EGU21-898
https://doi.org/10.5194/egusphere-egu21-898
EGU General Assembly 2021

Hydrogen gas bubble nucleation from corrosion of steel in compacted bentonite

Alexandros Papafotiou[1], Chao Li[1], Nikitas Diomidis[2], and Olivier Leupin[2]
[1]INTERA Inc. Swiss Branch, Hardstrasse 73, Wettingen, Switzerland
[2]Nagra, Hardstrasse 73, Wettingen, Switzerland

The reference concept for the deep geological disposal of spent fuel and high-level radioactive waste in Switzerland foresees carbon steel disposal canisters surrounded by compacted bentonite buffer material. In support of performance assessments, long-term in-situ corrosion experiments were conducted in Opalinus Clay at the Mont Terri Underground Research Laboratory (URL) in Switzerland, wherein carbon steel coupons were embedded in MX-80 bentonite. The preparation of the steel specimens and bentonite, the exposure in a sealed borehole in the URL, and the retrieval, dismantling and imaging of specimens were conducted under strictly anoxic conditions. Samples were removed for analysis after exposure durations of 372, 628, 1024, and 2008 days. A key finding was the development of visible reddish-brown corrosion fronts around the metal surfaces and along shrinkage cracks that extend up to approximately 0.5 cm into the bentonite. Iron that originated from the corroded surface was transported along the cracks and precipitated as Fe-hydroxides due to oxygen sorbed on bentonite.

The formation of shrinkage cracks is thought to result from a local desaturation of the bentonite near the steel surface. To test this hypothesis i.e., to test the likelihood of a separate gas phase forming in addition to hydrogen mass dissolved in liquid water, it is necessary to evaluate the fate of hydrogen in the bentonite adjacent to the steel surface. For this, a flow and transport numerical model of the steel coupon surface and surrounding bentonite was implemented for the simulation of hydrogen release with the simultaneous consumption of water at the steel surface. The effect of single- and (potentially) two-phase flow with the diffusive and advective transport of the hydrogen and water components in the gas and liquid phases were modelled in a fully coupled manner. The numerical simulations were performed probabilistically in a Monte Carlo framework to account for parametric uncertainty, comprising 1'000 perturbations of all flow and transport parameters used in the model for the bentonite.

Overall, the simulation results are consistent with the hypothesis of a link between cracks observed in the bentonite and a temporary formation of a gas phase that results in preferential pathways for the transport of iron corrosion products. The probability of gas formation in the model lies between 89% and 94% at the steel-bentonite interface and decreases significantly at distance of 1 cm from the steel coupon. Peak gas saturation at the steel-bentonite interface ranges up to approximately 1% with a mean value of approximately 0.18%. In all simulations, any gas

phase forming in the bentonite dissolves back into the liquid phase within 300 days.