FLASHING SLURRY RELEASES

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Abstract

The DOE Handbook, DOE-HDBK-3010-94, provides bases for evaluating respirable release fractions (RRFs) from accident releases of superheated fluids (flashing fluids). The Handbook includes flashing RRFs for catastrophic failures, small diameter jet sprays and boiling pools. Estimation of the RRF is important in quantifying consequences and identifying controls for preventing or mitigating a potential release. However, the Handbook does not provide sufficient details to estimate RRFs for a continuous range of fluid superheat (temperature), a continuous range of break sizes and locations (small, medium large and below, at or above liquid levels), or fluid properties (such as slurries versus solutions). In addition, evaluation of scaling effects of the data is not presented (scalability of small scale tests to full scale designs). This paper presents a methodology for evaluating the RRFs from flashing releases of hazardous superheated slurries based on the safety analysis work done on the Sludge Treatment Project (STP) at Hanford and presents solutions for a range of break sizes and locations.

INTRODUCTION

The Hanford K Basin Closure Project involves the retrieval, transfer and processing of radioactive contaminated slurries containing partially corroded spent nuclear fuel from the K Basin spent fuel pools. The spent fuel is primarily metallic fuel from the operation of the Hanford reactors. The Sludge Treatment Project is being designed to treat and package this material in preparation for ultimate disposal. The processing of the contaminated slurries includes further corrosion of the remaining uncorroded uranium metal in a heated pressure vessel to form a more stable metal oxide for packaging and storage. The corrosion process parameters used for the safety basis development were 1.65 MPa (225 psig) and 185^0C (365^0F).

Accident analysis to support the design process and PDSA required computing the potential respirable release fractions RRFs for the release of superheated slurry streams from the heated vessel. This involved reviews of available literature on release of superheated fluids, development of an analysis matrix for the potential release sizes and locations, and development of correlations for estimating the RRFs for differing break sizes including the effects of scalability of the test data. The methodology presented defines a 3x3 matrix of accidents for the release of superheated fluids from a heated vessel. This work extends the bases provided in the DOE Handbook for the evaluation of RRFs from superheated fluids.

DESIGN BASIS ACCIDENT SELECTION

Within the STP system design, the location of a flashing release accident is limited to the Corrosion Vessel Pressure Boundary (CVPB) as this is the only region that is both pressurized and heated. This boundary includes the corrosion vessel (CV) and connecting piping from the vessel wall penetration to the isolation valves in the pipe. The potential leak or failure points within the CVPB are essentially defined by three locations; 1) above the CV liquid level (vessel wall or connecting pipe), 2) at the CV liquid level interface (vessel wall only), and 3) below the CV liquid level (vessel wall or connecting pipe). The largest pipe penetration below the water line is a 1.5in (38.1mm) diameter pipe. The largest pipe penetration above the water line is a 1.5in (38.1mm) diameter pipe, excluding the non-pipe penetrations such as the agitator shaft penetration (>6 in) and other sealed access ports.

The hazard analysis examined the potential for uncontrolled releases during the corrosion process to identify the variety of potential releases for the purpose of selection a set of design basis accidents (DBA). Initially, a catastrophic rupture of the corrosion vessel at corrosion temperature and pressure was examined as the potential DBA for corrosion. However, as the accident analysis progressed, it became apparent that the phenomena associated a catastrophic rupture (a rapid depressurization of the system in less that 100 seconds) did not address potential phenomena associated with smaller failure and associated longer term releases.

By iterating with the accident analysis, the hazardous conditions for corrosion were eventually grouped under three DBAs:

1. Small Break: Small Leak below the liquid level (This release is treated as a flashing jet and is the bounding dose consequence.)
2. Medium Break: Double Ended Pipe Break below the liquid level (This release is treated as a flashing jet up to 1.5’’ in diameter)
3. Catastrophic Break: Catastrophic Vessel Rupture (This release is treated as a sudden/catastrophic failure in the vessel head space)

The fluid being released in each of these accidents consists of a slurry of water and sludge. The sludge, defined as any particulate which can pass through a ¼” strainer, consists of fuel corrosion products (including metallic uranium, and fission and activation products), small fuel fragments, iron and aluminum oxide, resin beads, concrete grit, sand, dirt, operational debris and biological debris. There are three “types” of sludge each with a different density and radioactive content, and the solids content of any particular corrosion batch is variable. The particle size distribution for the post corrosion slurry is not defined and one of the challenges was to identify an acceptable approach for quantifying the potential RRF for each of these DBAs.

