

Changes in Uranium(VI) Solution Speciation and Adsorption Behavior with Increasing Temperature

RUTH TINNACHER¹, BEN URICK¹ AND CHRISTOPHE TOURNASSAT^{2,3}

¹California State University East Bay

²ISTO, Université d'Orléans

³Earth and Environmental Sciences Area, Lawrence Berkeley National Laboratory, Berkeley, California

Presenting Author: ruth.tinnacher@csueastbay.edu

Most nuclear waste repositories will include compacted bentonite, mainly consisting of montmorillonite clay, as part of engineered barrier systems (EBS) surrounding waste canisters. These barriers will minimize the release of radioactive contaminants into the natural environment, given their high adsorption affinities and low permeability for many radionuclides. For early release scenarios, elevated temperatures have to be expected in the EBS due to the initial thermal loading of waste packages and the continued release of decay heat. As a result, contaminant transport predictions will have to include the complex effects of temperature on radionuclide solution speciation, adsorption behavior and diffusion coefficients. The goal of this modeling study is to improve our understanding of temperature effects on radionuclide mobility in the EBS using U(VI)-montmorillonite systems as examples.

First, we simulated U(VI) solution speciation as a function of pH, partial pressure of CO₂ and temperature (25-80 °C) in PHREEQC (ThermoChimie 11a database). We found that, with temperature, U(VI)-hydroxide solution complexes become dominant over a broader pH range, both in the absence and presence of CO₂. Next, we evaluated the individual contributions of various parameters and processes (e.g., changes in pH neutrality, the dissolution of CO₂ and U(VI) solution complexation constants with temperature) to the overall shifts in U(VI) solution speciation. Based on our analysis, changes in U(VI) solution complexation constants primarily drive the changes in U(VI) solution speciation with temperature; hence, uncertainties associated with these thermodynamic parameters directly affect our predictions. Finally, we also included temperature effects on U(VI) adsorption behavior by modifying log *K* values for U(VI) surface complexation and surface protonation reactions. While the predicted U(VI) adsorption decreases with temperature for most pH values, this change is primarily driven by temperature effects on solution-phase, not surface processes. We will compare these predicted U(VI) adsorption data at elevated temperatures with our own preliminary experimental results (companion presentation by Urick et al.), and existing data for comparable systems in the literature.

This research is funded through DOE's Nuclear Engineering University Program (DE-NE0008683 and DE-NE0008938).