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Mechanism and kinetics of smectite dissolution

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Dissolution process of stevensite particles was observed in acidic aqueous solution with atomic force microscope (AFM) and attenuated total reflection infrared (ATR-IR) spectrometer. In-situ observations with AFM were carried out to evaluate the reactive surface area of individual particles and to measure the dissolution rate under the acidic condition. In-situ observations with ATR-IR were carried out to reveal the mechanism of stevensite dissolution.

During dissolution experiment, the stevensite particles dissolved exclusively via the retreat of the edge surfaces, and basal surface area of the particles continuously decreased during the observation. In contrast, no change of the basal surface was observed within the dissolution experiment. Therefore, the observations were indicated that only edge surface contributed as reactive surface of the stevensite dissolution. The change in cross section profile of stevensite particle with reaction time indicated that dissolution of stevensite particle was apparently congruent. Solubility of octahedral sheet in stevensite is thought to be higher than that of tetrahedral sheet at acidic condition. However, the intensity of Si-O band (930-1050cm-1) in the IR spectra decreased without any change in shape and peak shift as a function of reaction time. Therefore, the dissolution of stevensite presumably progress by quick breakdown of octahedral sheet and subsequent breakdown of tetrahedral sheet.

The dissolution rate was calculated from the change in basal surface area during reaction time. The rate was normalized to reactive surface area and total surface area of individual particles obtained by AFM observations. From the reactive edge surface area determined by AFM, the dissolution rate under pH1.1 solution at 22C was calculated to 1.00 * 10-9 to 2.54 * 10-9mol/m2/sec (average value 1.85 * 10-9 mol/m2/sec). In contrast, from the total surface area determined by BET, the dissolution rate was ranged from 1.88 * 10-11 to 4.07 * 10-11 mol/m2/sec. (average value 3.14 * 10-11 mol/m2/sec). These in situ observations will help us to characterize the reactive surface area term in experimentally determined rate expressions more precisely.