonation would go airborne. In the case of the ion exchange column, the inert mass includes both resin and liquid in the resin bed. The mass of resin in a column is ~ 15,000 g. The previous resin void volume estimate of 50% is reduced by half to 25% in order to minimize the mass of liquid. The resulting estimate of 5.8 l of liquid would add an additional 5800 g of inert material if the density of water is assumed. Therefore, any one ion exchange column would contain ~ 20,800 g of inert material in the region where the explosion would take place. If total plutonium loading is distributed between two columns, a maximum of 3250 g of plutonium would be in any one column, resulting in an overall grams plutonium to grams inert ratio of 0.16.

The bounding release for the actual example was 65 g of plutonium. If an exotherm in one column leads to an exotherm in the second loaded column, the explosion of each column would have to generate 32.5 g of airborne plutonium. By the ratio just calculated, 203 g of total material would go airborne to generate 32.5 g of airborne plutonium. This would

require a TNT equivalent of 203 g, or almost 0.5 lb of TNT. In the more likely event of an exotherm in only one column, the required TNT equivalent would be 406 g, or almost 1 lb of TNT. Experimental studies performed at Rocky Flats (Tuck, G., et al, 1975) indicated a TNT charge of 227 g would destroy most internal equipment, blow out all windows, and badly bow structural walls of a typical glovebox. Standard overpressure plots indicate 227 g of TNT will produce at least partial demolition effects to a distance of 7 to 8 m. This level of damage is simply not observed in the historical record of nuclear processing, which uses small ion exchange columns. That level of damage is rarely approached even for very large industrial size ion exchange units.

The thermal explosion estimate, which has itself used very conservative assumptions, bounds the phenomena more than adequately. A realistic estimate would, in all likelihood, be significantly less than the 65-g bounding estimate. The thermal explosion model can be accused of being nonphysical, but the same criticism would ultimately be true of any claim to accurately specify TNT yield in a hypothetical detonation. The only physical basis for judgement is the historical record of occurrences for which significant experimental work has already been performed. At this point in the process, arguing over the appropriate theoretical model to apply to an observed phenomena for the source term is an unproductive activity. It inappropriately elevates the significance of source term at the expense of objectively evaluating the process for compliance with known empirical rules for preventing the phenomena. One does not need to know the exact source term to know that the ion exchange column explosion discussed is a highly undesirable event that deserves special attention. By the SAR preparation process outlined in DOE-STD-3009-94, the only question the source term may specifically be answering is whether or not certain preventive and/or mitigative controls will be labelled as safety-class structures, systems, and components by comparison to dose criteria, or safety-significant structures, systems, and components for defense in depth.

The danger of inappropriately focusing on source term arguments is real. At one extreme, mathematical arguments could be used to reduce the source term below a given value attributed special significance. If the non-definitive nature of almost any model is not appreciated, that fact alone could be used to preclude further examination of the system. At the other extreme, the source term could be just as inappropriately inflated. For example, historical experience and the conservatism of the burning resin assumption could be ignored. The heat of combustion of all the resin could be used in an adiabatic temperature calculation turning the process room into a furnace with resulting severe estimates of facility damage and releases from all processes. Such exaggerations can seriously distort evaluation by provoking very time-consuming arguments as, again, the non-definitive nature of the source term is not appreciated. How much actual safety information of real significance can be missed by

either extreme is indicated by a summary of the worst findings allowed for discovery for the mock ion exchange operation in the original hazard analysis training exercise:

1. With an ion exchange feed temperature of 46 °C, the process is already operating too close to the 60 °C empirical limit regardless of any claims about the reliability of the temperature sensors that alarm and secure steam flow to the heat exchanger (Note: The system has single failure if the steam valve sticks open). The process will operate at actual feed temperatures without additional heating, although overall process time will be longer.

2. Based on flowsheet values, the pressure and flowrate of steam through the heat exchanger compared to the ion exchange feedrate indicates that a major heat exchanger leak results in sending a very hot liquid/vapor slug (approaching 100 °C) into the ion exchange columns even if the steam valve shuts as it is supposed to.

3. In the course of discussing the operation of the ion exchange unit, several operators disagree that the valves on the vent lines leading to the vessel vent system are supposed to be left open when the process is operating. They state that they shut the valves when operating and open them when shutdown. Accordingly, another empirical correlation, that ion exchange columns are supposed to be vented at all times, is being defeated.

4. Other operators offer the opinion that the valve confusion exists probably because there are so many valve manipulations required to perform a complete process cycle, as is obvious from looking at the simple flow diagram of the process provided. It was further ascertained that procedures refer to each valve by a name because the valves are not numbered, and no figures are included in the procedures.

5. Upon examining the valve manipulation issue in more detail, the question is raised about the possibility of partially draining ion exchange columns and thus allowing some resin to dry. Investigation verifies that relative heights would allow liquid in the columns to drain into piping if shutdown valve lineups are incorrect.

From just these five summary issues, it is obvious that the example ion exchange system is not currently safe to operate with regard to an ion exchange exotherm, regardless of how high or low a source term may have been estimated or how much of the existing equipment had been designated safety-class or safety-significant. If it were necessary to operate it to prevent holding process solution in tanks indefinitely, examples of potentially appropriate compensatory measures include tagging shut valves on the steam line to the heat exchanger,

developing a clear checklist for valve positions at each step of the process, providing clear direction or controls to ensure the vent valves remain open, and requiring experienced supervision at all times while operating. If it was necessary to operate the system for the long term, consideration would have to be given to issues such as removing the heat exchanger system, possibly removing or caution tagging the isolation valves on the vent lines, formally upgrading procedures, installing elevated piping legs for drain prevention, and examining use of different resins to change loading patterns, optimize new process conditions or possibly even preclude the phenomena from occurring. In short, a serious effort to regain control of the process configuration for principal safety concerns would be necessary. While the example operation is admittedly flawed to an extreme level, the errors included are based on actual industrial experiences. An effective hazard analysis process, not estimation of an initial source term, is the most productive vehicle for coming to grips with such issues.

7.3.7 Calcination and Hydrofluorination

In the interests of brevity, the plutonium peroxide and evaporation operations are not examined because they do not involve phenomenological concerns not already dealt with in previous operations. The calcination and hydrofluorination operations are combined because they operate in series and the main phenomenological concern for each is the same. Release topics explored in this example are listed in Table 7-10.

Table 7-10. Calcination and Hydrofluorination Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- None	- N/A	- Thermal stress	- Thermal stress of contaminated cellulosics and plastics	- None

7.3.7.1 Hazard Summary

The calciner glovebox, part of the "dry" line, receives plastic jugs containing plutonium peroxide precipitate from the "wet" line. These jugs are temporarily stored while awaiting feed to the calciner. Material first goes through a low-temperature drying cycle in a small box drying unit and is then placed in the calciner hopper, where a screw auger feeds it through the calciner and the material is converted to plutonium oxide at elevated temperatures (~ 370 ºC). The output of the calciner enters a critically safe lag storage hopper. Another screw auger then feeds this material to the hydrofluorinator, where a counter-current flow of hydrogen fluoride reacts with the oxide to generate plutonium

fluoride. The hydrofluorinator operates over a temperature range of 400 °C to ~ 600 °C. Both the calciner and the hydrofluorinator are electrically heated.

64% nitric acid is directed to a scrubber in the calcination glovebox to dissolves entrained plutonium oxide from the calciner offgas. This solution is collected in a 100-l slab tank similar to those in the dissolving lines for recycle as ion exchange feed. Hydrogen fluoride is metered on the second floor to a flowrate of ~ 1 scf per minute. 30% potassium hydroxide solution is added to the offgas scrubber in the hydrofluorinator scrubbing glovebox to neutralize the excess hydrogen fluoride. Process air enters both the calcining and hydrofluorinating glovebox lines at 0.24 MPa_g (35 psig). Dry process air provides internal glovebox ventilation flow at 0.21 MPa_g (30 psig) for the calcination glovebox and 0.14 MPa_g (20 psig) for the hydrofluorination glovebox.

The internal sheath of the calciner rotates during operation to ensure even heating. This rotation is driven by a hydraulic unit mounted in the floor of the glovebox. The hydraulic fluid used has a flammability rating of 1, which means "materials that must be preheated before ignition can occur. Materials in this degree require considerable preheating, under all ambient conditions, before ignition and combustion can occur" (NFPA 704). Materials such as this are classified as Class IIIB combustibles, the lowest hazard rating available.

The similar rotation in the hydrofluorinator is driven by an all-electric unit that replaced the hydraulic unit 5 years ago. The hydrofluorination glovebox is also not normally visible from the operating room. Due to the high neutron density associated with the interaction of alpha particles and fluorine, the entire operating side is shielded by a large, Benelex wall with sliding cabinet doors to allow access. Other than windows, the actual surfaces of the glovebox are equipped with such shielding as well. This material is supposed to be painted with an intumescent paint to prevent it from serving as a source of fuel to initiate a large fire.

7.3.7.2 Release Estimation

The major energetic phenomenon of concern for the "dry" line is a large fire.

A. Large Room Fire. Plutonium in a damp or dry powder configuration is the primary material-at-risk in the calcining and hydrofluorination gloveboxes. Rounding upward, approximately 23,000 g of plutonium as plutonium peroxide can be stored in the 10 receipt storage positions available. Between 2 and 4 kg of predominantly plutonium oxide could be in the calcining vessel at any one time, with the 4 kg upper limit based on the size of the vessel. If this amount of material is assumed, it must be subtracted from the amount considered stored. A complete operating cycle up to

the calciner just fills the 10 storage positions, which are then emptied in a continuous run of the calciner. Plutonium nitrate solution could be accumulated with plutonium quantities up to ~ 1 kg by the end of a complete calciner operating cycle.

Approximately 2500 g of plutonium as plutonium oxide and fluoride can physically be in the hydrofluorinator at any one time. No special storage rack is provided in the hydrofluorination glovebox. By procedure, two containers holding ~ 2300 g of plutonium fluoride product (i.e., ~ 1800 g as Pu) apiece are allowed in the glovebox. There is no need to provide long-term storage because the button reduction furnaces operate as batch units, unlike the calciner that is fed continuously from an accumulated source of feed; and it is the hydrofluorinator, not the reduction operation, that is the "chokepoint" in the process flow. Hydrogen fluoride MAR is not specifically examined in this example, because it is a gas (ARF = 1), and the fundamental focus of these examples is radionuclide releases.

A large room fire where the room contains a glovebox with plutonium powders has already been examined for the feed preparation operation in section 7.3.1. In that example, the predominant release effect was heating of plutonium powders, for which the ARF and RF were 6E-3 and 0.01 (subsection 4.4.1.1). The ARF and RF values for heating of plutonium fluoride powders assessed to be bounding in subsection 4.4.1.1 are 1E-3 and 0.001. If the plutonium feed storage for the calciner were considered to experience this release mechanism, the total release would be:

$$(23{,}000\ g * 1.0 * 6E\text{-}3 * 0.01) + (3600\ g * 1.0 * 1E\text{-}3 * 1E\text{-}3) = 1.4\ g$$

Unfortunately, this model may seriously underestimate potential airborne releases because the powder is stored in light plastic jugs (i.e., non-hardened plastic). These jugs will pyrolyze quickly and burn provided the fire is of a minimum duration and heat generation rate. Intermingling of material with melting, bubbling, and burning plastic introduces more release stresses and mechanisms than simple heating of powders.

Polystyrene is reasonably representative of the type of plastics used in molded plastic containers. Subsection 5.2.1.4 assessed bounding ARF and RF values to be 1E-2 and 1.0 for powder contamination on burning polystyrene. The principal question is, therefore, how much of the material in the plastic jugs might experience this release mechanism? If sufficient powder is accumulated, it is conceivable that not all of the material would experience the effects associated with burning plastic. A large amount of powder may remain as a coherent mass as the plastic "disintegrates" around it. Another factor to consider is the geometry of storage. If the plastic jugs are elevated

and the storage racks have a significant amount of open space, the jugs will soften and fail and the powder contents will spill to the glovebox floor with limited, if any, interaction with plastic during combustion.

In this example, however, significant interaction with burning plastic is expected. The quantities of material in relation to container size is not large, and the containers sit upright in recessed wells in a solid storage rack. The powder will, therefore, sit in the well while the entire plastic jug burns. If the fire is presumed to happen with maximum material loading, the initial respirable source term is:

Calciner Feed: 23,000 g * 1.0 * 1E-2 * 1.0 = 230 g
Hydrofluorinator Product: 3600 g * 1.0 * 1E-2 * 1.0 = 36 g

The potential contribution of material inside the calciner or hydrofluorinator can be neglected as the only available release mechanism would be heating of powder in a vessel that is already at temperatures in excess of at least 370 ^{o}C. Likewise, the plutonium-contaminated liquid in the slab tank can be neglected. If it boils (ARF = 2E-3, RF = 1.0), the most likely result, the release to the vessel vent system is:

1000 g * 1.0 * 2E-3 * 1.0 = 2 g

Even if the release were treated as release of a superheated solution, the vessel blow-out plug failure pressure would be at less than the 0.76 MPa (110 psia) associated with 50 ^{o}C superheat. As previously noted in the ion exchange example in section 7.3.6, the ARF x RF for this phenomena is 6E-3, only a factor of three increase over that already postulated. Even these relatively small releases could not typically be concurrent with the calciner release postulated above as the cumulative quantity of 1 kg of plutonium is obtained only at the end of the calcining operating cycle, when the calciner feed storage rack has not been refilled.

7.3.7.3 Calcination and Hydrofluorination Example Assessment

For a large fire in the gloveboxes or glovebox rooms, a bounding initial plutonium source term is 230 g in the calciner and 36 g in the hydrofluorinator, or 266 g combined. This large fire, like the large fire in the maintenance room of the wet processing line (section 7.3.5), is very unrepresentative of the majority of accidental releases that could be postulated for the example facility. Clearly the calcination and hydrofluorination operating areas represent locations where special attention to minimizing the likelihood of even small

fires is warranted. An additional obvious conclusion is that the current practices for storing plutonium in the calcining glovebox need to be reexamined.

The appropriateness of those practices is particularly questionable when the overall operational environment is considered. Although well-insulated, there are vessels operating at very high internal temperatures in both gloveboxes. The calcining glovebox contains a pressurized oil that is combustible even if the likelihood of ignition is very small. Likewise, even though it has been painted with a fire-retardant paint, there is a large amount of combustible shielding surrounding the hydrofluorination glovebox. There is a much greater standing combustible loading in this process room than any of the other example rooms considered. Although not considered highly likely, an objective assessment will conclude there is a greater chance of a large fire in this process room than any others. Given the existence of combustible loading that does not have to be theoretically assumed, this operation may be an appropriate worst-case upon which to estimate temperature conditions that the ventilation filtration system may be exposed to.

As previously noted, potential considerations raised by this example begin with altering storage practices in the calcining glovebox. Potential improvements could include items such as limiting material storage quantities at any one time, reconfiguring the geometry of the storage rack vis-a-vis the containers, or even using different types of containers. It is undesirable to use metal containers for storing the peroxide precipitate, but the use of Pyrex or Pyrex-lined containers may be feasible. Other potential considerations include use of a noncombustible hydraulic fluid in the calciner hydraulic system, ultimate replacement of the hydraulic sheath rotating unit with an all-electric unit like that installed in the hydrofluorinator, and reexamination of man-rem vis-a-vis appropriate shielding for the hydrofluorinator.

