X-ray Diffraction Studies on Surrogates for Americium Oxides for European Radioisotope Power Systems

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Abstract. European Radioisotope Power Systems will be fuelled with an americium oxide heat source. This will require integral discs or pellets to be created with reproducible geometry, high relative density and well characterised mechanical properties. With few published investigations on americium oxides, conducting sintering studies with surrogate oxides helps to inform sintering trials with highly radioactive americium oxides by reducing the field of investigation.

Understanding the crystallography of a surrogate oxide prior to and post sintering is essential to understanding the crystallographic changes that may occur during the densification process. Such changes could impact disc/pellet integrity. Americium dioxide ($AmO_{2.00}$) loses oxygen at high temperatures, which can lead to sub-stoichiometric materials of the type $AmO_{2-(x/2)}$ where the americium is present as a mixture Am^{3+} and Am^{+4} ions occupying crystal lattice sites. It is in theory possible to make $CeO_{2-(x/2)}$ solids with similar mixed oxidation sites, however, these solids easily re-oxidise in air. To replicate this arrangement, non-radioactive surrogate materials have been used to simulate the +3 and +4 metal sites. Neodymium, Nd^{3+} and cerium, Ce^{4+} mixed oxides can be made by co-precipitating cerium and neodymium oxalate solids and decomposing them to the oxide $Ce_{1-x}Nd_xO_{2-(x/2)}$. These solids are stable in air and allow the crystal properties to be examined in detail.

Earlier studies outlined the synthesis and characterisation of a cubic (Ia-3) cerium-neodymium oxide, $Ce_{1-x}Nd_xO_{2-(x/2)}$, as a surrogate for certain $AmO_{2-(x/2)}$ species between $AmO_{1.65}$ and $AmO_{1.75}$, and also showed early X-ray diffraction (XRD) analysis results of spark plasma sintered (SPS) CeO_2. Quantitative X-ray fluorescence analysis of the Ce Nd oxides suggested they had an x-value with a small discrepancy (0.02) from the initial nominal target ratio of 0.60 dictated by the molar ratio of the Ce and Nd nitrate reagent inputs to the oxalate precipitation process. Initial XRD of the SPS discs indicated consistency with CeO_2 yet the grey colours suggested some reduction. Detailed XRD analysis (Rietveld refinement) results of the Ce-Nd oxide has been conducted and data are presented. An x-value for the material has been estimated from the lattice parameter using published Vegard-like laws for $Ce_{1-x}Nd_xO_{2-(x/2)}$ specific solid solutions. The results have been compared with the quantitative X-ray fluorescence results and checked for consistency. The SPS discs Rietveld refinement results are also presented. Lattice parameters have been measured and compared to CeO_2 to determine if there is an expansion that would be consistent with small amounts of reduction.

Keywords: cerium-neodymium oxide, cerium dioxide, surrogates, X-ray diffraction, americium.