

Data from Web Upload:

Project ID: **60326**

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

Lead Principal Investigator:

Dr. Jost O.L. Wendt
Professor and Head
Department of Chemical and Environmental Engineering
University of Arizona
P.O.Box 210011
J.W. Harshbarger Building
North Campus Drive
Tucson, Arizona 85721-0011
Telephone: 520-621-2591
e-mail: wendt@u.arizona.edu

Co Principal Investigators:

Dr. William P. Linak
National Risk Management Research Laboratory (NRMRL)
Environmental Protection Agency
Air Pollution Prevention and Control Division (APPCD)
Air Pollution Technology Branch
MD-65
Research Triangle Park North Carolina 27711
Telephone: 919-541-5792
e-mail: LINAK.BILL@EPA.GOV

Dr. Alan R. Kerstein
DOE Combustion Research Facility
Sandia National Laboratories - California
MS 9051
Livermore California 94551
Telephone: 510-294-2390
e-mail: ARKERST@sandia.gov

Dr. Arne J Pearlstein
Associate Professor
Department of Mechanical and Industrial Engineering
University of Illinois at Urbana-Champaign
Urbana Illinois 61801
Telephone: 217-333-3658
e-mail: arne@ajpiris.me.uiuc.edu

Dr. Alexander Scheeline
Professor
Department of Chemistry
University of Illinois at Urbana-Champaign
Urbana Illinois 61801
Telephone: 217-333-2999
e-mail: scheelin@aries.scs.uiuc.edu

ISOLATION OF METALS FROM LIQUID WASTES: REACTIVE SCAVENGING BY SORBENTS IN TURBULENT THERMAL REACTORS

Graduate Students

Jonathan M. Amos, University of Arizona, Department of Chemical and Environmental Engineering, Graduate student studying towards an M.S. degree.

John R. Schmidt, University of Arizona, Department of Chemical and Environmental Engineering, Graduate student studying towards a Ph.D. degree.

Art Fernandez, University of Arizona, Department of Chemical and Environmental Engineering, Graduate student studying towards a Ph.D. degree.

Min-Po Shiue, University of Illinois, Department of Mechanical and Industrial Engineering, Graduate student studying towards a Ph.D. degree.

W. Joseph Mantle, University of Illinois, Department of Mechanical and Industrial Engineering, Graduate student studying towards a Ph.D. degree.

Research Objectives

A large portion of the Department of Energy (DOE) radioactive waste inventory is composed of metal-bearing liquid wastes, which may or may not also contain organics. It is highly desirable to concentrate the metals in order to reduce the volume of these wastes and to render them into an environmentally benign form. One method for doing this is to exploit high-temperatures to reactively capture metals by sorbents, and thus to isolate them from the environment. The objective of this research is to provide the background information necessary to design a process that accomplishes this on a large scale, namely in hot turbulent flows, into which are injected the wastes to be treated and, also the sorbents that scavenge the metals. The current work focuses on cesium and strontium, which are present in the DOE inventory as radioactive isotopes.

The project involves five investigators at three institutions, and is comprised of the following parts:

- 1) Experimental research at the University of Arizona focuses on the chemistry of cesium and strontium sorption on kaolinite and lime sorbents in a laminar flow environment.
- 2) Theoretical research pursued jointly by the University of Arizona and Sandia Laboratories, Livermore, focuses on prediction of droplet trajectories in a turbulent flow environment.
- 3) Experimental research at the Air Pollution Technology Branch of the US Environmental Protection Agency, to investigate the process in turbulent flows.
- 4) Experimental research at the University of Illinois focuses on design, construction, and application of a laser based LIBS system for measuring droplet size, metal concentration in the gas phase, and metal concentration in the vapor phase. This analysis procedure will be used both at the University Of Arizona laminar flow reactor and the EPA turbulent flow reactor.
- 5) Theoretical research at the University of Illinois to provide input into the drag model to be used to predict droplet trajectories in hot turbulent flows.

Research Progress and Implications

This report summarizes work after 1¾ years of a 3-year project.

Experiments were performed in the 18 kW laminar downflow furnace at the University Of Arizona. Aqueous solutions the metals were atomized and injected into the flame. Powdered sorbents were injected into the post-flame region of the furnace. Particles were sampled from the furnace isokinetically and passed through a Berner low-pressure impactor. The samples were then analyzed. Extents of reaction for cesium (a semi-volatile metal) were determined through the method of aerosol size fractionation, whereby the fraction of a metal falling in the sub-micron particle size fraction range is considered to be unreacted. Reaction extents of the less volatile strontium were also inferred through the resulting strontium particle size distribution (psd).

