

Uranium Adsorption Experiments at Elevated Temperatures: Method Development and Lessons Learned

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Elevated temperatures have to be expected at future nuclear waste repositories due to the initial thermal loading and radioactive decay heat of spent nuclear fuel. Potential heat effects on engineered barrier systems, surrounding nuclear waste canisters, will influence repository design with respect to canister spacing, and subsequently determine the size and cost of future repositories. Engineered bentonite barriers will be designed to limit the release of radioactive contaminants after potential canister failures, largely driven by the high, expected adsorption of radionuclides onto clays and other minerals. In close collaboration with Dr. Florie Caporuscio's group at Los Alamos National Laboratory, we have characterized uranium(VI) adsorption onto heat-treated bentonite and montmorillonite clay at room-temperature over the past few years mimicking a late radionuclide release scenario. In a next step, we are investigating U(VI) adsorption onto montmorillonite at elevated system temperatures (up to 80 °C) for an early release scenario. These adsorption experiments typically include: (1) a pre-equilibration of solids with target chemical solution conditions, (2) the equilibration of solution-phase U(VI) with clay surfaces, followed by (3) a solid-liquid separation, and (4) the sampling and analysis of supernatant solutions in terms of pH, remaining U(VI) solution concentrations, etc.

In this presentation, we will provide an overview of the challenges involved with performing metal adsorption experiments at elevated temperatures, and discuss sources of experimental errors and bias for temperature-dependent metal adsorption data. We were able to test and resolve technical challenges with respect to pH calibrations and measurements, the loss of dissolved CO₂ from solution, and variability in solution temperatures depending on sample vial positions on thermal shakers. Additionally, it became apparent that the method selected for solid-liquid separations (i.e., centrifugation, filtration or clay settling) can introduce large experimental errors and bias for U(VI) adsorption data as a function of pH and temperature. If not corrected, then this could lead to a misinterpretation of temperature effects on the pH dependence of uranium(VI) sorption behavior, and the calibration of predictive models with inaccurate experimental data.

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