

ESTIMATING MAXIMUM DROPLET SIZE THAT CAN EVAPORATE TO A RESPIRABLE SIZE DROPLET

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ABSTRACT

The DOE Handbook, DOE-HDBK-3010-94, provides bases for evaluating respirable release fractions (RRFs) for accident releases of slurries and solutions. A slurry or solution release can create respirable-sized aerosol that is an inhalation hazard. By definition respirable aerosol has an aerodynamic equivalent diameter (AED) equal to or less than 10 μm , where an aerosol's AED is defined as the diameter of a spherical water droplet (density of 1 gram per milliliter) that has the same terminal free fall velocity in air as the aerosol droplet or particle. If evaporation is ignored, the respirable RRF includes only those droplets whose physical diameter is less than that corresponding to a 10- μm AED at the time of release. If evaporation is considered, then droplets larger than the AED criterion at the time of release can evaporate to a smaller physical size that may become respirable. Thus, the RRF of a release at the time it reaches the receptor may be larger than the RRF at the time of release. This paper presents a methodology for evaluating the effective RRF for evaporating droplets based on the safety analysis work done on the K Basin Closure Project at Hanford.

INTRODUCTION

The Hanford K Basin Closure Project involves the retrieval, transfer and processing of slurries and solutions containing partially corroded spent nuclear fuel from the K Basin spent fuel pools and dissolved contaminants such as cesium. The spent fuel is primarily metallic fuel from the operation of the Hanford reactors. The Sludge Treatment Project (STP) 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 large heated vessel to form a more stable metal oxide for packaging (grouting) and storage. Dissolved contaminants are treated with the slurries.

Slurries and solutions in the K Basin Closure Project contain radiologically hazardous solids or solute that become airborne under accident conditions. Accidental releases of these materials can create hazards from the inhalation of respirable aerosol generated by the release (in these accidents the liquid is water, not a hazardous material). Estimating the exposure consequence requires determination of the RRF (the amount of the release that is airborne **and** respirable). In many release types (spray, splash, flash, etc.), a distribution function can be derived to represent the droplet size distribution of the release. Such a function can be integrated to obtain the

fraction of respirable droplets; the required integration range is from zero to the droplet physical diameter that corresponds to a 10- μm AED. If evaporation is ignored, the maximum physical size of a respirable droplet is equal to that which has a 10- μm AED at the time of release. If evaporation is included the maximum initial physical size of a potentially-respirable droplet may be larger

In previous methodologies used at the Hanford Site for evaluating the RRF, a single large droplet size was selected based on conservative estimates of evaporation time and droplet rainout (HNF-8739). However, this did not account for densification of the droplet as it evaporated and was overly conservative for heavier slurries and solutions. Droplet density will increase if (1) the entrained solid or solute is denser than the carrier fluid, and (2) evaporation does not proceed so far as to produce a dry agglomerate particle with high void fraction. Droplet densification can counteract the effects of the decrease in diameter, thereby limiting the fraction of airborne respirable droplets.

Evaluation of the maximum initial physical droplet size that could become respirable was used to support evaluation of the dose consequences of potential accident releases of radiological material for the K Basin Closure Project. The droplet size evaluation was used in determining the RRF for evaluating the exposure consequence from inhalation of respirable droplets, and then used in the Preliminary Documented Safety Analysis for the safety classification and selection of controls for preventing or mitigating potential releases.

The methodology developed can be used to provide a simple assessment of the impact to the RRF for droplet evaporation. In the applied analyses this allowed the use of less restrictive assumptions without the need for complex modeling of the evaporation process. Note: this methodology was developed for accident releases in which the carrier fluid is itself not a hazardous material. If a volatile, hazardous carrier liquid or solvent is involved, then its vapor phase must also be considered.

