Simulation of Uranium(VI) Speciation and Sorption in Montmorillonite Clay in the Presence of Calcite Impurities

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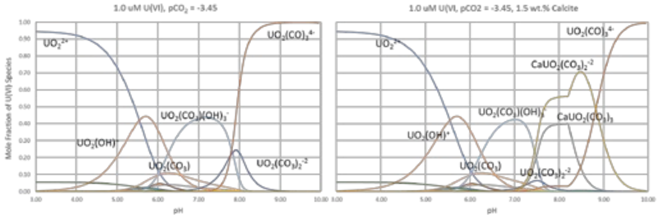
Montmorillonite is an abundant clay mineral that has been identified as the primary component in engineered bentonite barrier systems in future nuclear waste repositories. Selective sorption onto planar and edge surfaces of clay particles make it a great candidate for the attentuation of uranium(VI). Although previous works have investigated uranium(VI) interactions with this clay, few have done so in the presence of calcite, a mineral impurity in bentonite that is understood to alter U(VI) speciation, sorption, and consequently its mobility.

PHREEQC software is used to simulate the chemical solution speciation of U(VI) under various known repository conditions, in the absence and presence of calcite, and across a range of U(VI)concentrations (1.0E-7 to 1.0E-5 M), and partial pressures of CO_2 (zero to 2% CO_2) (Fig. 1). In addition, the kinetic dissolution of calcite is simulated to gain a better understanding of how quickly chemical equilibrium can be reached in these systems. Lastly, surface complexation models will be developed to characterize the influence of calcite impurities on the formation of ternary U(VI)-Ca-carbonate solution species on U(VI) sorption behavior.



Our results indiciate that the presence of Ca^{2+} and CO_3^{2-} ions released from calcite leads to U(VI)-Ca-carbonate species with a negative or neutral charge even without the presence of CO_2. Quantification of these species will allow for the development of more accurate U(VI)-montmorillonite sorption models. Calcite precipitation at alkaline pH further complicates speciation by creating a competation for Ca^{2+}. This research is being performed using funding received from the DOE Office of Nuclear Energy's NEUP.