Acceptable Endpoints for Metals and Radionuclides: Quantifying the Stability of Uranium and Lead Immobilized Under Sulfate Reducing Conditions

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The creation of sulfate-reducing conditions to immobilize metals has potential use at many contaminated sites because of the large number of metals (e.g., Hg, Pb, Cd, Cu, Ni, Zn) that form stable sulfide compounds. In addition, effective reduction and subsequent precipitation of U and Cr under these conditions has been shown. However, as with other possible treatments, the long-term stability of the immobilized metals and radionuclides and the factors that affect the immobilization/remobilization process must be quantified to determine whether the treatment can produce an acceptable endpoint.

Methods have been developed to test the hypothesis that the rate of metal remobilization will largely depend on the following factors: (1) aquifer reductive capacity (generated in the form of iron sulfides); (2) distribution and morphology of contaminant precipitates on mineral surfaces; and (3) accessibility of contaminant precipitates to dissolved oxygen.

Experiments using a new fluidized-bed "V-bottom" reactor that examine SRB growth and attachment to grains of quartz and feldspar are in progress. In preliminary anaerobic batch studies using lactate-C medium and quartz, significantly more (2x) growth occurred when quartz was present.

A green fluorescent protein (GFP), from the jellyfish *Aequorea victoria*, was introduced into *Desulfovibrio desulfuricans* for direct observation of SRB on hematite using fluorescence microscopy. Unlike fluorescent stains, the GFP allows non-destructive, real-time observation of active SRB colonization and biofilm growth on mineral surfaces (e.g., hematite) in both pure and mixed cultures. The GFP allows us to register the locations of SRB activity without disturbing the progress of experiments.

As another measure of SRB activity in mixed culture biofilms, microelectrode measurements of colony-scale H_2S profiles on surfaces will also be presented. Biofilms composed of *Desulfovibrio desulfuricans* and *Pseudomonas fluorescens* were grown on inert glass surfaces. We have measured profiles of H_2S, pH and local effective diffusivity at two locations in a cell cluster: at the center and near the edge, and at three locations in interstitial voids: in a large void (about 100 microns wide), open to the flow and parallel to the flow direction, in a mid size void, open and perpendicular to the flow direction, and in a small, closed void. The pH in the biofilm was nearly constant, around 7.2, (results not shown). H_2S concentration varied significantly among locations, indicating a heterogeneity that may be significant in the presence of redox-sensitive minerals.

Specially designed flat-plate flow cells that contain pure and mixed redox-sensitive and insensitive minerals allow us to determine the spatial relationships between SRB colony location and activity, and the location and type of contaminant and sulfide mineral deposits on a hematite, or other aquifer-relevant surface. A hematite coupon surface was exposed to SRB colonization for 17 days, then it was rinsed and examined using X-ray photoelectron spectroscopy (XPS) for surface characterization. XPS data indicate the formation of reduced iron sulfide on the hematite surface and also the presence of numerous sulfur species, including S^{2-}, S_2^{2-}, S_n^{2-}, SO_3^{2-} and SO_4^{2-}; however, no elemental S or thiosulfate were detected. Our initial results indicate the possible formation of pyrrhotite ($Fe_{1-x}S$, where $0< x < 0.125$) on hematite surfaces when SRBs are grown. We are currently replicating this test and also plan to confirm the result using grazing angle XRD. If confirmed, the presence of pyrrhotite contrasts with the results of most other work in homogeneous SRB solutions where the metastable sulfide phase is mackinawite (FeS_{1-x}, where $0 < x < 0.1$). Mackinawite is a precursor to both of the thermodynamically stable high-temperature iron-sulfide phases [pyrite (FeS_2) and pyrrhotite].