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

W.R. Bower[a,b,e], K. Morris[a], J.F.W. Mosselmans[c], O.R. Thompson[b], A.W. Banford[b,d], K. Law[e], R.A.D. Pattrick[a,*]

[a] Research Centre for Radwaste Disposal, School of Earth and Environmental Sciences, The University of Manchester, Oxford Road, Manchester M13 9PL, UK
[b] National Nuclear Laboratory, Chadwick House, Warrington Road, Birchwood Park, Warrington, WA3 6AE, UK
[c] Diamond Light Source, Harwell Science and Innovation Campus, Didcot, Oxfordshire OX11 0DE, UK
[d] School of Chemical Engineering and Analytical Science, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK
[e] Centre for Radiochemistry Research, Chemistry Building, The University of Manchester, Brunswick Street, Manchester M13 9PL, UK

## HIGHLIGHTS

- A sample from a spent fuel pond wall has been analysed via X-ray spectroscopy.
- Autoradiography shows a patchy distribution of radioactivity on the core face.
- μXAS across a 'hot spot' showed Sr associates with the $TiO_2$ pigment in the paint.
- Original concrete coatings prove effective at limiting radionuclide migration.
- Sorption studies show Sr immobilisation by the concrete and Cs by aggregate clasts.

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## ABSTRACT

Analysis of a radioactive, coated concrete core from the decommissioned, spent nuclear fuel cooling pond at the Hunterston-A nuclear site (UK) has provided a unique opportunity to study radionuclides within a real-world system. The core, obtained from a dividing wall and sampled at the fill level of the pond, exhibited radioactivity (dominantly $^{137}Cs$ and $^{90}Sr$) heterogeneously distributed across both painted faces. Chemical analysis of the core was undertaken using microfocus spectroscopy at Diamond Light Source, UK. Mapping of Sr across the surface coatings using microfocus X-ray fluorescence (μXRF) combined with X-ray absorption spectroscopy showed that Sr was bound to $TiO_2$ particles in the paint layers, suggesting an association between $TiO_2$ and radiostrontium. Stable Sr and Cs sorption experiments using concrete coupons were also undertaken to assess their interactions with the bulk concrete in case of a breach in the coating layers. μXRF and scanning electron microscopy showed that Sr was immobilized by the cement phases, whilst at the elevated experimental concentrations, Cs was associated with clay minerals in the aggregates. This study provides a crucial insight into poorly understood infrastructural contamination in complex systems and is directly applicable to the UK's nuclear decommissioning efforts.


## 1. Introduction

Decommissioning of legacy nuclear installations and the safe disposal of the resulting radioactive wastes is a significant challenge for many nuclear nations. Current estimates for the UK alone suggest that nuclear decommissioning will cost >$100 billion with a timescale of 100 years+ [1]. In the UK, power station and fuel handling facility wastes will be dominated in terms of their volume by cementitious materials, with an estimated total of ~2.5 million tonnes [2]. Waste minimisation, to limit the volume of radioactively contaminated material requiring disposal as low and intermediate level wastes, will be essential to maximise disposal capacity [3]. This can be achieved by effective decontamination and/or segregation of materials to minimise the radioactively contaminated fraction, provided the underpinning knowledge is available to inform these operations. Storage ponds for highly radioactive spent fuel are often constructed from concrete, with the dominant dose-yielding radioactive isotopes in the ponds being $^{137}Cs$ and $^{90}Sr$. A fundamental understanding of the interactions of these radionuclides (or their stable analogues) with the pond walls and

* Corresponding author.
*E-mail address:* richard.pattrick@manchester.ac.uk (R.A.D. Pattrick).

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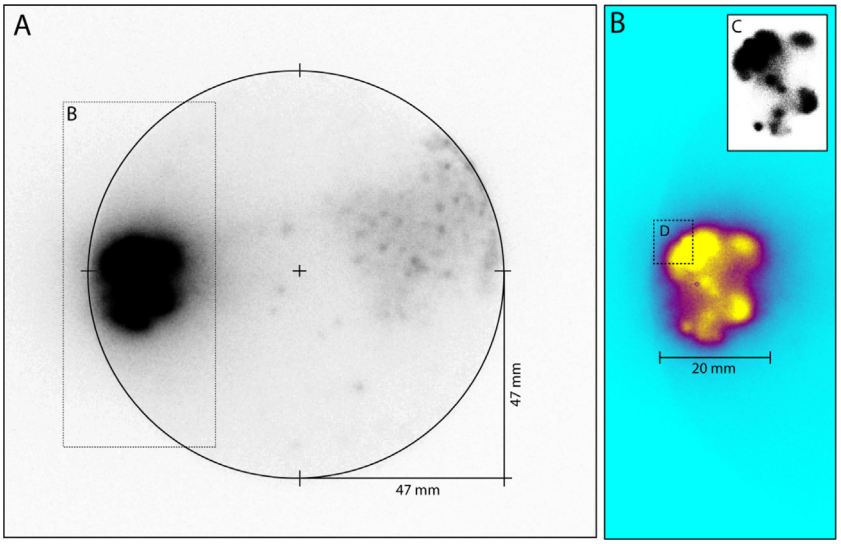


**Fig. 1.** A. Low resolution (50 μm$^2$ pixels), autoradiograph (24 h) showing areas of radioactivity from the most active face of the core. B. False-color, contrast adjusted image of region 'B'. C. High resolution (25 μm$^2$ pixel size) image over 24 h, contrast adjusted to highlight spots of elevated activity. Area 'D' was selected for spectroscopic analysis.

their coatings has the potential to significantly reduce decommissioning waste volumes along with associated disposal costs and therefore is the focus of this work.

