Factors Affecting Wollastonite Carbonation under CO_2 Supercritical Conditions

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The feasibility of a carbon dioxide disposal method based on combining carbon dioxide chemically with silicate rocks was tested in this experiment. The process uses a stirred batch reactor, in which supercritical carbon dioxide is absorbed into a rock slurry to cause dissolution of the mineral and precipitation of calcium carbonate. Wollastonite ($CaSiO_3$) was chosen in a systematic study because it had the highest conversion among the rocks investigated in a preliminary test. The effects of operating variables—including reaction time, reaction pressure, the particle size of wollastonite, reaction temperature, and solution composition—on the conversion of wollastonite were investigated, and a reaction mechanism was proposed to explain these effects. The conversion was determined by means of thermogravimetric analysis. A high conversion of 90% was achieved by using a solution composed of 1 M $NaHCO_3$ to prepare the wollastonite slurry when the carbonation reaction was operated at 8.6 MPa and 383 K for 6 h. © 2005 American Institute of Chemical Engineers *AIChE J,* 52: 292–299, 2006

Keywords: wollastonite carbonation, CO_2 fixation, liquid-phase reaction, supercritical CO_2

Introduction

For the last century, fossil fuels, such as coal, natural gas, and oil, have been widely used as energy sources, and vast amounts of carbon dioxide has been produced by burning these fuels. It is believed that the massive amounts of carbon dioxide released into the atmosphere have seriously aggravated the greenhouse effect. Actually, the average ground temperature on earth has increased by 0.5°C in the past century.[1] To solve this problem, a great deal of effort has been devoted to research areas related to the reduction of carbon dioxide. Several feasible methods are being developed to reduce the amount of carbon dioxide released into the atmosphere, including physical storage, biological fixation, and chemical fixation. Among them, the chemical fixation method seems to be potentially more effective than the others at the present stage.[2]

The use of carbon dioxide through chemical fixation can be divided into two types: that forming organic or inorganic compounds. Because the demand for organic chemicals is huge, catalytic reactions that convert carbon dioxide into organic compounds, such as methane, methanol, urea, and polymers, are potentially capable of solving the carbon dioxide problem. Although the conversion rates of these reactions are still low, they are being improved through intensive research efforts.[2] On the other hand, carbon dioxide can easily react with alkaline or alkaline earth elements dissolved in aqueous phase. The excess carbon dioxide can be used by either removing calcium ions from drinking water or recovering heavy metals from wastewater; that is, carbon dioxide is first absorbed by water and then reacts with calcium or heavy metal ions to form carbonate.[3,4] For a more realistic case, the weathering of alkaline rocks is thought to have played an important role in the reaction of atmospheric CO_2 on this planet. Many silicate rocks are available for the fixation of carbon dioxide, including wollastonite ($CaSiO_3$), olivine (Mg_2SiO_4), serpentine [$Mg_3Si_2O_5(OH)_4$], orthoclase ($KAlSi_3O_8$), albite ($NaAlSi_3O_8$), and talc [$Mg_3Si_4O_{16}(OH)_2$]. Kojima et al.[5] and Wu et al.[6] investigated a carbonation process at atmospheric pressure,

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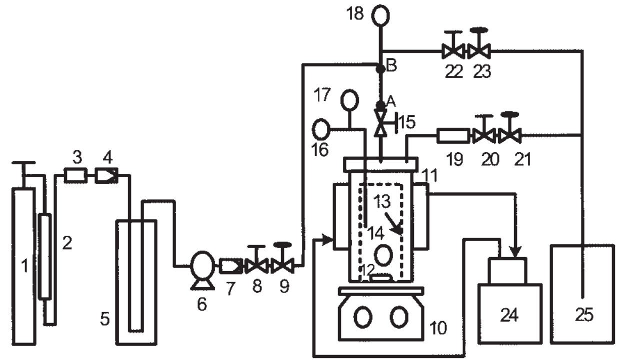

1. gas cylinder
2. dry tube
3. filter
4. check valve
5. condenser
6. pump
7. check valve
8. ball valve
9. micrometering valve
10. stirrer plate
11. reactor
12. stir bar
13. sample bottle
14. eye glass
15. ball valve
16. temperature indicator
17. pressure indicator
18. pressure indicator
19. filter
20. ball valve
21. micrometering valve
22. ball valve
23. micrometering valve
24. oil bath
25. sink

Figure 1. Experimental apparatus.

although the conversion rate was very low. Recently, a carbonation process for converting the magnesium in olivine and serpentine into $MgCO_3$ has been developed; it uses carbon dioxide dissolved in water under supercritical conditions.[7,8] The effects of operating variables, such as CO_2 pressure and reaction time, on the conversion of the silicates have been investigated, and a high conversion of 78% has been achieved in 30 min at 185 atm and 155°C.

