A Prediction of Uranium(VI) Diffusion in Montmorillonite at Various Chemical Solution Conditions

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Most nuclear waste disposal options include compacted bentonite, predominately consisting of montmorillonite clay, as part of a barrier system to minimize contaminant mobility. Uranium (U) is the primary element in spent nuclear fuel, and a potential contaminant of water resources. Uranium sorption onto clay and its slow diffusive transport away from waste canisters are expected to limit U(VI) mobility, and possibly also control nuclear fuel degradation rates based on mass action considerations.

A prediction of U(VI) diffusion in these systems is complicated by two main factors: (1) the complex mineralogical structure of montmorillonite clay, leading to two types of clay porosities and surface sites, and (2) the dependence of U(VI) sorption on U(VI) solution speciation and the specific chemical system conditions. For instance, depending on pH and ionic strength, U(VI) may sorb onto montmorillonite due to weak ion exchange reactions or the formation of stable surface complexes. Furthermore, in contrast to conventional porous media, U(VI) diffusion in montmorillonite is not only limited by contaminant sorption, but also by a (potential) lack of access to the full clay porosity for some U(VI) solution species. For instance, at the high degrees of clay compaction expected in future barrier systems, the partial or full exclusion of U(VI) from negatively-charged clay interlayer spaces may substantially decrease U(VI) fluxes.

In this modeling study, we specifically investigated how the charge of U(VI) solution species affects U(VI) sorption and diffusion behavior. For this purpose, we first simplified complex *chemical* solution speciation diagrams to *charge* speciation diagrams. Then, we coupled an existing U(VI) surface complexation model (Tournassat et al., *Geochim. Cosmochim. Acta*, 2018) with an analytical solution of the diffusion equation to simulate U(VI) mobility in lab-scale through-diffusion experiments for various chemical conditions. Our results suggest that there are two important parameters that govern U(VI) diffusion in these systems. First, U(VI) surface complexation on clay edge surfaces leads to contaminant retardation. On the field scale, this will determine how early a plume may cross site boundaries and pose risks to drinking water resources. Second, the degree of access of U(VI) solution species to diffuse layers in clay interlayer spaces largely controls U(VI) diffusive fluxes. In the field, this parameter will govern the level of U(VI) concentrations that can be expected in contaminated waters.

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