A large fire in this room, while unlikely, is probably the most severe, potentially feasible release examined as a result of internal initiators. As such, it would also be a good measuring point for judging how responsive integrated safety management is at the facility. For example, while not based on DOE experience, a worst-case facility walk-through and subsequent hazard analysis might find the following, based on similar large fire concerns at an actual industrial facility:

1. The storage rack in the calcining glovebox is made of a combustible material such as Benelex. Operators state that, in audits and inspections, they have requested it be replaced on multiple occasions.

2. The hydraulic rotating unit on the calciner leaks fairly regularly. Operators state that a standing collection of both clean and oily cleaning wipes is kept in the glovebox

because it often becomes a mess in a hurry otherwise. Numerous oil stains are visible on the calciner insulation. Extensive accumulations of tape are visible at several connection points on the hydraulic lines. A number of outstanding work requests exist on the unit.

3. The paint on combustible hydrofluorinator shielding is thin and obviously worn with numerous scratches and chips revealing the underlying material.

Such findings indicate that the integration of operations, maintenance, assessment, etc., functions is poor. The proper emphasis and support is not being given to safety management programs regardless of how much effort is being expended on "safety analysis." On the other hand, opposite findings would indicate that actual issues are being systematically addressed in an effective manner. Such considerations are real, directly relevant to the overall facility safety basis, and not amenable to effective assessment by source term considerations alone.

7.3.8 Reduction Line

Release topics explored in this example are listed in Table 7-11:

Table 7-11. Reduction Line Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- N/A	- Disturbed molten metal surfaces - Self-sustained oxidation	- Thermal stress - Explosive shock effects - Explosive blast effects	- None	- None

7.3.8.1 Hazard Summary

Plutonium fluoride is received from the hydrofluorination glovebox. A plutonium fluoride charge, an excess of calcium metal, and a pyrotechnic charge are mixed in a magnesium oxide crucible. The crucible is placed in a metal reduction can, which is then sealed by hydraulic pressure against the top of an induction furnace. Heating coils raise the furnace temperature until a thermite-type exothermic reaction is initiated between the plutonium fluoride and calcium. At temperatures between 1500 ^{o}C and 1700 ^{o}C, and pressures between 0.34 and 2.8 MPa_g (50 to 400 psig), the mixture of reaction products is molten and the

denser plutonium flows to the bottom of the crucible in a distinct phase. When the crucible is cooled, the plutonium solidifies as a mass of pure metal.

The cooled crucible is broken, and the product plutonium metal button is cleaned and passed through an airlock to the packaging station. There, the button is first dipped in a 2-l 35% nitric acid bath and then in an identical water bath to remove any slag contamination. A 3/8" drill is used to obtain a small sample of the metal for analysis. The metal is then sealed in a can and bagged out of the glovebox.

The small pyrotechnic charges used in the reduction operation contain magnesium metal. These charges generally ignite at temperatures between 60 ^{o}C and 150 ^{o}C, with an intense, white hot burn lasting less than a second. The purpose of these charges is to catalyze the exothermic calcium-plutonium reactions, so that more of the heat of reaction can be used to elevate the mixture temperature and less heat must be supplied by the furnace. This results in lower ultimate temperatures and pressures in the reduction vessel itself.

The induction furnace heating coils are supplied by a 30-kw motor-generator mounted in the floor of the glovebox. The hydraulic press that locks the reduction vessel in place in the induction furnace operates at pressures in excess of 2.1 MPa_g (300 psig) and uses a hydraulic fluid with a flammable rating of 0, which means that it is not practically combustible. Argon and nitrogen are supplied to the glovebox from 0.1 MPa_g (15 psig) for the purposes of inerting the reduction furnace and the overall glovebox respectively. Process water is also supplied to jacket lines that cool the exterior of the reduction furnace.

The use of calcium can be a problem if excessive contaminant moisture is present in the crucible or the plutonium fluoride. Hydrogen is a product of the reaction. The reaction of calcium and water is also exothermic. Under the right conditions, it can generate sufficient heat to ignite the pyrotechnic charge outside the induction furnace. As noted above, the charge typically would produce a localized high temperature region in the mixture upon ignition, potentially initiating the reduction reaction outside the furnace. Also, a defect in the charge, such as partial decomposition due to excessive storage time, could result in an explosive bang on the order of a small firecracker. It is noted that the charge can also be ignited for other reasons, such as failure to properly cool a magnesium oxide crucible.

Historically, the unit has experienced failure of the reduction vessel pressure seal four times during the reduction process. In the two worst instances before the glovebox was inerted, small fireballs were observed. In addition, relatively minor thermal damage occurred to several glovebox gloves, and contamination was released to the operating rooms. In all cases, however, the bulk of the molten plutonium mass remained in the crucible. These historical cases were largely due to gasket design difficulties and improper gasket installation

prior to initiating hydraulic seal of the reduction can. Only one incident occurred after standardization of gasket specifications and was due to installation of a previously used gasket. No incidents have occurred since the use of pyrotechnic initiators to lower ultimate reduction pressure began in 1977.

7.3.8.2 Release Estimation

The potential energetic phenomena considered in this example are pyrotechnic initiation outside the reduction furnace, failure of the pressure seal on the reduction can, and hydrogen explosion in proximity to plutonium powder.

A. Pyrotechnic Initiation. If the reduction reaction that generates molten products has initiated, it is not physically possible for an unfired pyrotechnic charge to remain in the crucible. Therefore, the pyrotechnic charge itself can affect only plutonium fluoride powder, although the heat generated by the onset of general reduction could affect plutonium metal as well. The exact mechanism for initiating a charge is immaterial and is considered to be one of the two previously discussed. If the charge fires as it normally should, the main effect will be heat. The reduction reaction itself may be initiated, in which case flames would be visible from the combustion of calcium and some plutonium if the glovebox were not inerted. Such an event has been observed in the history of the DOE complex. In one case, there was only a brief flash of flame, while in another the combustion had to be extinguished by pouring magnesium oxide sand over the crucible.

The amount of plutonium present as plutonium fluoride is between 1000 and 2000 g. If the release is considered a product of heating plutonium fluoride powders only, the ARF and RF from section 4.4.1 are, as previously discussed, a maximum of 1E-3 and 0.001. The initial respirable source term is:

(1000 to 2000 g) * 1.0 * 1E-3 * 0.001 = 1E-3 to 2E-3 g

If plutonium metal were assumed to rapidly form in a molten state, the molten metal could begin to burn. That would not happen in this example process due to the use of nitrogen inerting, but it will be evaluated for the sake of example. For burning plutonium metal, the ARF and RF are, as previously discussed, 5E-4 and 0.5 (subsection 4.2.1.1.3). The maximum initial respirable source term (i.e, the highly unlikely event that all material burns) is:

(1000 to 2000 g) * 1.0 * 5E-4 * 0.5 = 0.25 to 0.5 g

Therefore, even if general combustion of all plutonium produced by a completed reduction reaction is assumed, the overall initial source term estimated is still not large.

The final aspect of this problem that must be considered is the potential for a firecracker-type explosion of the initiator. The potential explosion involved is not large. It would not be expected to exceed a TNT-equivalent of a gram or two. The mass of inert material includes calcium as well as plutonium, and the crucible wall would receive a significant portion of the shock impact as well since charges are placed next to the wall. For the sake of simplicity, however, the plutonium fluoride powder is assumed to receive the full impact of the explosion. Subsection 4.4.2.1 assessed a combined ARF x RF value of 0.2 x TNT equivalent to be bounding for powder masses. If the charge is assumed to have a TNT equivalent of 2 g, the initial respirable plutonium source term would be 0.4 g. If it is assumed that the redundant charge on the opposite wall of the crucible also ignites, the value would increase to 0.8 g. The overall release to the glovebox atmosphere estimated is small, even using very conservative assumptions.

B. Pressure Seal Failure. Pressurized venting of a reduction furnace could occur due to improper gasket installation or a fast buildup of reaction gas byproducts from excessive moisture in the reduction charge. The venting will occur at elevated pressures, most likely in excess of 0.34 MPa_g (50 psig). However, the venting of this pressure is a phenomenon whose most severe effects are relegated to the vapor space in the furnace and the immediate surface of the reduction product mixture. The coalescing molten plutonium at the bottom of the plutonium will not be affected as significantly as surface material.

The historical experience cited supports the general observation made above. The incidents reported have not been associated with evidence that a significant portion of the plutonium mass has been driven airborne. The ARF and RF of 1.0 and 0.5 stated in subsection 4.2.1.1.5 for "the violent ejection of molten metal and vapor formation from droplets," or "explosive reaction of the entire metal mass" based on exploding wire experiments, is objectively inappropriate. Subsection 4.2.1.1.4 specifies an ARF and RF of 1E-2 and 1.0 for "airborne release of particulates from disturbed molten metal surfaces (flowing metal, actions resulting in continuous surface renewal), high turbulence at surface, violent airborne reaction." Even these conditions seem somewhat excessive for what is expected in the case of the pressure seal failure.

Another important factor to keep in mind is that the experimental conditions from which the ARF estimate of 1E-2 was obtained involved free fall of plutonium metal

droplets in an air atmosphere, where combustion was a significant factor in driving material airborne. In this example, the furnace contains an argon atmosphere, and the glovebox contains a nitrogen atmosphere. Overall releases under those conditions should be significantly less. If the conservative estimate of phenomena (i.e., "flowing metal or actions involving continuous surface renewal of molten metal") is assumed as the bound, and the glovebox atmosphere is assumed to be air, the initial respirable source term released to the glovebox atmosphere is:

(1000 to 2000 g) * 1.0 * 1E-2 * 1.0 = 10 to 20 g

If the atmosphere involved is inert (i.e., no molten plutonium combustion), the bounding release fraction could reasonably be considered at least an order of magnitude less without the need to provide detailed justifications.

Even if the atmosphere in the glovebox was air, the 10 to 20 g estimate is considered excessive. This handbook does not delve into damage ratio issues at great length, but this is one case where such issues must be confronted based on the limitations of the ARF database. The release estimate is considered excessive, but there is no other ARF value that can be neatly derived to cover this phenomena. The true release is a function of how exposed to turbulence the molten plutonium actually is during vessel venting. Given that the historical record does not include observations of significant splattering of molten material in the crucible, furnace, or adjacent glovebox floor, it is not unconservative to consider the effect largely a surface phenomena. Accordingly, it would not be obviously inappropriate to consider only some fraction of the plutonium, say 10% to 25%, to be in the affected surface layer. Use of the 10% value, for example, as a damage ratio would reduce the initial estimated release range to 1 to 2 g. It is not essential to produce a supposedly "defensible" calculations to incorporate actual experience and understanding of phenomenology into use of the ARF data in the context of providing "information to support general bases for decisionmaking."

C. Hydrogen Explosion. The total amount of calcium metal used in a given reduction charge, including stoichiometric excess, is ~ 740 g, or 18.5 moles. The specific reaction of concern for calcium and water is:

$$Ca^{o} + 2H_2O \rightarrow Ca(OH)_2 + H_2$$

Therefore, to completely react all of the calcium in a given charge would require 37 moles of water (670 ml) and would generate 18.5 moles of hydrogen (37 g). Such a complete reaction in a very short time period is not feasible, but is considered as an

initial baseline for evaluating the process-specific context of the explosive hazard. Seriously contemplating a detonation involving all 37 g of hydrogen would be inappropriate, as discussed below.

At an ambient temperature of 27 °C, one mole of hydrogen gas ideally occupies a volume of 24.5 l, and 18.5 moles would occupy ~ 450 l. Allowing for 25% equipment occupancy, the gas volume of the reduction glovebox is 30,600 l. The dispersed hydrogen concentration is therefore 1.5% by volume. The lower flammability limit of hydrogen is 4.0%. Accordingly, even if the reduction glovebox had an air atmosphere, the possibility of a detonation in the glovebox volume is essentially nonexistent. Even attaining a localized concentration to support a small deflagration in the glovebox volume is dubious.

The same basic examination can be applied to the maintenance room for this operation, where calcium metal is stored. A maximum of 9 kg (226 moles) of calcium, the amount needed for processing the output of hydrogen fluoride from one entire production cycle, is allowed in the storage locker. To completely react all of this calcium would require 552 moles of water (8 l) and would generate 226 moles of hydrogen (5500 l at 27 °C). Discounting space occupied by equipment, the overall maintenance room volume is at least 170,000 l. The dispersed hydrogen concentration is therefore 3.2%. Exceeding 4% would not have been of major significance, but the inability to do so under such an extreme set of assumptions again indicates that a large hydrogen deflagration in the room is very unlikely.

For the sake of example, the reduction glovebox is assumed to have an air atmosphere, as opposed to a nitrogen atmosphere, and a localized hydrogen accumulation above the lower flammable limit is assumed. The demarcation point for ARF and RF values for deflagrations occupying less than 25% of the available free gas volume is 0.17 MPa_g (25 psig). Glovebox confinement would easily fail well below this value, but it is also not even physically attainable from a grossly conservative calculation.

The combustion of 18.5 moles of hydrogen (68,317 cal/mole) yields 1.3E+6 calories. At 27 °C, the remaining 30,100 liters of gas space holds ~ 1230 moles of air, or 35,600 g. The constant volume heat capacity of air is ~ 0.17 cal/g °C. The heat liberated by the combustion of all the hydrogen that can be generated from complete reaction of a calcium charge therefore theoretically raises gas temperature by 215 °C. Using the ideal gas law, this temperature increase would only raise the glovebox pressure to 0.07 MPa_g (10.5 psig).

The ARF and RF values for venting of pressurized powders at confinement failure pressures less than 0.17 MPa_g are 5E-3 and 0.4 (subsection 4.4.2.3.2). It would be equally appropriate to use the ARF and RF values of 5E-3 and 0.3 (subsection 4.4.2.2.2) for powder shielded from a deflagration by a container. If the slightly higher value is used, the initial respirable source term is:

(1000 to 2000 g) * 1.0 * 5E-3 * 0.4 = 2 to 4 g

The last issue to be examined is the potential for hydrogen accumulation in the reduction furnace itself. This is closely related to the pressure seal failure case that has already been examined because the only way for hydrogen to be present in the furnace atmosphere is if the reduction can pressure seal has failed. In the example process, an explosion would not be possible as the furnace is inerted with argon and contained in a glovebox inerted with nitrogen. The use of air in the reduction furnace atmosphere would never be appropriate due to product quality concerns alone. For the sake of example, however, an air atmosphere is assumed present for some reason.

Maximum energy liberation occurs at stoichiometric mixing, which is a 33% by volume hydrogen concentration. If the free volume in the furnace is 85 l (3 ft^3), 28 l of hydrogen would be available to react. The high temperature of the furnace volume makes it conservative to assume that the given volume of hydrogen is ~ 1 mole, which has an available energy in combustion of 68,000 cal. At a standard conversion of 1100 cal/g of TNT, this corresponds to a TNT equivalent of 62 g if the reduction furnace will contain the reaction to significant pressures. This would be expected to blow out gloves in the vicinity and possibly fail windows in the immediate vicinity as well. The source term will not exceed the maximum already estimated for the pressure seal failure case (10 to 20 g) as it is the same phenomena. The molten plutonium is not itself reacting explosively with anything. It is being acted upon by an external force. In any case, a reasonable case can be made that this event is not physically feasible for the system as it is operated (i.e., inerted).

