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Radionuclide Waste Disposal: Development of Multi-scale Experimental and Modeling Capabilities

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This abstract provides a general overview of a DOE Experimental Program to Stimulate Competitive Research (EPSCoR) Implementation grant in South Carolina with supporting funding coming from the BES Heavy Element Chemistry program and the BER Subsurface Biogeochemical Research Program. The objective of this project is to quantitatively describe the influence of coupled physical, chemical, and biological reactions on the migration of radionuclides through porous media. Particular emphasis is placed on determining processes which limit the rates of radionuclide migration including:

1. Impacts of fluid flow, particularly preferential flow, which induce chemical gradients and control the availability of reagents which may influence the mobility of radionuclides.
2. Identification of rate limiting steps of chemical reactions which cause changes in radionuclide mobility as well as the extent of reaction reversibility
3. Characterization of the time and length scales over which non-equilibrium states are maintained based on a competition between chemical reaction rates and physical processes (i.e. flow).

To meet these objectives, we will develop a series of novel, multidimensional tools capable of real time monitoring radionuclide mobility in space and time. This effort will entail development of 2D sensors to monitor chemical states and distribution (i.e. pH, O₂(aq) concentrations, radionuclide concentrations) in 2D microfluidic cells and macroscale tanks as well as CT and SPECT medical imaging techniques to monitor flow, pore structure, and radionuclide distribution in 4D.

Highlights from several ongoing experimental and modeling studies will be discussed including:

1. Examination of uranyl phosphate dissolution by plant exudates using batch flow through reactors, 2D microfluidic systems, and soil column testing.
2. Development of novel 2D and 3D measurement techniques including x-ray CT and Single Photon Computed Tomography (SPECT) examining Tc-99m transport through porous media. We have evaluated the spatial and temporal resolution limitations of these techniques in an idealized system (i.e. Tc-99m transport through a column packed with glass beads of varying sizes) and also monitored redox driven retention of Tc-99m through soil columns with heterogeneous reducing zones.
3. Quantification of sorption hysteresis and rate limiting steps involved with Cs ion exchange on a sandy loam soil and iodine transformations to organo-iodine species in a wetland sediment. Both studies indicate there is a low concentration but highly reactive component of the soil which controls Cs and I behavior at environmentally relevant concentrations.
4. Field based lysimeter studies of Tc-99 and Np-237 which indicate Tc-99 and Np-237 mobility are highly controlled by oxidation of initially Tc(IV) and Np(IV) sources. The rate and extent of source term oxidation was examined through measurements of Np and Tc in the lysimeter effluents during field deployment as well as leaching, batch sorption, x-ray absorption spectroscopy, and electron microscopy techniques after the lysimeters were retrieved from, the field.