DROPLET AERODYNAMIC EQUIVALENT DIAMETER

In many release types (spray, splash, flashing, etc.), a distribution function can be derived to represent the droplet size distribution. In spray releases, for example, Lefebvre (1989) identifies several common functions that describe the droplet size distribution. These include the log-normal, Nukiyama and Tanasawa, and Rosin-Rammler functions. In each case, the volume fraction of respirable droplets is the integral of the distribution function over the range of droplet diameters from zero to the physical diameter that has an AED of 10 μm , which is considered to be an upper-bound for respirability. An AED limit of 10 μm means that no droplet of unit-density material that has a physical diameter of more than 10 μm can be respirable. Because the AED of an aerosol depends on its density as well as its physical diameter, the maximum respirable physical diameter of an aerosol with non-unit density will be something other than 10 μm .

The density correction factor for the AED can be derived using the coefficient of drag (C_D) for a sphere. White (1991) provides a definition for C_D that is reasonably accurate for the case of particles that are above sub-micron size (where slip factors must be considered). The definition is given in Equation 1 and a force balance for the droplet falling at terminal velocity is given in Equation 2. The simultaneous solution of the equations yields the droplet velocity.

$$C_D = \frac{24}{\text{Re}} + \frac{6}{1 + \sqrt{\text{Re}}} + 0.4 \quad (1)$$

$$C_D V_{\text{droplet}}^2 = \frac{4}{3} D_{\text{droplet}} \frac{\rho_{\text{droplet}}}{\rho_{\text{air}}} g \quad (2)$$

where:

Re = droplet Reynolds number, $V_{\text{droplet}} D_{\text{droplet}} \rho_{\text{air}} / \mu_{\text{air}}$

D_{droplet} = droplet diameter

V_{droplet} = droplet velocity

ρ_{droplet} = droplet density

ρ_{air} = air density

g = acceleration due to gravity

A respirable particle is small enough to have its drag coefficient controlled predominantly by the first term in Equation 1. Holding all other particle properties the same, two particles of different density can only have the same fall velocity if the ratio of their diameters equals the inverse square-root of the ratio of their densities (substituting for Re and C_D in Equation 2).

The more general equation for accounting for droplet properties in calculating the physical diameter of the droplet that has a 10- μm AED--i.e., the maximum respirable particle--is given by:

$$d_{\text{resp}} = 10 \mu\text{m} \left[\frac{1}{\gamma_{\text{droplet}}} \right]^2 \left[\frac{C_v}{C_{\text{AED}}} \right] \frac{1}{S} \quad (3)$$

where:

d_{resp} = physical diameter of the maximum respirable droplet, μm

γ_{droplet} = droplet specific gravity

C_v = Cunningham slip factor, volume equivalent diameter

C_{AED} = Cunningham slip factor, AED diameter

S = shape factor for drag.

If the droplet/particle has a density different than water, a non-spherical shape, or can be shown to have significant slip (molecular slip), then Equation 3 allows calculation of the droplet diameter having the same settling velocity as the maximum respirable water droplet. In Equation 3, C_v/C_{AED} is the Cunningham slip ratio to correct for potential molecular slip. Above the sub-micron size range, all particles impact with air molecules and the ratio of both Cunningham slip factors can be ignored (i.e., $C_v/C_{\text{AED}} = 1.0$). The shape factor, S, is a measure of the change in drag coefficient for non-spherical shapes. For a spherical shape, $S = 1.0$. A spraying jet or splashing liquid shatters to stable droplet sizes based on mechanical and thermal effects and the resulting droplets would be expected to be spherical. Therefore, the shape factor is typically set to 1.0 for the droplets at the source. Evaporation is a concern further downstream from the droplet source and could result in naked solid particles. In this case, it is assumed that the dry particulate agglomerates in a spherical shape. This is a potential non-conservatism. However, the overall conservatism of this methodology is considered to minimize this potential impact. With these simplifications, the calculation of the physical diameter of the maximum respirable droplet reduces to the form below in Equation 4.

$$d_{resp} = 10 \mu m \left[\frac{1}{\gamma_{droplet}} \right]^{\frac{1}{2}} \quad (4)$$

