

Examination of volatile fission and activation products within the fuel-cladding interface of irradiated PWR fuel rod segments

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Cladding integrity of spent nuclear fuel rods is one of the main criteria when relocating spent fuel assemblies from an interim storage cask into a final disposal cask. Amongst others, cladding integrity is affected by mechanical stresses due to swelling or elongation of the fuel pellet, hydride-induced cracking of the Zircaloy cladding, irradiation damage of alpha emitters present in the fuel pellet's rim zone, as well as deposition of fission or activation products responsible for fuel-cladding chemical interaction (FCCI). Latter occurs with increasing burn-up of the fuel resulting in the closure of the gap between fuel and Zircaloy cladding, and formation of a fuel-cladding interaction layer. These layers accumulate different isotopes such as caesium, iodine, tellurium and chlorine forming agglomerates that display an interface for corrosive processes, which possibly affect the cladding integrity.

In this study, two different types of nuclear fuel rod segments, namely UO_2 and mixed oxide fuel (MOX) rod segments, were examined using various spectroscopic techniques and compared to each other. In addition, the radionuclide inventory of the Zircaloy cladding was experimentally determined and compared to theoretical values obtained by means of MCNP CINDER / KORIGEN calculations.

Fuel and Zircaloy specimens were sampled from fuel rod segments irradiated in the pressurised water reactors of Gösgen (Switzerland) and Obrigheim (Germany) during the 1980s with average fuel rod burn-ups of $50.4 \text{ GWd/t}_{\text{HM}}$ and $38.0 \text{ GWd/t}_{\text{HM}}$, respectively.

Analysis of agglomerates found on the inside of the cladding samples was performed by optical microscopy, scanning electron microscopy and energy-dispersive X-ray spectroscopy, as well as X-ray photoelectron spectroscopy and X-ray absorption spectroscopy. Special focus was set on volatile fission and activation products e.g. caesium isotopes, ^{36}Cl and ^{129}I which were separated by a radiochemical separation method and analysed by means of liquid scintillation counting, γ -spectrometry, ion chromatography and inductive-coupled plasma mass spectrometry. Obtained results from this study aim at a further understanding of the FCCI layer in terms of radionuclide inventory and chemical speciation of the compounds present, as well as their conceivable degradation ability of the Zircaloy cladding during dry interim storage.