7.3.8.3 Reduction Line Example Assessment

The 10 to 20 gram source term considered excessively conservative for this example could exceed 25 rem at the site boundary for conservative meteorology and no deposition if the release is completely unmitigated. With deposition alone accounted for, the unmitigated release dose is less than 10 rem. The more reasonable spectrum of results examined might exceed 5 rem unmitigated for the no deposition case. The significance of any one of these numbers is limited. This operation does present more potential concerns than operations such as dissolution, but worker safety is still the principle focus for this operation.

7.3.9 Solid Waste Handling

Release topics explored in this example are listed in Table 7-12.

Table 7-12. Solid Waste Handling Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- N/A	- None	- None	- Shock effects to waste - Thermal stress of combustible waste both packaged and uncontained	- None

7.3.9.1 Hazard Summary

The majority of solid waste is bagged out of gloveboxes through typical bagout ports, sealed with tape in a second layer of plastic, and placed inside a hard plastic pail. The lid of this pail is also sealed with a thin strand of tape, which serves as much to indicate tampering as it does to prevent the lid from being removed. These pails are then temporarily stored in the pre-assay storage room, which can accommodate up to 25 pails. After assay, up to 40 pails can be stored in another room. Five pails can also be temporarily stored in the assay room.

The pails are eventually taken to the drum packaging room, where 20 pails can be temporarily stored on a floor array awaiting drumming. This room has a ventilated waste hut where operators in protective clothing remove individual waste bags from pails and place them in a lined 55-gallon drum. Up to 10 filled and sealed drums can be stored in a floor array in the drum packaging room, but normal practice is to take drums to the TRU-Waste storage annex after they are sealed and assayed. Up to 320 drums can be stored on fire-retardant coated pallets in the TRU-Waste storage annex.

Large glovebox waste, such as equipment that cannot be bagged out, is covered and placed inside sealed wooden crates for immediate removal. Low-level solid waste generated outside gloveboxes or in general maintenance is placed either in drums or in fire-retardant wooden crates depending on its size. There are three authorized locations in the hallways of the TRU-waste storage annex to temporarily hold a maximum of two wooden crates per location.

Even noncombustible waste will generally be mingled with combustibles, such as plastic bags or tape, to some degree. The plastic pails can obviously burn along with whatever combustible waste is inside them if an external heat source is provided. Internal heat sources can generate fires as well. Procedure does not allow free-flowing liquid in the solid waste, but liquids can be present in damp paper or other absorbents. Nitric acid-contaminated cellulosic materials can spontaneously ignite, and if any trace moisture was present with other trace waste such as calcium, an exothermic reaction could lead to ignition of combustible material.

Generation of significant combustion inside sealed drums is much less likely due to close packing and a limited oxygen supply. External fires will, however, result in pyrolization of combustible material inside the drum. In normal room fire conditions, this will cause a slow buildup of pressure that eventually fails seal joints resulting in emission of a torch flame around the lid of the drum. If the drum contains highly volatile material and/or if the flames are unusually intense with a high heat generation rate, the pressure buildup may be so rapid that the drum lid pops off and some fraction of the waste is ejected from the drum.

7.3.9.2 Release Estimation

The potential energetic phenomena considered in this example are shock and impact to waste containers and involvement of waste containers in fires.

A. Shock and Impact. For waste contained in taped plastic bags, very little release is expected from any event that does not significantly deform or puncture the bag. For waste in drums, the same expectation exists. In either case, if significant exposure of the waste internals does not occur, whatever material might momentarily be jarred airborne will be contained and filtered within the waste matrix. Estimating release would be intrinsically related to the matrix leakpath factor, which is not large.

The maximum plutonium quantity allowed in a waste pail is 100 g, as initially determined by a frisk reading at the point of generation and confirmed by subsequent assay. The historical average for waste pails in general is 2 g. The maximum amount of plutonium allowed in a waste drum is 200 g, with an historical average of 30 g.

Simply dropping a waste pail to the ground would not be expected to generate a significant release for reasons already discussed. However, if the pail spills its contents and the plastic bag deforms upon impact with a surface, some small release is possible. For impact on contaminated solids that are unpackaged or lightly packaged, the bounding ARF and RF are assessed to be 1E-3 and 1.0 (subsection

5.2.3.2). This bound is considered excessive for this circumstance, but is used because the resulting release is not large. The initial respirable source term for a maximally loaded pail is:

$$100\ g * 1.0 * 1E\text{-}3 * 1.0 = 0.1\ g$$

For an average pail, the release would be only 2E-3 g.

Less limiting values are used for waste drums, where a reasonably robust container shields the majority of the material from direct exposure to air or impact. The same value is assessed to be appropriate for a range of conditions; from a forklift crashing into and puncturing a drum, to a large object falling on and deforming the drum during a building collapse. This value is considered very conservative for the lower end of that range. The bounding ARF and RF are 1E-3 and 0.1 (subsection 5.2.3.2). For a maximally loaded drum, the initial respirable source term is:

$$200\ g * 1.0 * 1E\text{-}3 * 0.1 = 2E\text{-}2\ g$$

The estimated release for an average drum loading is 3E-3 g.

Multiple drum involvement could be postulated as well. If a forklift hit a stack of drums, it might topple, and the third and fourth levels might be assumed to lose their seals as well. In that case, if eight drums lost seals from the fall, and one drum was hit by the forklift, the initial respirable source term is ~ 0.2 g if all drums were maximally loaded, or 3E-2 g if all had average plutonium loadings.

B. Fires. The basic distinction between mixed waste fires is whether the waste is contained in a packaged/bulk form as opposed to loosely strewn. Subsection 5.2.1.1 assessed bounding ARF and RF values to be 5E-4 and 1.0 for packaged waste. Subsection 5.2.1.2 assessed bounding ARF and RF values to be 1E-2 and 1.0 for uncontained combustible waste. The latter value is considered very conservative for many applications, but is likely to bound virtually all applications of interest.

Any fire involving a pail of waste must be considered packaged unless some precursor event has destroyed the pail and dispersed the material within. The waste pail will burn and would not support any significant internal pressurization that would disperse the waste in the event of an external fire. The initial respirable source term for a single, maximally loaded pail is:

$$100\ g * 1.0 * 5E\text{-}4 * 1.0 = 5E\text{-}2\ g$$

The pre-assay storage room can hold 25 pails, so the maximum source term for a large room fire in this location is 1.25 g if all of the pails contained 100 g, or 2.5E-2 g if all of the pails contain the historical average loading of 2 g. If the fire involves the post-assay storage area with a full complement of 40 pails stored, the additional initial release is 2 g at maximum loading and 4E-2 g at average loading. Therefore, for a large fire involving both pail storage rooms, the total bounding initial respirable release is between 7E-2 g and 3.3 g for the range of average to maximum loading conditions.

Estimation of fire releases in drum storage must consider the issue of drum pressurization. Tests of drums under extreme fire conditions have been performed. Sandia National Lab (SNL, 1979) performed experiments where drums with and without liners were placed in square burn pans holding diesel fuel. In one test, close rings were not used on five drums in the flame zone, so these drums were not actually sealed. The lids lifted on all of these drums. In another test, no lifting of lids was observed, most likely due to stacking drums on top of the bottom layer of drums that were exposed to the most intense heat.

In the last large-scale test, 12 drums were sealed and placed in the diesel-soaked (190 l) salt bed without stacking. Three of these drums were unlined, and four had 1/8-inch-diameter vents drilled through the center of their lids. The fire burned for 45 minutes, with the majority of the visible flame zone centering on four drums due to wind conditions. Of these four maximally affected drums, the vented and unlined drum blew its lid 7 minutes into the burn, scattering burning debris over the area. Flaring was observed around the lid of a lined, unvented drum, and a flame torch emanated from the side of the upper lid of a lined, vented drum. The remaining lined, unvented drum experienced a rupture of the bottom seam on one side. In general, polyethylene liners in drums melted and badly pyrolyzed. However, it is possible the insulation provided by the liners prevents as rapid a buildup of temperature and pressure as in the unlined drums.

The dislodgement of drum lids or lack thereof is a function of the rate of pressure rise. A rapid pressure rise is more likely to blow off a drum lid than a slow pressure rise, which will cause localized failure at seal and seam edges followed by emission of a torch of pyrolyzed gases. Recent tests at LLNL (Hasegawa, Staggs, and Doughty, July 1993) used sealed 55-gal metal drums without a vent plug, loaded with combustible materials and "salted" with isopropyl alcohol. These drums were placed in an isopropyl alcohol flow flame and violently ejected their lids in some instances. The test configuration (drums in a pan with isopropyl alcohol) and the fuel are extreme. However, even in those instances where lids blew off, the filmed record

showed bulk waste landing on the ground, where it proceeded to burn. There was no fragmentation of the drum or instantaneous combustion of significant quantities of waste.

Sealed 55-gal metal drums containing a mixture of combustible materials did not lose their lids when placed in a wooden structure burned to the ground with combustibles purposefully stacked around the drums to produce a high fuel loading and associated heat flux (Greenhalgh, Demiter, and Olson, May 1994). These drums exhibited a more typical phenomena of lid seal failures producing torch flames from pyrolysis gases generated in the drums. After the fire consumed the entire building, examination of the drums revealed the majority of the contents to be uncombusted.

If a waste drum experiences seam or seal edge failure with evolution of flame jets, the bounding ARF and RF for packaged waste are very conservative because much of the material inside the drums may not actually burn. If a drum lid blows off at a sufficient pressure to eject waste from the drum, that portion of the waste that ejects is considered loosely strewn and assigned the bounding ARF and RF assessed for uncontained combustible waste. This may be conservative as well because some waste will land in clumps that may more accurately be characterized as packaged rather than loosely strewn. For a fire in the drum packaging room in the plutonium recovery facility, the combustible loading is not of a nature to support extreme heat generation rates over a short period of time. It consists of bulk-combustible solids, not highly volatile hydrocarbon fluids such as alcohol or machinery fuels in large quantities around the drums. Therefore, it would be appropriate to use the bounding release estimates for packaged waste. For a single maximally loaded drum in a fire, the initial respirable source term is:

$$200\ g * 1.0 * 5E\text{-}4 * 1.0 = 0.1\ g$$

The drum packaging room can store 10 drums in a floor array, so the maximum source term for a large room fire in this location is 1 g if all of the drums contained 200 g, or 0.15 g if all of the pails contain the historical average loading of 30 g. Assuming 20 pails were stored in the drum packaging room adds an additional 2E-2 to 1 g to the room fire total. Therefore, for a large fire involving both the pail storage rooms and the drum packaging room, the total bounding initial respirable release is between 0.25 g and 5.3 g for the range of average to maximum loading conditions. The credibility of such a large fire, and thus the utility of the release estimate, however, can legitimately be questioned. Further, even if the large fire is deemed credible, the damage ratio can significantly modify release estimates as well. For example, suppose facility-specific data indicated that 30% of the waste is

noncombustible material, such as metal tools, inorganic solids, HEPA filters, glass, etc. The predominant release mechanism for these materials is heating. The bounding ARF and RF for these phenomena are significantly below those for combustion, so accounting for this factor would reduce the overall source term estimate by 25% or more.

Up to 320 drums containing a maximum of 10,000 g of plutonium can be stored in the TRU waste annex. Therefore, if all 320 positions are used, the average drum loading is 31 g, which is essentially the historical average. Other than the material packaged inside the drums, the combustible loading in the Butler building annex is very limited. In the absence of a large external source of some kind, a fire involving all of the drums is not reasonable to postulate. A localized fire would be expected to be fueled by the same type of bulk combustibles postulated in the drum packaging room fire. As a result, the fire would not provide the rapid heat generation rate typical of conditions where drum lids are blown off. The release that would be estimated is a simple multiple of the average to maximum range (1.5E-2 g to 0.1 g) for a single drum based on how many drums are postulated to be involved.

The only potential source of a large fire involving all of the drums is an extreme event such as a jet airplane crash into the annex. The event itself is very unlikely, and the actual damage that might be inflicted is largely indeterminate and based on factors such as type of plane, angle of descent, point of ground impact, and skid distance prior to impacting the structure, etc. The assumptions can always be made worse by simply assuming the airplane nosedives directly into the center of the facility, thereby making an improbable scenario more improbable. If that temptation is resisted, and a type of airplane is specified, then basic modelling such as the amount and type of debris reaching the facility after the crash, amount of fuel from the fuel tanks, if any, spilling in direct proximity to the facility or its remains, and the fraction of drums directly in the flame zone can be attempted. Drums that are largely intact (i.e., waste still relatively packed in drum, drum shell intact with minor openings or lid jarred) would use the ARF and RF for packaged waste if they were in a fire zone, as would undamaged drums outside the intense flame zone. The contents of drums broken up by missile impact would be considered uncontained waste if they were in the fire zone. Shock-impact releases would be estimated for these drums as well and added to the total source term. It can be assumed for drums in the intense flame zone that a fraction of the drums blow their lids and some fraction of the combustible waste is ejected to burn as uncontained combustible waste.

7.3.9.3 Solid Waste Handling Example Assessment

The source terms estimated for the waste are not large, even when assuming a large number of waste containers will be at maximum procedural loading, which is an extreme assumption. These unmitigated releases are not large for the types of extensive room fires being examined. The use of a graded approach in managing solid waste handling issues at the example facility seems appropriate.

7.3.10 Seismic Release

Release topics explored in this example are listed in Table 7-13.

Table 7-13. Seismic Release Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Free-fall spill - Resuspension	- None	- Free-fall spill - Vibration-shock of bulk powder - Shock-impact of contained powder - Air turbulence from debris impact - Resuspension	- Vibration-shock	- None

7.3.10.1 Hazard Summary

For the plutonium recovery example facility, the one in 1000 years and one in 10,000 years seismic events have ground accelerations of 0.13g and 0.26g respectively. Ostensibly, existing studies estimate building damage for such events. For a 0.13g event, it is expected that the facility remains largely intact, ventilation flow is likely to continue, and damage to process equipment will be minimal, if any. For the 0.26g event, more significant damage is anticipated. Spalling of external walls is possible, non-concrete walls will fail, electrical power will be lost, external piping and wiring connections to gloveboxes will fail, gloveboxes may begin to sag, and one or two of the plutonium process tanks may collapse. For a 0.3g seismic event, all gloveboxes will sag, and between two and four of the plutonium process tanks may collapse, but the building is still expected to remain intact. There is, however, a small possibility that partial collapse of the building centered around the L-shaped hinge on the north face may occur. If this were to happen, processes in the potential collapse zone include the vessel vent system, the north half of the feed preparation

glovebox, the metal dissolving glovebox, the north half of the residue dissolving glovebox, and the liquid sampling glovebox.

7.3.10.2 Release Estimation

The use of release fraction information for estimating seismic releases will be examined in general. Possible release mechanisms will be considered for the 0.13g event, the 0.26g event, and the 0.3g event assuming partial collapse of the facility.