This respirable limit definition is consistent with DOE-HDBK-3010-94, and defines the limit of integration for a droplet distribution function assuming no evaporation. From Equation 4, a heavy slurry or solution (density greater than 1 gram per milliliter) yields a droplet respirable limit of less than 10 μm at the source. If evaporation also occurs, then the droplet diameter decreases and it is possible for droplets initially too large (above the initial respirable limit) to become respirable. Existing methodologies used for the Hanford Site (HNF-8739) defined a single large droplet physical diameter (e.g. 30 μm) that could evaporate and become respirable. This was based on dilute solutions with relative low specific gravities (1.0-1.03). However, for heavier slurries and solutions this is unnecessarily conservative due to droplet densification and a more reasonable but still bounding limit can be defined based solely on the droplet characteristics (evaluation of evaporation rate and droplet rainout is not needed).

DROPLET EVAPORATION AND DENSIFICATION

The definition of a respirable droplet is one that has the same settling velocity (terminal free fall velocity in air) as a 10- μm diameter droplet of water (density of 1 gram per milliliter). As a droplet of solution or slurry evaporates, the droplet density will increase for solids and solutes with densities greater than the carrier liquid or solvent. Because both diameter and density vary during evaporation, the respirable limit diameter for the aerosol does not necessarily increase or decrease monotonically, but may go through a maximum partway through evaporation. It can be shown that one of three potential conditions provides a bound on the physical droplet size that can become respirable.

- the initial droplet respirable limit (considering no evaporation)
- the physical droplet size at the respirable limit that applies after complete evaporation to the equivalent volume diameter of the solids (or solute) assuming a packing fraction (the center is gas filled)
- the physical droplet size at the respirable limit that applies at the intermediate point in evaporation that maximizes the RRF for the aerosol

The derivations that follow are shown for slurry (solid particulates in a carrier of water). However, this can easily be applied to solution as well. In developing this methodology, three assumptions are made.

- The carrier liquid (or solvent) is not a volatile, hazardous liquid. If the liquid is volatile and hazardous, then additional consideration must be given to the vapor created in a release.
- During evaporation, the hazardous material remains in the droplets. Solids remain entrained in the droplet due to surface tension effects (solute remains entrained if it has a lower vapor pressure than the solvent).
- As the liquid evaporates, a maximum packing fraction of 0.64 is assumed for the solids (or solute as it precipitates). This is based on the maximum random packing fraction of monodisperse spheres. If further evaporation would reduce the droplet diameter below

the equivalent volume diameter of the solids (or solute) with this packing fraction, then it is assumed an air pocket is formed in the interstitial space of the agglomerate.

Prior to evaporation, the specific gravity of a droplet is given by Equation 5.

$$\gamma_{droplet} = \alpha_s \gamma_s + (1 - \alpha_s) \gamma_l \quad (5)$$

where:

- α_s = initial solid volume fraction in the droplet
- γ_s = solid specific gravity
- γ_l = liquid specific gravity

Combining Equations 4 and 5 provides the definition of the droplet respirable limit immediately after release at the source based on the solid fraction.

$$d_{resp} = 10 \mu m \left[\frac{1}{\alpha_s \gamma_s + (1 - \alpha_s) \gamma_l} \right]^{\frac{1}{2}} \quad (6)$$

To account for evaporation, the initial volume of solids and carrier liquid are needed. The volume of suspended solids and liquid in the initial droplet are given by:

$$Vol_{solid} = \frac{\pi}{6} \alpha_s D_{droplet}^3 \quad (7)$$

$$Vol_{liquid} = \frac{\pi}{6} (1 - \alpha_s) D_{droplet}^3 \quad (8)$$

where:

- $D_{droplet}$ = initial diameter of a droplet
- Vol_{solid} = initial volume of solids in a droplet
- Vol_{liquid} = initial volume of liquid in a droplet

After release, droplets begin to evaporate. If the fraction of carrier liquid that evaporates is 'x', then the droplet volume is given by Equation 9. The droplet specific gravity is given by Equation 10 (the denominator in Equation 8 accounts for the change in volume as the droplet evaporates). These equations are correct as long as the diameter of the evaporating droplet is greater than the equivalent-volume diameter of the total solids at the assumed 0.64 packing fraction. This diameter is referred to as the critical diameter ($D_{critical}$). If the droplet evaporates to where the diameter reaches the critical diameter, then the calculation of the respirable limit must be modified (this is done below).

