Dissolution experiments were performed on serpentinite samples containing serpentine, brucite, olivine and pyroxene from Mt. Iwanai, Hokkaido, northern Japan. From the results, we obtained dissolution rate constants for serpentine \((\text{Mg}, \text{Fe})(\text{Si}_2\text{O}_5)(\text{OH})_4\), brucite \((\text{Mg}(\text{OH})_2\)) and olivine \((\text{Mg}, \text{Fe})\text{SiO}_4\) as functions of pH. These constants and obtained mineral surface area \((A)\) / mass of water \((M)\) ratio were used in a kinetic model (PATHARC)\(^1, 2\) to calculate the temporal variation of the amounts of minerals in serpentinite and the chemical composition of groundwater in which \(\text{CO}_2\) is injected \((P_{\text{CO}_2} = 100\ \text{bar} \text{ and total } \text{CO}_2 \text{ in groundwater} = 1 \text{ mol/kgH}_2\text{O}, \text{pH} = 3)\) at a depth of 1,000 m and a temperature of 40°C. The results indicate that the amounts of primary minerals (brucite, olivine, and pyroxene) decrease over time due to dissolution. Arctinite forms as a product of this dissolution in the early stages and then its amount decreases as magnesite forms. 100% of the injected \(\text{CO}_2\) will be fixed as magnesite in 22 years for sample A, a harzburgite serpentinite, and in 3 years for sample B, a dunite serpentinite. These periods are very short compared with other types of rocks like basalt, granite, and sedimentary rocks from Japan studied in our laboratory\(^3-5\). This indicates that the serpentinite rocks are potentially useful aquifer host rocks for underground sequestration of \(\text{CO}_2\).

**KEY WORDS:** \(\text{CO}_2\) Underground Sequestration, Serpentinite, Mineral Trapping, Ultramafic Rocks

1. Introduction

It is widely accepted that recent global warming is being caused by an increase in greenhouse gases such as \(\text{CO}_2\) emitted by anthropogenic activities (e.g., fossil fuel burning). Therefore, it is mandatory that humans reduce these anthropogenic \(\text{CO}_2\) emissions. One potentially useful method to reduce \(\text{CO}_2\) emissions and the greenhouse effect is underground sequestration of \(\text{CO}_2\) into groundwater aquifers. When \(\text{CO}_2\) is injected into an aquifer, the \(\text{CO}_2\) dissolves into the groundwater, reacts with the host rocks in the aquifer and, in the long-term, may precipitate as secondary minerals. If the reaction produces carbonate minerals, most of the \(\text{CO}_2\) will be trapped permanently in deep underground. So, over the long-term, \(\text{CO}_2\) injected into aquifers could be trapped in groundwater (solubility trapping) and in carbonate minerals (mineral trapping). Thus, this method is generally thought to have great potential as a highly useful and efficient method for long-term fixing of \(\text{CO}_2\) deep underground\(^6, 7\). However, one of this method’s problems is that the long-term behavior of \(\text{CO}_2\) underground is not well understood.

The trapping mechanism and the amount of carbon fixed underground depend on geologic environmental factors such as the properties of the host rocks, such as their constituent minerals, grain size, permeability, and porosity, and of the groundwater, such as its chemical composition and flow rate.

Among the various kinds of rocks, ultramafic rocks dissolve at higher rates than other igneous rocks such as granite and basalt, and sedimentary rocks such as sandstone and mudstone. Therefore, we expected that ultramafic rocks would react quickly with injected groundwater containing high concentrations of \(\text{CO}_2\), resulting in efficient \(\text{CO}_2\) fixation by solubility and mineral trapping in a relatively short period of time compared with other rocks. This is due to the high dissolution rate of the constituent minerals in ultramafic rocks (olivine and pyroxene) and high concentrations of Ca, Mg, and Fe (the main elements in carbonate minerals) in groundwater that has reacted with ultramafic rocks.

