AU-30-RPT-02

U.S. Department of Energy
Pressurized Spray Release Technical Report

April 2019

U.S. Department of Energy
Office of Environment, Health, Safety and Security
Office of Nuclear Safety
Washington, D.C. 20585

FOREWORD

This technical report summarizes several years of work by the Department of Energy (DOE) and its contractors to improve methods for analyzing pressurized spray leak phenomena in the context of accident analysis. The purposes of the report are twofold: (1) summarize DOE's analytical and experimental work on spray leak phenomena from 2009 to the present, and (2) provide technical findings and recommendations based on that work.

The report is divided into six main sections. Section 1 provides a brief introduction to the issues and events leading to issuance of this report. Section 2 reviews the technical basis for DOE-HDBK-3010-94, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities,* as well as the technical concerns from outside entities. Section 3 describes the methods and results of a series of experiments, conducted at the Pacific Northwest National Laboratory (PNNL) in 2012-2013, designed to provide new information on pressurized spray leaks. Section 4 describes parametric approaches to enhancing the model used in the 1994 handbook. Section 5 summarizes this report and provides recommended actions. Appendix A defines acronyms used frequently in the report. Appendix B expands on Section 1 by providing a detailed description of events beginning in 2009 related to the pressurized spray leak issue. Appendix C reprints the memorandum from Steven Krahn to Andrew Lawrence. Appendix D reprints the Mishima-Foppe technical paper. Both Appendices C and D are important reference documents for understanding of this report. Appendix E provides proposed text to be incorporated into a revision of DOE-HDBK-3010-94, Section 3.2.2.3.1, "Venting Below the Liquid Level."

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TABLE OF CONTENTS

1. INTRODUCTION

The analysis of accidents in DOE nuclear facilities sometimes requires close study of initiating events in which pressurized liquids are sprayed from ruptured piping. One effect of such spray releases may be toxic or radioactive exposure to operating personnel in or near the affected facility and the public should the material be vented to the atmosphere. Calculating the extent of this exposure requires an analysis of the volume of the release (i.e., MARxDR), the particle size distribution, and the respirability of the liquid droplets suspended in air (i.e., ARFxRF) and its subsequent evaporation. Arriving at a conservative approach for analyzing the hazard caused by such leaks has proven difficult, especially in cases where the released material is not a pure fluid but rather a non-Newtonian slurry or viscous fluid. Modeling these events is problematic both in terms of source term characterization, complicated by leak plugging, and of establishing the respirability of the released material over time.

Because pressurized spray leaks of radioactive and toxic materials have long been determined to be a safety hazard at DOE's nuclear facilities, guidance was needed to assist analysts preparing safety analyses of such facilities. Pressurized spray leaks were therefore addressed in DOE-HDBK-3010-94, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities*, Section 3 of which discusses the state of analytical and experimental knowledge as of the early 1990s. The Handbook's scope with respect to pressurized leaks is confined to analyzing pure Newtonian liquids over a limited application domain. The Handbook has been reaffirmed as recently as 2013, but has never been substantially revised or enhanced.

A pressurized spray leak producing respirable droplets is one of the unmitigated accident sequences required to be analyzed in the Preliminary Documented Safety Analysis (PDSA) for DOE's Waste Treatment and Immobilization Plant (WTP). Up to 2009, the PDSA for WTP had relied on an approach based on DOE-HDBK-3010-94 and on subsequent peer-reviewed studies appearing in published technical literature. Such studies, however, typically have assumed a pressurized release of pure liquids that behave as a Newtonian fluid and therefore do not extend the reach of the 1994 Handbook. Pressurized releases at WTP involve a wide range of slurries and viscous materials that do not exhibit Newtonian fluid behavior, may be subject to plugging, and are released through orifices outside the range of the 1994 Handbook.

In 2009, discussions between DOE and its WTP contractor with the technical staff of the Defense Nuclear Facilities Safety Board (DNFSB) focused attention on potential non-conservatism in the project's PDSA with respect to analysis of pressurized spray leak accidents. In February of 2010, DOE embarked on an effort to achieve a better understanding of the types of pressurized spray leaks that might occur at WTP and at other DOE facilities processing similar materials. This effort was to include conducting experiments to acquire important data. Such an enhanced understanding could then be used to establish a more comprehensive, yet sufficiently conservative analytical approach. This report summarizes the results of the effort.

2. TECHNICAL BASIS FOR DOE-HDBK-3010-94 AND TECHNICAL CONCERNS

2.1 DOE-HDBK-3010-94 Technical Basis

DOE-HDBK-3010-94 provides acceptable methodologies for various postulated events, and bounding values of ARF and RF to be used to determine the source term from various phenomenologies. The Handbook's ARFxRF recommendation for pressurized spray releases was established more than 20 years ago and was based on limited data from commercial hollow cone spray nozzles with three different orifice diameters (0.063 inch, 0.086 inch, 0.128 inch) at three different upstream pressures (50 psig, 100 psig, 200 psig). From these nine combinations of orifice diameters and upstream pressures, the Handbook selected the ARF and RF from the coarsest distribution generated by these commercial hollow cone spray nozzles; which is from the 0.128-inch diameter orifice at an upstream pressure of 200 psig, and a droplet diameter of concern (10-µm) as the bounding ARF and RF.[1]

Section 3.2.2.3.1 of the Handbook, "Venting Below the Liquid Level," provides the following bounding ARFxRF recommendation:

> *"For the purpose 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... It is not anticipated that drops formed from breaches, cracks, leaks would generate finer drop size distribution 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 MPag) upstream pressure.*
>
> *Other recent investigations ... using an analytical model suggest that, under some conditions, the fractions of drops in the finer size fractions (i.e., 10 µm and less) are greater for finer 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*

[1] Figure 3-4 of DOE-HDBK-3010-94 appears to be based on an incorrect interpretation of the source data for aerosol generation by hollow-cone nozzles. The original data (Perry's *Chemical Engineers' Handbook*) are tabulated in terms of the mid-point (average) diameters for droplet-size bins. A footnote to the original table states the upper-limit diameters for the bins. The y axis of Figure 3-4 is based on the mid-point diameters, not the upper-limit diameters that should have been used to show the cumulative distribution.

understanding the phenomenon, cannot presently be used predict the size distribution of sprays."

The Handbook also provides guidance on the range of release phenomenology for breaches venting pressurized liquids. It states that (1) the amount and Aerodynamic Equivalent Diameter (AED) of the pressurized spray release are a function of size and characteristics of the breach, the upstream pressure and the physical characteristics of the liquid, and (2) the size distribution of liquid drops becomes finer with decreasing orifice diameter and increasing upstream pressure.

The Handbook references 1993 analytical modeling studies suggesting that the fraction of drops in finer size fractions ($\leq$ 10 µm) are greater for fine orifices and possibly slot-type breaches, at high upstream pressures, and that evaporation and agglomeration affect the droplet size distribution. The studies taken as a whole suggest that uncertainties remain and analytical models cannot presently be used to predict size distribution of pressurized spray leaks outside of a limited application domain.

To address these many uncertainties, the Handbook recommends a bounding ARFxRF of 1E-4 for respirable droplets. Respirable droplets are defined as having AEDs $\leq$ 10 µm, based on the coarsest distribution generated by commercial spray nozzles shown in Figure 3-4 of the Handbook. While the 1E-4 value corresponds to a discharge from a nozzle of 0.128 inch diameter and an upstream pressure of 200 psig, no specific recommendations regarding leak size and pressure are made in the Handbook.

2.2 DNFSB Critique

In an April 5, 2011 letter and reporting requirement, (Reference 2-1), the DNFSB formally stated its concerns. The letter asserted that three major sources of uncertainty in the WTP methodology could affect accident dose consequences:

- Orifice configuration: A single rectangular slit orifice to represent all potential leak site geometries may be non-conservative. Different leak site geometries may result in higher dose consequences.
- Droplet size distribution (DSD): The Rosin-Rammler probability distribution to represent the DSD of a pressurized spray leak may not be the most appropriate distribution to use. Other equally viable droplet size distributions (e.g., log normal) may result in higher dose consequences.
- Agglomerate structure: The methodology applied to the process slurry assumes that dried agglomerates transform from multiple discrete particles into a solid monolith with no void space. The more probable situation is the formation of agglomerates rather than a monolithic particle upon drying. This may also result in higher dose consequences.

On the basis of its review, the DNFSB stated that

> *"… the WTP project needs to provide a well-formulated analysis that accounts for the uncertainties and reduces the potential for non-conservative results associated with the analysis of spray leaks. The Board believes it may be possible to reduce uncertainties to more manageable levels by completing additional research and development."*

Ultimately, in order to narrow the uncertainties for the DSA, DOE designed experiments to be performed by PNNL for the purpose of applying the resulting experimental data to the WTP model, as appropriate.

2.3 Mishima-Foppe Critique

The analytical study by Jofu Mishima and Terry Foppe (Reference 2-3 and included in Appendix D) concluded that the Handbook's spray release model may not be conservative in establishing an ARFxRF for certain WTP applications. This conclusion follows from these considerations:

- An ARFxRF value of 1E-4 for respirable droplets was originally selected for NRC evaluation by PNNL to address a seismic scenario in a specific facility (i.e., a mixed oxide fuel fabrication facility) and was actually incorrectly labeled as a "spray release." This ARFxRF value of 1E-4 represents an estimate of the stable post-interaction and deposition of a liquid aerosol in a glovebox, based on a 10 mg/m^3 "fog" limit due to breakage of glass and fragile equipment. The spray data were used to obtain perspective on ARFxRF in a glovebox for limited droplet evaporation applicable to such environment, and was not intended for determining the dose impact of a spray release.
- The ARFxRF 1E-4 value was used in 1992 analyses of potential releases from DOE weapons complex facilities. Shortly thereafter, this value was incorporated into DOE-HDBK-3010-94 as a bounding value for spray releases. Figure 3-4 of the Handbook displayed commercial spray data for water. This figure shows the size distribution of a spray formed by forcing liquid through a pressure nozzle or orifice. The size distribution becomes finer with decreasing size of orifice and increasing upstream pressure.
- The recommended bounding value of 1E-4 in DOE-HDBK-3010-94 remains valid only for the studied glovebox. It is not a bounding value for liquid droplets of respirable size generated by sprays from metal piping and vessels as a function of orifice size, configuration, and upstream pressure, with liquid properties that may be significantly different than water.

Mishima and Foppe also discussed the effect of physical properties of non-Newtonian fluids present at WTP. These fluids include supernatant liquids on top of undissolved solids, high-dissolved solid solutions, low solid slurries, and high solid slurries. The authors indicated there is a lack of physical data available for density, viscosity and surface tension for these liquid slurries. The Handbook, they pointed out, only addresses liquids and does not analyze non-Newtonian fluid behavior.

The authors identified these measures to enhance the Handbook for use at WTP:

- Establish a data base of relevant physical properties of the various fluids (e.g., slurries) anticipated for the tank farms and WTP;
- Consider application of empirical correlations using (a) appropriately conservative assumptions for input parameters specific to the waste solution physical properties and (b) applicable ranges of the correlations to calculate the bounding respirable diameter ($\leq$ 10 μm) for spray releases;
- Use caution in analyzing evaporation of larger droplet sizes to respirable sizes, using a drop size fitting parameter, to avoid excessive conservatism;
- Consider the plugging potential of the waste slurry with respect to smaller orifice diameters;
- Perform experimental studies to determine discharge rate droplet size distributions of various fluids for the range and types of breaks anticipated; and
- Consider publishing a change notice to DOE-STD-3010-94 to provide additional clarifications on the applicability of the current recommendations and alternative approaches to establish a bounding estimate. The change would increase the current ARFxRF value by a factor of 20 from 1E-4 to 2E-3 until a more general model is developed through a complex-wide consensus process.

2.4 References

2-1. Winokur to Triay, April 5, 2011, available on the DNFSB website.

2-2. Triay to Winokur, June 3, 2011, available on the DNFSB website.

2-3. Jofu Mishima and Terry L. Foppe, "Review of the DOE-HDBK-3010-94: Airborne Release Fractions and Respirable Fractions for Spray Releases from Hanford Waste Solutions," Jan. 20, 2010.

3.0 PNNL EXPERIMENTS [2]

3.1 Spray Leak Test Objectives

To address the DNFSB and Mishima-Foppe concerns, PNNL developed test objectives that would provide additional information in areas of uncertainty. The overall objective of the testing was to determine aerosol droplet size distribution and total droplet volume from prototypic breaches and fluids, including sprays from larger breaches and sprays of slurries for which literature data are mostly absent. To address this, the testing program collected aerosol generation data at two scales, commonly referred to as small-scale and large-scale testing. The small-scale testing and resultant data are described in PNNL-21367, and the large-scale testing and resultant data are presented in PNNL-21333. In tests at both scales, simulants were used to mimic the relevant physical properties projected for actual WTP process streams. The small-scale system was used initially for as much testing as possible. The large-scale testing was then used to confirm and expand these results. The test objectives were:

Small-scale spray leak testing:

1. Breach plugging due to slurries (note: no aerosol characterization was performed for this objective). The objective is to determine the size of circular and slot-shaped breaches that will plug and not form appreciable sprays with slurry simulants with an appropriate time period (such as 15 minutes).
2. Aerosol quantification of small-scale spray leaks of water, non-hazardous salts (such as sodium nitrate), and non-hazardous slurries (such as Gibbsite). The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for a range of smaller breach sizes for circular and rectangular breaches, liquid and slurry simulants, and process conditions.
3. Aerosol quantification of small-scale spray leaks with chemical slurry simulants. The test objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for a chemical slurry simulant representative of a washed and leached process stream. The results will be compared with the results from the non-hazardous simulants.

Prototypic large-scale spray leak testing with aerosol characterization:

4. Aerosol quantification of large-scale spray leaks of water. The objective is to determine the size distribution of aerosol droplets and total droplet volume concentration as a fraction of the total spray volume for a range of circular and rectangular breach sizes.

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[2] Portions of this section have been extracted from the text of the referenced PNNL test reports.

5. Aerosol quantification of large-scale spray leaks of non-hazardous slurries (such as Gibbsite) and liquids. The objective is to determine the size distribution of aerosol droplets and total droplet volume concentration as a fraction of the total spray volume for a range of circular and rectangular breach sizes with non-hazardous slurries and liquids.

After the original suite of small-scale and large-scale testing was completed, additional test objectives were developed. The second phase of testing (Phase II) focuses on quantifying the effect of the spray and chamber length on aerosol generation and evaluating aerosol generation from additional non-Newtonian and Newtonian slurries. This additional scope is being conducted to reduce the uncertainty in the estimated aerosol generation from long-distances sprays of both Newtonian and non-Newtonian slurries and to evaluate additional slurry sprays that may potentially give higher release fractions than the slurries evaluated earlier. The additional testing scope is identified as Phase II.

Phase II - Small-scale spray leak testing:

6. Aerosol quantification of small-scale spray leaks with chemical slurry simulant at 6 Pa/6 cP and 30 Pa/30 cP. The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for a chemical iron-rich slurry simulant of sludge waste representative of a washed and leached process stream. Also to determine the droplet size distribution directly in the spray (in-spray measurement).
7. Aerosol quantification of small-scale spray leaks with clay slurry simulants achieving rheology limits of 6 Pa/6 cP and 30 Pa/30 cP. The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for non-Newtonian clay slurries. Also to determine the droplet size distribution directly in the spray (in-spray measurement).
8. Aerosol quantification of small-scale spray leaks of a non-hazardous slurry at 27 wt% UDS. The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for a non-hazardous slurry simulant with 27 wt% UDS. Also to determine the droplet size distribution directly in the spray (in-spray measurement) for this simulant.
9. Aerosol quantification of small-scale spray leaks of a non-hazardous slurry with a small fraction of very dense particles (such as stainless steel (SS) or molybdenum (Mo) metal). The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for a slurry simulant with 20 wt% UDS including a target quantity of about 1 wt% of very dense particles. Also to determine the droplet size distribution directly in the spray (in-spray measurement) for this simulant.
10. Malvern validation testing. The objective is to assess the capability of the Malvern Insitec-S in-process particle size analyzer, which is the instrument used in the aerosol

testing, to measure accurately the concentration and size distribution of samples. This will be accomplished by measuring carefully controlled dilute aqueous slurries of known concentration and particle size distribution and comparing the Malvern result to the known values. Testing will include mono- and poly-disperse suspensions and will evaluate all four Malvern configurations used in the testing.

11. Compare aerosol measurements from old and new Malvern lenses. The aerosol results from the Malvern Insitec-S using the new 500 mm lens that has a nominal measurement range of 2.5 - 2500 µm (Malvern Instruments, Ltd. 2010) are compared to aerosol results using the 100 mm lens employed in Phase I that provided a nominal range of 0.5 to 200 µm (Malvern Instruments, Ltd. 2010). Tests will use one or more orifices.

Phase II - Large-scale spray leak testing:

12. Aerosol quantification of the effect of chamber size and spray length. The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for water sprays for a range of different chamber sizes and for sprays traveling different distances within the largest chamber. Also determine the droplet size distribution directly in the spray (in-spray measurement) for the largest sprays that can be measured. Testing will be conducted using at least one circular hole and multiple rectangular breaches up to the 1x76 mm breach at target pressures between 100 and 380 psi. The chamber length will be varied from 39 ft to 10 ft and perhaps as small as 5 ft if the data show this will help the extrapolation to long-distance sprays.
13. Aerosol Quantification of Large-Scale Spray Leaks with Clay Slurry Simulants at 6 Pa/6 cP and 30 Pa/30 cP. The objective is to determine the size distribution of aerosol droplets and the total droplet volume concentration as a fraction of the total spray volume for non-Newtonian clay slurries. Also to determine the droplet size distribution directly in the spray (in-spray measurement) for these simulants. The rheology of the simulant will be adjusted so that one slurry, at the beginning of testing, has at least one Bingham parameter near 30 Pa/30 cP (target range is 30 ± 4 Pa or cP) and the second Bingham parameter should be less than or equal to the upper 30 ± 4 Pa or cP target. The second simulant will be adjusted so that one slurry has at least one Bingham parameter near 6 Pa/6 cP (target range is 6 ± 2 Pa or cP) and the second Bingham parameter should be greater than or equal to the lower 6 ± 2 Pa or cP target. Testing will be conducted using at least one circular hole and multiple rectangular breaches up to the 1x76 mm breach at target pressures between 100 and 380 psi.
14. Compare aerosol measurements from old and new Malvern lenses. The aerosol results from the Malvern Insitec-S using the new 500 mm lens that has a nominal measurement range of 2.5 - 2500 µm (Malvern Instruments, Ltd. 2010) are compared to aerosol results using the 100 mm lens employed in Phase I that provided a nominal range of 0.5 to 200 µm (Malvern Instruments, Ltd. 2010). Tests will use one or more orifices.

3.2 Spray Leak Testing Summary

The spray testing program was conducted in 2012-2013 by PNNL in two phases. Experiments were conducted at three pressures (100, 200, and 380 psig), using several circular orifices (ranging from 0.2 to 4.46 mm in diameter) and slot-shaped orifices (0.3 × 5, 1 x 10 mm, 1 x 20 mm, 1 x 76 mm, and 2.74 x 76.2 mm), and using water, solutions, and Newtonian and non-Newtonian slurry simulants.

