
The impacts of pore-scale physical and chemical heterogeneities on the transport of radionuclide-carrying colloids

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ABSTRACT:

This proposal addresses the *Used Fuel Disposition Campaign Disposal Research and Development Roadmap*, which ranked the transport of backfill materials (e.g., bentonite clay) in porous media as "highly important" for safety, since their high affinity to radionuclides makes them potential hazards when these materials are released into groundwater. Previous studies of colloid transport have focused solely on the physical heterogeneities (e.g., pore space geometry and grain size) within porous media. However, all natural porous media possess inherently coupled physical and chemical heterogeneities (e.g., surface charge and hydrophobicity). While the chemical heterogeneities determine whether a specific location is favorable for colloidal retention, the physical heterogeneities determine the site accessibility. Therefore, both heterogeneities must be considered together. The lack of understanding in how these heterogeneities influence transport is a major source of uncertainty in developing models to predict radionuclides distribution in the geosphere.

To address the above fundamental knowledge gap, we will identify the dominant transport mechanisms of radionuclide-carrying colloids in porous media under the influence of coupled physical and chemical heterogeneities. The proposed work is structured as a series of complementary experiments and numerical simulations, which span length scales from 100 nm to 10 cm. We will first build pore-scale sediment analogs using a bead-based microfluidic approach so that both physical and chemical heterogeneities can be precisely defined. Measurements from the microfluidic sediment analogs will be used as the input of a pore-scale program that includes the essential hydrodynamic and colloidal interactions needed to faithfully simulate the fluid flow and colloidal transport. Outputs from the pore-scale model will then be averaged to develop a continuum-scale simulator for predicting colloidal transport at larger scales and for estimation of uncertainties in parameters. Such prediction will be tested and improved against measurements from centimeter scale column experiments. We will focus on ^{137}Cs and ^{90}Sr as the representative monovalent and multivalent radionuclides. These radionuclides will be adsorbed to both model silica colloids and bentonite clay colloids. The transport of these colloids will be measured as a function of chemical and physical heterogeneities, as well as fluid properties (e.g., pH, ionic strength, and solution chemistry) to simulate relevant repository environment. Understanding gained from our studies, however, can be readily extended to other types of radionuclides and colloids.

The expected outcome of this work will be experimental and computational models of near-field environments that couple pore-scale geometric and interfacial structures with the transport of radionuclide bearing colloids. Key deliverables include a validated pore-scale simulator that captures key transport mechanisms identified in experiments using microfluidic sediment analogs and a continuum-scale model that rigorously upscales the averaged adsorptive properties to 10 cm column scales. The delivery of such simulators that are rigorous at the microscopic scale and predictive at the intermediate scale is a necessary step towards fully predictive models for field-scale applications in future.

The proposed work builds on a solid track record of prior work carried out by Colorado School of Mines (\$640,000) and Pacific Northwest National Laboratory (\$160,000). Together, the investigators have extensive publications in colloidal science/engineering, microfluidics, porous media simulation, radiochemistry, and radioactive waste management. The collaborating groups have all the state-of-the art equipment necessary for successful implementation of the research plan.