

## **Annual Report of EMSP Project 55396-CA**

### **Title: Reactive Transport of Uranium in Waste Plumes of Hanford Site**

PI: **Jiamin Wan**, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., MS 70-108B, Berkeley, CA 94720, (510) 486-6004, jwan@lbl.gov

Co-PI: **Tetsu K. Tokunaga**, Lawrence Berkeley National Laboratory

Co-PI: **Carl I. Steefel**, Lawrence Berkeley National Laboratory

Co-PI: **Peter C. Burns**, University of Notre Dame

### **Research Objectives**

The overall objective of this renewal proposal is to construct an experimentally supported conceptual model of U fate and transport, during and after the tank leakage, at heavily U-contaminated areas within the Hanford Site. Through simulating the tank leakage event in laboratory columns, we will better understand the spatial and temporal distribution of the spilled U, and its mobility. These laboratory results will specifically be used to compare with and help explain borehole data from underneath tank BX-102. Our research focuses on the following basic issues:

1. Determining the spatial distribution of U within the plume during the early stage of waste plume development.
2. Determining the forms and amount of U partitioned in the pore water relative to the matrix-associated U. Identify dissolved U and suspended mobile U-rich colloids in the aqueous phase, and characterize matrix-associated forms (deposited U colloids, sorbed and precipitated U species).
3. Determine how all the above change with time.
4. Determining the mobility of U (release and transport) as function of aging.
5. Developing a mechanistic reactive transport model capable of accounting for both dissolved and particulate U(VI) in the Hanford vadose zone.

An overall understanding of U-precipitation vs. adsorption, stability of nanoparticles and their transport, will improve our ability to predict current and future development of U-waste plumes.

### **Research Progress and Implications**

This report summarizes progress during the first year (2004) of our project. As of June 2004, we have been focused on two tasks.

#### **1. Preparatory work for column experiments.**

- We have prepared a large batch of sediment for all the future experiments with desired moisture content, and measured the chemical composition of its water extract.
- We have constructed a 2 m long column, and six 0.5 m long columns. We prepared all equipment needed for running the experiment at 70°C, sectioning columns, extracting pore liquids, and aging the sediment.
- We tested the possibility of adding NaBr in the tank waste solution (TWS) as a tracer, but found it interacted excessively with other solution components.
- We are currently testing what flow rate to be used in the column experiments, using newly synthesized TWS (although the solution has not reached equilibrium yet). We are currently testing 3 flow rates; 25 cm/d, 5 cm/d, and 1cm/d in three 25 cm long columns. The 1 cm/d column is still on going. We will have the flow rate for the main experiments chosen soon.

## **2. Synthesizing the U Tank Waste Solution (BX-102)**

This has been a very difficult task. In order to have the most relevant TWS to begin with, we decided to follow the recipe of historical procedure by going through entire  $\text{BiPO}_4$  and neutralization processes. After 8 months of experimenting, we now have solutions close to the target composition. Within the next month, we will have the synthesized BX-102 tank waste solution ready, and begin the column and batch experiments.

The DOE's clean up efforts at the Hanford Site include the Tank Farm Vadose Zone Characterization Project. Because our proposed experiments closely simulate field contamination processes, the results and analyses will permit direct comparisons with field-based observations. The quantitative reactive transport model developed for U based on the experiments in this study will provide an important bridge to the field-scale problem.

### **Planned Activities**

In order to achieve the goal of understanding uranium reaction and transport within waste plumes, we plan to take two general approaches: the batch equilibrations and column flow experiments using the simulated waste solution and Hanford sediments. The batch approach will provide information for well-mixed and near-equilibrium systems, and provide comparisons to reactive transport (column flow) results. The column experiments are of 2 types: stopped-flow with immediate profile sampling, and flow-through with later natural recharge (dilute, uncontaminated soil water). Geochemical modeling will be done on each of these batch and column experiments to help understand laboratory and field measurements, and to assist in improving future experimental procedures.

### **Publications**

Although the following papers are from our previous EMSP project which just ended, the methods and approaches will be used in this new project.

- Wan, J., J.T. Larsen, T.K. Tokunaga, Z. Zheng, pH Neutralization and Zonation in Alkaline-Saline Tank Waste Plumes. *Environ. Sci. Technol.* 38, 1321-29, 2004.
- Wan, J., T. K. Tokunaga, J.T. Larsen, and R. J. Serne, Geochemical Evolution of Highly Alkaline and Saline Tank Waste Plumes during Seepage through Vadose Zone Sediments. *Geochim.Cosmochim. Acta.* 68, 491-502, 2004.
- Wan, J., T. K. Tokunaga, J.T. Larsen, E. Saiz, J. T. Larsen, Z. Zheng, R. A. Couture, Colloid Formation at Waste Plume Fronts. *Environ. Sci. Technol.*, submitted May 2004.