Sandia National Laboratories

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Airborne Release Fraction (ARF) of Nuclear Waste Surrogates

Joshua Hubbard, Timothy Boyle, Ethan Zepper, Alexander Brown, Joshua Santarpia, Gabriel Lucero, Taylor Settecerri, Joseph Zigmond, Fernando Guerrero, and Xavier Robinson

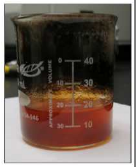
Test Setup

An aerosol chamber was designed to capture particulate from fires containing nuclear waste surrogates, thereby enabling studies of Airborne Release Fractions (ARF). The inner chamber surface is made of porous stainless steel (Mott). The outer shell serves as a plenum and air flows up around the fire, sweeping effluent up through a 185 mm filter at the top of the chamber. This chamber is unique because air flow is perpendicular to the internal surfaces of the chamber, thereby minimizing, and possibly eliminating, wall deposition. Filters were selected from cellulose (Whatman 41) and glass fiber (Whatman EPM-2000) to facilitate chemical analysis of effluents (acid digestion, or leaching, and inductively coupled plasma mass spectrometry). Each filter has published values for aerosol collection efficiency. Optical access is provided through several flange mounted windows. After the aerosol collection filter, air is passed through a stainless steel filter, HEPA filter, and carbon filter, before it is exhausted into a fume hood.

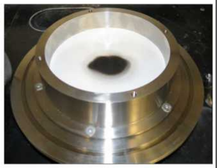
For preliminary experiments, aerosols were extracted from the chamber through a 0.25 inch diameter sampling probe and characterized with TSI aerosol measurement instrumentation: Laser Aerosol Spectrometer (LAS; 90-7000 nm), Scanning Mobility Particle Sizer (SMPS; 10-1000 nm), and Aerodynamic Particle Sizer (APS; 500-10000 nm). The LAS, SMPS, and APS size particles based on light scattering, electrical mobility, and fluid mechanical mobility. A 47 mm filter sample was also taken from the aerosol sampling probe onto glass fiber filters for analysis with Scanning Electron Microscopy (SEM).

No Vigorous Boiloff

Three materials were burned in the SNL test chamber to characterize and validate chamber performance: 30% tributyl phosphate (TBP) diluted in kerosene, 0.1 g/mL lutetium nitrate in 30% TBP/kerosene, and 0.1 g/mL ytterbium nitrate in 30% TBP/kerosene. Lutetium nitrate and ytterbium nitrate serve as surrogates for uranium and plutonium compounds present in PUREX waste. These two metals were selected because their respective oxides have densities more similar to uranium and plutonium as compared to other commonly used surrogates like cerium.



20 mL of surrogate waste was placed in a 50 mL beaker and inserted into the chamber. A hot plate, set to 400°C was turned on for two minutes to heat the kerosene mixture. Upon visual observation of vapors, a butane lighter was inserted into the chamber through a side port, and used to ignite the vapors. Chamber air flow was turned on after ignition, and ramped from 15-25 cfm over 3 minutes. The experiments were performed to self-extinguishment (no heating after initiation of flaming combustion) similar to section 3.3.1 of DOE-HDBK-3010-94 (no vigorous boiloff). Two tests were performed with TBP/kerosene, and three tests were performed for both lutetium nitrate and ytterbium nitrate in solution.



Flame heights were nominally one to two inches, and cavity effects (flame recessed inside the beaker) likely caused the fire to self-extinguish before the fuel was totally consumed. Ytterbium fires were distinguishably shorter than lutetium fires (10-20 minutes vs. 40-60 minutes), which suggests the surrogate metal may have significant effects on fire characteristics and ARF. The chamber air flow concentrated the effluent in the middle of the 185 mm cellulose filter, which was the intent of the design. Only small amounts of the liquid were lost during experiments (2-4 mL). ARF values published by Mishima and Schwendimann (June 1973) were on the order of 2.3E-4 – 2.7E-4 for uranium. Air flow rates used in this study were scaled to provide the same updraft velocity as the original 1973 experiments.

