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Selenium speciation in a spent UO2 fuel and in non-irradiated UO2 reference samples: a synchrotron-based (micro-)XRF / -XAS feasibility study

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In the safety case for underground radioactive waste repositories, aqueous corrosion of the waste plays a central role, since it determines the source term of radionuclide release to the environment. The direct disposal of spent nuclear fuels is an option adopted in many countries, implying that radionuclide release studies from spent UO2 and mixed oxide fuels are critical for building confidence in the safety case. One of the major issues to be solved is the reliable determination of the «Instant Release Fraction» (IRF), i.e. the cumulative inventory fraction of potentially easily soluble nuclides (129I, 135Cs, 36Cl, 79Se, 14C), which will be released on contact with aqueous solutions penetrating a breached canister soon after failure of the waste overpack. Whereas I, Cs and Cl are soluble within the entire natural Eh range, C and particularly Se mobilization on contact with aqueous solution strongly depends on oxidation potential. Especially, under oxic conditions the easily soluble Se(IV) and Se(VI) form, whereas under reducing conditions the sparingly soluble and thus almost immobile Se(0) and Se(-II) are stabilized.

In the present feasibility study, X-ray absorption spectroscopy (XAS) and X-ray fluorescence (XRF) investigations have been conducted at the Se K-edge, partly at micrometric beam resolution. The samples used were (i) a high burn-up (˜ 79 GWd/t) UO2 spent fuel and (ii) several non-irradiated UO2 reference samples doped with Se in amounts covering the concentration ranges expected in spent fuel (˜ 50 -150 ppm). Meaningful data were obtained in spite of the very low Se concentrations and the heavy sample matrix (UO2). For instance, the micro-XANES spectra obtained on particles of the irradiated sample -not larger than a few micrometers- consistently indicate the presence of mixed valence selenium (possibly Se(0) and Se(IV)). Moreover, acceptable and consistent bulk EXAFS data have been obtained for the UO2 reference sample doped with 100 ppm SeO2.

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