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Characterization of neoformed illite from hydrothermal experiments at 250 °C and $P_{v, soln}$: An HRTEM/ATEM study

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ABSTRACT

Solid products from hydrothermal experiments conducted at 250 °C and $P_{v,soln}$ were characterized by powder X-ray diffraction (XRD) and ATEM/HRTEM. Experiments were conducted with muscovite, kaolinite, and quartz or amorphous silica in 2M KCl solutions for 43 to 176 d. Post-experiment solution compositions lie either within the illite(0.88 K) stability field or on the illite(0.88 K)-kaolinite or illite(0.88 K)-diaspore univariant boundaries in log (a_{K}^{+}/a_{H}^{+}) vs. log $a_{H_4SiO_4}$ activity space. Transmission electron microscopy (TEM) observations of muscovite grain edges reveal the neoformation of illite crystals with a range of compositions (ATEM) from 0.31 to 0.89 $K/O_{10}(OH)_2$. The range of K-contents appears to narrow toward 0.88 $K/O_{10}(OH)_2$ with increased experiment duration. HRTEM suggests the presence of 2 to 11 layer fundamental particles composed of illitic layers with 10 Å periodicity. Fundamental particle thicknesses increase toward an average of 8 layers/particle with increased experiment duration. In the longer duration experiments, fundamental particle thicknesses were normally distributed about thicknesses of 4 and 8 layers, whereas fundamental particles with thicknesses <4 layers were common in a shorter duration experiment.

The compositions and structure of the illites are consistent with the multiphase model, which states that the smectite-to-illite transition occurs through the step-wise formation of solubility-controlling phases consisting of fundamental particles with thicknesses of 1, 2, 4, and ≥8 layers. The increase in K-content and fundamental particle thickness with the extent of reaction suggests that the illite crystals underwent a prograde reaction culminating in the formation of end-member illite [0.88 $K/O_{10}(OH)_2$]. This reaction, in conjunction with the previously observed, retrograde reaction from muscovite to end-member illite, demonstrates the stability of end-member illite in the system K_2O-Al_2O_3-SiO_2-H_2O at 250 °C.