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Project Title: **Microbial Mineral Transformations at the Fe(II)/Fe(III) Redox Boundary for Solid Phase Capture of Strontium and Other Metal/Radionuclide Contaminants**

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Research Objective

Determine microbiological and geochemical controls on carbonate mineral precipitation reactions, and identify contributions of these processes to the solid phase capture of strontium and other metal/radionuclide contaminants. The study is relevant to the development of new clean-up strategies for DOE sites where strontium and other metal/radionuclides exist as ubiquitous and often mobile contaminants.

Research Progress and Implications

The work summarized in this report pertains to the final year of a three-year project investigating the use of bacteria to concentrate and immobilize Sr^{2+} , as well as other metal/radionuclide, contaminants. Major accomplishments over the first two years of the project included completion of sorption studies with bacteria and hydrous iron oxides (HFO), assessment of the impact of Sr^{2+} on bacterial Fe(III)-reduction, induction of carbonate mineral precipitation and solid phase capture of Sr^{2+} under Fe(III)-reducing conditions, and discovery of a procedure to attain rapid high-level concentration of Sr^{2+} in microbiologically produced calcite. In the past year, studies have focused on the influence of ionic strength on Sr^{2+} sorption by bacteria and HFO, the impact of Ca^{2+} as a competing ion in Sr^{2+} immobilization under iron-reducing conditions, and measurement of the extent of Sr^{2+} solid phase capture in saturated porous sand media in the presence of iron-reducing bacteria.

The sorption studies with iron-reducing bacteria and HFO established that the solid phase partitioning of Sr^{2+} at bacterial and HFO surfaces can be quantitatively described using surface complexation theory. These experiments documented further that the scavenging of Sr^{2+} is strongly pH dependent, showing rapid increases in Sr^{2+} sorption as pH increased; however, significant Sr^{2+} sorption occurs at the bacterial surfaces at much lower pH values than at the HFO surfaces, emphasizing their importance for contaminant modeling, and for possible remedial applications. In recent experiments, sorption of Sr^{2+} by bacteria was found to be extremely sensitive to ionic strength, whereas Sr^{2+} sorption by HFO was not affected.

Moreover, composite mixtures of bacteria and HFO were found to exhibit Sr^{2+} sorption behavior intermediate between the pure bacteria and HFO, but showed a lower sorptive capacity than anticipated. This result is explained by a masking of bacterial sorption sites by the surface associated HFO, which has a lower relative sorptive capacity than the bacterial surface.

Other results indicate that the concentration of HFO precipitated at the bacterial surface will ultimately determine the scavenging ability of the bacteria-HFO complex. However, the fundamental concept is that microbial mediated reactions involving dissolved inorganic contaminants must be considered if accurate contaminant fate models are to be developed.

The potential for immobilization of aqueous Sr^{2+} coupled to microbial reduction of a synthetic crystalline Fe(III) oxide (goethite) was examined using a bicarbonate-buffered anaerobic artificial groundwater, containing 1.0 mM, 0.1 mM, or 0.01mM Sr^{2+} . Experiments were also conducted using 10 mM Ca^{2+} simulate a natural Ca-rich groundwater. Although Fe(III) reduction was initially slower in Ca-amended cultures, the overall amount reduction was similar in both systems. In the Ca-containing cultures, there was only modest removal (20-30%) of Sr^{2+} from solution, despite the removal of a significant amount of Ca^{2+} (5-9 mM) through the formation of CaCO_3 . The pH remained constant at 6.8-6.9 in these cultures due to buffering of the system by CaCO_3 precipitation. In contrast, pH rose to much higher levels in the cultures without Ca, and a much greater amount of Sr was immobilized. At all three levels of Sr in the absence of Ca^{2+} , 90 % of aqueous Sr was removed after 51 days of incubation. Between 10 and 15 mmol/L solid phase carbonate was formed in the cultures, which presumably accounted for the Sr^{2+} immobilized during Fe(III) oxide reduction. In the 0.1 mM and 0.01mM Sr cultures there was an initial release of sorbed Sr as Fe(III) was reduced, followed by subsequent Sr immobilization. In the 1.0 mM Sr^{2+} - Ca^{2+} -free cultures, there was immediate solid-phase capture of Sr^{2+} . This initial sorption and release was not observed in the 1.0 mM Sr systems due to saturation of sorption sites.

In saturated sand, the extent of Fe (III) reduction was not changed; however, sand enhanced the rate and extent of subsequent removal of Sr^{2+} from solution. These findings demonstrate the potential for solid-phase capture of Sr during carbonate formation coupled to microbial reduction of HFO and crystalline Fe(III) oxide which are common to many soil and subsurface sedimentary environments. This process may provide a mechanism for retarding the migration of Sr in carbonate-rich groundwater aquifers.

Demonstration of Sr^{2+} solid phase capture during Fe(III) reduction suggests that considerable potential exists to adapt this novel immobilization concept to the development of new clean-up strategies for DOE sites where strontium and other metal/radionuclides exist as ubiquitous and often mobile contaminants. Further laboratory and field investigations focused on assesment of performance under *in situ* conditions are considered to be the most appropriate course of action.

Planned Activities

There are two graduate students presently working on the project that are in the final stages of their thesis research (one each at the University of Alabama and the University of

Toronto). Several manuscripts are expected to be prepared for journal submission based on the work of these students. In order for these students to complete their work and write papers, a no cost extension has been requested. Two new EMSP proposals have been submitted based on the results of this project, one through the University of Alabama and the other through INEEL where groundwater and vadose zone Sr^{2+} contamination is recognized as a problem.

Information Access

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