Evolution of Fundamental-Particle Size during Illitization of Smectite and Implications for Reaction Mechanism

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Abstract: Area-weighted thickness distributions of fundamental illite particles for samples of illite and illite-smectite from seven locations (including bentonites and hydrothermally altered pyroclastics) were measured by Pt-shadowing technique, by transmission electron microscopy. Most thickness distributions are described by lognormal distributions, which suggest a unique crystallization process. The shapes of lognormal distributions of fundamental illite particles can be calculated from the distribution mean because the shape parameters α and β^2 are interrelated: $\beta^2 = 0.107\alpha - 0.03$. This growth process was simulated by the mathematical Law of Proportionate Effect that generates lognormal distributions. Simulations indicated that illite particles grow from 2-nm thick illite nuclei by surface-controlled growth, *i.e.*, the rate of growth is restricted by how rapid crystallization proceeds given a near infinite supply of reactants, and not by the rate of supply of reactants to the crystal surface. Initially formed, 2-nm thick crystals may nucleate and grow within smectite interlayers from material produced by dissolution of single smectite 2:1 layers, thereby transforming the clay from randomly interstratified (Reichweite, R = 0) to ordered (R = 1) illite-smectite after the smectite single layers dissolve. In this initial period of illite nucleation and growth, during which expandable layers range from 100 to 20%, illite crystals grow parallel to [001]* direction, and the dimensions of the (001) plane are confined to the size of the original smectite 2:1 layers. After nucleation ceases, illite crystals may continue to grow by surface-controlled growth, and the expandable-layer content ranges from 20 to 0%. This latter period of illitization is characterized by three-dimensional growth. Other crystal-growth mechanisms, such as Ostwald ripening, supply-controlled growth, and the coalescence of smectite layers, do not produce the observed evolution of α and β^2 and the observed shapes of crystal thickness distributions.

Key Words: Crystal Growth • Fundamental Particle • Illite-Smectite • Illitization Mechanism • Lognormal Distribution • Ostwald Ripening

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