$$Vol_{vapdrop} = \frac{\pi}{6} [\alpha_s + (1 - x)(1 - \alpha_s)] \alpha_s D_{droplet}^3 \quad (9)$$

$$\gamma_{vapdrop} = \frac{\alpha_s \gamma_s + (1 - x)(1 - \alpha_s) \gamma_l}{\alpha_s + (1 - x)(1 - \alpha_s)} \quad (10)$$

where:

- $Vol_{vapdrop}$ = volume of a droplet during the evaporation process
- $\gamma_{vapdrop}$ = specific gravity of a droplet during evaporation, if the diameter is greater than the critical diameter

Combining Equations 4, 9 and 10 provides the definition of the initial droplet physical diameter that, after partial evaporation, produces a maximum respirable droplet. The physical droplet volume can be equated to the maximum respirable droplet volume by combining Equations 4 and 9 to give Equation 11. Substituting Equation 10 for the specific gravity gives Equation 12.

$$Vol_{resp} = \frac{\pi}{6} d_{resp}^3 \quad (11)$$

$$\frac{\pi}{6} \left[10 \mu m \left(\frac{\alpha_s + (1-x)(1-\alpha_s)}{\alpha_s \gamma_s + (1-x)(1-\alpha_s) \gamma_l} \right)^{0.5} \right]^3 = \frac{\pi}{6} [\alpha_s + (1-x)(1-\alpha_s)] \alpha_s D_{droplet}^3 \quad (12)$$

Rearranging for $D_{droplet}$, Equation 13 is the initial physical droplet diameter at the respirable limit that exists after partial evaporation.

$$D_{droplet} = 10 \mu m \left(\frac{\alpha_s + (1-x)(1-\alpha_s)}{\alpha_s \gamma_s + (1-x)(1-\alpha_s) \gamma_l} \right)^{\frac{1}{2}} \frac{1}{\sqrt[3]{[\alpha_s + (1-x)(1-\alpha_s)]}} \quad (13)$$

This equation has a local maximum based on the fraction of liquid evaporated and the density of the solids and requires an additional condition to determine the maximum droplet diameter. The equation can be differentiated with respect to 'x' and set equal to 0. The resulting droplet maximum diameter can be shown to occur when the specific gravity of the droplet equals 3 times the specific gravity of the liquid (the complete derivation is not presented).

$$\frac{\partial D_{droplet}}{\partial x} = 0 \quad (14)$$

yields

$$\gamma_{vapdrop} = 3\gamma_{liquid} \quad (15)$$

If evaporation continues, the diameter of the droplet eventually reaches the critical diameter, where the equivalent-volume diameter is equal to the diameter of the solids plus some interstitial air. If complete evaporation is experienced the solids may form a semi-hollow shape filled with air. At this point a packing fraction is assumed and the non-solid volume is treated as a void (i.e. neglect the density of air as it is several orders of magnitude less than the solids). Given a packing fraction, ϕ , the final volume after complete evaporation is given by Equation 16.

$$Vol_{packing} = \frac{\pi}{6} \frac{\alpha_s}{\phi} D_{droplet}^3 \quad (16)$$

Again equating this volume to the maximum respirable droplet volume defines the initial physical droplet diameter at the respirable limit that exists after complete evaporation.

$$D_{droplet} = 10 \mu m \frac{1}{\alpha_s^{\frac{1}{3}} \phi^{\frac{1}{6}} \gamma_s^{\frac{1}{2}}} \quad (17)$$

Equations 6, 13, and 17 define the three initial droplet diameters that could potentially set the respirable limit. The maximum diameter of these three definitions is the bounding droplet diameter that could become respirable under any condition of evaporation.

