

Field experiments for studying CO₂ mineral trap at high temperature at Ogachi, Japan

Hideshi Kaieda^{1*}, Akira Ueda², Kenji Kubota¹, Hiroshi Wakahama³, Saeko Mito³, Kazunori Sugiyama⁴, Akiko Ozawa⁴, Yoshihiro Kuroda², Yoshikazu Kaji⁵, Heigo Suzuki⁵ and Hisao Sato⁶

¹ Central research institute of electric power industry, 1646, Abiko, Abiko-shi, Chiba, 270-1194, Japan

² Kyoto university, C1-2-155, Katsura, Nishikyo-ku, Kyoto 616-8540, Japan

³ Research Institute of Innovative Technology for the Earth, 9-2 Kizugawadai, Kyoto, 619-0292, Japan

⁴ Mitsubishi Material Techno Corp., 1-14-16 Kudan-kita, Chiyoda-ku, Tokyo 102-8205, Japan

⁵ Chuo Kaihatsu Corp., 3-4-2 Nishi-aoki, Kawaguchi, Saitama, 332-0035, Japan

⁶ Mitsubishi Material Corp., 1-297, Kitabukuro, Ohmiya-ku, Saitama, 330-8508, Japan

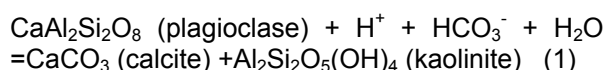
* corresponding author : kaieda@criepi.denken.or.jp

Previous experiments at Ogachi, up until 2007, generated two important results obtained by injecting CO₂ dissolved water into a 1,100 m deep well, OGC-2 at Ogachi in Japan, for which the bottom-hole temperature was measured at about 210 degree C. One result was that Ca concentration of the CO₂ dissolved (3 weight %) water, at a depth of 1,030 m in OGC-2, increased to a maximum of 85.2 mg/L in a few hours. The Ca was considered to be dissolved from rock. The second result was that calcite precipitation on calcite crystal samples was observed when the samples were held at depths of 850 m and 950 m for an hour in the CO₂ dissolved water in OGC-2. In 2008, we reconfirmed that calcite precipitation occurred on calcite crystal samples at a depth of 950 m in OGC-2. From these results, we constructed a model for describing a CO₂ mineral trap mechanism at high temperature. When CO₂ is injected underground at high temperature four zones are created, one is a super critical CO₂ zone, second is a dense CO₂ dissolved water zone, third is a thin CO₂ dissolved water zone, and fourth is a formation water zone. The CO₂ dissolved water zones react with rock. Ca dissolves from rock in the dense CO₂ dissolved water zone by high CO₂ concentration (low pH) but in the thin CO₂ dissolved zone (nearly neutral pH), by mixing with formation water, Ca precipitates as calcite minerals.

Keywords: CO₂, Mineral trap, Geothermal, Calcite, Ogachi

Concept of CO₂ Mineral Trap at High Temperature

Under high temperature conditions, the following reaction can proceed from the upper to the lower of the formula (1) (Gale and Shane, 1905).



According to Ueda et al., (2005), carbonate-rich rock formations can be observed in most Japanese geothermal fields. Reaction according to formula (1) moves towards the lower side at higher temperatures, reflecting the fact that calcite solubility decreases with increasing temperature. Calcite and kaolinite (clay)-rich rock is produced

