An experimental study of the chemical evolution of the fluid plume from a repository for low- and intermediate level radioactive waste

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Many concepts for the geological disposal of low- and intermediate-level radioactive waste will employ cements both in repository construction and within the engineered barrier system. Following closure, groundwater will re-saturate the repository and leach the cement, producing a hyperalkaline plume that will then react with the host rock producing an alkali disturbed zone (ADZ). The hyperalkaline cement leachate will evolve from an initially K-dominated 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 the incongruent dissolution of calcium silicate hydrate hydrogels. Eventually, the alkaline porewater in the ADZ will be displaced by background groundwater, which may then react with the secondary alteration products. The chemical evolution of the ADZ fluids 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-HCO_3-type) background groundwater (GW). The columns were reacted at 50°C, for a total of 900 days. Samples for fluid chemistry were collected throughout the experiment and analysed to determine the evolution of the outflowing fluid.

The pH of the reacted fluids generally reflects that of the initial fluids. [Na], and [Cl] are controlled by the initial fluid chemistry and remain close to the input concentration to the columns. [Si], and [Al] show large gains in the early YCL fluids, but levels then decrease and are below detection in the ICL reacted fluids. Both Si and Al reappear at low levels in the final GW reacted fluids. This behaviour suggests secondary phase formation followed by later reaction. Ca is removed from the YCl and ICL fluids, this being most apparent in large reductions seen with the ICL fluid but small gains are seen with the GW. Thus, these experimental data show evolution of the ADZ fluids is controlled by both primary and secondary mineral reaction.