Although a large number of experimental studies on the dissolution of the minerals which are contained in ultramafic rocks (e.g. olivine, pyroxene, and serpentine) have been performed, there are few experimental studies on the dissolution of ultramafic rocks (Marini, 2007).

Therefore, in this study themselves, serpentinite, a common ultramafic rock, was selected for the dissolution experiment. The serpentinite samples collected from Mt. Iwanai, Hokkaido, northern Japan were analyzed and experimentally studied. The dissolution rate constants of the serpentinite samples were obtained by experiment. Based on those dissolution rate constants, kinetic calculations were performed to estimate the
temporal variation in the amount of carbon that could be fixed over the long-term by mineral trapping (formation of carbonate minerals) and mass transfer in water-rock reaction.

2. Experimental details

2.1 Samples

We experimented on two samples, harzburgite serpentinite (sample A) and dunite serpentinite (sample B), which were collected from drill-cores at depths of 99.0-99.4 m and 85.0-85.5 m, respectively, in Mt. Iwanai, Hokkaido, northern Japan.

The samples were powdered by pot-mill, then the powdered samples were sieved out to grain sizes of 90-100 μm and 400-600 μm. After that the fine-grains on the powdered samples were removed by ultrasonic method. The sample A and B whose average grain size of 100 μm and 500 μm is called sample A1, A2 and sample B1, B2, respectively. We used powdered samples with 100 μm and 500 μm average grain size for the experiments.

The specific surface area and average grain size of these samples were determined by the laser (LP920 unit, Horiba) and BET methods. Based on the specific surface area data and density of the samples, A/M, where A= the surface area of the powdered rock, and M= the mass of water, ratios were calculated for the samples. Table 1 displays the data gathered on specific surface area, and A/M for the starting powdered serpentinite samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Density</th>
<th>Lazer method</th>
<th>BET method</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Specific area</td>
<td>Specific area</td>
<td>A/M</td>
</tr>
<tr>
<td>g/cm³</td>
<td>m²/g</td>
<td>m²/g</td>
<td>m²/kg</td>
</tr>
<tr>
<td>SampleA → SampleA1</td>
<td>2.81</td>
<td>3.09</td>
<td>25</td>
</tr>
<tr>
<td>SampleB → SampleB1</td>
<td>2.92</td>
<td>7.21</td>
<td>25</td>
</tr>
</tbody>
</table>

2.2 Experimental procedure

0.7 g ± 0.0003 g of samples A and B were reacted with 50 ml of an aqueous solution in plastic bottles in a furnace at 25.0 ± 0.1°C.

The pH of the aqueous solution was adjusted to 3, 5, 7, or 9. HCl and NaOH were used to prepare acid and alkaline solutions, respectively. Each run lasted 1, 10, or 30 minutes, or 1, 2, 4, 8, 10, 15, or 20 hours. The final solution was filtered through a 0.22 μm membrane. pH and total CO₂ concentration were measured using a pH meter (portable pH meter HM-21P, Koa DKK, Keio University), and a CO₂ meter (Ti-9004, Koa-Chemical Institute, Keio University). The concentrations of H₄SiO₄, total Al (Al³⁺+Al(OH)₄⁻), Ca²⁺, Mg²⁺, and total Fe (Fe²⁺ + Fe³⁺) were measured by an inductively coupled plasma atomic emission spectroscopy (ICP-AE) unit (Seiko Instruments, Geological Survey of Japan). The grain size and specific surface area of the residue remaining after filtration were measured with the laser method and mineral identification was done by XRD.

The reaction experiments conducted on serpentinite samples A and B were also performed using pH buffer solutions following the procedure described above. Phthalic acid (pH 4.021), a phosphoric acid solution (pH 6.86), and a boric acid solution (pH 9.18) were used as pH buffer solutions.

3. Analytical and experimental results

3.1 Analyses of the samples

Table 2 shows the result of XRF. The major constituent minerals of sample A calculated based on XRF data and XRD data are serpentine (47% by mass), olivine (28%), pyroxene (18%), brucite (5%), and magnetite (1%). Sample B contained serpentine (73%), brucite (15%), magnetite (5%), and olivine (3%).

