
Quantifying radionuclide sorption to engineered barrier materials under elevated temperature and ionic strength conditions

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ABSTRACT: The objective of this research is to examine mechanisms and thermodynamics of actinide sorption to engineered barrier materials (iron (oxyhydr)oxides and bentonite clay) for nuclear waste repositories under high temperature and high ionic strength conditions using a suite of macroscopic and microscopic techniques, which will be coupled with interfacial reaction models. Gaining a mechanistic understanding of interfacial processes governing the sorption/sequestration of actinides at mineral-water interfaces is fundamental for the accurate prediction of actinide behavior in waste repositories. Although macroscale sorption data and various spectroscopic techniques have provided valuable information regarding speciation of actinides at solid-water interfaces, significant knowledge gaps still exist with respect to sorption mechanisms and our ability to quantify sorption, particularly at high temperatures and ionic strengths. *This proposed work seeks to characterize actinide sorption to mineral surfaces and examine possible mechanisms of sorption at high temperature/ionic strength through examination of the effects of hydration and hydrolysis on actinide sorption.* This will be achieved by quantifying actinide sorption using a thermodynamically based surface complexation model and determining sorption enthalpy and entropy from variable temperature sorption experiments and calorimetric titrations. There has been little effort to measure such thermodynamic data, with the majority of past efforts using empirical approaches (discussed below) with data collected only at room temperature and only over a limited range of ionic strengths. Therefore, the thermodynamic data collected in this work at high temperature and ionic strength will greatly improve the technical basis for prediction of waste performance in a repository under near field conditions.

Hypotheses: The overarching hypothesis of this research is that strong actinide interactions with metal (oxyhydr)oxide surfaces are manifested by large stability constants for the actinide surface complexes. These large stability constants are due to positive entropies which are mechanistically driven by displacement of solvating water molecules from the actinide ion and the mineral surface during sorption and/or surface precipitation. Such entropies are accessible through measurement of sorption enthalpies and stability constants using surface complexation modeling and calorimetric titration techniques.

Outcomes: The primary deliverable will be a set of thermodynamically based sorption and ion exchange constants describing radionuclide sorption to engineered barrier materials. These data will provide an understanding of the fundamental reaction mechanisms occurring at the mineral-water interface. A greater understanding of these processes will reduce the uncertainty in strategies for sequestration of radionuclide bearing wastes. Overall this project will increase our understanding of radionuclide interfacial reactions and help to ensure human and environmental health are protected during treatment and disposal of radionuclide bearing wastes.