Several simulants were developed and characterized for use in the small-scale Phase I plugging and aerosol tests. The simulants have been selected to represent a range of relevant physical and rheological properties. Table 3-1 lists the target simulants.

Table 3-1. Phase I Small-Scale Target Simulants

Material	Target Property Range
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
Solutions of water and sodium nitrate/sodium thiosulfate	Viscosities of ~1.5 and ~2.5 mPa·s (cP).
Gibbsite and boehmite particulates in water	The particle size distribution (PSD) of the slurries were selected to match small treated Hanford slurries with 8 and 20 wt% solids.
A washed and leached simulant (PNNL-18894) (Reference 3-1)	Solids loading was adjusted to meet target Bingham yield stresses of 6 and 30 Pa.

Because of the hazards and costs associated with some of the simulants developed for the small-scale test stand, only a small subset of simulants was used in the large-scale test stand during Phase I testing. Table 3-2 lists those simulants.

Table 3-2. Phase I Large-Scale Target Simulants

Material	Target Property Range
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
Solution of water and sodium thiosulfate	Viscosity of ~2.5 mPa·s (cP).

Table 3-2. Phase I Large-Scale Target Simulants

Material	Target Property Range
Boehmite particulates in water	The PSD of the slurries was selected to match small treated Hanford slurries with 8 and 20 wt% solids.

The simulants used in the small-scale Phase II aerosol tests are listed in Table 3-3.

Table 3-3. Phase II Small-Scale Target Simulants

Material	Target Property Range
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
Boehmite particulates in water	The PSD of the slurries was selected to match small treated Hanford slurries at a concentration of 27 wt% solids.
Small fraction of Mo in water and a boehmite-water slurry	1 wt% (in the slurry) Mo particles included to represent dense particles in the waste such as plutonium oxide. The boehmite slurry had a total solids loading of 20 wt% (i.e., 19 wt% boehmite and 1 wt% Mo).
Clay slurries composed of a solid phase with 80 wt% kaolin and 20 wt% bentonite in water	The total solids loadings were adjusted, via dilution, before testing began so that one simulant had at least one Bingham parameter near 30 Pa/30 cP (target range was 30 ± 4 Pa or cP) and the second Bingham parameter less than or equal to the 30 ± 4 Pa or cP target. The second simulant was adjusted so that at least one Bingham parameter near 6 Pa/6 cP (target range was 6 ± 2 Pa or cP) and the second Bingham parameter greater than or equal to the 6 ± 2 Pa or cP target.
A washed and leached simulant (PNNL-18894)	Same.

The simulants used in the Phase II large-scale aerosol tests are listed in Table 3-4.

Table 3-4. Phase II Large-Scale Target Simulants

Material	Target Property Range
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
80/20 solids blend of a kaolin/bentonite clay slurry, 32 wt%	The solids loading was adjusted to meet target Bingham yield stress of 30 Pa.
80/20 solids blend of a kaolin/bentonite clay slurry, 27 wt%	The solids loading was adjusted to meet target Bingham yield stress of 6 Pa.

Tests were conducted by varying the distance between the orifice and a splash wall perpendicular to the spray, and also by varying the chamber size in which tests were conducted. Two methods were used to determine the aerosol generation rate, termed "in-spray" and "in-chamber."

Three experimental methods were considered to measure the aerosol net generation rate and release fraction: 1) direct in-spray measurements, 2) steady-state aerosol concentration measurements in a chamber with different volumetric purge rates, and 3) transient aerosol concentration measurements in a chamber with no purge flow. The first experimental method measures the aerosol directly in the spray, providing an explicit measurement of the aerosol droplet size distribution at a specific position. The release fraction for any given size of droplet is equal to the volume fraction of it in the spray, as given by the droplet size distribution. The second experimental method is to generate a steady spray and measure the steady-state concentration within a chamber by varying the flow rates of clean air introduced into the chamber to dilute the aerosol. The net generation rate is calculated from the measured aerosol concentration with different purge rates. The third experimental method consists of measuring the rate of increase in aerosol concentration in a closed chamber of known volume. Using a simple material balance, the rate of concentration increase gives the aerosol net generation rate from a spray. The first and third methods were used in the testing.

The in-spray measurement technique for the jet centerline was capable of measuring the larger droplets but not droplets less than 50 µm. The in-chamber measurements used the time history of aerosol concentration in the chamber together with a first order rise model to determine the generation rate. The in-chamber measurements accounted for wall deposition, and any net loss or gain from splash and splatter on the far wall as well as any splash of droplets in the pool on chamber floor. The in-chamber method was found to be biased low for larger droplet due to gravitational settling.

A reasonably conservative correlation for aerosol generation rate was developed based on the in-chamber test data and extrapolations of the in-chamber data to 100 ft chambers. The correlation was developed for water, but it is appropriate for all the liquids and slurries tested because the aerosol generation from the other fluids is overwhelmingly always the same or less than water sprays. The primary exception is the result set for the non-Newtonian chemical slurry simulants. These slurries had unusual rheology in comparison to actual waste and the clay simulants, making the applicability of these results questionable. The correlation was compared to in-spray data and was found to match the in-spray data for the range of orifices and spray pressures tested. The good comparisons in the regions of overlap for different size orifices and different spray pressures confirm that the conservative correlation has orifice area, spray pressure, and droplet size dependences that agree with in-spray data. Because the conservative correlation matches the in-spray results, it can be concluded that the conservative correlation accounts for the potential biases (humidity and method bias) with the in-chamber method without actually quantifying them.[3]

Because the upper confidence interval correlation and values account for the uncertainty in fitting the chamber concentration data, the correlation for the upper confidence interval will be used in developing a reasonably conservative correlation. One approach for obtaining a conservative correlation is to adjust the correlation so that all, or the majority, of the measured values are less than the correlation. The most sensible adjustment is to increase the leading coefficient and not adjust the exponents for the individual parameters of orifice area, spray pressure, and droplet size. The following result (see PNNL-22415, Equation 10.4, Reference 3-2) was selected to have nearly all of the measured values in the upper confidence interval be the same or less than the conservative correlation.

$$GR_C = 3.26 \times 10^{-16} (A)^{0.793} (P_s)^{2.18} \left(d_p\right)^{2.40} \qquad \text{Equation 3-1}$$

where:

GR_C is the conservative correlation for generation rate (m^3/s)

A is the orifice(s) area (mm^2)

P_s is the spray pressure (psig)

d_p is the aerosol droplet diameter (μm)

[3] Venting of superheated liquid (flashing spray) is not within the scope of this technical report and was not tested

Figure 3.1 shows the comparison of the measured values in the upper confidence interval with the conservative correlation. The results from the individual tests are less than or equal to the conservative correlation, with the exception of a couple of individual points.

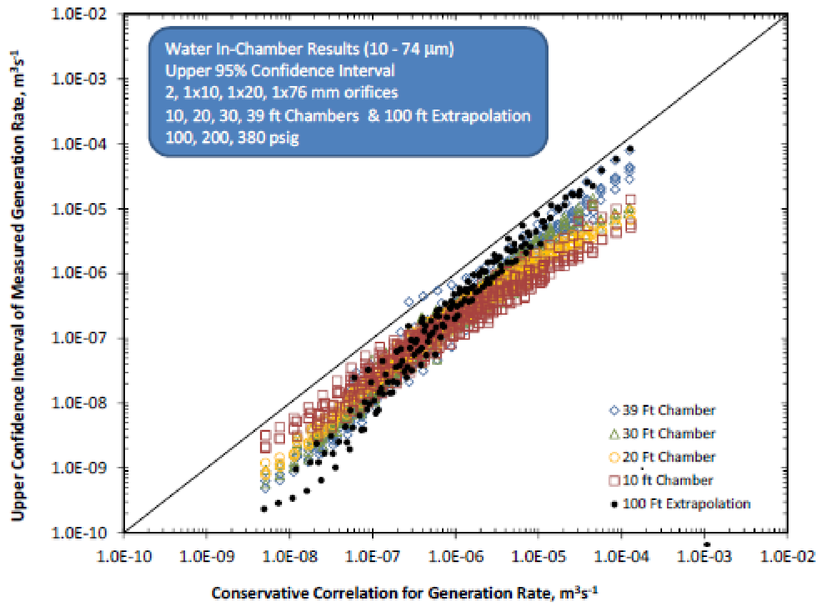

Figure 3.1 Comparison of Measured Upper Confidence Interval of Generation Rate with the Conservative Correlation for Generation Rate

Figure 3.2 shows a comparison of the measured generation rates with the conservative correlation. The measured generation rates are lower than the upper confidence interval values, and this figure shows that all of these measured generation rates are farther below the diagonal line, as expected.

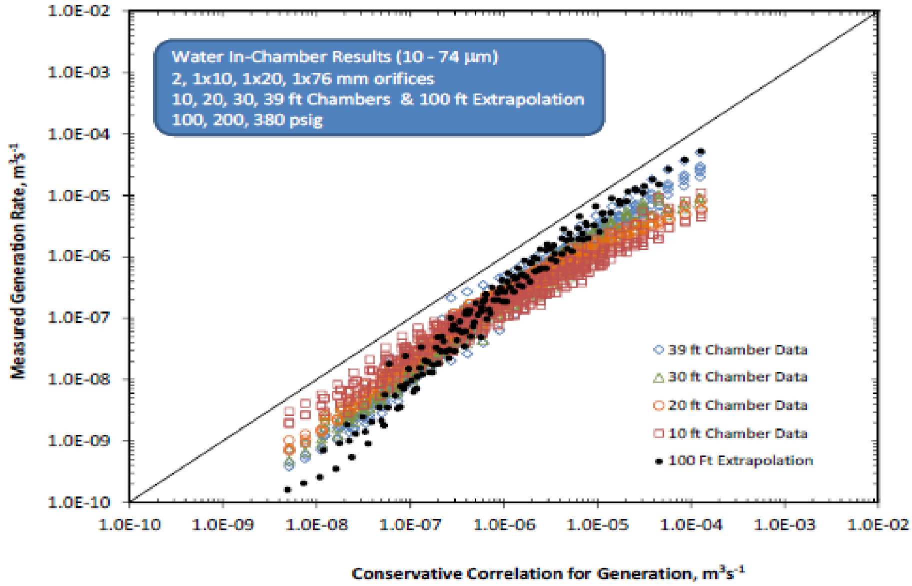

Figure 3.2 Comparison of the Measured Generation Rates with the Conservative Correlation

3.3 Range of Parameters within Experimental Protocols

3.3.1 Breach Size

A spray leak scenario postulates a small breach in the primary confinement boundary of a pressurized liquid, resulting in a spray of hazardous material. The small opening may be caused by corrosion, erosion, jumper misalignment, weld crack, seal leak at pump or valve stem, impact, or other initiators. Due to the large differences in the design, operating conditions, and failure mechanisms, the size and morphology of the breach are indeterminate. However, the postulated breaches can be divided into two broad groups: those resulting from a sudden change such as an impact, seismic event, or jumper misalignment; and those resulting from gradual deterioration such as corrosion or erosion. The approach described here assumes that the breach in the confinement boundary occurs suddenly. This approach provides significant conservatism for the cases where the breach develops over a period of time, such as a breach due to corrosion.

Given the above, three different approaches, all of which are used in this report, can be used to establish the postulated breach size.

1. **Use of historical data**. Past failure data may be analyzed for causes and resulting breach sizes to determine some "worst case" or representative scenarios. However, available historical data are very limited, and may not apply to the design and operating conditions for a facility. A criterion to determine the "worst case" or representative scenario is also needed.

2. **Optimization approach**. The breach area is determined by optimizing the source term. The flowrate through the breach increases with breach area, provided pressure can be maintained. Provided the release fraction does not decrease with flow area more strongly than the inverse of the flow area, then the optimized area is the largest breach area. However, the atomization becomes less efficient with increase in orifice area. In this case an optimum area may be found when the release fractions are a strong function of the orifice area. The PNNL correlation shows an increase in aerosol generation rate with increased orifice area. That is, the conservative PNNL correlation shows a weak dependence of cumulative release fraction (CRF) on the orifice area $\left(\mathrm{CRF} \propto \mathrm{A}^{-0.207}\right)$ so that the optimum breach area is at the upper boundary of postulated sizes. In this case, the breach area can be based on a credible range. However, with further increase in breach area, the release fractions are expected to decrease significantly and the release will transition from spray to spill because the pressure at the breach location will decrease with increasing flow rate, and the atomization will become coarse due to both reduced jet speed and larger jet size. With increasing orifice area, a sharp transition from spray to spill is not expected. The length and optimized width are discussed in more detail below.

3. **Regulatory Guidance**. Such guidance is often based on historical data. As mentioned above, it is conservative to assume that loss of confinement occurs suddenly, as in the case of a jumper misalignment. Sudden breaches are more likely to be slit type failures rather than a large hole with equal area. For these reasons, the spray analyses will assume that a slit type breach occurs in the piping being evaluated. This approach is consistent with some past practices, and with the guidance in NRC's NUREG-0800 (Reference 3-3; see, e.g., Sections 3.6.1 and 3.6.2 and Branch Technical Positions 3-3, 3-4). The aerosol generation rate depends on breach area. Random holes produced by corrosion or erosion are normally closely spaced. The correlation for aerosol generation rates found in PNNL-22415 shows a weak dependence of cumulative release fraction on orifice size. Furthermore, the experimental data show that the aerosol generation rate from several nearby small holes are not much different than the aerosol generation rate from a larger hole of equal cross sectional area. These results support modeling of the postulated breach for spray leak source term as a single slot. Assumption of slit-type failure provides a method to determine a reasonably conservative size of breach, and otherwise has no effect on source term calculation. Postulating the largest breach is more conservative when event duration is constant; that is, when there is enough material to spray over the event duration.

   The objective of NRC guidance is similar but not identical to the objective of the spray leak source term methodology. Process piping with operating pressures greater than 275 psig does not meet NRC's definition of moderate energy fluid systems. However, the criteria for high energy line breaks in NUREG-0800 address a double-ended guillotine break or a longitudinal break with surface area equal to the cross-section of the pipe. Such large breaks are more appropriately treated as a line break scenario rather than a spray-generating scenario.

a) Length

The total mass flow for a slit-type breach depends on the breach dimensions. It has been postulated that a leak could occur at a jumper or connection with a length equal to the circumference of the pipe. This would require the realization of leakage along the entire circumference; such an arrangement is inherently unstable and would lead to larger slit width. NRC's criteria for leakage cracks in moderate energy pipes (Reference 3-4) specify maximum breach areas for the events postulated for moderate energy fluid systems. The NRC specifies a length of one half the pipe diameter and a width equal to one-half of the pipe wall thickness for evaluating the impacts of medium energy line breaks on adjacent equipment. (Reference 3-5) A review of historic information in Reference 4-5 concerning leaks in Hanford's waste transfer system pipes and jumpers concludes that it was appropriate to assume a length equal to the pipe diameter with a 3 in. upper limit on length. This assumption was also considered appropriate for a jumper or flange connection leak, such as that from misalignment or sagging. The breach length is based on a combination of Reference 3-3 and 3-5 approaches as follows: (1) one pipe diameter for pipes less than 3 inches in diameter, (2) three inch for pipes from 3 inches to 6 inches in diameter, and (3) one-half of the pipe diameter for pipes greater than 6 inches in diameter.

b) Width

As stated earlier, the aerosol generation rate is increased with increased orifice area provided the spray pressure is maintained and event duration is constant. As already noted, NUREG-0800 criteria for leakage cracks in moderate energy pipes specify the breach width as one half the pipe wall thickness for moderate energy fluid systems. Consequently, the breach width is selected as one half the pipe wall thickness, provided there is sufficient material available that spray is sustained for the event duration.

c) Optimized Width

In some postulated spray release scenarios, the available material at risk may be exhausted in less than the event duration when the slit length and width described above are used. If this is the case, a narrower breach size is used so that the material at risk is exhausted at the end of the event duration. This approach will produce larger consequences over the event duration as compared with larger breach. For example, it can be shown, using the PNNL conservative generation rate correlation, that there is a 15% increase in the source term when the amount of release is constant and breach area is reduced in half. An air-based Weber number[4] of 60 represents the transition to a jet breakup regime where full atomization can occur. However, jet breakup may occur with the Weber number as low as 10 (Liu, Reference 3-7). For Hanford tank waste, a Weber number $\leq$ 10 corresponds to a width of 0.1 mm. As such, the adjusted width is chosen based upon Equation 3-2:

$$w_b = \max\left(0.0001,\ \frac{V_{max\_op}}{v_j \cdot l_b \cdot t}\right) \qquad \text{Equation 3-2}$$

where:

w_b is the width of the breach opening (m)

V_{max_op} is the bounding maximum operating volume of the vessel (m^3)

v_j is the jet velocity (m/s)

l_b is the length of the breach opening (m)

t is the spray duration (s)

PNNL-21361 states that the smallest round orifice tested (a diameter of 0.188 mm or 0.000188 m) plugged in 9 of the 11 tests and all of the tests at 20 wt% UDS plugged this orifice. In some instances, even the adjusted breach opening may result in an event duration shorter than the desired evaluation duration due to early exhaustion of the material. Optimizing the breach size only pertains to calculating radiological dose consequences. If toxicological consequences are being calculated, the original crack width is used to determine the peak 15-minute release.

3.2.2 Pressure

The differential pressure term in Equation 3-1 is determined using the maximum possible gauge pressure in the pipe. A calculated leak pressure may be used for the spray leak calculation or, if no calculation is available and a pump is the pressure source, then the shutoff pressure of the pump may be used. Alternatively, a hydraulic pump curve could be used to determine the pressure at the spray location.

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[4] The Weber number (We) is a dimensionless number in fluid mechanics that is often useful in analyzing fluid flows where there is an interface between two different fluids, especially for multiphase flows with strongly curved surfaces. It can be thought of as a measure of the relative importance of the fluid's inertia compared to its surface tension. The quantity is useful in analyzing thin film flows and the formation of droplets and bubbles.

3.2.3 Droplet Diameter

RPP-37897 (Reference 3-5) accounts for evaporation using a model that treats the suspended solids and solution residue as a single drop. The RPP-37897 model assumes that the maximum drop size that could become respirable, d_p, is given by:

$$d_p = \frac{10\ \mu m}{(f_{solid} + 0.1)^{1/3}} \qquad \text{Equation 3-3}$$

where f_{solid} is the volume fraction of suspended solids.

