**Aerosol Formation from High-Pressure Sprays for Supporting the
Safety Analysis for the Hanford Waste Treatment and Immobilization Plant - 13183**

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ABSTRACT

The Waste Treatment and Immobilization Plant (WTP) at Hanford is being designed and built to pretreat and vitrify waste currently stored in underground tanks at Hanford. One of the postulated events in the hazard analysis for the WTP is a breach in process piping that produces a pressurized spray with small droplets that can be transported into ventilation systems. Literature correlations are currently used for estimating the generation rate and size distribution of aerosol droplets in postulated releases. These correlations, however, are based on results obtained from small engineered nozzles using Newtonian liquids that do not contain slurry particles and thus do not represent the fluids and breaches in the WTP. A test program was developed to measure the generation rate, and the release fraction which is the ratio of generation rate to spray flow rate, of droplets suspended in a test chamber and droplet size distribution from prototypic sprays. A novel test method was developed to allow measurement of sprays from small to large breaches and also includes the effect of aerosol generation from splatter when the spray impacts on walls. Results show that the release fraction decreases with increasing orifice area, though with a weaker dependence on orifice area than the currently-used correlation. A comparison of water sprays to slurry sprays with 8 to 20 wt% gibbsite or boehmite particles shows that the presence of slurry particles depresses the release fraction compared to water for droplets above 10 μm and increases the release fraction below this droplet size.

INTRODUCTION

The U.S. Department of Energy Office of River Protection manages the River Protection Project, which has the mission to retrieve and treat the Hanford tank waste for disposal and close the tank farms [1]. The Waste Treatment and Immobilization Plant (WTP) is a major element for achieving this mission. One of the events postulated in the hazard analysis for the WTP is a breach in process piping that produces a pressurized spray generating small aerosol droplets that remain suspended in the air in an enclosure and therefore can become airborne releases when transported into ventilation systems. The piping breaches are postulated to result from a number of causes, including pipe erosion/corrosion, mechanical impact, and seal/gasket failures. The breaches are expected to be rough in texture and irregular in shape.

The formation of aerosol droplets for sprays from a range of nozzles and fluids has been widely studied and good summaries of correlations and mechanisms are available in textbooks [2-4] and review articles [5]. Though droplet formation from nozzles may be different than from potential pipe breaches, the general behavior is expected to have similarities. When a high-pressure (and high velocity) fluid stream exits a breach in a process pipe a number of mechanisms occur as the stream, or jet, breaks up into droplets of different size. Initially, the stream breaks up into primary droplets. These droplets, which are moving faster than the surrounding air, may then be distorted by aerodynamic forces and breakup to form even smaller droplets. Some of these droplets, preferentially the larger ones, may impact on nearby surfaces and make additional smaller droplets by splattering on the surface or, more probably, be substantially captured by the surface. Within a chamber or facility, the net generation of droplets that

become suspended in the chamber and the size distribution of the droplets depend on all of the generation and capture mechanisms.

The current approach for predicting the size and generation rate of aerosols produced in a postulated spray leak in the WTP involves using literature correlations [6-7]. These correlations are based on results obtained from small engineered spray nozzles using Newtonian fluids. The narrow ranges of physical properties, on which the correlations are based, do not cover the wide range of slurries and viscous materials that will be processed in the WTP and across processing facilities in the DOE complex. In addition, the postulated breaches for the WTP have a much larger size range and have different geometries than the engineered spray nozzles used in the correlation. Finally, the postulated sprays may impact on walls, or other equipment, and the combined effect of droplet formation from the spray, droplet formation from splatter, and droplet capture by the impact surface has not been included in previous correlations.

The purpose of this paper is to summarize test results for net aerosol generation (total generation minus capture) for two technical objectives that address limitations in existing literature and correlations. The first technical objective is to determine the aerosol droplet size distribution and net generation rate from prototypic breaches, specifically including sprays from larger breaches for which data in the literature are scarce. These results are compared to the literature correlation for total generation currently being used for hazard analyses for the WTP. The second technical objective is to quantify the role of slurry particles on the aerosol net generation rate for slurries that are representative of some of the expected waste streams in the WTP. The objectives, methods, and results discussed here are part of a broader testing effort on postulated spray leaks in the WTP [8-10].

