Attenuating Cesium Migration with Bentonite in Nuclear Repositories

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Radionuclide migration in deep borehole (DBH) and engineered barrier systems (EBS) is a concern regarding bentonite seal stability and nuclear waste isolation after emplacement. Zeolites are common products of bentonite in repository conditios and in the presence of a Cs-rich hydrothermal fluid, Cs-rich zeolites may crystalize as authegenic phases within the bentonite seal material. Determining the interaction between bentonite and a Cs-rich fluid is important for evaluating Cs isolation during mineral alteration of bentonite in nuclear waste repositories.

The experiments were conducted in cold-seal pressure vessels from 200 to 450 °C and 500 to 1000 bar, typically of P, T condition of both repositories. Gold capsules were loaded with a 2:1 water:rock ratio of unprocessed bentonite and an aqueous fluid containing 2 equilimolar $CaCl_2$-CsCl-NaCl. The run products were characterized via SEM (images) and EMPA (mineralogy).

EMPA at 200 °C and 1 kbars observed minor zeolites, but instead a Cs-rich glass and pumice (13.63 ± 2.36 wt.% Cs, n=14 and 13.27 ± 0.44 wt. % Cs, n=10, respectively) were observed. At temperatures of 300 °C and 1 kbar, Cs-rich zeolites was produced (19.01 ± 0.59 wt.% Cs, n=6 and 18.25 ± 1.25 wt.% Cs, n=9). Cs – Ca-rich zeolites formed during the 400 °C experiments (13.56 ± 1.69 wt. % Cs, n=24). Preliminary SEM-EDS analyses were performed on the other experiments. The 250 °C and 1 kbar show Cs wt.% of 21.61 ± 9.65 (n= 15). At 400 °C and 500 bars, Cs concentrations are 18.15 ± 6.72 wt.% (n=6). The experiment at 450 °C and 500 bars have Cs concentrations of 15.61 ± 5.27 wt.% (n=8).

The results demonstrate Cs entrapment in mineral alteration products varies with temperature and pressure. Cs was incorporated into zeolites and glass in the presence of a Cs-rich brine. The results demonstrate the temperature and pressure dependence of the formation of Cs-rich phases in which Cs is better incorporated at moderate temperature and presssures typical of respositories.