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CO_2 Sequestration into Hydrothermal System at Ogachi HDR Site

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ABSTRACT

Field experiments of CO2 sequestration in the Ogachi hot dry rock (HDR) site with a temperature of 200°C were performed to investigate the mineralization of CO2 as carbonates through interaction with rocks (Georeactor – Ca extraction from rocks and carbonate fixation). In 2007, carbonated water (1 wt% CO2 – river water with dry ice) was directly injected into well OGC-2 during Test #1 (September 2-9) and Test #2 (September 11-16). Several tracers were also injected simultaneously. Water samples were collected at the depth of about 850m by a sampler (500ml in volume) and monitored for their chemical and isotopic compositions. During Test #2, river water was injected into OGC-1 2 days after the injection of CO2 water into OGC-2. The CO2 concentrations in fluids collected decreased with duration time and were almost 2/3 of the expected concentration from the behavior of tracers. This provided evidence that CO2 injected into well OGC-2 can be removed from fluid by carbonate fixation.

During the field experiments, the dissolution and precipitation rates of calcite were determined using a technique of “in site analyses.” Calcite crystals covered with Au film were held in a crystal cell and set in a crystal sonde. The crystal sonde was then put into well OGC-2, and water samples were introduced into the sonde at the specified depth. After 1 hour, the sonde was recovered and the calcite crystals were observed using a newly developed phase shift interferometer to analyze the dissolution and precipitation rates of calcite in relation to the reservoir fluids. The “in situ analyses” show that calcite precipitation was observed within 2 days of the injection. This supports the view that a majority of injected CO2 might be fixed as carbonate.

1. INTRODUCTION

In recent years, much research has been directed at finding a solution to global warming caused by anthropogenic CO_2 emissions. Several research programs have focused on the possibility of sequestering such CO_2 underground, where pressures are a few tens to hundreds of bars (Holloway, 1996). Under such conditions, CO2 is thought to be stable as a supercritical fluid, due to the slow rate at which it reacts with reservoir rocks. Experimental and theoretical investigations of the chemical reaction of CO2-saturated water and rocks have been reported (Gunter et al, 1996; Perkins and Gunter, 1996; Sorai et al., 2002).

In countries with many active volcanic areas such as Japan, one possibility is to sequester CO2 in hydrothermal regions. The chemical reaction rates between CO2-saturated water and hot rocks are usually faster than those at room temperature. For example, Reaction 1 can proceed to the right at high temperatures.

CaAl2Si2O8 (plagioclase) + H+ + HCO3- + H2O =CaCO3 (calcite) +Al2Si2O5(OH)4 (kaolinite)   (1)

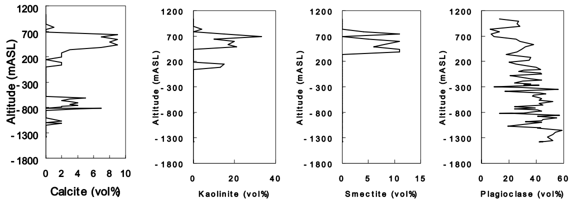

Figure 1. Abundances of calcite, kaolinite, smectite and plagioclase in rocks from Sumikawa geothermal field

Carbonate-rich rock formations can be observed in most Japanese geothermal fields. In the Sumikawa field in Akita, 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, as shown in Figure 1. 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).

2. THE CONCEPT OF “GEOREACTOR”

Reaction 1 proceeds to the right side at higher temperatures, reflecting the fact that calcite solubility decreases with rock is produced in this reaction and acts as a cap rock for the geothermal reservoir. These considerations, together with the increasing reaction rates as temperatures are elevated and the fact that precipitation of carbonate minerals fixes CO_2, suggest that CO_2 sequestration could be practicable by injection into geothermal fields (Georeactor; Fig.2).

In Japan, total rock volume in high (>150°C) and intermediate (90-150°C) temperature fields are estimated to be 2100 and 2800 km^3 respectively. CO_2 storage capacity is calculated to be about 20 billion tons of CO_2 (about 17 times Japan's total annual CO_2 emissions). The CO_2/(Ca+Mg) mole ratio of typical igneous rocks is 0.012-0.12. If 1-12% of Ca and Mg could be released, CO_2 may be fixed in carbonate minerals.