To address these technical objectives, small- and large-scale test systems were constructed and operated with water and slurry simulants to determine aerosol net generation rates and release fractions from a range of breach sizes and geometries (the release fraction is defined as the net generation rate divided by the total spray flow rate). In the section below, the literature correlation currently used by the WTP for hazard analyses is discussed together with other correlations, followed by a summary of previous studies on the effect of slurry particles on aerosol formation. The experimental method is then summarized, including details on the experimental approach, data analysis method, apparatus, simulants, and test conditions. The test results for the effect of orifice size and role of slurry particles are then summarized.

Effect of Orifice Size on Droplet Size Distribution

There are a number of correlations in the literature for the effect of orifice size on droplet formation from orifices and nozzles. For plain-orifice atomizers [2, 3], the correlations typically give the Sauter mean diameter (SMD) as a function of orifice diameters, fluid properties, and spray velocities. These correlations typically show decreasing droplet size with decreasing orifice diameter, which generally agrees with the original data from hollow-cone nozzles [11] used in developing the DOE Handbook for analyzing spray releases [12]. In contrast, the recent work by Epstein and Plys [13] and the original work of Merrington and Richardson [14] show no effect of orifice size.

Larson and Allen [6] summarize the methodology used by the WTP for estimating the aerosol release fraction and generation rate of spray releases, and McAllister [7] provides additional details on the equations and method. The method uses the theoretically based correlation by Dombrowski and Johns [15] for estimating the SMD (assuming a spray angle of 150° for the nozzle parameter [16]) and

then estimates the aerosol droplet size distributions using the Rosin and Rammler [17] distribution (assuming an exponent of 2.4 for the measure of the spread of the distribution [2]). This methodology used by the WTP, which does not include impact capture of droplets, gives release fractions that decrease with orifice diameter and will be compared with the experimental results below.

In the experimental method discussed below, the net generation rate of aerosol droplets is measured together with the flow rate of the spray. To compare with predictions from the current WTP model, the release fraction is determined by the ratio of the measured net generation rate to the measured spray flow rate. The experimental net generation rates include the effect of capture and are therefore expected to be less than the model predictions, which do not include capture. For the experimental results presented later in this paper, the results are presented as cumulative release fractions (releases for all droplets smaller than a stated diameter).

PREVIOUS WORK ON THE EFFECT OF SLURRY PARTICLES

A few previous studies have evaluated the role of slurry particles on aerosol droplet formation. While there are many specific observations, there are some overall general findings. One group of studies evaluated the role of slurry particle size and generally determined that, if the slurry particles are smaller than the droplets, the slurry particles do not tend to affect the droplet distribution [18-21]. When the slurry particles become progressively larger and specifically larger than droplets that would be generated in the absence of the slurry particles, droplet formation is naturally influenced. Droplets smaller than the slurry particles can still be formed, but these droplets will not contain slurry particles, and the larger droplets will contain the slurry particles. Breitling et al. [22] presented both computational fluid dynamics and experimental results for shear-thinning lime slurries sprayed from hollow cone pressure swirl nozzles. The results showed little difference between the slurry and water, but there was no information on the size of the lime particles. Hecht et al. [23] found that increasing solids loading resulted in a small decrease in the droplet size. Son and Kihm [24] studied the effect of coal slurry particles on spray formation. The coal slurries were non-Newtonian and had progressively higher apparent viscosities for progressively smaller particles. The aerosol results showed that larger aerosol droplets were generated as the coal particle size became smaller, and Son and Kihm [24] suggested that the primary reason was the increase in apparent viscosity for the smaller particle slurries. Dombrowski and Munday [25] found that a small volume fraction of wetting particles did not change the breakup of a fan jet but that a high particle concentration in the slurry changed the behavior markedly and resulted in larger droplets. Finally, Hecht and Bayly [26] discussed how aerosol formation from concentrated non-Newtonian slurries is affected by a range of phenomena associated with the particles, their interaction, and the slurry rheology. Overall, the literature on aerosol formation with slurries suggests that slurry particles can affect droplet formation in ways that can either increase or decrease the size of the droplets.

