Mineralogical evolution of the alkali disturbed zone around a geological disposal facility for radioactive waste

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Several concepts for a geological disposal facility (GDF) of low- and intermediate-level radioactive waste will employ cementitious materials both in repository construction and within the engineered barrier system. Groundwater resaturation of the GDF following closure will result in leaching of the cement, producing a hyperalkaline plume that will interact with the surrounding host rock to produce an alkali disturbed zone (ADZ). The mineralogical evolution of the ADZ has been investigated experimentally within the NERC **Bi**ogeochemical **G**radients and **Rad**ionuclide Transport (BIGRAD) Project. A series of replicate PEEK columns packed with gently disaggregated "model" host rock containing quartz, feldspars, mica, chlorite, kaolinite and minor hematite were sequentially-reacted with: (1) K-Na-Ca-OH fluid (~pH13.1) representing a young cement leachate (YCL); followed by; (2) a $Ca(OH)_2$-saturated fluid (pH12.3) representing intermediate cement leachate (ICL), and finally; (3) $Ca\text{-}HCO_3$-type fluid, representing eventual resaturation of the ADZ by groundwater. The columns were reacted at 50 °C, for ~300 days at each stage, and fluid chemistry and flow rate were monitored throughout the experiment. After each stage, a number of columns were examined by scanning electron microscopy, optical microscopy and energy-dispersive X-ray microanalysis, to characterise the mineralogical alteration and its impact of the pore structure and flow paths.

Quartz, feldspar, muscovite, illite and kaolinite dissolved to varying degrees in the YCL. This correlated with a large initial rise in Si and Al in solution, after which Ca, Si and Al were removed (accompanied by a decrease in K) by the precipitation of a K-Al-bearing calcium silicate hydrate (CSH) gel. This early-formed CSH gel is replaced by CSH gel with a higher Ca:Si ratio during subsequent reaction with ICL. Well-crystallised CSH was also formed during the ICL stage. Following reaction with dilute $Ca\text{-}HCO_3$ groundwater, the HCO_3 is rapidly removed from the fluid by reaction with CSH to form calcite close to the inlet of the column. Further along the flow path, the CSH dissociates Ca to solution, to form a lower Ca:Si ratio CSH gel. The permeability of the columns was progressively reduced, as a result of pore blocking by secondary phases, as the experimental system evolved.