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Degradation of concrete-based barriers by Mg-containing brines: From laboratory experiments via reactive transport modelling to overall safety analysis in repository scale

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The Morsleben nuclear waste repository (ERAM) for low- and intermediate-level radioactive waste is located in an old rock salt and potash mine in Northern Germany. From 1971 to 1998, approximately 36 800 m^3 of waste have been disposed of. Now, waste disposal is finished, and the repository has to be backfilled and sealed. The closure concept is based on extensive backfilling of the salt mine with an inexpensive concrete mixture. The major disposal areas, containing most of the waste, will be isolated from the rest of the mine building by sealing the connecting access tunnels.

The good geological situation (an intact cap rock with very small flow rates) provides an excellent basic condition for a safe repository. However, the access of water to the remaining parts of the mine cannot be excluded. The brines that are formed in contact with potash salts will contain Mg in a concentration that depends on various factors and cannot be predicted. For backfill and closure of the repository, no common material chemically stable against brines of all possible compositions is available. Salt concrete has a low permeability but is corroded by brines containing Mg, whilst concrete based on Sorel phases is decomposed if the Mg-content of the brines is too low. In each of these alternatives, corrosion results in a strong increase of permeability and a loss of the mechanical integrity of the material. However, for large hydraulic seals the flow of corroding brines is limited because of the high hydraulic resistance of the barrier. Thus, the barriers will persist for a long time in spite of the chemical incompatibility of building material and brine.

For the planning of the backfill and closure measures as well as for the license application procedure it is crucial to demonstrate that the seals of the major disposal areas will keep their function for a sufficient long time. This lifetime depends among others on the corrosion capacity of the brine to the building material, on the initial hydraulic permeability and possibly on inhomogeneities like fractures and the hydraulic behaviour of the excavation damage zone (EDZ).

Experimentally, the corrosion capacity of the brines to the concrete cannot be directly determined by throughflow experiments because the initial hydraulic permeability of the original building material is far too low. Instead, the decrease of magnesium, the main corroding agent in the brines, has been measured in cascade experiments with grounded cement mortar. The results of these experiments have been reproduced with geochemical modelling. However, those model calculations reveal that the stoichiometry of this reaction strongly depends on the assumptions about the relative stability of the potentially formed mineral phases, especially the various magnesium-silicate-hydrate-phases. As a pragmatic approach, the probability density function of the corrosion capacity has been estimated by stochastic calculations including the variation within a reasonable bandwidth of the chemical composition of each mortar components and the thermodynamic data of the critical mineral phases.

Subsequently, the corrosion of the sealed access drifts in repository scale has been simulated by a reactive transport model, combining advective / dispersive transport and variable hydraulic permeability as function of the reaction progress. In the model, the chemistry of the corrosion process has been abstracted to one single equation. This allowed a fine discretisation – more than 10’000 nodes in an auto-adapting 2D-FE-mesh with axial symmetry. The parameter for the reaction rate was chosen on basis of experimental observations and turned out to be nonsensitive. The calculations show that the reaction zone is quite narrow – less than 5 m in a seal of 130 m length – as the reaction rate is much faster than the transport processes.

With this model, the influence of a persistent EDZ in the host rock on the degradation of the hydraulic seal was studied. Corrosion in radial direction towards the axis of the seal would quickly create an expanding preferred flowpath if it occurred over the whole length of the seal simultaneously and would thus destroy the building in a rather short time. The model results show, however, that the diffusion process from the EDZ into the seal is fast compared to the transport velocity along the EDZ. Thus the corroding agent is extracted from the EDZ within a transport distance of a few meters what means that the reaction zone is almost vertically planar and that along the remaining length the barrier keeps intact for a sufficiently long time. This justifies the use of a 1D-approximation of the corrosion process in the complex transport model for the safety analysis for the whole repository system which comprises all relevant processes taking place in the salt structure and the surrounding geosphere.