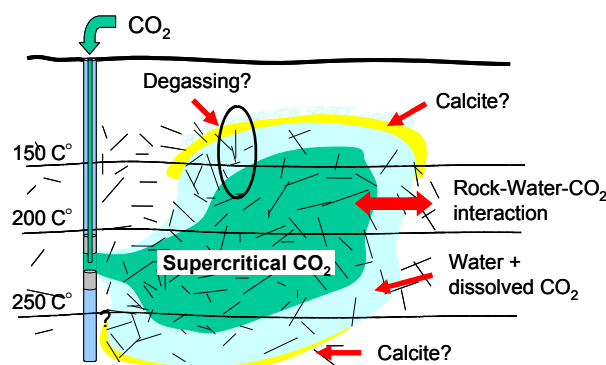


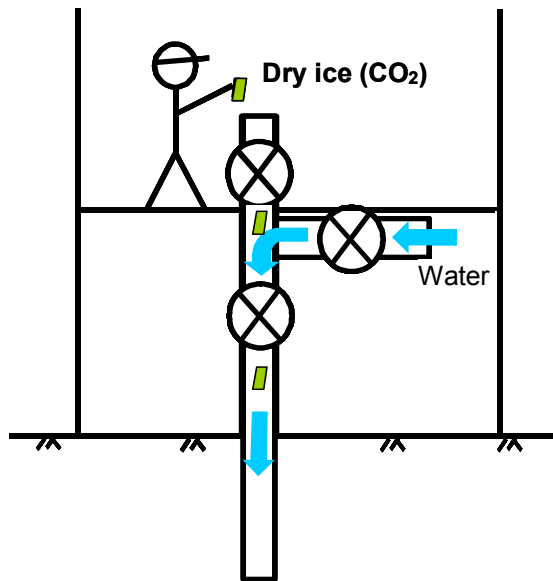
Figure 1: Concept of CO₂ injection into high temperature rock (Ueda et al., 2005)

through the reaction and forms a cap rock for the geothermal reservoir. These considerations, together with the increasing reaction rates at high temperature conditions 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).

We have conducted some field experiments from 2002 to 2008 to study CO₂ sequestration in solid minerals by injecting CO₂ dissolved in water into a high temperature borehole, OGC-2, at the Ogachi Hot Dry Rock geothermal site in Japan (Kaieda et al., 2008 and Kaieda et al., 2009). OGC-2 was drilled into granitic rock to a depth of 1,100 m and was cased from the ground surface to 700 m depth. The bottom 400 m interval of OGC-2 was left uncased and the bottom-hole temperature was measured around 210 degree C.

CO₂ dissolved water injection in 2008

The surface water (nearly neutral pH) was injected into OGC-2 at a flow rate of 50 kg/min at atmospheric pressure on September 16th, 2008. During the water injection solid CO₂ (dry ice) blocks of a few centimetres in size were injected at a rate of 0.4 kg/min to create 0.8 weight percent CO₂ dissolved water (see Figure 2). A total of 20 tons of 0.8 % CO₂ dissolved water with a total of about 160 kg-dry ice was injected into the open-hole region between 700 m and 1,100 m (bottom) in OGC-2 and into the surrounding rocks.

Figure 2: CO₂ (Dry ice) injection with water

Calcite Precipitation Measurement

After the CO₂ dissolved water was injected and replaced pre-existing formation water, a specially designed test sonde (see Figure 3) was inserted into OGC-2 and calcite crystal growth tests were conducted using the sonde. Calcite crystal samples partially covered with Au (Gold) film were set in the sonde to prevent the covered area reacting with the CO₂ dissolved water. He (Helium) gas filled the inside the sonde at an adequate pressure for protecting outside water coming in. A valve (rupture disk) of the sonde breaks when the outside pressure increases high enough relative to the He gas pressure in the

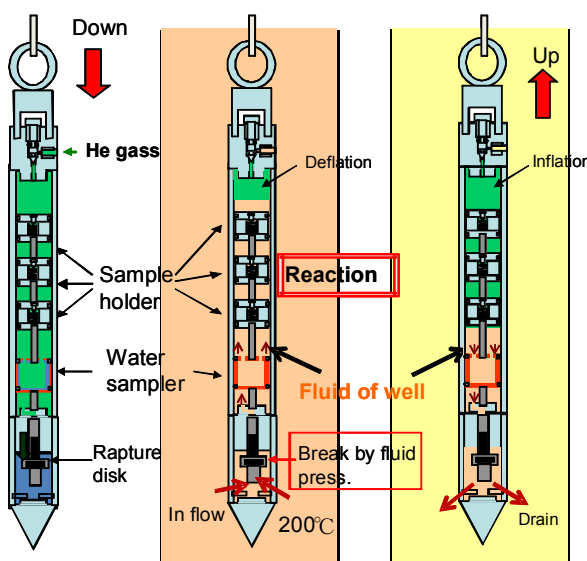


Figure 3: Test sonde for Calcite sample react with fluid in borehole

sonde. In this test we set the valve to break at a pressure of 950 m depth in OGC-2 where the well

