

FIELD EXPERIMENTS FOR STUDYING ON CO₂ SEQUESTRATION IN SOLID MINERALS AT THE OGACHI HDR GEOTHERMAL SITE, JAPAN

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ABSTRACT

We have conducted some field experiments to study CO₂ sequestration in solid minerals by injecting CO₂ dissolved water into a high temperature borehole, OGC-2, at the Ogachi Hot Dry Rock geothermal site in Japan. OGC-2 was drilled into granitic rock to a depth of 1,100 m and the bottom-hole temperature was measured around 210 degree C.

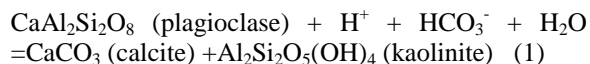
Ca concentration of the water sampled at 1,030 m depth in OGC-2 increased to a maximum of 85.2 mg/L in a few hours after CO₂ dissolved water (3 weight %) injection. Then the Ca concentration decreased in a few days. The Ca concentration increase was considered that Ca was supplied from Ca-feldspar in granitic rock by chemical reaction of the injected CO₂ dissolved water with the rock.

Calcite precipitation on calcite crystals was observed when the crystal samples were suspended in CO₂ dissolved water at 850 m in OGC-2. Calcite precipitation rate on calcite crystals was measured a maximum of 1 micro meters per an hour in CO₂ dissolved water at a temperature of around 210 degree C. This supports the view that injected CO₂ might be fixed as carbonate minerals.

INTRODUCTION

For the global warming problems, it is considered to reduce CO₂ emission into atmosphere by injecting CO₂ into underground. In Japan, there are many volcanoes and many geothermal areas. If CO₂ is injected into underground at high temperature condition, then the reaction of CO₂ dissolved water with rocks will be faster than normal temperature area and CO₂ can be stored in carbonate minerals in relatively short time.

Under high temperature condition, the following reaction can proceed from the upper to the lower (*Gale and Shane, 1905*).



Carbonate-rich rock formations can be observed in most Japanese geothermal fields. In the Sumikawa field in Akita prefecture, Japan, for example, CO₂-rich ground waters are thought to have reacted with reservoir rocks according to reaction (1), to form a carbonate and kaolinite alteration assemblage. An isotopic investigation of calcites at Sumikawa indicated that the reaction occurred at 100 to 250 °C by interaction of reservoir rocks with meteoric waters (*Ueda et al., 2001*).

We conducted some field experiments at Ogachi Hot Dry Rock geothermal site to study chemical reaction between CO₂ dissolved water and granitic rock and creation of carbonate minerals by injecting CO₂ dissolved water into a high temperature borehole.

CONCEPT OF CO₂ MINERAL TRAP AT HIGH TEMPERATURE

Reaction according to formula (1) moves towards the right side at higher temperature, reflecting the fact that calcite solubility decreases with increasing temperature. The calcite and kaolinite (clay)-rich rock is produced through the reaction and forms a cap rock for the geothermal reservoir. These considerations, together with the increasing reaction rates at high temperature condition and the fact that precipitation of carbonate minerals fixes CO₂ suggest that CO₂ sequestration could be practicable by injection into geothermal fields (see Figure 1).

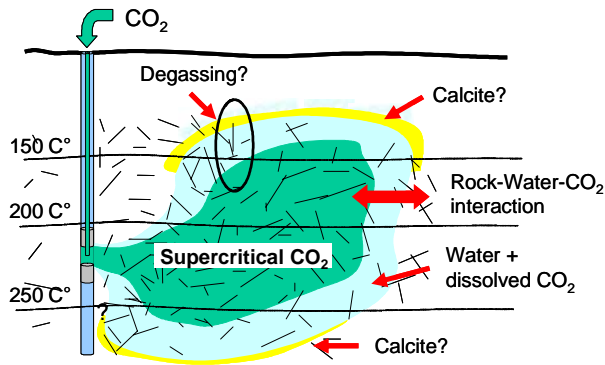


Figure 1: Concept of CO₂ injection into high temperature rock

OGACHI TEST SITE

Geology

The Ogachi site is located in northern Japan. There is a small hot spa, Akinomiya-onsen, 2 km away from the site. Many geothermal wells were drilled around the site for geothermal promotion survey projects. One geothermal plant, Uenotai geothermal power generation plant, is located about 10 km from the site.

The Ogachi site is situated inside a Neogen caldera which was formed during 6-2 Ma due to an intensive felsic volcanic activity. The bedrock below 300 m from the surface at the site consists of pre-Tertiary granitic rocks and mylonite, which underlines Neogene and Quaternary felsic volcanic rocks.