ANALYSIS APPLICATION

To evaluate the potential benefit of this approach, two types of spray release are assumed, a cold spray and a flashing spray. A cold spray (sub-cooled liquid) is dominated by mechanical stresses and is typically characterized using the Rosin-Rammler (RR) distribution function, Equation 18. A flashing spray (super-heated liquid) is dominated by thermal shattering and is typically characterized using log-normal (LN) distribution function, Equation 19. Both Equations represent the volume fraction of droplets contained in drops less than diameter D (the respirable limit diameter of the release). For the purpose of this evaluation these functions can be considered equivalent to the RRF of the release (the airborne release fraction times the respirable fraction).

$$RRF = 1 - \exp\left(-\left(\frac{D}{X}\right)^q\right) \quad (18)$$

$$RRF = \frac{1}{2} \left(1 + \operatorname{erf} \left(\frac{\ln\left(\frac{D}{SMD}\right) - \frac{1}{2}\sigma^2}{\sqrt{2}\sigma} \right) \right) \quad (19)$$

In Equation 18, parameters q and X are constants where q is a measure of the droplet spread and X can be related to the Sauter Mean Diameter (SMD) to characterize the droplet size. The SMD is the ratio of volume to surface area for the droplet distribution. The SMD can be defined from measured data or empirical correlations. The following values are assumed for the Rosin-Rammler distribution function. These values are representative of a small diameter water jet at room temperature and several hundred psi above ambient.

$$\begin{aligned} q &= 2.4 \\ X &= SMD/0.65415 \\ SMD &= 200\mu\text{m} \end{aligned}$$

In Equation 19, parameters σ and SMD are constants where σ is the variance of the droplet diameter to the geometric mean and SMD is the Sauter Mean Diameter. Again, the SMD can be defined from measured data or empirical correlations and are typically different for the two releases assumed here. The following values are assumed for the log-normal distribution function. These values are representative of a small diameter flashing water jet with high super-heat ($\sim 100^\circ\text{C}$ above ambient saturation).

$$\begin{aligned} \sigma &= 0.457 \\ SMD &= 20\mu\text{m} \end{aligned}$$

Other parameters assumed for a comparison are:

$$\gamma_l = 1.0, \text{ carrier liquid specific gravity}$$

$\gamma_s = 9.6$, solid phase specific gravity
 $\phi = 0.64$, solid phase packing fraction
 $\alpha = 0 - 25$ vol%, potential range of initial solid fraction in droplet

The diameter D would be defined as the maximum diameter from Equations 6, 13, and 17. A spreadsheet was created using Mathcad to solve for the maximum diameter. The equations were evaluated at increments of 0.5% solid volume fraction from 0-25% and the calculated maximum diameter is plotted in Figure 1. Also shown are the reference 30- μm diameter limit and the liquid fraction remaining in the droplet for evaporation that yields the maximum diameter. Note: the solutions would tend toward infinity if the initial solids fraction were set to 0. To avoid a numerical discontinuity, the minimum initial solid fraction was limited to 0.1 volume percent (0.001) and results in a diameter of $\sim 36\mu\text{m}$ displayed at 0 solids fraction.