A. 0.13g Event. As described in the hazard summary, this event will have little substantial impact on process equipment or glovebox lines, even though it will do some damage to structure internals. The majority of plutonium driven airborne will come from vibration of uncontained plutonium powder or surface contamination. The latter topic has been largely ignored in examples to this point because of the large amount of process material involvement in the accidents examined.

The only process material that may be expected to release non-trivial amounts of airborne plutonium are loose bulk powders exposed to the glovebox atmosphere during the seismic vibration. In the feed preparation glovebox, a maximum of 2000 g of loose bulk powder can be exposed in both the weighing and batching and furnace stations. Another 1320 g of plutonium as oxide might feasibly be in a charge can in the conveyor or on the glovebox floor. The can holding this oxide could tip over, but the spring-hinged lid will not dislodge. To be very conservative, this phenomena could be accounted for as well. It is more properly shock-vibration than free-fall spill because no real spill in air or falling distance is involved.

Up to 2300 g of plutonium in peroxide cake can be exposed outside containers in the precipitation line. This material is damp, but can be included if more conservatism is desired. The containers in the calcining and hydrofluorination gloveboxes have screw-on lids and are unlikely to be affected. Up to 2300 g of predominantly plutonium oxide in the calciner may be sitting in the first-cycle dryer pan. The same material may be being loaded into the calciner hopper at the precise moment the event occurs and may spill. A maximum of 2000 g of plutonium as plutonium fluoride may be sitting in a reduction crucible awaiting loading into the reduction furnace.

For all of the above to be happening requires considerable synchronicity between operations and the seismic event. Even if it does, however, the resulting source term is not large. The bounding ARF and RF values assessed for vibration-shock impact of bulk powder on an unyielding surface are 1E-3 and 0.1 (subsection 4.4.3.3.1). The bounding ARF and RF for free-fall spill of powder less than 3 m are 2E-3 and

0.6 (subsection 4.4.3.1). If all of the events described above occur, with the 2300 g in the calciner spilled as opposed to being vibrated, the cumulative initial respirable source term within the glovebox atmospheres is:

(9,620 g * 1.0 * 1E-3 * 0.1) + (2300 g * 1.0 * 2E-3 * 0.6) = 3.7 g

The precise amount of plutonium surface contamination cannot be determined. Even survey measurements cannot accurately identify quantities due to shielding uncertainties and the sheer size of the potentially contaminated areas. The type of surface contamination is relevant as well. Unlike loose surface contamination, fixed surface contamination is unlikely to be dislodged to an appreciable degree except by extremely violent events. The inability to dislodge such material by normal cleaning actions testifies to the difficulty of removing such material. Another type of contamination is material holdup in piping and vessels. This material does not receive the direct or undamped type of stress that other contamination would. Further, whatever portion of this material is driven airborne is unlikely to be released from initial confinement if vessel or piping failure does not occur.

This example will concentrate on loose surface contamination outside vessels and process piping, as it is a reasonable assumption that this is the primary material of concern for airborne release due to seismic vibrations. Existing studies (Allen, Arrowsmith, Charlot, and Hooper, 1978) indicate that a surface contamination level of 0.7 g/ft^2 is the level of contamination that would be visible to the naked eye. Historical data from a mixed oxide fuel fabrication facility indicated a surface contamination level of 0.1 g/ft^2 inside gloveboxes was representative of powder handling operations (Mishima, Schwendiman, and Ayer, 1978). There is no firm basis for estimating contamination in less dirty, non-powder operations, but common practice is to consider contamination levels in such operations at least an order of magnitude less than those for powder operations. Typical estimates for glovebox holdup magnitudes are in the range of 0 to 50 g, with interior equipment holdup estimates going to as high as hundreds to thousands of grams for large furnace and incinerator operations.

The internal surface area of the gloveboxes is determined from their measurements. In this construct, that value is multiplied by a factor of 1.25 or 1.5 depending on how much equipment relative to volume there is in a given glovebox. None of the plutonium recovery facility gloveboxes have visible surface contamination over significant areas. Therefore, the available surface area estimate is multiplied by 0.1 g/ft^2 for powder handling operations, and 0.01 g/ft^2 for liquid operations. Resulting surface contamination estimates are listed on the following page.

Glovebox	Surface Area (ft^2)	Multiple	g Pu/ft^2	Contamination (g Pu)
Feed Prep.	470	1.25	0.1	59
3 Dissolvers	840	1.25	0.01	11
Sample	560	1.25	0.01	7
IX and Evap.	880	1.50	0.01	13
H_2O_2 Precip.	440	1.50	0.1	66
Dry Line	1320	1.50	0.1	200
Reduction	1400	1.25	0.1	175
Total				531

The bounding ARF and RF values assessed for vibration-shock impact of loose surface contamination on an unyielding surface are 1E-3 and 1.0 (subsection 5.2.3.2). The resulting cumulative initial respirable source term in glovebox atmospheres due to surface contamination is:

$$531 \text{ g} * 1.0 * 1E\text{-}3 * 1.0 = 0.5 \text{ g}.$$

Similar estimates could be made for surface contamination in a HEPA filter plenum. For a plenum 15 ft long, 10 ft wide, and 10 ft high, the basic enclosure surface area is 600 ft^2. An individual wall for a bank of HEPA filters has a surface area, discounting HEPA filter face area, of 100 ft^2, or 200 ft^2 on both sides. If there are four banks of HEPA filters, the total additional surface area is bounded by an estimate of 800 ft^2. Given that there is not visible surface contamination in a significant part of the plenum, the 1400 ft^2 could be considered powder handling area, yielding an overall surface contamination estimate of 140 g Pu, which would yield an airborne source term of 0.1 g Pu in the plenum, as no release is anticipated from the filters themselves.

The same logic could be applied to plutonium contamination in ventilation ductwork if significant quantities are assumed or known to be present. If the material in the ductwork was a hard, rock-like substance, or a sticky oily residue, no significant release from the residue would be postulated. If, however, it was fine, loose surface contamination, the source term could be estimated in the same manner it was for gloveboxes or the filter plenum.

B. 0.26 g Event. The only significant addition to the release phenomena postulated for the 0.13g event is the failure of one or two plutonium process solution tanks. It is noted, however, that the failure of glovebox connections may yield a higher glovebox

leakpath factor for the airborne surface contamination than would be the case for static glovebox conditions.

The worst-case condition in terms of MAR is for the two plutonium peroxide feed tanks (tanks 13 and 14) to be full (11,650 g Pu each) and to be the tanks that fail. The release phenomena is conservatively estimated as a free-fall spill of 3 m or less. As previously noted, these solutions are not considered concentrated heavy metal solutions by the simple definition assigned in this handbook. The ARF and RF values assessed to be bounding for free-fall spill of aqueous solutions are 2E-4 and 0.5 (subsection 3.2.3.1), making the initial respirable source term from such an event:

$$23{,}300 * 1.0 * 2E\text{-}4 * 0.5 = 2.3 \text{ g}$$

C. 0.3g Event with Partial Collapse. For this event, two to four process solution tanks are assumed to fail. The two tanks with the next largest MAR values are the ion exchange eluate tanks (tanks 7 and 8), each of which can hold 5800 g by flowsheet parameters or 6500 g by procedural limit. It would be very unusual to have these tanks and the peroxide feed tanks full at the same time. The process would be backed up because the destination of the ion exchange eluate tank material as it exits the next process, eluate evaporation, is the peroxide precipitation feed tanks. However, if this condition existed, the maximum initial respirable source term for process tank failures increases to 3.6 g of plutonium.

The partial collapse assumption, however, entails significant additional damage to the facility. This damage will be examined by major process, starting with the vessel vent system. Significant plutonium accumulations are not present in the vessel vent system. The hazard identification estimates were 30 g of plutonium in liquid form, and from 5 to 20 g of plutonium as a solid in each of the three major tanks. It would be conservative to consider the 30 g estimated as aqueous liquid that experiences a free-fall spill with ARF and RF of 2E-4 and 0.5 (subsection 3.2.3.1).

The solid material is unlikely to spill as it is adhering to the raschig rings at the bottom of the tanks, but that same adherence makes it an unlikely candidate for shock-vibration release. The ARF and RF for this circumstance will be small. Given the limited amount of material, the shock-vibration ARF and RF of 1E-3 and 1.0 (subsection 5.2.3.2) will be used solely for the purpose of proceeding with computation. If 20 g of solid plutonium are assumed present in each tank, the maximum initial respirable source term is:

$$(90 * 1.0 * 2E\text{-}4 * 1.0) + (60 * 1.0 * 1E\text{-}3 * 1.0) = 8E\text{-}2 \text{ g}$$

Contamination in piping can also generate airborne releases due to shock-vibration, but these releases are also small for the process as it exists and are ignored in this example.

The north half of the feed preparation glovebox contains the weighing and batching and furnace stations. As previously noted in the examination of the 0.13g event, these stations can hold up to 4000 g of plutonium as loose bulk powder. The impactor station is in the north half of the glovebox as well, and it could be processing as much as 1000 g of plutonium as residue contamination. In addition, 59 g of plutonium as loose surface contamination are also assumed in the glovebox.

The glovebox and residue surface contamination are assumed to release material due to shock-vibration impact with an associated ARF and RF of 1E-3 and 1.0 (subsection 5.2.3.2). This is a very conservative assumption for the residue material, much of which may not be amenable to release, and was not considered as available surface contamination for the 0.13 g event. For example, plutonium metal in crucible residues may be embedded in the pores of the crucible. Air current effects are neglected because the material is routinely subject to forced draft ventilation.

The loose bulk powder experiences three release effects. The first is shock-vibration of bulk-powder, for which the ARF and RF are 1E-3 and 0.1 (subsection 4.4.3.3.1). The second is free-fall spill as the glovebox structure in the free-fall zone collapses. The ARF and RF for this phenomena are 2E-3 and 0.6 (subsection 4.4.3.1). The final release phenomena is air turbulence generated by the impact of debris. The bounding ARF and RF for this phenomena are 1E-2 and 0.2 (subsection 4.4.3.3.2). The initial respirable source term estimated is:

(1059 g * 1.0 * 1E-3 * 1.0) + [4000 g * 1.0 * (1E-4 + 6E-4 + 2E-3)] = 11.9 g

An additional release assumption that does not occur for the facility is specifically examined for the feed preparation glovebox to demonstrate use of another release fraction consideration. Suppose the plutonium charge can storage compartment and one charge can outside the compartment awaiting transfer were located in the north half of the glovebox as well. If each can contained a charge for the oxide dissolver, 8000 g of plutonium as oxide could be in the 6 cans in the holding compartment. An additional 1320 g of plutonium as oxide could be in the can outside the holding compartment.

This material is not loose bulk powder. It is contained, and thus will not experience the full vibration and shock-impact effects that loose material experiences. It will not

actually experience free-fall spill as well. The material in the holding compartment may not experience any direct impact at all. The bounding ARF and RF values assessed for shock-impact on powder confinement are 1E-3 and 0.1 (subsection 4.4.3.3.2) even if the confinement ultimately fails. If this value is applied to all of the material, the additional initial respirable source term is:

$$(9320 \text{ g} * 1.0 * 1E\text{-}3 * 0.1) = 0.9 \text{ g}$$

The dissolution product solution can be considered spilled in the metal line as it is completely within the partial collapse zone. Product spillage is assumed in the residue line as well since the final accountability slab tank is in the collapse zone. If a complete dissolution operation is just completed within each glovebox at the moment of collapse, the total solution MAR is 2200 g. The maximum amount of solution allowed in the sampling line slab tank is 5000 g, although it is typically much less. In this zone of the partial collapse, a total of 7200 g of plutonium in solution can be considered to experience a free-fall spill of less than 3 m. The ARF and RF for free-fall spill of concentrated TRU solutions are 2E-5 and 1.0 (section 3.2.3), and the initial respirable source term is:

$$7{,}200 * 1.0 * 2E\text{-}5 * 1.0 = 0.1 \text{ g}$$

The possibility of liquid being pumped to the sampling line at the moment of the earthquake is discounted as electric power will be lost and the small amount of liquid in lines will not significantly increase the source term estimate. The source term from surface contamination in the liquid lines is also not significant compared to the other releases calculated.

One final release mechanism is resuspension. If a substantial amount of time passes before intervention occurs to minimize further release, the material outside of confinement will be exposed to airflows that can resuspend the material. Between the process tank failures and damage to the residue and metal dissolving lines and the sampling glovebox, 43,200 g of plutonium in liquid can be exposed for resuspension. In addition, 7059 g of plutonium powder and surface contamination are exposed as well if a material holding can (i.e., procedural maximum of 2000 g plutonium vice 1320 g dissolving charge) is assumed to be fully open to the environment. The bounding resuspension rate assessed for liquids covered with debris is 4E-8/hr (subsection 3.2.4.5) and 4E-6/hr for powders covered by debris (subsection 4.4.4.1.2). The respirable source term is:

$$(43{,}200 * 1.0 * 4E\text{-}8/\text{hr}) + (7059 * 1.0 * 4E\text{-}6/\text{hr}) = 3E\text{-}2 \text{ g/hr}$$

If a standard resuspension time of 48 hours is assumed, the total additional release is 1.4 g.

At this point, the initial airborne releases that have been estimated are 3.6 g for the process solution tank failures, 8E-2 g for damage to the vessel vent system, 11.9 g from damage to the feed preparation line (excluding contained powder damage that does not occur for process as is), and 0.1 g from liquid spillage in the metal and residue dissolving lines and the sampling line. The total initial release estimated is therefore 15.7 g. Another 1.4 g could be released over 48 hours if no intervention occurs. Additional material could be considered airborne in gloveboxes outside the collapse zone if some of the conservative assumptions made for the 0.13 g event are made for this event as well.

7.3.10.3 Seismic Release Example Assessment

The seismic release example most clearly brings out an inherent factor in any of the release estimates made in this handbook. The source term is most directly affected by assumptions regarding what material is where. For most examples in this handbook, bounding assumptions have routinely been made. However, the associated likelihood of the release goes down, in some cases by an order of magnitude or more, as a result of these assumptions. For process operations, there is no such thing as "the amount of material present." How much material is where is a highly variable function of time. Variability is inherent even in storage operations, as was demonstrated by the examination of solid waste in section 7.3.9. The chance of having a large number of pails and or drums loaded to the maximum limit is, as a minimum, small. It is very easy to loose sight of the accumulated unlikely conditions that have been assumed in source term calculation when bounding results are desired.

The basic scenarios examined in this example did not add additional events, such as fires and explosions, to the seismic phenomena because, for the level of damage postulated, there was no obvious mechanism for such an event to occur. The 0.13g seismic event did little actual damage to the facility. The 0.26g event did more damage, but again there was no obvious condition created that would result in an additional event. Strong vibration would not be expected to initiate an ion exchange resin exotherm. There are few locations where sufficient flammable material is present to allow electrical equipment damage and residual sparking as power is lost to cause a significant fire. Even for the calciner, there is no compelling reason to believe that a large fire would spontaneously initiate or that the calciner hydraulic system fluid would be sprayed as a fine mist from piping damage to allow fire initiation from sparking. For the final earthquake examined, the 0.3g partial collapse, the collapse zone contained no high combustible loading or major energy sources. These

decisions reflect on the overall issue discussed in the first paragraph. In this case, however, the explicit question is how many poorly definable assumptions need to be piled on top of each other before the source term estimated is practically bounding? These types of questions cannot be answered without carefully defining the purpose of the estimate within an overall context. As previously noted, source term estimates are not capable of providing proof of safety; they can only provide information that supports general bases for decisionmaking.