Well bores

At the Ogachi site there are three 1,000 m class wells (OGC-1, 2, 3) which were drilled into granitic rocks from around 300 m below the ground surface. These wells were used for basic experiments for Hot Dry Rock geothermal energy development. The temperature of the wells at 1,000 m depth was measured at around 200 degree C. In one of these experiments, river water at temperature of about 15 degree C was injected into OGC-1 and recovered form OGC-2 at a temperature of 165 degree C (Kaieda et al., 2005).

OGC-2 was used for this research. OGC-2 was cased with full-hole cementing from the ground surface to a depth of 700 m and below 700 m to the bottom of the well of 1,100 m was left uncased (open-hole). The bottom-hole diameter is 98 mm (see Figure 2). Water level in OGC-2 is 173 m below the ground surface. OGC-2 accepts water at flow rate of around 100 kg/min at atmospheric pressure. The volume of the cased region is about 5 m³ and 3 m³ for the open-hole region in OGC-2.

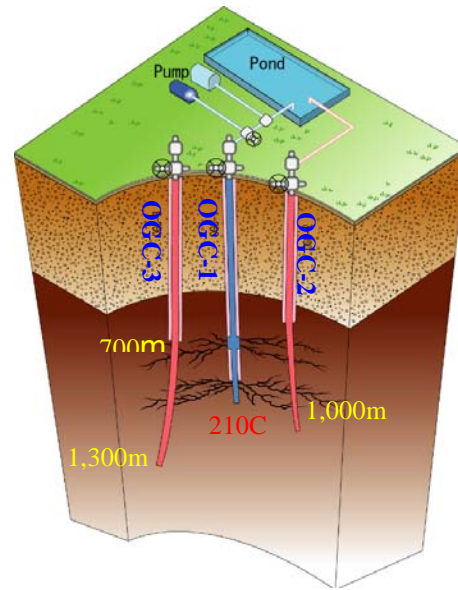


Figure 2: Wellbores at Ogachi HDR site

CHEMICAL REACTION BETWEEN CO₂ DISSOLVED WATER AND GRANITIC ROCK

Water injection

The river water of pH 4.7 was neutralized with NaOH (hereafter river water) and injected into OGC-2 at flow rates of 80-100 kg/min. Total 19.6 tons of the river water was injected. After the river water replaced pre-existence water in OGC-2, water samples in OGC-2 was recovered at a depth of 1,030 m in OGC-2 for 12 days by using a water sampler.

The results of chemical components of the sampled water with time after the water injection are shown in Figure 3. From this figure, we can see that Ca concentration increased quickly to a maximum of 7 mg/L at 22 hours after the river water injection.

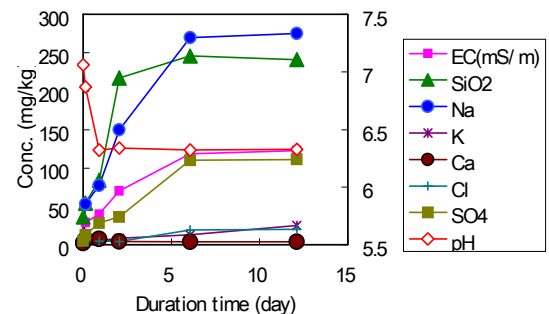


Figure 3: Chemical component change with time after the river water injection

CO₂ dissolved (1wt%) water injection

The neutralized river water was injected into OGC-2 at a flow rate of 50 kg/min. During the water injection solid CO₂ (dry ice) blocks in some centimeter size were injected at a rate of 0.5 kg/min to create 1 weight percent CO₂ dissolved water (hereafter 1 % CO₂ water). (see Figure 4) After a total of 8.6 tons of 1 % CO₂ water was injected, a total of 4 tons of the neutralized water was injected to push down the 1 % CO₂ water to open-hole region in OGC-2 and into surrounding rocks (see Figure 5).

After the 1 % CO₂ water replaced the pre-existed water, water samples at 1,030 m depth in OGC-2 were sampled for 12 days.