3.2 Variation of chemical composition of aqueous solution over time

3.2.1 pH Variation of pH over time for samples A and B with initial pH values of 3, 5, 7, and 9 are shown in Figs. 1 and 2.

The pH increased very rapidly in the early stages (within 1...
hour) to 9.5-10.5. This rapid pH increase is thought to be caused by the dissolution of brucite (Mg(OH)2) which is represented by

\[ \text{Mg(OH)}_2 + 2\text{H}^+ \rightarrow \text{Mg}^{2+} + 2\text{H}_2\text{O}. \]

As will be shown later, brucite’s dissolution rate is comparatively higher than that of the other minerals (serpentine, olivine, and pyroxene). In contrast, the pH values of the pH buffer solution interacting with the serpentinite samples did not change over time.

3・2・2 \( \text{H}_4\text{SiO}_4 \) concentration

Fig. 3 shows how the \( \text{H}_4\text{SiO}_4 \) concentration varies over time.

The \( \text{H}_4\text{SiO}_4 \) concentration increased very rapidly within the first hour. After that, step-wise increases and decreases were observed. The increase is due to the liberation of Si by the dissolution reactions of olivine, serpentine, and pyroxene, which are expressed as

\[ \text{Mg}_2\text{SiO}_4 (\text{olivine}) + 4\text{H}^+ \rightarrow 2\text{Mg}^{2+} + \text{H}_4\text{SiO}_4 \]
\[ \text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 (\text{serpentine}) + 6\text{H}^+ \rightarrow 3\text{Mg}^{2+} + 2\text{H}_4\text{SiO}_4 + \text{H}_2\text{O} \]
\[ \text{MgSiO}_3 (\text{pyroxene}) + 2\text{H}^+ + \text{H}_2\text{O} \rightarrow \text{Mg}^{2+} + \text{H}_4\text{SiO}_4 \]

Considering the dissolution rate constants of these minerals\(^8\)\(^-\)\(^1\)\(^1\) the dissolution occurred in the following order: olivine, serpentine, and then pyroxene.

3・2・3 \( \text{Mg}^{2+} \) concentration

The concentration of \( \text{Mg}^{2+} \) increased very rapidly during the first day and then increased slowly with two to three step-wise increases and decreases (Fig. 4). This behavior seems similar to that of \( \text{H}_4\text{SiO}_4 \). The variations of \( \text{Mg}^{2+} \) over time were caused by the dissolution of the Mg-bearing minerals brucite, olivine, serpentine, and pyroxene and the precipitation of Mg-Si minerals or amorphous phases.

4. Discussion

4・1 Dissolution rate constant

It is well known that the dissolution rate of a mineral is simply expressed by

\[ \frac{dm_i}{dt} = k(A/M)(1 - (\text{IAP}/\text{Ksp})) \]  \hspace{1cm} (1)

where \( m_i \) is the concentration of element i in aqueous solution, t is time, \( k \) is a rate constant, \( A \) is the surface area of the mineral, \( M \) is the mass of water, IAP is the ion activity product, and \( \text{Ksp} \) is the solubility product of the dissolving mineral.

In the early stage, \( m_i \) is negligibly small compared with \( m_{\text{eq}} \). Thus, in this case, IAP/\( \text{Ksp} \) is assumed to be zero. Thus, equation (1) becomes

\[ \frac{dm_i}{dt} = k(A/M) \]  \hspace{1cm} (2)

\[ m_i = \frac{k(A/M)t + m_0(A/M)t}{k(A/M)t} \]  \hspace{1cm} (3)

where \( m_0 \) (the initial concentration before dissolution) is assumed to be zero.