bore wall was relatively smooth. The sonde was inserted to a depth of 950 m in OGC-2 and held there for an hour. During this hold time CO₂ dissolved water in OGC-2 came into the sonde and reacted with the samples. The sonde was recovered to the surface after the hold time and the CO₂ dissolved water drained from the sonde during retrieval from OGC-2. At the surface the samples were removed from the sonde and the surface features of the samples were observed by stereo microscope. Roughness of the sample surfaces was measured by phase shift interferometer, comparing the Au film covered area to the remaining area (see Figure 4).

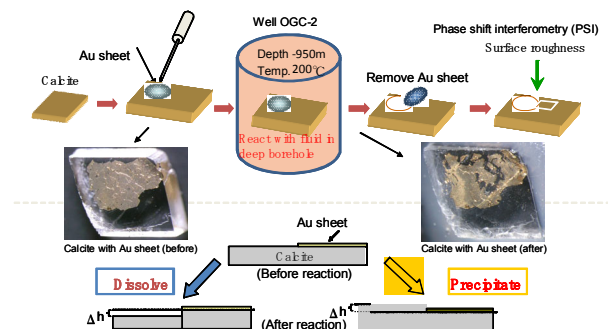


Figure 4: Calcite sample treatment

Calcite Precipitation Results

We conducted calcite precipitation tests five times before and after the CO₂ dissolved water injection from September 15 to 19, 2008. Figure 5 shows an example of the test result which was observed when the crystal test sonde was set 3.6 hours after the CO₂ dissolved water was injected into OGC-2. The top picture of Figure 5 shows an example of the surface feature of the calcite crystal sample from the stereo microscope before the test. X is a horizontal distance in micro-meter and Z is the elevation in nano-meter. The term 'masked' refers to the area covered with Au film. The middle picture shows the sample's surface at the same point as the above picture but following the test. The bottom graph shows the measured roughness of the sample surface along line A to B measured by the phase shift interferometer. The blue line represents the before test condition and the red line represents the after test condition. In the masked area, the roughness of the sample surface is relatively flat both before and after the test, but in the other area elevation of the sample surface was measured to increase after the test to a maximum of about 2,224 nano-meters (2.224 micro meters). This means that the calcite crystal samples grew in the test.

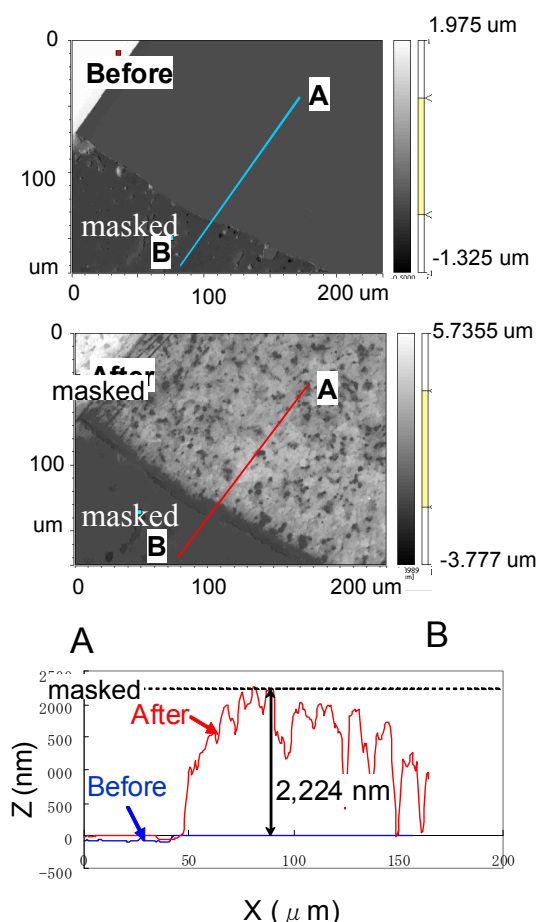


Figure 5: Stereo microscope pictures and the surface roughness along the A-B line of the calcite sample before and after reaction 3.6 hours after CO_2 dissolved water injection into OGC-2. X is horizontal distance and Z is elevation along the A-B line.

