Framework for Estimating ARF and RF of Chloride Molten Salts in Pyroprocessing Based on DOE-HDBK-3010

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1. Introduction

Chloride-based molten salts are present as liquids in pyroprocessing operations such as oxide reduction/electrorefining salt handling. For safety analysis of normal and off-normal events (e.g., overheating, pressure transients, spills), the airborne source term is commonly parameterized by an airborne release fraction (ARF) and a respirable fraction (RF, typically particles with aerodynamic equivalent diameter ≤10 μm). DOE-HDBK-3010 [1] provides experimentally derived ARF/RF values for various nonreactor scenarios. This paper reorganizes the DOE-HDBK-3010 liquid data for aqueous solutions into three stress groups relevant to molten salts—thermal stress, explosive-type stress (shock/blast/pressurized venting), and free-fall spill—and proposes a practical framework to screen ARF/RF for chloride spent nuclear fuel (SNF) species in molten salt, together with an experimental test concept.

2. Stress Categories for Liquids in DOE-HDBK-3010

DOE-HDBK-3010 [1] treats aerosol release from liquids mainly as mechanical disintegration of the liquid surface followed by aerodynamic entrainment. For thermal stress, it distinguishes (i) heated liquids without continuous bubble breakup at the surface and (ii) boiling with persistent bubble rupture, and provides median and bounding ARFs for each case [2]. For pressurized venting of liquids, it classifies releases as venting below the liquid level, venting above the liquid level (or container rupture with a headspace), and depressurization of superheated liquids. These categories map to different atomization mechanisms such as spray/jet breakup, flashing spray, and splash/entrainment [3]. For free-fall spills, impact-induced splashing dominates, and ARF/RF depend on density (water-like vs concentrated heavy-metal solutions), viscosity, and slurry solids content [4].

Representative values used in this study are summarized in Table I. For thermal stress, heating without surface bubble breakup yields median ARF 6×10^{-7} and bounding ARF 3×10^{-5} (RF conservatively 1.0), while boiling yields median ARF 1×10^{-3} and bounding ARF 2×10^{-3} (RF 1.0) [2]. For pressurized venting, venting below the liquid level is bounded by ARF 1×10^{-4} (RF 1.0). For venting above the liquid level, a low-pressure case (≤0.35 MPa(g)) gives a bounding ARF 5×10^{-5} with RF 0.8, and higher-pressure cases show medians around 2×10^{-4}–3×10^{-4} depending on solution density. Depressurization of superheated liquids can increase ARF to the 10^{-2} range (e.g., bounding ARF 1×10^{-2}, RF 0.6 for ≤50°C superheat). For a 3 m free-fall spill, water-like solutions show median ARF 4×10^{-5} (RF 0.7) and bounding ARF 2×10^{-4} (RF 0.5), while high-density heavy-metal solutions are lower.

Table I: Summary of representative ARF/RF values for aqueous solutions in DOE-HDBK-3010

Condition	ARF (median /bounding)	RF (median /bounding)	Notes
Stress type			
Thermal stress			
Heated, non-boiling	6×10^{-7} / 3×10^{-5}	1.0 / 1.0	Surface heating without sustained bubble rupture; entrainment is typically limited.
Boiling (bubble rupture)	1×10^{-3} / 2×10^{-3}	1.0 / 1.0	Persistent bubble rupture drives surface disruption and droplet entrainment (boiling-driven aerosolization).
Explosive-type / Pressurized venting			
Pressurized venting below liquid level	— / 1×10^{-4}	— / 1.0	Spray/jet (mist/jet) through a breach
Venting above liquid level, low P (≤0.35 MPa(g))	— / 5×10^{-5}	— / 0.8	Droplet entrainment in depressurization flow; dissolved gas/turbulence/shear
Venting above liquid level, higher P(aqueous // heavy metal solution)	3×10^{-4} / 2×10^{-3} // 2×10^{-4} / 1×10^{-3}	0.9 / 1.0 // 0.3 / 0.4	
Depressurization of superheated liquid (≤50°C // 50–100°C superheat)	— / 1×10^{-2} // 2×10^{-2} / 1×10^{-2}	— / 0.6 // 0.6 / 0.7	Flashing spray can dominate
Free-fall spill			
3 m free-fall spill, water-like solutions (~1.0g/cm^3 // ~1.2g/cm^3)	4×10^{-5} / 2×10^{-4} // 1×10^{-6} / 2×10^{-5}	0.7 / 0.5 // 0.3 / 1.0	Uranine and UNH
3 m free-fall spill, slurries (<40% solids)	2×10^{-5} / 5×10^{-5}	0.7 / 0.8	TiO2 and glass frit slurry in uranine
3 m free-fall spill, high-viscosity solutions (>8 CP)	3×10^{-6} / 7×10^{-6}	0.8 / 0.8	Sucrose solution

