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Modeling a CO2 mineralization experiment of fractured peridotite from the Semail ophiolite/ Oman

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Most geologic CO2 sequestration technologies focus on sedimentary rocks, where the carbon dioxide is stored in a fluid phase. A possible alternative is to trap it as a mineral in the subsurface (in-situ) in basaltic or even (ultra)mafic rocks.

Carbon dioxide in aqueous solution reacts with Mg-, Ca-, and Fe-bearing silicate minerals, precipitates as (MgCa,Fe)CO3 (carbonate), and can thus be permanently sequestered. The cation donors are silicate minerals such as olivine and pyroxene which are abundant in (ultra)mafic rocks, such as peridotite. Investigations are underway to evaluate the sequestration potential of the Semail Ophiolite in Oman, utilizing the large volumes of partially serpentinized peridotite that are present. Key factors are the rate of mineralization due to dissolution of the peridotite and precipitation of carbonate, the extent of the natural and hydraulic fracture network and the accessibility of the rock to reactive fluids.

To quantify the influence of dissolution rates on the overall CO2 mineralization process, small, fractured peridotite samples were exposed to supercritical CO2 and water in laboratory experiments. The samples are cored from a large rock sample in the dimension of small cylinders with 1 cm in height and diameter, with a mass of ~2g. Several experimental conditions were tested with different equipment, from large volume autoclave to small volume cold seal vessel. The 650 ml autoclave contained 400-500g of water and a sample under 10 MPa of partial CO2 pressure up to 150. The small capsules in the cold seal vessel held 1-1.5g of water and the sample under CO2 partial pressure from 15MPa to 70 MPa and temperature from 60 to 200°C. The samples remained for two weeks in the reaction vessels. In addition, bench acid bath experiments in 150 ml vials were performed open to the atmosphere at 50-80°C and pH of ~3.

The main observation was that the peridotite dissolved two orders of magnitude slower in the high pressure and temperature cell of the cold seal vessel than comparative experiments in large volume autoclaves and bench acid bath vials under lower and atmospheric pressure conditions. We attributed this observation to the limited water availability in the cold seal vessel, limiting the aqueous reaction of bi-carbonate formation and magnesite precipitation. To test this hypothesis, one of the cold seal vessel experiments at 20 MPa and 100°C was simulated with a reactive transport model, using TOUGHREACT.

To simulate the actual experimental conditions, the model used a grid on mm and 100's of µm scale and a fractured peridotite medium with serpentine filling the fractures. The simulation produced dissolution comparable to the experiment and showed an effective shut down of the bi-carbonation reaction within one day after the start of the experiment. If the conditions of limited water supply seen in our experiments are applicable in a field setting, we could expect dissolution may be limited by the buffering of the pH and shut down of the bi-carbonate formation. Under field conditions water and CO2 will only flow in hydraulic induced fractures and the natural fracture network that is filled with serpentine and some carbonate. The simulation result and potential implication for the field application will require further experimental investigation in the lab or field in the future.