Nuclear Energy University Program

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Developing a Thermochemical Database of Radionuclide Reactions at the Mineral–Water Interface for Improved Nuclear Waste Repository Performance Assessment

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ABSTRACT:

The goal of this project is to develop a robust and self-consistent database of thermodynamic constants describing radionuclide reactions at the mineral–water interface (e.g., surface complexation reactions). The development of this thermochemical database will increase the accuracy of input parameters for nuclear waste repository performance assessments and will provide a vast resource for advancing understanding of the fundamental chemistry driving radionuclide reactions at the mineral–water interface. No such database currently exists, precluding efforts to include surface complexation modeling in safety assessment calculations. To remedy this problem, this work will create a publically–accessible database of thermodynamic constants (i.e., free energies, enthalpies, and entropies) for surface complexation reactions. The database will be fully consistent with thermodynamic data describing radionuclide reactions in the aqueous phase that are currently available in other databases, and the database will provide surface complexation constants that can be directly incorporated into geochemical modeling software (e.g., PHREEQC, MINTEQA2, etc.). Initially, the database will be populated with data describing the sorption of several risk driving radionuclides (^{79}Se, ^{152}Eu, ^{226}Ra, ^{237}Np, ^{234,235,238}U, ^{238,239,242}Pu, and ^{241}Am) onto iron (hydro)oxide minerals that form during steel corrosion, with the expectation that continued development of the database will expand the available thermodynamic constants to other radionuclide/mineral systems. Specifically, this project will normalize radionuclide sorption data as a function of specific surface area, globally fit the cumulative data using a single self–consistent surface complexation model (SCM), validate the SCM by testing the fit quality with select datasets, and output the data into a publicly–accessible and software–ready database. Key aspects of this project include selection of the electrical double layer (EDL) model (e.g., diffuse-double layer model (DLM), triple-layer model (TLM), etc.), incorporation of thermodynamic constants for radionuclide–mineral–surface interactions at elevated temperatures, and inclusion of raw radionuclide sorption metadata (that can be used by other researchers for additional fitting exercises). Ultimately, this work will facilitate an improved assessment of the human and environmental risks associated with nuclear waste disposal and will establish a resource for the geochemical community to further explore fundamental concepts and questions related to radionuclide reactions at the mineral–water interface.