

New Evidence of Orthoclase Dissolution Mechanisms by In Situ X-Ray Reflectivity and Atomic Force Microscopy

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Feldspar minerals are abundant in the Earth's crust. The reaction of feldspars with aqueous solutions produces clay minerals and has a strong influence on the chemistry of natural waters. The dissolution of feldspar is incongruent under some conditions and the dissolution mechanism apparently depends on both solution and feldspar compositions. An altered layer (0.1 to 1000 nm thick) may form at the feldspar-water interface during dissolution. Structural and compositional characterization of this altered layer at the feldspar-water interface is critical for understanding mechanisms of feldspar dissolution and clay mineral formation. We have begun to apply in situ X-ray specular reflectivity and atomic force microscopy (AFM) measurements to study structure and dissolution of the orthoclase (001)-water interface at 25 to 60 °C (Fenter et al., 2000). These studies of gem-quality orthoclase indicate that the orthoclase (001) cleavage surface has minimal roughness and exposes a single cleavage plane. The X-ray data for the deionized water-orthoclase interface show that (1) only one of the two possible surface terminations is exposed, (2) dangling Si and Al bonds are hydroxylated, and (3) the outermost layer of K⁺ ions is absent. A complex relaxation profile affects the near-surface structure and propagates ~2.6 nm into the orthoclase. Dissolution kinetics

were measured in situ by X-ray reflectivity at pH 1 and 13 between 48 and 60 °C. Layer by layer dissolution was observed at both pH 1 and pH 13. At pH 1, an altered layer about unit cell thick or less was observed by X-ray reflectivity, whereas at pH 13 there was no evidence for such residual altered layer. AFM images made at 25 °C show that the mechanism of dissolution at pH 1 involves mainly etching of random sites on atomically flat terraces, whereas at pH 13 the mechanism involves mainly dissolution at step edges. Residual material at pH 1 is easily moved by the AFM tip in contact mode and also by an increase in the flow rate of fluid in the AFM cell.

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