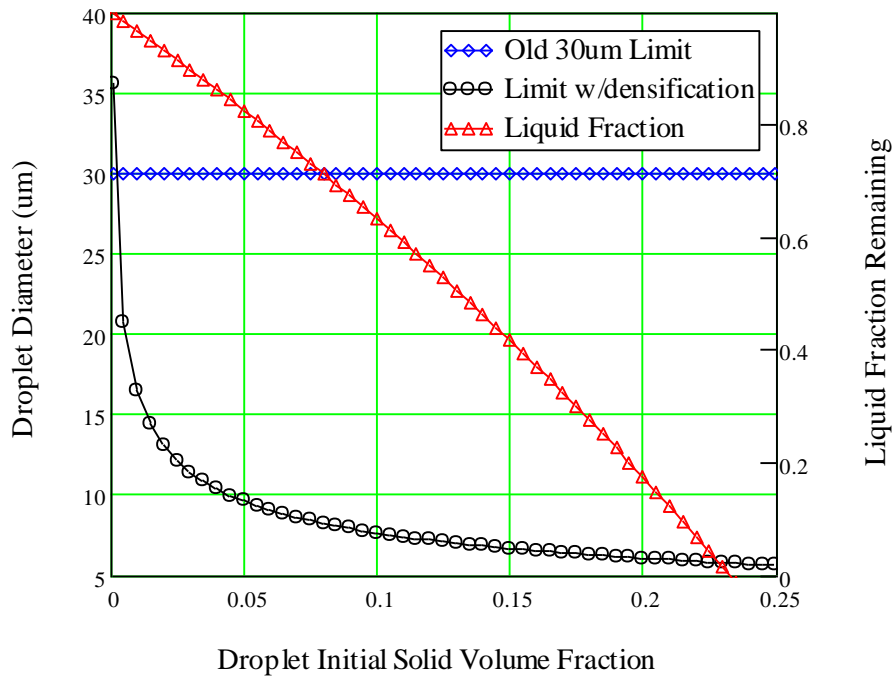


Figure 1. Maximum Droplet Diameter for AED with Densification

The results show that as the initial solids fraction in the droplets increases, the maximum droplet size that can evaporate to a respirable size decreases quickly. At 1% initial solids volume, and the solid phase specific gravity of 9.6 assumed here, the maximum droplet size that can become respirable is less than 17 μm . At 5% initial solids volume, the maximum droplet size is less than 10 μm . The existing methodology would have assumed 30 μm at both initial solids fractions. The optimal evaporation fraction is also plotted in Figure 1. As plotted, this is the liquid fraction remaining that defines the maximum droplet diameter that can evaporate to a respirable size. At a packing fraction of 0.64, the maximum naked particle that is respirable is $\sim 6 \mu\text{m}$. Although not specifically addressed here, sensitivity to the assumed packing fraction was small for values >0.5 for the slurry assumed. However, this would require evaluation on a case by case basis.

The RRF from Equations 18 and 19 are plotted in Figure 2. At the 30- μm diameter limit the cold spray (RR) and flashing spray (LN) RRFs are 3.8×10^{-3} and 0.75, respectively, regardless of the

solids fraction. These are considered conservative estimates for dilute solution, but are unnecessarily conservative for heavy slurry (or heavy solution). If droplet densification is included the RRF decreases to 9.0×10^{-4} and 0.26 at 1% initial solids, and to 2.5×10^{-4} and 0.03 at 5% initial solids, for cold and flashing sprays respectively.