RRF COMPUTATIONAL APPROACH

Summary of Analysis Approach

The RRF is a combination of the Airborne Release Fraction (ARF) and Respirable Fraction (RF) used in the common five factor release approach to compute respirable amount of material released (Q) (e.g., Q = MAR*DR*ARF*RF*LPF)[1]. The corrosion process will reduce the potential particulate size in the slurry. It was conservatively assumed that all post-corrosion particulates will be of a respirable size. Superheated liquid leaks that occur below the CV liquid level are bounded by the small flashing leak, and therefore only a small flashing leak was analyzed for flashing liquid jet releases. The intermediate or medium size breaks, up to a guillotine rupture of the largest pipe, is bounded by the small leak. In addition, the consequences appear to asymptotically approach the results of a catastrophic failure as the break size is increased. Therefore, the medium sized leaks were not explicitly evaluated. Leaks that occur in the vapor space are bounded by the catastrophic failure. However, this is a complete release that is virtually instantaneous and is applicable to a lower head failure or vessel side wall failure (that are catastrophic failures and not small leaks). It is argued that the RRF is dependent upon the orientation of the catastrophic break for the liquid depth normal to the release direction. The 3x3 matrix provided below that describes the potential break locations and analysis approaches used (Table 1) for each. Note, these discussions include STP design specific considerations not provided in detail here.

Table 1. Combinations of Break Sizes and Locations

	Below the Liquid level	At the Liquid level	Above the Liquid level
Small Break	This release is treated as a flashing jet and is the bounding flashing release. A small leak maximizes the RRF as both thermal and mechanical breakup mechanism can be important. The low volumetric release rate allows for minimum self-interference of respirable droplets material being released (droplet collisions and agglomeration). In addition, the low volumetric release rate allows the N2 sparge system to maintain system pressure, and heaters to maintain temperature, until all liquid contents are released.	This release is bounded by the small leak below the water level. The initial phase of the transient would be similar to a leak below the water line. However, as level decreases, it would reduce to an entrainment type release with lower RRF (see “Above the Liquid Level”).	This release would be treated as entrainment of droplets in the flowing offgas system. A small leak in the vapor space of the vessel leads to either a slow depressurization or no depressurization, depending upon leak size (normal offgas flow is >15 cfm). RRFs would be low as the dominant release mechanism would be surface bubbling creating aerosols with entrainment in the offgas flow. Surface bubbling would be mild and head space gas velocities would be low due to the slow depressurization.
Medium Break	This release is treated as a flashing jet. However, the	This release is bounded by the small leak below the water	This release would be treated as boiling release. The break

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[1] MAR: material at risk, DR: damage ratio, LPF: leak path factor

