

Saturation dependence of feldspar dissolution rate in supercritical CO₂-water system

Masao Sorai[1]; Munetake Sasaki[2]; Yasuko Okuyama[3]; Toshiyuki Toshi[1]

[1] AIST; [2] AIST, GeoResour. Dep.; [3] Geological Survey of Japan, AIST

<http://unit.aist.go.jp/georesenv/>

Carbon dioxide underground sequestration to aquifer formations has been proposed recently as a global warming countermeasure. However, a true understanding of the true potential of geological sequestration including the ultimate fate of the injected CO₂ demands consideration of various geochemical processes with much longer timescales (from several to several tens of millennia). Especially, if we restrict assessments to a more immediate timescale (ca. 1,000 years), the verification of initial dissolution processes of reservoir minerals is essential. Among these, the dissolution of feldspar is most important because feldspar, which has high reactivity, is the most abundant mineral in the upper crust. Regarding this, we have devoted attention to anorthite as a Ca-rich end member of feldspar, and have sought to determine its dissolution rate based on observations of crystal surface using phase-shift interferometer (PSI). Especially in this study, the saturation dependence of anorthite dissolution rate was examined for the data accumulation toward a more realistic safety assessment.

A series of batch-type dissolution experiments were done at 50°C in solution with various undersaturations (25 ml) using a CO₂-water-mineral reaction system (SCF-Get, SCF-ROV; Jasco Inc.). A constant CO₂ pressure of 10 MPa was maintained for one month. The solution was stirred continuously during the experiments using a magnetic stirring system. We used cleaved anorthite crystals from Miyake-jima, Japan as feldspar samples. After being washed ultrasonically in acetone, these samples were glued onto an 8*32 mm stainless steel plate. This plate was fixed on the sample holder in a stainless reaction vessel (50 ml). A part of the sample surface was covered with a Pt-Pd thin film by sputtering for an inert reference area. Before and after the reaction, several regions on the sample crystal surface were observed using PSI and a laser microscope. These measurements were performed sequentially for several specific points of the sample surface. The sample was taken out after 24, 72, 168, 336, 504, and 672 h and observed in air after washing with pure water, then dried at room temperature.

We found that anorthite dissolution rate changed significantly up to one order between near equilibrium point and far from equilibrium. Our results shows that the saturation dependence of anorthite dissolution rate follows to a sigmoidal curve function rather than a linear function. Such a difference of the function form of dissolution rate is important because the change of saturation state in formation water caused by a CO₂-water-rock interaction is expected to be large. Therefore the sensitivity of the dissolution rate change was checked using a numerical simulation code to analyze the geochemical processes caused by CO₂ geological sequestration.

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