Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.8103

Alteration of uranyl-phosphate source from environmental exposure

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The formation of uranyl-phosphate precipitate is a remediation strategy to limit the mobility of uranium in contaminated soils under oxidizing conditions. This study uses a combination of field lysimeters and laboratory studies to explore the changes in the uranyl-phosphate mineral chernikovite [$(H_3O)(UO_2)(PO_4)•3H_2O$] from exposure to soil, natural weather conditions, and the organic ligand citrate to gain a better understanding of the geophysical, chemical, and biological processes that control the fate of this uranium source in the sub-surface.

Synthesized chernikovite sources were placed in the middle of lysimeter columns that were packed with a one-part sand and one-part sandy clay loam sediment representative of the soil found at the U.S Department of Energy Savanah River Site. The sources were characterized before deployment using x-ray diffraction (XRD) and scanning transmission electron microscopy (STEM) coupled with energy dispersive spectroscopy (EDS). After one year of exposure, the sources were recovered, and the solid phase characterization revealed an altered uranyl-phosphate compared to the starting chernikovite source. Peak shifts and additional peaks in the XRD diffraction pattern suggest possible changes in the crystal structure of the source. STEM images show a weathered more heterogeneous morphology than the initial source, and the EDS analysis reveal an increase in potassium associated with the recovered uranyl-phosphate. Based on these findings, we propose that cations from the soil interact with the uranyl-phosphate source and alter the mineral phase.

Preliminary results have shown that organic ligands promote the dissolution of the initial chernikovite material. The ligand citrate, with a concentration as low as 0.1 mM, increased dissolved uranium concentrations by a factor of 4.9 in batch experiments. Therefore, batch dissolution experiments were performed using the recovered uranyl-phosphate to determine the effect of these observed changes on the solubility and citrate-promoted dissolution of the altered uranyl-phosphate compared to the starting chernikovite. These results provide valuable insight on the long-term stability of uranyl-phosphate in the environment.