Table 1. Combinations of Break Sizes and Locations

	Below the Liquid level	At the Liquid level	Above the Liquid level
	flashing jet correlations were derived from data for 0.1-2 mm diameter orifices and appear to under predict the RRF for break sizes of 38.1 mm (1.5 in). Nagai et al. indicate an asymptotic behavior in the Sauter Mean Diameter (SMD) as superheat and orifice diameter increase, but there was insufficient date to adequately characterize the droplet distribution. Comparison against the small orifice correlations shows qualitative agreement that these larger leaks have a smaller RRF with deceasing trend as the leak diameter increases.	level. The initial phase of the transient would be similar to a leak below the water line. However, as level decreases, it would degenerate to an entrainment type release with lower ARF*RF (see "Above the Liquid Level").	size would be large enough to depressurize the corrosion vessel as the vapor release rate would greatly exceed the normal N2 supply. A 1.5 in diameter break would release ~1040 cfm of steam/N2. However, as the system depressurizes the release rate would follow the vapor generation/flashing rate of the liquid in the vessel as it cools (plus the N2 supply flow). Ultimately this would drop to only the N2 flow if no heat is added as the liquid cools to 100C, or with heat, the steam rate would balance the heat rate of the heaters. At 100 kW, the steaming rate would be ~0.05 kg/s (~177 cfm at 1atm).
Catastrophic Break	This release is a vessel lower head failure. While this may not appear to mimic the reference DOE-HDBK-3010-94 experiments for head space failures, this is simply an extension of a head space failure with a full vessel. However, this release would be an inherent 'downward release' that could be argued to impact a physical structure (i.e. the ground) that would act to reduce the respirable release fraction. Therefore, it is considered to be bounded by the head space failure (failure above the liquid level).	This release is a vessel wall failure that discharges sideways. While a radial unzipping may appear to be different than an axial failure, the release is still a catastrophic failure with liquid depth normal to the direction of the release (the vessel ID). The liquid is rapidly ejected from the side of the vessel forming potentially a fan shape instead of a cone or cylinder. However, the vessel geometry (90in ID, or 2.3m) is such that the diameter is larger than the liquid depth (~2m) assumed in the analysis and phenomena arguments suggest that the respirable release is a function of the liquid depth normal to the direction of release for a catastrophic failure.	This release is a vessel head space failure that mimics the data set from the reference DOE-HDBK-3010-94 experiments with a MAR and rupture size ~4-5 orders of magnitude larger. Plots show an inverse relationship between the MAR volume and RRF. This is attributed to the speed of the release and free-surface to volume ratio (inherent self-shielding) for the surface normal to the release direction. Rapid depressurization ejects all the material and creates extremely high particulate densities. As fluid flashes, the free surface has a large mean free path to the environment and respirable droplets easily escape. However, flashing liquid in the lower vessel volume is still surrounded by large quantities of liquid resulting in more collisions and agglomeration than for droplets created near the 'original' liquid surface.

STP System Design Considerations

Ambient pressure during operation is 1 atm, with a saturation temperature of 100ºC. The STP nominal operating process temperature is 185ºC, and the nominal process pressure is 225 psig. This process pressure corresponds to a saturation temperature of 203ºC. A reasonably bounding maximum process temperature of 203ºC (103ºC superheat) was defined for the accident analyses and is well above the thermal shatter criteria for water.

The potential size of a leak or failure is categorized as small, medium or catastrophic.

- Small leaks have been defined as on the order of <50-80mm^2 in area (<8-10mm equiv. diameter). At 225psig, an 80mm^2 rupture of a pipe below the vessel liquid level would blowdown at 30-50gpm (0.02-0.03m^3/s). This is 3-5.5kg/s depending upon the slurry density, 1.0-3.0gm/cc. This would take several hours to empty the vessel liquid contents. An 80 mm^2 rupture of a pipe above the vessel liquid level would release steam at ~73cfm (0.034m3/s) at ~8.3kg/m3. This is ~0.3kg/s and would take approximately 10x longer to remove the vessel contents (than the liquid release). At 73cfm of steam, this is a velocity of ~0.84cm/s in the vessel head space immediately above the liquid level (vertical flow in the 90in diameter vessel).
- Medium leaks are defined as leaks >8-10 mm diameter up to a guillotine rupture of the largest pipe penetration (a 1.5in diameter leak, or 38.1mm). At 225psig, a guillotine rupture of a 1.5in diameter pipe penetrating below the vessel liquid level would blowdown at 400-700gpm (liquid phase). This is 45-75kg/s depending upon the slurry density, 1.0-3.0gm/cc. This would take several minutes to empty the vessel. A guillotine rupture of a pipe above the vessel liquid level would release steam at ~1040cfm (0.5m3/s) at ~8.3kg/m3. This is ~4kg/s and would take approximately 10x longer to remove the vessel contents (than the liquid release). At 1040cfm of steam, this is velocity of ~0.12m/s in the gas head immediately above the liquid level (vertical flow in the 90in diameter vessel).
- A catastrophic failure is defined as a large break capable of releasing the vessel contents in a few seconds or less. For reasons specific to the STP accident analysis this is defined as a release <100 second (i.e. it was conservatively bounding to assume a catastrophic failure could take up to 100s to release the vessel contents). A 100s time frame for a liquid space release equates to ~1930mm^2 (~50mm / 1.95in equiv. diameter). A 100s time frame for a vapor space release equates to ~2.1*10^{4}mm^2 (~164mm / 6.5in equiv. diameter) for steam flow only.

Flashing Leak Characterization

A flashing release is defined as a release of superheated liquid above a critical temperature that results in shattering of the liquid due to vapor expansion (thermal shattering). It is the liquid breakup and bubble bursting at surfaces that create respirable droplets. This can be for either stagnant pools (e.g. a vessel) with sudden depressurization or a jet from a pressurized leak. Brown and York, and others, have identified that the critical temperature to initiate thermal shattering for water is ~10ºC superheat (this process is at ~100ºC superheat).

