

Dissolution of well and poorly crystallized kaolinites: Al speciation and effects of surface characteristics

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ABSTRACT

This study compared surface characteristics and dissolution behavior of well-crystallized (KGa-1b) and poorly crystallized (KGa-2) kaolinite standards. Atomic force microscopy (AFM) revealed that particles of KGa-1b generally have nicely hexagonal micromorphology and crystallographically controlled microtopographic features. Particles of KGa-2 are also hexagonal, but their micromorphology tends to be more rounded. Basal-plane surfaces tend to be more irregular with fewer clearly crystallographically controlled features. KGa-1b particles tend to be larger in diameter and thicker than KGa-2 particles. Micromorphologic measurements showed that both KGa-1b and KGa-2 have modal edge- to total surface-area ratios of approximately 0.1 (mean ~ 0.2), although these measurements did not include the potentially large contribution of basal-plane step edges (additional 20% or more).

Dissolution experiments were conducted in oxalic acid and inorganic acids at pH 3, 22 °C, $I = 0.01$ M, under batch dissolution conditions. Dissolution rates (measured as Si release) in 1 mM oxalic acid were approximately twice as fast for KGa-2 as for KGa-1b (2.27 vs. 0.96 nmol/m²·h). Rates for KGa-2 and KGa-1b were similar in HNO₃ (0.86 and 1.16 nmol/m²·h, respectively). The comparable rates for these two sedimentary kaolinites and for a hydrothermal kaolinite studied by Wieland and Stumm (1992) suggests that the fundamental structure of kaolinite, rather than specific surface details, exerts the greatest influence on dissolution kinetics.

High-performance cation exchange chromatography (HP-CEC) was used to determine the distributions of monomeric Al species over the course of kaolinite dissolution. For dissolution in 1 mM oxalate, Al-oxalates were observed almost exclusively in agreement with results of equilibrium speciation calculations. For dissolution in HNO₃, the peak representing uncomplexed Al species, Al₁, was predominant but not exclusive, as predicted by calculations. A peak having a retention time characteristic of species with +2 charge may be evidence for an AlOSi(OH)₃²⁺ species, and warrant further investigation.