Near field evolution of a spent fuel repository in an argillaceous rock formation and impact on radionuclide migration

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In August 2015, the German government approved the national programme for the responsible and safe management of spent nuclear fuel (SNF) and radioactive waste proposed by the Federal Ministry for the Environment, Nature Conservation, Building and Reactor Safety (BMU). The assumption is that about ~ 1 100 storage casks (10 500 tons of heavy metal) in the form of spent fuel assemblies will be generated in nuclear power plants and will have to be disposed. However, a decision on the disposal concept for high-level waste is pending and an appropriate solution has to be developed with a balance in multiple aspects. All potential types of host rocks, clay and salt stones as well as crystalline formations are under consideration. In the decision process, evaluation of the risk of different waste management options and scenarios play an enormous role in the discussion. Coupled physical and chemical processes taking place within the engineered barrier system of a repository for high-level radioactive waste will define the radionuclide mobility/retention and the possible radiological impact. The objective of this work is to assess coupled processes occurring in the near-field of a generic repository for spent nuclear fuel in a high saline clay host rock, integrating complex geochemical processes at centimetre-scale. The scenario considers that radionuclides can be released during a period of thousands of years after full saturation of the bentonite barrier and the thermal phase.

Transport parameters and the discretization of the system, are implemented in a 2D axisymmetric geometry. The multi-barrier system is emplaced in clay and a solubility limited source term for the selected radionuclides is assumed. Kinetics and chemical equilibria reactions are simulated using parameters obtained from experiments. Additionally, porosity changes due to mineral precipitation/dissolution and feedback on the effective diffusion coefficient are taken into account. Protonation/deprotonation, ion exchange reactions and radionuclide inner-sphere sorption is considered.

Numerical simulations show, that, when the canister corrosion starts, the redox potential decreases, magnetite precipitates and H\textsubscript{2} is formed. Furthermore, the aqueous concentration of Fe(II) increases due to the presence of magnetite. By considering binding to montmorillonite via
ion exchange reactions, the bentonite acts as a sink for Fe(II). Additionally, magnetite forms a chemical barrier offering significant sorption capacity for many radionuclides. Finally, a decrease of porosity in the bentonite/canister interface leads to a further deceleration of radionuclide migration. Due to the complexity of reactive transport processes in saline environments, benchmarking of reactive transport models (RTM) is important also to build confidence in those modelling approaches. Development of RTM benchmark procedures is part of the iCROSS project (Integrity of nuclear waste repository systems - Cross-scale system understanding and analysis) funded by both the Helmholtz Association and the Federal Ministry of Education and Research (BMBF).