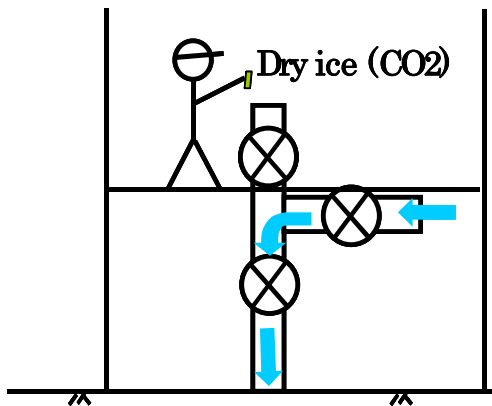


Figure 4: Procedure of CO₂ dissolved water injection into OGC-2

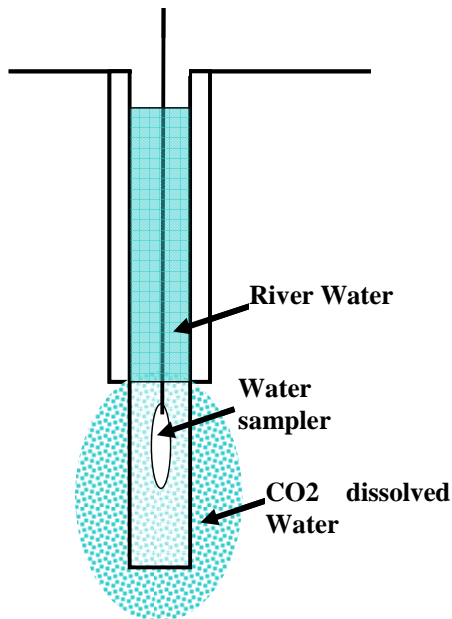


Figure 5: Image in OGC-2 after CO₂ dissolved water injection

The results of chemical components of the sampled water are shown in Figure 6. From this figure, we can see that Ca concentration increased quickly to a maximum of 21 mg/L at 17 hours after 1 % CO₂ water injection.

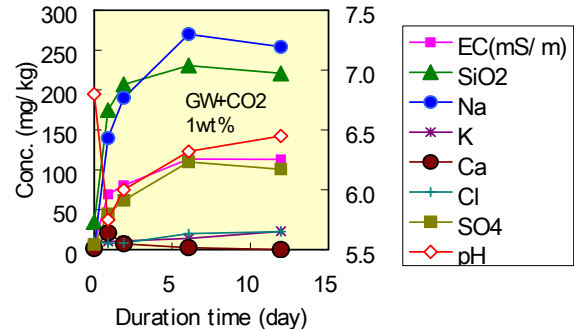


Figure 6: Chemical component change with time after 1 % CO₂ water injection

CO₂ dissolved (3wt%) water injection

The neutralized river water and dry ice were injected into OGC-2 at flow rates of 50 kg/min and 1.5 kg/min to create 3 weight percent CO₂ dissolved water (hereafter 3 % CO₂ water). A total of 8.6 tons of 3 % CO₂ water was injected and a total of 4 tons of river water was injected.

After the 3 % CO₂ water replaced the pre-existed water, water samples at 1,030 m depth in OGC-2 were sampled for 8 days.

The results of chemical components of the water are shown in Figure 7. Ca concentration increased to the maximum of 85.2 mg/L at 1 hour after 3 % CO₂ water injection.

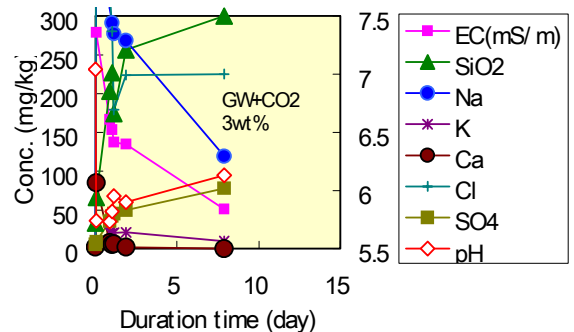


Figure 7: Chemical component change with time after 3 % CO₂ water injection

From the results of chemical components change of the sampled water after the neutralized river water and CO₂ dissolved water injections, it was found that Ca concentration of the sampled water increased quickly in some hours. The peak values of Ca concentration of the sampled water were 7 mg/L for the river water, 21 mg/L for 1 % CO₂ water and 85.2mg/L for 3 % CO₂ water. After the increase of Ca concentration of the sampled water Ca concentration decreased in some days.

The Ca concentration decrease may be caused by water flow in and/or out between OGC-2 and surrounding rock, or by precipitation as CaCO₃. We considered that Ca was supplied from Ca-feldspar in granitic rocks.

CALCITE PRECIPITATION TEST IN CO₂ DISSOLVED WATER AT HIGH TEMPERATURE

Procedure of calcite crystal grow test

In this experiment we intended to study the possibility for CO₂ sequestration as calcite. As previous experiments, 1 % CO₂ water was created by the neutralized river water and dry ice and injected into OGC-2. A total of 14 tons of 1 % CO₂ water was injected and a total of 4 tons of river water was injected (see Figure 4 and 5).

After the 1 % CO₂ water replaced pre-existed water, a specially created test sonde was inserted into OGC-2 and calcite crystal grow tests were conducted. Calcite crystals partially covered with Au film were held in the sonde. The sonde was filled with He gas at an adequate pressure. A valve of the sonde opens depend on pressure difference between in and out of the sonde. In this test we set the valve to open at a pressure of 850 m depth in OGC-2. The sonde was put into OGC-2 to a depth of 850 m and held there for an hour. During this hold time CO₂ dissolved water come into the sonde and reacted with calcite crystals. The sonde was recovered to the surface after one hour and the CO₂ dissolved water removed from the sonde. On the surface calcite crystals in the sonde was removed and roughness of the calcite crystal surface was observed by the phase shift interferometer comparing with Au film covered area and not covered area.

