Atomic-Resolution Imaging of Calcite Dissolution Processes by High-Speed Atomic Force Microscopy

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Calcium carbonates play critical roles in the global carbon cycle and thereby influence the Earth's aerial, aquatic and geologic environments. Calcite is among the most abundant and reactive mineral and hence its crystal growth and dissolution are important. In particular, the calcite dissolution has attracted growing interests due to the recent efforts in geologic carbon sequestration: the storage of carbon dioxide into the subsurface, as it may form leakage paths for the stored carbon. To accurately simulate such long-term and large-scale carbon cycles, it is important to understand the mechanism and kinetics of the calcite dissolution processes.

So far, calcite dissolution processes have been investigated by various methods. The macroscopic behavior has been investigated by monitoring changes in the ion concentration in the solution during the dissolution processes. The nanoscale behavior has been studied by imaging real-time and real-space movements of the single atomic steps by atomic force microscopy (AFM) or optical measurement techniques. However, the establishment of the atomic-scale crystal dissolution model requires understanding of the atomic-scale behavior near the step edges. However, conventional measurement techniques do not allow to directly image such behavior with sufficient spatial and temporal resolution.

Frequency modulation atomic force microscopy (FM-AFM) has traditionally been used for atomic- or molecular-scale investigations on the surface structures and properties of various materials in ultrahigh vacuum. In 2005, we enabled to operate FM-AFM in liquid with true atomic resolution. Furthermore, we recently improved its imaging speed from ~1 frame/min to ~1 frame/sec without losing its high spatial resolution. With the developed high-speed FM-AFM, we succeeded in imaging calcite dissolution processes with atomic-scale resolution. The results reveal the existence of a transition region with a width of 1-8 nm near the step edges. Although the origin of the transition region is still under investigation, this finding should greatly improve our understanding on the atomic-scale behavior of calcite crystal dissolution processes. The results opens up broad range of future applications of the high-speed FM-AFM on crystal growth and dissolution processes.

Keywords: Atomic Force Microscopy, Calcite, Crystal Dissolution

