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Characterisation of simulant Chernobyl nuclear meltdown materials

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1 Introduction

In order to support decommissioning operation at the Chernobyl nuclear power plant (NPP) work is being conducted to understand the properties of materials formed during nuclear meltdowns. In this study small-scale batch samples of lave-like fuel containing materials (LFCM) have been prepared and characterized by X-ray absorption spectroscopy (XAS). XAS is a powerful technique for investigating local physicochemical properties, such as valence state and local coordination environment, which will have an influence on important material characteristics such as the durability. Comparison is made with data acquired from material retrieved from the Chernobyl NPP to provide some insight into the suitability of these synthetic samples as simulants.

2 Experiment

Synthetic LFCM was prepared from reagent grade precursor to match bulk chemical analyses reported in the literature [1-3]. The sample was melted in an alumina crucible under reducing conditions (5% H_2 – 95% N_2) at 1500 °C for 4 hours and annealed at 720 °C for 72 hours. U L_{III} edge XANES spectra of the synthetic LFCM sample was measured in transmission mode on BL-27 at the Photon Factory (Tsukuba, Japan). To aid the analysis standards with known oxidation state (UO_2 – U^{4+}; $Yb_{0.5}U_{0.5}Ti_2O_6$ – U^{5+}; $CaUO_4$ – U^{6+}) were also measured. All the samples were prepared by mixing finely ground powders with polyethylene glycol (PEG) and pressing into 13mm diameter disks. Subsequent data reduction and analysis was performed using the Athena software package [4].

3 Results and Discussion

Figure 1 shows the U L_{III} edge XANES data for the synthetic LFCM sample. For comparison the data from the standards are also shown. The position of the absorption edge (determined as the energy position at a normalised absorption of 0.5) was similar to the value measured from the UO_2 standard which suggested that the average uranium oxidation state in both samples is close to four (Figure 2). There are also marked similarities in the features in the post edge range which indicated that the local coordination environment of uranium in the synthetic LFCM sample was similar to that in UO_2 (VIII-fold). Previous studies on real LFCM particles has indicated that the uranium tended to partition into zircon crystals often referred as chernobylite ($Zr_{1-x}U_xSiO_4$) [5, 6]. The local structural environment around the Zr cations in zircon is VIII-fold [7], which is consistent with the findings in this study, suggesting that our synthetic sample is a promising analogue for real LFCMs.

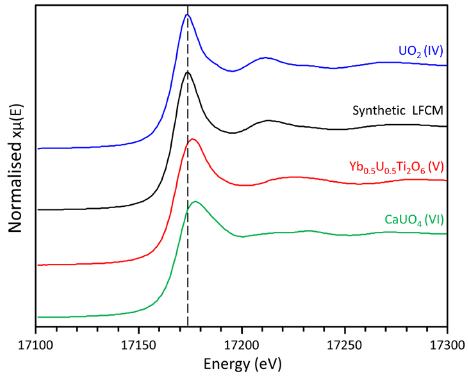

Fig. 1: Uranium LIII edge XANES data for synthetic LFCM, with reference to uranium standards of known oxidation state.

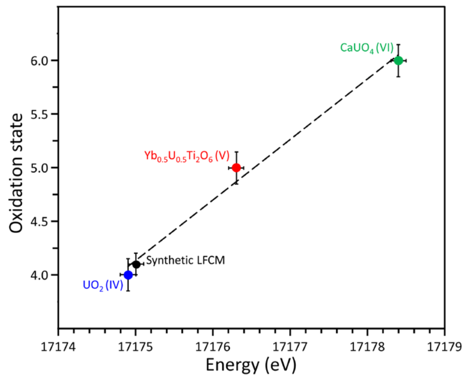

Fig. 2: Linear regression of the extracted normalised edge position of standards and LFCM sample.

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