7.3.11 Production Support Lab

Release topics explored in this example are listed in Table 7-14.

Table 7-14. Production Support Lab Example Topics

Liquid	Metal	Powder	Surface Contamination	Criticality
- Thermal stress - Venting superheated liquid - Free-fall spill - Resuspension	- Self-sustained combustion	- Thermal stress - Accelerated airflow parallel to surface - Free-fall spill - Shock-impact - Air turbulence from falling debris - Resuspension	- HEPA thermal stress - Crush-impact enclosed HEPA filters	- None

7.3.11.1 Hazard Summary

The Production Support Lab conducts small sample analysis on materials generated in the Plutonium Recovery Facility. There are eight lab rooms that conduct liquid sampling, solid sampling, emission spectroscopy, mass spectrometry, thermogravimetric analysis, atomic absorption testing, and raschig ring testing. Plutonium is present in gloveboxes in liquid form in 20-ml glass sample vials and 2-l leftover sample collection jugs for recycle to plutonium recovery processes. Solid plutonium is present in at most 10-g quantities in small glass vials. Small amounts of plutonium in liquid and solid are also handled outside containers as part of the analytical gloveboxes in the form of samples in transfer containers, waste pails, and small counting planchettes containing trace quantities. Waste pails are sent to the Plutonium Recovery Facility for storage.

The lab facility, while not characterized by heavy combustible loading, has more combustible material than the processing lines in the plutonium recovery facility plutonium-handling

areas. The atomic absorption unit also uses a 125-scf cylinder of acetylene that is stored in the lab room, where it can be both a fire and explosive hazard. The ventilation confinement, criticality monitoring, fire protection, and air sampling systems are similar to those in the plutonium recovery facility. The lab facility, however, does not possess the structural strength of the process facility. The southeast quadrant of the ventilation and filter plenum room is estimated to collapse in the 1 in 1000 year 0.13g seismic event. The overall facility is estimated to collapse for seismic events in excess of 0.15g.

7.3.11.2 Release Estimation

The purpose of this example is to demonstrate use of release fraction values for a relatively low-hazard facility. This example will evaluate the facility as a whole instead of examining individual processes or activities. The three specific events examined are a large fire involving all lab rooms, an acetylene explosion, and total collapse of the facility.

A. Large Fire. A large fire involving all of the laboratory rooms and the filter room is very unlikely given the level of combustible loading in the facility, the use of fire walls between lab rooms, and the existence of a building sprinkler system. To provide immediate perspective on the hazard potential of the lab facility, however, such a large fire is arbitrarily assumed to occur.

The release phenomena for liquids in gloveboxes is assumed to be simple boiling. The 2-l plastic jugs of leftover sample solution will not support internal pressures sufficient for superheat phenomena and will fail structurally due to the heat flux of the fire. The spilled liquid will then boil due to that heat flux. The small 20-ml sample vials will also not support any significant internal pressure and their plastic caps will likewise fail due to the heat flux of the fire. The bounding ARF and RF for this phenomena are 2E-3 and 1.0 (subsection 3.2.1.3).

The release phenomena for solids will vary by type. Powders will experience heating only, with a maximum associated ARF and RF of 6E-3 and 0.01 (subsection 4.4.1.1) for oxide. Small metal samples will burn with an ARF and RF of 5E-4 and 0.5 (subsection 4.2.1.1.3) for self-sustained oxidation of metal. Therefore, all solids will be considered to be metal, even though this condition will not be approached in operation. Waste pails will burn with an ARF and RF of 5E-4 and 1.0 for combustion of packaged waste (subsection 5.2.1.1). The ARF and RF for thermal stress of HEPA filters are 1E-4 and 1.0 (section 5.4.1).

7.0 Application Examples; Production Support Lab Example

For the sake of simplicity, the MARs are the maximum listed in Table C-2, "Production Support Lab Facility Hazard Identification." The specific releases by lab room are:

Liquid Sampling Room 1

Liquid	150 g * 1.0 * 2E-3 * 1.0	= 0.3 g
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.3 g

Liquid Sampling Room 2

Liquid	150 g * 1.0 * 2E-3 * 1.0	= 0.3 g
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.3 g

Solid Sampling Room

Liquid	120 g * 1.0 * 2E-3 * 1.0	= 0.24 g
Metal	150 g * 1.0 * 5E-4 * 0.5	= 0.04
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.3 g

Emission Spectroscopy

Metal	250 g * 1.0 * 5E-4 * 0.5	= 0.06 g
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.07 g

Mass Spectrometry

Liquid	120 g * 1.0 * 2E-3 * 1.0	= 0.24 g
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.25 g

Raschig Ring Testing - No significant MAR

Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.01 g

Thermogravimetric Analysis

Metal	150 g * 1.0 * 5E-4 * 0.5	= 0.04
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.05 g

Atomic Absorption

Liquid	20 g * 1.0 * 2E-3 * 1.0	= 0.04 g
Waste	20 g * 1.0 * 5E-4 * 1.0	= 0.01 g
Total		0.05 g

HEPA Plenum Room

Filters	18 g * 1.0 * 1E-4 * 1.0	= 2E-3 g

The total initial respirable source term conservatively estimated for a building-wide fire is 1.3 g of plutonium.

B. Acetylene Explosion. Before proceeding to estimate a source term, the concept of an acetylene explosion in the context of this facility must be understood. The characteristics of explosive decomposition and propensity for self-detonation associated with industrial acetylene processing are not relevant to this discussion, which relates to industrial acetylene cylinders.

By themselves, acetylene cylinders do not represent an explosive threat due to standards for stable loading codified as law in all industrialized nations. Acetylene cylinders are filled with an inert, porous material with interstices too small to allow reaction propagation. The porous mass is soaked with acetone, and the acetylene is then dissolved and stabilized in the acetone. The overall design precludes explosions initiating inside the bottle, and cylinder designs are required to pass an ignition test.

In this test, acetylene in an explosion chamber is connected to the acetylene cylinder. The acetylene in the chamber is detonated, and the detonation propagates into the cylinder. The test passes if there are no cracks or swelling in the cylinder after 24 hours. Industrial literature on the subject of cylinder stability notes:

> For some reason, acetylene has retained an image of danger and accident proneness which it does not deserve. Modern methods of cylinder construction and porous mass manufacture have produced cylinders which resist standard bonfire tests and tests in which a torch is played on the cylinder wall until it bulges with red heat. With regard to shock, a generally believed hazard, cylinders have been dropped from 40 ft and even from aircraft without incident. They have punched their way through concrete floors when accidentally dropped from the top of construction sites.

Accordingly, the accident in question for this event is not a catastrophic detonation of the cylinder contents. That is why the large fire scenario examined did not consider an acetylene explosion a common cause consequence of the fire. Acetylene cylinders are not high-pressure cylinders at ambient conditions. The response to large fires is for a fuse plug in the cylinder to melt, after which acetylene gas ignites and forms a visible jet torch emanating from the fuse plug. The bottle eventually empties the acetylene contents, with the depletion rate a function of the rate at which acetylene comes out of solution with acetone, not a function of pressurized gas flow through a nozzle.

The acetylene explosion of concern is an explosion external to the cylinder. If acetylene leaks from the bottle, it can form a flammable vapor cloud like any other flammable gas. Discounting equipment, the total volume of the atomic absorption unit room is ~ 2000 scf. If the entire content of the acetylene cylinder (125 scf) is uniformly distributed throughout this volume, the resulting acetylene concentration by volume is ~ 6%, which is above the lower flammable limit for acetylene in air (2.5%). Exceeding the lower flammable limit means an acetylene deflagration is physically possible. Typical building ventilation flow and the physical inability of the cylinder to rapidly void its contents, however, make such an explosion an unlikely event. It would not, under any circumstances including the unlikely development of a flammable gas cloud in the glovebox, merit modelling as a confined condition with progression to detonation.

The exact strength of the explosion is of interest only for estimating the degree of physical damage to the overall facility. It is of interest in source term determination

only to the extent it limits the number of plutonium-bearing locations that are affected. The explosion will not affect all of the laboratory sampling rooms, but even if it did, the resulting release is not large. Liquid material inside gloveboxes will not be significantly affected. The limited shielding provided by the gloveboxes will result in the blast wave largely passing over the glovebox by the time penetration occurs. As the duration of the pressure wave can be measured in fractions of a second, the release can be considered bounded by treating all liquid in the blast zone as if it has spilled. The ARF and RF for free-fall spill of liquids are 2 E-4 and 0.5 or 2E-5 and 1.0 (subsection 3.2.3.1) depending on the density of the solution. For this case, the higher value is arbitrarily selected.

The phenomena of concern for powder is accelerated airflow parallel to the surface for powders shielded from direct effects of the blast. However, in actuality, the small hardened glass containers holding the solid samples may be largely unaffected outside the immediate blast zone. Metals will be unaffected. Accordingly, all solids will be conservatively considered to be powders, for which the ARF and RF are 5E-3 and 0.3 (subsection 4.4.2.2.2). Waste pails in the blast zone are assumed to experience surface contamination shock-impact, for which the ARF and RF are 1E-3 and 1.0 (subsection 5.2.3.2). A waste pail in the immediate proximity of the blast may release its contents, which may also burn as localized debris in the aftermath of the explosion, for which the ARF and RF are 1E-2 and 1.0 for combustion of unpackaged cellulosic waste (subsection 5.2.1.2). Only the waste pail in the atomic absorption room is probably eligible for these effects, but a waste pail in each of the eight labrooms will be considered affected for the purposes of obviously bounding potential releases.

For the sake of simplicity, the MARs are the maximum listed in Table C-2, "Production Support Lab Facility Hazard Identification." The blast release effects identified above are applied to all of the material in the lab rooms for a conservative initial screen to estimate a source term of:

Liquid	560 g * 1.0 * 2E-4 * 0.5	= 0.06 g
Powder	550 g * 1.0 * 5E-3 * 0.3	= 0.8 g
Waste	160 g * 1.0 * 1E-3 * 1.0	= 0.16 g
Waste	160 g * 1.0 * 1E-2 * 1.0	= 1.6 g
Total		2.6 g

Even with the use of ridiculously conservative assumptions, the release estimated is very small for a low probability, severe phenomena event.

C. Total Collapse. The release phenomena for liquids in gloveboxes is again considered to be bounded by free-fall spill. The ARF and RF for free-fall spill of 2 E-4 and 0.5 (section 3.2.3) for aqueous solutions is again arbitrarily selected.

The release phenomena for solids will vary by type. Metals will not experience any significant release, and therefore all solids are considered to be powders. Although these powders are contained inside glass vials and, in some cases, metal containers, they are assumed to be loose powder for the sake of simplicity. The loose powder experiences three release effects. The first is shock-vibration of clump powder, for which the ARF and RF are 1E-3 and 0.1 (subsection 4.4.3.3.1). The second is free-fall spill as the glovebox structure in the free-fall zone collapses. The ARF and RF for this phenomena are 2E-3 and 0.6 (subsection 4.4.3.1). The final release phenomena is air turbulence generated by the impact of debris. The bounding ARF and RF for this phenomena are 1E-2 and 0.2 (subsection 4.4.3.3.2). The combined cumulative ARF x RF value for all three phenomena is 2.7E-3. Waste pails are assumed to experience surface contamination shock-impact, for which the ARF and RF are 1E-3 and 1.0 (subsection 5.2.3.2).

The facility HEPA filter media is encased in the filter housing and loaded in a steel plenum housing. Even if the plenum was on a second level of the collapsing facility, this effect cannot be considered unenclosed HEPA filter media. Accordingly, the ARF and RF of 5E-4 and 1.0 (subsection 5.4.4.1) for crush-impact of enclosed HEPA filters is used.

For the sake of simplicity, the MARs are the maximum listed in Table C-2, "Production Support Lab Facility Hazard Identification." The blast release effects identified above are applied to all of the material in the lab rooms for a conservative initial screen to estimate an initial source term of:

Liquid	560 g * 1.0 * 2E-4 * 0.5	= 0.06 g
Powder	550 g * 1.0 * 2.7E-3	= 1.5 g
Waste	140 g * 1.0 * 1E-3 * 1.0	= 0.1 g
Filters	18 g * 1.0 * 5E-4 * 1.0	= 9E-3 g
Total		1.7 g

Following the initial release, resuspension releases are evaluated as well. The bounding resuspension rate assessed for liquids covered with debris is 4E-8/hr (subsection 3.2.4.5) and 4E-6/hr for powders covered by debris (subsection 4.4.4.1.2). No resuspension is assumed from HEPA filter media. If all of the waste

plutonium is treated as powder for the purpose of estimating resuspension, the respirable source term is:

(560 g * 1.0 * 4E-8/hr) + (690 * 1.0 * 4E-6/hr) = 2.8E-3 g/hr

If a standard resuspension time of 48 hours is assumed, the total additional release is 0.1 g. Again, the total release estimated is small for such a severe event.

7.3.11.3 Production Support Lab Example Assessment

The treatment of releases for the lab facility is generally more superficial than that given to the individual process phenomena in the plutonium recovery facility, which is considered appropriate. From a release perspective, the potential issues of concern in the lab facility do not involve a potential complexity or hazard level approaching the process facility. It would be a waste of time and resources to treat source term estimation for the lab facility at a level of analytical detail commensurate with the process facility. There is simply very little information of actual value in formal source term estimates for the lab facility.

7.3.12 Conclusion

A number of comments on the initial draft of this document requested that it be made more "user-friendly," with examples being specifically requested on multiple occasions. This chapter is a direct result of those requests. It is hoped that the examples provided will be of assistance to users of this handbook.

There is, however, an inherent limit on the level of "user-friendliness" that can be incorporated. The values provided cannot become truly generic. A useful cookbook prescription is not feasible, if for no other reason than the simple fact that there is not only one proper approach to using this information. As has been stated, the degree of modelling performed is a function of the ultimate use to which the information will be put. In that sense, the most valuable insight obtained from these examples is the sterility of this release fraction information in a vacuum. Actual experience and understanding of the types of activities being analyzed and the practical limits of phenomena on an industrial scale are as critical to proper application as the experimental data itself. No amount of data or source term algorithms can take the place of such knowledge. The data itself was originally developed within the context of that knowledge, and it is that context that provides information supporting general bases for decisionmaking as opposed to mathematical gamesmanship of little ultimate utility.

8.0 REFERENCES

AIChE. 1992. Guidelines for Hazard Evaluation Procedures, 2nd Edition, Center for Chemical Process Safety of the AIChE, New York, NY, 10017.

Allen, R.P., H.W. Arrowsmith, L.A. Charlot, and J.L. Hooper. April 1978. Electropolishing as a Decontamination Process: Progress and Applications, PNL-SA-6858, Pacific Northwest Laboratory, Richland WA 99352.

Alonso, A., R. Bolado and Hontanon. July 1981. Aerosol Resuspension in the Reactor Cooling System of LWR's Under Severe Accident Conditions, EUR 13789 EN, Directorate-General for Science Research and Development, Joint Research Centre - Ispra Site.

