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Corrosion behaviour of irradiated mixed oxide (MOX) fuels: Effects of environmental conditions on radionuclide leaching

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In the context of spent nuclear fuel (SNF) disposition, a deep geological repository (DGR) based on a multi barrier concept is considered in many countries as the safest and most sustainable disposal option. The licensing of such a DGR is challenging and demands amongst others, safety assessments considering time frames of up to one million years. Moreover, a profound understanding of the corrosion behaviour of SNF coming into contact with groundwater, when the waste canisters are eventually breached, is crucial. During the last decades, a number of studies addressed this topic, leading to a good phenomenological understanding of the long-term behaviour of SNF in a DGR. However, these studies addressed mainly uranium dioxide-based fuels, and various processes contributing to the (radiolytic) matrix corrosion of SNF in the generally reducing repository environment are still not fully understood.

Since corrosion data on irradiated mixed oxide (MOX) fuels under reducing conditions are scarce to date, the SF-ALE project (Spent Fuel Autoclave Leaching Experiments) was initiated. MOX fuel with a well-known irradiation history was characterised by optical microscopy, SEM and EPMA before leaching experiments were performed. The latter aim at elucidating the impact of the environmental conditions on SNF corrosion, addressing the instant release of radionuclides as well as the (long-term) matrix corrosion. Three cladded fuel rod segments with burn-ups ranging between 29 GWd/t_{HM} and 52 GWd/t_{HM} were selected for leaching experiments and were exposed to bicarbonate water as reference groundwater at neutral pH, and a synthetic cementitious water (pH 13.5), to address repository conditions for disposal concepts with different engineered barrier systems. Since repository conditions are expected to be reducing as a consequence of hydrogen generation due to anoxic corrosion of metallic waste canisters, the autoclave leaching experiments, which lasted for about 3.5 years, were performed under a gas phase consisting of an argon/hydrogen mixture at a pressure of 40 bar. During the leaching experiments, both the leachates and the gas phases were regularly sampled in order to determine the release of more than 30 relevant radionuclides from the irradiated MOX by various analytical techniques.

Results regarding the initial release fractions of the fission products caesium and iodine observed within the first two years of leaching were published recently \[1\]. In this contribution, we report results on the release behaviour of further selected fission products and actinides obtained throughout the whole duration of SF-ALE.

References

1. Schreinemachers, C., Leinders, G., Mennecart, T. et al. "*Caesium and iodine release from spent mixed oxide fuels under repository relevant conditions: Initial leaching results*". MRS Advances (2022). DOI: 10.1557/s43580-022-00220-7

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