3. Applicability to Chloride Molten Salts

DOE-HDBK-3010 provides a systematic, experiment-informed compilation of airborne release fractions (ARF) and respirable fractions (RF) for liquids under a range of stress categories relevant to nonreactor source-term evaluations, including thermal stress (heated non-boiling and boiling), explosive-type stress (e.g., pressurized venting), and free-fall spills. The value of DOE-HDBK-3010 for this study lies in its physics-based characterization of liquid aerosolization across these event classes—through surface disruption, jet/spray atomization, impact splashing, and aerodynamic entrainment—which can be mapped to analogous mechanisms in molten-salt systems. However, the underlying experimental basis in DOE-HDBK-3010 largely involves materials such as uranium nitrate hexahydrate (UNH) and other aqueous/solution systems, rather than chloride molten salts containing dissolved SNF species. This difference implies potentially significant deviations in operating temperature, thermo-physical properties (viscosity, surface tension, density), and the importance of species-dependent volatilization–condensation pathways. Accordingly, we adopt a staged strategy: we first use the DOE-HDBK-3010 stress-category framework (Sections 3.1–3.3) to screen ARF/RF ranges via a physics-based mapping to pyroprocessing molten-salt events, and then refine the parameters through targeted experiments that account for molten-salt properties and species-specific behavior.

3.1 Thermal Stress [2]

3.1.1. Heated, non-boiling liquids (no persistent bubble rupture at the surface)

DOE-HDBK-3010 assigns relatively low ARFs to heated liquids without persistent bubble rupture at the surface. In molten-salt systems, however, mechanical aerosolization can still occur without boiling due to surface agitation, inert-gas (Ar) purging, or process-gas flows. In addition, volatile chloride species may evaporate at high temperature and subsequently condense along thermal gradients in the headspace or off-gas lines, producing fine particulates via nucleation and condensation. Therefore, for non-boiling heating conditions, DOE-HDBK-3010 values can be applied as conservative baseline estimates for mechanical aerosolization, while volatilization–condensation contributions should be treated separately and quantified for molten-salt-specific conditions.

3.1.2. Boiling with persistent bubble rupture at the surface

Persistent bubble rupture repeatedly fragments the liquid surface and generally yields higher ARFs than non-boiling heating. In molten salts, bubble rupture may arise not only from thermal boiling but also from gas generation associated with electrochemical reactions or impurity ingress, producing similar surface disruption mechanisms. Because boiling can affect not only total release but also the particle size distribution (and thus RF), a conservative initial approach is to apply DOE-HDBK-3010 boiling values, followed by experiments that characterize bubble dynamics (pool vs localized boiling, gas flow conditions) and the resulting size distributions.

3.2 Explosive-type stress (pressurized venting) [3]

From an operational standpoint, pressurized venting is therefore a comparatively plausible initiating class. DOE-HDBK-3010 categorizes pressurized venting into (i) subsurface venting, (ii) venting above the liquid surface into a headspace, and (iii) depressurization of superheated liquids, with dominant mechanisms mapping to splashing/entrainment, jet/spray atomization, and flashing spray, respectively.

3.2.1 Subsurface venting (venting below the liquid level)

DOE-HDBK-3010 treats venting below the liquid level as spray/jet formation through a breach (from pinhole mist to large jets). For conservatism, the droplet size distribution is bounded using commercial spray-nozzle behavior, and bounding ARF/RF values are selected accordingly. For chloride molten salts in pyroprocessing, this mapping is relevant to overpressure relief or leak/breach events occurring within a contained molten-salt inventory.