In this study, experiments were performed to investigate the feasibility of reacting natural rocks with supercritical CO_2. In a preliminary test, different rocks were investigated and wollastonite, which had the highest conversion rate, was selected for further study. Wollastonite, which is abundant on the earth, was pulverized and then suspended in water, where it reacted with carbon dioxide under supercritical conditions. The overall reaction is given by the following equation:

$$CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2 \qquad (1)$$

The effects of several variables, including reaction temperature, rock particle size, CO_2 pressure, and reaction time, on the conversion of wollastonite were investigated. The conversion of wollastonite was determined through thermogravimetric analysis (TGA).

Experimental

Wollastonite, a natural rock imported from mainland China, was obtained from the Feng-Deng Ceramic Company in Taiwan. The rock was pulverized and sieved into three different sizes, as determined by a particle-size analyzer (Coulter LS-230) as being 147.6 (sample a), 40.5 (sample b), and 34.1 μm (sample c), respectively. These materials were used directly without any further treatment.

A schematic diagram of the apparatus used in this study is shown in Figure 1. The system is mainly composed of three parts: (1) a feeding system, (2) a pressurized reactor, and (3) a pressure releasing system. The feeding system consists of a CO_2 cylinder, a filter for filtering out fine particles, a cooler for condensing CO_2, and a piston pump (LDC analytical minipump) for feeding liquid CO_2. The reactor is a 275-mL cylindrical vessel with viewing windows. The temperature of the reactor is controlled by circulating hot oil through the jacket around the reactor. A 220-mL glass container is placed in the reactor to hold the reacting slurry and to facilitate removing the slurry sample from the reactor. The slurry is agitated by a magnetic bar during a run. The working pressure of the reactor can be increased to 21 MPa (3000 psig). The pressure-releasing system consists of a micrometering valve for controlling the depressurization rate and a water sink for absorbing the released CO_2.

At the beginning of an experiment, the glassware containing 20 mL of 10 wt % wollastonite slurry was placed in the reactor after the reactor temperature reached a setting level. Liquefied CO_2, which came from a gas cylinder, was pumped continuously into the reactor to achieve a desired pressure, after which the reaction time began to be measured. After the slurry reacted for a certain period of time, the temperature was lowered to room temperature, and the pressure was slowly decreased to atmospheric pressure. We made sure that no slurry was carried out by the released gas. Then the reacted slurry was filtered and dried overnight at 105°C. Finally, the dried solid was subjected to TGA analysis to determine the weight loss at the $CaCO_3$ decomposition temperature, X-ray diffraction analysis to determine the crystal phase, and SEM examination to determine the particle morphology.

The conversion of wollastonite to $CaCO_3$ was based on the weight loss determined through TGA analysis (TGA-7, Perkin–Elmer Cetus Instruments, Norwalk, CT). A typical TGA plot of a reacted wollastonite sample is shown in Figure 2, in which ΔW stands for the weight loss of the sample in a temperature range between 600 and 900°C caused by the decomposition of $CaCO_3$, and W_R stands for the weight after

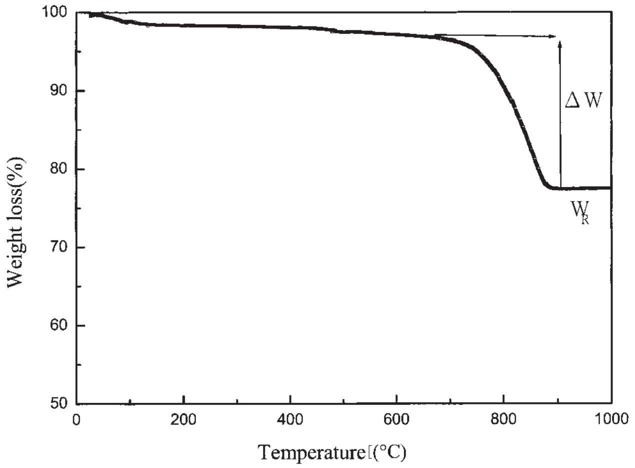

Figure 2. TGA plot of wollastonite (sample b) reacted at P = 8.6 MPa and T = 433K for 6 h.

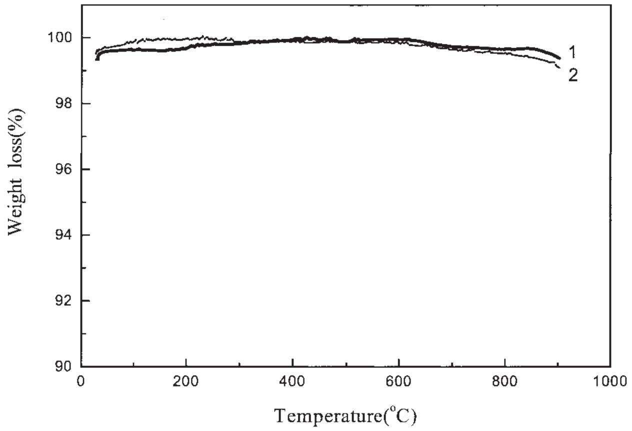

Figure 3. TGA plot of reacted and unreacted samples of talc.

