Reactive transport investigations of the long-term geochemical evolution of a multibarrier system and impacts on radionuclide migration

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Engineered barrier materials including highly-compacted and Homogenized Bentonite (HB) and low-alkali concrete, such as Low-Heat High-Performance Concrete (LHHPC), are considered in the Canadian concept for a deep geological repository for used nuclear fuel. The difference in initial mineralogy and porewater compositions of bentonite, LHHPC and potential host rocks, such as granite and limestone, results in chemical alterations at the interfaces between these materials. Two scenarios were considered to investigate the alterations at material interfaces by reactive transport modelling using MIN3P-THCm: 1) HB/LHHPC/host rock; and 2) HB/host rock. For both scenarios, excavation damage zones (EDZs) were taken into consideration. Scenarios including LHHPC showed significant local porosity reduction due to mineral precipitation. No significant porosity changes; however, occurred in the simulations without LHHPC. The predicted geochemical evolution depends mainly on the mineralogy and porewater chemical composition of the host rock. In the case of granitic host rock, Calcium Silicate Hydrate (CSH) phases, initially present in LHHPC, are predicted to transform into tobermorite, phillipsite and saponite within 1,000 years. The simulations indicate a complex evolution of porosity, ultimately leading to pore clogging at the HB/LHHPC interface. In the case of limestone host rock, saponite and sepiolite are the dominant minerals formed in the LHHPC and clogging occurs at the LHHPC/limestone interface. A sensitivity analysis on initial effective diffusion coefficients in LHHPC indicates that enhanced mobility of ions, due to higher diffusion coefficients, results in earlier pore clogging. The location and timing of pore clogging relative to radionuclide release have a strong impact on the migration of radionuclides across the interfaces. The impact of porosity reduction on radionuclide migration is most significant, if clogging occurs before radionuclide release.