EXPERIMENTAL METHOD

The experimental method for measuring the net generation rate of droplets from the combined effect of droplet formation in the spray, droplet formation from splatter, and droplet removal by capture consists of measuring the rate of increase in aerosol concentration in a closed chamber of known volume. By applying a simple material balance for the aerosol in the chamber, and assuming uniform concentration or that an average concentration can be measured and represent the overall chamber, the rate of concentration increase gives the aerosol generation rate from a spray. Fig. 1 shows a conceptual example of concentration increase with time, for different cumulative droplet volumes, where the initial rate of

increase can be estimated from the initial increase in concentration. Eventually, the concentrations no longer increase linearly with time and approach steady-state values. This behavior results from aerosol losses in the chamber. Measuring the accumulation rate in a chamber is different from the more typical method of measuring the droplet size distribution within the spray and then assuming that the generation rate is a product of the size distribution, or release fraction, and flow rate. The primary advantage of using the transient chamber concentrations is the inclusion of droplet formation from splatter, and this approach can be used to evaluate sprays from large breaches that might be difficult to study with in-spray measurements. Some disadvantages of the method are the uncertainty caused by non-uniform concentration in the chamber and that the measured net generation can depend on the chamber size.

To determine the net generation rate of droplets within the chamber, a simple material-balance model for the overall accumulation of aerosol droplets in a chamber in terms of droplet concentration, a loss term that is linear in concentration, and the net generation of droplets accumulating in the chamber was derived from a material balance. The net generation rate is calculated from the initial slope of the model fit and the chamber volume. The material balance equation was used to fit the experimental data. For the small-chamber data, the first 20 s of the sprays were typically used in the fit, subject to some constraints, while the fitting period was typically 60 s for large-scale data. The choice of the fit period was made to constrain the values of the adjustable parameters for net generation rate and loss rate constant so that most or all of the rise period would be included, but the scatter in the post-rise steady-state concentration would not overly influence the fit. Additional details on comparing the two fitting approaches and evaluating the goodness-of-fit are discussed in Mahoney et al. [9] and Schonewill et al. [10].
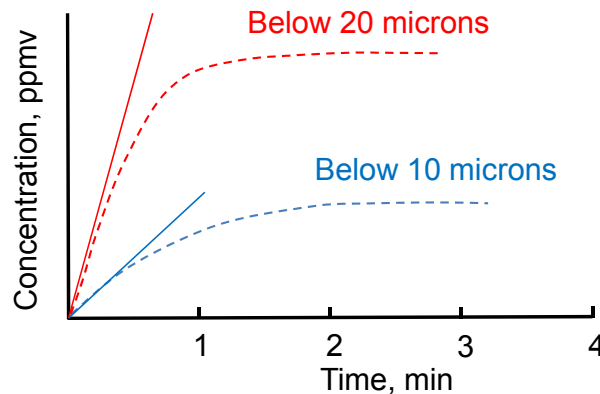

Fig. 1. Schematic of Aerosol Concentration Increasing with Time where the Aerosol Generation Rate is Calculated from the Initial Slope (solid lines)

EXPERIMENTAL TEST SYSTEMS

Both a small- and large-scale test systems were developed to create high-pressure sprays inside enclosures and to measure the transient increases in droplet concentrations within the chambers (see Bontha et al. [27] for additional details). Each system had the same general configuration and primary components provided the same functions, although at different sizes. Each system had a recirculating flow loop connected to a mechanically agitated feed tank containing test liquids and slurries. The flow loops had pumps that were capable of maintaining pressure up to 2.62 MPa (380 psig) while delivering flow through the loop and back to the feed tank. Each flow loop had exchangeable test orifices with a range of sizes and shapes. The flow rate through the orifice during a spray could be measured by either mass

change in the feed tanks or with the difference between flow meters upstream and downstream of the orifice. For each test system, the sprays could be quickly initiated. In the small-scale system the orifices were placed in a by-pass section of the flow loop where hand-operated valves redirected the flow to the orifice almost instantly. In the large-scale system a spring-loaded clamp positioned a plug over the orifice that could be released almost instantly. The size and concentration of aerosol droplets were measured with Malvern Insitec-S instruments. The small-scale chamber used a single Malvern and three Malverns (the data were averaged for use with the material balance model) were used in the large-scale chamber. Additional details on the experimental systems are given in Mahoney et al. [9] and Schonewill et al. [10].

Figure 2 shows the small-scale system assembled in a walk-in fume hood together with a typical water sprays from of an orifice. Figure 3 shows a drawing of the large-scale test system with a 6.10 m (20 ft) chamber. The overall dimensions of the small-scale chamber were approximately 0.76 m wide x 0.76 m high and 1.45 m long (30 in. x 30 in. x 57 in.), with an internal volume of 0.702 m^3 (24.8 ft^3) after accounting for the sloped bottom. The large scale chamber was approximately 2.44-m x 2.44-m x 6.10 m (8-ft x 8-ft x 20-ft) long, with an internal volume of 27.5 m^3 (970 ft^3) after accounting for the slope in chamber walls and other internal components.