In laboratory experiments, CO_2 can be partially precipitated as carbonate during interaction with rocks at 100 to 250°C (Ueda et al., 2005). To examine this phenomena in the field, CO_2 saturated water was injected into the Ogachi HDR site.

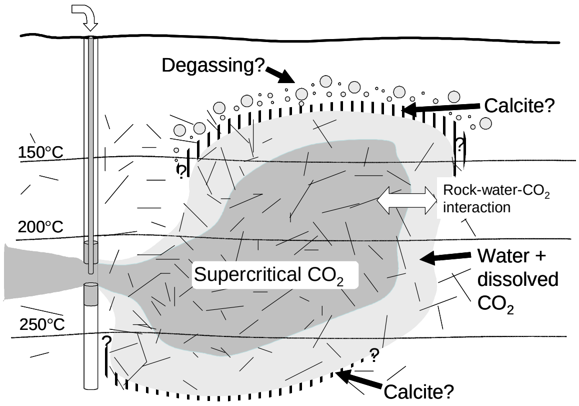

Figure 2. Outline of CO2 sequestration into geothermal fields

3. FIELD EXPERIMENTS

The Ogachi hot dry rock (HDR) field is situated in northeastern Japan and its geothermal electricity production has been studied by the Central Research Institute of the Electric Power Industry for 20 years. There are two injection/production wells (OGC-1 and -2) and an observation well (OGC-3). Two major feed zones were formed via hydro fracturing at depths of 700m and 1100m, where their temperatures are 170 and 210°C, respectively.

Field experiments of CO_2 sequestration in the Ogachi HDR have been examined for 4 years by this group (Metcalfe et al., 2006; Ueda et al., 2007). In 2006, river waters were continuously injected (380 L/min, 15 MPa) into an injection well (OGC-1) and produced (40 L/min) from a prodcution well (OGC-2) during the 2 week experiment. After the fluid flow rate became constant, CO_2 dissolved water (0.2 wt%) was injected with tracers (KI; 2 kg/200 L and uranine; 150 g/18 m^3). The fluids from OGC-2 were geochemically monitored for their isotope and chemical compositions. The final output temperature at the OGC-2 well site was 127.5°C. In this field experiment, the fixation rate of CO_2 as carbonate could not be determined due to the high dilution rate with reservoir fluids, as mentioned later.

In 2007, CO_2 dissolved water (river water with dry ice) was directly injected into OGC-2. A sketch of the injection scheme is shown in Figure 3 for Run #1 (September 2-9) and Run #2 (September 11-16). Several tracers were also injected simultaneously. Water samples were collected at a depth of about 800m by a sampler (500ml in volume) and monitored for their chemical and isotopic compositions (δD, $\delta^{18}O$, $\delta^{13}C_{CO2}$). During Run #2, river water was injected into OGC-1 (2 days after injection of CO_2 water into OGC-2).

During the field experiments, the dissolution and precipitation rates of calcite were determined using a technique called "in site analyses" (Satoh et al., 2007). Calcite crystals covered with Ti rod or Au film were held in a crystal cell and set in a crystal sonde. The crystal sonde was then put into OGC-2, and water samples at the specified depth were introduced into the sonde. After 1 hour, the sonde was recovered and the calcite crystal was observed using a newly developed phase shift interferometer (Ueda et al., 2005; Satoh et al., 2008) to analyze the dissolution and precipitation rates of calcite from the reservoir fluids.

4. RESULTS AND DISCUSSION

Test in 2006

During the flow test experiments in 2006, the major chemical compositions of fluids collected at the well site of OGC-2 were monitored. The tracers were first detected 34 hours after the injection and showed a maximum concentration at 67 hours. The dilution rates of the two tracers (KI and uranine) were calculated to be 6×10^{-5} and 1×10^{-3}, respectively. CO_2 concentrations in the fluids varied from 600 to 200 mg/L with time and showed no correlation with those of the tracers. This is due to the high dilution rate with reservoir fluids. In this field experiment, the fixation rate of CO_2 as carbonate could not be determined.

