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Experimental investigation of uranium release from spent nuclear fuels under conditions expected in a deep geological repository

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Abstract. Direct disposal of spent nuclear fuel (SNF) in a deep geological repository is planned for the long-term management of the highly radioactive waste discharged from the light water reactors (LWRs) of nuclear power plants in Germany. The aim is to safely isolate the radionuclide inventory from the biosphere, ad infinitum, making use of a multi-barrier system consisting of technical, geotechnical and geological barriers interposed between the emplaced waste and the environment. Still, reversibility measures including retrievability of the waste products are considered in this concept. Internationally, in safety assessments for deep underground repositories (DGRs), the access of groundwater, followed by failure of canisters and finally loss of the SNF's cladding integrity is considered in the long term. Hence, the evaluation of the performance of SNF in the near field of a deep geological repository requires process understanding of SNF dissolution and rates, in addition to the quantification of the radionuclide release from SNF under reducing conditions of a breached container. With the aim to derive a radionuclide source term, the SNF dissolution and alteration processes can be assigned to two steps, namely the (i) instantaneous release of radionuclides upon cladding failure from gap and grain boundaries and (ii) a long-term release that results from the dissolution of the fuel grains themselves (Ewing, 2015). Since most radionuclides produced during irradiation of nuclear fuel in a reactor are trapped within the SNF matrix, understanding the processes leading to the dissolution of the matrix and its dissolution rates are of particular interest for post-closure safety assessments (Pastina and LaVerne, 2021).

Research at the Institute for Nuclear Waste Disposal (INE) at the Karlsruhe Institute of Technology (KIT) has focused, for more than 25 years, on the behaviour of SNF (irradiated UO_2 and mixed-oxide, MOX, fuels) under geochemical conditions (pH, redox milieu, gas atmosphere and ionic strength) representative of various repository concepts and radiolytic reactions on SNF dissolution mechanisms.

This contribution will give an overview on the radionuclide release from dissolving SNFs under repository conditions. In particular, uranium released into solution from UO_2 and MOX fuels during up to 1629 d of leaching in salt brines, granitic/bentonitic or cementitious groundwaters under Ar or Ar/H_2 atmospheres.

As observed in other published studies on SNF dissolution under reducing conditions, a large scatter of the uranium concentration is seen in the initial stages of our experiments. After about 1 year of leaching, the aqueous concentration of uranium slowly approaches the solubility limit of U(IV), independent of the type of studied SNF samples (various irradiated UO_2 and MOX fuels). A similar behaviour is observed for other redox-sensitive actinides. The release rate of uranium decreases significantly in all experiments within first 400 d of leaching. Although the rate is very low, it is > 0, and a continuous release is observed. A comparison of the results from SNF experiments under various redox conditions demonstrates that oxidative matrix dissolution is inhibited due to the presence of hydrogen. Our results are discussed in the context of the outcome of international research programmes on SNF corrosion. These findings are considered to be of relevance for the definition of source terms for various irradiated LWR fuels in safety investigations for a DGR in Germany.

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