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Montmorillonite stability in bentonite under high-temperature hydrothermal conditions: Effect of initial composition and Mg-rich solutions

CHANYOUNG SEO, YUJIN BYUN AND HO YOUNG JO

Department of Earth and Environmental Sciences, Korea University

Bentonite buffers in geological disposal systems rely on the structural stability of montmorillonite under long-term thermo–hydro–chemical conditions. This study evaluated montmorillonite alteration at 200 °C and examined whether K-feldspar–montmorillonite interaction and an Mg-rich solution induce illitization or structural degradation. Batch hydrothermal experiments were conducted for one year at 200 °C using mixtures of K-feldspar with Ca- and Na-montmorillonite in deionized (DI) water and a 1 M $MgCl_2$ solution. Reaction solutions were analyzed for pH and electrical conductivity (EC), and dissolved element concentrations were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES). Mineralogical changes were determined by X-ray diffraction (XRD) through comparison of basal spacing (d001) before and after ethylene glycol (EG) solvation. Additional mineralogical characterization was performed using scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS) and nuclear magnetic resonance (NMR) to further assess alteration features, and cation exchange capacity (CEC) was measured to quantify changes in buffer performance. In DI water, pH remained between 7 and 8, and EC increased to 10–500 μS/cm, due to release of K^+, Na^+, and Ca^{2+} during K-feldspar–montmorillonite interaction. In 1 M $MgCl_2$, pH decreased to 4–5 and EC increased markedly, reflecting extensive Mg^{2+} exchange and related reactions. No 10 Å illite (001) peak developed in any sample. The expansion of d001 from 12–14 Å to 16–17 Å after EG treatment indicates that interlayer swelling capacity was largely preserved and that no clear evidence of illitization was observed. In Mg^{2+}-bearing systems, EG-induced expansion was partially restricted, indicating limited interlayer modification by Mg^{2+}. These results show that K^+ released from K-feldspar under the tested conditions is insufficient to induce illitization, even at 200 °C. Mg^{2+} exchange reduces swelling capacity but does not transform montmorillonite into illite. Montmorillonite structure remained stable under restricted K^+ supply from K-feldspar, possibly limiting the extent of buffer alteration in high-temperature disposal environments.