The added value of 0.1 presumes that, based on precipitation of a typical amount of dissolved solids, the droplet will lose no more than 90% of its initial volume to evaporation (i.e., the radius is reduced to 46.4% of the initial radius). The RPP-37897 model is based on the physical diameter of the drop rather than the AED because it does not account for drop density. The AED is equivalent to the diameter of a sphere of 1 g/cm^3 that exhibits the same terminal velocity as the drop in question. According to Stokes' law, the terminal velocity is proportional to the density and the square of the diameter and inversely proportional to a dynamic shape factor. The dynamic shape factor for spheres is 1. If the terminal velocity of a particle with diameter D, density ρ and shape factor κ is the same as the terminal velocity of a unit density sphere with diameter D_{AED}, then

$$\frac{D^2 \cdot (\rho/\kappa)}{(D_{AED})^2} = 1 \qquad \text{Equation 3-4}$$

After solving for D, the equation reduces to:

$$D = \frac{D_{AED}}{(\rho/\kappa)^{1/2}} \qquad \text{Equation 3-5}$$

Accordingly, d_p is defined as:

$$d_p = \frac{\dfrac{10\ \mu m}{(f_{solid} + V_{sol})^{1/3}}}{\left(\rho_{final}/\kappa\right)^{1/2}} \qquad \text{Equation 3-6}$$

where:

f_{solid} is the volume fraction of the suspended solids in the initial liquid slurry drop (dimensionless) and is calculated using:

$$f_{solid} = \frac{\rho_{slurry} \cdot wt\%_{solids}}{\rho_{solids}} \qquad \text{Equation 3-7}$$

V_{sol} is the fraction of the initial drop volume represented by the volume of solution remaining when the concentration reaches 19 M (dimensionless)

ρ_{final} is the droplet density after evaporation (g/cm^3)

κ is the dynamic shape factor value (1.5) consistent with the shape factor used for the human lung model in International Commission on Radiological Protection (ICRP) Publication 66, page 49 (Reference 3-6).

The expression for d_p has a singularity if both the Na molarity and solids content are zero. d_p is set equal to 100 µm for streams with little or no Na content (< 0.02 molar) and low concentrations of suspended solids (< 0.1 wt%).

3.3 Application Domain

The PNNL testing was performed at three pressures; 100, 200, and 380 psig. In Phase I, the tested orifices were circular holes ranging from 0.2 to 4.46 mm in diameter and rectangular slots (width × length) ranging from 0.3 × 5 to 2.74 × 76.2 mm. Phase II tested four orifices: a 2 mm diameter circular hole, and 1 x 10 mm, 1 x 20 mm, and 1 x 76 mm rectangular slots. The current test data estimates the aerosol generation rates for droplets between 10 and 100 µm, which corresponds to the size range needed for accident analyses. The particles sizes were chosen as 10 to 100 µm, since 10 µm is considered respirable, particles up to 100 µm are capable of evaporating to reach a respirable size, and particles larger than 100 µm tend to settle and not be transportable.

PNNL-22415 states that the conservative correlation for aerosol generation rate (Equation 3-1) can be used to extrapolate to larger orifices and higher pressures that were not tested and can also be used to interpolate to other conditions that were not specifically tested. A given example in PNNL-22415 uses a pressure of 540 psig and an orifice area of 290 mm^2. These are both approximately 40 % larger than what was tested.

3.4 References

3-1. PNNL-18894 (WTP-RPT-197, Rev 0), *Pretreatment Engineering Platform Phase-1 Final Test Report*, Dec. 2009, Pacific Northwest National Laboratory, Richland, WA.

3-2. PNNL-22415 (WTP-RPT-221, Rev 0), *Large Scale Spray Releases: Additional Aerosol Test Results*, August 2013, Pacific Northwest National Laboratory, Richland, WA.

3-3. NUREG-0800 (formerly issued as NUREG-75/087), *Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Power Plants*: LWR Edition, June 1987, U.S. Nuclear Regulatory Commission.

3-4. NUREG-0800, Branch Technical Position 3-3, Revision 3, 2007, Appendix A to J.F. O'Leary Letter of July 12, 1972, Section C(2)(2).

3-5. RPP-37897, Rev 1 and Rev 2, *Waste Transfer Leak Analysis Methodology Description Document*, Washington River Protection Solutions LLC. Richland, Washington, 2009 and 2010.

3-6. ICRP Publication 66, 1995, *Human Respiratory Tract Model for Radiological Protection*, Pergamon Press, New York.

3-7. Liu, H, 2000, *Science and Engineering of Droplets – Fundamentals and Applications,* William Andrew Publication/Noyes.

4.0 PNNL TESTING APPROACHES

The source term from a spray release event depends on five main factors: the breach in the primary confinement boundary (breach configuration), pressurized release of liquid through this breach causing spray or atomization of liquid into droplets (atomization), settling and transport of these droplets to the receptor location, evaporation during this settling and/or transport, and behavior of semi-dry or dry particulate during their transport to the receptor location. Other than the breach configuration and atomization process, these factors are common with other release scenarios for liquid waste or chemicals.

These factors are coupled and depend on many parameters; most of these parameters are scenario specific. For example, DSD is a function of breach configuration. Evaporation and rainout from a plume of droplets are closely coupled with the jet velocity, entrainment of air into spray jet, DSD, and other scenario specific variables, such as chemical composition of the droplets, size of the room in which spray is introduced. Conservative assumptions that can decouple some of these phenomena will greatly simplify source term calculations. However, decoupling cumulative release fraction (CRF) from the orifice size (and possibly from operating conditions) will become excessively conservative for many situations.

Below, some parameters and phenomena related to spray leak scenario are discussed. These parameters were evaluated in the testing program performed at PNNL.

4.1 Plugging of Small Breaches

Plugging of an orifice or slot from solids in a slurry is a function of slurry properties, driving pressure, and orifice size. The slurry properties include particle size and shape, cohesive/non-cohesive nature of suspended solid particles, and their concentration. Clearly a breach will plug if the size of the breach is smaller than the largest particle in the slurry and the pressure is not large enough to shear or fracture these particles, plugging time being proportional to the concentration of these large particulates in the slurry.

PNNL-21361 test results (Reference 4-1) for four circular orifices (0.188 mm to 0.706 mm) and two rectangular orifices (0.260 x 4.946 mm and 0.357 x 5.021 mm) indicated that orifice plugging may be a function of solids concentration, but there is no observable plugging trend as a function of simulant particle size, over the size range tested. Four simulants were tested: water, solutions of water and non-hazardous salts (sodium nitrate and sodium thiosulfate), slurries (Gibbsite and boehmite particulates in water), and a washed and leached chemical slurry simulant where the solids loading was adjusted to meet target Bingham yield stresses of 6 and 30 Pa. PNNL noted that, given that some of the actual wastes have particles larger than the maximum particle size of the simulants, it is likely that some, but not all actual wastes could consistently plug the orifices tested. No combination of simulant and pressure produced plugging for round orifices $\geq$ 0.382 mm or for either of the slots tested. The smallest orifice (0.188 mm) plugged in 9 of the 11 tests. All tests at 20 wt% UDS plugged the smallest orifice.

In general, no consistent distinction could be made between the simulants in terms of plugging behavior, nor was there any recognizable trend with particle size or cohesiveness. There also was no clear trend for the effect of pressure on plugging. The orifice dimensions that can be assumed to consistently plug are, therefore, smaller than the orifice dimensions tested with the range of simulants and pressures employed.

4.2 Fluid Viscosity

The mean droplet size correlations available in the literature for pressurized atomization provide considerably different functional dependence of mean droplet diameter on fluid viscosity: a Sauter Mean Diameter $\propto \mu^c$ with the exponent c in the range 0 to 0.5. A smaller value of this exponent implies weak dependence.

PNNL-21361 test results indicated that for a salt solution, as viscosity and density increased, the cumulative release fraction was unchanged. PNNL-21333 test results (Reference 4-2) also indicated that for a salt solution, as viscosity and density increased, cumulative release fraction and generation rate varied by a slight amount that depended on droplet size. Considering uncertainty in test data, there was essentially no difference in the test results for water and salt solution. Not enough data was collected to determine the separate roles of viscosity and density.

4.3 Surface Tension and Anti-Foam Agent

For low viscosity fluids, breakup of fast-moving droplets into smaller droplets involves a competition between the destabilizing aerodynamic force and the stabilizing surface tension force, or alternately, between kinetic energy and the surface energy, i.e., the Weber number. (In this discussion, the Weber number is air-based.) The estimates for critical Weber number range from 10 to 100. (Note: PNNL tests with small-area orifices and 100 psig pressure generated respirable droplets at air-based Weber numbers of ~8.) For $We_a \approx 12$, a droplet may break into a few large fragments due to bag breakup; shear breakup requires a $We_a > 80$. Due to aerodynamic forces, the high speed liquid jet or sheet breaks up into large droplets, which break up into smaller droplets in "secondary atomization." Secondary breakup of droplets plays a significant role in the spray release atomization; and fluid surface tension is one of the main parameters.

A concern expressed by the DNFSB staff was that the greatly reduced equilibrium surface tension due to an anti-foam agent (AFA) may lead to much finer aerosol from postulated spray releases. PNNL tests (PNNL-21367) added AFA to either to two simulants, which approximately halved the equilibrium surface tension. The addition of AFA did not cause an increase in the release fraction. To the extent that an effect could be distinguished, the presence of AFA caused a slight decrease in the release fraction.

4.4 Orifice Discharge Coefficient

PNNL-21367 investigated the orifice coefficient. Average and standard deviation for orifice coefficients were 0.59 ± 0.05 when orifice area was >2 mm², consistent with the conventional value of 0.62. The discharge coefficient for orifices of <2 mm² area was 0.76 ± 0.06, or ~20% greater. Similarly, PNNL-21333 also investigated orifice coefficients. Overall the average orifice coefficient for all tests was 0.66 and 0.62 (two methods were used to determine the coefficient) and if several outliers, which occurred in 11 of 246 tests, are disregarded the overall average orifice coefficients were 0.65 and 0.63 for the two methods. PNNL-22402 (Reference 4-3) reports the results of small-scale additional testing. The conclusion was that the small-scale system had accumulated a large data set of orifice coefficients which suggests that, particularly for slurries, orifice coefficients significantly greater than 0.62 occur in orifices regardless of type. The conservative generation rate correlation developed by PNNL uses an empirical approach that does not explicitly use an orifice discharge coefficient.

4.5 Slot Orientation and Aspect Ratio

The orientation of the postulated slot is needed to discern any change in atomization due to slot orientation or flow state near the slot. Three situations were tested: (a) the slot length aligned with the pipe axis, water flowing through the pipe and spraying through the slot, (b) the slot aligned along the pipe circumference, water flowing through the pipe and spraying through the slot, and (c) an axial slot in the 'dead-end' configuration.

Tests to compare the release fractions with change in slot length or width, i.e., the aspect ratio, were also conducted. These tests were conducted under similar conditions: in the same test chamber, with identical distance from the spray header to the splatter wall. By their nature, these tests included any contribution from splash and splatter.

PNNL-21367 testing indicated that for round holes, as orifice area increased, the cumulative release fraction was essentially constant for <10 µm drops. Cumulative release fractions for <30 µm and <100 µm drops showed an area dependence. Cumulative net generation rate increases with orifice area due to increase in total spray flow. The test report notes that the cumulative release fraction correlates reasonably well with orifice area for slots and round holes. Dependence on orifice area varies between smaller and larger areas. However, there is insufficient data to determine whether the difference in dependence at small and larger areas is due to orifice area or geometry. For drops between 10 µm and 100 µm, as the slot orifice area increased, cumulative release fraction decreased and cumulative net generation rate increased slightly. PNNL-21333 testing indicated that as orifice area increases, cumulative release fraction decreases while cumulative generation rate increases.

The dependence on area was found to hold regardless of orifice shape and slot orientation and did not have an appreciable effect on the release fraction. Finally, PNNL-22415 (Reference 4-4) compared the correlation to in-spray data and found a match for the range of orifices and spray pressures tested. The use of a conservative generation rate correlation developed by PNNL captures the tested behavior.

4.6 Multiple Nearby Orifices

Jumper misalignment may lead to a slot-type breach of varying thickness along the connector circumference. Local corrosion or erosion may cause one or several small nearby holes, which may increase in number and size, and may merge to form a breach with irregular edges, varying width, multiple connected branches, and a rough flow path. It is a formidable task to define and manufacture realistic and representative breaches resulting from significantly different failure mechanisms and operating conditions. Tests to determine impact on release fractions for several nearby holes in order to simulate an irregular breach were conducted by PNNL; see PNNL-21367 and PNNL-21333. Tests were performed using an array of five round holes lined up with the header axis, with each orifice separated by a distance equal to the hole diameter. The use of a test piece with an array of closely spaced orifices is an attempt to mimic an actual breach. The array of 1-mm orifices had a cross-sectional area that was very similar to the 2-mm orifice (3.69 vs. 3.50 mm^2). As expected, the release fractions of the 2-mm orifice and the array of 1-mm orifices are very similar over the entire droplet size range and are higher than the release fractions measured for a single 1-mm orifice. That they are in good agreement suggests that only cross-sectional area is important for predicting the release fraction of aerosol.

4.7 Solids in Newtonian Slurries

Waste contains undissolved particulate, and most of the radioactive material is in this solid phase. Therefore, the slurry streams are of most concern from the radiological source term perspective. Pressure atomization of slurries involves interactions of the three different phases (suspended solids, carrier liquid and dissolved salts) with air at atmospheric pressure.

No data was found in open literature for pressure atomization of Newtonian or non-Newtonian slurries from simple orifices. Although slurry atomization is extensively used in industry to manufacture spray-dried goods, there is very limited data on hollow-cone and air-assisted atomization of slurries. This data is not applicable to spray leak scenarios, but may be useful in qualitative judgments about the effects of various parameters.

One direct effect of suspended solids is increased viscosity. Many factors influence the slurry viscosity; these factors include viscosity of the carrier fluid and the parameters related to the suspended particulate such as density, shape, size, size distribution and concentration. The slurry viscosity generally increases with decreasing particulate size and increasing solids loading, this trend is attributed to increased number density and surface area of the solids.

PNNL-21333 states that overall, the literature on aerosol formation with slurries suggests that slurry particles can increase and decrease the size of droplets.

4.8 Surface Tension

Another fluid parameter that effects atomization is surface tension. Data found in the literature measured both static and dynamic surface tension of coal-water-slurries containing 40 wt% coal. Three different particulate sizes were used: 32-45 μm, 45-63 μm, and 63-90 μm. The measured static surface tension values were equal to that of carrier fluid (water) within the measurement uncertainty, and the dynamic surface tension was measured to be marginally (5%) higher than that of the carrier fluid. Spray tests were also conducted using co-flowing sonic air to assist atomization of liquid jet discharged at relatively low pressure. The result was an increase in Sauter Mean Diameter with decreasing size of solid particulate. Smaller coal particles strongly coalesce to one another and strongly retain water between them. The high capillary effects and tight packing is attributed to larger Sauter Mean Diameters of coal-water slurries containing smaller particles (Son and Kihm, Reference 4-7).

PNNL-21333 testing indicated a straightforward functionality of cumulative release fraction on pressure and orifice area. Viscosity and weight fraction of solids had a negligible effect. As the weight fraction of solids increases, the cumulative release fraction is unaffected for droplets >10 μm. There was some deviation from water at droplet sizes <10 μm, but differences below this droplet size are of minor concern for spray release accident analyses. Configuration and geometry of spray are also important, as evidenced by splash wall distance and in-spray aerosol tests.

4.9 Small Concentration of Dense Particles in Slurries

Waste contains insoluble solids consisting primarily of oxides and hydroxides of metals used in the fabrication and processing of nuclear fuels. Some of the undissolved solids in the slurry have a large density compared to that of the carrier fluid. The concentration of these dense particulates is small as compared to the other undissolved solids. During atomization, the droplets decelerate due to aerodynamic drag. A concern was raised that the dense solid particles in the droplets may not decelerate, leading to fission of the droplet into smaller droplets. Testing is needed to determine the effect of dense solid particles on aerosolization.

PNNL-21367 (Reference 4-5) indicated that low solids concentrations (such as 8 wt%) appeared to depress release fractions below those of water over most or all of droplet size range for baseline slot and round orifices. Increasing solids content to 20 wt% increased the release fraction. PNNL-22402 indicated that addition of a small fraction (nominally 1 wt%) of dense particles to water and 19 wt% small treated simulant (STR) did not result in a significant effect on measured release fractions when compared to simulants devoid of dense particles.

4.10 Effect of Non-Newtonian Rheology

During processing at WTP, the waste slurry will be concentrated by filtering the supernate. The concentrated slurry will be washed and leached to dissolve non-radioactive constituents from the solid phase. The leached slurry will be concentrated by filtering the supernate. Filtering will be done in a high pressure and high flow rate ultra-filtration recirculation loop. The washed and leached slurry streams will contain relatively fine particulate at solid concentrations of up to 27 wt% in aqueous solutions that exhibit non-Newtonian behavior, with Bingham yield stress/ consistency in the range of 6 Pa / 6 cP to 30 Pa / 30 cP. Because of the higher process pressure and the higher concentration of radioactive material, these washed and leached non-Newtonian slurry streams present some of the largest hazards from the radiological source term perspective. Available literature data and correlations point to reduced release fractions due to increased viscosity, but these predictions are based on the viscosity of liquid phase and not on interaction of liquid phase with the suspended solids. Consequently, tests were conducted to understand the role of Bingham rheology caused by relatively fine solid particulate in water or dilute caustic solution.

PNNL-21367 testing showed that for water sprays, as pressure increased, the cumulative release fraction increased; this is consistent with large-scale testing results. However, for non-Newtonian simulants, the effect of pressure was variable with release fraction, sometimes increasing and sometimes decreasing with increasing pressure. PNNL-22415 testing measured in-spray release fractions for water and for two non-Newtonian clay slurries as a function of downstream distance from the orifice. For many test pressures, orifices, and simulant combinations, the in-spray release fraction did not change substantially with increasing distance from the orifice. Also, for in-spray measurements, the release fraction for clay slurries at 380 psig is the same as water. At 100 and 200 psig, the release fraction for clay slurries decreases with increasing solids content, which shows that water results are typically the same or larger than release fraction measured for non-Newtonian clay slurries. Finally, PNNL-22415 states: "A reasonably conservative correlation for aerosol generation rate was developed based on in-chamber test data and extrapolations of the in-chamber data to 100 ft chambers. The correlation was developed for water, but is appropriate for all liquids and slurries tested. The primary exception is results for non-Newtonian chemical slurry simulants that have unusual rheology in comparison to actual waste and the clay simulants, making the applicability of these results questionable." PNNL-22402 indicated that 27 wt% STR slurry had cumulative release fractions very similar to water and 20 wt% STR slurry. Comparison of clay slurry and water cumulative release fractions in 10 µm to 100 µm droplet size range showed that clay release fractions are less than or equal to those of water at both 6 Pa and 30 Pa yield stress.

4.11 Contribution from Splash and Free Fall

The pressurized fluid released from a breach may impact nearby surfaces (such as walls or other structural elements or piping) or it may splash in the expanding liquid pool on the floor. The DNFSB expressed concerns regarding the contribution to the source term of additional droplet formation, caused by (a) impingement on surfaces and (b) impingement on a liquid pool. Testing was done to determine the effect, if any, from impact on nearby surfaces.

PNNL-21367 testing indicated that as distance between spray and splash wall decreased, cumulative release fraction remained essentially constant between 42 in and 18 in, increased slightly between 18 in and 3 in, and increased significantly at a distance of 1 in. PNNL-21333 states that for the large-scale chamber, the spray distance varied from 43 in. to 227 in., essentially the full length of the chamber. For the five different orifices tested at pressures of 200 and 380 psig, the largest release fraction always occurred with sprays that traveled the full length of the chamber.