Dissolution of the other Mg-bearing minerals (serpentine,
olivine, and pyroxene) is very slow compared with brucite (8, 9, 11). Thus, the dissolution rate constants of olivine and serpentine were calculated using equation (3) (Table 3, 4). However, brucite dissolves very fast. Thus, its dissolution rate constant was determined using $\frac{d[mg]}{dt} = k[A/M] \{1 - \frac{(mg^2+ / mh^2+)}{K_{sp}}\}$ where $K_{sp}$ is brucite’s solubility product ($\log K_{sp} = 16.84$ at 25°C) (Table 5).

Fig. 5 presents a schematic diagram of the variations in the Mg and Si concentrations over time in relation to the dissolution of brucite, olivine, and serpentine and the precipitation of Mg-Si minerals or amorphous phases. Considering the schematic representation and experimental data on the change in the concentration of Si, Mg with time due to the dissolution of silicates (Fig. 5) and using equations (1) and (5), we estimated dissolution rate constants for brucite, serpentine and olivine.

It was found that the dissolution rate constants lie in the decreasing order: brucite, olivine, and serpentine. Brucite’s dissolution rate constant was determined from the variation in $mg^{2+}$ concentration during the first four hours of the test. Tables 3, 4 and 5 summarize the dissolution rate constants calculated for each run.

We compare the dissolution rate constants of brucite, olivine and serpentine as functions of pH obtained by this study with published values (8-10) in Fig. 6.

It was confirmed that generally the dissolution rate constants for brucite, olivine and serpentine obtained in this study are similar to previously published values.

The dissolution rate constants we obtained for olivine and serpentine in acidic conditions are similar to those in the literature, but in neutral-alkaline conditions, our values are influenced by brucite dissolution, which increases the concentration of $mg^{2+}$.

4.2 The influence of precipitation on the variation of $mg^{2+}$ and Si concentrations

As mentioned earlier, $mg^{2+}$ and Si concentrations generally increase with time due to the dissolution of minerals. However, they occasionally decrease, which is thought to be caused by the precipitation of Mg-Si bearing phases.

Fig. 7 shows the thermochemical stability field of minerals in the Mg-Si-O-H system with the experimental data plotted on the figure. It is found in Fig. 7 that with the proceeding of the dissolutions in pure water and buffered solution experiments $mg^{2+}$ and $H_2SiO_4$ concentrations become to be oversaturated with respect to chrysotile talc, antigorite and sepiolite. Activity ratio, $amg^{2+} / ah^{2+}$ (in logarithmic unit) and activity $a_{H_2SiO_4}$ increase with the proceeding of the reaction due to the increases in $mg^{2+}$, $H_2SiO_4$ concentration and decrease in $H^+$ concentration. These changes in the concentration area considered to be caused by the dissolution of Mg-silicates (olivine pyroxene, serpentinite) and brucite.

The $H_2SiO_4$ concentration in acidic solutions (phthalic acid and phosphoric pH buffered solutions) is mostly oversaturated with respect to quartz. However, quartz does not precipitate due to its very slow precipitation rate (12), but the Mg-Si amorphous phase is likely to precipitate. The $mg^{2+}$ and $H_2SiO_4$ concentrations in alkaline solutions (boric acid) and distilled water that interacted with the serpentinite samples were oversaturated with respect to chrysotile and antigorite (Fig. 7), indicating that these minerals or Mg-Si amorphous phases with compositions similar to them may precipitate. From the decrease in the amounts of Mg and Si during a given period we were able to make rough estimates of the Mg/Si precipitate ratio. Field emission scanning electron microscope (FE-SEM) analyses of the surface of sample B2 before and after the experimental runs indicate Mg/Si ratios of about 0.5 and 2.1, respectively. The data indicate that the phase analyzed in the sample before the
experiment is crystalline serpentine while afterward we probably have the amorphous phase with an Mg/Si ratio of about 2.

4.3 CO₂ behavior during underground sequestration in a serpentinite aquifer

4.3.1 Estimated amounts of carbon fixed by mineral trapping based on a kinetic model CO₂ behavior when sequestered underground in a serpentinite aquifer was simulated using the dissolution rate constants of brucite, olivine and serpentine obtained by this study in the PATHARC model. PATHARC is useful in simulating CO₂ behavior during underground sequestration. PATHARC is briefly described below.