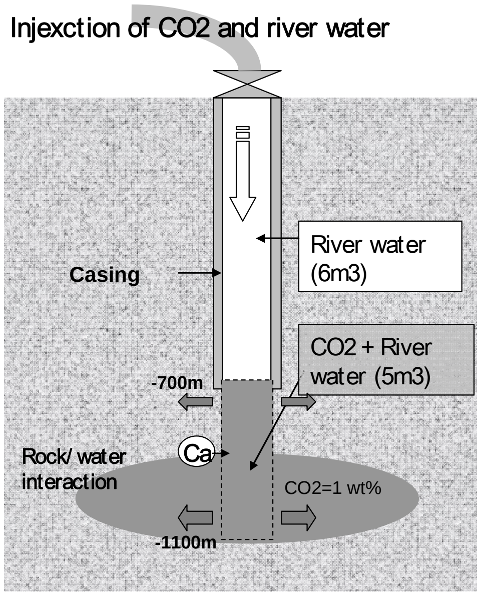

Figure 3. CO2-water injection test at OGC-2 in 2007

TEST IN 2007

After injection of CO_2 water, the tracer concentrations decreased with time. The tracer concentrations are plotted with time in Figure 4. It is noted that the behavior of natural tracers such as Cl, δD and $\delta^{13}C_{CO2}$ are different from artificially added tracers such as I, Br and fluorescein. The reason for this is not yet clear, but one possibility is that river water in the casing pipe could have mixed with CO_2 water (river water and dry ice).

During the 2007 field experiments, CO_2 concentrations in the water samples under the reservoir pressure could not be correctly monitored due to malfunction of the water sampler. This means that water samples were collected at a depth of about 800m, and part of the CO_2 had to be degassed while ascending to the surface from the sampler. Therefore, CO_2 concentrations of the reservoir fluids were calculated on the basis of the observed pH and either the charge balance of each sample (Case 1) or the tracer concentration (Case 2). In Case 2, CO_2 in the injected water did not react with rocks. The calculated CO_2 concentration with iodine is shown in Figure 5. Iodine concentration decreased from 1000 to 200μg/L with elapsed time. In Figure 4, the fraction of the injected CO_2 water (1 wt%CO_2) is almost zero. This means that the injected water was completely diluted with the reservoir fluid. The calculated CO_2 concentrations varied from 900 to 600 mg/L, as shown in Figure 5.

The saturation index of calcite was calculated from the chemical compositions of water samples as follows;

$$\text{S.I. (Saturation Index)} = \text{Log}\,(Q/K), \qquad (2)$$

where Q and K are the activity and solubility products of calcite, $(Ca^{2+})(CO_3^{2-})$, respectively. If S.I.>0, the fluid is saturated with respect to calcite (calcite precipitation).

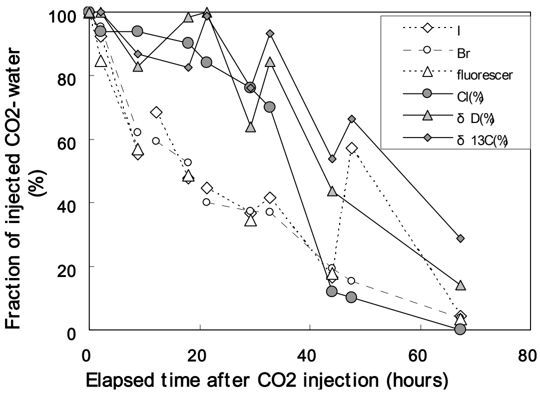

Figure 4. Tracer concentrations after CO_2 injection

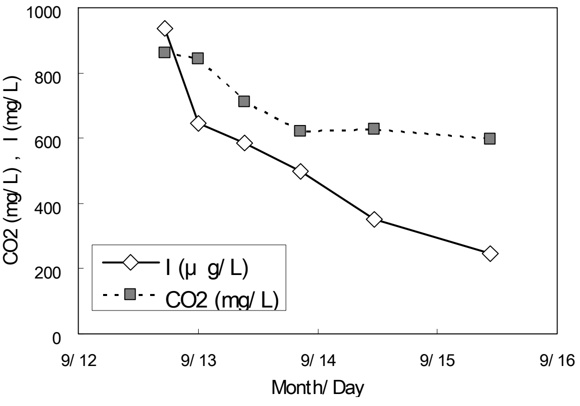

Fig.5 Concentrations of iodine and CO_2

The change of the S.I. during the experiments is shown in Figure 6 for Run 2. In Case 1, the S.I. was above zero except for the sample collected just after CO_2 injection. In contrast, the S.I. was close to zero in Case 2, implying that the reservoir fluid mixed with the injected CO_2 water was almost in equilibrium with calcite. The observed dissolution and precipitation rates of calcites determined by in site analyses are also shown in Figure 6. From these results, it can be inferred that the injected CO_2 water was diluted with the reservoir fluid within 3 days, and most CO_2 in them might have been deposited by interaction with granitic rocks. According to the material balance, a granitic rock thickness of only 1μm is enough to react with the CO_2 water and precipitate carbonate minerals. In the previous field experiments in 2002 and 2003, Ca concentration in the water samples increased from 10 to 80 mg/L within 1 day and quickly decreased to the background. These results also support the view that injected CO_2 was precipitated as carbonates within a few days.

