Simulating uranium(VI) diffusion in sodium-montmorillonite as a function of chemical solution conditions

Ruth M. Tinnacher[1], Jonathan C. Pistorino[1], Christophe Tournassat[2]

[1]Department of Chemistry and Biochemistry, California State University East Bay, Hayward, CA 94542
[2]UMR 7327 Institut des Sciences de la Terre d'Orléans, Université d'Orléans – CNRS/INSU – BRGM, Orléans

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 in these systems. However, their prediction is complicated by the complex mineralogical structure of montmorillonite clay, leading to two types of surface sites and clay porosities, and by the dependence of U(VI) solution speciation on chemical 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, at the high degrees of clay compaction expected in future barrier systems, the ability of U(VI) species to access small, highly negatively charged interlayer spaces may ultimately control diffusive fluxes.

In this modeling study, we investigate how the charge of U(VI) solution species affects U(VI) sorption and diffusion behavior. For this purpose, we simplified complex *chemical* solution speciation diagrams to *charge* speciation diagrams. Furthermore, we coupled an existing U(VI) sorption model (in PHREEQC) with U(VI) diffusion calculations (in GLE) to simulate U(VI) mobility in lab-scale through-diffusion experiments over various chemical conditions. Our results suggest that there are two important parameters that govern U(VI) diffusion in these systems. Uranium(VI) surface complexation on clay edge surfaces leads to contaminant retardation, while the access of U(VI) solution species to diffuse layers in clay interlayer spaces controls diffusive fluxes. Hence on the field scale, these parameters will determine how early a plume may cross site boundaries and pose risks to drinking water resources, and the levels of U(VI) concentrations to be expected in contaminated waters.