According to the PATHARC model, a rate equation is given by

\[ R = R_{\text{acid}} + R_{\text{neutral}} + R_{\text{base}} = A^*k_a[\text{H}^+]^n + k_{\text{OH}}[\text{OH}^-] + k_{\text{HCO}_3^-}[\text{PCO}_2]^m(1 - \sigma)^p \]  

where \( R \) is the dissolution rate; \( R_{\text{acid}}, R_{\text{neutral}}, \) and \( R_{\text{base}} \) is dissolution rates for acidic, neutral and basic conditions, respectively; \( \sigma \) is the saturation index; \( A^* \) is the reactive surface area of the solid phase \( \text{m}^2/\text{kgH}_2\text{O} \); \( s, y, \) and \( z \) are empirically determined reaction coefficients; \( n \) and \( m \) are orders of reaction; and \( k_a, k_{\text{OH}}, \) and \( k_{\text{HCO}_3^-} \) are reaction rate constants for acidic, neutral, and alkaline conditions, respectively. Reaction coefficients \( s \) and \( y \) for serpentine were determined experimentally from the dependencies of dissolution rate on pH obtained here and in previous studies.

We derived the specific surface area, \( A^* \) and mineral surface area / mass of water ratio, \( A/M \) as functions of porosity \( \phi \) and density of mineral grain size \( r: \text{radius of grain} \).

According to equation (3), the concentrations of dissolved species increase over time. However, if the saturation index exceeds 1, the mineral precipitates and reaches close to its equilibrium (saturation) condition.

Modeling with PATHARC requires initial conditions to be given. These conditions are grain size, constituent minerals and their proportions, the chemical composition of the groundwater (pH, cation and anion concentrations etc.), and kinetic parameter values (dissolution rate constants and how they vary with pH).

If the system reaches equilibrium conditions or the minerals dissolve out, the calculation run terminates.

The calculations were performed for the following conditions.

- Depth from the surface: 1,000 m; host rocks: serpentinite (samples A and B in this study); initial PCO₂: 100 bar; equilibrium total dissolved carbon concentration \( \Sigma\text{CO}_2 \): 1 mol/kgH₂O initial pH: 3, and temperature: 40°C. For initial groundwater conditions, we used the initial ion concentrations in the groundwater at Mt. Iwanai, Hokkaido: \[ \text{[Na}^+] = 0.11135 \text{mM, } [\text{K}^+] = 0.036 \text{mM, } [\text{Ca}^{2+}] = 0.06 \text{mM, } [\text{Mg}^{2+}] = 0.66 \text{mM, } [\text{H}_2\text{CO}_3] = 1 \text{M, } [\text{Cl}^-] = 0.001 \text{M} \]. Constituent minerals and their proportions (mass%) were calculated using the chemical data from the XRF testing and minerals as identified by XRD. The mineral compositions of the samples are as sample A; serpentine (47% by mass), olivine (28%), pyroxene (18%), and magnetite (1%), sample B; serpentine (73%), brucite (15%), magnetite (5%), olivine (3%). Average grain size was 500 μm, and the porosity of the serpentinite aquifer was set at 2% and 10%, estimated from the grain size and density of the samples.

4.3.2 Simulation results The calculated variations in pH, anion, cation, and amounts of precipitated minerals produced...
The pH of the solution containing sample A increases and artinite forms very rapidly, resulting in sequestration of all injected CO₂. After that, artinite changes gradually to magnesite, resulting in 100% fixation of carbon as magnesite.

The pH of the solution reacting with sample B as host rock also increases and artinite forms very rapidly at an early stage. Artinite changes gradually to magnesite. Almost all the injected CO₂ is fixed as magnesite within about 31 years after the CO₂ is injected.

The Mg²⁺ concentration decreases rapidly due to the initial dissolution of brucite and olivine.

