Project Title: **Colloid Genesis/Transport and Flow Pathway Alterations Resulting From Interactions of Reactive Waste Solutions and Hanford Vadose Zone Sediments**

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Research Objectives

Leakage of underground tanks containing high-level nuclear waste solutions has been identified at various DOE facilities. The Hanford Site is one the main facilities of concern, with about 2,300 to 3,400 m^3 of leaked waste liquids. Radionuclides and other contaminants have been found in elevated concentrations in the vadose zone and groundwater underneath single shell tank farms. We do not currently know the mechanisms responsible for the unexpected deep migration of some contaminants through the vadose zone, and such understanding is urgently needed for planning remediation. Due to the extreme chemical conditions of the tank waste solutions (very high pH, aluminum concentration, and ionic strength), interactions between the highly reactive waste solutions and sediments underneath the tanks can result in dissolution of primary minerals of the sediments and precipitation of secondary phases including colloidal particles. Contaminants can sorb onto and/or co-precipitate with the secondary phases. Therefore transport of strongly associated contaminants on mobile colloids can be substantially greater than without colloids. The overall objective of this research is to improve our understanding on the effects of interactions between the tank waste solution and sediments on deep contaminant migration under Hanford Site conditions. This objective will be achieved through the following four tasks: (1) colloid generation and transport studies, (2) studies on sediment permeability and chemical composition alterations, (3) quantifying associations of contaminants with secondary colloids, and (4) studies on the combined effects of the aforementioned processes on deep contaminant migration.

Research Progress and Implications

This report summarizes progress during the first year (2001) of our project, focusing on task 1, colloid generation and transport. In pursuit of our research objective, the process of a leaking tank waste solution interacting with underlying sediments was simulated through infiltrating simulated REDOX tank waste solutions into columns packed with Hanford vadose zone sediments. Since the sediments immediately underlying some STX tanks experience elevated temperatures from heat released by reactions within tanks, experiments were conducted at elevated (70 •C) and room temperatures. Effluents from columns were collected continuously and analyzed for turbidity, pH and chemical composition. Secondary colloids collected from effluents were characterized for their major elements using an electron microprobe, and mineralogy was determined by x-ray diffraction. The morphology of the particles was studied using a scanning electron microscope. Changes in major elemental composition within the sediment columns were measured using x-ray fluorescence.

We identified three stages of waste solution and sediment interactions: the leading edge of the waster solution, the waste plume, and dilution stages. We found that the secondary colloids formed during the different infiltration stages are different in term of quantity, chemical composition, mineralogy, morphology, and size distribution. At the leading edge, small amorphous particles are the dominate phase. Within the waste plume stage, sodium aluminum silicates are dominant at both temperatures. During the dilution stage bayerite is abundant. The majority of precipitates collected from effluents were submicron to micrometer sized. Maximum particle formation occurred at the leading edge of the waste plume, where maximum geochemical disequilibrium exists. Maximum transport of the secondary colloids also occurred at the leading edge, where and when pores have not yet been plugged by particle production and new colloids have not yet attached to primary grain surfaces. Our laboratory studies revealed the complex processes of reaction-dependent colloid generation/release, permeability change, and flow pathway alteration that can strongly influence flow and transport of REDOX tank contaminants in the Hanford vadose zone. In comparison to the large quantity of secondary colloids, the inventory of indigenous colloids is often small.

Planned Activities

The recent studies described above focused on colloid generation, under conditions of relatively high sediment water saturation. Ongoing studies are quantifying sediment alteration with respect to permeability and chemical composition. During FY 2002 we plan to conduct studies of colloid transport in sediments at lower saturations, more typical of the Hanford vadose zone.

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