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Se K-edge micro XANES on high-burnup UO2 spent nuclear fuel: First results in the framework of the FIRST-Nuclides project

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In the context of the FIRST-Nuclides collaborative project (EURATOM FP7 programme) Paul Scherrer Institut is carrying out leaching experiments on high-burnup UO2 spent nuclear fuel (SNF) from Swiss nuclear power plants, in order to determine the short-term release of easily accessible nuclides (instant release fraction, shortly IRF). The focus is on nuclides such as 129I, 135Cs, 14C and 79Se. Previous experiments (Johnson et al., 2012) had shown that, contrary to expectations, no detectable Se was released to the aqueous phase even after 100 days of leaching.

In order to understand the behaviour of 79Se during aqueous leaching of SNF in a geological repository, X-ray absorption experiments were carried out at the MicroXAS beamline (SLS) on high burnup UO2 spent fuel from the Leibstadt nuclear power plant. As the comparison of the Se K-edge micro XANES data obtained on SNF with the spectra of reference compounds yielded ambiguous results, the experimental spectra were modelled ab initio via FEFF or FDMNES calculations coupled with geometrical optimization procedures provided by the FitIt package.

Best fits of the experimental SNF XANES spectra were obtained by assuming substitution of Se(-II) (selenide) in oxygen sites of the UO2 lattice, with Se-U distances very similar to those of crystalline U selenide compounds. These results suggest that 79Se may be stabilized as selenide in SNF, explaining the failure to detect aqueous selenium in leaching experiments. Release of 79Se to aqueous solutions under geological storage conditions would then proceed by matrix dissolution rather than via IRF.

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