Water and slurries of gibbsite or boehmite in water were used in testing. Gibbsite and boehmite materials were selected because they are common components in Hanford tank waste and powders with appropriate PSDs are available. The target particle size distributions (PSD) for specific slurry simulants were selected to match targets based on PSDs of actual Hanford tank waste (Mahoney et al. 2012b). Three different target PSDs were selected for the simulants to match: 1) a typical particle size as-received feed to the WTP – this simulant will be referred to as TAR; a small particle size as-received feed – referred to as SAR; and a small PSD representing the smallest expected for waste after pretreatment operations in the WTP – referred to as STR for small treated. Table I summaries specific gibbsite and boehmite materials used in the testing and the relative proportions for these specific simulants. These simulants were tested at a target concentration of undissolved solids (UDS) of 8 and 20 wt%. The simulants identified in the Results and Discussion section as TAR8, SAR8, and STR8 are 8 wt% slurries and SAR20 and STR20 are 20 wt% slurries. These slurries had essentially Newtonian fluid properties.
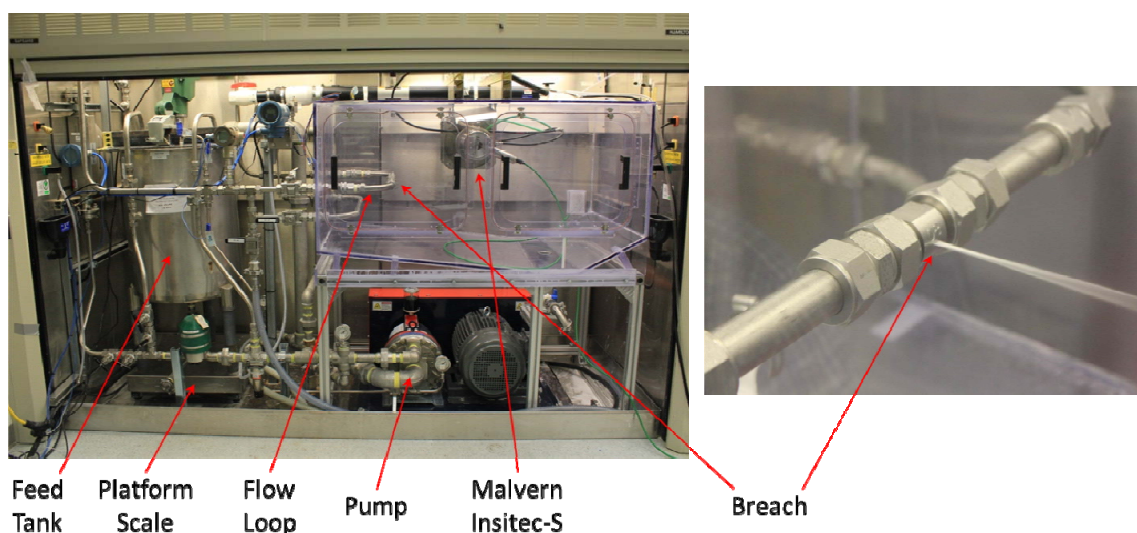

