

Study on reactive surface area of smectite dissolution and its quantitative evaluation

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In previous studies about mineral dissolution, BET surface area has been generally measured to calculate a rate of the dissolution. A great number of dissolution rates have therefore been reported by using BET surface. However, BET surface area of smectite is strictly not consistent with reactive surface area of smectite because smectite is normally exfoliated into single plate in reactive solution for its extreme expandability. Recently, from Atomic Force Microscopy (AFM) observation, it is reported that smectite particles are dominantly dissolved from edge surface (Bosbach et al., 2000). Therefore, it is necessary to estimate the reactive surface for assessment of smectite dissolution rate and for considering the dissolution rate of smectite in compacted bentonite established at radioactive waste disposal. In this context, we intend to quantify the reactive surface area by various methods and understand the reactive surface area for the dissolution of smectite in this study. Starting materials with different BET surface areas were prepared by grinding of Kunipia-P (from Tsukinuno, Yamagata) for 3, 6, 15 and 24hrs. Stirred-flow-through dissolution experiments were carried out to measure the smectite dissolution rates under highly alkaline conditions ($\text{pH} = 13.3$). In situ (fluid-cell) AFM were used to investigate the total surface area (TSA) and edge surface area (ESA). For characterization of the starting materials, X-ray diffractometry (XRD), scanning electron microscopy (SEM), infrared spectroscopy (IR) and Cs adsorption test were also conducted. The BET surface areas increase from 4.3 to 109 m^2/g (about 25 times higher than that of the starting material) as function of grinding time. Si concentrations obtained from the dissolution experiments also increase as function of grinding time. However, the concentration registers a modest 4.5 times increase from 8.0 to 46.1 μM . Calculated rates of the dissolution based on their own BET surface area are different even though using the same starting material with different grinding time. On the other hand, TSA of samples obtained by in situ AFM observation is constant for all samples with different BET surface area. Therefore, if we calculate the dissolution rates based on TSA obtained by AFM, the rate is considered to be increased as function of grinding time. However, ESA obtained by AFM, considered being dominant reactive surface in the previous study, increases as function of grinding time, and the increasing rate in ESA is correlated to that in Si concentration obtained from the dissolution experiment. Consequently, it is necessary to obtain edge surface area of smectite at the same condition with that of dissolution experiment when we accurately calculate the rate of smectite dissolution.