4.12 Evaporation

Evaporation effects for the case of a spray at 200 psi through a 0.128 in. diameter hole were investigated. This investigation considered three different fluids: water, waste with high suspended solids but no NaOH, and waste with suspended solids and small amounts of NaOH. The analysis considered a cell with the dimensions and ventilation flow rate representative of a processing hot cell. The assumed initial conditions in the cell were 45 °C and 5% relative humidity (RH). The report concludes that, if the hygroscopic nature of NaOH is ignored, droplets with initial diameters 100 μm or smaller will evaporate to near dryness, while larger drops will retain a substantial fraction of their initial volume. The report also notes that the droplets will not in fact dry completely and that the suspended solids are not expected to become a "truly dry dust." Finally, the report shows that the evaporation rate will decrease rapidly from an initial peak to roughly two thirds of the peak rate in less than one hour. Evaporation is further reduced as the relative humidity increased in the experiments to roughly 90 % after three hours.

This model effectively describes the behavior of essentially aqueous slurries. The results suggest that a very conservative model for such solutions is to assume that drops with diameters less than or equal to 100 μm generated in the first three hours become respirable, that is, to assume that the suspended solids become a truly dry dust. Following that period, the atmosphere approaches saturation and only drops with initial diameters equal to or less than 10 μm are considered respirable.

Evaporation was investigated during the tests. PNNL-22402 testing indicated that the initial RH in the chamber affects the measured release fraction, in particular for RH <80 %. Extrapolating to 100 % initial RH and interpolating to 80 % initial RH the decrease in release fraction is approximately a factor of two across the range of typical initial RHs for all droplet sizes. PNNL-

22415 also discusses testing of RH impact on water sprays was conducted in the 20, 30, and 39 ft chambers and humidity tests with the 20 ft chamber and 6 Pa clay simulant. All RH tests were performed using a nominal 2 mm hole and a 380 psig spray.

The results indicated that, regardless of chamber size, release fraction is reduced at low humidity across all droplet sizes. While release fraction measurements for water appear to be affected more strongly at smaller droplet sizes (<20 μm), the difference in the reduction for small and large droplets does not appear to be great. In other words, the impact of humidity on droplet size is significant for both large and small aerosols. The degree of reduction appears to scale proportionally to the difference between the test humidity and 100 percent, and any divergence from this behavior appears to derive from measurement uncertainty rather than a phenomenological mechanism. Low RH affects the clay release fraction differently than it does water release fraction.

Therefore, it is not surprising to observe different humidity correction factors for the 6 Pa clay tested. The difference derives both from different median and minimum RH values for 6 Pa clay, which are 92 percent and 81 percent, respectively, and from the presence of particulate that limits the role of evaporation with respect to aerosol size below aerosol diameters of ~50 μm. The presence of non-volatile clay solids also greatly reduces the effect of humidity on release fraction and reduces the overall magnitude of the worst-case humidity correction from ~1.7 for water to ~1.3 for clay. Evaporation caused nearly equal factor decreases in the RF for droplet sizes below 100 μm.

Two approaches for recommending a single correction factor for low humidity bias were taken. The first approach is based on the minimum RH, while the second represents the median RH observed in all the water and clay matrix testing. For the first and second approaches, the correction factors for water are 1.5 ± 0.2 and 1.2 ± 0.2, respectively. For clay, these same correction factors are 1.2 ± 0.1 and 1.1 ± 0.1, respectively. These correction factors are relatively small compared to the suspected magnitude of the method bias and the experimental variability noted between different experimental conditions.

4.13 Deposition

Aerosol is generated by primary and secondary jet breakup and by “splatter” droplets formed when the jet, or droplets formed by jet breakup, hit the splash wall at the downstream end of the enclosure. The in-flight and impact breakup events have not been distinguished, though it has been observed in previous testing that in-flight events appear to have a greater effect on aerosol generation than do impact events. Aerosol is “lost” from the bounded control volume through deposition onto chamber surfaces. The loss rate is proportional to the surface area, the droplet convective velocity, some form of a capture coefficient, and the droplet concentration. Aerosol also settles out at a rate proportional to the floor area, the droplet settling velocity, and the droplet concentration.

4.14 References

4-1. PNNL-21361 (WTP-RPT-219, Rev 1), Small-Scale Spray Releases: Orifice Plugging Test Results, September 2012, Pacific Northwest National Laboratory, Richland, WA.

4-2. PNNL-21333 (WTP-RPT-217, Rev 0), Large Scale Spray Releases: Initial Aerosol Test Results, December 2012, Pacific Northwest National Laboratory, Richland, WA.

4-3. PNNL-22402 (WTP-RPT-222, Rev 0), Small-Scale Spray Releases: Additional Aerosol Test Results, August 2013, Pacific Northwest National Laboratory, Richland, WA.

4-4. PNNL-22415 (WTP-RPT-221, Rev 0), Large Scale Spray Releases: Additional Aerosol Test Results, August 2013, Pacific Northwest National Laboratory, Richland, WA.

4-5. PNNL-21367 (WTP-RPT-216, Rev 1), Small-Scale Spray Releases: Initial Aerosol Test Results, May 2013, Pacific Northwest National Laboratory, Richland, WA.

4-6. "Consequences of Sprays from Pipe or Vessel Breaks," 24590-CM-HC4-W000-00176-T03-01-00001, Rev A.

4-7. Son, S. Y. and R. D. Kihm, "Effect of Coal Particle Size on Coal-Water Slurry (CWS) Atomization," Atomization and Sprays, Volume 8, pp. 503-519, 1998.

5.0 SUMMARY AND RECOMMENDATIONS

5.1 Revision of DOE-HDBK-3010-94

Taking into account all evidence presented in this report, the conclusion can be made that the ARFxRF value of 1E-4 provided in DOE-HDBK-3010-94 is not sufficiently conservative for all potential spray leak phenomenology. Appendix E provides a proposed revision to the Handbook's Section 3.2.2.3.1, "Venting Below the Liquid Level", and is recommended for use until the Handbook is revised, approved and issued. Appendix E incorporates the PNNL conservative correlation for generation rate (Equation 3-1) for spray leaks from pressurized process pipes. The conservative correlation is believed to provide a reasonable bounding estimation of ARFxRF, consistent with the approach to providing values in HDBK-3010-94. Use of conservative values is also consistent with the methodology described in DOE-STD-3009-2014, *Preparation of Nonreactor Nuclear Facility Documented Safety Analysis*, which requires bounding estimates of ARFxRF be used "unless a different value is provided in an applicable standard or is otherwise technically justified."

Appendix E recommends revising DOE-HDBK-3010-94 to limit use of the ARFxRF value of 1E-4 only for certain scenarios, specifically aerosol generation from breakage of glass-like equipment in a closed environment, such as a glovebox.

5.2 General Approach to Designing Controls for Spray Leaks

The design of controls to mitigate potential effects of spray leaks can be accomplished effectively by applying the following DOE-accepted safety principles:

- Hazardous material inventory should be minimized at all times,
- Safety SSCs are preferred over administrative controls,
- Passive SSCs are preferred over active SSCs,
- Preventive controls are preferred over mitigative controls,
- Controls closest to the hazard may provide protection to both workers and the public,
- Facility safety SSCs are preferred over personal protective equipment, and,
- Controls that are effective for multiple hazards can be resource effective.

The subsections below provide details on potential control methods and approaches.

5.2.1 Piping

The transfer pipe is the primary confinement boundary for liquids in transit in a facility or between facilities. Sturdy pipe such as ASME Schedule 40 or Schedule 80, procured to high quality standards, provides assurance that a spray leak will not develop. Additional assurance is provided by using co-axial piping, because if a leak does occur in the primary piping, it is contained in the secondary piping. Leak detection in the annular space between the primary pipe and the secondary pipe alerts operators so that the transfer can be terminated.

5.2.2 Confinement

The ceiling, walls and floor act in concert with the cascade ventilation system to provide secondary confinement of the aerosols from the spray leak. These structures ensure that aerosols generated by the spray leak event remain confined to the cell's air space, and that this contaminated air is exhausted by the cascade ventilation system. The cell walls passively maintain this function, provided confinement velocity is maintained by the cascade ventilation system across all potential leakage paths into the cell. The cell walls provide secondary confinement of the sprayed liquid waste and resulting aerosols. The cell walls also provide a control barrier effective for the accidents in which the waste is sprayed into the annular region of the coaxial pipe.

5.2.3 Cascade Ventilation

The cascade ventilation system is required to maintain the cell at a negative pressure with respect to the adjacent, lower contaminated areas of the plant, and to maintain confinement velocity across all penetrations in the cell wall. The design of the cell penetrations along with the depression and cascade airflow provided by the cascade ventilation ensure secondary confinement of aerosols within the cell.

5.2.4 Filtration

The active function of the cascade ventilation system also is required to ensure contaminated air is directed to and passes through HEPA filters. The HEPA filters provide secondary confinement by ensuring the air released to the environment is filtered. The filters are required to remove aerosols from the cell air before discharge to the environment. This system is a mitigative control for the spray leak accident.

5.2.5 Spray Shrouds

Piping often includes flanges, valves, and jumpers. The areas where pipes join together are more prone to spray leaks than uninterrupted pipe runs. Spray shrouds are devices that surround these areas and cause the spray to be directed into a waste collection system. In essence the spray leak is changed into a spill event. Leak detection is provided to alert operators so that the transfer can be terminated.

5.2.6 Moisture Monitors

A spray leak event causes the moisture content of the cell air to increase substantially. A moisture monitor in the cascade ventilation system can detect the increase in moisture and terminate the transfer.

5.2.7 Leak Detectors

Leak detection equipment is installed in the annular space in a coaxial pipe. If a spray leak occurs during the transfer of waste, the secondary pipe contains the fluid and it is detected in the annular region. Upon detection of the leak, the pump is shut down, thus terminating the release. Leak detection can also be provided in sumps. Again, if a leak occurs during the transfer of waste, liquid accumulating on the floor of the process cell migrates to the sump and is detected. The detection of a leak results in a termination of the transfer.

5.3 Value of Additional Testing and Experimentation

Three areas were identified as candidates for additional testing. Each is discussed below and reasons are given why no further testing needs to be conducted.

5.3.1 Higher Pressure Sprays

A reasonably conservative correlation for aerosol generation rate was developed based on the in-chamber test data and extrapolations of the in-chamber data to 100 ft chambers. The correlation was developed for water, but it is appropriate for all the liquids and slurries tested because the aerosol generation from the other fluids is overwhelmingly always the same or less than water sprays. The correlation was compared to in-spray data and was found to agree with the in-spray data for the range of orifices and spray pressures tested. This agreement tends to confirm (a) that the dependences of the conservative correlation on orifice area, spray pressure, and droplet size are consistent with in-spray data and (b) potential biases (humidity and method) have been accounted for without quantification.

The desired flow rates in the loop of >6.5 ft/sec and pressures of up to 380 psi at the test section were achieved using three Krebs millMAX centrifugal pumps connected in series, as illustrated in Figure 6.1. The 50 hp, 200 gpm slurry pumps were capable of producing 133 psig (with water) and handling non-Newtonian fluids with a Bingham rheology (consistency of 6 cP, yield stress of 6 Pa) with 50 μm, 2.5 specific gravity particles at a solids loading of 20 wt%. The flow rate through the pumps is controlled by Honeywell Variable Frequency Devices. These devices were connected in a master/slave configuration with the downstream pumps frequencies slaved to match the frequency of the upstream or master pump. Pressure in the loop is regulated by two globe valves located downstream of the test/bypass sections.

The flow rate through the loop is measured both upstream and downstream of the breach using two Coriolis mass flow meters. The locations of the Coriolis meters provides for sufficient pressure to minimize interference with the meter readings from entrained air/gas.

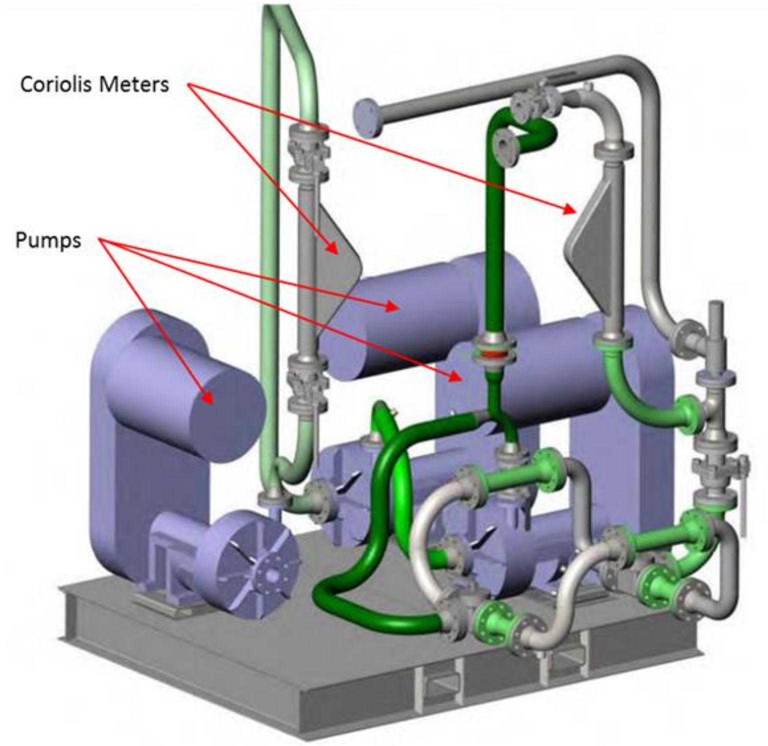

Figure 5.1: Schematic of the Flow Loop with Centrifugal Pumps on the Pump Skid

To test at higher pressures, the pump skid would need to be redesigned to accommodate additional pumps, a major endeavor. PNNL-22415 states:

> All results indicate that release fraction shows a statistically significant increase with increases in spray test pressure over the range tested in Phase II studies (100 to 380 psig). Evaluation of the pressure dependence of the results with a power-law model indicates that release fraction scales with pressure raised to a power that generally ranges from 1 to 2. Results for tests where chamber size was varied indicate that this pressure scaling depends on chamber and aerosol size. Release fractions for smaller aerosols show greater increase with increasing pressure relative to larger aerosols. Likewise, release fractions for aerosols generated in large chambers shows greater pressure dependence than those generated in confined spaces. Analysis of pressure scaling factors for tests in which spray length was varied in a chamber of fixed length indicate similar aerosol size dependence but do not suggest a statistically significant variation in scaling factors with actual spray length.

PNNL-22415 indicated that the conservative correlation for aerosol generation rate can be used for higher pressures up to 540 psig, hence no additional high pressure testing data is needed.

5.3.2 Spill/Spray Aerosol Ratio Quantification

Water spray tests demonstrated that the most optically dense region of the spray containing the largest droplets reaches the chamber floor at ~27.5 feet for 100 psig sprays from a 2 mm circular hole and at ~33.6 feet for the 380 psig sprays from 2 mm circular holes, 10 mm slots, and 20 mm slots. The mixing fans installed near the floor to improve chamber homogenization may have increased this distance, while the air backflow from the splash wall towards the orifice header may have decreased this distance. Data measurements in the 39 foot test chamber for these orifices include effects of splatter from the shallow pool at the chamber bottom. This shallow pool is created by the pre-spray used to wet the chamber and by the high initial relative humidity in the chamber. The PNNL testing did not separate the two effects or provide a ratio. However, a comparison of the conservative correlation for generation rate for spray leaks with the very conservative methodology in DOE-HDBK-3010-94 (for spills utilizing the Archimedes number for heights up to 24 m) shows that the conservative correlation is bounding. Hence, no further testing is recommended for this topic.

5.3.3 Evaporation Effect Quantification

The PNNL data does not include evaporative effects and the data are not expected to have been significantly affected by evaporation. This is because of wetting the test chamber walls and, when the chamber is closed, humidifying it to >80% RH before collection of baseline test data and subsequent test execution.

PNNL did a humidity correction when the in-chamber humidity during a test was less than 100% RH. Humidity measurements were collected during each test, but these measurements are for information only because they are based on readings instruments that do not meet NQA-1 calibration requirements. However, the humidity sensors were procured with a factory calibration. Additionally, a performance check of the sensors toward the end of large-scale testing (and before the final humidity tests) indicated that the humidity sensors were providing reasonably accurate measurement of chamber humidity. By comparing test results at different humidity conditions, a humidity correction was estimated for the test results. PNNL-22415 corrected the measured values by the for information only (FIO) humidity measurement maximum correction factor of 1.5. Note the humidity meter was designated as FIO since it did not have a project recognized NQA-1 pedigree. These corrected values were then compared with the conservative correlation. The comparison showed that the conservative correlation is reasonably conservative and bounds the test data, even if it is corrected for humidity using the estimated maximum correction of 1.5. Less than 1% of the individual humidity corrected data points exceed the conservative correlation. The majority of the individual corrected data points that exceeded the conservative correlation correspond to extrapolated 100 foot chamber data.

This demonstrates that the conservative correlation has accounted for the potential biases (humidity and method bias) without actually quantifying them, i.e., no correction factors to the conservative correlation were found to be necessary. .

The effects of evaporation on the resulting aerosol can be quantified separately. No further testing to further quantify evaporation is required.

Appendix A: Acronym List

AED	Aerodynamic Equivalent Diameter
AFA	Anti-Foam Agent
ARF	Airborne Release Fraction
ASME	American Society for Mechanical Engineers
CRF	Cumulative Release Fraction
DNFSB	Defense Nuclear Facility Safety Board
DOE	Department of Energy
DSA	Documented Safety Analysis
DSD	Droplet Size Distribution
HDBK	Handbook
HEPA	High Efficiency Particulate Air
ICRP	International Commission on Radiological Protection
INL	Idaho National Laboratory
ISL	Information Systems Laboratory
MAR	Material at Risk
NNSA	National Nuclear Security Administration
NRC	Nuclear Regulatory Commission
ORP	Office of River Protection
PNNL	Pacific Northwest National Laboratory
PSD	Particle Size Distribution
RF	Respirable Fraction
RH	Relative Humidity
STD	Standard
STP	Sludge Treatment Plant
STR	Small Treated (Simulant)
WTP	Waste Treatment and Immobilization Plant

Appendix B: Issue Evolution, 2009 – 2015

B.1 Discussions with the DNFSB

In 2009, discussions were held between the staff of the Defense Nuclear Facilities Safety Board (DNFSB) and the staff of Waste Treatment and Immobilization Plant (WTP) at the Hanford site concerning the calculation of accident consequences resulting from pressurized spray leaks. Specifically, the following topics were addressed:

- Whether the breach configuration used in the spray leak analysis calculation was conservative;
- Whether the respirable fraction (RF) and airborne release fraction (ARF) used from DOE-HDBK-3010-94 adequately represents the droplet size distribution (DSD) produced by possible WTP process leak geometries;
- Whether evaporation from transported droplets could substantially change spray DSD, increasing the amount of respirable material release; and,
- Whether agglomerates formed by drying of droplets might de-agglomerate during transport, further increasing respirable material at the receptor location.

