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Dissolution and reaction in a CO₂-brine-clay mineral particle system under geological CO₂ sequestration from subcritical to supercritical conditions

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ABSTRACT

To support effective geological CO₂ sequestration design and operation, dissolution and reaction in CO₂-brine-clay mineral particle systems (sepiolite and montmorillonite) were studied under subcritical to supercritical CO₂ conditions (10 bar to 150 bar at 45 $^\circ\,$ C and 65 $^{\circ}$ C). The order of ion dissolution from the framework of sepiolite in the brine was slightly different under each experimental condition, whereas the order of dissolved ion concentration from the montmorillonite was not varied. The solubility of CO₂ was lower in the CO₂-brine-clay mineral particle system than in a CO₂-brine system. Precipitation of amorphous silica as a secondary mineral formation was observed after the reaction of both sepiolite and montmorillonite. The CO₂ solubility model, considering ion concentration and aqueous silica, reasonably predicted the CO₂ solubility from subcritical to supercritical conditions. The kinetic rate constant of the dissolution reaction of sepiolite was correlated with the initial pH of the brine. After reaction with high-pressure CO_2 -saturated brine, the micro-crystallinity of sepiolite did not change, whereas the basal (001) plane of montmorillonite showed deformation in micro-crystallinity after the dissolution reaction. By contrast, reaction with the CO₂-saturated brine led to a decrease in the surface area of sepiolite and an increase in the surface area of montmorillonite.

Key words: clay mineral; carbon dioxide; dissolution; solubility trapping; supercritical condition

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