Clay Seals Mineralogy in Deep Borehole Repository

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The U.S. Used Fuel Disposition Campaign is conducting research to evaluate disposal of smaller DOE-managed waste forms in deep boreholes. Mineral alteration is one of the concerns regarding bentonite seal stability and radionuclide capture after emplacement, therefore we focused on borehole seals conditions at 3 and 5 Km depth.

Two experiments were performed: 1) Bottom of borehole (1 K bar and 400 °C) and 2) Base of isolation seals (300 bar and 150 °C). These baseline experiments contained brine (CaCl, NaCl, and Cs-Na-Ca-Cl 2 molar brine) + clay at a 2:1 water:rock ratio, and low oxygen fugacity. The experiments were designed to determine the stability of clay (smectite /illite) and zeolite minerals (analcime-pollucite-wairakitess) under repository conditions.

Potassium was mobilized and exchanged with interlayer Na, transitioning the clay from Na-montmorillonite to Ksmectite. Mineral products in the high pressure experiment were hornfels type, while the lower pressure experiment were diagenetic zeolite. Clinoptilolite (and remnant glass) in the starting bentonite mixture was unstable at 400 °C. Hence, clinoptilolite underwent recrystallization at 150°C, producing a Si-rich analcime in addition to authigenic silica phases in the Na-rich brine. The reaction is:

 $\begin{array}{l} \textbf{clinoptilolite} + 0.92 \ Na + 0.02 \ Ca_2 = 8 \ \textbf{analcime} + 0.14 \ K + \\ 11.62 \ \textbf{SiO}_2 + 12.64 \ H_2O + 0.40 \ H \end{array}$

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. Cesium released from waste containers would incorporate into the pollucite crystal structure.