Volatile activation and fission products ^{36}Cl and ^{129}I present in fuel-cladding interfaces of spent UO_2 fuel – separation method and first results

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In Germany, spent nuclear fuel (SNF) is stored in dual-purpose casks (e.g. CASTOR®, GNS) after discharge from a nuclear reactor until a repository for final disposal will be available. Commissioning of a repository for high-level waste is expected by 2050 at the best. Yet, cask and facility licenses for dry storage of SNF expires after forty years following loading of the cask and delivery to a storage facility. Thereafter, a thorough revaluation of all safety relevant aspects regarding the long-term behaviour of the casks and their inventory is required. Due to considerable delays in the development of a selection process for a disposal site and an estimated duration of the construction and commissioning of a final disposal system of at least two decades, a significant amount of SNF has to be kept in dry storage facilities for about 65 to 100 years (ESK, 2015). Integrity of the irradiated Zircaloy cladding after such long periods of dry interim storage is essential e.g. for conditioning of the fuel assemblies for final disposal. However, various processes affect cladding integrity during reactor operation and beyond e.g. mechanical stress on the cladding exerted by swelling of the fuel pellet as well as fuel-cladding-chemical-interactions (Ewing, 2015). With increasing burn-up, the gap between the fuel pellets and the cladding closes and so called fuel-cladding interaction layers do form. Precipitates of fission/activation products such as caesium, chlorine, iodine, and tellurium present at the fuel-cladding interface possibly exhibit corrosive properties and thus affect the cladding integrity.

In order to determine the amount and chemical speciation of such radionuclide precipitates, Zircaloy specimens were sampled from fuel rod segments irradiated in the pressurised water reactor of Gösgen, Switzerland, during the 1980s. Samples were taken from the cladding tube in contact with SNF (denoted as Zry-fuel) and from the plenum section (denoted as Zry-plenum) of an UO_2 fuel rod segment (irradiated at an average linear power of 260 W/cm and a final burn-up of 50.4 GWd/t_{HM},).

In this study, a separation method involving liquid-liquid extraction, precipitation as well as chromatographic extraction steps was developed and tested in order to isolate ^{36}Cl and ^{129}I from other radionuclides present in digestion liquors involving highly radioactive specimens. Applying this analytical method, Zry-fuel samples were analysed for their ^{36}Cl and ^{129}I contents.

Inactive salts of the halides were added to the leachate in order to study the extraction yield of the method. The analytical procedure was performed by means of ICP-MS, IC, LSC and γ-spectrometry leading to an effectiveness of the separation method of 90 ± 2 % for chlorine and 92 ± 5 % for iodine. Similar results were obtained for solutions spiked with ^{36}Cl and ^{129}I. Furthermore, the separation technique was applied to a digestion liquor of a Zry-plenum sample as well as to a leaching solution that had been in contact with a cladded SNF segment for 333 days of the same fuel rod segment leading to a clean separation of the radionuclides from the solutions.

References:

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