Biotic controls on uranium sequestration and release by framboidal pyrite in bioreduced sediments

University-Led Research

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Uranium-containing framboidal pyrite, recently discovered in both IFRC/SFA sites at Rifle and Hanford, may play a significant role in controlling the fate and transport of subsurface contaminant U(VI). Biofilm-associated bacteria present on the framboidal pyrite surfaces could potentially influence the fate of uranium through enzymatic redox processes in the subsurface. Alternatively, framboidal pyrite-associated microbes could also play an indirect role in governing U(VI) bioreduction by catalyzing the initial formation of framboidal pyrite via iron-monosulfide precursors that can subsequently engage in non-enzymatic redox processes leading to U reduction and immobilization. Identifying framboidal pyrite-associated microbes and their distribution in biofilms may provide key insight into electron transfer processes driving U uptake and/or release.

An initial batch microcosm study was conducted to examine the relationship between biostimulants (lactate and acetate), pyrite (non-framboidal) amendment, and the microbial community dynamics using Rifle background sediment that was initially devoid of pyrite. A subsequent study will be conducted using synthetic-framboidal pyrite to amend the batch microcosms and compared with naturally bioreduced sediments to gain insight into the broader indigenous microbial community associated with framboidal pyrite. Several chemical synthesis methods involving aging of amorphous iron monosulfide are being employed to reproducibly generate substantial yields of pure framboidal pyrite for additional microcosm and parallel column studies. A key challenge in most synthesis methods involves scaling-up the production and ensuring phase purity. Flow-through column studies are being designed to evaluate uranium uptake and release by framboidal pyrite and compare uranium sorption and desorption behavior to the field site. Multivariate and statistical approaches applied to analyze microbial communities observed in the Rifle sediment and column studies will identify key groups of microbes associated with framboidal pyrite and help explain uranium removal under different test conditions. The biofilms formed on the surface of framboidal pyrite will also be visualized using fluorescence in-situ hybridization (FISH) and confocal microscopy. Additionally, advanced microscopy and surface-sensitive spectroscopic techniques will be applied to probe molecular-scale redox interactions between framboidal pyrite and uranium. This exploratory research effort will help gain an understanding of interfacial biogeochemical processes impacting framboidal pyrite, the microbial population and activity, and U interactions in heterogeneous natural systems. This new insight in turn will aid in the development of molecular models to predict the rate and extent of U attenuation, as well as its long-term mobility, in bioreduced sediments. Further, the ability to stimulate these interactions may offer a new remediation strategy for DOE sites.