Structure of hydrated UO₂ surfaces

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Corrosion of uraninite (UO_2) is fundamentally important to environmental remediation strategies for uranium-contaminated soils and waters, to nuclear fuel systems, and for waste disposal technologies. The structure of hydrated uraninite-water interfaces under pristine and corroded conditions is of seminal importance to uranium corrosion, and thus to nuclear and environmental chemistry. To date, UO_2 surface studies have been limited to vacuum-based techniques, generally conducted at elevated temperatures. UO_2 surfaces have not previously been studied in the presence of liquid water.

The crystal truncation rod (CTR) x-ray diffraction method is ideally suited to such measurements, as it specifically probes structures of 2-dimensional interfaces. The high energy and high brilliance of the x-rays available at the Advanced Photon Source allow for the accurate measurement of the weak signals generated at the liquid-solid interface. The (111) surface structure of pure, synthetic UO_2, determined under both dry and hydrated conditions, will be presented. Complementary atomic force microscopy (AFM) and low energy electron diffraction (LEED) studies will be discussed.

Spent Nuclear Fuel corrosion – The influence of Pd ε-particles on dissolution processes

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The safety assessment of a nuclear waste repository requires the reliable prediction of the overall spent nuclear fuel (SNF) corrosion behaviour since the dissolution of the oxidic fuel matrix can result in the release of radionuclides (RN) into the environment. SNF is a complex system composed of a UO_2 matrix that is doped with RNs and fission products (FPs; ε-particles). In the present study the corrosion of different UO_2 based fuels (UO_2, $Th_xU_yO_z$) and the influence of nano-sized Pd ε-particles on corrosion processes is investigated in cyclovoltammetric (CV) measurements.

Model fuel oxide surfaces are prepared by sputter co-deposition of U, Th and Pd thin films. The films are characterized by application of microscopic (AFM, SEM), spectroscopic (XPS) and diffraction methods (XRD). The CV measurements indicate that for $Th_xU_yO_z$ it is U (VI) which preferentially dissolves. The resulting change in surface morphology of the film is monitored by SEM. The influence of the inhomogeneous dissolution on the release of FPs is further investigated. It was already shown by the authors that UO_2 oxidation is inhibited by interaction of the oxide surface with H_2 due to the presence of Pd nano-particles (Gas-Adsorption-Experiments). The catalytic activity of the noble metal towards hydrogen activation is now investigated in solution experiments (CV measurements).