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Airborne Release Fraction of Nuclear Waste Surrogates

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Background

Airborne contaminants are a major concern for the design, transportation, and storage of hazardous nuclear waste materials. Actinide and other metal contaminants represent major health hazards. The safety basis analysts throughout the Department of Energy (DOE) complex rely heavily on information provided in the DOE Handbook, DOE-HDBK-3010, Airborne Release Fractions/ Rates and Respirable Fractions for Nonreactor Nuclear Facilities.

This study targets data from the DOE Handbook which outlines data sources for release fractions of nuclear waste contaminants in a fire. Much of the data in the DOE Handbook is from testing performed over 40 years ago. Also, there is a persistent question regarding the adequacy of common surrogates (e.g., cerium oxide or CeO_2) for hazardous nuclear wastes. This study seeks to reassess aerosol generation, aerosol release fraction, and respirable release fraction, for relevant fire release scenarios.

Drums of nuclear waste in a salt shaft at New Mexico's Waste Isolation Pilot Plant (Los Angeles Times, August 22, 2016).

Additionally, this study also seeks to assess the adequacy of historically tested surrogates to help provide confidence in applying contaminant release data from surrogates, and mockup experimental testing, to actual fires and accident scenarios at nuclear waste holding facilities.

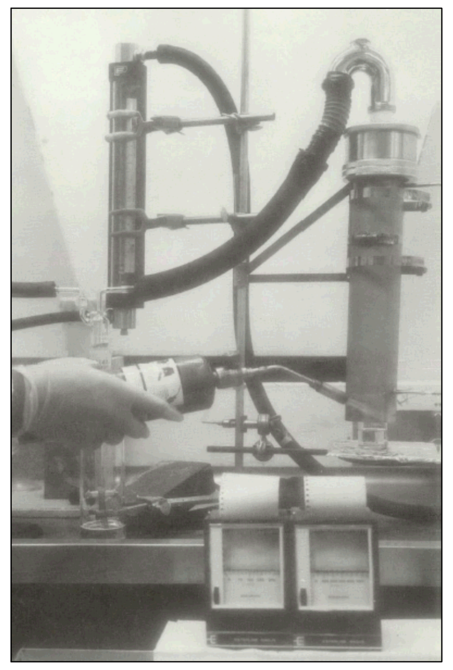


Image of original test apparatus taken from BNWL-B274

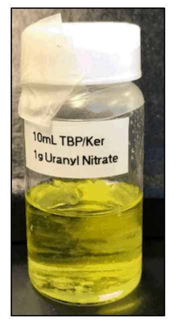
Our objective is to reassess the results documented in Mishima and Schwendiman (The Fractional Airborne Release of Dissolved Radioactive Materials During the Combustion of 30 Percent Normal Tributyl Phosphate in a Kerosene -Type Diluent, BNWL - B274, June 1973). The PUREX (Plutonium Uranium Redox EXtraction) process is used to separate actinides from contaminants. Organic solvents (e.g., tributyl phosphate, TBP) are used to separate the actinides from other contaminants.

Literature reports clearly demonstrate the coordination of TBP to the actinide metal nitrates, thereby enabling separation. TBP is commonly diluted to a concentration of 30% where the diluent is kerosene (referred to as TBP-kerosene). The solubility of uranium in 30% TBP-kerosene is reportedly ~120 g/L. The 30% TBP-kerosene solution, with actinides and trace metal contaminants, represents a flammable hazardous material generated during the solvent-solvent extraction of actinides. Mishima and Schwendiman initiated small beaker fires of the liquid. The effluent from the fire was captured and the release fraction of uranium was quantified. Release fractions were on the order of 0.03 to 0.3% by activity.

Surrogate Selection

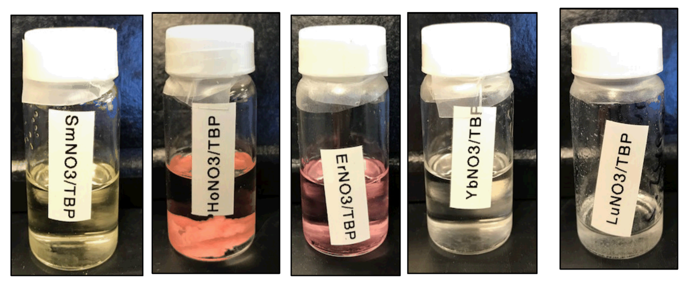
Studies with depleted uranium (d-U) would be ideal for predicting airborne release fractions from wastes containing plutonium and enriched uranium. However, concerns with the safety and additional management protocols in working with d-U, U, and Pu, makes finding non-radioactive surrogates attractive. Lanthanide materials are frequently presented as reasonable alternatives to the actinide cations. This is due to their similar size, electronics, densities, and additional inherent properties. Cerium is a common surrogate due to its similar structure and oxidation state. For aerosol transport, the density of the metal-oxide is an important parameter.

