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Examination of High Temperature Structural Phase Transformations in Strontium Uranium Oxides Using Synchrotron X-ray Diffraction and Spectroscopy

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Interest in nuclear power has recently increased as many argue it will play a pivotal role in the transition away from fossil fuel energy. However, little progress has been made regarding the understanding of the fundamental solid state chemistry of UO_2 fuel matrices, the interaction they have with fission daughters such as Sr-90, and the solid-phases that form during operation of reactors. This has resulted in significant challenges for nuclear waste immobilisation methods, currently there are no viable long term solutions for nuclear waste.

Our investigations have revealed SrUO_4 exists in two structural types, a metastable rhombohedral () and a stable sensitive to oxygen vacancies orthorhombic () form. The structural transformation between the polymorphs is unusual and appears to be facilitated by loss of oxygen and subsequent generation of reduced forms of uranium. Once significant oxygen de-occupation has occurred the lattice is able to transform into the orthorhombic variant but only if significant atmospheric oxygen is available for reabsorption for it become stoichiometric. Without oxygen reabsorption the material cannot obtain thermodynamic stability, continuing as a high temperature structurally metastable rhombohedral material with extensive oxygen vacancies and unstable forms of uranium.

Understanding this process has involved a combination of in situ synchrotron x-ray and neutron diffraction, as well as X-ray absorption spectroscopy under normal and reducing conditions. Uranium demonstrated surprising structural flexibility uranium in controlling oxide lattice reactivity and thermodynamic stabilisation and highlights the importance of ionic conductivity in uranium oxides.

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