Phosphates as an Effective Add-on to Backfill Materials: Immobilization of uranium and iodine by bentonite/phosphate mixtures.

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ABSTRACT:

The aim of this proposal is to advance the functionality of engineered barrier systems (EBS) by investigating how the addition of phosphate minerals (monetite and apatite) to bentonite would increase sorption/uptake efficiency of radionuclides by backfill material in the scenario of water breaching the repository at saturated conditions. This is the next step in solving radionuclides attenuation problem through EBS improvement. During our current NEUP project (Phase-I) we have demonstrated that apatite, an extremely insoluble mineral, has high affinity for iodine (^{129}I, a mobile fission product) and uranium (a primary component of spent nuclear fuel, SNF) uptake, at both ambient and hydrothermal conditions. In addition, our results showed that uptakes of oxidized species (IO_3^- and UO_2^{2+}) by apatite are higher than uptake of reduced species (I^- and U^{4+}). To extend our current NEUP project to the next level, we propose to initiate a set of modelling experiments mimicking fluid flow through phosphate-doped (amended) bentonite engineered barrier. More specifically, we will evaluate the degrees of I and U entrapment in the course of transformation of monetite to apatite in water saturated bentonite matrix via column and batch experiments. Previous experiments on I and U sorption on bentonite have largely limited to adsorption mechanism of radionuclides by phosphate-free bentonite. **We propose to fill this knowledge gap by providing quantified information on UO_2^{2+}, I^-, and IO_3^- uptake by amended bentonite, where phosphates and potentially other newly formed minerals will serve as a permanent storage medium for these radionuclides.** In particular, we intend to assess breakthrough time of I and U and determine the sorption/uptake capacity of amended bentonite at ambient and hydrothermal conditions. To achieve the above goals, we will employ a number of experimental techniques, which include benchtop and hydrothermal crystallization experiments and post experimental characterization with: X-ray diffraction (XRD), scanning electron microscopy (SEM), electron backscattered diffraction (EBSD), inductively coupled plasma mass spectrometry (ICP-MS), ultraviolet–visible (UV-Vis) spectrophotometry, electrochemistry, and atom probe tomography (APT). Expected results will provide novel quantitative information on the interaction of dissolved radionuclides with amended bentonite at different temperatures (elevated from radioactive decay) as an immediate consequence of ground water breaking through EBS. Obtained data will permit us to optimize the composition of a new backfill mixture for improved effectiveness. In addition, the proposed research will improve the current knowledge of the following mineralogical and geochemical aspects critical to nuclear environmental hazard mitigation and remediation: 1) The mechanisms of I and U incorporation into mineral phases and quantitative constants permitting to evaluate this process under a range of relevant conditions; 2) The role of I and U incorporation in formation of associated mineral phases. The proposed collaboration is strongly beneficial for both execution of the project and for study on bentonite interaction with hydrothermal solutions, which is being performed in EES-16 at LANL.