To shed light on these questions, DOE's Office of River Protection (ORP) sponsored an evaluation of DOE-HDBK-3010-94 (1994 handbook) with respect to RFs and ARFs for pressurized spray leaks. Preliminary results of that evaluation indicated that the Handbook's spray release model might not be conservative in establishing ARFs and RFs for WTP process leak geometries. DOE then requested that a review of this subject be written by recognized subject matter experts in the field.

B.2 Mishima-Foppe Technical Paper

The subject matter experts chosen to conduct the review were Jofu Mishima, one of the authors of DOE-HDBK-3010-94, and Terry Foppe, a widely-known and respected safety analyst. Mishima and Foppe delivered their results in the form of a white paper to DOE in January 2010, presented as Appendix D in this report. They concluded that while the Handbook's recommended 1E-4 ARFxRF value remains valid for the fully-enclosed glovebox studied in the Handbook, it could not be taken as a bounding value for liquid droplets of respirable size generated by sprays from metal piping and vessels such as may be expected at WTP. For this situation, Mishima and Foppe recommended a more conservative bounding value 20 times larger than the Handbook's multiple of ARF and RF. They stated, in addition, that (1) the existing database should be expanded, (2) experiments should be conducted, (3) more comprehensive analytical models should be developed and used, and (4) orifice plugging should be addressed.

B.3 Deputy Assistant Secretary for Environmental Management Memorandum

The Mishima-Foppe paper was included in a February 1, 2010 memorandum sent by Steven Krahn, Deputy Assistant Secretary for Environmental Management, to Andrew Lawrence, Director of the Office of Nuclear Safety, Quality Assurance, and Environment within the Office of Health, Safety and Security. The memorandum, entitled, "DOE Guidance for Performing Dose Consequence Analyses," presented as Appendix C in this report, summarized the shortcomings and application domain limitations of the 1994 handbook and concluded that revised DOE guidance was needed to reduce uncertainties in spray leak analyses. In response to this memorandum, ORP directed WTP to initiate a research program aimed at developing technically justified and reasonably conservative ARFs and RFs for spray leak scenarios from its process geometries.

B.4 Hanford Refinements to the Pressurized Spray Leak Model

Pending the outcome of laboratory research that would take several years to accomplish, DOE sought to improve in the near-term the methods used for analyzing spray leaks. Over the period March – August 2010, four DOE-sponsored papers were published. (References B-1, B-2, B-3, and B-4); three of these papers dealt with WTP, while the fourth dealt with the nearby Sludge Treatment Plant (STP) at Hanford. Taken together, these papers formed the basis of a near-term methodology that DOE believed could be used in lieu of DOE-HDBK-3010-94. This revised method of analyzing spray leaks provided an adequately conservative model for facility preliminary design and a reasonable basis for conservative control selection. The authors of these papers agreed that additional research was still needed to reduce uncertainties.

B.5 Independent Evaluation of Hanford Modified Spray Leak Model

Following receipt of the four papers discussed in B.4, DOE requested an independent evaluation of the revised methodologies, which was conducted by consultants W. Arcieri and M.A. Azarm of Information Systems Laboratories, Inc. (ISL). Specifically, DOE directed ISL to: (1) review for technical soundness the results of the revised WTP spray leak calculations; and, (2) report on Nuclear Regulatory Commission (NRC) requirements and guidance for analyzing spray releases in non-reactor nuclear facilities.

The requested report was released by DOE in January of 2011. (Reference B-5) The authors found weaknesses in the WTP analyses, particularly in the use of certain input data developed by Baker Engineering and Risk Consultants. The authors recommended that: (1) the methodology used in the SPRAY code, which is the starting point in the WTP analyses, needs to be revised, and; (2) DOE should consider updating this code to reflect more recent work. With respect to relevant NRC guidance which can help to better understand what may constitute the break characteristics of a spray accident, the report identified techniques in NUREG/CR-6410 (1998), *Nuclear Fuel Cycle Facility Accident Analysis Handbook*. (Reference B-6).

B.6 DOE Draft Paper and DNFSB Response

To summarize work to date, DOE released on March 27, 2011, a draft paper entitled "Technical Analysis of the Airborne Release Fraction and Respirable Fraction for Spray Release Accident Scenarios." (Reference B-7). The draft paper stated two main conclusions:

- The ARF and RF values used in DOE-HDBK-3010-94 for a spray leak accident was based on analysis of a release of small droplets of water-like solutions from a glass vessel rather than a pressurized spray release of liquids unlike water from metal vessels or piping. As a result, the handbook's ARF and RF values may not be conservative in all cases. Hence, a combined ARF and RF value appropriate for the conditions and fluids released in a specific spray leak accident scenario needs to be determined for accident analysis calculations, and,

- The current desired approach for arriving at a conservative combined ARF and RF value for spray leak accidents in the DOE complex is to use a methodology similar to the approach recently developed and used by WTP, adjusting the parameters as needed to account for differences in the fluids and facility conditions. In addition, a facility-specific evaporation effects model needs to be used in the dose calculations.

The paper argued that this approach addressed the DNFSB's concerns by incorporating the DSD and evaporation effects into a combined ARF and RF value. This action, the paper contended, satisfied the conservatism requirements of DOE-STD-3009-94 (Reference B-8).

On March 30, 2011, this draft paper was discussed with the DNFSB staff. The DNFSB formalized its technical position in a letter to DOE dated April 8, 2011. (Reference B-9). In its response dated June 3, 2011 (Reference B-10), DOE informed the DNFSB that it had directed WTP to engage PNNL in a test program to narrow the uncertainties under discussion.

B.7 PNNL Experiments and Results

The PNNL test plan (Reference B-11), issued in July of 2011, indicated that testing was needed to reduce the uncertainty caused by extrapolation of results reported in the technical literature. Two general goals were established for the research:

- Quantify the role of slurry particles in small breaches, where plugging by the particles may result in substantially reduced, or even negligible, RF formed by high-pressure sprays; and,

- Determine the aerosol DSD and total droplet volume from prototypic breaches and fluids, specifically including sprays from larger breaches and slurries.

The experiments were designed to provide empirical data for a range of orifice sizes and orientations. Five specific test objectives were identified:

1. Determine the size of circular and slot-shaped breaches that will plug and not form appreciable sprays with slurry simulants within an appropriate time period;

2. Determine the DSD of aerosol droplets and the total droplet volume concentration as a fraction of total spray volume, for a range of smaller breach sizes for circular and rectangular breaches, liquid and slurry simulants, and WTP process conditions;

3. Determine the DSD of aerosol droplets and the total droplet volume concentration, as a fraction of the total spray volume for a chemical slurry simulant, representative of a washed and leached process stream, to compare with the results from non-hazardous simulants;

4. Determine the DSD of aerosol droplets and the total droplet volume concentration, as a fraction of the total spray volume for a range of circular and rectangular breach sizes; and,

5. Determine the DSD of aerosol droplets and the total droplet concentrations, as a fraction of the total spray volume for a range of circular and rectangular breach sizes.

From September 2012 to August 2013, PNNL conducted experiments and issued five test reports and one revision to an earlier test report:

- September 2012: PNNL-21361 (WTP-RPT-219, Rev 0), *Small-Scale Spray Releases: Orifice Plugging Test Results*, September 2012, Pacific Northwest National Laboratory, Richland, WA;
- November 2012: PNNL-21361 (WTP-RPT-219, Rev 1), *Small-Scale Spray Releases: Orifice Plugging Test Results*, September 2012, Pacific Northwest National Laboratory, Richland, WA ;
- December 2012: PNNL-21333 (WTP-RPT-217, Rev 0), *Large Scale Spray Releases: Initial Aerosol Test Results*, December 2012, Pacific Northwest National Laboratory, Richland, WA;
- May 2013: PNNL-21367 (WTP-RPT-216, Rev 1), *Small-Scale Spray Releases: Initial Aerosol Test Results*, May 2013, Pacific Northwest National Laboratory, Richland, WA;

- August 2013: PNNL-22402 (WTP-RPT-222, Rev 0), *Small-Scale Spray Releases: Additional Aerosol Test Results*, August 2013, Pacific Northwest National Laboratory, Richland, WA; and,
- August 2013: PNNL-22415 (WTP-RPT-221, Rev 0), *Large Scale Spray Releases: Additional Aerosol Test Results*, August 2013, Pacific Northwest National Laboratory, Richland, WA.

In order to widely share the insights gained in the PNNL research, DOE conducted a complex-wide workshop in Oak Ridge, Tennessee, July 17-18, 2013.

B.8 Spray Leak Release Technical Report

On June 3, 2014, DOE directed that a technical report be prepared to collect all relevant information on spray leak efforts that has been assembled. The following topics were to be addressed:

- Spray leak analysis methodologies used at DOE sites and in the commercial nuclear and chemical industries (information collected but not included in this report);
- DOE-HDBK-3010-94 approach to spray leak ARFs and RFs;
- Spray leak characterization parameters;
- Factors affecting spray droplet size distributions;
- Factors affecting the measurement of spray DSD;
- Results of PNNL experiments;
- Recommended approaches to establishing a conservative value of ARFxRF,
- Recommended control selection; and
- Recommendations for additional testing and experimentation.

Work on this report commenced on June 2, 2014, and was completed in December 2018.

B.9 References

B-1. Larson, A.R., and Allen, B.T., "WTP Methodology for Spray Leak Scenarios," March 11, 2010.

B-2. McAllister, J.E., "Severity Level Calculations for the Pretreatment Facility Based on Updated MAR," May 7, 2010.

B-3. Crowe, R.D., "Sludge Treatment Project (STP) Methodology for Spray Leak Scenarios," June 29, 2010.

B-4. Larson, A.R. and Allen, B.T., revision of Reference B-2, August 11, 2010.

B-5. W. Arcieri and M. A. Azarm, "WTP Methodology for Spray Leak Scenarios," Information Systems Laboratories, Inc., Rockville, MD, 2011.

B-6. U.S. Nuclear Regulatory Commission, NUREG/CR-6410, *Nuclear Fuel Cycle Facility Accident Analysis Handbook*, March 1998.

B-7. DOE Office of Health, Safety and Security, "Technical Analysis of the Airborne Release Fraction and Respirable Fraction for Spray Release Accident Scenarios (Draft)," March 27, 2011.

B-8. DOE-STD-3009-94, *Preparation Guide for U.S. Department of Energy Nonreactor Nuclear Facility Documented Safety Analyses* (1994).

B-9. Winokur to Triay, April 5, 2011, available on the DNFSB website.

B-10. Triay to Winokur, June 3, 2011, available on the DNFSB website.

B-11. Test Plan TP-WTPSP-031.

Appendix C: Krahn-Lawrence Memorandum C

Department of Energy
Washington, DC 20585

FEB 0 1 2010

MEMORANDUM FOR ANDREW LAWRENCE
DIRECTOR
OFFICE OF NUCLEAR SAFETY, QUALITY ASSURANCE AND ENVIRONMENT
OFFICE OF HEALTH, SAFETY AND SECURITY

FROM: DR. STEVEN L. KRAHN
DEPUTY ASSISTANT SECRETARY FOR SAFETY AND SECURITY PROGRAM
ENVIRONMENTAL MANAGEMENT

SUBJECT: Department of Energy Guidance for Performing Dose Consequence Analyses

DOE-STD-3009, *Preparation Guide for U.S. Department of Energy Nonreactor Nuclear Facility Safety Analysis* provides guidance for estimating the radiological consequences of material released from nuclear facilities under accident conditions. Additional guidance for performing dose consequence analyses is provided in other Department of Energy (DOE) documents, such as DOE-HDBK-3010, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities.* Some of these guidance documents are directly referenced in the DOE-STD-3009 (such as DOE-HDBK-3010) and some are not. This DOE guidance, if properly utilized, is intended to ensure an estimated dose consequence that is conservative. However, recent issues raised during the technical review of unmitigated dose consequence results for the Waste Treatment Plant (WTP) at the Hanford Site have identified that this may not currently be the case. As a result of these reviews, two parameters recommended for use by the DOE guidance are being questioned. These two parameters are described briefly below.

Airborne Release Fraction for a Pressurized Spray Leak:

DOE-HDBK-3010-1994 provides an airborne release fraction (ARF) and respirable fractions (RF) to be used for a spray leak. Handbook Section 3.2.2.3.1, "Venting Below the Liquid Level," provides the following bounding ARFxRF recommendation, along with limited additional guidance related to the geometry of the leak and evaporation:

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"... a conservative assumption would be the pressurized release of the liquid via a very fine hole as occurs in a commercial spray nozzle... It is not anticipated that drops formed from breaches, cracks, leaks would generate finer drop size distribution. 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.

Other recent investigations ... 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."

Given this guidance, and the normally assumed maximum respirable size of 10µm, the spray leak has been modeled as a 0.128-inch orifice with an ARFxRF of 1E-4. Consideration of evaporation and a smaller or slit-type leak are not presently encouraged by the DOE-HDBK-3010 guidance. During the review of the WTP, the Defense Nuclear Facility Safety Board (DNFSB) staff provided comments on the unmitigated spray leak analysis that have called into question the ARF value in DOE-HDBK-3010. Specifically, the staff commented that the DOE-HDBK-3010 ARF may not conservatively represent the droplet size distribution produced by a slit or crack shaped spray leak; also, evaporation from the droplets, as they settle, could change the size distribution of the spray, increasing the amount of respirable material available for release.

Jofu Mishima, one of the principal authors of DOE-HDBK-3010-1994, has also observed that the Handbook may not be conservative in establishing an ARFxRF for a spray leak (see attached white paper). Limited experimental data relevant to liquid waste fluids (e.g., slurries, high-salt content solutions, mixtures, etc.) has been identified upon which a technical basis can be formulated. There is experimental data from industry with respect to spray nozzle droplet distributions and there are textbook correlations for some parameters but this information may not be directly applicable for the range of fluids of interest in high-level waste applications (for example). Parameters to be explored include leak size and shape, dissolved and suspended solids, plugging, surface tension, viscosity, density, and pressure.

Deposition Velocity:

In June 2004, the Department issued MACCS2 Code Guidance in response to DNFSB Recommendation 2002-1, "Software Quality Assurance." This guidance recommended using a deposition velocity of 1 cm/s to estimate exposures for unmitigated releases from DOE facilities. Deposition velocity is a simplified factor for representing plume depletion and is affected by the size distribution of particles released, wind speed, and the roughness of the surface upon which the plume is travelling.

At Hanford, the WTP incorporated a deposition velocity of 1 cm/s into MACCS2 dispersion calculations, consistent with this DOE MACCS2 code use guidance. The DNFSB staff raised a concern that this value was not conservative for unmitigated releases at the Hanford site, since the value did not bound measured values associated with a known and documented 1985 Hanford Tank Farm radiological release incident. The staff also referenced modeling assumptions cited in NUREG/CR 3332/ORNL-5968, and suggested that a value of 0.1 cm/s would be more appropriate.

Concluding Thoughts:

1) The uncertainty regarding these two parameters has potential impact across the complex. Pending resolution, guidance is needed regarding these parameters and what actions, if any, should be taken in the interim.
2) DOE needs to establish the appropriate value(s) for these two parameters and issue revised guidance. This will require the development of a suitable technical basis, and will likely require some research and development.
3) More broadly, it is essential that the proper implementation of DOE guidance produce results that are predictable and reasonably conservative. As such, it would appear that an ongoing effort to update and/or confirm the technical basis of its guidance related to nuclear safety is needed.
4) In addition, there is need to establish a mechanism for assessing and dispositioning future challenges to this guidance using a controlled, complex-wide approach as opposed to facility-by-facility adjustments or corrections.

If you have any questions, please contact me at (202) 586-5151.

Attachment

Appendix D: Mishima-Foppe Paper

Review of the DOE-HDBK-3010-1994 Airborne Release Fractions and Respirable Fractions for Spray Releases from Hanford Waste Solutions

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January 20, 2010

1.0 INTRODUCTION

The U.S. Department of Energy (DOE) Handbook, DOE-HDBK-3010-1994, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities* (DOE 1994), provides guidance for modeling spray leak scenarios involving pressurized liquid releases and recommends bounding airborne release fractions (ARF) and respirable fractions (RF) to be used in accident consequence analysis. In response to review comments on the application of Handbook methodology for the Hanford Waste Treatment and Immobilization Plant (WTP) Preliminary Safety Analysis Report, the DOE Office of River Protection (ORP), sponsored a review of the technical basis of the Handbook guidance by Mr. Jofu Mishima, one of the principal authors of DOE-HDBK-3010, and Mr. Terry Foppe, support service subcontractor to ORP. The review was to support WTP project modeling of potential spray releases to consider whether a change to the previous approach was needed when accounting for leakage from pipes carrying pumped viscous waste slurries at pressures up to several hundred psig.

In December 2009, preliminary conclusions were provided to ORP that the Handbook spray release model may not be conservative in establishing the [ARF][RF] value for the WTP application. The purpose of this paper is to document the review findings, to provide recommendations regarding path forward for the WTP project, and to consider implications of potential revision to the DOE-HDBK-3010.

2.0 DOE-HDBK-3010 BOUNDING ARF/RF RECOMMENDATION

The Handbook Section 3.2.2.3.1, "Venting Below the Liquid Level", provides the following bounding ARF and RF recommendation and additional guidance related to the geometry of the leak and evaporation *{note: includes minor editorial changes made for clarity}*:

> "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 *{water drops from}* some commercial spray nozzles as a function of orifice diameter and upstream pressure were shown *{in a document}* by Mishima, Schwendiman and Ayer (October 1978). The size distribution of the liquid drops *{becomes finer (the fraction of small droplets increases)* ~~decreases~~ with *decreasing}* 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.