The characterization of flashing leaks is divided into 3 broad categories; small leaks (<8-10 mm in diameter), medium leaks (up to a guillotine rupture of 1.5” diameter pipe), and large leaks (catastrophic vessel failure). The flashing phenomenon itself is similar between the different leak sizes (i.e. thermal shattering). However, other phenomena can become more or less important depending upon the slurry density and both the leak size and location (such as mechanical shattering, self shielding, etc.). Slurry density is important because as a droplet evaporates after release it is possible for densification to occur, inherently limiting the RRF of droplets. This is discussed in more detail in a second paper (Schmitt 2007). For small leaks, both thermal and mechanical breakup can be important. This is seen in the York-Brown and Bushnell-Gooderum data where the RF is a function of superheat and orifice diameter. Their experiments included orifice diameters primarily in the range of 0.02-0.06 inches (0.5-1.5mm). Correlating their data estimates a range of SMD of ~15μm for a 2mm orifice to ~50μm for an 8mm orifice. This is consistent with the observations of Touil (2004)[2]. Touil (2004) reported on flashing leaks and included data from Nagai et al. (1985) that extended to leaks 10mm in diameter. As the nozzle diameter and superheat increased, Nagai reported that the SMD appeared to reach an asymptotic value, and the limiting SMD for water was ~36.8 μm. In addition, NUREG/CR-1607 reports an estimated range of droplet sizes for higher superheat (>300ºC superheat) that results in droplet sizes between 16 and 76 μm for the assumption of homogeneous and inhomogeneous bubble nucleation, respectively.

Unfortunately, the Nagai raw data could not be obtained and Touil did not include an evaluation of the droplet distribution which is needed to correlate the fraction of respirable sized droplets for larger diameter releases. Using a diameter of 38.1 mm (1.5 in), the small leak correlations give a SMD >200 for 100ºC superheat, nearly 3-6x larger than the NUREG/CR-1607 and Nagai data would indicate. The conclusion reached from this review is that for orifice (break) sizes larger than ~4mm the respirable release fraction is better represented by the Nagai limit.

A superheated jet will expand due to the vapor formation from depressurization of the jet. At high superheat the jet will expand quickly to pressure equilibrium. The expansion process will be a thermodynamic non-equilibrium expansion, but the expansion at pressure equilibrium can be estimated assuming an isentropic expansion (~17% vapor) or isenthalpic (~20% vapor). The dose consequence assumes isentropic expansion as this was more conservative (steam venting calculations assumed isenthalpic expansion). At the STP superheat conditions the isentropic vapor formation is ~17% (mass) and the expansion is approximately 200/1. The two-phase jet will continue to expand at roughly a 10 degree half angle (ANSI/ANS-58.2) due to air entrainment, maintaining roughly a cylindrical shape. As the jet diameter is increased, the volume to surface ratio increases for a cylindrical release. Thus, as the material released increases a larger fraction of the droplets created are located within the cylinder of the two-phase jet. This would result in more droplet collisions and agglomeration during the initial expansion and throughout the two-phase jet expansion. Evaporation and condensation effects are treated separately, but the initial droplet distribution would be expected to show an increase in effective

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[2] Touil reference provided by email communication with Bigot, Jean-Pierre, dated 3/6/2006, subject: background data for a paper, "Rain-out Investigation: Initial Droplet Size Measurement," by Jean-Pierre Bigot, Abdellah Touil et al.

droplet diameter (and SMD) with increasing jet diameter, resulting in a decrease in respirable fraction released.

Catastrophic flash releases are analyzed using experimental data from DOE-HDBK-3010-94 with a modification for scaling effects. A catastrophic flash release is essentially a rapid or sudden depressurization of a superheated, stagnant fluid (such as in vessel). The experiments used to define the RRF were performed in an 800mL vessel (NUREG/CR-4779). The vessel was a devise called the “pressurized-airborne release equipment” (PARE). In the experiments, as the volume of liquid was increased the RRF decreased by an apparent 1/volume relationship. However, after reviewing the performance the experiments the vessel diameter was held constant and only the depth of liquid (height) was varied in the experiments. Thus, only a 1/’liquid height’ relationship could be established.