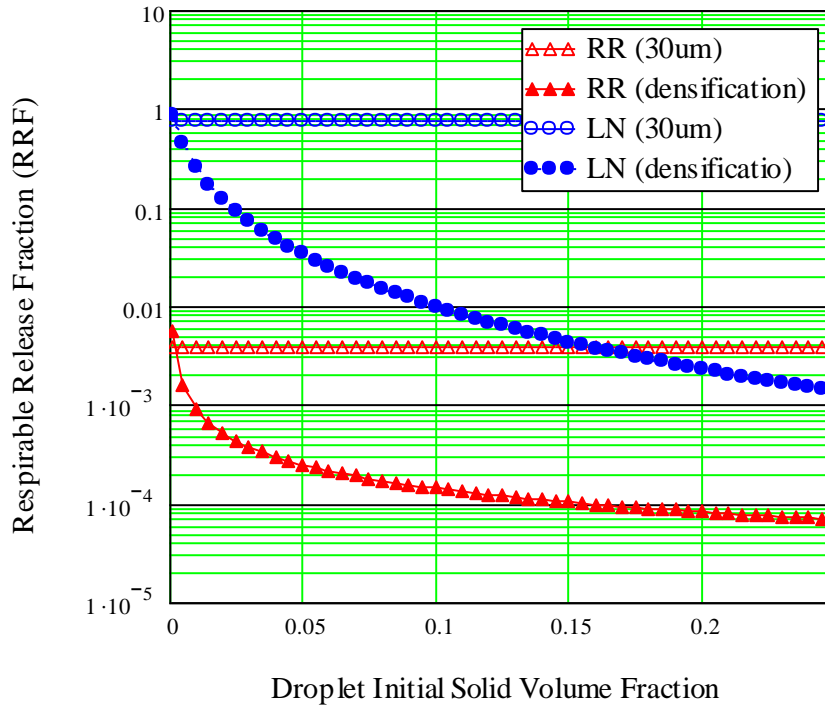


Figure 2. RRF Impact from Densification

The parameters that affect dose consequence are the RRF and unit liter dose (ULD), rem/liter. The ULD for the contaminated slurry increases proportional to the initial solids volume fraction. A relative impact to the dose consequence can be estimated by multiplying the relative change in solids content to the relative change in RRF, referenced at 1% solids.

$$DoseChange \approx \frac{\% solids}{1\%} \frac{RRF (\% solids)}{RRF (1\% solids)} \quad (20)$$

The results are plotted in Figure 3. Using the existing methodology for evaporation the dose consequence would increase linearly with solids content and at 25% solids the dose consequence would be predicted to be 25 times higher than at 1% solids (right side scale in Figure 3). However, correcting for densification of the droplets as they evaporate results in a self-limiting increase. Over the range of 1-25% solids, the Rosin-Rammler results show less than a factor 2 increase in dose, and the log-normal results in a local peak at 1-1.5% solids, and then decreasing dose thereafter (left hand scale in Figure 3).

The impact to the dose consequence is a function of the parameter values used to represent the two spray releases. However, this comparison illustrates the importance of crediting a physical phenomenon such as densification during droplet evaporation. In this example, an order of magnitude benefit is seen for crediting a physically real phenomenon. The key assumption

required were: 1) the release is a heavy slurry or solution, specific gravity >1.0, 2) solids remain entrained during evaporation due to droplet surface tension (or low vapor pressure for a solute), 3) a minimum packing fraction be defined for the potential case of complete dryout with formation of an interior vapor space, and 4) the carrier fluid itself was not a volatile hazardous material.

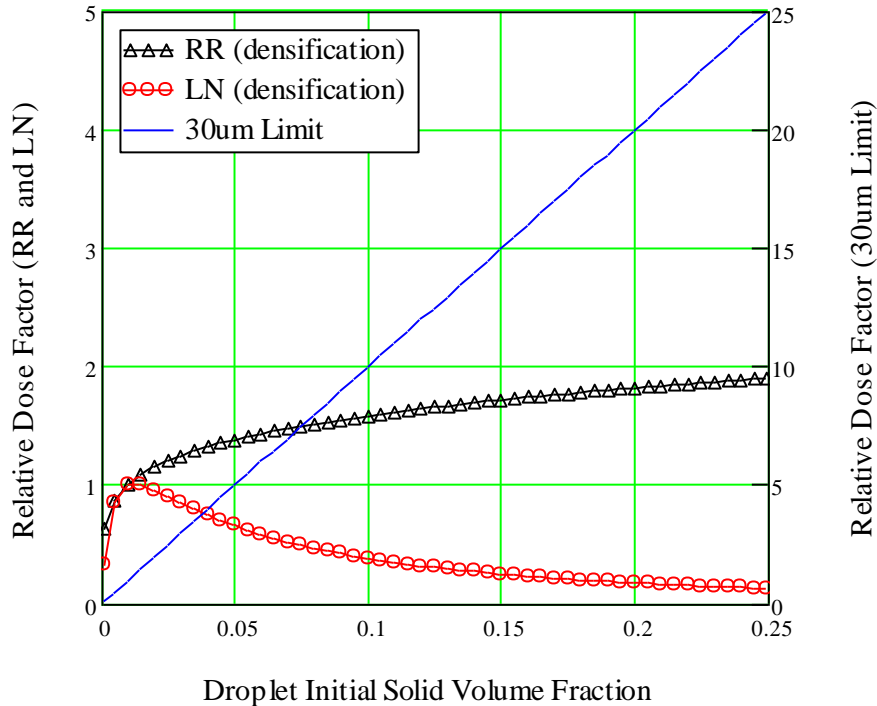


Figure 3. Potential Dose Impact with Densification

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