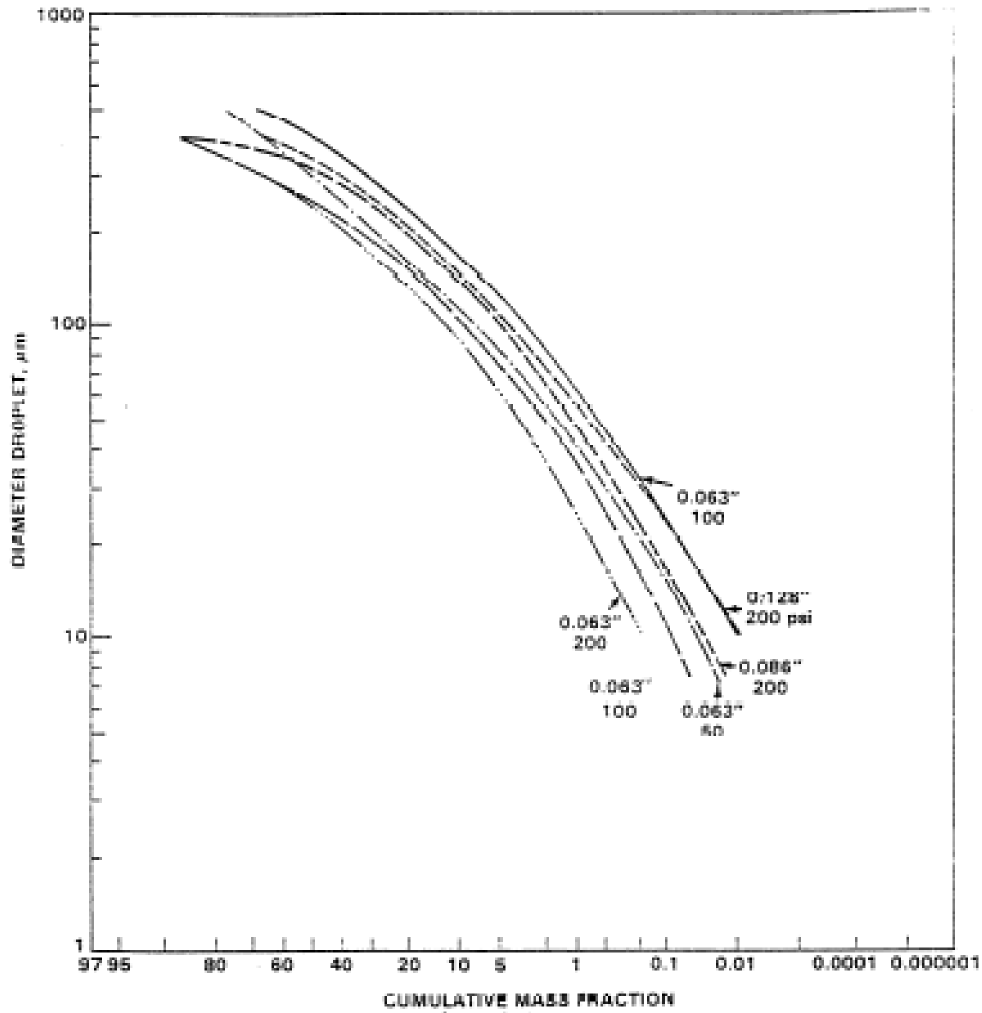

Figure 3-4. Mass Fraction vs. Droplet Diameters for Sprays as a Function of Orifice Diameter and Upstream Pressure (Mishima, Schwendiman, and Ayer October 1978)

{Note 1: The x-axis is labeled "Cumulative Mass Fraction". As shown on the scale, the value increases to the left to a maximum of 97.95. These values are in units of percent, i.e., the maximum value is 97.95% of the mass. For example, the 200 psig and 0.128" curve shows that the cumulative mass fraction for particles less than 10 um is 0.01 on the x-axis, which is 0.01% or 1E-4 of the mass.
Note 2: The liquid drop size distribution, of those plotted here, is considered the coarsest distribution since it has the smallest fraction of droplets $d_{AED} \leq 10$-um.}

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 Aerodynamic Equivalent Diameters [AED] 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."

To summarize the above, the Handbook [ARF][RF] recommendation is based on limited data from commercial hollow cone spray nozzles with orifice diameters of 0.063 inch, 0.086 inch, and 0.128 at three pressures (i.e., 50, 100, and 200 psig). The Handbook selects the respirable release fraction from the coarsest distribution generated by these commercial spray nozzles (i.e., 0.128 inch and 200 psi) as the bounding [ARF][RF].

3.0 REVIEW AND DISCUSSIONS

The situations and events considered for this type of phenomenon has expanded greatly over the 15 years since the DOE-HDBK-3010 was issued. Leaks from metal piping and vessels holding liquids of significantly different properties (e.g., slurries, high-salt content solution, mixtures, etc.) are now analyzed. Several questions regarding the validity of the DOE-HDBK-3010 spray release methodology for such waste solutions have resulted in a critical examination of the basis for the recommended bounding value.

It is apparent from examination of Figure 3-4 on page 3-20 of the handbook that the bounding value of 1E-4 [ARF][RF] does not bound all potential sprays from nozzles and therefore may not be bounding for metal vessels and piping. As illustrated in the figure and stated in the Handbook discussion, the size distribution of a spray formed by forcing liquid through a pressure nozzle/orifice becomes finer with decreasing size of the orifice and increasing pressure. The Handbook data and discussions were adopted from previous accident consequence studies for nonreactor nuclear facilities as discussed below.

The following information from the *Chemical Engineers' Handbook* (Perry 1941) was considered for the original evaluations:

- Pg 1982 – "Other Methods of Comminution"
- Pg 1983 – "Spray Nozzles"
- Pg 1985 – "Pressure Nozzles" - "Hollow cone Nozzles" This is the most common type of pressure nozzle in use. Fluid is passed into a whirl chamber through tangential passages or through fixed spiral so that it acquires a rapid rotation. The orifice is placed on the axis of the whirl chamber, and the fluid exits in the form of a hollow, conical sheet which then breaks up into drops.
- Pg 1988 Table 1, "Discharge Rates and Included Angle of Spray of Typical Pressure Nozzles" (reproduced at the end of this report)
  The data listed demonstrate that the discharge rate and included angle (the area covered by the spray which increases with distance from the nozzle to some maximum) increase with upstream pressure and orifice diameter. Also note that the discharge rate and

included angle of the various types of pressure nozzle vary with the hollow cone nozzle having the largest discharge rate of the three nozzles listed.

A graph of the data first appeared as Figure 6 (same as the Handbook Figure 3-4) in the evaluation of a mixed oxide fuel fabrication facility, *Increment of Analysis – An Estimate of Airborne Release of Plutonium from Babcock and Wilcox Plant as a Result of Severe Wind Hazard and Earthquake* (Mishima, Schwendiman & Ayer 1978). The evaluation was performed by the Pacific Northwest (National) Laboratory (PNL) for the U.S. Nuclear Regulatory Commission (NRC). It was used to confirm a bounding release estimate from seismic shaking of a glove-box with liquids in fragile containers and vessels such as glass based on the "fog limit" and a small enclosure. Relevant excerpts include *{note: includes minor editorial changes made for clarity}*:

> “The volume of the average enclosure is assumed to be (3-ft X 3-ft X 8-ft = 72-ft^3) 2-m^3." *{pg 30}*
>
> “For liquids held in a fragile container (those that could be ruptured by the impact of debris), it is assumed the entire volume of the enclosure is filled with a mass of respirable particles equivalent to the maximum mass formed in nature – fog, 10-mg/m^3 – and **size distribution of a coarse spray** *{bold emphasis added}*. Figure 6 *{pg 3-21}* is a plot of the cumulative mass fraction versus droplet diameters for hollow cone nozzles of orifice diameter ranging from 0.063- to 0.178-inches [1.6- to 3.25-mm, (1,600- to 3,250-µm)] at various liquid upstream pressures. The orifice diameter appears small and the pressures high for the conditions envisioned for most situations resulting in the break-up of fragile containers. The distribution of the coarsest spray (0.128-inches diameter at 200 psig) indicates the mass of droplets 100-µm or less is 50 times the mass of droplets 10-µm and less. Particles 100-µm could be carried beyond the remnants of the structure from wind hazards scenario and it was assumed that the airborne mass concentration of the particle d_{AED} and less in the enclosure was 500-mg/m^3." *{pg 33}*

The mass fractions for the various drop-size bins in Perry’s *Chemical Engineers' Handbook* are not cited and the mass of the various numbers of drops in each size fraction must be converted using the volume of the drops and the density of water (1-g/cm^3). The mass fractions upon which the graph is based are cited in another PNL study, *Source Term and Radiation DOE Estimates for Postulated Damage to the 102 Building at the General Electric Vallecitos Nuclear Center* (Mishima, Schwendiman & Ayer 1979). The data are presented in Table A.1 of the Appendix A, “Discussion of Factors Used to Estimate Potential Airborne Release from Seismic Activity at the Vallecitos Nuclear Center” on page A.4 (reproduced at the end of this report). This data came from Table 4.1 “Drop Size of a Hollow Cone Nozzle at Various Pressures" from the 1943 printing of Perry's *Chemical Engineers' Handbook* in the section "Spray Nozzles authored by H.G. Houghton (Houghton 1943).

In the discussion in the Appendix A, under "AIRBORNE MASS CONCENTRATIONS WITHIN ENCLOSED SPACES, Liquids" *{note: includes minor editorial changes made for clarity}*:

> {*Pg A.1*} – "Oak Ridge National Laboratory has been able to demonstrate … that the meta-stable aerosol concentration of 10-mg/m^3 (approximately equivalent to fog) and has size distribution shown in Figure A.2 *{pg A.3, 'Particle Size Distribution of a Stable Aerosol that has Encountered Several Changes in Direction in a Pipeline'.}* … {*Pg A.3*} Table A.1 shows the cumulative masses associated with droplets less than various size ranges for three orifice diameters ranging from 0.063-in. (1.6-mm) to 0.129-in. (3.3-mm) at various pressures. These size distributions become coarser with increasing orifice diameters and decreasing pressure. … These conditions appear to greatly exceed the pressures and are much finer than openings found for the breakage of glass equipment. Thus, an assumption of 10^{-4} of the inventory is conservative."

The above excerpts from the previous PNL evaluations of mixed oxide fabrication facilities in the late 1970s justified the bounding [ARF][RF] of 1E-4 for releases from liquids in glass equipment using two approaches, a "fog limit" and perspectives from hollow cone nozzles. The value is an estimate of the stable (post interaction and deposition) liquid aerosol in a glove-box. The value has been incorrectly labeled as a "spray release" and has been used in similar evaluations of NRC and DOE nonreactor nuclear weapons facilities since that time until the present day. Additional experimental studies have been performed at Pacific Northwest (National) Laboratory and are reported in Sutter (1983) and Ballinger, Sutter, and Hodgson. (1986). The information and data were compiled in NUREG-1320 and its update NUREG/CR-6410 (1998). This value was carried over to the guidance for investigators for the DOE Safety Survey in 1992 for engineering analysis of the potential releases from DOE Weapons Complex facilities. The Safety Survey guidance was shortly thereafter formalized into DOE-HDBK-3010 which included the commercial spray data for water (Figure 3-4) and recommended the 1E-4 as a bounding value for spray releases. The reliance on the bounding values cited for these 1970's studies without careful examination of the basis led to the selection of the bounding value for the Handbook and its long-time use.

4.0 CONCLUSIONS AND RECOMMENDATIONS

Although the value cited, 1E-4 [ARF][RF], was appropriate for the conditions postulated at the time (i.e., airborne release of material inside an enclosure due to seismic shaking or toppling of glass equipment containing water-like solutions), the value cited, fraction $\leq d_{AED}$ 10-µm AED, is not a bounding value for airborne releases from a spray of liquids with properties significantly different than water (e.g., neutralized and processed High-Level Waste). The reliance on the bounding values cited without careful examination of the basis led to the selection of the bounding value for the Handbook and its long-time use.

Summary of key review findings.

- The Handbook recommended bounding [ARF][RF] of 1E-4 of respirable droplets (≤10 µm AED) is based on "the coarsest distribution generated by commercial spray nozzles shown in Figure 3-4." While the 1E-4 value corresponds to discharge from a nozzle of 0.128" diameter and 200 psig, no specific recommendations regarding leak size and pressure was intended in the Handbook.
- The [ARF][RF] of 1E-4 of respirable droplets was originally selected for the PNL/NRC evaluation of a seismic scenario in a specific facility, Babcock and Wilcox mixed oxide fuel fabrication, and was incorrectly labeled as a "spray release". The value is an estimate of the stable (post interaction and deposition) liquid aerosol in a glove-box based on 10 mg/m^3 "fog" limit due to breakage of glass/fragile equipment. Droplet evaporation is limited in such an environment.
    - It was considered conservative by comparing to the commercial spray nozzle data for largest diameter coarse sprays which showed that a 1E-4 respirable value would be bounding.
- The 1E-4 value was carried over to the guidance for investigators for the DOE Safety Survey in 1992 for engineering analysis of the potential releases from DOE Weapons Complex nonreactor nuclear facilities.
- The Safety Survey guidance was shortly thereafter formalized into DOE-HDBK-3010 that included the commercial spray data for water (Figure 3-4) and recommended the 1E-4 as a bounding value for spray releases.
- Figure 3-4 shows that the size distribution of a spray formed by forcing liquid through a pressure nozzle/orifice becomes finer with decreasing size of the orifice and increasing pressure.
- The recommended 1E-4 value in DOE-HDBK-3010 remains valid for the studied glove-box, but is ***not*** a bounding value for liquid droplets of respirable size generated by sprays from metal piping and vessels as a function of opening size, configuration, and upstream pressure, with liquid properties that may be significantly different than water.

Resolution of the problem is made difficult by the fact that there are at least four types of liquids that must be addressed;

- Supernatant liquids that over-lie un-dissolved solids – these liquids may range from water-like fluids to high-dissolved solid solutions;
- High-dissolved solids solutions – the dissolved materials may cover a wide range of compounds but are primarily caustic/neutral sodium salts that may also contain organic compounds used to treat the waste at various times;
- Low solids (≤7-wt%) slurries; and,
- High solids (up to 20-wt%) slurries.

There is a lack of data available for the relevant physical properties (densities, viscosities, surface tension, etc.) of the liquids that are necessary to use in analytical models.

Some potential remedial measures for the WTP Project and for consideration of potential revision to the DOE-HDBK-3010 are:

Establish a Data Base of Relevant Physical Properties of the Various Fluids Anticipated for the Tank Farms and WTP. Some data may currently exist for properties of the fluids anticipated and should be compiled, technically supported, and documented.

Analytical Models - Rather than relying on commercial spray nozzle data using water, consider application of empirical correlations from the literature, using appropriately conservative assumptions for input parameters specific to the waste solution physical properties and applicable ranges of the correlations, to calculate the bounding $d_{AED} \leq 10$-µm for spray releases. Although each method is not fully supported and simplifications need to be made to make the engineering calculations tractable, better experimental data for these types of event and materials is not currently available.

- An example of an empirical correlation is one similar to the SPRAY code developed for the Hanford Tank Farms in *A Model for Predicting Respirable Releases from Pressurized Leaks* (Hey and Leach. 1994), and its current modifications using a Microsoft Excel® spreadsheet. Other correlations may also be suitable. Prior to use, this methodology should be critically reviewed to assure that the selection of input parameters results in an overall bounding value, e.g., one approach is to consider using the 90th percentile-type value for up to 3 parameters and technically based average values for the remainder.
- It is acknowledged that the recent concern of evaporation of larger droplet sizes to respirable sizes can be addressed using these empirical models, however, as stated in the Handbook discussion, there is considerable uncertainty as to the value to assign the critical factor (Q, a drop size fitting parameter), which is also true for many other input parameters. Caution is urged to select appropriate input values such that the overall result is not unrealistically high or even physically not plausible, which would significantly over-estimate the release potential. Grossly conservative assumptions (e.g., 5% RH at 30° C) may skew the results and yield results that are misleading. It should be borne in mind that in ventilated areas, the air is conditioned to a comfort level for the personnel (70° F, 50% RH) and liquids sprayed into this environment would rapidly saturate. Liquids sprayed into a confined volume (even with a low ventilation rate) rapidly saturate the air from the liquid evaporated from the drops, liquids impacted on surfaces, and the pool formed by rainout. Only for liquids sprayed into the ambient atmosphere would evaporation be a significant concern for the entire release duration.

Potential Plugging of Breaks by Solids - Consider the plugging potential of the waste slurry, e.g., base the bounding $d_{AED} \leq 10$-µm for spray [ARF][RF] on the ratio of the largest particles and the minimum dimension (i.e., orifice diameter or crack width) with the expectation that if the ratio is >1, the leak will plug. Use the [ARF][RF] value for the orifice diameter that exceeds the size of the largest particles.

Experimental Studies - Perform experimental studies to determine the discharge rates droplet size distributions of the various fluids or their surrogates for the range and types of breaks anticipated. . Such experimental studies would face some severe difficulties such as:

- Providing fluids to use as surrogates without knowledge of the range of chemical composition and their effect on the physical properties to be defined; and,
- Determining the drop sizes of sprays generated – liquid drops splatter when impacting hard surfaces and potentially large number of drops in any location during any time.

Recommendations:

- Prepared a documented estimate/methodology for the maximum mass fraction of droplets in the respirable size range ($d_{AED} \leq 10$-µm) to appropriately bound Hanford waste solution spray releases.
- Consider publishing a "Change Notice" to DOE-STD-3010 to provide additional clarifications on the applicability of the current recommendations and alternative approaches to establish a bounding estimate.
    - Consider increasing from the current value of 1E-4 to 2E-3 (an increase by a factor of 20) based on the depressurization of containment via a failure above the liquid level or overall containment failure with the highest [ARF][RF] for a release from aqueous solutions (< 1.2 g/cm^3) from up to 500 psig (DOE-HDBK-3010, page 3-3). This is believed to be bounding, if not overconservative for many situations in the DOE Complex, but may not be appropriate as bounding for some unique situations since the ≤10-µm fraction for spray increases with decreasing orifice diameter and increasing upstream pressure.
    - As an alternative to a single fixed value, consider establishing a more general model through a complex-wide consensus process.

5. REFERENCES

Ballenger, Sutter & Hodgson 1986	Ballinger, M.Y., S.L. Sutter, and W.H. Hodgson. 1986. *New Data for Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air*, NUREG/CR- 4779 (PNL-6065), Pacific Northwest (National) Laboratory, Richland, WA.
DOE 1994	DOE Handbook *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities*, 1994, DOE-HDBK-3010-94, Change Notice 1, March 2000, U.S. Department of Energy, Office of Environment, Safety and Health, Washington, DC.

Hey and Leach 1994	B.E. Hey and D.S. Leach. 1994. *A Model for Predicting Respirable Releases from Pressurized Leaks*, WHC-SD-GN-SWD-20007, Westinghouse Hanford Company, Richland, WA.
Houghton 1943	Houghton, H.G. 1943. "*Spray Nozzles*" in Chemical Engineers' Handbook, 2nd Edition, (later printing than the 3rd Impression by J.H. Perry, Editor, cited below), McGraw-Hill, New York, NY.
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Sutter 1983	Sutter, S.L. August 1983. *Aerosols Generated by Release of Pressurized Powders and Solutions in Static Air*, NUREG/CR-3093 (PNL-4566), Pacific Northwest (National) Laboratory, Richland, WA.