Metal	Assumed Oxide	Density (g/cm^3)	Solution (g/mL)	Notes
Ce	CeO_2	7.2	N/A	Common surrogate, not tested here
Ho	Ho_2O_3	8.1	N/A	Insoluble
Sm	Sm_2O_3	8.3	0.1	
Er	Er_2O_3	8.6	0.1	
Yb	Yb_2O_3	9.2	0.1	Better surrogate
Lu	Lu_2O_3	9.4	0.1	Better surrogate
Th	ThO_2	10.0	-	Not yet tested
U	UO_2	11.0	0.1	99.7% 238-U

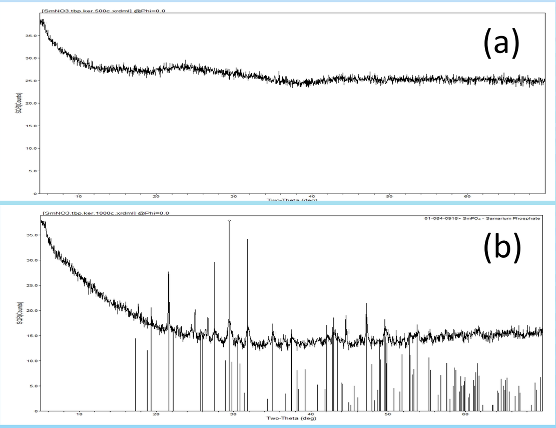


As can be discerned from the density of the oxides listed in the table above, CeO_2 may not be the best choice in surrogate for uranium and plutonium oxide. The heavier, late lanthanide cations (ytterbium and lutetium) are the most similar in terms of oxide density. Critically missing for this study, is the solubility of lanthanide nitrates in the TBP-kerosene solution. Shown to the left is the $U(NO_3)_3{\bullet}6H_2O$ solution dissolved recently at SNL.

A representative set of lanthanide (Ln) nitrate hydrates were selected for this study, with an emphasis on the later Ln cations. For this study, 1g of the sample was mixed in 10 mL of the 30% TBP-kerosene stock solution.



Solutions of $Ln(NO_3)_3{\bullet}6H_2O$ in the TBP-kerosene stock solution. Note insoluble material in Ho sample. Left to right: Sm, Ho, Er, Yb, Lu.

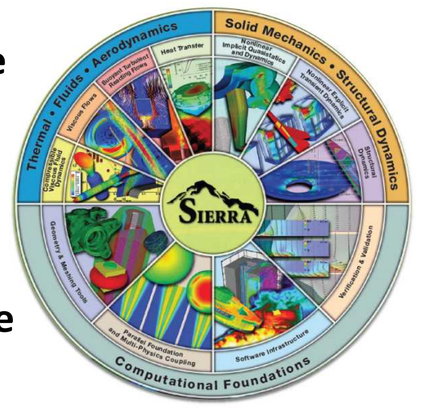


PXRD pattern of the pyrolyzed Sm solution at (a) 500 °C amorphous and (b) 1000 °C $Sm(PO_4)$.

Additional work has been done on characterizing the products generated from the pyrolysis of these solutions. For each sample, the solution was placed in a box furnace and heated to 1000 °C. The final material was collected and analyzed by powder X-ray diffraction. The sample proved to be samarium phosphate (PDF 01-084-0918). Studies on the d-U solution are underway.

