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Isolation of Metals from Liquid Wastes: Reactive Scavenging in Turbulent Thermal Reactors

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Research Objective

The objective of this project is to develop the fundamental science base necessary to assess the utility of high-temperature processes to volatilize metals in DOE metal-bearing liquid wastes, so that they can be reactively scavenged by sorbents. The problem is addressed through a collaborative research program involving a team of five senior scientists and their respective laboratories, at four institutions.

Specific goals are to:

1.) Understand high-temperature reaction kinetics between sorbent substrates and certain volatile and semi-volatile metals in the DOE liquid waste inventory (e.g., Cs and Sr), using a laminar-flow reactor for which extraction of kinetic data is not complicated by turbulence;
2.) Develop models to predict both trajectories of individual droplets in turbulent high-temperature reactors, and rates of metal evolution from droplets, and compare model predictions with experimental data from a pilot-scale turbulent thermal reactor;
3.) Connect the reaction kinetic models with the droplet trajectory/mass evolution models, in order to predict and optimize metal scavenging processes in turbulent-flow reactors, and to test these combined models against data taken from a turbulent high temperature reactor.

Research Progress and Implications

This report summarizes work at a point midway through the first year of a 3-year project.

At the University of Arizona (UA), two tasks are underway. The first task is concerned with attempting to understand high-temperature reaction kinetics between sorbent substrates and certain volatile and semi-volatile metals. The second task is concerned with applying Kerstein's One Dimensional Turbulence model to prediction of droplet trajectories in turbulent flow.

A UA doctoral student has commenced working on the first task. The high temperature laminar-flow reactor has been refurbished and is now available for screening tests. Nonradioactive isotopes of cesium and strontium were chosen as representative metals of interest to DOE. Kaolinite was chosen as a representative sorbent, since its interactions with alkali metals such as sodium and potassium have already been studied in this laboratory. The metals are introduced as aqueous solutions of cesium acetate and strontium nitrate and the metal-containing solution is then sprayed through a natural gas flame. The metal vaporizes, and then either reacts with sorbent substrates or nucleates and condenses further downstream. For the screening tests, which are about to commence, particulate samples are isokinetically withdrawn at the end of the reactor, and passed through a Berner low pressure impactor for size fractionation into multiple sub-micron and super micron aerodynamic size classes. The size-segregated distribution of cesium or strontium then allows the extent of scavenging by sorbent to be inferred.

We have conducted a multi-component equilibrium analysis on the partitioning of cesium and strontium at high temperatures. Prediction of dewpoints is important, since only the metal vapors will react with sorbents. Taken from the most recently updated thermodynamic data available, the strontium compounds considered were: Sr, SrCl, $SrCl_2$, SrO, SrOH, $Sr(OH)_2$, SrS, Sr(s), Sr(l), $SrCl_2$(s),

$SrCl_2$(l), SrO(s), SrO(l), $Sr(OH)_2$(s), $Sr(OH)_2$(l), SrS(s). The cesium compounds considered were: Cs, CsCl, CsO, CsOH, Cs_2, Cs_2Cl_2, Cs_2O, $Cs_2(OH)_2$, Cs_2SO_4, Cs(s), Cs(l), CsCl(s), CsCl(l),CsOH(s), CsOH(l), Cs_2SO_4(s), Cs_2SO_4(l). In the absence of chlorine, 100 ppm strontium began to condense at 2000K, with condensation complete at 1500K. Addition of 2500 ppm chlorine reduced the dewpoint to 1410K. Hence, chlorine addition may be required to facilitate strontium capture by sorbents, a procedure which has been shown to be effective for the scavenging of nickel, which is a similarly refractory metal. Sulfur had little effect on strontium dewpoints. 100 ppm of cesium began to condense at a very low temperature of 700 K. Although chlorine and sulfur both increased the condensation dewpoints to 900K and 1300K respectively, these predictions suggest that cesium would be an excellent candidate for sorbent scavenging.

A laser diagnostic system, to allow in-situ measurement of solid, droplet, and vapor metal species has been designed by the University of Illinois (UI) for use with the UA laminar flow reactor. Optical components and arrangements have been specified which will provide illumination sheets 5 cm across and less than 100 microns thick for probing the spatial distribution of droplets, soot, OH radical, free aluminum atoms, and free copper atoms. Additional components also allow for laser-induced breakdown at a fixed position within this sheet. Design is nearly complete and fabrication of the system has started. Work has begun on designing modifications to the reactor, to allow optical access and to ensure safe and secure operation. A soon to-be-appointed post-doctoral research associate, to be supervised by UI, will be responsible for the operation of the laser and related spectroscopic apparatus, and the extraction of data from the UA reactor. Two meetings, one at UA and one at UI, have enabled this task to advance. A third meeting at EPA in North Carolina helped ensure that the design of system to be used at UA is also compatible with the larger turbulent flame reactor at EPA.

A meeting between Dr Kerstein and Professor Wendt in October initiated modeling activities for the second task. A graduate student at UA is now collaborating with personnel at Sandia National Laboratories (Livermore) and will spend six weeks this summer working with Dr. Kerstein and his team there. Efforts hitherto have focussed on becoming familiar with Kerstein's One Dimensional Turbulence Model and with using the UA mini supercomputer cluster.

Computational work at Illinois focuses on the development of models for computing the trajectories of evaporating drops over a wide range of initial sizes, including drops large enough for deformation and inertial effects to be important. To that end, we have extended our earlier computations of axisymmetric drop motion to the case where the drop density is much larger than that of the surrounding medium, and found that elongated prolate drops can occur in this case. Work is underway to account for drop acceleration and heat transfer in the finite-element code.

Planned Activities

The UA screening tests for scavenging of cesium and strontium by kaolinite should be complete by the end of the first year. Fabrication of the optical diagnostic system should also be one third complete at the end of the first year. Preliminary calculations using the One Dimensional Turbulence Model to predict droplet trajectories in turbulent flows should be well underway by the end of the Summer. Modeling work will continue throughout the duration of the project.