Structural insights into UO_2-based model systems for spent nuclear fuel

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Demonstrating the long-term safety of a deep geological repository for spent nuclear fuel (SNF) requires a sound understanding of radionuclide release mechanisms from SNF. The long-term matrix corrosion of SNF is controlled by the electrochemical properties of the system, which are influenced by the concentration and distribution of fission products in the UO_2 matrix, such as, lanthanides and metallic epsilon-particles. Due to its chemical and structural complexity, SNF is unsuitable for entirely unravelling concurrent corrosion mechanisms. Here, we present a new approach *via* simplified UO_2-based model systems, which enable single-effect studies of long-term matrix corrosion.

Samples were synthesized by various wet chemical routes to complement studies on "real" SNF. UO_2 ceramics were doped with Nd (1-7 wt%), which serves as surrogate for fission lanthanides. The Nd-doping modifies the oxygen sublattice and influences the electrochemical properties. In order to probe these local structural changes, neutron total scattering was applied. The pair distribution function analysis indicated that lower dopant concentrations (1 wt% Nd) cause higher local distortions in the UO_2 structure. Complementary information was obtained by Raman spectroscopy. These new structural insights allow one to link structural properties of the UO_2-based model systems to corrosion behavior, thus, contributing to the understanding of highly-relevant long-term matrix corrosion mechanisms of SNF.