Table 1 Discharge Rates and Included Angle of Spray of Typical Pressure Nozzles
(Perry's pg 1988)

Type	Orifice Diameter, in.	Discharge, gal/min, and included angle of spray							
		10 psig		25 psig		50 psig		100 psig	
		Discharge	Angle	Discharge	Angle	Discharge	Angle	Discharge	Angle
Hollow cone	0.046	---	---	0.10	65	0.135	68	0.183	25
	0.140	0.535	82	0.81	88	1.1	90	1.50	93
	0.218	1.25	83	1.88	86	2.55	89	3.45	92
	0.375	7.2	62	11.8	70	16.5	70	---	----
Solid cone	0.047	---	---	0.167	65	0.235	70	---	---
	0.188	1.60	55	2.46	58	3.43	60	4.78	60
	0.250	3.35	65	5.40	70	7.50	70	10.4	75
	0.500	17.5	86	27.5	84	38.7	87	---	---
Fan	0.031	0.085	40	0.132	90	0.182	110	0.252	110
	0.093	0.70	70	1.12	76	1.57	80	2.25	80
	0.187	2.25	50	3.70	59	5.35	65	2.70	65
	0.375	9.50	66	15.40	74	22.10	75	30.75	75

Table A.1 Drop Size Distribution of 3 Hollow Cone Nozzles at Various Pressures
(Pg A.4 Mishima, Schwendiman, and Ayers 1979)

Drop Size, µm)[a]	Percent Drops in Size Fraction											
	0.063-in. (1.6-mm)						**0.086-in. (-mm)**				**0.128-in. (3.3-mm)**	
	50-psig		**100-psig**		**200-psig**		**100-psig**		**200-psig**		**200-psig**	
	Vol%	**Wt%**[b]	**Vol%**	**Wt%**[b]	**Vol%**	**Wt%**[b]	**Vol%]**	**Wt%**[b]	**Vol%**	**Wt%**[b]	**Vol%**	**Wt%**[b]
10	0.038	0.038	0.079	0.08	0.17	0.2	0.01	0.01	0.03	0.03	0.01	0.01
25	0.31	0.35	0.44	0.5	0.9	1.1	0.09	0.1	0.24	0.3	0.12	0.1
50	2.0	2.4	2.2	2.7	3.2	4.3	0.5	0.6	1.3	1.6	0.73	0.8
100	5.0	7.4	6.0	8.7	7.0	11.3	2.6	3.2	3.4	5.0	3.5	4.3
150	9.1	16.5	10.4	19.2	11.8	23.1	4.6	7.8	6.1	11.1	6.5	10.8
200	15.2	31.7	18.3	37.5	21.5	44.6	7,19	14.9	9.6	20.7	11.3	22.1
300	21.7	53.4	24.5	82.0	29.9	74.5	13.5	28.4	21.4	42.6	21.1	43.2
400	12.8	66.2	25.5	87.5	25.5	100	25.3	53.9	44.9	87.5	24.6	67.7
500	12.5	78.7	12.5	100	---	---	24.8	78.6	12.6	100	32.2	100
600	21.5	100	---	---	---	---	21.4	100	---	---	---	---

[a] The Test fluid is water with a density of 1-g/cm^3 ∴ $d_G = d_{AED.}$
[b] Cumulative fraction associated with drops ≤ than the stated size.

Appendix E: Proposed Revision to DOE-HDBK-3010-94 (R2006) Section 3.2.2.3.1, "Venting Below the Liquid Level", Regarding Spray Leak Releases

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/ducts (generating a mist) to drips from very slow leaks to large jets of liquids that may gush from large holes.

MAR can be defined in two ways. One method is by the total amount of material-of-concern in the vessel involved or the amount of the material-of-concern that is passed through the pipe/duct for some specific duration (e.g., up to 8-hr, or until the vessel empties) during the period of the event. For this MAR definition, the DR is the fraction of the MAR released through the breach. The DR needs to be defined consistent with the MAR definition, (e.g., DR is 1.0 for a spray release if the MAR is alternately defined as the total amount leaked from the crack over the accident duration, not to exceed the liquid volume available for discharge).

Sections A and B are based on two different spray release experimental data sets. Section A, Leaks from Breakage of Glass-Like Equipment, uses spray data which has been determined to bound aerosol generation from water-like releases from breakage of glass in a closed environment such as a glovebox. Section B, Spray Leaks from Pressurized Process Pipes, is based on experiments that are applicable for sprays generated from piping and vessels as a function of orifice size, pipe configuration, and upstream pressure.

A. Leaks from Breakage of Glass-like Equipment For the purposes of airborne suspension for some aerosol generating releases, a conservative assumption is that it is bounded by the pressurized release of the liquid via a very fine hole as occurs in a commercial spray nozzle, or by a "fog limit" as described below. This section is only applicable to releases from breakage of glass equipment (such as ion exchange column, jars, beakers), or similar fragile equipment, inside a small enclosure such as a glovebox containing liquid that may or may not be pressurized. The aerosol generation is primarily due to the initial breakage of the primary glass containment and release as a spray if it is pressurized, or it is from the free-fall spill of the liquid inside glovebox (i.e., a "splash and splatter" type of stress). The original basis for the bounding estimate of release for this situation is discussed below, and it is intended to include both the spray and spill contributions to aerosol generation.

For an initial estimate to bound the respirable liquid release from glass breakage, 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 droplet size distribution of the liquid drops becomes finer (i.e., the fraction of small droplets increases) with decreasing orifice diameter and increasing upstream pressure. It is not anticipated that drops formed from breaches, cracks, of other leak paths 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 releases as described in this section.

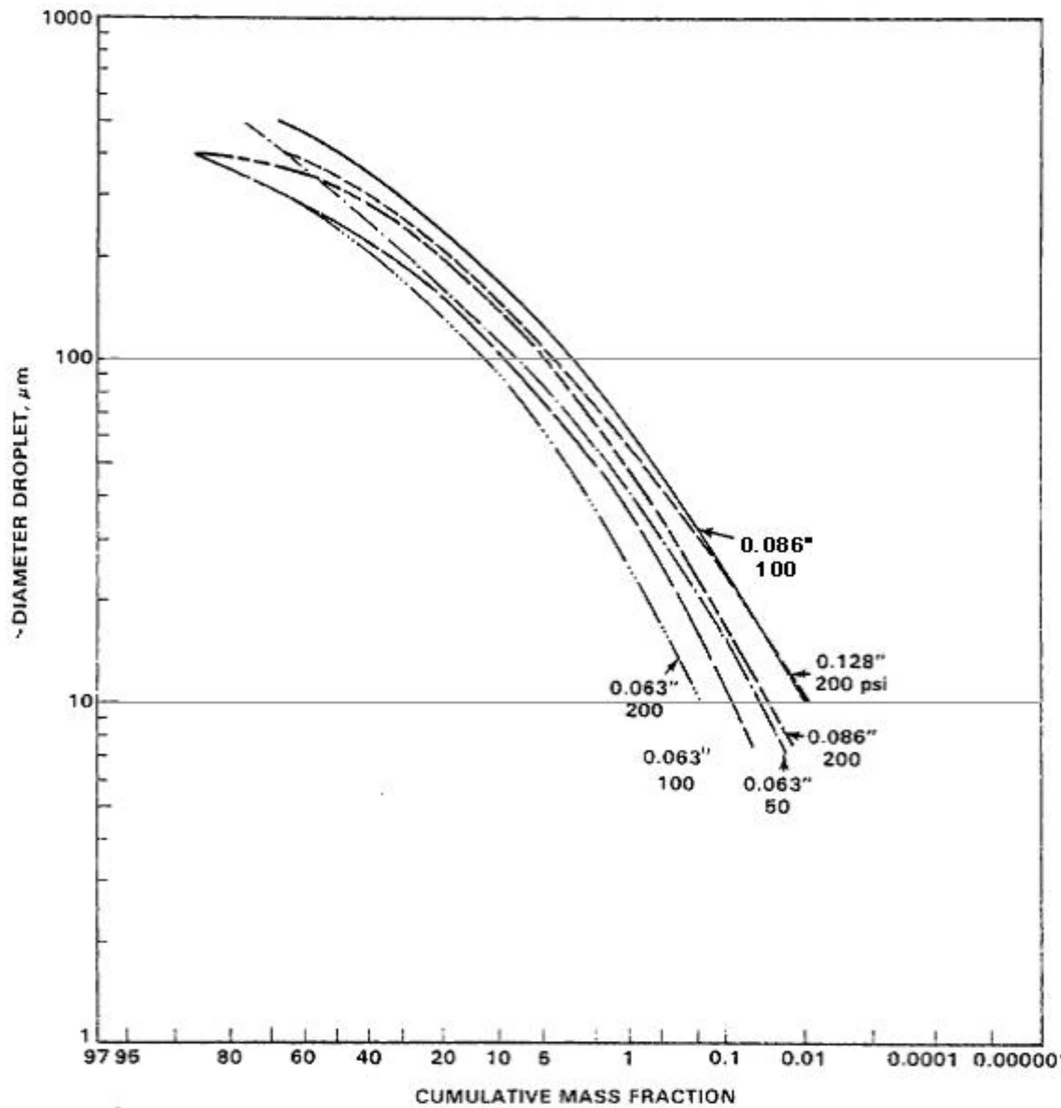

Figure 3-4. Mass Fraction vs. Droplet Diameters for Sprays as a Function of Orifice Diameter and Upstream Pressure (Modified from Mishima, Schwendiman, and Ayer October 1978)

Note 1: The x-axis is labeled "Cumulative Mass Fraction". As shown on the scale, the value increases to the left to a maximum of 97. These values are in

units of percent (i.e., the maximum value is 97% of the mass). For example, the 200 psi and 0.128" curve shows that the cumulative mass fraction for liquid drops less than 10 µm is 0.01 on the x-axis, which is 0.01% or 1E-4 of the mass.
Note 2: The liquid drop size distribution, of those plotted here, is considered the coarsest distribution since it has the smallest fraction of droplets $d_{AED} \leq 10$-µm.
Note 3: In HDBK-3010-94, the right-most curve (solid line) was incorrectly labeled. This revision corrects the Figure per the original data reproduced in Mishima, et al., 1979

It is apparent from examination of Figure 3-4, that the bounding value of 1E-4 ARFxRF from the coarsest spray distribution does not bound all potential sprays from nozzles, and therefore may not be bounding for all spray release scenarios, such as from steel piping and vessels. As illustrated in Figure 3-4, the curves show that for the same orifice size, the respirable mass fraction increases as the pressure increases, and for the same upstream pressure, the cumulative mass fraction increases with decreasing orifice size. As stated earlier, the droplet size distribution of a spray formed by forcing an incompressible liquid through a nozzle/orifice becomes finer with decreasing size of the nozzle/orifice and/or increasing upstream pressure.

Figure 3-4 and above discussions were adopted from previous accident consequence studies for nonreactor nuclear facilities. A graph of the data which led to Figure 3-4 first appeared as Figure 6 in the consequence evaluation of a mixed oxide fuel fabrication facility, *Increment of Analysis – An Estimate of Airborne Release of Plutonium from Babcock and Wilcox Plant as a Result of Severe Wind Hazard and Earthquake* (Mishima, Schwendiman and Ayer, 1978). The original data for the Handbook figure were reproduced in Table A.1, *Drop Size Distribution of 3 Hollow Cone Nozzles at Various Pressures* (Houghton 1943), of *Source Term and Radiation Dose Estimates for Postulated Damage to the 102 Building at the General Electric Vallecitos Nuclear Center* (Mishima, et al., 1979). The evaluations were performed by the Pacific Northwest (National) Laboratory (PNNL) for the Nuclear Regulatory Commission (NRC).

The aforementioned reports justified a bounding estimate for releases from liquids in glass equipment using two approaches, a "fog limit" and perspectives from hollow cone nozzles. The 1E-4 ARFxRF from the coarsest distribution (i.e., 0.128" diameter, 200 psig) was used to confirm a bounding release estimate from seismic shaking of a glove-box with liquids in fragile containers and vessels, such as glass, calculated using a 10 mg/m^3 "fog limit" and volume of a small enclosure. The value is an estimate of the stable post-interaction and deposition liquid aerosol in a glovebox. It was considered conservative by comparing the release estimate using the "fog limit" to the commercial spray nozzle data for largest diameter coarse sprays, due to glass breakage, which showed that a 1E-4 respirable value would be bounding for this situation and includes both spray and spill contributions.

Therefore, the conservative 1E-4 ARFxRF is valid for water-like sprays in a closed environment such as a glovebox, but is not a representative bounding value for liquid droplets of respirable size generated by sprays from metal piping and vessels as a function of orifice size, pipe configuration, and upstream pressure. Moreover, it is not representative for liquid properties that may be significantly different than water (e.g., slurries, neutralized and processed liquid waste).

For other spray release situations not involving glass breakage in a small enclosure, the desired approach for arriving at a conservative combined ARF/RF value for spray leak accidents in the DOE complex is to use a methodology similar to the approach discussed in the next section that is based on recent experimental testing, adjusting the parameters, as needed, to account for differences in the fluid characteristics and in-facility conditions.

B. Spray Leaks from Pressurized Process Pipes. A pressurized spray leak testing program was conducted in 2012-2013 by the Pacific Northwest National Laboratory (PNNL) (see PNNL-21361, PNNL-21333, PNNL-21367, PNNL-22402, and PNNL-22415). While the testing was performed for the Hanford Waste Treatment and Immobilization Plant (WTP), the results are applicable to other DOE facilities, subject to limitations and assumptions used in developing the spray leak predictive methodology presented in this section. Experiments were conducted at three pressures (100, 200, and 380 psig), using several circular orifices ranging from 0.2 to 4.46 mm in diameter, and several slot-shaped orifices (i.e., 0.3 × 5 mm, 1 x 10 mm, 1 x 20 mm, 1 x 76 mm, and 2.74 x 76.2 mm). Experiments were conducted with various liquids which included water, solutions, and Newtonian and non-Newtonian slurry simulants. Testing was carried out in two phases: (1) Phase I, plugging and aerosol tests, and, Phase II, additional testing deemed necessary after examining the Phase I results.

Pressurized Spray Leak Testing Summary

Several liquid simulants were developed and characterized for use in the small-scale Phase I plugging and aerosol tests. The simulants were selected to represent a range of relevant physical and rheological properties. Table A.56 in Volume II of this Standard, the Appendix, lists the small-scale and large-scale target simulants used in both Phase I and Phase II aerosol tests.

It should be noted that because of the hazards and costs associated with some of the simulants developed for the small-scale test stand, only a small subset of simulants was used in the large-scale test stand during Phase I testing.

Tests were conducted by varying the distance between the orifice and a splash wall perpendicular to the spray, and by also varying the chamber size (i.e., a small-scale and large-scale chamber) in which tests were conducted. In the small-scale chamber, the distance varied from 1-inch to 42-inches, while in the large-scale chamber, the distance varied from 3.5-feet to 38-feet. Two methods were used to determine the aerosol generation rate, termed "in-spray" and "in-chamber." In-chamber measurements employ aerosol analyzers installed several feet above the spray at both upstream and downstream measurement locations. In this configuration, the aerosol analyzers preferentially observe the smaller aerosols capable of being suspended and mixed throughout the chamber. In-spray tests employ aerosol analyzers that are located in the direct downstream path of the spray leak jet.

Prior to the conduct of the experiments, three experimental methods were considered to measure the aerosol net generation rate and release fraction: 1) direct in-spray measurements; 2) steady-state aerosol concentration measurements in a chamber with different volumetric purge rates; and 3) transient aerosol concentration measurements in a chamber with no purge flow. The first experimental method measures the aerosol directly in the spray, providing an explicit measurement of the aerosol droplet size distribution at a specific position. The release fraction for any given size of droplet is equal to the volume fraction of it in the spray, as given by the droplet size distribution. The second experimental method is to generate a steady spray and measure the steady-state concentration within a chamber by varying the flow rates of clean air introduced into the chamber to dilute the aerosol. The generation rate is then calculated from the measured aerosol concentration with different purge rates. The third experimental method consists of measuring the rate of increase in aerosol concentration in a closed chamber of known volume. Using a simple material balance, the rate of concentration increase gives the aerosol net generation rate from a spray. The first and third methods were used in the testing.

The in-spray measurement technique for the jet centerline was capable of measuring the larger droplets but could not measure droplets with diameters less than 50 µm. The in-chamber measurements used the time history of aerosol concentration in the chamber together with a first order rise model to determine the generation rate. The in-chamber measurements also accounted for plateout due to wall deposition, and any net loss or gain from splash and splatter on the far wall, as well as any splash of droplets in the pool on chamber floor. The in-chamber method was determined to be biased low for larger droplet sizes due to gravitational settling effects.

Parameters Evaluated by PNNL Testing

To investigate the possible parameters that might be needed in developing a new correlation, a number of parameters were tested including fluid viscosity, fluid rheology, orifice discharge characteristics, and contributions from evaporation and splash/splatter.

A full list of parameters tested by PNNL is summarized in Table A.57 in Volume II of this Standard, the Appendix. The testing showed that water bounds, or is equal to, the results from the other substances tested. In addition, there was some early concern regarding whether any orifices would become plugged from solid particles, although testing did not bear this out.

Pressurized Spray Leak Correlation

A reasonably conservative correlation for aerosol generation rate was developed based on the in-chamber test data and extrapolations of the in-chamber data to 100 ft chambers. The correlation was developed for water, but it is appropriate for all the liquids and slurries tested because the aerosol generation from the other Newtonian and non-Newtonian fluids is overwhelmingly always the same or less than water sprays. The correlation was compared to in-spray data and was determined to match the in-spray data for the range of orifice sizes and spray pressures that were tested. The good comparisons in the regions of overlap for different size orifices and different spray pressures confirm that the conservative correlation has orifice area, spray pressure, and droplet size dependences that agree with the in-spray data. Since the conservative correlation matches the in-spray results, it can be concluded that the conservative correlation accounts for any potential biases (humidity and method bias) with the in-chamber method, without actually quantifying them.[5]

Because the upper confidence interval correlation and values account for the uncertainty in fitting the chamber concentration data, the correlation for the upper confidence interval can be used in developing a reasonably conservative correlation. One approach for obtaining a conservative correlation is to adjust the correlation so that all, or the majority, of the measured values are less than the correlation. The most sensible adjustment is to increase the leading coefficient and not adjust the exponents for the individual parameters of orifice area, spray pressure, and droplet size. The following result, presented in Equation 3-5a (see PNNL-22415, Equation 10.4), was selected to have nearly all the

[5] Venting of superheated liquid (flashing spray) was not within the scope of the PNNL testing.

measured values in the upper 95% confidence interval either the same or less than the conservative correlation.

$$GR_C = 3.26 \times 10^{-16} (A)^{0.793} (P_s)^{2.18} \left(d_p\right)^{2.40} \qquad \text{(3-5a)}$$

where:

GR_C is the conservative correlation for generation rate (m^3/s)

A is the orifice(s) area (mm^2)

P_s is the spray pressure (psig)

d_p is the aerosol droplet diameter (μm)

Figure 3-4a shows the comparison of the measured values in the upper 95% confidence interval with the conservative correlation. The results from the individual tests are less than or equal to the conservative correlation with the exception of two individual points which are barely above the correlation line.

