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Mineralogical and geochemical evolution of engineered barrier materials in an argillite-hosted nuclear waste repository: Wyoming Bentonite and Opalinus Clay hydrothermal experiments

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High temperature (200–300°C) and pressure (∼150 bar) hydrothermal experiments were conducted to evaluate the stability of engineered barrier system (EBS) materials within an argillite-hosted nuclear waste repository. Eight experiments involving Opalinus Clay (shale) from Mont Terri, Switzerland and Wyoming Bentonite from Colony, WY were designed to mimic hydrothermal reactions within Opalinus Clay wall rock and at the interface between wall rock and Wyoming Bentonite, both in the presence of synthetic Opalinus Clay groundwater. Experiments were reacted at high temperature repository conditions for between 6 weeks and 6 months.

Fluid samples were collected periodically throughout the experiment duration and analyzed for major cations and anions. The pH (25°C) of the solution in each experiment started at ∼7.5 and decreased to 5–6 by the end of the experiment. Aqueous SiO_2 remained saturated with respect to quartz throughout the 200 and 300°C experiments. The solid-reaction products and were characterized post experiment via XRD, XRF, SEM, and EMPA. The major mineralogical products observed in the 6-week, Opalinus Clay-only 300°C experiment was the formation of abundant Ca-rich analcime (Na/Na+Ca = 0.21, Si/Al = 2.22) along fractures and edges of Opalinus Clay fragments. In comparison, analcime with an intermediate composition (Na/Na+Ca = 0.36–0.64, Si/Al = 2.67–2.97) formed in 6-week to 6-month 300 °C experiments that included Wyoming Bentonite and Opalinus Clay. The 6-month experiment produced analcime with the highest Si/Al ratio. Zeolite formation was not observed in 8-week, 200°C experiments. Preliminary XRD results from the clay-mineral fraction from the 6-month 300°C experiment do not exhibit significant smectite peak shifts or evidence of illite-smectite interlayering. Further, the 100% expandability estimates from 002/003 glycol-saturated smectite peak positions indicate silica cementation did not occur. In comparison, XRD analyses of Opalinus Clay fragments indicated some authigenic illite growth.

The transformation of Na-montmorillonite to illite within the Wyoming Bentonite EBS material was likely limited by the bulk chemistry of the system (i.e. low potassium and aluminum, silica saturation). Authigenic illite within Opalinus Clay fragments likely nucleated on pre-existing illite in the shale. Previous experiments with only Wyoming Bentonite documented the transformation of precursor clinoptilolite into analcime with a sodium- and silica-rich composition (Na/Na+Ca = 0.80–0.90, Si/Al = 3.05–3.38). Results from Opalinus Clay-only 300°C experiments indicate that the dissolution and re-precipitation of other phases, such as kaolinite, calcite, and smectite, may also contribute to zeolite formation, as Opalinus Clay does not contain the clinoptilolite precursor. Further, the low permeability of the Opalinus Clay rock limited authigenic analcime formation to fractures. These results are significant for understanding zeolite formation, clay mineral stability, and silica precipitation within EBS materials of a high-temperature repository.