

257th ACS National Meeting

View Abstract

ABSTRACT SYMPOSIUM NAME: Fundamental Research in Colloids, Surfaces & Nanomaterials

ABSTRACT SYMPOSIUM PROGRAM AREA NAME: COLL

CONTROL ID: 3113412

PRESENTATION TYPE: Poster Only : Consider for Sci-Mix

TITLE: Influence of Calcite on Uranium(VI) Sorption onto Montmorillonite Clay

AUTHORS (FIRST NAME, LAST NAME): Nicolas Hall¹, Allen C. Shaw¹, Diem Quynh N. La¹, Christophe Tournassat², Ruth M. Tinnacher¹

INSTITUTIONS (ALL): 1. Cal State East Bay, Hayward, CA, United States.

2. BRGM Epi/Des, Orleans, France.

ABSTRACT BODY:

Abstract: Currently, most proposed methods for the long-term storage of nuclear waste include engineered buffer systems (EBS) in underground geologic repositories. The design of EBS typically involves a layer of geological buffer material such as bentonite. Bentonite is an ideal material for these systems, since it largely consists of the swelling smectite clay montmorillonite, which provides two types of surface sites available for the sorption of radionuclides, such as uranium(VI) (U(VI)). However, the presence of calcite, a common accessory mineral in bentonite, may potentially alter the sorption of U(VI) onto montmorillonite in multiple ways. First, calcite provides an additional surface for U(VI) sorption reactions. In addition, calcite dissolution will lead to dissolved calcium in porewater solutions, which may in turn affect U(VI) solution speciation, U(VI) ion exchange reactions, and the stacking and interlayer spacing of smectite layers. Last, a potential formation of U(VI)-Ca-carbonate co-precipitates may remove U(VI) from solution.

As a first step, we characterized the sorption of U(VI) onto montmorillonite in the presence and absence of dissolved calcium at various ionic strength conditions in batch sorption envelope experiments. The presence of dissolved calcium resulted in decreased U(VI) sorption onto montmorillonite in acidic environments, but in no apparent changes in U(VI) sorption in near-neutral pH ranges. At higher pH values, there may be a possible decrease in U(VI) sorption, which needs to be further investigated. The observed trends at low pH can be explained by the preferential binding of Ca^{2+} cations relative to UO_2^{2+} , which resulted in a competition between the two cations for clay surface sites. This hypothesis is further supported by the results of additional sorption experiments, which were performed at various ionic strengths, but in the absence of calcium. In a next step, we will expand our experiments to include mixtures of calcite and montmorillonite as solids.

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