Zeolite capture of Cs in high temperature contaminated environment

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The U.S. Used Fuel Disposition Campaign is conducting a Deep Borehole repository demonstration project for disposal of CsCl waste forms. Experimental work will constrain the P, T conditions which "seal" material will experience in deep borehole repositories. Mineral alteration is one of the concerns regarding bentonite seal stability and radionuclide capture after emplacement and we therefore focused on borehole seals conditions at 5 Km depth.

Experiments were performed at 1 K bar and 400 °C over a period of ~2 weeks. An unprocessed bentonite from Colony, Wyoming and CaCl, NaCl, and Cs-Na-Ca-Cl 2 molal brine was used at a 2:1 water:rock ratio. The baseline experiments contained brine + clay, and buffered at low oxygen fugacity. The experiments were designed to determine the stability of zeolite minerals (analcime-pollucite-wairakitess) under repository conditions.

Potassium was mobilized and exchanged with interlayer Na, transitioning the clay from Namontmorillonite to K-smectite. Clinoptilolite (and remnant glass) present in the starting bentonite mixture are unstable at 400 °C. Hence, precursor clinoptilolite underwent recrystallization at 150°C producing, a Si-rich analcime in addition to authigenic silica phases in the Na-rich brine. The analcime reaction is:

clinoptilolite + 0.92 Na+ + 0.02 Ca₂₊ → 8 **analcime** + 0.14 K+ + 11.62 **SiO**₂ + 12.64 H₂O + 0.40 H+

Wairakite and pollucite were formed by similar phase transformations. When clinoptilolite alters at the 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.