Another result is shown in Figure 6. This result was obtained when the test sonde was set 22.6 hours after the CO_2 dissolved water injection into OGC-2. In this test calcite crystal growth was also measured to a maximum of about 300 nano meters (0.3 micro meters).

From the above results, it was considered that CO_2 precipitation on crystals occurred quickly and CO_2 precipitation rate was faster a few hours after CO_2 dissolved water injection but the rate gradually decreased.

The sample dissolutions which were observed just after the CO_2 dissolved water injection in the 2007 tests were not observed in the 2008 tests.

We summarised the reaction process of CO_2 dissolved water with rock as follows: 1) water conditions in the open-hole region in OGC-2 for a few hours after CO_2 dissolved water injection was that of high CO_2 concentration and low pH. For this condition Ca is dissolved from the rock and calcite samples are also dissolved; 2) a few hours

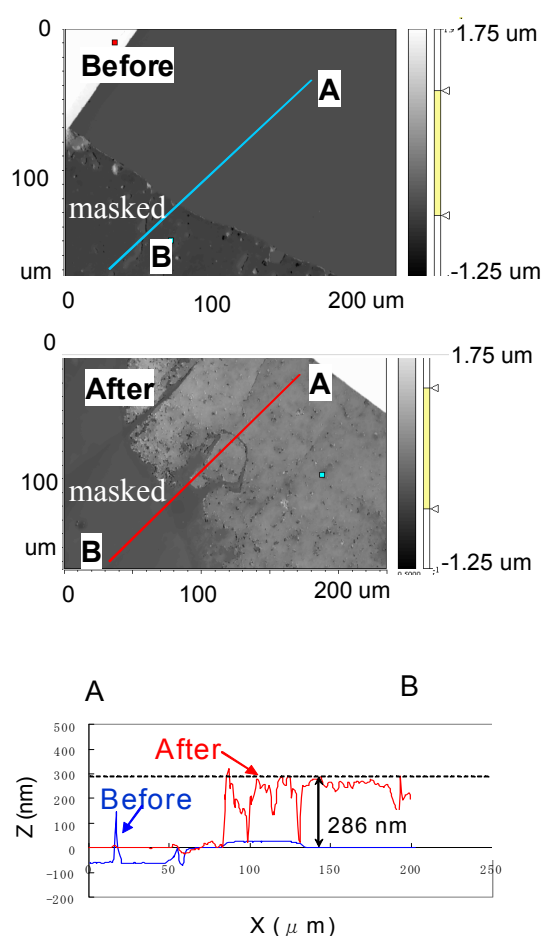


Figure 6: Stereo microscope pictures and the surface roughness along the A-B line of the calcite sample before and after reaction 22.6 hours after CO_2 dissolved water injection into OGC-2. X is horizontal distance and Z is elevation along the A-B line.

after CO_2 dissolved water injection CO_2 concentration decreased and pH increased to near neutral because of mixing with formation water, and calcite solubility decreases with temperature increase. For this condition, calcite precipitates quickly, 3) after Ca and CO_2 precipitation occurred, calcite precipitation rate gradually decreased.

CO_2 mineral trap mechanism at high temperature

Using these results we constructed a model to explain the CO_2 precipitation (mineral trap) mechanism at high temperature. The concept of this mechanism is shown Figure 7.

When CO_2 is injected deep underground (more than 800 m deep) at high temperature, four zones are created. One is a super critical CO_2 zone, second is a dense CO_2 dissolved water zone, third is a thin CO_2 dissolved water zone, and fourth is a formation water zone. Ca dissolves from rock in the dense CO_2 dissolved water zone

by high CO_2 concentration (low pH) but in the thin CO_2 dissolved zone (nearly neutral pH) where mixing with formation water occurs, Ca precipitates as calcite minerals.

