Sorption studies of actinides onto clay minerals under saline conditions

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Abstract

Actinide retention at clay mineral surfaces is a well established process, which has been intensely investigated in the context of nuclear waste disposal. Up to now, however, no mechanistic sorption model exists describing reliably actinide retention under ionic strength conditions in groundwater upto ~ 5 mol/kg. Such conditions are to be expected e.g. in the vicinity of a repository in rock salt formations, in the Jurassic and lower Cretaceous clay rock layers in Northern Germany or in sedimentary rocks in Canada, all identified as potential host rocks for high level nuclear waste disposal.

Within the present study lanthanide/actinide ion (Eu(III), Cm(III), U(VI)) interaction with montmorillonite and illite in aqueous solution ($I \leq 4.5$ mol/kg NaCl, $CaCl_2$ and $MgCl_2$) in absence of carbonate is investigated. Batch sorption and time-resolved laser fluorescence spectroscopy (TRLFS) experiments are carried out for a given ionic strength at fixed metal concentration and constant solid to liquid ratio for $3 < pH_m < 12$ (for $MgCl_2$ solutions up to pH=8). Highly saline conditions have a surprisingly small impact on K_d—values in the NaCl system. A significant decrease in sorption extent is observed in solutions with elevated Mg/$CaCl_2$ concentrations, notably for the trivalent cations. Nonetheless, K_d values remain high ($\log K_d \geq 4.5$) for all systems in the pH range 7-11. TRLFS studies using Cm(III) as a fluorescent probe point to the invariant metal ion first coordination sphere at given pH_c if ionic strength is increased. As a consequence, we assume no significant change in the speciation of surface sorbed Cm(III) at elevated ionic strength. Sorption data are described using the non-electrostatic 2SPNE/SC CE approach. The impact of elevated ionic strength on dissolved metal ion activity is taken into account by applying the specific ion interaction theory as well as the Pitzer approach.