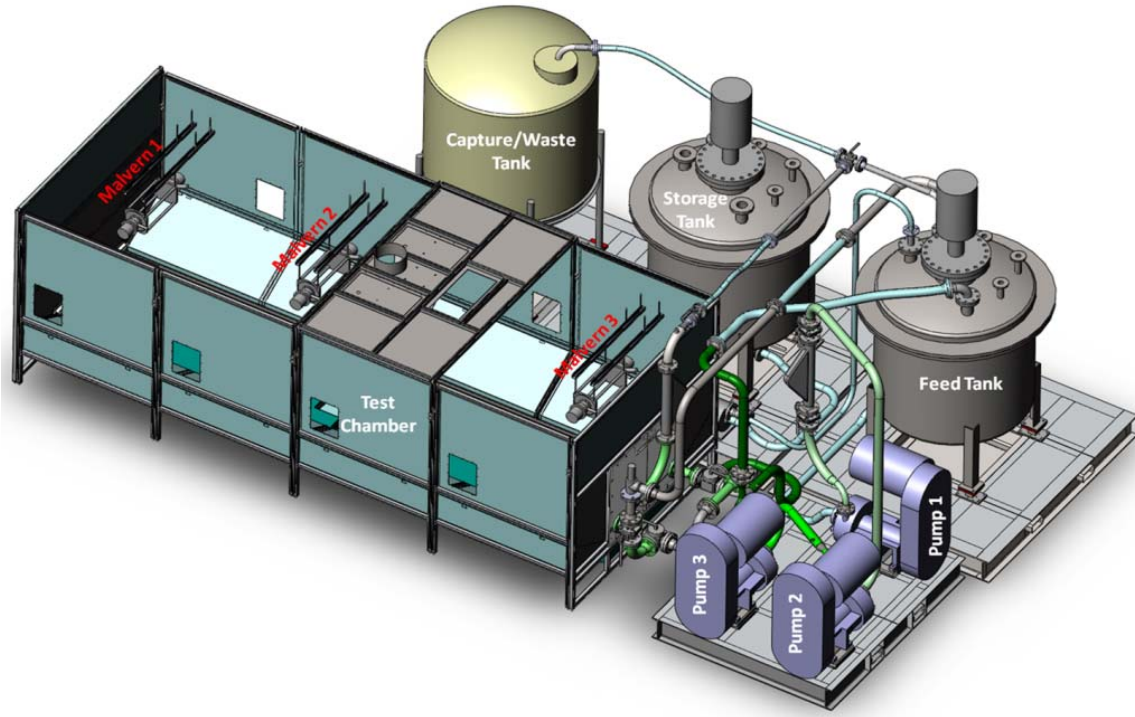Fig. 2. Small-Scale Test System Located Inside a Walk-in Fume Hood and a Typical Water Spray

Fig. 3. Large-Scale Test System Showing Feed Tank, Slurry Pumps and Piping, 6.1-m (20-ft) Test Chamber, and Approximate Location of Malvern Instruments (Breach is just Inside the Chamber near the Pumps)

Table I. Gibbsite and Boehmite Materials and the Proportions for Simulants

Simulant Particle	Component	PSD d_{50} (µm)[a]	Simulants and Target Proportions		
			Small Treated (STR)	Small As-Received (SAR)	Typical As-Received (TAR)
Almatis C333 gibbsite	gibbsite	5.9			100%
Nabaltec APYRAL 40CD	gibbsite	1.3		65%	
NOAH Technologies R6011	gibbsite	9.6		35%	
Nabaltec APYRAL AOH60	boehmite	0.78	80%		
NOAH Technologies R6000	boehmite	0.21	20%		
(a) based on vendor data					

Table II shows the orifices used in the spray tests, the target dimension, actual dimensions, and cross-sectional areas. These orifices span a 200-fold range in area. For the large-scale tests, these orifices were machined in sections of 3-in. (76.2 mm) Schedule 40 SS pipe, which is a common pipe size in the WTP. For the small-scale tests, the orifices were machined from test pieces that had the same wall thickness (5.49 mm, 0.216 in.) as the 3-in. (76.2 mm) pipe used in the large-scale tests. Spray tests were conducted at 2.62 MPa (380 psig) and at ambient temperature. The test method included conducting a pre-spray to wet the chamber walls, which also humidified the chamber prior to testing. This minimized any role of evaporation on the test results. Testing is ongoing to quantify the role of humidity and potential evaporation on the test results.

Table II. Test Orifices for Small and Large Chambers

Orifice Designation	Test Chamber	Target Orifice Dimensions (mm)	Measured Diameter[a] (mm)	Cross-Sectional Area (mm²)	Type
OTP-31	Small	0.3	0.306	0.074	Circular
OTP-13	Small	0.5	0.531	0.22	Circular
OTP-03	Small	0.5	0.534	0.22	Circular
OTP-05	Small	1.0	0.975	0.75	Circular
OTP-06	Small	2.0	2.015	3.2	Circular
S1A	Large	1	1.00	0.78	Circular
S1D	Large	2	2.11	3.50	Circular
S1C	Large	2.74	2.74	5.89	Circular
S1B	Large	4.46	4.67	17.13	Circular
(a) average diameter from analysis of magnified optical images of the orifices					

RESULTS AND DISCUSSION

Results were obtained to compare droplet net generation in the different size test chambers and to quantify the roles of orifice size and the presence of slurry particles on droplet generation. Results for additional test conditions have been obtained as part of a broader study (Mahoney et al. [8-9] and Schonewill et al. [10]).

