NEUP Nuclear Energy University Program
U.S. Department of Energy

Surface Complexation of Radionuclides on Mineral Surfaces at High Temperature

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Program:
Spent Fuel, Waste Science & Technology, and Integrated Waste Management System

ABSTRACT:
In a typical multi-barrier system for a geological repository of high-level radioactive waste, retardation of radionuclides due to adsorption by bentonite buffer and natural barrier is key for assessing the long-term safety of the repository. Process models and performance assessment models have been used for the safety assessment, which parameterize the adsorption process with different models, such as the linear solid/liquid partition model and surface complexation models. Thus, accurate adsorption constants in these models are critical for accurate and reliable predictions of the migration of radionuclides in a repository. Adsorption constants in adsorption models vary with temperature. In the life span of a repository, bentonite buffer and the near field area of host rock undergo heating from the waste package, and the temperature therein is expected to go as high as 100 ºC or even higher (up to 200 ºC). Depending on the assumed time that canister is fully breached, migration of radionuclide could happen when the temperature in bentonite remains relatively high, particularly in the case of early failure of the waste package. Thus, it is necessary to use temperature-dependent adsorption constants in those models. However, all currently available adsorption constants for radionuclide adsorption on barrier materials were determined from experimental adsorption data obtained at ambient temperature. The constants cannot be extrapolated reliably to a high temperature using Van't Hoff equation that applies only to a narrow temperature range. The lack of the data limits the model predictability of radionuclide migration under high temperature conditions. Therefore, the goal of the proposed project is to determine the adsorption constants of typical radionuclides on repository-relevant minerals at temperatures up to 200 ºC using a combination of batch adsorption experiments with a hydrothermal reactor and surface complexation modeling. The molecular structure of surface complexes at high temperatures, which are required by accurate surface complexation models, will be determined using in situ spectroscopic measurements with a hydrothermal cell, which will be complemented by molecular simulations. The obtained surface complexation constants at various temperatures will be further incorporated into reactive transport modes and performance assessment models for the safety assessment. The obtained parameters will enable a more accurate assessment of the long-term safety of the repository.