Images: (from top to bottom) beaker flame for no vigorous boiloff conditions, residual beaker waste after self-extinguishment, 47 mm filter sample, 185 mm filter showing effluent is concentrated in the central region of the chamber (away from the walls)

Vigorous Boiloff

In two separate tests, the hot plate was left on to heat the kerosene mixture after the onset of combustion. Under these conditions, vigorous boiling could be observed in the beaker where in previous tests it was not observed. Flame heights grew rapidly to the point they reached the aerosol collection filter (6-8 inches in height). Experiments were terminated at this point.

In one experiment containing lutetium nitrate (images to the right), we allowed the beaker and chamber to cool, and then relit the mixture without heating from the hot plate. The flame burned with a much lower flame height similar to previous experiments for no vigorous boiloff. The lutetium nitrate fire described here also produced an inordinate amount of soot compared to the other tests without vigorous boiloff. For this test, the high soot loading reduced the airflow to 15 cfm rather than 25 cfm in previous tests.

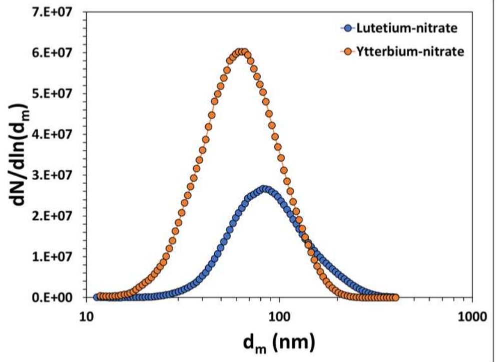
It is unknown if the high temperatures resulted in chemical transformations in the waste material which could have led to a more sooty flame. The residual (unburned waste) had much less liquid, and appeared more solid/gelatinous. AML will analyze post-combustion residues.

These tests conducted under vigorous boiling illustrate that vaporization rates have a substantial impact on flame height, and may result in chemical transformations that generate more soot, and possibly higher ARFs. The aim of our current work is to reproduce results from no vigorous boiloff conditions. In the future, to operate our chamber at vigorous boiloff conditions, we would use glass fiber filters rather than cellulose filters, and propose to add a 12" extension to the top of the chamber to reduce the possibility of damaging the filter with heat or flame.

Images: (from top to bottom) beaker flame for vigorous boiloff conditions, residual waste after self-extinguishment showing a more solid/gelatinous form compared to no vigorous boiloff, 47 mm showing high soot concentration, and 185 mm filter showing significant soot loading which reduced the air flow rate to 15 cfm

Particulate Analyses

Aerosol distributions observed with the LAS and SMPS showed high concentrations of particles smaller than 200 nm. Representative data are shown below for experiments with lutetium-nitrate and ytterbium-nitrate. We will attempt to corroborate these data with SEM images from 47mm filter samples. We will also compare SEM from 185 mm and 47 mm filter samples to determine what sampling and transport losses may have occurred between the test chamber and aerosol measurement instrumentation. Generally speaking, soot aerosols aggregate to form larger particles with tenuous, fractal, structures. The presence of low-volatility materials like TBP and metals could alter particle sizes and morphology appreciably.



Methods used in preliminary experiments, shown here, are referred to as extractive aerosol sampling. This requires that a sample be drawn from the chamber and transported to aerosol measurement instruments. Combustion aerosols present unique challenges due to the temperature of the gas, condensation of volatile species, and the possibility of thermophoretic losses. Particles outside the respirable range (>10 µm) generally are less easily sampled and transported. Thus, we used extractive sampling to better understand aerosol characteristics but we do not regard these measurements as quantitative in terms of characterizing ARF values. In upcoming ARF tests, the aerosol sampling probe will be removed. All aerosol will be captured by the 185 mm collection filter.

Future Work

SEM will be used to analyze soot samples from experiments described here. SEM data will be used to corroborate aerosol measurements from LAS and SMPS instruments.

ARF's will be measured for fires (no vigorous boiloff) in triplicate for the following mixtures:
(1) 0.1 g/mL ytterbium nitrate in 30% TBP/kerosene,
(2) 0.1 g/mL lutetium nitrate in 30% TBP/kerosene, and
(3) 0.1 g/mL depleted uranium nitrate in 30% TBP/kerosene.

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Joshua A. Hubbard, Ph.D.
jahubba@sandia.gov
(505) 844-0879