Hydrothermal interaction of Opalinus Clay, Wyoming Bentonite, and Portland Cement

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The U.S. Department of Energy aims to characterize fluid-solid interactions at elevated temperatures of candidate engineered barrier system materials to isolate heat-generating spent nuclear fuel in a deep geological repository. Four hydrothermal experiments examining the interaction of Opalinus Clay (argillite wall rock), Wyoming Bentonite, and ordinary Portland Cement were conducted to understand the mineralogical and geochemical evolution of a high temperature repository with cement pore fluids. Experiments were conducted at ~9:1 synthetic Opalinus Clay groundwater:rock ratio, 200°C, and 150 bar for 8 weeks. Coupons of either 316 stainless steel (SS), 304 SS, or low carbon steel were added to simulate the presence of a waste cansiter. Results are compared to previous experiments with Wyoming Bentonite ± Opalinus Clay.

The aqueous geochemistry of the reaction fluid was monitored during each experiment. Quench pH (at 25 °C) dropped from initial values of ~10 to 8.5. Aqueous silica activities calculated at 200°C indicate undersaturation with respect to quartz. Characterization of the solid-phase run products show significant mineralogical changes in the Opalinus Clay fragments and bentonite clay. Quantitative X-ray diffraction (XRD) estimates show dissolution of montmorrilonite and XRD of the oriented clay fraction indicate the formation of illite-smectite and chlorite in the bentonite groundmass. Zeolite formation was abundant; large analcime crystals (up to ~0.5 mm diameter) are observed coating the Opalinus Clay fragments and within the clay matrix. Electron microprobe analyses of analcime crystals coating the Opalinus Clay fragments have higher Na/(Na+Ca) values but similar Si/Al ratios relative to the grains in the clay matrix.

In contrast, previous experiments with only Opalinus Clay and Wyoming Bentonite at 200 °C did not produce zeolites and no structural changes to montmorillonite were observed. These results highlight the potential for a highly reactive environment at the bentonite-cement interface within engineered barrier systems.