



Contribution ID: 45

Type: Oral

XAS studies of radionuclide incorporation into mineral phases

Wednesday 21 May 2014 16:25 (25 minutes)

Many countries have a legacy of radioactively contaminated land and nuclear wastes. Current policy is to dispose of waste in a geological disposal facility, and decommission legacy nuclear sites over the next decades. These processes could lead to the release of radionuclides (e.g. ^{137}Cs and Tc). Radionuclide mobility in the environment is often controlled by adsorption onto, and incorporation into mineral phases, which can significantly limit their transport and bioavailability. Key to understanding the long-term behaviour of radionuclides within environmental systems is a molecular-scale understanding of the mechanisms by which they become incorporated into minerals.

In this study the incorporation of U, Tc and Cs into iron oxide (e.g. hematite and magnetite) or aluminosilicate clay (e.g. illite) minerals have been characterised at the atomic scale using X-ray Absorption Spectroscopy (XAS) in conjunction with aberration corrected (scanning) transmission electron microscopy ((S)TEM), and density functional theory modelling. The results of this work provide a new mechanistic understanding of U and Tc incorporation into hematite and magnetite, respectively. We show that U(VI) and Tc(IV) substitute directly for Fe within the iron oxides. Firstly, U(VI) was incorporated into a distorted, octahedrally coordinated site replacing Fe(III) in hematite. XAS showed the uranyl bonds lengthened from 1.81 to 1.87 Å, in contrast to previous studies. Secondly, Tc(IV) was incorporated into octahedrally coordinated sites replacing Fe(III) in magnetite, with the majority of the Tc located near the particle surface. In addition, XAS and STEM analysis showed that following initial sorption onto the frayed edges, Cs migrates into the illite interlayer becoming incorporated. This process is irreversible as Cs is held in the collapsed interlayers and is not exchangeable. During longer term experiments Cs did not remain at the edge of the illite particle, but migrated into the collapsed interlayer through exchange with K.

Primary author: Dr SHAW, Sam (University of Manchester)

Co-authors: Mr FULLER, Adam (University of Leeds); Prof. LIVENS, Francis (University of Manchester); Prof. MOSSELMANS, Frederick (Diamond Light Source); Dr LAW, Gareth (University of Manchester); Dr BURKE, Ian (University of Leeds); Prof. MORRIS, Katherine (The University of Manchester); Mr MARSHALL, Timothy (University of Manchester)

Presenter: Dr SHAW, Sam (University of Manchester)

Session Classification: Actinides in Environmental and Life Sciences

Track Classification: Actinides in Environmental and Life Sciences