Instant release fraction and matrix release of high burn-up UO₂ spent nuclear fuel: Effect of high burn-up structure and leaching solution composition

Abstract:
Two weak points in Performance Assessment (PA) exercises regarding the alteration of Spent Nuclear Fuel (SNF) are the contribution of the so-called Instant Release Fraction (IRF) and the effect of High Burn-Up Structure (HBS). This manuscript focuses on the effect of HBS in matrix (long term) and instant release of a Pressurised Water Reactor (PWR) SNF irradiated in a commercial reactor with a mean Burn-Up (BU) of 60 GWd/tU. In order to study the HBS contribution, two samples from different radial positions have been prepared. One from the centre of the SNF, labelled CORE, and one from the periphery, enriched with HBS and labelled OUT. Static leaching experiments have been carried out with two synthetic leaching solutions: bicarbonate (BIC) and Bentonitic Granitic Groundwater (BGW), and in all cases under oxidising conditions. IRF values have been calculated from the determined Fraction of Inventory in Aqueous Phase (FIAP). In all studied cases, some radionuclides (RN): Rb, Sr and Cs, have shown higher release rates than uranium, especially at the beginning of the experiment, and have been considered as IRF. Redox sensitive RN like Mo and Tc have been found to dissolve slightly faster than uranium and further studies might be needed to confirm if they can also be considered part of the IRF. Most of the remaining studied RN, mainly actinides and lanthanides, have been found to dissolve congruently with the uranium matrix. Finally, Zr, Ru and Rh presented lower release rates than the matrix. Higher matrix release has been determined for CORE than for OUT samples showing that the formation of HBS might have a protective effect against the oxidative corrosion of the SNF. On the contrary, no significant differences have been observed between the two studied leaching solutions (BIC and BGW). Two different IRF contributions have been determined. One corresponding to the fraction of inventory segregated in the external open grain boundaries, directly available to water and very significant during the first days of the experiment; and a second one corresponding to a less accessible, most probably located at the internal grain boundaries, one order of magnitude lower than the first one at equal given dissolution times but of much longer period of incidence. Unlike matrix release results, higher Cs IRF release was found for OUT than for CORE sample. This effect can be attributed to thermal migration of Cs to the periphery of the fuel during irradiation. In the case of Rb no clear differences were observed between CORE and OUT showing equilibrium between the opposing thermal migration and matrix effects. Finally, Sr CORE/OUT release ratio showed similar behaviour to matrix release, thus proving no significant thermal migration during
irradiation.

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