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# Clay Mineral Thermometry—A Critical Perspective<sup>1</sup>

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**Abstract:** Diagenetic clay minerals usually occur as heterogeneous assemblages of submicroscopic layers consisting of different structure types such as illite, smectite and chlorite, with variable composition within a given structure type, and with highly variable concentrations of imperfections. The dimensions of mixed-layering, the semi-coherent to coherent nature of the structures across the layering, and compositional heterogeneity occur at a scale well below that of an individual thermodynamic phase. These relations imply that most clays are not distinct minerals or phases, and that assemblages of clays in shales and mudstones are incompatible with the phase rule. Such relations are better evaluated in terms of the formation of metastable materials with each small unit having unique chemical properties, rather than as a small number of stable homogeneous phases. Consequently, treatment of most clay minerals in terms of equilibrium stability with either a thermodynamic or experimental approach is subject to error.

Chemical reactions involving most clay minerals are best understood with kinetic models. These involve a great variety of parameters such as time, fluid/rock ratio, deformation history, nature of starting materials and transformation mechanisms, as well as the variables, such as temperature, pressure and composition, that are commonly used to define equilibrium. Solubility experiments on the stabilities of clay minerals are unlikely to attain equilibrium at low temperatures. Moreover, the activity of soluble species may be controlled by surface equilibria, or by absorbed or exchangeable cations. Interpretations of available experiments on the solubility of illite vs. other mineral assemblages are in violation of Schreinemakers' rules and indicate lack of equilibrium.

Predictable sequences of clay minerals as a function of temperature are best understood through the Ostwald step rule, in which clay mineral assemblages undergo reactions in response to kinetic factors that represent reaction progress rather than an approach to equilibrium. Currently used clay mineral thermometers (illite crystallinity, smectite/illite reaction, chlorite composition) are not based on equilibrium reactions. Such systems are not accurate thermometers and therefore have questionable utility.

**Key Words:** Chlorite thermometry • Clay thermometry • Illite crystallinity • Illite solubility • Metastability • Ostwald step rule • Smectite solubility • Stability

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