Experimental and numerical validation of the radionuclide inventory in irradiated high burn-up UO_X and $(U, Pu)O_X$ pressurised water reactor fuel claddings

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Currently, spent nuclear fuel (SNF) assemblies discharged from German power reactors are stored in dual-purpose casks (DPC) in interim dry storage facilities and kept there until their conditioning for final disposal. Licenses for both, the DPCs and interim dry storage facilities, will expire after forty years consecutive to the loading of the cask and the first emplacement of a DPC in the storage location. Due to considerable delays in the site selection process and the resulting absence of a final repository in Germany so far, a prolongation of interim dry storage of a timespan of about 100 years is inevitable. Regarding this long timespan, the integrity of the SNF cladding is of significant importance in respect of the latter conditioning of the fuel assemblies for final disposal.

With increasing burn-up, the gap between the nuclear fuel pellet and Zircaloy cladding tube closes under the formation of an interaction layer leading to chemical and mechanical interactions between pellet and cladding. Within this fuel-cladding interface, possible cladding degrading elements can accumulate and induce pitting or stress corrosion processes. In addition, the cladding is exposed to alpha irradiation damage induced by the decaying actinides present at the periphery of the fuel pellet. Precise knowledge of the radionuclide content in SNF and the cladding is of utter importance for a thorough revaluation of all safety relevant aspects regarding the long-term behaviour of the casks and their inventory. Moreover, knowledge of the SNF radionuclide inventory is important in safety assessments for final disposal, in order to scrutinise the individual radionuclide source terms in case of ground water intrusion into the repository and a consecutive cladding failure. However, uncertainties in calculated radionuclide inventories in SNF can be about 2 % for uranium and plutonium, 7 % for fission products and up to 11 % for minor actinides [1]. Uncertainties of activation products, present in the claddings, can be much higher since they depend solely on the precursor content within the material [1].

Within our contribution, we provide radionuclide inventory data of Zircaloy-4 claddings taken from a high burn-up UO_X (50.4 GWd/t_{HM}) and an $(U, Pu)O_X$ (38.0 GWd/t_{HM}) fuel, previously irradiated in commercial pressurised water reactors during the 1980s. Furthermore, the obtained experimental data is compared to results received by MCNP/CINDER and webKORIGEN calculations in order to validate the performance of calculation codes against radiochemical analyses. For activation products, such as ^{14}C, ^{36}Cl or ^{125}Sb, a good agreement between experimental analysis and numerical calculations is apparent. However, in case of fission products and actinides, the radiochemical determined activities differ significant from values obtained by MCNP/CINDER and webKORIGEN calculations.

References

[1] Spahiu, K., State of the Knowledge (SoK) Report, European Joint Programme on Radioactive Waste Management (EURAD) (2021).