

Carbonate reaction experiments in hot springs as a natural analogue field of CO₂ geological sequestration

SORAI, Masao^{1*} ; SASAKI, Munetake¹

¹National Institute of Advanced Industrial Science and Technology

Carbon dioxide geological sequestration (CGS) into aquifer formations involves various geochemical processes starting from dissolution of injected CO₂ in underground waters. Among such processes, the reaction of carbonates is most important from both perspectives of storage potentials and leakage risks. However, there have remained many uncertainties on its kinetics under conditions of CGS. On this subject, we applied the approach to measure reaction rates in a condition cloth to CGS. The approach has two important concepts. First, we focus on carbonated or bicarbonated springs as a reaction field because they are exactly regarded as a natural analogue of CGS. Another point is that seed crystals are introduced into spring waters artificially, thereby allowing us to get highly reliable reaction rates in a short time.

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. The CO₂ is degassed during the drainage, which causes the degree of supersaturation with respect to carbonates to be progressively higher downstream. We set the tank interior, and the middle and lower end of a pipe, as observation points. For each point, cleaved samples of major carbonate species, calcite and aragonite (CaCO₃), dolomite (CaMg(CO₃)₂), and magnesite (MgCO₃), were dipped into the water over a period up to 24 hours. Then, the sample was taken out one by one at predetermined time. Present experiments included additional reactions adding respectively the CO₂ gas and magnesium chloride to the spring water within a tank, 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 result showed that the reaction rates of carbonates change sensitively to the change of the water composition. Specifically, the calcite reaction rate in nature was much lower than that calculated from the empirical equation, which was compiled based on laboratory experiments. This is possibly because the current equation includes no inhibitor effect of dissolved ions (mainly magnesium ions) and uses unrealistic function form with respect to the saturation-state dependency. We also found that although dolomite generally indicated the highest degree of supersaturation, its growth rate was significantly lower than that of calcite and aragonite. This suggests that judging the possibility of any carbonate precipitation only from its degree of supersaturation would mislead a prediction of long-term behavior of injected CO₂.

This research is funded and supported by Ministry of Economy, Trade and Industry (METI).

Keywords: CO₂ geological sequestration, natural analogue, carbonate, reaction rate, mineral trapping, hot spring