The formation of droplets during flashing (thermal shattering) is a surface effect and occurs when steam bubbles burst or shatter the liquid surface. When a deep pool of superheated liquid is depressurized the entire contents rapidly flash and expand. At the superheat temperatures associated with the corrosion process (185ºC), it would be expected that a majority (if not all) of the material initially in the vessel would be ejected given a catastrophic vessel failure. Surface breakup and droplet formation near the original liquid surface, or near the ‘outer surface’ of the expanding /ejected flashing liquid can more easily escape to the environment than droplets formed internal to the flashing liquid mass. Surface breakup and the droplets formed in the interior of the flashing liquid are still surrounded by large quantities of liquid that did not flash, and are therefore more likely to collide/agglomerate with the surrounding liquid. For a fixed geometry (e.g. vessel diameter), as the depth of liquid is increased more of the droplets created in the interior of the flashing pool are removed (collide/agglomerate) due to the increase in total mass of non-flashed liquid (i.e. self-shielding), and a reduction in respirable fraction released.

A plot of the experimental data is provided in Figure 1, showing the inverse relationship between release fraction and initial volume. Statistical analysis of the experimental data for different liquids (density and viscosity) supports the inverse relationship. Figure 1 is a plot of the bounding RRF from the PARE experiments (circle) versus the correlation for RRF (solid line).

The RRF relationship is likely a function of the volume to surface ratio of the release (surface area normal to the release direction). For an initial cylindrical release shape (e.g. a vessel head failure), this degenerates to the depth of liquid. However, a release from the side of cylinder (a lateral unzipping of a vessel) would be expected to have a different dependency to the volume of liquid present. A bounding condition would be to assume a rectangular geometry where the opening is a rectangular shape. The volume to surface dependency would again be the depth of liquid, but for the lateral release of a cylindrical shape this would be the diameter of the vessel instead of the liquid depth. In this interpretation, a long narrow cylinder would have a larger release fraction than an equal volume cylinder that is shorter with a wider diameter. This would seem to be physically consistent. Qualitatively then, the geometry of a catastrophic failure that should yield the highest RRF is that geometry with the minimum liquid depth normal to the release direction. This discussion is only applicable to catastrophic, large releases. As the release size decreases it would reduce to a flashing jet release. This is physically consistent because as the release rate is slowed down, the residence time of the material released is short compared to when the remaining material in the vessel is released and not available to interact/collide with droplets from the remaining material. This then reduces to the jet release as the diameter of the release decreases as well.

