Characterising legacy spent nuclear fuel pond materials using microfocus X-ray absorption spectroscopy

W. R. BOWER1,2,3, K. MORRIS1, J. F. W. MOSSELMANS3, O. R. THOMPSON2, A. BANFORD2,4, K. LAW5, R.A.D. PATTRICK1

1Research Centre for Radwaste Disposal, The University of Manchester, Manchester M13 9PL william.bower@manchester.ac.uk
2National Nuclear Laboratory, Chadwick House, Warrington Road, Birchwood Park, Warrington, WA3 6AE
3Diamond Light Source, Harwell Science and Innovation Campus, Didcot, Oxfordshire OX11 0DE
4School of Chemical Engineering and Analytical Science, The University of Manchester, Manchester, M13 9PL
5Centre for Radiochemistry Research, The University of Manchester, Manchester M13 9PL

A core of radiouclide contaminated, coated and painted concrete taken from the decommissioned, spent nuclear fuel cooling pond wall at the Hunterston-A nuclear site (Ayreshire, UK) has provided a unique opportunity to study strontium and caesium behaviour in a real system. The core, extracted from a dividing wall at a few cm above the original fill level of the pond, exhibited radioactivity (dominantly from $^{137}\text{Cs}$ and $^{90}\text{Sr}$) which was heterogeneously distributed across both painted faces. Autoradiography suggests that activity was splashed onto the surface.

At beamline I18 of the Diamond Light Source (UK), the Sr distribution across the painted core surface was mapped using microfocus X-ray fluorescence ($\mu$XRF) and this was combined with Sr K-edge XANES. This revealed that Sr on the painted core is bound to regions of high TiO$_2$ (the dominant pigment in the paint) and XAS spectra exhibit similarities with outer-sphere complexation of Sr on TiO$_2$ reported elsewhere. Stable Sr and Cs reactivity experiments were also undertaken using uncontaminated blocks of the concrete in order to analyse the interactions of these elements as proxies for $^{137}\text{Cs}$ and $^{90}\text{Sr}$ in case of a failure in the surface sealants. Using $\mu$XRF and scanning electron microscopy, Sr was shown to be immobilized by the cement phases, whilst at the concentrations used, Cs was taken up by clay minerals within the altered igneous rocks clasts in the concrete aggregate.

Studies of challenging real world samples such as this are crucial to ensure the safe management of nuclear wastes and limit the uncertainties associated with post-operational clean out and decommissioning.