Calcite crystal test results

Figure 8 shows an example of a picture of the calcite crystal surface by a stereo microscope (upper picture) and roughness of the crystal surface along the red line measured by the phase shift interferometer (lower figure). The 'masked surface' means the area covered with Au film. In this masked area, crystal surface

roughness is relatively smooth, but other area shown as 'reacted area' calcite crystal grew to a maximum height of about 1,100 nano-meters (1.1 micro-meter).

This result was observed when the crystal test sonde was set 23 hours after 1 % CO₂ water injected into OGC-2. Calcite crystal grew to a maximum of almost 1 micro meters in an hour. However, a few hours after CO₂ dissolved water injection, calcite crystal was solved. Figure 9 shows another example of a picture of the calcite crystal surface. In this stereo microscope picture (upper) we can see a special feature of depression (etch pit).

From these results we considered as follows, 1) water condition in the open-hole region in OGC-2 for a few hours after CO₂ dissolved water injection was high CO₂ content and low pH, in this condition calcite solved, 2) some hours after CO₂ dissolved water injection CO₂ concentration decreased and pH increased to near neutral, in this condition calcite grew quickly, 3) after most of Ca and CO₂ precipitated as calcite, calcite grow rate decreased.

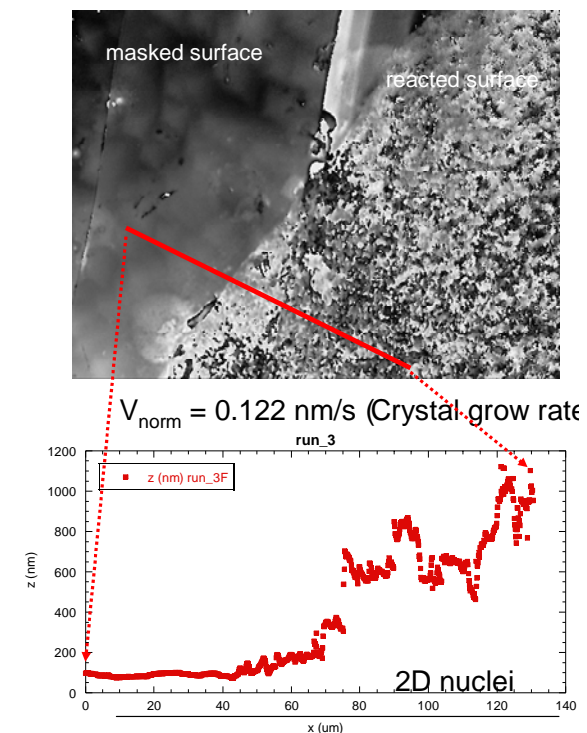
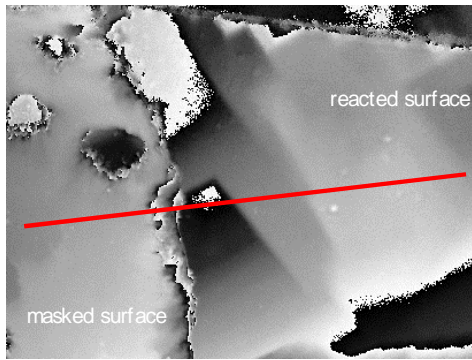


Figure 8: An example of stereo microscope picture of calcite crystal grew surface (upper) and roughness measured by phase shift interferometer (lower)



$$V_{\text{norm}} = -0.174 \text{ nm/s}$$

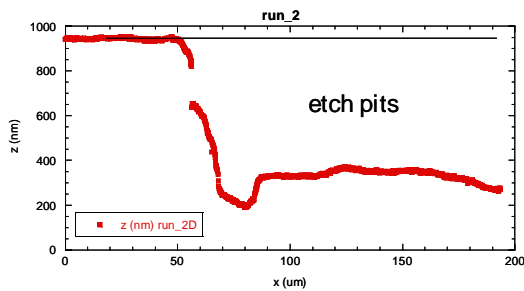


Figure 9: An example of stereo microscope picture of calcite crystal dissolved surface (upper) and roughness measured by phase shift interferometer (lower)

CONCLUSION

CO₂ dissolved water was injected into an open-hole interval between 700 m to 1,100 m depths of OGC-2 at Ogachi which was drilled into granitic rock. The bottom hole temperature was measured around 210 degree C.

Ca concentration of water sampled at 1,030 m depth increased in a few hours after CO₂ dissolved water injection then decreased in a few days. For the Ca increase it was considered that Ca was supplied from Ca-feldspar in granitic rock. Calcite precipitation on calcite crystals was observed in CO₂ dissolved water at 850 m for one hour in OGC-2.

ACKNOWLEDGMENT

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