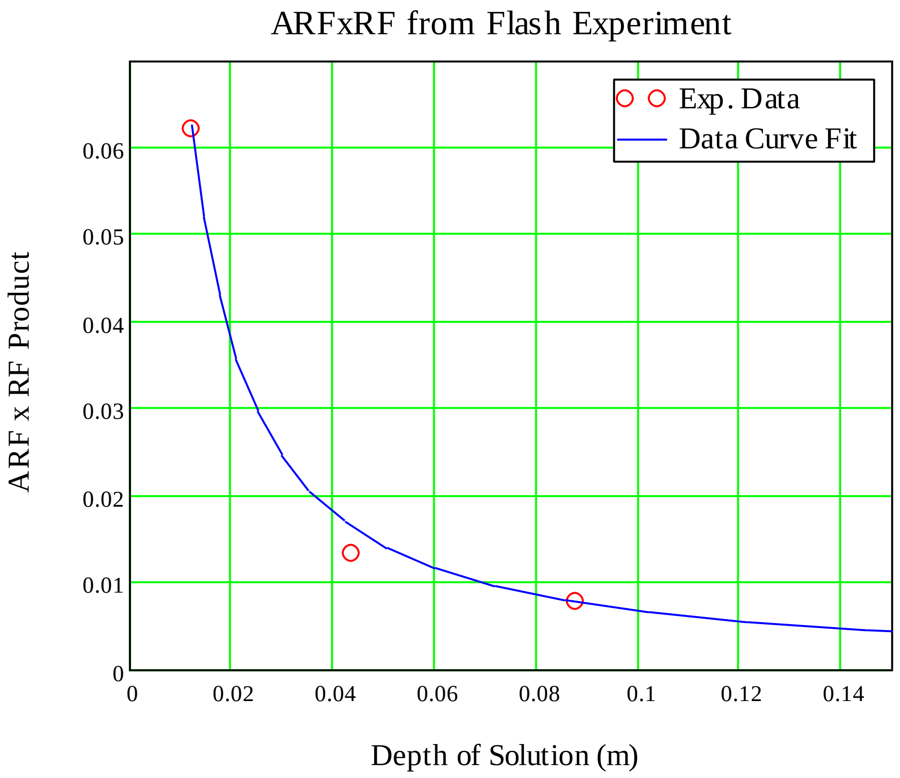

Figure 1. Catastrophic Flash RRF Scaling

The scaling of the experimental data is a concern. The corrosion vessel is 4-5 orders of magnitude larger than the experimental device. However, phenomenological arguments would support the conclusion that as more material is released greater self-shielding is inherent and the respirable release fraction should decrease. Statistical analysis of the experimental data for different liquids (density and viscosity) supports the inverse relationship. In addition, the plume dispersion modeling is considered very conservative as this release is not a true point source and the plume density would exceed the density of the air. A volume source could be credited and the high plume density near the source would allow for significant rainout to occur.

The medium break sizes were not explicitly evaluated for dose consequences. Based on increasing Sauter Mean Diameter (SMD) and decreasing RRF with break size, it is argued that dose consequences for the medium size breaks are bounded by the small leaks and quickly approach the release consequences from a catastrophic failure. A Mathcad spreadsheet was developed to plot qualitative comparisons. A plot of the RRF for different breaks size is shown in Figure 2. The RRF for small leaks is plotted for diameters of 2, 3 and 4mm. The medium break size RRF was estimated using the SMD reported from Nagai, SMD = 36.8 µm, and an assumed lognormal droplet distribution similar to the small leaks (“Nagai data”). The catastrophic RFF is plotted for the scaled RRF of the vessel (“Catastrophic”). Sludge properties for the STP were used in this comparison. This comparison is used to support the conclusion that the medium break sizes could be classified as the same dose consequence category as the large leaks and did not require explicit analysis.

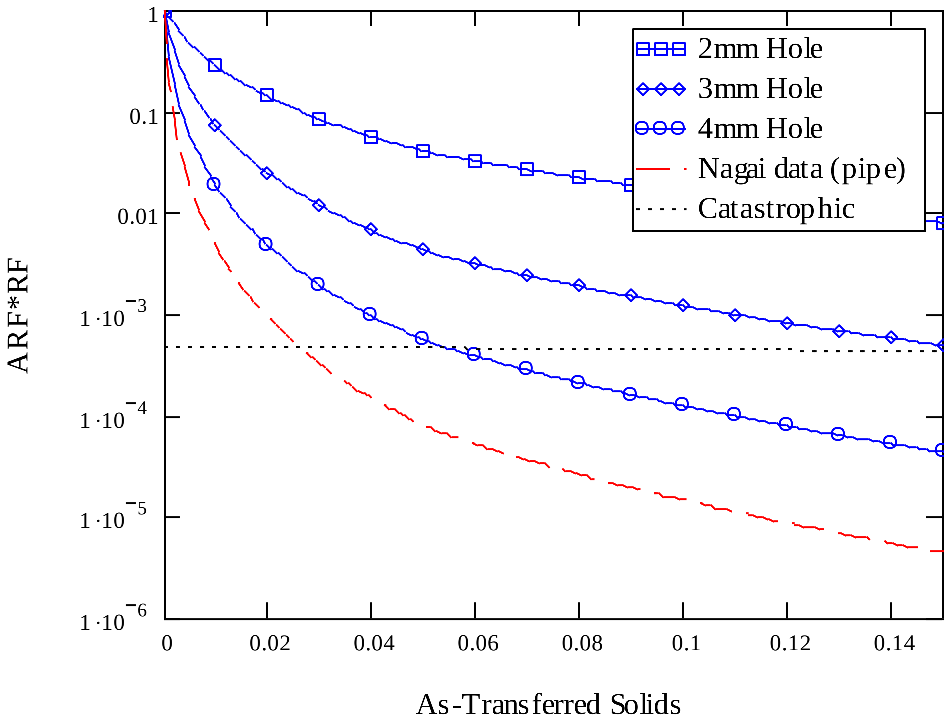

Figure 2. Comparison of RRF for Small and Large leaks

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