Model Development

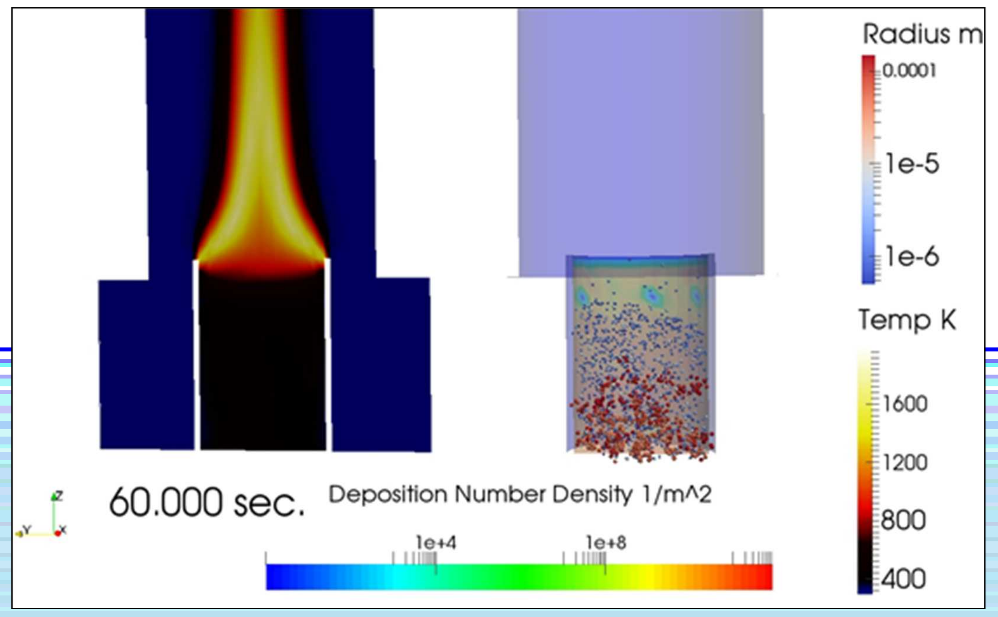
Models can be used to help bridge the gap between the tested surrogates and the hazardous material of interest. The planned experiments in this series will be modeled using SIERRA/Fuego, a low-Mach, fire CFD tool, part of the Thermal Fluids code suite. SIERRA/Fuego is a DOE model created and maintained at Sandia National Laboratories. All SIERRA tools are held to high quality control standards (DOE 414.1D), with nightly regression tests to ensure continuous functionality.



Fuego has been used previously to simulate the 1973 tests performed by Mishima and Schwendiman in support of an assessment of DOE-HDBK-3010[1-4]. Fuego's Lagrangian-Eulerian particle model was used to track the aerosolized contaminant-TBP- kerosene solution due to surface boiling.

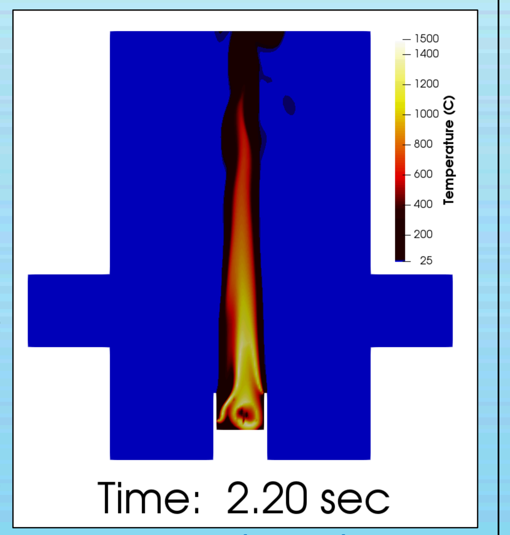
Particle deposition and contaminant concentration were tracked on all surfaces and outflows of the simulated domain. The contaminant that passed through the outflow boundary was considered airborne, and counted towards the total airborne release fraction (ARF). Using this model, a parameter study indicated that the model was very sensitive to variations in initial liquid level, a feature not explored in any experiments.

Leveraging the simulations done in previous years, the models have aided in test design through pre-test simulations which have helped size the chimney to manage the temperature of the gas at the outflow (cellulose filter). Further simulations will be performed in support of the experimental measurements with the intent of using the models to help interpret the experiments with surrogate materials considering the actual materials of interest.



