

Dissolution kinetics of anorthite in a supercritical CO₂–water system

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ABSTRACT

The Gibbs free energy change, ΔG_r , dependence of the anorthite dissolution rate in a supercritical CO₂–water system was measured as part of a geochemical assessment of CO₂ geological sequestration. Being bounded at ΔG_r^{crit} , the critical ΔG_r required for an opening of the etch pit at a screw dislocation, it has been accepted that mineral dissolution follows etch pit formation assisted by dislocations below ΔG_r^{crit} , whereas the horizontal step retreats without the etch pit above ΔG_r^{crit} . The experiment described herein, however, revealed that another mode of dissolution occurs more distant from equilibrium by spontaneous formation of the etch pit over the entire surface, as observed on calcite. The dissolution rate is higher by more than one order of magnitude than that in the dislocation-assisted mode. Therefore, including the rate gap at ΔG_r^{crit} , a nonlinear curve with three steps instead of a sigmoidal curve is proposed for the ΔG_r dependence of the anorthite dissolution rate. Extremely slow rates were observed depending on observed points for the same ΔG_r condition. Although the reason for such a rate difference remains unknown, it is likely related to the defect density on the crystal surface. It is possible that initial spreading of the dissolved surface attributable to the etch pit formation assisted by defects provides some trigger for subsequent explosive etch pit formation. These findings suggest that the initial transient process can strongly influence the kinetics of geochemical reactions that occur during CO₂ geological sequestration.

Keywords: CO₂ geological sequestration, phase-shift interferometry, dissolution rate, Gibbs free energy, anorthite, feldspar