To confirm this model, we need to inject super critical CO_2 (not CO_2 dissolved water) into rock and further need to observe the CO_2 concentration distribution in CO_2 dissolved water zone around the super critical CO_2 .

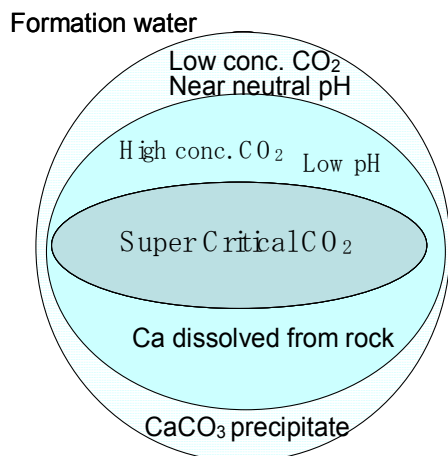


Figure 7: A model for describing the reaction of CO_2 dissolved water with rock around super critical CO_2 .

We also think that the following studies are needed as future work: the effect of calcite precipitation on permeability changes of rock: Ca dissolution rate from rock in CO_2 dissolved water; effective Ca volume in rock, and confirmation of calcite precipitation phenomena at different temperatures and with different geology.

We plan to conduct permeability tests of rock in OGC-2 before and after CO_2 dissolved water injection in Autumn 2009.

Conclusion

CO_2 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 at around 210 degree C. In 2008, we reconfirmed that calcite precipitation occurred on calcite crystal samples which was observed in previous experiments up until 2007. From these results, we constructed a model for describing a CO_2 mineral trap mechanism at high temperature. When CO_2 is injected underground at a high temperature four zones are created, one is super critical CO_2 zone, second is a dense CO_2 dissolved water zone, third is a thin CO_2 dissolved water zone, and fourth is a formation water zone. The CO_2 dissolved water zone reacts with rock. Ca dissolves from rock in the dense CO_2 dissolved water zone by high CO_2 concentration (low pH), but in the thin CO_2 dissolved zone (nearly neutral

pH), where mixing with formation water occurs, Ca precipitates as calcite minerals. In 2009 we will conduct permeability tests of rock in OGC-2 before and after the CO_2 dissolved water injection.

Acknowledgement

This research has been conducted as a collaboration work between the Central Research Institute of the Electric Power Industry and Research Institute of Innovative Technology for the Earth in the Research & Development Program "Development of innovative technology for the CO_2 fixation by Georeactor" under the fund from Ministry of Economy, Trade & Industry of Japan.

References

- Gales, C. and Shane, H., 1905, About a case of kaolinitisation in granite by a cold CO_2 bearing water, *Zenlt. Geol. Min. Palisant.*, 427-467.
- Kaieda, H., Ito, H., Kiho, K., Suzuki, K., Suenaga, H. and Shin, K., 2005, Review of the Ogachi HDR Project in Japan, *Proceedings World Geothermal Congress 2005*.
- Kaieda, H., Kubota, K., Wakahama, H., Mito, S., Ueda, A., Ohsumi, T., Yajima, T., Satoh, H., Kaji, Y., Sugiyama, K. and Ozawa, A., 2008, Experimental Study on CO_2 Injection into HDR Geothermal Reservoir, *Proceedings, 2008 Australian Geothermal Conference*.
- Kaieda, H., Ueda, A., Kubota, K., Wakahama, H., Mito, S., Sugiyama, K., Ozawa, A., Kuroda, Y., Sato, H., Yajima, T., Kato, K., Ito, H., Ohsumi, T., Kaji, Y. and Tokumaru, T., 2009, Field Experiments for Studying on CO_2 Sequestration in Solid Minerals at the Ogachi HDR Geothermal Site, Japan, *Proceedings, Thirty-Fourth Workshop on Geothermal Reservoir Engineering, Stanford University*.
- Ueda, A., Kato, K., Ohsumi, T., Yajima, T., Ito, H., Kaieda, H., Metcalfe, R. and Takase, H., 2005, Experimental studies of CO_2 -rock interaction at elevated temperatures under hydrothermal conditions. *Geochemical Journal*, Vol. 39, pp.417-425.