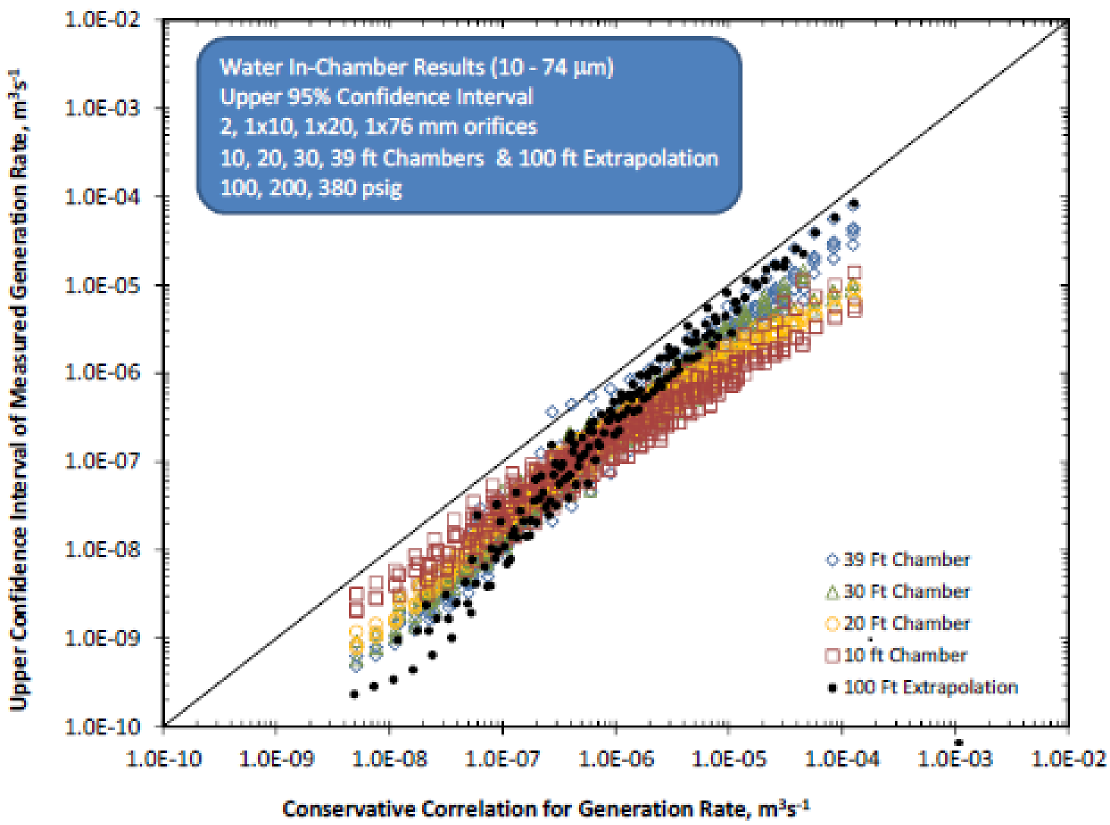

Figure 3-4a Comparison of Measured Upper Confidence Interval of Generation Rate with the Conservative Correlation for Generation Rate

Figure 3-4b shows a comparison of the measured generation rates with the conservative correlation. The measured generation rates are all lower than the upper confidence interval values, and this figure shows that all of these measured generation rates are even farther below the diagonal line, as expected.

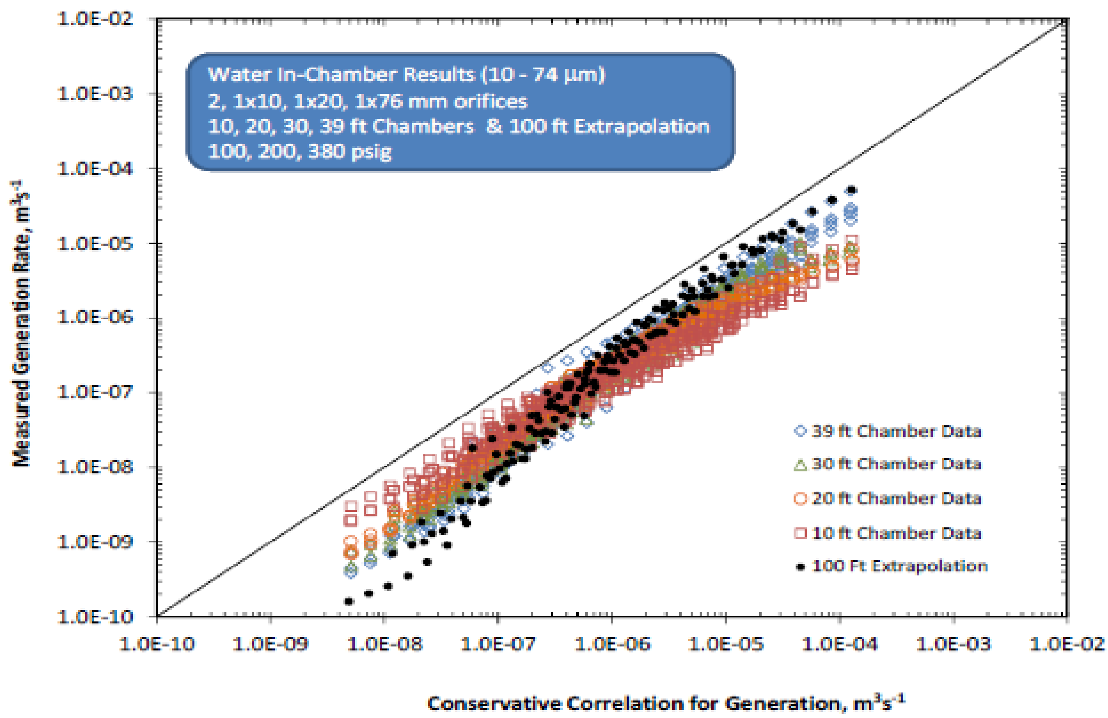

Figure 3-4b Comparison of the Measured Generation Rate

Guidance on Correlation Spray Breach Area, Pressure and Droplet Size Parameters

<u>Spray Breach Area</u>

The PNNL correlation requires an input value for the orifice area. The PNNL spray leak test data in PNNL-22415 indicates that the crack shape has minimal impact on the aerosol generation rate. For convenience, the orifice can be modeled using the guidance in NUREG-0800, *Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Power Plants*, Branch Technical Position (BTP) 3-3, "Protection Against Postulated Piping Failures in fluid Systems Outside Containment" for determining the orifice size. The BTP 3-3, Appendix B, "Size and Types of Pipe Breaks and Cracks," states:

- "… The critical crack size is taken to be 1/2 the pipe diameter in length and,
- 1/2 the wall thickness in width."

Use of this criterion is corroborated by historical data on piping system breaches. RPP-5667, *Stochastic Consequence Analysis for Waste Leaks*, provides data on crack sizes from historical transfer piping system failures. RPP-5667, Appendix B, “Probability Density Functions for Leak Length, Width and Depth”, summarizes data from several reports, and the data provides an indication of the range of crack sizes that may result in pipe failures from any “non-mechanical” cause. This provides a gauge of the reasonableness of following the NUREG-0800 guidance. The historical data found in RPP-5667, Appendix B, on piping system fracture sizes show most of the breach sizes were under 100 mm^2.

Using this guidance from NUREG-0800 determines the area[6] for use in the correlation.

The critical crack size guidance is for rigid pipes larger than 1-inch diameter. However, a conservative approach for pipes with a diameter smaller than 1 inch, would be to use the correlation utilizing 1 inch as the pipe diameter, not the actual smaller diameter. The use of the correlation is also considered conservative for non-rigid pipes (i.e., flexible pipe or rubber hosing).

Pressure

The conservative correlation is based on testing at three pressures, 100, 200, and 380 psig. Below a certain pressure a spray will transition to a spill. A reasonable lower bound for this transition point is that less than the lowest tested pressure of 100 psig (i.e., at below 100 psig), treat the event as a spill, and at greater than or equal to 100 psig, treat it as a spray. However, when pump pressures are close to 100 psig, a conservative approach would be to use the correlation utilizing 100 psig as the pressure. For other pressures less than 100 psig, another conservative approach is to select the larger estimate of airborne release from the free-fall spill (Chapter 3) and the PNNL correlation for the facility-specific conditions, or if additional conservatism is deemed not necessary for the situation being evaluated, follow the free-fall spill recommendations. Additionally, PNNL indicates that the correlation can be extrapolated to higher pressures but does not state an upper bound for the pressure. Since PNNL-22415 gives an example that uses a pressure of 540 psig, establishing an upper bound of 600 psig is reasonable. Accordingly it

[6] Note: the PNNL testing showed that a circular orifice with an equivalent area gives the same results as a rectangular slot.

is strongly suggested that the maximum pump pressure be applied in the correlation to represent an unmitigated release.

Droplet Diameter

Consistent with the discussion in Section 3.2.2.3.1 of the handbook, the droplet diameter is the actual measured diameter of the aerosol. With respect to RF, the size of interest is $\leq$ 10 μm, therefore the aerosol droplet diameter used in the correlation is set to 10 μm. No conversion to AED is necessary since the correlation was developed for water and it bounded all other liquids and slurries tested. Based on the conclusions of the PNNL experiments, the effects of humidity and evaporation on droplet size can typically be ignored. However, if the accident scenario involves extreme energetic conditions (i.e., fire) that warrant consideration of significant evaporation due to the additional sensible heat, the droplet diameter can be adjusted to account for evaporation (i.e., larger droplet sizes can evaporate to $\leq$ 10 μm at the collocated worker or the site public boundary location).

Note: The PNNL spray correlation provides the bounding source term for spray release scenarios such that contribution from splash or splatter does not need to be accounted for separately.

PNNL testing was conducted specifically to address concerns regarding contribution to the source term from additional droplet formation due to impingement on surfaces and additional droplet formation due to impingement on a liquid pool (i.e. splash and splatter). The testing considered a range of orifice sizes, pressures, chamber sizes, and distance from the spray location to a perpendicular surface. Large-scale testing concluded that the largest release fraction always occurred with sprays that traveled the full length of the chamber (PNNL-21333).

Calculating Pressurized Spray Release Source Term

The radiological source term (ST) is determined by multiplying the generation rate times the duration of the event (t):

$$ST\ (m^3) = GR_C\ (m^3/s) \times t\ (s)$$

limited by the total flow rate through the orifice over the event duration based on a bounding estimate of MAR x DR, such as maximum tank volume. The event duration is determined consistent with DOE Standard DOE-STD-3009-2014, *Preparation Guidance* *for U.S. Department of Energy Nonreactor Nuclear Facility Safety Analysis*, and DOE-HDBK-1224-2018, *Hazard and Accident Analysis Handbook*, but is not to exceed eight (8) hours for scenarios that are slow to develop. Convert the volume of the source term into its radiological constituents as input to atmospheric dispersion codes to estimate radiological dose consequences to the collocated worker and MOI.

Additional Tables for inclusion in a revision to DOE-HDBK-3010-94, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities, Volume II-Appendices*:

Table A.56. Target Simulants

Liquid	Target Property Range
Phase I Small-Scale Target Simulants	
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
Solutions of water and sodium nitrate/sodium thiosulfate	Viscosities of ~1.5 and ~2.5 mPa·s (cP).
Gibbsite and boehmite particulates in water	The particle size distribution (PSD) of the slurries were selected to match small treated Hanford slurries with 8 and 20 wt% solids.
A washed and leached simulant (PNNL-18894)	Solids loading was adjusted to meet target Bingham yield stresses of 6 and 30 Pa.
Phase I Large-Scale Target Simulants	
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
Solution of water and sodium thiosulfate	Viscosity of ~2.5 mPa·s (cP).
Boehmite particulates in water	The PSD of the slurries was selected to match small treated Hanford slurries with 8 and 20 wt% solids
Phase II Small-Scale Target Simulants	
Water	Viscosity 1 mPa·s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
Boehmite particulates in water	The PSD of the slurries was selected to match small treated Hanford slurries at a concentration of 27 wt% solids.
Small fraction of Mo in water and a boehmite-water slurry	1 wt% (in the slurry) Mo particles included to represent dense particles in the waste such as plutonium oxide. The boehmite slurry had a total

Table A.56. Target Simulants

Liquid	Target Property Range
	solid loading of 20 wt% (i.e., 19 wt% boehmite and 1 wt% Mo).
Clay slurries composed of a solid phase with 80 wt% kaolin and 20 wt% bentonite in water	The total solids loadings were adjusted, via dilution, before testing began so that one simulant had at least one Bingham parameter near 30 Pa/30 cP (target range was 30 ± 4 Pa or cP) and the second Bingham parameter less than or equal to the 30 ± 4 Pa or cP target. The second simulant was adjusted so that at least one Bingham parameter near 6 Pa/6 cP (target range was 6 ± 2 Pa or cP) and the second Bingham parameter greater than or equal to the 6 ± 2 Pa or cP target.
A washed and leached simulant (PNNL-18894)	Same.
Phase II Large-Scale Target Simulants	
Water	Viscosity 1 mPa-s (cP), density 1,000 kg/m^3, and surface tension 73 mN/m.
80/20 solids blend of a kaolin/bentonite clay slurry, 32 wt%	The solids loading was adjusted to meet target Bingham yield stress of 30 Pa.
80/20 solids blend of a kaolin/bentonite clay slurry, 27 wt%	The solids loading was adjusted to meet target Bingham yield stress of 6 Pa.

Table A.57. Parameters Tested

Parameter	Discussion	Reference
Plugging of Small Breaches	Small circular orifices, 0.188-mm in diameter, plugged in 9 of 11 tests. However, no combination of simulant and pressure plugged orifices $\geq$ 0.382 mm or either of the slots tested. The orifice dimensions that can be assumed to consistently plug are, therefore,	PNNL-21361

Table A.57. Parameters Tested

Parameter	Discussion	Reference
	smaller than the orifice dimensions tested with the range of simulants and pressures employed.	
Fluid Viscosity	Test results indicated that for a salt solution, as fluid viscosity and density increased, the cumulative release fraction was unchanged. PNNL-21333 test results also indicated that for a salt solution, as fluid viscosity and density increased, cumulative release fraction and generation rate varied by a slight amount that was dependent on droplet size. Considering uncertainty in test data, there was essentially no difference in the test results for water and a salt solution.	PNNL-21361 PNNL-21333
Surface Tension and Anti-Foam Agent (AFA)	The addition of AFA did not cause an increase in the release fraction. To the extent that an effect could be distinguished, the presence of AFA caused a slight decrease in the release fraction.	PNNL-21367
Orifice Discharge Coefficient	The PNNL-31367 report notes that the cumulative release fraction correlates reasonably well with orifice area for slots and round holes The PNNL-22402 report states that orifice coefficients significantly greater than 0.62 occur in orifices regardless of type. However, the conservative generation rate correlation uses an empirical approach that does not explicitly use an orifice discharge coefficient. .	PNNL-21367 PNNL-22402
Slot Orientation and Aspect Ratio	Three situations were tested: (a) slot length aligned with the pipe axis, water flowing through the pipe and spraying through the slot,	PNNL-21367

Table A.57. Parameters Tested

Parameter	Discussion	Reference
	(b) slot aligned along the pipe circumference, water flowing through the pipe and spraying through the slot, and (c) an axial slot in the 'dead-end' configuration. The dependence on area was found to hold regardless of orifice shape and slot orientation and did not have an appreciable effect on the release fraction.	PNNL-21333 PNNL-22415
Multiple Nearby Orifices	Tests were performed using an array of five round holes lined up with the header axis, with each orifice separated by a distance equal to the hole diameter. The array of 1-mm orifices had a cross-sectional area that was very similar to the 2-mm orifice (i.e., 3.69 vs. 3.50 mm^2). As expected, the release fractions of the 2-mm orifice and the array of 1-mm orifices are very similar over the entire droplet size range. That they are in good agreement suggests that only cross-sectional area is important for predicting the release fraction of aerosol.	PNNL-21367 PNNL-21333
Solids in Newtonian Slurries	Waste contains undissolved particulate, and most of the radioactive material is in this solid phase. Therefore, the slurry streams are of most concern from the radiological source term perspective. Pressure atomization of slurries involves interactions of the three different phases (i.e., suspended solids, carrier liquid and dissolved salts) with air at standard atmospheric pressure. Overall, the results indicate a straightforward functionality of the cumulative release fraction on pressure and orifice area; whereas, liquid viscosity and the weight fraction of solids had a negligible effect on the cumulative release fraction.	PNNL-21333

Table A.57. Parameters Tested

Parameter	Discussion	Reference
Surface Tension	When the formation of an interface is rapid compared to the time it takes surface-active species to diffuse to and adsorb at the interface, the surface tension is different than the equilibrium value; this time dependent surface tension often is called the dynamic surface tension. The dynamic surface tension approaches that of the pure fluid as the time scale for interface formation becomes progressively shorter. Because of this, the addition of surfactants, such as AFA, caused little change in droplet size distribution compared to that for water because the surface tension was equal to that of water at the time scale for droplet formation.	PNNL-21333
Small Concentration of Dense Particles in Slurries	PNNL-21367 indicated that low solids concentrations appeared to depress release fractions below those of water over most or all of droplet size range for baseline slot and round orifices. Increasing solids content to 20 wt% increased the release fraction. PNNL-22402 indicated that addition of a small fraction of dense particles to water and 19 wt% small-treated simulant did not result in a significant effect on measured release fractions when compared to simulants devoid of dense particles. However, as mentioned, water bounded or equaled the testing done with other substances.	PNNL-21367 PNNL-22402
Effect of Non-Newtonian Rheology	PNNL-22415 testing measured in-spray release fractions for water and for two non-Newtonian clay slurries as a function of downstream distance from the orifice. Tests at	PNNL-22415

Table A.57. Parameters Tested

Parameter	Discussion	Reference
	100 and 200 psig showed the release fraction for clay slurries decreases with increasing solids content. Water release fraction results are typically the same or larger than release fractions measured for non-Newtonian clay slurries. PNNL-22402 compared clay slurry and water cumulative release fractions in the 10 μm to 100 μm droplet size range and stated that clay release fractions are less than or equal to those of water at both 6 Pa and 30 Pa yield stress.	PNNL-22402
Contribution from Splash/Splatter	PNNL-21367 states that aerosol is generated by primary and secondary jet breakup and by "splatter" droplets formed when the jet, or droplets formed by jet breakup, hit the splash wall at the downstream end of the enclosure. Testing indicated that as distance between spray and splash wall decreased, cumulative release fraction remained essentially constant between 42 in. and 18 in., increased slightly between 18 in. and 3 in., and increased significantly at a distance of 1 in. PNNL-21333 states that for the large-scale chamber, the spray distance varied from 43 in. to 227 in., essentially the full length of the chamber. For the five different orifices tested at pressures of 200 and 380 psig, the largest release fraction always occurred with sprays that traveled the full length of the chamber.	PNNL-21367 PNNL-21333
Evaporation	PNNL-22402 testing indicated that the initial relative humidity (RH) in the chamber affects the measured release fraction.	PNNL-22402

Table A.57. Parameters Tested

Parameter	Discussion	Reference
	PNNL-22415 also discusses testing of RH impacts on water sprays that was conducted in the 20, 30, and 39 ft chambers and humidity tests with the 20 ft chamber and 6 Pa clay simulant. All RH tests were performed using a nominal 2 mm hole and a 380 psig spray. The results indicate that, regardless of chamber size, release fraction is reduced at low humidity across all droplet sizes. While release fraction measurements for water appear to be impacted more strongly at smaller droplet sizes ($\leq$ 20 µm), the difference in the reduction for small and large droplets does not appear to be great. The degree of reduction appears to scale proportionally to the difference between the test humidity and 100 percent, and any divergence from this behavior appears to derive from measurement uncertainty rather than a phenomenological mechanism. It is concluded that correction factors for RH are relatively small.	PNNL-22415
Deposition	Size distribution can change with time as aerosols are preferentially retained or removed from the system. Loss in the region outside the spray is assumed to occur through several means: • deposition • settling • entrainment into the jet • coalescence or aggregation • evaporation, minimal because of pre-wetting of the chamber before the tests	PNNL-21367

Table A.57. Parameters Tested

Parameter	Discussion	Reference
	The overall aerosol balance is derived by considering the sum of generation and loss terms. The testing evaluated an aggregate aerosol generation rate that takes into account generation and removal mechanisms.	

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