### 1.1. Hunterston-A core sample

Hunterston-A, which ceased power generation in 1990, was a twin MAGNOX power reactor (with spent fuel storage ponds on site) located in Ayrshire, Scotland. After closure, post-operational clean-out (POCO) and decommissioning activities have been ongoing. In approximately 2013, the spent fuel storage ponds were drained and the opportunity arose to sample a unique series of cores from the pond walls. Specifically, a core was extracted from the level of the original water surface in the fuel pond. This study has exploited a range of analyses, in particular advanced spectroscopic techniques, to define likely radionuclide speciation and binding in the fuel pond wall sample and characterise the poorly understood infrastructural sealants.

The core sample (drilled perpendicular to the wall face and through the dividing wall) was sampled from a fuel pond

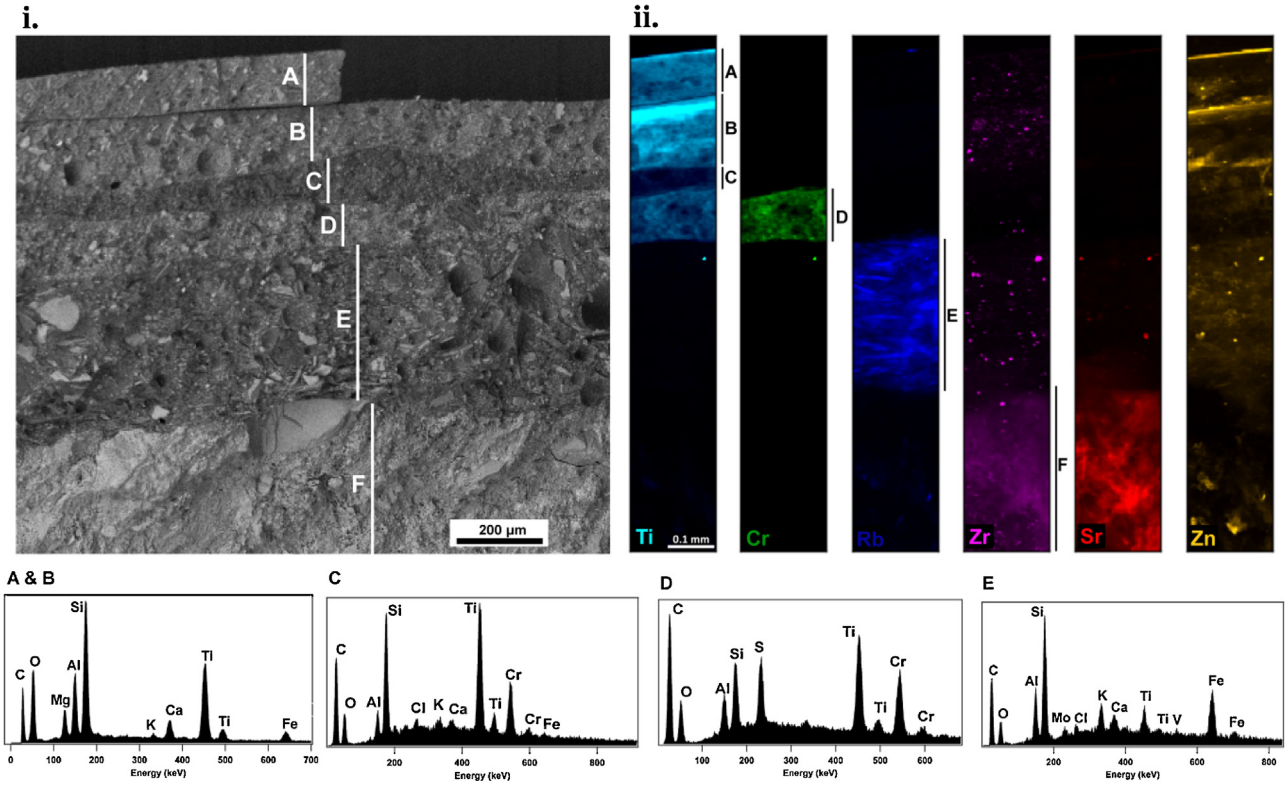


**Fig. 2.** i. ESEM BSE image showing a section through the core coating to the underlying concrete. Layers A & B are paint, layer C an undercoat, layer D paint and layer E chlorinated rubber seal with layer F the underlying concrete. EDAX spectra from representative points across each layer are presented below the images. **ii.** μXRF maps from beamline I18 (Ti, Cr, Rb, Zr, Sr & Zn) along the same cross section. The Ti XANES displayed in Fig. 4 was collected from region A.