Field-Scale Demonstration of In Situ Biological Uranium(VI) Reduction at Area 3, NABIR Field Research Center, Oak Ridge, TN

W. Wu[1], M. Ginder-Volgel[1], M. Fiene[1], J. Nyman[1], M. Gentile[1], J. Luo[1], O. Cirpka[1], S. Fendorf[1], P. Kitanidis[1], C. Criddle (PI)[1], J. Carley[2], T.Gentry[2], T. Mehlhorn[2], S. Caroll[2], H. Yan[2], J. Zhou[2], B. Gu[2], D. Watson[2], P. M. Jardine , and M. Fields[3]

[1]Stanford University, Stanford, CA; [2]Oak Ridge National Laboratory, Oak Ridge, TN; [3]Miami University, Oxford, OH

Mixed wastes and extreme conditions pose a significant challenge for remediation of uranium-contaminated soils at many contaminated sites. We have evaluated the potential for field-scale in situ biological reduction of uranium(VI) in Area 3 of the DOE NABIR Field Research Center in Oak Ridge, TN. At this site, high levels of uranium are present in the groundwater (~60 mg/L) and soil (up to 700–1,000 mg/kg). Many other contaminants are also present. Notably, the groundwater contains high levels of nitrate (8–10 g/L), calcium (0.9 g/L), and aluminum (0.5 g/L) at a pH of 3.4. To evaluate options for site remediation, we carried out slurry and column experiments. These studies suggested the feasibility of a three-step approach: (1) flushing with a low pH water to remove aluminum and bulk nitrate; (2) adjustment of pH to a range favorable for microbial activity; and (3) ethanol addition to stimulate U(VI) reduction. This strategy was then implemented at the field site, using an aboveground treatment system to remove nitrate, calcium, and aluminum, and to adjust pH, and belowground recirculation wells for ethanol addition.

The field experiment began with a clean-water flush. Water from the subsurface was extracted and treated to remove Al, Ca, and nitrate, supplemented with tap water and injected into the well system. The pH of the injected water was then gradually increased. After three months, nitrate concentrations had fallen to less than 1.0 mM. In situ bioremediation was then initiated by intermittent ethanol addition. A vacuum stripper was installed between the innerloop extraction well and injection wells to remove nitrogen gas created by denitrification of residual nitrate. Immediately after ethanol injection, nitrate levels dropped to 0.3 mM or lower. After three months of ethanol addition, ethanol addition was accompanied by a reduction in aqueous U(VI). Sulfate consumption and sulfide formation also occurred concomitantly. A tracer test with bromide and ethanol confirmed that the removal of ethanol was due to biological activity. Most Probable Number (MPN) analyses of the groundwater from extraction well and multiple level sampling (MLS) wells indicated the growth of denitrifying bacteria, sulfate-reducing bacteria (SRB) and iron-reducing bacteria (FeRB) in the subsurface. In the extraction well, recent cell numbers (cell/mL) are: denitrifiers, 107; SRB, 104; and FeRB, 105. More SRB and FeRB were detected in MLS wells. During biostimulation operations, we generally maintained the pH of the well system at 5.8–6.0 to prevent growth of methanogens. The growth of these organisms is of concern because they compete for ethanol with U(VI)-reducing bacteria and they produce methane, a gas that could clog the aquifer. To date, we have not detected methanogenesis.

Chemical clogging by aluminum precipitates was observed in the injection and extraction wells during the initial phases of operation. Biological clogging was observed during biostimulation. To restore hydraulic conductivity, we moved surge blocks up and down within the clogged wells. This operation was repeated as needed, with a frequency of about once every 1.5 months. This cleaning operation released biomass and sediment from around the well screens and into the well itself. The resulting slurry was removed by pumping. X-ray absorption near edge structure spectroscopy of the slurry solids confirmed that U(VI) was reduced to U(IV). After 9 months of operation, up to 42% of the uranium in injection-well slurry samples was present as U(IV); uranium in extraction well slurry samples remained as U (VI).