For tests with identical orifices and test conditions conducted in the small and large chambers, the results would give, preferably, the same net generation rates and release fractions in both chambers. Because of the increased volume of the larger chamber, the aerosol concentrations and the rate of concentration increase will be smaller. In actual tests, of course, the dynamics of the aerosol droplets in the different chambers will influence the test results. Figure 4 shows a comparison of the transient cumulative droplet concentrations in the small and large test chambers for 2.62-MPa (380 psig) water sprays from a 1-mm circular orifice where the sprays traveled essentially the full length of each chamber. The two results are shown with the same ranges for both the x- and y-axis to allow a direct comparison. Note that the concentrations are higher in the small-scale chamber and the rate of concentration increase, or the slope of the droplet concentration with time, also is much higher. This is expected based on the nearly fortyfold difference in the large- and small-scale chamber volumes.

The transient droplet concentrations were analyzed to determine the droplet net generation rate and release fractions. Comparisons between the small- and large-chamber release fractions are shown in Fig. 5 for 2.62-MPa (380 psig) water sprays from 1-mm circular orifices. This plot also shows comparisons to WTP model predictions of total generation rate that are identical for both chamber sizes. In these experiments, the sprays traveled essentially the full length of each chamber.

For these water sprays, the comparison of release fraction results for the small and large test chambers shows a difference in release fraction varying from none to as much as fivefold for droplets between 10 and 100 μm (large-scale tests gave lower release fractions for droplets below about 30 μm and higher release fractions above this size). While differences exist between these release fraction results in the different chambers, given the fortyfold difference in chamber volume the overall agreement in the release fraction results is reasonably good. Other tests have shown that the generation rate and release fraction depend on the position of the spray within the chamber (Mahoney et al. [9] and Schonewill et al. [10]), so the comparison of results for the different chambers in Fig. 5 are specific for sprays that traveled the full

length of each chamber. Additional tests are ongoing for a range of chamber sizes to quantify the effect of chamber size on release fractions.
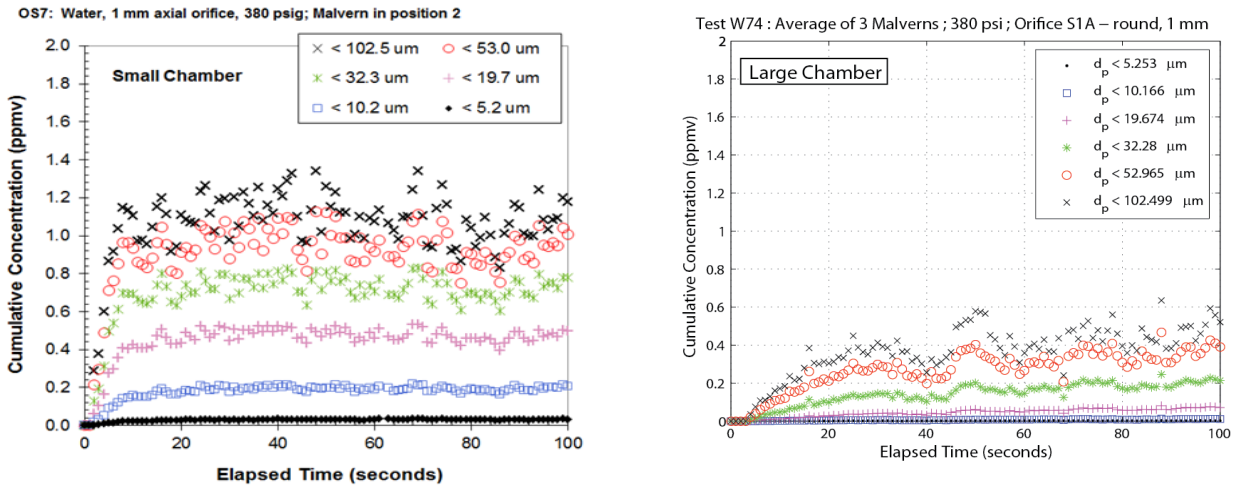

Fig. 4. Comparison of the Transient Cumulative Droplet Concentrations in the Small and Large Chambers for 2.62-MPa (380 psig) Water Sprays from 1-mm Circular Orifices