Previous work: beaker fire contaminant modeling[1]

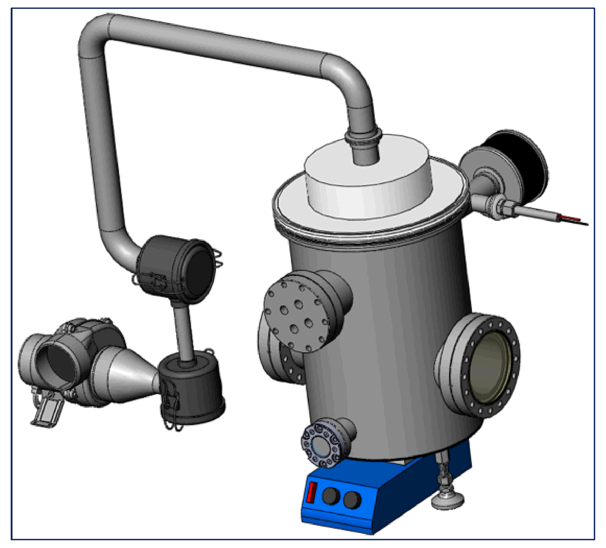
The modeling effort tied to this test series will simulate the various surrogate contaminants and directly compare to experimental observations. Test comparisons will provide model confidence through application of validation techniques. The validated model will be used for non-surrogate assessments to aid in understanding the performance of real hazards.



Current work: preliminary simulation results

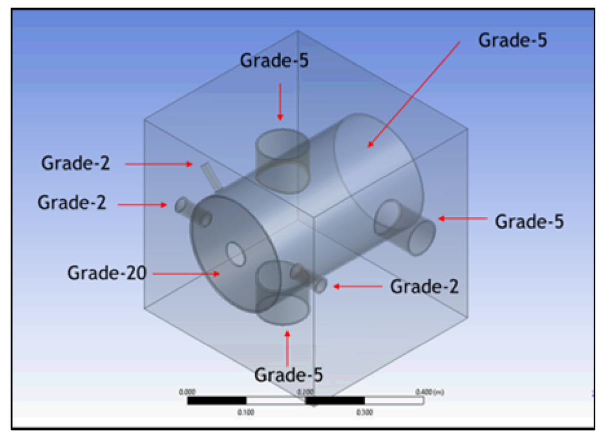
Test Chamber

A custom chamber was designed for this test series to enable in-situ optical diagnostics, extraction aerosol sampling, and total particulate filter collection. The main chamber is approximately 8 inches in diameter and 14 inches tall. A small beaker containing 10-20 milliliters of TBP-Kerosene-Metal solution is ignited at the bottom center of the chamber.



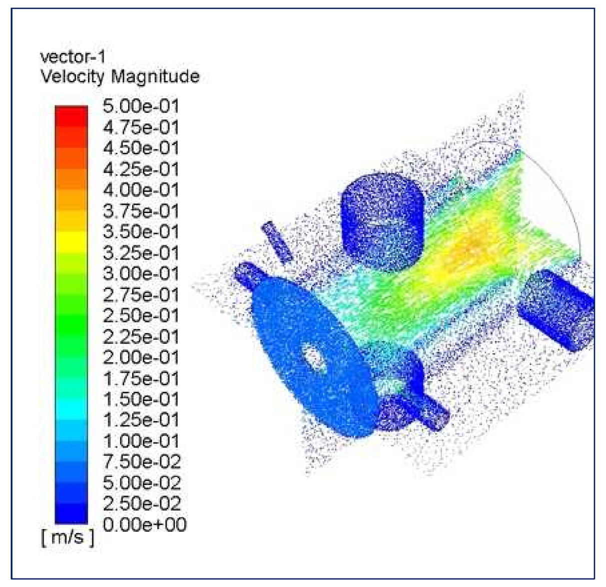
Solid model of custom chamber utilized to capture aerosolized materials from fires

Optical access ports on the sides of the chamber enable Mie Scattering measurements of particle size distributions, and optical spectroscopy. Quantitative grade cellulose filters capture 98% of the aerosol at the top of the chamber. Filters are then digested in acid and analyzed with ICP-MS.



CFD geometry used to study air flow within the chamber and optimize porous material selection

The inner surfaces of the chamber are porous stainless steel. This provides a source of uniform dilution air to cool the combustion products and keep materials from depositing on the inside of the chamber rather than being captured on the filter. Computational Fluid Dynamics (CFD) simulations were utilized to design the porosity of the metal and obtain more optimal air flow patterns.



CFD simulation results showing air velocity magnitude within the chamber

The inner porous sleeve has been welded and the test apparatus will be completed in FY18. Of the lanthanide series, lutetium and ytterbium nitrates were soluble in TBP-Kerosene and have oxides with densities more similar to uranium and plutonium oxides than traditional surrogates. We plan to conduct four airborne release fraction experiments in triplicate: lutetium, ytterbium, thorium, and depleted uranium.

Porous metal sleeve fabricated to introduce dilution/shaping air to capture particulates more effectively

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Acknowledgements

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