Ammerich, M., A. Briand, J.C. Laborde, Ph. Mulcey and J. Savornin. 1989. "Contamination Releases From HEPA Filters Under High Temperature Conditions", Proceedings of the 20th DOE/NRC Nuclear Air Cleaning Conference (M.W. First, Ed), NUREG/CP-0098 (CONF-880822), Harvard Air Cleaning Laboratory, Boston, MA.

ANSI. 1983. American National Standards for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI/ANS-8.1-1983, American Nuclear Society, La Grange Park, IL.

ANS. 1986. American National Standard - Criticality Accident Alarm System, ANSI/ANS-8.3-1986, American Nuclear Society, La Grange Park, IL.

ANS. 1987a. American National Standard - Guide for Nuclear Criticality Safety in the Storage of Fissile Materials, ANSI/ANS-8.7-1987, American Nuclear Society, La Grange Park, IL.

ANS. 1987b. American National Standard - Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixture Outside Reactors, ANSI/ANS-8.12, American Nuclear Society, La Grange Park, IL.

Anspaugh, L.R., P.L. Phelps, N.C. Kennedy, J.H. Shinn and J.M. Reichman. 1976. "Experimental Studies on the Resuspension of Plutonium from Aged Sources at the Neveda Test Site." in Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants - 1974 (R.J. Englemann and G.A. Sehmel, Ed.), CONF-740921, pp 727-743, National Technical Information Services, Springfield, VA.

8.0 References

Anspaugh, L.R., P.L. Phelps, G. Holladay and K.O. Hamby. 1970. "Distribution and Redistribution of Airborne Particulate from the Schooner Cratering Event", In Proceedings of the 5th Annual Health Physics Society Midyear Topical Symposium: Health Physics Aspects of Nuclear Facility Siting, 2:pp 428-446, Eastern Idaho Health Physics Society, Idaho Falls, ID.

Anspaugh, L.R., J.H. Shinn, P.l. Phelps and C. Kennedy. 1975. "Resuspension and Redistribution of Pu in Soils", Health Physics, 29:pp 571-582.

Arnold, P.M. September 1986. Characterization of Spent HEPA Filters From Rocky Flats Plant, AO 6629 UNIF, Rockwell International - Rocky Flats Plant, Golden, CO.

Ayer, J.E., A.T. Clark, P. Loysen, M.Y. Ballinger, J. Mishima, P.C. Owczarski, W.S. Gregory, B.D. Nichols. May 1988. Nuclear Fuel Cycle Accident Analysis Handbook, U.S.N.R.C. - Office of Nuclear Material Safety and Safeguards, Washington, DC 20555.

Aylor, D.E. 1976. "Resuspension of Particles from Plant Surfaces by Wind," in Proceedings of the Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants -- 1974 (R.J. Englemann and G.A. Sehmel, Ed.), CONF-740921, pp 791-812, U.S. Energy Research and Development Administration, National Technical Information Services, Springfield, VA.

Ballinger, M.Y., J.W. Buck, P.C. Owczarski and J.E. Ayer. January 1988. Methods for Describing Airborne Fractions of Free Fall Spills of Powders and Liquids, NUREG/CR-4997 (PNL-6300), Pacific Northwest Laboratory, Richland, WA 99352.

Ballinger, M.Y. and W.H. Hodgson. December 1986. Aerosols Generated by Spills of Viscous Solutions and Slurries, NUREG/CR-4658 (PNL-5910), Pacific Northwest Laboratory, Richland, WA.

Ballinger, M.Y., S.L. Sutter and W.H. Hodgson. May 1987. New Data for Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air, NUREG/CR-4779 (PNL-6065), Pacific Northwest Laboratory, Richland, WA 99352.

Bennett, B.G. 1976. "Transuranic Element Pathways to Man", in Transuranium Nuclides in the Environment Symposium Proceedings, pp 376-383, U.S. Research and Development Administration and International Atomic Energy Agency.

8.0 References

Borkowski, R., H. Bunz and W. Schoeck. May 1986. Resuspension of Fission Products During Severe Accidents in Light-Water Reactors, KfK 3987 (EUR 10391 EN), Kernforschungszentrum Karlsruhe, Germany.

Braaten, D.A., R.H. Shaw and K.T. Paw U. 1986. "Particle Detachment in Turbulent Boundary Layer, Proceedings of the Second International Aerosol Conference - West Berlin.

Brockman, J.E. February 1985. "Range of Possible Resuspension and Reentrainment of Fission Product During Containment Depressurization", Appendix G of Uncertainty in Radionuclide Release Under Specific LWR Accident Condition, Volume II - TMLbeta' Analyses, SAND84-0410, Sandia National Laboratory, Albuquerque, NM.

Brown, R. and J.L. York. 1962. "Sprays Formed by Flashing Liquid Spray", A.I.Ch.E. Journal, 8:No. 2 pp 149-153.

Brunskill, R.T. 1967. "The Relationship Between Surface and Airborne Contamination", in Proceedings of Surface Contamination Symposium (B.R Fish, Ed.), pp 93-105, Pergamon Press, New York.

Buijis, K., B. Chavane de Dalmassy and H.E. Schmidt. October 1983. "The Characterization of Radioactive Aerosols from Fires", Proceedings of the CSNI Specialist Meeting on Interaction of Fire and Explosion with Ventilation Systems in Nuclear Facilities, LA-9911-C Volumes I & II (CSNI Report No. 83), Los Alamos National Laboratory, Los Alamos, NM.

Carter, R.F. 1970. "The Measurement of Dust Levels in a Workshop Environment.", AWRE 028/70, United Kingdom Atomic Energy Authority, Aldermaston, Berkshire, England.

Carter, R.F. and K. Stewart. 1970. "On the Oxide Fume Formed by the Combustion of Plutonium and Uranium", Inhaled Particles III (Proceedings of an International Symposium, British Occupational Hygiene Society, London, 9/14-23/70), Unwin Brothers Limited - The Gresham Press, Old Working, Surrey, England.

Chambers, D.R., R.A. Markland, M.K. Clary and R.L. Bowman. October 1982. Aerosolization Characteristics of Hard Impact Testing of Depleted Uranium Penetrators, ARBRL-TR-02435, Ballistic Research Laboratory, Aberdeen Proving Grounds, MD 21005.

8.0 References

Chan, M.K., M.Y. Ballinger and P.C. Owczarski. February 1989. User's Manual for FIRIN, NUREG/CR-3037 (PNL-4532), Pacific Northwest Laboratory, Richland, WA 99352.

Chatfield, E.J. 1968. "The Generation of Particulate Aerosols From Plutonium Surfaces", Atmospheric Environment, 2:pp 97-101.

Chatfield, E.J. 1969. "Some Studies on the Aerosol Produced by the Combustion or Vaporization of Plutonium-Alkali Mixtures", Journal of Nuclear Materials, 32:pp 228-246.

Chepil, W.S. 1945. "Dynamics of Soil Erosion II -- Initiation of Soil Movement", Soil Science, 60:pp 397-411.

Clayton, E.D. June 1979. Anomalies of Nuclear Criticality, PNL-SA-4868, Rev 5, Pacific Northwest Laboratory, Richland, WA.

Cleaver, J.W. and B. Yates. 1973. "Mechanism of Detachment of Colloidal Particles From a Flat Surface in a Turbulent Flow", Journal of Colloid and Interface Science, 44:p 464.

Condit, R.H. October 1986. Plutonium Dispersal in Fires - Summary of What's Known, LLNL.

Corino, E.R. and R.S. Brodkey. 1969. "A Visual Investigation of the Wall Region in Turbulent Flow", Journal of Fluid Mechanic, 37:No. 1 ...

Corn, M. 1966. "Adhesion of Particles" in Aerosol Science (C.N. Davies, Ed), Academic Press, New York.

Corn, M. and F. Stein. July 1965. "Re-Entrainment of Particles From a Plane Surface" American Industrial Hygiene Association Journal, 26:pp 325-336.

Dahneke, B. January 1975. 'Kinetic Theory of the Escape of Particles From Surfaces", Journal of Colloid and Interface Sciences, 50:No. 1, pp 89-107.

Dempsey, E. and A.E. Kay. 1958. "Some Investigations on Plutonium Metal", Journal Institute of Metals, 4:p 379.

DOE. December 1992. Hazard Categorization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports, DOE-STD-1027-92, U.S. Department of Energy, Washington, DC.

8.0 References

DOE. September 1993. Example Process Hazard Analysis of a Department of Energy Water Chlorination Process, DOE/EH-0340, U.S. Department of Energy, Washington, DC.

DOE. November 1993. Guidelines for Preparing Criticality Safety Evaluations at Department of Energy Nonreactor Nuclear Facilities, DOE-STD-3007-93, U.S. Department of Energy, Washington, DC.

DOE. July 1994. Preparation Guide for U.S. Department of Energy Nonreactor Nuclear Facility Safety Analysis Reports, DOE-STD-3009-94, U.S. Department of Energy, Washington, DC.

Eidson, A.F. and G.M Kanapilly. February 1983. Plutonium Aerosolization Studies: Phase I Final Report, ITRI - Lovelace Laboratory, Albuquerque, NM.

Eidson, A.F., H.C. Yeh and G.M. Kanapilly. 1988. "Plutonium Aerosol Generation in Reducing and Oxidizing Atmospheres", Journal of Nuclear Materials, 152:pp 41-52.

Einstein, H.A. 1942. "Formulas for the Transportation of Bed-Load", Trans. Am. Soc. Civil Eng., 107:pp 561-597.

Elder, J.C. and M.C. Tinkle. December 1980. Oxidation of Depleted Uranium Penetrators and Aerosol Dispersal at High Temperatures, LA-8610-MS, Los Alamos National Laboratory, Los Alamos, NM 87545.

Ellison, P.G., M.L. Hyder, P.R. Monson, A.B. DeWald, Jr., T.A. Long and M. Epstein. 1994. "Aluminum-Uranium Fuel Melt Behavior During Severe Nuclear Reactor Accidents", Nuclear Safety, 35.

ERDA. October 1976. Report of the Investigation of the Chemical Explosion of an Ion Exchange Column and Resulting Americium Contamination of Personnel in the 242-Z Building, August 30, 1976, Energy Research and Development Administration Richland Operations Office, Richland, WA 99352.

Ettinger, H.J. 1974. "Resuspension of Particulates, in Aerosol Studies, Annual Report of the Biomedical and Environmental Research Program of the LASL Health Division, January through December 1974, LA-5883-PR pp 27-29, Los Alamos National Laboratory, Los Alamos, NM.

Factory Mutual. July 1981. Loss Prevention Data.

8.0 References

Fairchild, C.I. 1982. "Wind Tunnel Measurement of Ideal Particles", Atmospheric Environment, 16: pp. 229-238.

Fish, B.r., R.L. Walker, G.W. Royster and J.l Thompson. 1967. "Redispersion of Settled Particles.", in Proceedings of the Surface Contamination Symposium (B.R. Fish, Ed.), pp 75-81, Pergamon Press, New York.

Fish, B.R., T.D. Calton, C.F. Parrish, R.L. Walker and F.G. Karitoris. October 1966. "Adhesion of Particles", in Aerosol Physics, Health Physics Division Annual Report for Period Ending July 31, 1966, ORNL-4007, Oak Ridge National Laboratory, Oak Ridge, TN.

Freeman, D.E., et al. 1972. Dynamic Dust Measurements for Middle Gusts II, Defense Nuclear Agency Report, Washington, D.C.

Fromentin, A. September 1989. Particle Resuspension from a Multi-Layer Deposit by Turbulent Flow, PSI-Bericht Nr. 38, Paul Scherer Institut, Wuerenlingen, Switzerland.

Fry, M.A. 1991. Ramifications of Acetylene Explosion Inside Plutonium Storage Facilities, SAIC report 91/1188, SAIC, New York, NY 10173.

Fry, T.M. February 1980. Quantitative Assessment of the Magnitude of Aerosol Processes, Associated Nuclear Services Report No. 183, Associated Nuclear Services, Epsom, Surrey, England.

Garger, Yek, V.P. Gavrilov and G.P. Zhukov. 1992. "Estimation of the Secondary Contamination of Resuspension within 30 km Zone of Chernobyl NPP and Its Comparison with Measured Data", Precipitation Scavenging and Atmosphere-Surface Exchange, Volume 3. Summer Volume: Applications and Appraisals (S.E. Schwartz and W.G.N. Slinn, Coordinators), Hemisphere Publishing Corp., Washington, DC.

Garland, J.A. 1982. "Some Recent Studies of the Resuspension of Deposited Materials from Soil and Grasses", Proceeding of the Fourth International Conference on Precipitation Scavenging, Dry Deposition, and Resuspension - St. Monica.

Garland, J.A. and K. Pigford. 1992. "Resuspension Following the Chernobyl Accident", Precipitation Scavenging and Atmosphere-Surface Exchange Volume 3. The Summer Volume: Applications and Appraisals (S.E. Schwartz and W.G.N. Slinn, Coordinators), Hemisphere Publishing Corp., Washington, DC.

8.0 References

Garner, F.H., S.R.M. Ellis and J.A. Lacey. 1954. "The Size Distribution and Entrainment of Drops", Trans. Inst. Chem. Eng., 32:p 222

Garner, F.H., S.R.M. Ellis and D.B. Shearn. 1959. "Entrainment in Evaporators", Trans. Inst. Chem. Eng., 37:p 247.

Gerrard, J.H. 1963. "An Experimental Investigation of the Initial Stages of the Dispersion of Dust by Shock Waves." British J. of Applied Physics, 14: pp 186-192.

Gido, R.G. and A. Koestel. November 1978. "LOCA-Generated Drop Size Prediction - A Thermal Fragmentation Model", Transactions of ANS, 30:pp 371-372.

Gieseke, J.A., V. Kogan, and M. Shaw. September 1993. Spray Release Considerations for the Hanford Tank Farm, Battelle Columbus Laboratory, Columbus, OH.

Gilchrist, R.l. (Project Manager), G.B. Parker and J. Mishima. March 1978. Radiological and Toxicological Assessment of an External Heat (Burn) Test of the 105-mm Cartridge, APFSDS-T, XM-744, PNL-2670, Pacific Northwest Laboratory, Richland, WA 99352.

Gilchrist, R.L. (Project Manager) and P.W. Nickola. January 1979. Characterization of Airborne Depleted Uranium From April 1978 Test Firings of the 105-mm, APFSDS-T, M735E1 Cartridge, PNL-2881, Pacific Northwest Laboratory, Richland, WA 99352.

Glauberman, H., W.R. Bootman and A.J. Breslin. 1967. "Studies on the Significance of Surface Contamination.", in Proceedings of the Surface Contamination Symposium (B.R. Fish, Ed.), pp 169-178, Pergamon Press, New York.

Glissmeyer, G.A. and J. Mishima. November 1979. Characterization of Airborne Uranium From Test Firings of XM774 Ammunition, PNL-2944, Pacific Northwest Laboratory, Richland, WA 99352.

Graf, W.H. and E.R Acaroglu. 1968. "Sediment Transport in Conveyance Systems", Bull. Intern'l Assoc. Sci. Hydrol., 23(2):pp 20-39.

