Chemical and spectroscopic investigations on the distribution and enrichment of radionuclides in fuel-cladding interfaces of irradiated high burn-up UO_X and MOX fuels

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After utilisation of nuclear fuel in a power reactor and subsequent storage in a spent fuel pool to dissipate decay heat, many countries, including Germany, envisage the spent nuclear fuel (SNF) assemblies to be stored in an interim dry storage facility. At this, the SNF assemblies are emplaced in storage casks or dual-purpose casks to provide shielding from the radioactivity and a safe temporary storage mode until their conditioning for final disposal in a deep geological repository. Regarding the inevitable prolongation of interim dry storage of SNF due to the absence of a final repository, the impact of 65 to 100 years of storage on the cladding integrity is one of the most crucial aspects. Several physical and chemical effects may alter the integrity of the zirconium alloy throughout the storage period, such as irradiation damage caused by alpha emitters or the re-orientation of hydrides, incorporated into the zirconium matrix of the cladding tube during reactor operation. With increasing burn-up, the gap between the nuclear fuel pellet and Zircaloy cladding tube closes under the formation of an interaction layer. Within this interface between fuel pellet and cladding, possible cladding degrading elements, such as chlorine and iodine, can accumulate and induce pitting or stress corrosion processes.

In this work, we provide experimental data on the inventories of high burn-up UO_X (50.4 GWd/t_{HM}) and mixed oxide (MOX, 38.0 GWd/t_{HM}) fuel segments, irradiated in commercial nuclear power plants during the 1980s. Special regard is given to the local distribution and enrichment of various radionuclides within the fuel pellet, with particular emphasis on radionuclides regarded as highly mobile under repository conditions, e.g. ^{36}Cl and ^{129}I. In addition, the experimentally determined inventories are compared to data derived from calculations using MCNP / CINDER and webKORIGEN codes. Furthermore, spectroscopic measurements on the respective fuel and cladding specimens were performed by scanning electron microscopy coupled with energy and wavelength dispersive X-ray spectroscopy, as well as X-ray absorption spectroscopy.

Within this study, an enrichment of ^{129}I in the peripheral, cladding adherent area of the SNF pellets was identified. Furthermore, first results on the occurrence and distribution of the activation product ^{36}Cl in SNF are provided. The obtained results from this study contribute to an increased knowledge on the distribution and speciation of possible cladding degrading elements / phases within the fuel-cladding system.