Effects of pore-scale physics on uranium geochemistry in Hanford sediments

University-Led Research

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The Hanford 300 Area sediments are mainly river cobble and gravel, with sands and fines (silt + clay) variably filling the voids in between. Fines comprise only 1.78% of the total sediment mass, but hold a disproportionately large fraction of the total U. However, the coarser sediments (e.g., >2 mm), with their larger mass fraction, may serve as a long-term U release pool despite their lower U concentration. We hypothesized that slow U release at the 300 Area is partly due to low connectivity of intra-granular pores. The objectives of this project are to evaluate the U distribution in 2-8 mm size sediments, to determine whether low pore connectivity affects U distribution and release from Hanford 300 Area sediments, and if so, to assess its implications for long-term release. Our approach integrates laboratory experiments (ICP-MS instrumentation to measure multiple elemental concentrations in liquid samples, laser ablation (LA)-ICP-MS for elemental mapping of 2-8 mm sized basaltic clasts) and pore-scale network modeling.

Over the last year we performed the following tasks with associated results:

1) Conducted complementary laboratory tests (stirred flow cell, saturated column, and batch reactor) to study U release behavior from each of five size fractions (<75 µm, 75-500 µm, 500-2000 µm, 2-8 mm, and a <2 mm composite). The stirred flow cell results indicate that 8.7% of the total U is in the 2-8 mm fraction, significantly reducing uncertainty in the U inventory. In addition, released U concentration shows a power-law relationship to elution volume (i.e., elution time) for all size fractions (published in *J. Environ. Radioact.*). Numerical simulations are being developed to recreate both stirred cell and saturated column experiments to better characterize U release rate constant distributions, and evaluate rate constant uncertainty for different size fractions.
2) Assessed concentration distributions of U and other elements in a 2-8 mm basaltic clast as a function of distance from the grain's surface. A second 2-8 mm basaltic clast was mapped and compared to the first mapped clast, examining fewer layers but to deeper depths. U concentration was power-law distributed with distance from the grain surface (published in *Environ. Sci. Technol*.).
3) Tested a pore-scale network model to examine pore connectivity issues in contaminant retention and release. The model supports our hypotheses about accessible porosity and anomalous patterns of contaminant release, and predicts U concentration distributions consistent with those observed in (2). A finite-difference model closely matches the network model results, indicating that we correctly captured how porosity and diffusion change with distance (published in *Water Resour. Res.*).
4) Carried out 3-D elemental mapping of a 2-8 mm basaltic clast, after vacuum saturation with a suite of nonsorbing and sorbing tracers, to better assess the edge-accessible porosity distribution; data processing and analysis are underway.
5) Performed unsaturated column transport experiments for 2-8 mm size fraction (at water saturations of 77% and 14%) and <2 mm size fraction (at 88% water saturation). Uranium release from 14% saturation is greater than that from 77% saturation under otherwise identical conditions, probably because at lower saturation, flow paths have more contact and interaction with the grains.