Greenhalgh, W.D., J.A. Demiter and W.W. Olsen. May 1994. Exposure of Drummed Solid Waste to Fire, WHC-SD-WM-DTR-032, Westinghouse Hanford Company, Richland, WA.

8.0 References

Gregory, W.S., R.A. Martin, P.R. Smith and D.E. Fenton. October 1983. "Response of HEPA Filters to Simulated Accident Conditions", Proceedings of the 17th DOE Nuclear Air Cleaning Conference (M.W. First, Ed), CONF-820833, Harvard Air Cleaning Laboratory, Boston, MS.

Hackney, S. 1983. "Fire Testing of HEPA Filters Installed in Filter Housings", Proceedings of the 17th DOE Nuclear Air Cleaning Conference (M.W. First, Ed.), CONF-820833, Harvard Air Cleaning Laboratory, Boston, MS.

Haggard, D.L., W.M. Herrington, C.D. Hooker, J. Mishima, M.A. Parkhurst, R.I. Scherpelz, L.A. Sigalla and D.E. Hadlock. July 1986. Hazard Classification Test of the 120-mm, APFSDS-T, M829 Cartridge: Metal Shipping Container, PNL-5928, Pacific Northwest Laboratory, Richland, WA 99352.

Hall, D. 1988. "Measurements of the Mean Force on a Particle Near a Boundary in Turbulent Flow", Journal of Fluid Mechanics, 187:pp 451-466.

Halverson, M.A., and J. Mishima. 1986. Initial Concepts on Energetics and Mass Releases During Nonnuclear Explosive Events in Fuel Cycle Facilities, NUREG/CR-4593, Pacific Northwest Laboratory, Richland, WA 98352.

Halverson, M.A., M.Y. Ballinger and G.W. Dennis. March 1987. Combustion Aerosols Formed During Burning of Radioactively Contaminated Materials - Experimental Results, NUREG/CR-4736 (PNL-5999), Pacific Northwest Laboratory, Richland, WA.

Hanson, W.C., J.C. Elder, H.J. Ettinger, L.W. Hantel and J.W. Owens. October 1974. Particle Size Distribution of Fragments from Depleted Uranium Penetrators Fired Against Armor Plate Targets, LA-5654, Los Alamos National Laboratory, Los Alamos, NM 87544.

Haschke, J.M. July 1992. Evaluation of Source-Term Data for Plutonium Oxidation, LA-12315-MS, Los Alamos national Laboratory, Los Alamos, NM.

Hasegawa, H.K. July 1993. Fire Testing of 55 Gallon Metal Waste Drums for Dry waste Storage, UCRL-CR-115037, Lawrence Livermore National Laboratory, Livermore, CA.

Healy, J.W. 1974. A Proposed Interim Standard for Plutonium in the Soils, LA-5483-MS, Los Alamos National Laboratory, Los Alamos, NM.

Heger, R., et al. 1982. "Bestimmung der Quellterme bei verfahrenstechnischen Operationen im Weideraufbeitungsprozess", KfK Nachrichten, 14 (3):p 143.

8.0 References

Heger, R. and Th. Waescher. 1983. "Charakteriserung der Aerosolemissionen im Bahaelterabgas einer Wiederaufarbeitungsanlage", Jahrestagung Jerntecknik, Berlin, Tagungsband, p 423.

Hooker, C.D., D.E. Hadlock, J. Mishima and R.L. Gilchrist. November 1983. Hazard Classification Test of the Cartridge, 120-mm, APFSDS-T, XM829, PNL-4459, Pacific Northwest Laboratory, Richland, WA 99352.

Hubbe, M.A. 1984. "Theory of Detachment of Colloidal Particles From Flat Surfaces Exposed to Flow", Colloids and Surfaces, 12: pp 151-178.

Iranzo, E and S. Salvador. 1970. "Inhalation Risk to People Living Near a Contaminated Area." presented at the Second International Congress International Radiation Protection Association, May 3-8, 1970, Brighton, England.

Iwasaki, M., T. Sakurai, N. Ishikawa, and Y. Kobayashi. December 1968. "Oxidation of UO_2 Pellets in Air," Journal of Nuclear Science and Technology, 5:No. 5, pp 652-653.

Jardine, L.J., W.J. Mecham, G.T. Reedy and M.J. Steindler. 1982. Final Report of Experimental Laboratory-Scale Brittle Fracture Study of Glass and Ceramics, ANL-82-39, Argonne National Laboratory, Argonne, IL.

Jette, S.J., J.A. Glissmeyer, J. Mishima, D.J. Sherwood and R.E. Williford. August 1989. Potential Aerosolization (Airborne Suspension) of Depleted Uranium During Test Firings, PNL-7137 Informal Letter Report, Pacific Northwest Laboratory, Richland, WA 99352.

Jette, S.J., J. Mishima and D.E. Hadlock. August 1990. Aerosolization of the M829A1 and XM900E1 Rounds Fired Against Hard Targets, PNL-7452, Pacific Northwest Laboratory, Richland, WA 99352.

John, W., D.N. Fritter and W. Winklmayr. 1991. "Resuspension Induced by Impacting Particles", J. Aerosol Sciences, 22:No. 22, pp 723-736.

Johnson, K.L., K. Kendall and A.D. Roberts. 1971. "Surface Energy and the Contact of Elastic Solids", Proceedings of the Royal Society, London, Series A, 324:pp 301-313.

Jones, I.S. and S.F. Pond. 1967. "Some Experiments to Determine the Resuspension Factor of Plutonium from Various Surfaces", in Proceedings of the Surface Contamination Symposium (B.R. Fish, Ed), pp 83-92, Pergamon Press.

8.0 References

Jordan, D.W. 1954. "The Adhesion of Dust Particles." British J. of Applied Physics, 3, Supplement 3: pp S194-S198.

Jordan, S. and W. Lindner. October 1983. "The Behavior of Burning Kerosine, Aerosol Formation and Consequences", Proceedings of the CSNI Specialist Meeting on Interaction of Fire and Explosion with Ventilation Systems, LA-9911-C Volumes I & II (CSNI Report No. 83), Los Alamos National Laboratory, Los Alamos, NM.

Jordan, S. and W. Lindner. February 1985. "Aerosols Released from Solvent Fire Accidents in Reprocessing Plants", Proceedings of the CSNI Specialists Meeting on Nuclear Aerosols in Reactor Safety (W.O. Schikarski and W. Shock, Ed.), KfK 3800 (CSNI Report No. 95), Kerforschungszentrum Karlsruhe, FRD.

Kalinske, A.A. 1947. "Movement of Sediment as Bed Load in Rivers", Trans. Am. Geophys. Union, 28:615.

Kanapilly, G.M. March 1982. Pu Aerosolization Studies, ITRI - Lovelace Laboratory, Albuquerque, NM.

Kataoka, I. and M. Ishii. April 1983. Mechanistic Modeling for Correlations for Pool Entrainment Phenomenon, NUREG/CR-3304 (ANL-83-37), Argonne National Laboratory, Argonne, IL 60439.

Knief, R.A. 1985. Nuclear Criticality Safety - Theory and Practice, American Nuclear Society, La Grange Park, IL.

Krey, P.W., R. Knuth, T. Tamura and L. Toonkel. 1976. "Interrelationships of Surface Air Concentrations and Soil Characteristics at Rocky Flats.", Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants - 1974 (R.J. Englemann and G.A. Sehmel, Ed.), CONF-740921, pp 744-756, U.S. Energy Research and Development Administration, National Technical Information Services, Springfield, VA.

Langer, G. November 1987. Simulated Seismic Event Release Fraction Data, Progress Report April 1986 - April 1987, RFP-4161, Rockwell International - Rocky Flats Plant, Golden, CO 80402-0464.

Langham, W.H. 1971. "Plutonium Distribution as a Problem in Environmental Sciences, Proceedings of Environmental Plutonium Symposium (E.B. Fowler, R.W. Henderson and M.F. Milligan, Ed.), LA-4756, Los Alamos National Laboratory, Los Alamos, NM.

8.0 References

Larsen, R.I. August 1958. "The Adhesion and Removal of Particles Attached to Air Filter Surfaces", American Industrial Hygiene Association Journal, 19:pp 265-270.

Leach, D.S. 1993. "Predicting Droplet Size Distributions From Pressurized Leaks," Appendix C in WHC-SD-WM-ISB-001, Vol. 1, Westinghouse Hanford Company, Richland, WA.

Long, K.S., J.B. Mason, and B.P. Durst. September 1984. Results of Fort Carson, Colorado, Terrain Dust Obscuration Tests Using Explosives: Soil, Crater, and Dust Cloud Properties, Miscellaneous Paper EL-84-6, U.S. Army Corps of Engineers - Waterway Experiment Station, Vickbury, MI.

Luna, R.E. February 1994. A New Analysis of the VIXEN A Trials, SAND93-2528, Sandia National Laboratory, Albuquerque, NM.

Malet, J.C., G. Duverger de Cuy, R. Gasteiger and J. Janberg. April 1983. "Solvent Pool Fire Testing", Proceedings of the CSNI Specialist Meeting on Interaction of Fire and Explosion with Ventilation Systems in Nuclear Facilities, LA-8911-C Vol 1 & 2 (CSNI Report No. 83), Los Alamos National Laboratory, Los Alamos, NM.

Manowitz, B., R.H. Bretton and R.V. Horrigan. 1955. "Entrainment in Evaporators", Chem. Eng. Progress, 51 (7):p 313.

Martin, R.A., P.K. Tang, A.P. Harper, J.D. Novat and W.S. Gregory. October 1983. Material Transport Analysis for Accident-Induced Flow in Nuclear Facilities, NUREG/CR-3527 (LA-9913-MS), Los Alamos National Laboratory, Los Alamos, NM 87545.

McLaughlin, T.P. 1991. Process Criticality Accident Likelihood, Consequences, and Emergency Planning, LA-UR-91-2325, Los Alamos National Laboratory, Los Alamos, NM 87545.

Mecham, W.J., L.J. Jardine, R.H. Pelto, G.T. Reedy and M.J. Steindler. October 1981. Interim Report of Brittle Fracture Impact Studies: Development of Methodology, ANL-81-27, Argonne National Laboratory, Argonne, IL.

Mendel, J.E., M.D. Hertz, R.F. Hazelton and L.E. Thomas. December 1988. Characterization of the Distribution of Fission Products in the Gap and Grain Boundaries in Spent Fuel, PNL-6771, Pacific Northwest Laboratory, Richland, WA 99352.

8.0 References

Milham, R.C., J.F. Schubert, J.F. Watts, J.R. Boni and J.C. Corey. 1976. "Measured Plutonium Resuspension and Resulting Dose from Agricultural Operations on an Old Field at the Savannah River Plant in the Southeastern United States of America.", Transuranium Nuclides in the Environment, pp 409-421, International Atomic Energy Agency, Vienna, Austria.

Mishima, J. August 1964. A Review of Research on Plutonium Releases During Overheating and Fires, HW-83668, General Electric Company - HAPO, Richland, WA.

Mishima, J. December 1965. Plutonium Release Studies - I. Release From the Ignited Metal, BNWL-205, Pacific Northwest Laboratory, Richland, WA 99352.

Mishima, J. November 1966. Plutonium Release Studies II. Release From Ignited, Bulk Metallic Pieces, BNWL-357, Pacific Northwest Laboratory, Richland, WA 99352.

Mishima, J. June 1973. Interim Report: The Fractional Airborne Release of Dissolved Radioactive Materials During the Burning of 30 Percent Normal Tributyl Phosphate in a Kerosine-Type Diluent, BNWL-B-274, Pacific Northwest Laboratory, Richland, WA 99352.

Mishima, J. and L.C. Schwendiman. October 1970. The Amount and Characteristics of Plutonium Made Airborne Under Thermal Stress, BNWL-SA-3379, Pacific Northwest Laboratory, Richland, WA.

Mishima, J. and L.C. Schwendiman. April 1973. Fractional Airborne Release of Uranium (Representing Plutonium) During the Burning of Contaminated Waste, BNWL-1730, Pacific Northwest Laboratory, Richland, WA.

Mishima, J. and L.C. Schwendiman. August 1973. Some Experimental Measurements of Airborne Uranium (Representing Plutonium) in Transportation Accidents, BNWL-1732, Pacific Northwest Laboratory, Richland, WA 99352.

Mishima, J., L.C. Schwendiman and C.A. Radasch. July 1968. Plutonium Release Studies III. Release From Heated Plutonium Bearing Powders, BNWL-786, Pacific Northwest Laboratory, Richland, WA.

Mishima, J., L.C. Schwendiman and C.A. Radasch. November 1968. Plutonium Release Studies IV. Fractional Release From Heating Plutonium Nitrate Solutions in Flowing Air Stream, BNWL-931, Pacific Northwest Laboratory, Richland, WA 99352.

8.0 References

Mishima, J., L.C. Schwendiman and J.E. Ayer. October 1978. Increment of Analysis - An Estimate of Airborne Release of Plutonium from Babcock and Wilcox Plant as a Result of Severe Wind Hazard and Earthquake, PNL-2812, Pacific Northwest Laboratory, Richland, WA 99352.

Mishima, J., M.A. Parkhurst, R.I. Scherpelz and D.E. Hadlock. March 1985. Potential Behavior of Depleted Uranium Penetrators under Shipping and Bulk Storage Accident Conditions, PNL-5415, Pacific Northwest Laboratory, Richland, WA 99352.

Mitchell, R.N. and B.C. Eutsler. 1967. "A Study of Beryllium Surface Contamination and Resuspension.", in Surface Contamination Symposium Proceedings (B.R. Fish, Ed.), Pergamon Press, New York.

Myers, D.S., W.J. Silver, D.G. Coles, K.C. Lamson, D.R. McIntyre and B. Mendoza. 1976. "Evaluation of the Use of Sludge Containing Plutonium as a Soil Conditioner for Food Crops, in Transuranium Nuclides in the Environment, pp 311-323, International Atomic Energy Agency, Vienna, Austria.

Nelson, C.T. 1973. Resuspension Studies, AI-AEC-13108, Atomics International, ...

Nourbakhish, H.P., M. Khatib-Rahbar and R.E. Davis. March 1988. Fission Product Release Characteristics Into Containment Under Design Basis and Severe Accidents, NUREG/CR-4881 (BNL/NUREG-52059, Brookhaven National Laboratory, Upton, NY 11973.

NFPA 68-1988. Guide for Venting Deflagrations, National Fire Protection Association.

NFPA 704-1990. Identification of the Fire Hazard of Materials, National Fire Protection Association.

NRC. April 1977. Assumptions Used for Evaluating the Potential Radiological Consequences of Accidental Nuclear Criticality in a Fuel Reprocessing Plant, Regulatory Guide 3.33, USNRC - Office of Standards Development, Washington, DC.

NRC. April 1979. Assumption Used for Evaluating the Potential Radiological Consequences of Accidental Nuclear Criticality in Uranium Fuel Fabrication Plant, Regulatory Guide 3.34, USNRC - Office of Standards Development, Washington, DC.

8.0 References

NRC. July 1979. Assumption Used for Evaluating the Potential Radiological Consequences of Accidental Nuclear Criticality in a Plutonium Processing and Fuel Fabrication Plant, Regulatory Guide 3.35, USNRC - Office of Standards Development, Washington, DC.

