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High resolution grain boundary investigations of UO_2 based model systems for spent nuclear fuel

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For many oxide ceramics, dissolution at the grain boundaries (GBs) is the dominant dissolution mechanism. The dissolution of spent nuclear fuel (SNF) which can be considered as a UO_2 based ceramic, is relevant for the save disposal of nuclear waste. However, the details of the GB structures have been the subject of a few studies only [e.g. 1]. A detailed mechanistic understanding of the GB nanostructures including their doping with fission products and their role during SNF corrosion, is still missing.

In this study, pure UO_2 samples as well as Nd and Gd doped samples were synthesized as model systems for SNF using a co-precipitation method [2]. Extensive electron backscatter diffraction analyses were conducted to determine grain orientations and types of GBs. Statistical comparisons of GB misorientation angles and coincidence site lattice (CSL) boundary distributions were made between the pure UO_2 and Nd/Gd doped UO_2 samples. In addition, some specific GBs, such as Σ3 and random boundaries with a high misorientation angle, were selected for further analysis. Thin cross-section lamellae containing these GBs were extracted via focused ion beam milling to facilitate a simultaneous alignment of the boundary plane and crystal planes for the subsequent transmission electron microscopy (TEM). High-resolution scanning TEM imaging was combined with energy-dispersive X-ray spectroscopy (EDS).

A typical GB distribution for cubic polycrystalline materials was observed, with Σ3 being the most frequent CSL boundary. In high-resolution TEM, the Σ3 grain boundary in pure UO_2 was not highly symmetric and curved owing to small deviations from the Σ3 angle and due to an apparent fragmentation of the bulk crystal structure ranging in size from 10 to 50 nm. No amorphous intergranular phase was observed for the random high-angle GB. TEM-EDS mappings of a Nd-doped sample were collected across randomly chosen high misorientation angle GBs. The profiles show a significant collapse of the U-M signal at the GB (consistent with the HAADF signal), while the Nd-L and O-K signals show no significant change at the GB.

References

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