

XAFS and μ -XANES/XRF investigation of highly radioactive spent nuclear fuel fragments at the KIT synchrotron source

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Spent nuclear fuel (SNF) is a highly hazardous material which needs to be safely stored until final disposal facilities become operational. Deep geological disposal is deemed the preferred option for safe SNF management in many countries, enabling efficient isolation of the waste from the biosphere for a period of one million years. Until the phase-out of the use of nuclear power in Germany in 2022 about 1.05×10^4 tons of irradiated SNF from nuclear power plants will have been accumulated.

During irradiation in a nuclear power plant, several weight percent of the initial U atoms of fresh nuclear fuel are transmuted by neutron induced nuclear fission of U-235, producing different fission products (FP), and actinides by neutron capture of U-238. Xe, Kr, Br, Cs and I and other volatile / mobile FPs might segregate into the gap between the fuel pellets and the Zircaloy cladding, the fuel fractures and to grain boundaries depending on the steep temperature gradient from the centre to the periphery of the fuel pellet and the burn-up reached. Other FPs form metallic precipitates comprising noble metals as well as Mo, Te and Tc (known as “epsilon phases”). Fission lanthanides and actinides tend to stay incorporated in the UO_2 lattice by substituting the U. Speciation of uranium, actinides and, e.g., technetium as a long-lived radioactive FP depends on the temperature during irradiation in the reactor and on the oxygen availability and competitive reactions between the FPs. The expected prolongation of SNF interim storage puts a high demand on methods establishing a better understanding how thermal and radiological stress and chemical composition alters the integrity of irradiated fuel rods - with a focus on the chemical deterioration and embrittlement of the Zircaloy cladding. To this end, XAFS and related X-ray techniques are ideally suited as speciation methods for the investigation of SNF, as they are capable to provide in-situ information on oxidation states and short-range structural parameters of SNF constituents. X-ray fluorescence spectroscopy (XRF) delivers information on the elemental composition of the sample. XANES/XRF combined with a μ -focus incident beam provide additional spatial resolution of element and chemical species distribution.

Research at KIT-INE benefits from the unique situation where a shielded box-line with instrumentation for SNF sample manipulation and analysis operated at KIT-INE controlled area laboratories is situated in close proximity to X-ray spectroscopy experimental stations dedicated to radionuclide research - the INE-Beamline and the new ACT laboratory at the KIT synchrotron source on KIT Campus North. In this presentation, examples for XAFS/XRF investigations of mm sized SNF fragments and irradiated cladding tube specimens – both from the fuel rod segment top (plenum) and from a fuel pellet, where the cladding had been in contact with the UO_2 matrix – will be discussed. Bulk samples with contact dose rates up to several mSv/h can be safely handled at both experimental stations. It will be demonstrated that performing bulk measurements helps to minimize possible artefacts expected for the investigation of abrasive material or dust particles peeled off from the fuel pellet surface.