Effect of Orifice Size

Figure 6 shows the test results for cumulative release fractions for all the circular orifices tested in the small and large chambers. These results are shown for three different cumulative size ranges for droplets. For both the small- and large-chamber tests, the release fraction decreases with increasing orifice area. The trend in the test results (based on net generation in the chamber) is weaker than that in the WTP model (based on total generation of the spray). The results show similar trends of release fraction decreasing with orifice area in both the small and larger chamber results over about a 200-fold range in orifice area.
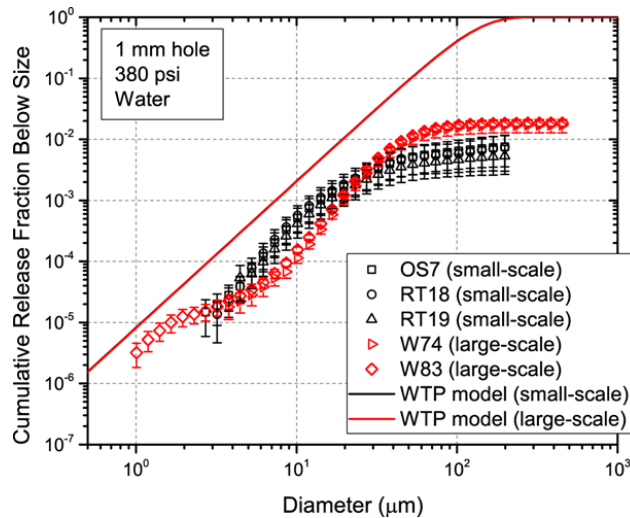

Fig. 5. Comparison of Release Fractions in the Small and Large Chambers for 2.62-MPa (380 psig) Water Sprays from a 1-mm Circular Orifice where the Sprays Traveled Essentially the Full Length of each Chamber
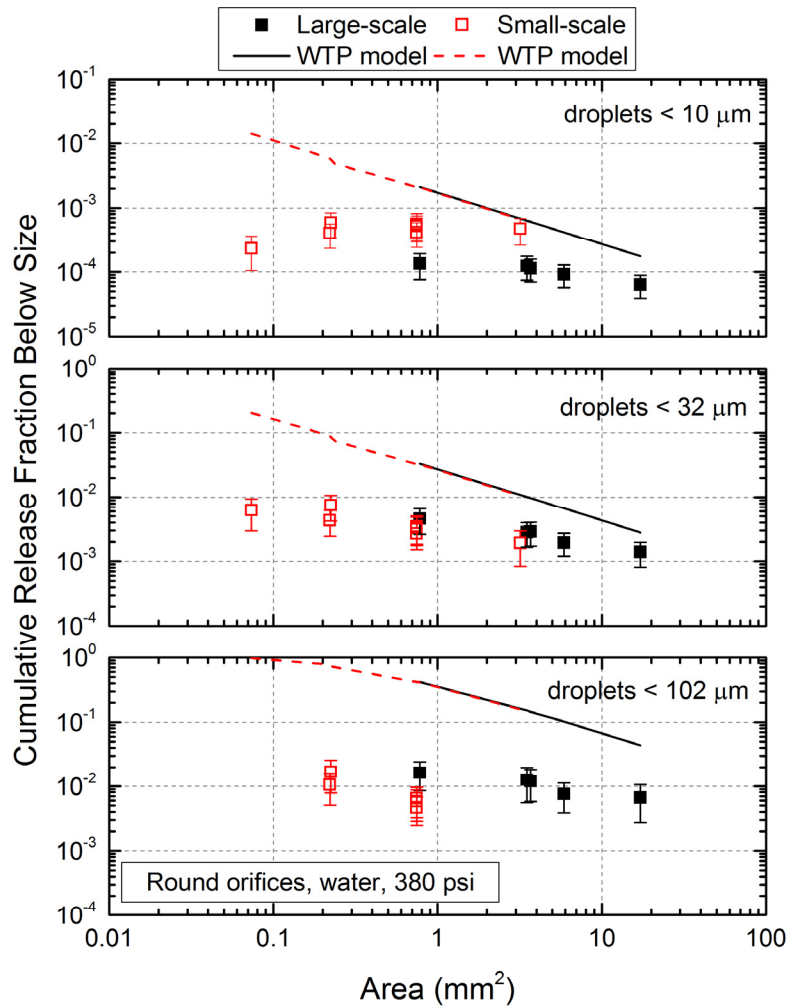
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Fig. 6. Comparison of the Effect of Orifice Area on Release Fractions in the Small- and Large-Scale Chambers for 2.62 MPa (380 psig) Water Sprays from Circular Orifices (results shown separately for droplets <10 μm [top], <32 μm [middle], and <102 μm [bottom]).