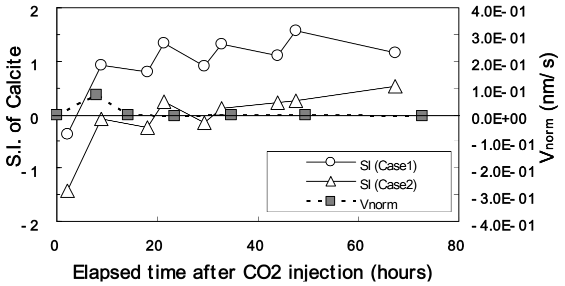

Figure 6. Saturation index of calcite

5. CONCLUSIONS

The injected CO_2 water was diluted with the reservoir fluid within 3 days. Most CO_2 in this fluid may have been deposited as calcite due to interaction with granitic rocks. Ca concentration in the water samples increased from 10 to 80 mg/L within 1 day and quickly decreased to the background. These results also support the view that injected CO_2 was precipitated as carbonates within few days.

REFERENCES

Gunter, W.D., Wiwchar, B and Perkins, E.H. (1996) Autoclave experiments and geochemical modeling. Aquifer disposal of carbon dioxide (Hitchon B. ed.), 115-141.

Holloway, S. ed. (1996) The underground disposal of carbon dioxide, Final report, Contract No. JOU2 CT92-0031, 355p.

Metcalfe1,R., Ueda,A., Kato,K., Ohsumi,T., Yajima,T., Ito,H., Kaieda,H., Takase,H., Savage,D. and Benbow,S. (2006) The feasibility of sequestering CO_2 in geothermal systems: a theoretical evaluation, abst. GHGT8

Perkins, E. H. and Gunter, W. D. (1996) Mineral traps for carbon dioxide. Aquifer disposal of carbon dioxide (Hitchon B. ed.), 93-113.

Satoh,H., Yajima,T., Ueda,A., Ozawa,A., Kato,K. and Kaji,Y. (2007) CO2 injection experiment at Ogachi hot dry rock: In-situ test of calcite production using a crystal-growth sonde, Abst. Meetings Geothermal Soc. Japan

Satoh,H., Nishimura,Y., Tsukamoto,K., Ueda,A., Kato,K. and Ueta,S. (2007) In-situ measurement of dissolution of anorthite in Na-Cl-OH solutions at 22℃ using phase-shift interferometry, American Mineralogist, 92, 503-509.

Sorai, M., Ohsumi, T. and Ishikawa, M. (2002) Nanoscale surface observation of feldspar dissolved under supercritical CO_2 - water - mineral system. Proccedings 5th International symposium on CO_2 fixation and efficient Utilization of energy, 381-384.

Ueda, A., Ajima, S. and Yamamoto, M. (2001) Isotopic study of carbonate minerals from the Sumikawa geothermal area and its application to water movement. J. Geotherm. Res. Soc. Japan 23, 181-196.

Ueda,A., Kato,K., Ohsumi,T., Yajima,T., Ito,H., Kaieda,H., Metcalf,R. and Takase,H. (2005) Experimental studies of CO_2-rock interaction at elevated temperatures under hydrothermal conditions. Geochemical Journal, 39, 417-425.

Ueda,A., Yajima,T., Satoh,H., Ozawa,A., Kaieda,H., Ito,H., Ohsumi,T. and Kato,K. (2007) Preliminary geochemical results on the CO_2 Georeactor sequestration test at the Ogachi HDR site, Abst. Goldschmidt Conf.

Ueda,A., Ueta,S., Kato,K., Tsukamoto,K., Satoh,H. and Nishimura,Y. (2005) A preliminary report of development of high resolution phase shift interferometry for the assessment of nuclear waster disposal. Journal Geochemical Exploration,