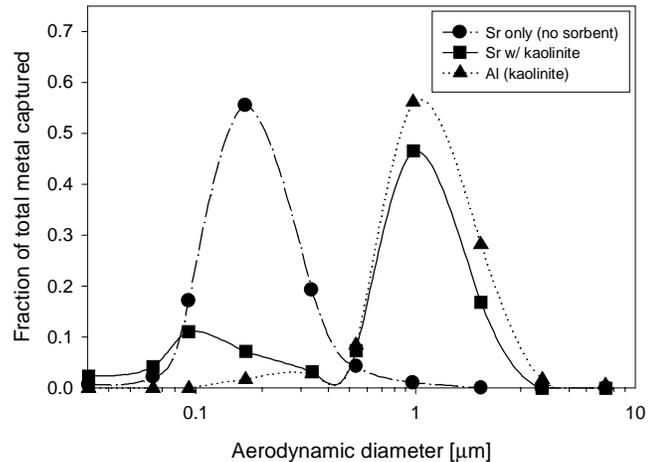


Figure 1: Strontium can be captured by kaolinite in a thermal reactor, but only in the presence of chlorine.

Without chlorine or sulfur present, strontium shows a broad peak in the exhaust psd, indicating that the strontium fails to vaporize. Addition of the sorbents creates a sharper peak, but this is probably due to coagulation of the strontium compound onto the sorbent, since the strontium does not vaporize. The experimental evidence suggests that sulfur allows the strontium to vaporize and then nucleate to form small particles, which is different from the equilibrium predictions. However, this enhanced vaporization does not appear to enhance the capture of strontium. Chlorine, however, acts to reduce the dewpoint of strontium. Thus, the metal is in the vapor form (as shown on Fig 1 by the psd around 0.1 μm, without sorbent present) over appreciable contact times with the sorbent. In the presence of chlorine, kaolinite can reactively capture strontium as shown in Fig 1 by the shift in the strontium psd to match the sorbent psd. Lime, however, does not capture strontium.

The psd of cesium alone forms a peak at 0.2 μm. This indicates that the cesium volatilizes, and then nucleates in the probe to form a fume of smaller particles. When kaolinite is added, the cesium peak shifts to match that of the kaolinite. This shows that the cesium is reacting with the kaolinite and is scavenged. Lime shows only a minimal amount of capture. When chlorine is added to the system, cesium is still captured onto kaolinite, but not on lime. Sulfur produces a double peak in the distribution curve. In the presence of both sorbents, this appears to enhance the capture.

The significance of these results lies in that they show that high temperatures can be exploited to reactively capture both cesium and strontium on substrates, which can be

captured, and effectively isolated from the environment. This is contrary to the conventional wisdom that high temperature processes have no application to the isolation of metal containing radioactive wastes.

Installation of laser diagnostics at the Arizona facility is progressing. Components are being fabricated at the University of Illinois at Urbana-Champaign as outlined in the original proposal, and both the gated CCD and excimer laser have been received and found to meet specifications. Transfer of major equipment items from Urbana to Tucson is anticipated for late summer. Optical engineering is effectively complete, following a site visit by Scheeline and Pearlstein in late May.

A key part of this effort is a model representing a liquid droplet of metal containing hazardous waste transversing a highly turbulent combustor. Even though the combustion of liquid fuels in most practical combustors is not dominated by the trajectory of individual droplets, this knowledge is necessary here in order to predict both the complete vaporization of all the hazardous waste containing drops and the optimum placement downstream of the flame of the injected sorbent to take advantage of the availability, and therefore optimize the capture of the metal vapor. The success of such a model would therefore allow the use of the kinetic data gathered from the University of Arizona's laminar flow reactor to predict and help design larger turbulent flow reactors.

A One Dimensional Turbulence model, devised by Kerstein at Sandia Laboratories, forms the basis on which a computer program describing individual droplet trajectories in turbulent flows has been written and is currently being tested. The purpose of this model is to predict the range of droplet burnout points in the EPA turbulent flow reactor, as measured previously. This program will form the basis for future predictions.

Research at University of Illinois has focused on drag prediction. We have developed a technique for efficiently computing two-dimensional, axisymmetric, or three-dimensional bluff-body wake flows on unbounded domains. The approach makes use of the fact that the vorticity is largely confined to a relatively small subdomain that is either parabolic (for two-dimensional flows) or paraboloidal (in the axisymmetric or three-dimensional cases). We then solve the full Navier-Stokes equations on this domain, subject to the condition that the solution matches the far-field analytical solution of the inviscid equations on the parabolic or paraboloidal boundary. The technique is being demonstrated for rigid bluff bodies (circular cylinders and spheres), and will then be implemented in the drop flows of interest.

Planned activities

The laser diagnostic system will be applied to the Arizona reactor before December 99, and to the EPA turbulent reactor before July 00. The droplet trajectory model will be calibrated against literature data by October, 99. Several papers on interim results obtained to date are being prepared for publication this year.