

Siderophore and oxalate mediated desorption of uranyl from goethite

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Uranium is the major contaminant in soils and groundwaters at sites associated with the processing and disposal of materials during nuclear energy and nuclear weapons production. The immobilization and removal of radionuclide contaminants in soils by microbial transformations, sorption and mineralization show the remediation potential some natural microbes may have.

In this study uranyl (UO_2^{2+}) batch sorption and desorption experiments on goethite surfaces were performed in the presence and absence of organic ligands to estimate their effect on the mobility of U in soils. The ligands were oxalate and desferrioxamine B (DFO-B). The latter is a strong iron chelator produced by bacteria and other microbes to overcome low iron availability but which also complexes actinides. As model soil constituent a synthetic goethite with a BET specific surface area of $34.4 \text{ m}^2/\text{g}$ was employed.

Experiments were performed at pH 6 (buffered with MES) in a NaNO_3 background electrolyte solution of 10mM. The goethite concentration was set at 0.5 g/L and the adsorbate concentration ranged between 24 ppb and 12 ppm. Sorption of uranyl under these circumstances is nearly 100% and the surface concentration of U(VI) is a function of the amount of adsorbent in the solution.

Desorption of uranyl by DFO-B versus time and ligand concentration reveals desorption to be immediate and the U recovery rate reaches a plateau at ~60% of the initial U(VI) concentration irrespective of any further increase in the aqueous DFO-B concentration. Comparing the desorptive effect of DFO-B with oxalate on an equimolar basis (240 μM), oxalate is a more effective desorbent than the siderophore. However, it might be rather dissolution of the underlying goethite surface than desorption that causes detachment of uranyl molecules. Dissolution experiments of goethite in the presence of oxalate or DFO-B anions reveal that oxalate shows hardly any dissolution promoting effect at low concentrations (< 200 μM) whereas DFO-B significantly enhances dissolution of goethite at any concentration (13-1200 μM). At high ligand concentrations (>2000 μM), however, oxalate out-competes DFO-B in the dissolution of goethite and this observation might explain the higher uranyl detachment with oxalate. U(VI) desorption with DFO-B is not related to an increase in the aqueous Fe content indicating that goethite dissolution takes place at uranyl-free sites.