Research on High-level Waste at KIT-INE in the Context of Prolonged Interim Storage and the New Site-selection Process for a Deep Geological Repository in Germany

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1. Background

In the year 2011, the Federal Parliament of Germany decided, based on a broad societal consensus, to phase-out nuclear electricity production with the last German nuclear power plant to be shut down in 2022. According to projections for the nuclear power phase-out, about a total of 17,000 tons of spent nuclear fuel (SNF) and about 600 cubic metres of vitrified reprocessing waste will accumulate in Germany. A new site selection process for final disposal of the aforementioned high level waste (HLW) was started in 2013 by billing a site selection act which has been amended in 2017. A specific challenge of the German process being distinctly different to international approaches is the provided comparison of different disposal concepts in the different host rock types (rock salt, crystalline and argillaceous rock).

The site selection process shall be based on scientific criteria and transparent procedures with public participation. It will comprise three phases, where the number of eligible regions/sites will be successively reduced until a final decision for a site will be made. According to plans of the Federal Ministry for Environment, Nature Conservation and Nuclear Safety, a deep geological repository is planned to be operational already in the second half of this century. Due to expected delays in site selection, licensing and construction of a deep geological repository, one cannot exclude that interim storage of HLW has to be extended for 100 or more years.

2. Scientific profile and mission of KIT-INE

At KIT-INE, the focus of research lies on fundamental aspects of radionuclide chemistry in aquatic systems, on applied studies related to radioactive waste forms (including SNF and HLW-glass) and radionuclide behavior in various repository barrier systems, on the development of tools contributing to a geochemistry based safety case and to a science-based site selection process. In collaboration with social science teams of a partner institute at KIT a more comprehensive view towards specific topics related to management of HLW and the site selection process is developed.

In the present communication we outline aspects of prolonged interim storage of spent nuclear fuel and the site-selection process for a deep geological repository for HLW in Germany, and we present selected research highlights of KIT-INE with respect to HLW behavior under conditions of extended dry storage and final disposal.

3. Research on high level waste behavior under repository conditions

At KIT-INE, investigations on the behavior of HLW under repository conditions focus on experimental determination, process understanding and numerical modelling of radionuclide release and retention during corrosion of SNF, activated metallic components of nuclear fuel rods and HLW glass. Experiments with these irradiated materials are conducted in the shielded box-line of KIT-INE.

Amongst other international projects, SNF behavior in various repository systems has been investigated within KIT-INE coordinated European projects (e.g. the 7th FP project FIRST-Nuclides). In our experiments, both the Instant Release Fraction (IRF) of radionuclides and the relatively slow release of radionuclides from the SNF matrix were quantified. IRF of twelve different types of high burn-up SNF were experimentally determined within FIRST-Nuclides. KIT-INE compiled a database of all available information of the studied fuels, including their IRF, fuel fabrication data and irradiation histories. Our evaluation of the database showed a close correlation between IRF, fission gas release (FGR) and in-reactor operation parameters (in particular the linear heat generation rate).

In an unique approach, KIT-INE scientists determined both the Kr and Xe inventories in the plenum of an irradiated fuel rod segment and in leaching experiments with samples of the same SNF segment. Experimental results showed a relatively high FGR to I-129 release ratio, whereas the release ratio of the fission gases to Cs-137 was 1:1.

In experiments with irradiated Zircaloy and stainless steel, about 99% of the C-14 was found as gaseous or dissolved organic C-14 bearing compounds after release from the samples. These results are of high relevance for the role of C-14 in safety assessments for SNF repositories, because volatile and dissolved C-14 bearing organic species possess a high potential mobility through various geo-engineered and geological barriers.

For the first time ever, a real HLW glass sample with a dose rate of 0.6 mSv/h (sampled from the Karlsruhe vitrification plant) was analyzed by means of XAS/XRF at the KIT synchrotron facility. Based on high quality XANES data, speciation of actinides (Am(III), Pu(IV), Np(V), U(VI)), Zr (Zr(IV)), Se (identified as selenite), and Tc (identified as pertechnetate) in the HLW glass was determined.

4. Research on spent nuclear fuel behavior under dry interim storage conditions

Experimental studies were initiated to examine possible mechanisms of cladding corrosion at the SNF / zircaloy interface and the potential relevance for the fuel pin integrity during extended interim storage conditions. Such scenarios have to be considered in view of the long interim storage times exceeding licensed periods of storage canisters by far according to the present plans for site selection in Germany.

In recent studies pellet-cladding-interactions and cladding corrosion by volatile fission products were studied using Zircaloy-4 specimens, which had been sampled from irradiated UO2 and MOX fuel rod segments. Composition of agglomerates found at the inner surface of plenum sections and fuel-cladding interaction layers were analyzed by means of μ-XAS and μ-XRF at the KIT synchrotron facility, as well as by means of SEM-EDS and XPS. It was found that a significant amount of Cs had been volatilized during irradiation from subjacent fuel pellets and deposited at the Zircaloy of the fuel rod plenum, whereas in lower parts of the pellet pile Cs precipitated as Cs-halogenide at the pellet-cladding interface.