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Accelerated Illite-Smectite Transformation in Bentonil-WRK for Nuclear Waste Repository Applications

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Bentonite is considered a buffer material in high-level radioactive waste (HLW) repositories due to its swelling capacity, low permeability, and cation exchange ability. Composed mainly of dioctahedral smectite, it may undergo smectite-to-illite (I-S) transformation under long-term heat and groundwater exposure, altering its properties. Since natural illitization occurs over geological timescales, this study investigates accelerated hydrothermal conditions for synthesizing I-S using Bentonil-WRK, a candidate bentonite for the Korean HLW repository.

A systematic approach was employed to investigate the effects of temperature and reaction time on the I-S transformation. The reaction temperature was varied from 150°C to 330°C while maintaining a fixed reaction duration of 7 days to evaluate the impact of temperature. Based on these results, 250°C was selected as a representative condition to examine the influence of reaction time, which was extended up to 42 days. The experiments were conducted in a 1 M KCl solution to promote illitization under hydrothermal conditions. After the reactions, the interlayer cations in the samples were replaced with Ca^{2+}.

X-ray diffraction (XRD) analysis showed progressive I-S formation with increasing temperature and reaction time, accompanied by a decrease in smectite (001) peak intensity and an increase in I(001)/S(002) and I(003)/S(005) peak ratios. Ethylene glycol-saturated oriented samples confirmed the expansion behavior of the altered clays. The results further demonstrated that I-S transformation predominantly occurs up to 250°C. However, at temperatures above 300°C, smectite dissolution becomes more pronounced, accompanied by the precipitation of orthoclase. At 250°C, even when the reaction time was extended to 42 days, no distinct orthoclase precipitation was observed, indicating that illitization remains the dominant process under these conditions. Solid-state nuclear magnetic resonance (NMR) spectroscopy was employed to investigate changes in the atomic environments of Si and Al during the I-S transformation. The results revealed an increase in $^{[4]}Al$ content as illitization progressed, suggesting a gradual substitution of Al in the tetrahedral layer.

These findings provide insights into the thermally induced alteration of bentonite and the key factors governing I-S transformation under repository-relevant conditions. The results contribute to refining long-term safety assessments of bentonite buffer materials in HLW repositories.