Impact of nano-scale iron sulfides on abiotic oxidative dissolution of UO₂

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Nano-scale iron sulfide precipitates formed under sulfate reducing conditions may buffer U(IV) insoluble solid phases from re-oxidation when oxidants reenter the reducing zone. In this study [1], sediment column experiments were performed to quantify the effect of biogenic mackinawite on U(IV) stability in the presence of nitrite or dissolved oxygen (DO). Two columns, packed with sediment from abandoned U contaminated mill tailings site near Rifle CO, were biostimulated for 62 days with an electron donor (3 mM acetate) in the presence (BRS+) and absence (BRS-) of 7 mM sulfate. The bioreduced sediment was supplemented with synthetic uraninite $(UO_2(s))$, sterilized by gamma-irradiated, and then subjected to sequential oxidation by nitrite and DO. Biogenic iron sulfides produced in the BRS+ column, mostly as mackinawite, inhibited U(IV) reoxidation and mobilization by both nitrite and oxygen. Most of the influent nitrite (0.53 mM) exited the columns without oxidizing UO₂, while a small amount of nitrite was consumed by iron sulfides precipitates. An additional 10-day supply of 0.25 mM DO influent resulted in the release of about 10% and 49% of total U in BRS+ and BRS- columns, respectively. Influent DO was effectively consumed by biogenic iron sulfides in the BRS+ column, while DO and a large U spike were detected after only a brief period in the effluent in the BRS- column. Α mechanistic interpretation for the inhibited UO₂ dissolution by iron sulfides will be provided based on the recently completed continuously-mixed flow-through reactor studies [2] [3] using synthetic FeS and UO₂ nanoparticles.

[1] Carpenter, Bi, & Hayes (2015), *ES&T* 49, 1075-1085.[2] Bi & Hayes (2014), *ES&T* 48, 632-640. [3] Bi & Hayes (2014), *ES&T* 48, 13402–13411.