Effects of calcite impurities in bentonite buffer systems on uranium(VI) solution speciation and sorption behavior

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In many nuclear waste disposal scenarios, bentonite is the proposed buffer material surrounding nuclear waste containers. Its main mineralogical component, Na-montmorillonite clay, is expected to limit radionuclide mobility to slow diffusion processes due to the clay's low porosity and permeability, and the high sorption affinity of clay surface complexation and cation exchange sites for many radionuclides. Given its mineral structure, montmorillonite provides two types of porosities that can be accessible to radioactive contaminants: macropores *between* clay particles and interlayer spaces *within* clay particles, which become dominant at increasing degrees of bentonite compaction. Despite the fact that bentonite naturally contains a variety of mineral impurities (e.g., quartz, feldspars and calcite), most previous studies have only focused on radionuclide interactions with pure Na-montmorillonite phases. Hence, in this study, we will investigate the influence of calcite impurities on uranium(VI) (U(VI)) solution speciation and sorption behavior in bentonite systems.

In the presence of water, calcite impurities are expected to lead to milli-molar concentrations of dissolved calcium over a wide range of pH conditions. Consequently, Ca^{2+} could – at least partially – replace Na^{+} on clay cation exchange sites, with further implications for exchange reactions of cationic U(VI) species. In addition, ternary U(VI)-Ca-carbonate solution species ($CaUO_2(CO_3)_3^{2-}$ and $Ca_2UO_2(CO_3)_3$) will largely replace U(VI)-carbonate species ($UO_2(CO_3)_3^{4-}$ and $UO_2(CO_3)_2^{2-}$) at alkaline pH. These larger, but lower-charged U(VI) species, may show different sorption and diffusion characteristics, e.g. if their access to negatively-charged clay interlayer spaces is different from binary U(VI)-carbonate complexes. In this presentation, we will first demonstrate the impacts of calcite impurities on U(VI) solution speciation based on PHREEQC simulations. We will then discuss first results from U(VI) sorption studies in these systems.

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