

Thermodynamic and kinetic factors during smectite evolution: A study based on experiments and thermodynamic calculations

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The stability of clay minerals (e.g., smectite) has great significance on clay geochemistry and industrial applications due to their abundance on the surface of the Earth and wide ranges of usages in various fields. Thermodynamic and kinetic factors during clay evolution essentially control the stability of clay minerals. Thus, the comprehensive assessments on thermodynamic and kinetic driving force are necessary to predict the physic-chemical behaviors of clay minerals. In present study, Four series of Mg,Ni-saponite (i.e., 150-150, 220-220, pMg220+pNigel, pNi220+pMggel) were obtained by hydrothermal treatment of the mechanical mixture of various endmember saponite (Mg-saponite and Ni-saponite) at 150 or 220 °C. The crystal chemistry and morphology of all the saponites were analyzed by XRD, FTIR, and HAADF-STEM. Experimental results indicated that Mg-Ni saponite solid solution only appear in two series (i.e., 150-150 and pMg220+pNigel). The evolution of Mg-saponite and Ni-saponite was relatively independent in Series 220-220 and Series pNi220+pMggel. The possible reaction path in each series was hypothesized and corresponding thermodynamic of each reaction was calculated based on theoretical thermodynamic data. The theoretical thermodynamic data suggested that solid solution is naturally most stable phase in all series. The controlling factors were systematically analyzed by combining the experimental data and thermodynamic results. The rapid formation of solid solution in Series 150-150 and Series pMg220+pNigel may attribute to kinetic or special thermodynamic factors. Small size of particles in Seires 150-150 could drive growth of saponite by some patterns (e.g., Ostwald ripening or particle attachment) to form solid solution. One special thermodynamic path, i.e., the spontaneous replacement of Ni²⁺ for structural Mg of saponite, facilitated the formation of solid solution in Series pMg220+pNigel. However, the formation of solid solution in other series (i.e., 220-220 and pNi220+pMggel) had to rely on a long period of hydrothermal time. Because the large size of particles in Series 220-220 cannot provide effective driving force and Mg²⁺ cannot spontaneously exchange for structural Ni of saponite in Series pNi220+pMggel. The analytical methods in present study can be applied to predict the behaviors of clay minerals in nature systems or industrial applications.