Radionuclide uptake during the formation and crystallisation of ferrihydrite

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Ferrihydrite is the most common iron oxyhydroxide found in soil and is a key sequester of contaminants in the environment. Ferrihydrite formation is also a common component of many treatment processes for cleanup of industrial effluents. The pathways of ferrihydrite nanoparticle formation and contaminant sequestration are numerous and complex. It is critical to understanding these processes at the molecular scale to underpin our knowledge of ferrihydrites environmental behaviour and technological applications.

In this study we have utilised a number of synchrotron-based techniques to determine the mechanisms by which ferrihydrite nanoparticles form, and the role of Fe_{13} Keggin clusters, during the controlled hydrolysis of an acidic ferric nitrate solution in an experimental protocol designed to mimic ferric oxyhydroxide effluent treatment systems. In addition, we have characterised the uptake and sequestration of uranium during the ferrihydrite precipitation and crystallisation process. *In situ* SAXS measurements supported by TEM indicate that ferrihydrite nanoparticles form via Fe_{13} Keggin prenucleation clusters (radius ~ 0.45 nm) leading to the formation of mass fractal aggregates of ferrihydrite nanoparticles (~3 nm) in which the Fe_{13} Keggin motif is preserved. During the ferrihydrite formation process uranium adsorbs to the surface of the nanoparticles. EXAFS spectroscopy indicates that the uranium initially forms bidentate edge-sharing adsorption complexes at the ferrihydrite surface. During crystallisation the uranium speciation changes with a significant proportion becoming incorporated into the crystalline phase.

The results from these studies have direct relevance to the fundamental understanding of ferrihydrite formation and contaminant sequestration in environmental, engineered, and industrial processes.

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