



Cesium Entrapment in Zeolites within Engineered Barrier Systems

Marlena Rock (1), Florie Caporuscio (1), Kirsten Sauer (1), Katherine Norskog (2), and James Maner (3)

(1) Los Alamos National Laboratory, EES-14, Los Alamos, United States (mrock@lanl.gov), (2) Environmental Resources Management (ERM), Nashville, United States, (3) University of Texas, Austin, Austin, United States

Mineral alteration in engineered barrier systems (EBS) is a concern regarding bentonite seal stability and nuclear waste isolation after emplacement. In EBS designs, bentonite may be present as a barrier between the canister and host rock to provide: 1) a physical barrier to prevent natural fluid from interacting with the waste package and 2) a chemical barrier by attenuating radionuclide migration, such as Cs, if a release occurs. At elevated repository temperatures and in the presence of a Cs-rich hydrothermal fluid, the mineral pollucite may crystallize within the bentonite EBS material. Pollucite, $(\text{Cs,Na})_2\text{Al}_2\text{Si}_4\text{O}_{12} \cdot 2\text{H}_2\text{O}$, is a zeolite mineral and is a common hydrothermal product. Determining the interaction between bentonite and potential Cs-rich hydrothermal fluid is important for evaluating Cs-isolation during mineral alteration in a high temperature nuclear waste repository.

Experimental work was conducted at the pressure and temperatures conditions that EBS materials would experience in underground repositories. The experiments were conducted in Udimet 870 cold-seal pressure vessels from 150 to 400°C and 300 to 1000 bar from two weeks. Gold capsules were loaded with a 2:1 water:rock ratio of unprocessed bentonite from Colony, Wyoming and an aqueous fluid containing 2 molal CaCl + CsCl + NaCl with a starting pH of 5.76. At the conclusion of the experiments, the run product was removed from the capsules and characterized via SEM and EMP.

The results demonstrate Cs entrapment in mineral alteration products varied with temperature and pressure. At 200°C and 1000 bars, pollucite did not form, but a Cs-rich glass ($13.62 \pm 2.36 \text{ Cs}_2\text{O wt.}\%$) was observed. At temperatures of 300 and 400°C and 1000 bars, pollucite was produced ($19.01 \pm 2.36 \text{ Cs}_2\text{O wt.}\%$ and $13.56 \pm 1.72 \text{ Cs}_2\text{O wt.}\%$, respectively).

The results of these experiments document the stability of zeolite minerals (analcime-pollucite-wairakite_{ss}) under repository conditions. Clinoptilolite (and remnant glass) are present in the starting bentonite material and are observed to recrystallize to analcime above 300°C in the absence of a Cs-rich brine. In the presence of a Cs brine, we have documented the incorporation of Cs in glass and of pollucite. Further, the results of this study demonstrate the temperature dependence of the formation of Cs-rich phases. When clinoptilolite alters from a high temperature thermal pulse of a repository, analcime/wairakite will form and exist as radionuclide sorbing phases, while pollucite would incorporate Cs into the crystal structure if released from the waste container.