Goldschmidt 2022 Abstract
https://doi.org/10.46427/gold2022.12709

Effects of temperature on uranium(VI) solution speciation and sorption behavior

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Elevated temperatures have to be expected at future nuclear waste repositories due to the initial thermal loading of waste packages and the continued release of decay heat over time. As a result, contaminant transport models for early-release scenarios will have to consider potential effects of temperature on contaminant mobility, which may include changes in mineral surface characteristics, radionuclide adsorption behavior and diffusion coefficients. Furthermore, the predictions of any temperature-dependent radionuclide surface complexation or diffusion model will largely depend on an accurate representation of radionuclide solution speciation at elevated temperatures. Hence, the goal of this study is to gain a better understanding of potential changes in radionuclide solution speciation as a function of temperature, using uranium(VI) as example.

In a first step, we evaluated a series of underlying parameters and processes that could contribute to the apparent changes in U(VI) solution speciation as a function of temperature. For this purpose, we simulated U(VI) solution speciation as a function of pH, partial pressures of CO_2 and temperature (25 – 80 °C), using PHREEQC with the ThermoChimie 10a database. Our results suggest that the temperature-dependence of U(VI) solution complexation constants is primarily responsible for the shifts in U(VI) solution speciation with temperature in many systems. For instance, at atmospheric CO_2 conditions, we observed that U(VI)-hydroxide complexes become dominant over a broader pH range at elevated temperatures.

In a next step, we are currently evaluating how the reported uncertainties associated with deltaH values in the Van't Hoff equation can affect predicted changes in U(VI) solution speciation with temperature. Last, we will also discuss how temperature-induced changes in U(VI) solution speciation alone may affect U(VI) sorption affinities and U(VI) surface speciation.

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