Summary of experimental Fe-Saponite growth at the bentonite / steel canister interface. Nuclear repository canister corrosion implications

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The United States has initiated the Spent Fuel and Waste Storage and Transport (SFWST) Campaign to evaluate various generic geological repositories for the disposal of spent nuclear fuel. Most previous international work describes Engineered Barrier Systems (EBS) for repositories focused on low temperature conditions. Our hydrothermal experiments on EBS materials were conducted to characterize high temperature interactions of bentonite clay with candidate waste container steels (304SS, 316SS, low-C steel).

Over eight years of experiments were performed using Dickson reaction cells at temperatures and pressure of up to 300° C and 15 - 16 MPa, respectively, for five to eight weeks. Wyoming bentonite was saturated with a 1,900 ppm K-Ca-Na-Cl solution in combination with stainless and low-C steel coupons.

No smectite illitization was observed in these reactions. However, a partial K-enrichment of -smectite occurred, providing a precursor to illitization. It would appear that illitization was retarded due to a limitation of K^+ in the closed-system.

Notable clay mineral reactions occurred at the steel surfaces. Authigenic chlorite and Fe-saponite grew with their basal planes near perpendicular to the steel plate, forming a $10 - 100 \mu m$ thick 'growth' layer. Partial dissolution of the steel plates was likely the iron source for chlorite/saponite formation with steel plates acting as a substrate for chlorite/saponite growth. Steel corrosion / saponite growth values will be presented. XRD and microprobe analyses of the silicate mantling on the low-carbon steel indicates the phase is a Fe saponite with a composition of (Na_{0.09},Ca_{0.03}) (Fe_{2.20}Mg_{0.12}Al_{0.86}) (Al_{0.58} Si_{3.42})O₁₀(OH)₂. Stainless steel (304) is mantled by a chlorite/Fe saponite mixture. This phyllosilicate mix is high in Fe (33.99 wt% FeO), Cr (1.35 wt % Cr₂O₃) and Ni (1.34 wt % NiO).

Mineral growth on the waste containers was influenced by the container, buffer, and fluid compositions, in addition to pressure and temperature conditions. No significant clay mineral alteration was apparent away from the steel-smectite interface. This research shows that the waste container may act as a substrate for mineral growth in response to corrosion. The role of these Fe-rich minerals as passivating agents on steel canisters at