NUREG-1140 (McGuire, S.). January 1988. A Regulatory Analysis on Emergency Preparedness for Fuel Cycle and Other Radioactive Material Licensees, U.S. Nuclear Regulatory Commission, Washington, DC.

NUREG-1320 (See Ayer, et al). May 1988. Nuclear Fuel Cycle Facility Accident Analysis Handbook, U.S. Nuclear Regulatory Commission, Washington DC.

Nyer, N.E., G.O Bright and R.J. McWhorter. 1965. "Reactor Excursion Behavior" in Proceedings of the Third International Conference on the Peaceful Uses of Atomic Energy, Vol 13. Nuclear Safety, United Nations, New York, NY.

Olsen, A.R., R.L. Hooper, V.O. Uotinen and C.L. Brown. July 1974. Empirical Method for Estimating the Total Number of Fissions From Accidental Criticality in Uranium and Plutonium Systems, BNWL-1840, Pacific Northwest Laboratory, Richland, WA.

Parkhurst, M.A., J. Mishima, D.E. Hadlock and S.J. Jette. April 1990. Hazard Classification and Airborne Dispersion Characteristics of the 25-mm, APFSDS-T XM919 Cartridge, PNL-7232, Pacific Northwest Laboratory, Richland, WA 99352.

Parthenaides, E. and R.E. Passwell. 1970. "Erodibility of Channels with Cohesive Boundaries", J. Hydraulics Div., Proceedings of Am. Soc. Civil Eng., 96(H):pp 755-771.

Paxton, H.C. 1980. "Historical Perspective of Criticality Safety in the United States" in Proceedings of ANS Topical Meeting Nuclear Criticality Safety, SAND80-1675, Sandia National Laboratory, Albuquerque, NM.

Paxton, H.C., J.T. Thomas, D. Callihan and E.B. Johnson. June 1964. Critical Dimensions of Systems Containing U^{235}, Pu^{239}, and U^{233}, TID-7028, Los Alamos National Laboratory & Oak Ridge National Laboratory, Los Alamos, NM/Oak Ridge, TN.

Pinkston, D. (ed.), et al. November 1993. U.S. Department of Energy Defense Programs Safety Survey Report, DOE/DP/70056-H1.

Plinke, M.A.E., D. Leith, D.B. Holstein, and M.G. Boundy. December 1991. "Experimental Examination of Factors That Affect Dust Generation," American Industrial Hygiene Association J., 52: No. 12 pp 521-528.

8.0 References

Punjrath, R.F. and D.R. Heldman. 1973. "Mechanisms of Small Particle Reentrainment from Flat Surfaces", Aerosol Sciences, 3:pp 429-440.

Raabe, O.G., S.V. Teague, N.L. Richardson and L.S. Nelson. November 1978. "Aerodynamic and Dissolution Behavior of Fume Aerosols Produced During the Combustion of Laser-Ignited Plutonium Droplets in Air:, Health Physics, 35:pp 663-674.

Rader, D.J. and D.A. Benson. June 1988. Aerosol Production by High-Velocity Molten-Metal Droplets, SAND88-0678, Sandia National Laboratory, Albuquerque, NM 87185.

Reeks, M.W., J. Reed and D. Hall. May 1985a. On the Long Term Resuspension of Small Particles by a Turbulent Flow, Part I - A Statistical Model, TPRD/B/0638/N85, Central Electric Generating Board, Berkeley Nuclear Laboratory, Gloucesterhire, UK.

Reeks, M.W., J. Reed and D. Hall. May 1985b. On the Long Term Resuspension of Small Particles by a Turbulent Flow, Part II - Determination of the Resuspension Rate Constant for an Elastic Particle on a Surface Under the influence of van der Waals Forces, TPRD/B/0639/N85, Central Electric Generating Board, Berkeley Nuclear Laboratory, Gloucestershire, UK.

Reeks, M.W., J. Reed and D. Hall. June 1985a. On the Long Term Resuspension of Small Particles by a Turbulent Flow, Part III - Resuspension for Rough Surfaces, TPRD/B/0640/N85, Central Electric Generating Board, Berkeley Nuclear laboratory, Gloucestershire, UK.

Reeks, M.W., J. Reed and D. Hall. June 1985b. On the Long Term Resuspension of Particles by a Turbulent Flow, Part IV - CAGR Plant Application, TPRD/B/0641/N85, Central Electric Generating Board, Berkeley Nuclear Laboratory, Gluocestershire, UK.

Reeks, M.W., J. Reed and D. Hall. 1988. "On the Resuspension of Small Particles by a Turbulent Flow", Journal of Physics, D, Applied Physics, 21:pp 574-589.

Restrepo, L. 1991. Building 771, Subteam 6 Report, Chapter 7, Criticality Assessment, EG&G-RF, Golden, CO.

Restrepo, L.F. August 1992. The Annular Core Research Reactor (ACRR) Limiting Event for Workers and Public Consequences, SAND91-0572, Sandia National Laboratory, Albuquerque, NM.

8.0 References

Roblyer, S.P. and P.C. Owczarski. 1992. Determining Wind Spray Release From Effluent Disposal Ponds Using AIRSOURCE Model, paper presented at Hazmacon '92, March 30-April 2, 1992, Long Beach, CA.

Royster, G.W. and B.R. Fish. 1967. "Techniques for Assessing 'Removable' Surface Contamination", Surface Contamination (B.R. Fish, Ed) pp 201-207, Pergamon Press, New York, NY.

Rozen, A.M., S.I. Golub, I.F. Davydov, and G.I. Gostinin. 1970. "Some Laws Governing Drop Carryover." Soviet Physics - Doklady, 14:(7)648.

Sackman, J.F. 1960. "The Atmospheric Oxidation of Plutonium Metal", Proceedings Conference Internationale Metallurgie du Plutonium, Grenoble, France.

SAND. September 1987. Appendix F "Preliminary Preclosure Safety Analysis", Site Characterization Plan Conceptual Design Report (H.R. MacDougall, L.W. Scully and J.R. Tillerson, Compilers), SAND84-2641 Vol 4, Sandia National Laboratory, Albuquerque, NM.

Schmitt, R.C. May 1975. "Carbon Microspheres as Extinguishing Agents for Fissionable Material Fires", Fire Protection, 11:pp 95-98.

Schrock, V.E., S.T. Revanker, R. Mannheimer, and C-H. Wang. August 1986. Small Break Critical Discharge - The Role of Vapor and Liquid Entrainment in the Stratified Two-Phase Region of the Break, LBL-22024, Lawrence Berkeley Laboratory, Berkeley, CA.

Sehmel, G.A. 1973. "Particle Resuspension from an Asphalt Road Caused by Car and Truck Traffic", Atmospheric Environment, 7:pp 291 - 301.

Sehmel, G.A. 1979. Deposition and Resuspension Process, PNL-SA-6746, Pacific Northwest Laboratory, Richland, WA.

Sehmel, G.A. 1980. "Particle Resuspension: A Review", Environment International, 4:pp 107 - 127.

Sehmel, G.A. and F.D. Lloyd. 1975. "Initial Particle Resuspension Rates -- A Field Experiment Using Tracer Particles.", Pacific Northwest Laboratory Annual Report for 1974: Atmospheric Sciences, BNWL-1950-Pt 3, pp 203-207, Pacific Northwest Laboratory, Richland, WA.

8.0 References

Sehmel, G.A. and F.D. Lloyd. 1976. "Resuspension Rates from a Circular Field Source", Annual Report 1975 to the USERDA Division of Biomedical and Environmental Research, Part 3. Atmospheric Sciences, BNWL-2000, Part 3, Pacific Northwest Laboratory, Richland, WA.

Sehmel, G.A. and F.D. Lloyd. 1976. "Particle Resuspension Rates", Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants (R.D. Englemann and G.A. Sehmel, Ed), ERDA Symp. Series #38, Technical Information Center - Energy Research and Development Center (Now U.S. Department of Energy), Washington, D.C.

Sehmel, G.A. and M.M. Orgill. 1973. "Resuspension by Wind at Rocky Flats.", in Pacific Northwest Laboratory Annual Report for 1972; Atmospheric Sciences, BNWL-1751-Pt-1, pp 15-22, Pacific Northwest Laboratory, Richland, WA.

Shor, A.J., H.T. Ward, D. Miller and W.A. Rodger. 1957. "Radioactive Carry-Over from Borax III and Test Systems", Nucl. Sci. Eng., 2:pp 126-142.

Singer, J.M., E.R. Cook and J. Grumer. 1972. Dispersal of Coal and Rock Dust Deposits, BMRI-7642, Bureau of Mines, Washington, D.C.

Singer, J.M., M.E. Harris and J. Grumer. 1976. Dust Dispersal by Explosion-Induced Airflow, Entrainment by Air Blast, BMRI-8130, Bureau of Mines, Washington, D.C.

Skidmore, E.L. and N.P. Woodruff. 1968. Wind Erosion Forces in the United States and Their Use in Predicting Soil Loss, Agricultural Handbook No. 346, U.S. Department of Agriculture, Washington, D.C.

Soffer, L. July 1993. "Revision of Reactor Accident Source Terms and Implications for Nuclear Air Cleaning Requirements", Proceedings of the 22nd DOE/NRC Nuclear Air Cleaning Conference (M.W. First, Ed), NUREG/CP-0130 (CONF-9020823), Harvard Air Cleaning Laboratory, Boston, MS 02115.

Spenser, H.W. 1976. 'Rapping Entrainment Studies", in Symposium on Particulate Control in Energy Processes (D.E. Blake, ED), ...

Steindler, M.J. and W.H. Seefeldt. 1980. "A Method for Estimating the Challenge to an Air Cleaning System", Proceeding of the 16th DOE Air Cleaning Conference (M.W. First, Ed.), Harvard Air Cleaning Laboratory, Boston, MS.

8.0 References

Stewart, K. 1963. "The Particulate Material Formed by the Oxidation of Plutonium", Progress in Nuclear Energy Series IV, Vol 5, pp 535-579.

Stewart, K. 1967. "The Resuspension of Particulate Material from Surfaces.", in Proceedings of the Surface Contamination Symposium (B.R. Fish, Ed.), pp 63-74, Pergamon Press, New York.

Strange, J.N. and A.D. Rooke, Jr. November 1988. Battlefield Dust from Exploding Munitions: Contribution by Cratering from Artillery and Mortar Projectiles, Technical Report EL-88-16, Science and Technology Company, Hampton, VA.

Stratton, W.F. September 1967. A Review of Criticality Accidents, LA-3611, Los Alamos National Laboratory, Los Alamos, NM.

Sutter, S.L. August 1980. Potential Airborne Release from Soil-Working Operations in a Contaminated Area, PNL-3498, Pacific Northwest Laboratory, Richland, WA 99352.

Sutter, S.L. May 1982. Accident Generated Particulate Materials and Their Characteristics -- A Review of Background Information, NUREG/CR-2651 (PNL-4154), Pacific Northwest Laboratory, Richland, WA 99352.

Sutter, S.L. August 1983. Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air, NUREG/CR-3093 (PNL-4566), Pacific Northwest Laboratory, Richland, WA 99352.

Sutter, S.L., J. Mishima and L.C. Schwendiman. June 1974. Fractional Airborne Release of Strontium During the Combustion of 30 Percent Normal Tributyl Phosphate in a Kerosine-Type Diluent, BNWL-B-358, Pacific Northwest Laboratory, Richland, WA.

Sutter, S.L., J.W. Johnston and J. Mishima. December 1981. Aerosols Generated by Free Fall Spills of Powders and Solutions in Static Air, NUREG/CR-2139 (PNL-3786), Pacific Northwest Laboratory, Richland, WA 99352.

Sutter, S.L., J.W. Johnston, J.A. Glissmeyer and G.F. Athey. January 1985. BTD Building Uranium Mass Balance Study, PNL-5344, Pacific Northwest Laboratory, Richland, WA 99352.

8.0 References

Teleyarkhan, R.P. 1992. "Analysis and Modeling of Fission Product Release from Various Uranium-Aluminum Plate-Type Reactor Fuel," Nuclear Safety, 33:No. 1.

Thomas, J.T., Ed. June 1978. Nuclear Safety Guide, TID-7016, Revision 2, NUREG/CR-0095, Oak Ridge National Laboratory, Oak Ridge, TN.

Tuck, G. August 1974. "Simplified Methods of Estimating the Results of Accidental Solution Excursions", Nuclear Technology, 23:pp 177-188.

Tuck, G., W.D. Dennison, N.J. Mossoni, P.A. Patton, R.E. Boyd, W.R. Sheets, and D.E. Payne. May 1975. Rocky-Flats Glove-Box Explosion Experiment, NSIR-1, Dow Chemical, Rocky Flats Division, Golden, CO 80401.

Waber, J.T., W.M. Olson, J.A.C. Davidson, P.M. French, E.N. Hodkin and M.J.F. Notley. 1960. Atmospheric Corrosion Tests of Several Delta-phase Alloys of Plutonium, Journal of Nuclear Materials, 3:p 201.

Walker, E. 1978. A Summary of Parameters Affecting the Release and Transport of Radioactive Material From an Unplanned Incident, Bechtel Inc., San Francisco, CA.

WHC. 1993 (DRAFT). Preliminary Safety Analysis Report for Hanford Waste Vitrification Plant, WHC-SD-HWV-PSAR-001, Westinghouse Hanford Company, Richland, WA 99352.

Wilsey, E.F. and E.W. Bloore. May 1989. M774 Cartridges Impacting Armor-Bustle Targets: Depleted Uranium Airborne and Fallout Material, BRL-MR-3760, Ballistics Research Laboratory, Aberdeen Proving Grounds, MD 21005-5066.

Woodcock, E.R. 1966. Potential Magnitude of Criticality Accidents, AHSB (RP) R-14, United Kingdom Atomic Energy Authority.

Woodley, R.E. June 1986. The Release of Fission Product From Irradiated SRP Spent Fuels at Elevated Temperature - Data Report on the First Stage of the SRP Source Term Study, HEDL-7598, Westinghouse - Hanford Engineering Development Laboratory, Richland, WA 99352.

Woodley, R.E. March 1987. Release of Fission Products From Irradiated SRP Fuels at Elevated Temperature - Data Report on the Second Stage of the SRP Source Term Study, HEDL-7651, Westinghouse Hanford Engineering Development Laboratory, Richland, WA 99352.

8.0 References

Wright, A.L. and W.L. Pattison. 1984. 'Results from Simulated Upper-Plenum Aerosol Transport and Aerosol Resuspension Experiment", Proceeding of the CSNI Specialist Meeting on Nuclear Aerosols in Reactor Safety, Karlsruhe.

Zimon, A.D. 1969. Adhesion of Dust and Powder (Translation by M. Corn), Plenum Press, New York.

CONCLUDING MATERIAL

Review Activities:

DOE	Field Offices	National Laboratories	Area Offices
DP	AL	ANL	Argonne
EH	CH	BNL	Brookhaven
EM	ID	INEL	Golden
ER	NV	LANL	Kirtland
NE	OR	LLNL	Los Alamos
NS	RFO	ORNL	Rocky Flats
	RL	PNL	
	SR	SNL	
	SF		
	SRO		

Preparing Activity:

DOE DP-31

Project Number:

SAFT-0013