Effect of Non-Cohesive Slurry Particles

Figures 7 and 8 show small-chamber release fraction comparisons between slurry and water sprays for gibbsite (SAR and TAR) and boehmite (STR) slurries, respectively. A comparison is also shown for the WTP model, where the model used physical properties for the slurries. For these tests, there is only a small difference between the release fractions for the slurries and water.   The presence of slurry particles generally depresses the release fractions below those of water for the droplet size range above 10 μm and tend to be above the water results for droplets below this size.
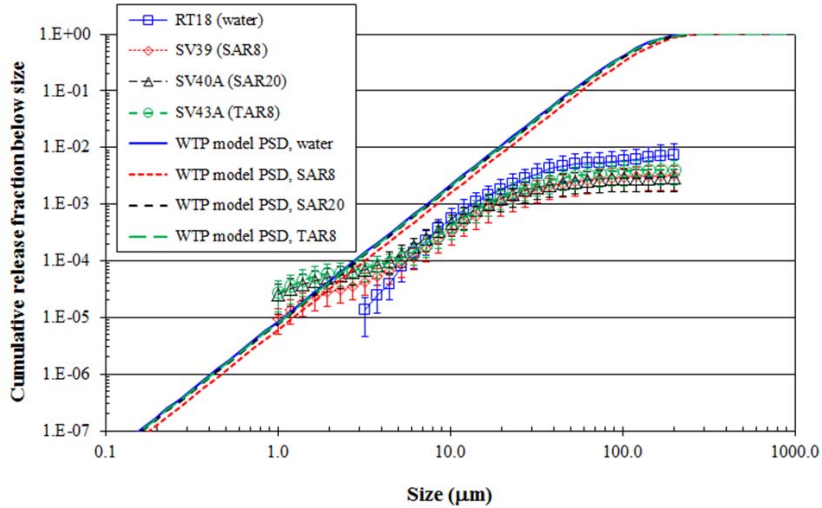
Fig 7. Effect of SAR and TAR Gibbsite Solids on Release Fractions for Water Sprays at 2.62 MPa (380 psig) from a 1-mm Round Hole and Comparison to WTP model Predictions
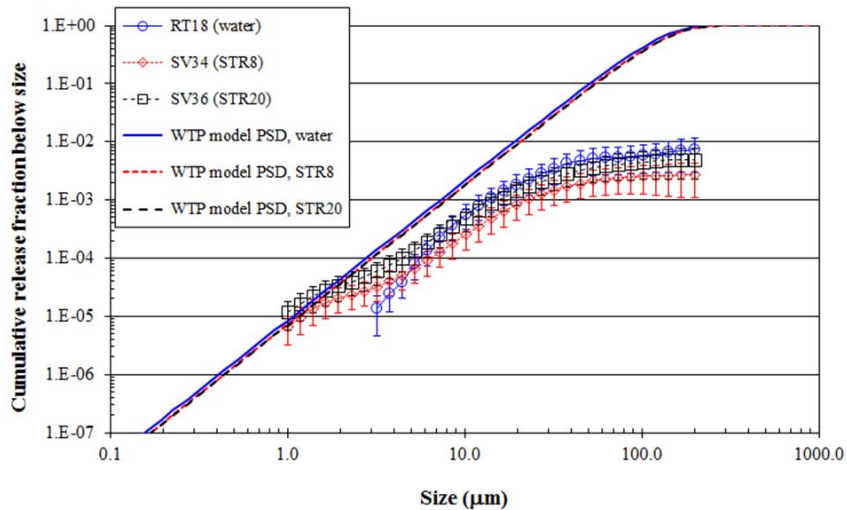

Fig 8. Effect of STR Boehmite Solids on Release Fractions for AFA-Free Sprays at 2.62 MPa (380 psig) from a Target 1-mm Round Hole and Comparison to WTP model Predictions

CONCLUSIONS

A novel test method based on the transient accumulation of aerosol droplets in a closed chamber was developed to measure the net generation rate and release fraction of aerosol droplets created by high-pressure sprays. The method includes the effect of droplet capture and droplet generation from splatter when the high-velocity sprays impact walls. The key conclusions of this study are the following:

- The release fraction decreases with orifice area, though not as strongly as predicted by the current WTP model. The orifice area spanned nearly a 200-fold range for the circular holes tested.

- The presence of slurry particles generally had only a small effect on droplet net generation and generally depresses the release fraction below that of water for droplets above 10 μm and increases the release fraction above the water results for droplets below this size.

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