Comparison of Structural Models of Mixed-Layer Illite/Smectite and Reaction Mechanisms of Smectite Illitization

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Abstract: This paper compares mechanisms of the reaction of smectite to illite, in light of structural models for interstratified illite/smectite (I/S). The crystal structure of I/S has been described previously by a nonpolar and polar 2:1 layer model. In a nonpolar model, individual 2:1 layers are chemically homogeneous, whereas a polar model assumes a 2:1 layer can have a smectite charge on one side and an illite charge on the other side. Several kinds of data support the polar model; however, more determinations of the negative charge of expandable sites in I/S are needed to confirm such a model.

Assuming a polar 2:1 layer model for I/S, we compare the mineralogical and geochemical consequences of several reaction mechanisms for smectite illitization: 1) solid-state transformation (SST), 2) dissolution and crystallization (DC) and 3) Ostwald ripening (OR). Features of an SST model are the replacement of smectite interlayers by illite interlayers, resulting in gradual changes in interlayer ordering, polytype, chemical and isotopic composition and crystal size and shape. Several SST models are possible depending on the nature of the reaction site (framework cations, polyhedra or interlayers). In contrast, DC models allow for abrupt changes in the structure, composition and texture of I/S as illitization proceeds. Several DC models are possible depending on the nature of the rate-controlling step, for example, diffusional transport or surface reactions during crystal growth. The OR model represents the coarsening of a single mineral where the smallest crystals dissolve and nucleate onto existing larger crystals, allowing for evolution in the overgrowth but not in the template crystal.

An SST mechanism, involving either reacting polyhedra or reacting interlayers, seems to best model illitization in rock-dominated systems such as bentonite. A DC mechanism seems to best model illitization in fluid-dominated systems such as sandstone and hydrothermal environments. Both DC and SST mechanisms can occur in shale. Differences in reaction mechanism may be related to permeability. An OR model poorly describes illitization because of the progressive mineralogical and chemical changes involved. For many geologic environments, it is important to consider alternate origins for I/S such as kaolinite illitization and detrital. Further work is needed to clarify the DC crystal growth process in terms of a structural model of I/S and to determine which specific SST or DC model best characterizes illitization in geologic systems.

Key Words: Illite • Illite/Smectite • Mixed-Layer • Reaction Mechanism • Smectite

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