

Field experiment of carbonate reactions in the CO₂-injected hot spring waters*Masao Sorai¹, Munetake Sasaki¹

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On geological CO₂ storage (GCS), the evaluation of the injected CO₂ behavior over a long time requires the numerical simulation considering geochemical processes. Among such geochemical processes, both precipitation and dissolution of carbonate minerals are important from each perspective of storage potentials and leakage risks. However, there have remained many uncertainties on their kinetics under conditions of GCS.

In this regard, to obtain reliable dataset on carbonate kinetics, we have performed reaction experiments of carbonate minerals at bicarbonated springs, which can be regarded as a natural analogue of GCS. Specifically, an input of CO₂ gas and Mg ions into spring waters allowed to highlight the effects of these chemical species on the reaction rate and precipitating phase of carbonate minerals under natural conditions. Previous experiment injected CO₂ at an atmospheric pressure. Under this condition, the solubility of CO₂ was too low (i.e., the pH was too high) to dissolve carbonate minerals. This time, we tried to change the pH lower by injecting high pressure CO₂ using a stainless-steel tube.

The experiment was performed at the Utoro hot spring in Hokkaido, Japan. In this site, the spring water from a well was stored once in a tank, and then it was flew down through a pipe 50 meters long. We created the bypass system, where the spring water pumped up from the tank was flowed through a stainless-steel tube. After passing through the tube, the water was thrown down the existing pipe. Three sample holders were arranged in series within a tube; each holder set cleaved crystals of major carbonate species, calcite and aragonite (CaCO₃), dolomite (CaMg(CO₃)₂), and magnesite (MgCO₃). These samples were dipped into the flowing water over a period up to 24 hours. Then, the holder was taken out one by one at predetermined time. Present experiments included reactions injecting the CO₂ gas at 0.3 MPa with and without addition of magnesium chloride (i.e., Mg/Ca = 0.5, and 3, respectively), along with reactions in the untouched spring water.

Reaction rate was estimated from measurements of the height level difference between original and reacted surfaces by using a phase-shift interferometer and a laser microscope. The CO₂ injection tests resulted in the dissolution of calcite and aragonite. Although both dissolution rates were almost equal, addition of Mg ions induced an opposite trend: calcite dissolution rate was reduced by nearly half, whereas aragonite dissolution rate was slightly increased. On the other hand, the optical microscopic observation showed no obvious changes on dolomite and magnesite surfaces. These results suggest that the dissolution rate of carbonate minerals depends on both water compositions near a CO₂ injection well and mineral species of carbonates, and that the kinetics for carbonate reactions associated with GCS cannot be uniformly defined.

Keywords: Geological CO₂ storage, Bicarbonated spring, Dissolution rate, Reaction kinetics, Carbonate minerals