Sorption behavior and geochemical reaction from subcritical to supercritical conditions in CO_2-clay minerals and CO_2-brine-clay minerals systems

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When CO_2 is injected into geological formation for geological sequestration, the upper plum of the injected CO_2 directly contacts with caprock, and the lower part of the CO_2 plume dissolves into brine. Since the sorption and reaction of CO_2 are different for each environment, the CO_2 sorption on dry clay minerals (llite, sepiolite, and montmorillonite) and the dissolution-induced reaction in a CO_2-brine-clay minerals system (sepiolite and montmorillonite) were studied under subcritical to supercritical CO_2 conditions.

The sorption rates of CO_2 on clay minerals were independent of pressure, showing variation within a single order of magnitude. Based on the excess sorption isotherms, the CO_2 sorption amount on clay minerals reached a maximum near the critical pressure. And the absolute sorption isotherms moved towards saturation over the critical density of CO_2. The desorption isotherm of montmorillonite clearly showed a positive hysteresis. The significant hysteresis implied a security for the sorbed CO_2 on clay minerals even though the pressure in sequestration site was decreased by the injected CO_2 diffusion along the strata.

In the brine-containing system, the rate constant of sepiolite dissolution increased with lower initial pH of the brine. Preference of ion dissolution from the framework of sepiolite changed slightly at various experimental conditions, whereas montmorillonite showed preferred dissolution of the edge plane to the basal plane under all conditions. Due to the increased ion concentration, the solubility of CO_2 became lower in the CO_2-brine-clay mineral system than in the corresponding CO_2-brine system. The effect of aqueous silica on CO_2 solubility should also be considered for further understanding of reduced CO_2 solubility.

To study the structural change of clay minerals after being exposed to high pressure CO_2, their physical properties were compared to those of original clay minerals. Irreversible deformation of pore structure was different, depending on the exposed CO_2 experimental conditions.