

An experimental study of the rock-water interaction of an evolving hyperalkaline leachate plume from a cementitious repository for low- and intermediate level radioactive waste

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Several concepts for a geological disposal facility (GDF) for low- and intermediate-level radioactive waste will employ cement in repository construction and within the engineered barrier system. Following closure, groundwater will resaturate the GDF and leach the cement, producing hyperalkaline plume that may react with the host rock producing an alkali disturbed zone (ADZ). The hyperalkaline cement leachate will evolve from an initial high-K leachate (pH~13), to a Ca-OH fluid (pH 12.5 to 10) buffered initially by the dissolution of portlandite (Ca(OH)₂) and subsequently by dissolution of calcium silicate hydrate phases. Eventually, the alkaline porewater in the ADZ will be displaced by background groundwater, which may react with the alkali alteration products within the ADZ.

The long-term mineralogical evolution of the ADZ has been studied in a series of flow-through experiments using PEEK columns packed with a "model" host rock containing quartz, feldspars, mica, chlorite, kaolinite and minor hematite, which were sequentially-reacted with: K-Na-Ca-OH young cement leachate (YCL: ~pH 13.1), followed by; Ca(OH)₂-saturated intermediate cement leachate (ICL: pH 12.3), and finally by; a dilute (Ca-HCO₃-type) or a saline (Na-Mg-Cl-SO₄-HCO₃-type) background groundwater. The columns were reacted at 50°C, for 300 days during each stage, and the fluid chemistry and flow rate were monitored throughout the experiment. After each stage, a number of columns were petrographically-examined to characterise the mineralogical alteration and its impact of the pore structure and flow paths.

Significant mineralogical reaction was observed with the dissolution of quartz, feldspar and clay minerals) and the precipitation of secondary Ca-K-silicate during reaction with YCL, followed by replacement of these early products by a higher Ca:Si Ca-silicate gel during the ICL stage. Reaction with background groundwater in the final stages of the experiment resulted to the replacement of the Ca-silicates by calcite, ettringite-thaumasite-like phase. A progressive reduction in permeability, associated with pore blocking by secondary phases was observed as the system evolved.