Dissolution kinetics of feldspar on CO2 geological sequestration

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We measured the deltaG_r dependence of the anorthite dissolution rate under a supercritical CO2-water system using phase-shift interferometry as a part of the geochemical assessment of CO2 geological sequestration (CGS). Being bounded at delta$G_r{}^{crit}$, it has been accepted that mineral dissolution follows the etch pit formation assisted by dislocations below delta$G_r{}^{crit}$, whereas the horizontal step retreats without an etch pit above delta$G_r{}^{crit}$. The present experiment, however, revealed another dissolution mode in the dislocation-assisted region: much higher dissolution occurs by explosive formation of etch pits over the entire surface. This mode probably corresponds to the two-dimensional spontaneous nucleation of etch pits observed on calcite. Its rate is higher than that in dislocation-assisted mode by more than one order. Therefore, including the rate gap at delta$G_r{}^{crit}$, a nonlinear curve with three steps, instead of a sigmoidal curve, is proposed for the deltaG_r dependence of the anorthite dissolution rate.

In this study, we observed the surface showing a slight change during one month's reaction, in addition to the existence of some induction period before etch pit formation. Although the reason for such a difference of rate depending on observed points remains uncertain, the defect density on the crystal surface likely suggests an explanation: it is possible that initial spreading of the dissolved surface because of the etch pit formation assisted by defects might provide a trigger for subsequent explosive etch pit formation.

These findings provide important implications for geochemical assessments of CGS. In particular, CO2 injection causes the dissolution of feldspar in acidified formation water, but its rate decreases with time because leached components raise the degree of saturation of the formation water. Therefore, the rate function form of the saturation dependence is important for accuracy enhancement of the timescales of various geochemical processes on CGS. Hereafter, we should examine the effects of temperature, pH, and other dissolved species on the saturation dependence of the dissolution rate. This study highlighted the importance of heterogeneity of mineral surfaces. Natural crystal shows various defect densities, and apparently never has a fresh surface. For that reason, it is a challenge for the future to elucidate what mechanism works to reach a steady state dissolution process.