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$_{H_2}$O 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$_2$O-Al$_2$O$_3$-SiO$_2$-H$_2$O at 250 °C.

INTRODUCTION

The formation and stability of end-member illite were investigated experimentally in recent studies by Yates and Rosenberg (1996, 1997). These experiments combined muscovite, kaolinite and quartz, or amorphous silica with 2M KCl solutions from 100 to 250 °C to define mica-like, solubility-controlling phases with compositions of 0.29, 0.51, and 0.88 K/O$_{10}$(OH)$_2$. The experiments at 200 and 250 °C were of particular interest because formation of roughly parallel sets of illite crystals on the edges of altered muscovite grains resulted. The present study characterizes these illites using high resolution transmission electron microscopy (HRTEM) and analytical transmission electron microscopy (ATEM) techniques to gain a better understanding of the smectite-to-end-member illite transition. Goals include determination of solid-state properties, such as composition, structure, and interlayering of illites for which the equilibrating solution compositions are well known.

Nadeau et al. (1984a, 1984b, 1984c) proposed that interstratified I/S is made up of illitic fundamental particles in which the basal interfaces between units behave as smectite interlayers. In this fundamental particle model, smectite, R1 I/S, and R3 I/S are made of populations of discrete fundamental particles with thicknesses of 1, 2, and ≥4 silicate layers, respectively, whereas end-member illite consists of ≥5-layer fundamental particles. Numerous HRTEM studies of mixed-layer I/S, however, reveal sequences of illitic layers with thicknesses greater than would be predicted by the fundamental particle model (Klimentidis and MacKinnon 1986; Ahn and Peacor 1989; Ahn and Buseck 1990). In an HRTEM study of mixed layer I/S Veblen et al. (1990) observed crystallographic coherence across stacked fundamental particles, including their smectitic interfaces, and suggested that fundamental particles should be redefined as purely illitic units that are coherently stacked within I/S crystallites. Invoking this modified form of the fundamental particle model of Nadeau et al. (1984a, 1984b, 1984c), Rosenberg et al. (1990) proposed a multiphase model for mixed-layer I/S. In this model, the smectite-to-illite transition is