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The Defect Structures of Cr/Mn doped UO_2 Nuclear Fuels – New Insights Utilising Novel Single Crystal Methods with High Resolution Spectroscopy

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As a part of modern nuclear fuels, the use of transition metal elements as dopants, such as Cr and Mn, have been shown to increase the in-reactor fuel performance over traditional non-doped variants. In the case of chromia doped fuels, the small size of the Cr cation compared to the U^{4+} cation results in a low solubility of Cr into UO_2 of approximately 750-1000 ppm depending on oxygen potential and temperature [1]. Despite advances in the science behind Cr doped UO_2 modern nuclear fuels, significant paucities of information remain such as even the mechanism for incorporation and the structure of Cr-doped UO_2. Pertinently prior to this investigation, there is no definitive conclusion as to the oxidation state, local environment and structure of Cr within the fuel matrix [2],[3]. Notwithstanding this, we have managed to fabricate Cr single crystals, in addition to Mn doped crystals and have studied these using a combination of, single crystal X-ray diffraction, electron paramagnetic resonance (EPR) and X-ray absorption spectroscopy (HERFD-XAS and EXAFS). Accordingly, we provide the first conclusively resolved measurements of the Cr and Mn chemical states within the UO_2 matrix, assigning both oxidation state and describe the defect structures. We have further systematically examined the incorporation of both Mn and Cr transition metal dopants providing detailed observation of changes to the UO_2 structure, microstructure, and density relevant to their use as (spent) nuclear fuels. The results of these investigations will be discussed in detail regarding the local atomic behaviour of Cr and Mn in UO_2 to microstructure behaviour in addition to the relevance they have to both modern fuels and spent nuclear fuel disposal.

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