Comparing Figs. 10 and 13 with Figs. 14 and 15 it is found that porosity does not influence the change in the chemical composition of groundwater nor the mineral amounts during the interaction of groundwater containing CO₂ with serpentinite.

In order to clarify the effect of CO₂ concentration, a similar simulation was performed assuming [H₂CO₃] = 0.01 M and porosity = 0.02.

In this case, most of the injected CO₂ was fixed as carbonate minerals within 8.5 years for sample A and 10 years for sample B. These are very short periods compared with other main rock types in Japan (basalt, granite, sedimentary rocks) which have been studied in our laboratory 3-5, 11).

As mentioned already, uncertainty about the reactive surface area is large, however, it is certain that the injected CO₂ could be fixed in very shorter periods for the serpentinite aquifer, because the dissolution rate of the serpentinite is significantly higher than those of the other rock types and is the most important parameter which controls the efficiency of the CO₂ fixation in underground CO₂ sequestration in deep underground aquifer.

5. Conclusions

Dissolution experiments were conducted on two serpentinite drill-core samples from Mt. Iwanai, Hokkaido, northern Japan. Sample A contained 47% serpentine, 28% olivine, and 18% pyroxene, and sample B contained 15% brucite and 73% serpentine.

We determined the dissolution rate constants of brucite, serpentine, and olivine as a function of pH from tests on the samples. The results agree well with published values in acidic conditions, but are higher in neutral-alkaline conditions than previous data in the literature.

A kinetic model (PATHARC) using dissolution rate constants and surface area (A) / mass of water (M) ratio experimentally obtained by this study was used to calculate changes in the amounts of minerals in the serpentinite samples and the chemical composition of groundwater after CO₂ has been injected (P_{CO₂}=100 bar and ΣCO₂=1 mol/kgH₂O, pH=3) at a depth of 1,000 m and a temperature of 40°C. The calculations were performed using dissolution rate constants for the constituent minerals of the two samples obtained in this study.

Over time are shown in Figs. 8-15.

The pH of the solution containing sample A increases and artinite forms very rapidly, resulting in sequestration of all injected CO₂. After that, artinite changes gradually to magnesite, resulting in 100% fixation of carbon as magnesite.

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The calculations indicate that pH initially increases very rapidly from 3 to about 10 due to the dissolution of brucite, serpentine and olivine, and then remain relatively constant. The Mg$^{2+}$ and Si concentrations follow the same trend as pH; they increase very rapidly initially and then remain constant.

The amounts of the primary minerals (brucite, olivine, and pyroxene) decrease over time due to dissolution. Serpentine is formed when brucite dissolves (sample A). Artinite forms early on, then decreases and disappears after about 7 years (assuming porosity = 0.02) and magnesite forms. 100% of the CO$_2$ is fixed as magnesite in 22 years for sample A and three years for sample B. These are very short periods compared with those of other rock types like basalt, granite, and sedimentary rocks in Japan 3-5, 11). This indicates that serpentinite rocks are potentially useful aquifer host rocks, very suitable for underground sequestration of CO$_2$.

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References

蛇紋岩の溶解と蛇紋岩帯水層へのCO₂地中貯留*  

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蛇紋岩の溶解実験を行い、ブルーサイド、カンラン石、輝石の溶解速度定数のpH依存性を求めた。この溶解速度定数を用いて、カイネティックスモデルにより、1,000m、40℃の蛇紋岩帯水層へCO₂を圧入（PCO₂=100パール）した時の鉱物量と地下水水質の時間的変化を求めた。ブルーサイド、カンラン石、輝石の量は時間と共に減少し、アーチナイトが初期に生成し、その後はマグネタイトの生成と共に減少した。試料Aで22年後、試料Bで3年後に圧入されたCO₂の100％が炭酸塩として固定化された。したがって、蛇紋岩は、CO₂地中貯留に適した母岩といえる。

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キーワード: CO₂地中貯留、蛇紋岩、鉱物トラッピング、超塩基性岩

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