Alteration of Clay Minerals and Zeolites in Hydrothermal Brines

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Abstract: Clay minerals and zeolites, candidate backfill minerals for nuclear waste repositories, were treated with saturated NaCl brine and Mg-rich (Mg-Ca-Na-K) brine at 200 ° C and 300 ° C for 4 weeks under a confining pressure of 30 MPa. The Al concentrations released in NaCl brine were lower than those in Mg-rich brine at both temperatures indicating that the Mg-rich brine is more acidic than the NaCl brine under these hydrothermal conditions. The Si concentrations in both brines were low because of the relatively acidic conditions developed during the hydrothermal treatment. As determined by X-ray powder diffraction or by specific Cs and Sr sorption measurements, no alteration could be detected in clay minerals treated with NaCl brine at 200 ° C. Among the zeolites tested, only phillipsite and erionite altered to analcime in NaCl brine at 200 ° C. Zeolites and most of the clay minerals tested did not alter in the Mg-rich brine treated at 200 ° C. Vermiculite altered to randomly interstratified vermiculite/K-vermiculite (mica-like) by selective K uptake from the Mg-rich brine.

At 300 ° C, the clay minerals did not greatly alter, whereas the zeolites altered to analcime and/or albite in the presence of the NaCl brine. In the Mg-rich brine, Al-rich montmorillonite from Wyoming did not alter, whereas Al-poor montmorillonite from Texas altered to randomly interstratified montmorillonite/illite at 300 ° C. Vermiculite collapsed to form K-vermiculite (~ 10.2 Å) by the selective uptake of K from the Mg-rich brine at 300 ° C. Most of the zeolites altered to smectites in the Mg-rich brine at 300 ° C because of the acidic conditions generated by the hydrolysis of Mg. The selective Cs-sorption K_d decreased from 11,700 for untreated phillipsite to 240 and 15 for the hydrothermally produced analcime/albite mixtures from the phillipsite at 200 ° and 300 ° C, respectively, in NaCl brine. These results suggest that montmorillonites and mordenites are relatively more resistant than vermiculite or other zeolites at elevated temperatures and pressures in concentrated hydrothermal brines expected in a salt repository.

: Key Words • Backfill • Bentonite • Cesium • Clinoptilolite • Hydrothermal • Mordenite • Salt repository • Smectire • Strontium • Vermiculite • Zeolite

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