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Synthesis and detailed investigation of the microstructure of Nd- or Gd-doped UO_2 based model systems for spent nuclear fuel

FELIX BRANDT[1], ROBERT THÜMMLER[1], JURI BARTHEL[1], PHILIP KEGLER[1], ANDREY BUKAEMSKIY[1], JOACHIM MAYER[2], DIRK BOSBACH[3] AND MARTINA KLINKENBERG[1]

[1]Forschungszentrum Jülich
[2]RWTH Aachen University
[3]Forschungszentrum Jülich GmbH

Presenting Author: f.brandt@fz-juelich.de

In safety assessments for the deep geological disposal of high-level nuclear waste, the unlikely ingress of water and corrosion of spent nuclear fuel (SNF) are considered. For many oxide ceramics, grain boundary dissolution plays an important role, which can continuously increase the reactive surface area and sometimes even leads to a disintegration of the microstructure. Due to the complexity of SNF, simplified UO_2 based model systems were developed to enable a detailed study of the role of the microstructure during SNF corrosion. Here we present new results about the synthesis and microstructure of pure UO_2 and Nd or Gd doped UO_2 ceramics - mimicking the presence of fission products in SNF.

Polycrystalline UO_2 ceramic pellets were synthesized using a co-precipitation method [1] with doping levels of x = 0% to 8% in $(U_{1-x}Nd_x)O_2$ or $(U_{1-x}Gd_x)O_2$, respectively. The synthesis route was the same for all pellets to enable a comparison of the doping effects with different Nd or Gd concentrations. A special focus was put on the comprehensive analysis of microstructural features like grain size, pore size, grain shape and the chemical composition via scanning electron microscopy and energy dispersive X-ray spectroscopy.

Pure UO_2 ceramics exhibited a monomodal grain size distribution. Doping reduces the average grain size from 11 µm for pure UO_2 to 9.3 µm for Nd, and to 9.6 µm for Gd at a doping level of x = 8%. Particularly noteworthy is the formation of a heterogeneous microstructure at 2% and 4% doping levels due to clusters of fine grains. These clusters constitute significant portions of the sample area at 2% and a smaller fraction at a 4% doping level. Typically, these fine grains contain less than the nominal concentration of Nd or Gd, while large grains exhibit slight dopant enrichment. This significant change of the UO_2 microstructure may be explained by the formation of different uranium oxide phases during calcination and sintering, which are known to be affected by small variations of the oxygen potential.

[1] Kegler, P., et al. (2021). Materials, 14(20), p.6160.