Saf. Nucl. Waste Disposal, 2, 153–154, 2023
https://doi.org/10.5194/sand-2-153-2023

SaND
Safety of Nuclear Waste Disposal
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Experimental investigation of fission gas release from spent nuclear fuels under conditions expected in a deep geological repository

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Received: 9 April 2023 – Accepted: 21 May 2023 – Published: 6 September 2023

Abstract. In Germany, as well as several other countries (e.g. Sweden, Finland and Switzerland), the disposal of spent nuclear fuel (SNF) in deep geological formations, intertwined with a resilient multi-barrier concept, is considered to be the favourable option for the safe and profound isolation of such wastes from the biosphere. However, the intrusion of groundwater, followed by a failure of canisters and loss of cladding integrity, has to be considered in the long-term evaluation of such an underground repository. Therefore, a thorough process understanding of SNF dissolution, radionuclide source terms and alteration processes is necessary in order to evaluate the performance of SNF in the repository system. The aforementioned processes can be assigned into two steps: (i) a fast, initial release of radionuclides, located in accessible structures of the SNF upon cladding failure, and (ii) a slower, long-term release, resulting from the dissolution of the fuel matrix itself (Ewing, 2015).

During the first stages of breaching of the container and the resulting contact of groundwater with the waste form, a fraction of radionuclides, located in easily accessible parts of the SNF such as grain boundaries or the pellet-cladding gap, will be released relatively quickly. This fraction is comprised of mostly volatile radionuclides, such as ^{79}Se, ^{129}I, ^{135}Cs, and the fission gases Kr and Xe. Especially the release of fission gases from SNF is often correlated to the release properties of safety-relevant radioisotopes such as ^{129}I or ^{135}Cs and is, therefore, an indicator of their release rates. In particular, ^{129}I shows a release behaviour similar to the fission gases.

Within our contribution, we provide results regarding the release of fission gases during the dissolution process of two different types of SNF, i.e. UO_X ($50.4\,GWd\,t_{HM}^{-1}$) and mixed-oxide (MOX) ($38.0\,GWd\,t_{HM}^{-1}$, where HM denotes heavy metals) fuels, under simulated geochemical conditions, as they are expected in a deep geological repository. Both examined fuel types were irradiated in commercial pressurised water reactors in Switzerland and Germany during the 1980s. The shown data were compiled over more than a decade during experimental campaigns performed at the Institute for Nuclear Waste Disposal at Karlsruhe Institute of Technology (KIT-INE) within the framework of several international programmes. Results obtained in our study indicate that, independent of the type of SNF, a comparable, fast initial release of fission gases is evident in all experiments. However, throughout the experiments, this prompt release slows down significantly, although a continuous release can be observed. Furthermore, the obtained data on fission gas release are compared to the release behaviour of iodine and caesium. An attempt is made to identify and qualify the role of fission gas release rates as indicator of the radionuclide source term for SNF by comparing and interpreting respective data collected over various experimental campaigns.

Published by Copernicus Publications on behalf of the Federal Office for the Safety of Nuclear Waste Management (BASE).

Author contributions. TK, EB, EGR, MH, LIP and AW performed the experiments and measurements; TK wrote the manuscript draft; and MH, VM and HG reviewed and edited the manuscript.

Acknowledgements. The authors would like to thank Annika Fried, Markus Fuss, Frank Geyer and Cornelia Beiser (all KIT-INE) for their invaluable analytical work in this study. Nikolaus Müller (formerly KIT-INE), Volker Krepper, Stefan Schmitt (both KIT-INE) and Gerhard Christill (KIT-SUM – Sicherheit und Umwelt, Karlsruhe Institute of Technology) are acknowledged for their contribution to the operation and maintenance of the shielded box-line facility as well as radiation protection work.

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