3.2.2 Above-surface venting into a headspace

DOE-HDBK-3010 frames above-surface venting primarily as droplet entrainment in depressurization flow, influenced by (i) release of dissolved/trapped gases (bubble formation and fine droplets), (ii) surface turbulence, and (iii) stratified two-phase flow and shear near the liquid surface, with sensitivity to vent geometry and the distance to the liquid surface (freeboard). In pyroprocessing hot-cell operations, event definition should explicitly specify headspace volume, cover-gas conditions, and thermal gradients. Because molten salts are at elevated temperature, the analysis should also consider cooling and possible condensation of volatile chloride species in the headspace/off-gas path as an additional contribution distinct from mechanical entrainment/atomization.

3.2.3 Depressurization of superheated liquids

Depressurization of superheated liquid can produce flashing spray via rapid phase change, potentially increasing ARF substantially. In molten salts, saturation characteristics depend on salt composition and operating pressure; thus, the superheat definition and range should be explicitly stated, and depressurization rate and nucleation conditions should be included as event parameters. A conservative approach is to adopt DOE-HDBK-3010 flashing-related values for screening and then derive molten-salt-specific correlations through experiments that reproduce representative superheat levels and pressure-transient pathways.

3.3 Free-fall spill (3 m drop height) [4]

DOE-HDBK-3010 indicates that impact-driven splashing dominates aerosol generation for 3 m free-fall spills and reports different ARF/RF values for aqueous solutions, slurries, and viscous liquids. For molten salts, free-fall spill scenarios can similarly be governed by impact splashing; therefore, classification by rheology and solids content provides a defensible starting point.

3.3.1 Aqueous solution, 3 m (benchmark)

The aqueous-solution case serves as a benchmark for the "impact splashing–entrainment" mechanism and provides a reference to assess how molten-salt property differences (particularly viscosity and surface tension) affect ARF/RF.

3.3.2 Slurry, 3 m

Molten-salt systems may contain suspended solids, precipitates, corrosion products, or fine fuel particulates, making slurry-like behavior a realistic consideration. Because solids can influence atomization and size distributions, solids content (wt%) should be treated as a primary variable for evaluating and refining ARF/RF.

3.3.3 Viscous liquid, 3 m

Molten-salt viscosity can vary substantially with temperature and composition; increased viscosity (or partial freezing) may suppress droplet breakup and tend to reduce ARF. Therefore, viscous conditions should be defined over explicit temperature–composition ranges, and the applicability of DOE-HDBK-3010 values should be validated and corrected through experiments that account for temperature-dependent viscosity.

4. Source Term Experimental Concept

We propose a complementary test concept that couples INL MSTEC (Molten Salt Thermophysical Examination Capability) [5] with HFEF (Hot Fuel Examination Facility) infrastructure. MSTEC's strong instrumentation support can be used to screen temperature-dependent mass loss and evolved species for Oxide Reduction (OR)/ Electrolytic Reduction (ER) salts and fuel-bearing salts using simultaneous thermal analysis (STA) (optionally with in-line MS), and to identify key variables (temperature/superheat, pressure–depressurization path, gas flow) and capture/analysis requirements. A small-scale ARF/RF test fixture with off-gas and particulate capture is then used for repeated tests under representative thermal-stress and pressurized-venting conditions. Finally, HFEF's material and installed infrastructure (e.g., availability of SNF-bearing molten salts and existing remote-handling rigs) can be leveraged to expand conditions (overheating, depressurization/venting, and spills). Collected material is quantified by gravimetry, inductively coupled plasma-optical emission spectrometry (ICP-OES) and ICP-mass spectrometry (MS), with electron probe micro-analysis (EPMA) as needed.

5. Conclusions

DOE-HDBK-3010 provides stress-category-dependent ARF/RF ranges for liquids (thermal stress, pressurized venting, and free-fall spills), which are useful for initial screening of source terms for chloride molten-salt operations. Because the experimental basis is dominated by aqueous/solution systems (e.g., UNH), applications to SNF-bearing molten salts require refinement to account for thermo-physical differences and additional mechanisms such as volatilization–condensation and flashing. This paper outlines an MSTEC–HFEF integrated test concept to support a defensible ARF/RF database and reduce uncertainties in normal and off-normal source-term evaluations.

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