The heating rate was 10 K/min in N_2. Curve 1: unreacted; Curve 2: reacted at $P = 13.9$ MPa and $T = 338$ K for 6 h.

the temperature reached 900°C. The conversion was calculated using the following equation:

$$X\ (\%) = \frac{(\Delta W/44) \times 40}{(W_R/M) \times 40} \times 100 = \frac{\Delta W \times M}{W_R \times 44} \times 100 \quad (2)$$

where 40 and 44 are the atomic weights of calcium and molecular weight of carbon dioxide, respectively; X stands for the conversion; and M stands for the weight of wollastonite, which contains 1 gmol of calcium, that is, 40 g of calcium. The values of M for the three wollastonite samples were 128.16, 136.03, and 134.58 g/gmol of calcium, respectively, as determined through EDTA titration.[9] If a rock is composed of pure wollastonite, M is equal to 116.16 g/gmol of calcium, which is the molecular weight of $CaSiO_3$. A further explanation of Eq. 2 is given as follows. Because the reaction mechanism assumes that the calcium contained in the wollastonite is converted to calcium carbonate, which will decompose completely when heated to 900°C, the weight loss ΔW is the amount of CO_2 that escapes into the atmosphere after $CaCO_3$ decomposition. Thus, the term $\Delta W/44$ is the number of moles of CO_2 that escape or the number of gmol of reacted calcium. The weight remaining at 900°C, that is, W_R, is equal to the weight of the original wollastonite sample because the CO_2 captured by the sample in the reactor has been released in the TGA analyzer. Thus, W_R/M is the number of gmol of calcium in the original wollastonite sample.

The analysis procedure for determining M is as follows. A weighted wollastonite sample was dissolved in HCl solution at a pH value of 3 until dissolution was completed. Then, some EDTA solution was added, and the solution pH was adjusted to about 12.0–12.5 by adding NaOH solution, using hydroxynaphthol blue as the indicator. The solution was then titrated with EDTA solution until its color changed from violet to clear blue as the end point. Then, the value of M was calculated using the following formula

$$M = \frac{W}{(V_E \times C_E)/40} \quad (3)$$

where W is the weight of wollastonite (in g), V_E is the titration volume of EDTA solution (in L), and C_E is the equivalent weight of calcium per unit volume of EDTA solution (in mg/mL).

Results and Discussion

Preliminary testing of rocks

In a preliminary test, three different silicate rocks, pulverized and sieved to attain a size range of 88–105 μm, were studied, including wollastonite ($CaSiO_3$), albite ($NaAlSi_3O_8$), and talc [$Mg_3Si_4O_{10}(OH)_2$]. Samples of albite and talc were treated in an aqueous solution at 65°C and 13.9 MPa for 6 h. Both unreacted and reacted samples were subjected to TGA analysis, and the weight losses of the samples were found to be negligible. For example, Figure 3 shows TGA plots of reacted and unreacted samples of talc. It is obvious that carbonation of talc did not take place because there was almost no weight loss up to 900°C for both samples. On the other hand, the weight losses of the reacted wollastonite samples, which were treated at 8.6 MPa for 6 h and at temperatures of 50 and 80°C, respectively, were significant—as shown in Figure 4, about 10 and 17%, respectively. Therefore, wollastonite was selected for further study.

Mechanism of wollastonite carbonation

Kojima et al.[5] reported that no reaction occurred between gaseous CO_2 and olivine under atmospheric pressure at 515 K. On the other hand, Tanner et al.[10] found that carbonation of rocks proceeded to some extent in the presence of water vapor under 1–3 kbar at 500–850°C. Therefore, experiments were designed in this study to see how effective water vapor would be for inducing wollastonite carbonation. In an experimental run, a sample of wollastonite was placed in aqueous phase, whereas another sample was placed in supercritical phase, which was saturated with water vapor. The reactions proceeded at 11.1 MPa and 373 K for 10 h. TGA plots of the samples are shown in Figure 5, indicating that the carbonation of wollas-

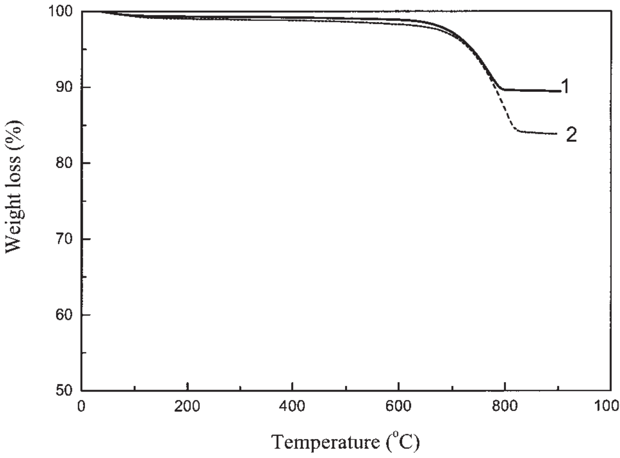

Figure 4. TGA plot of wollastonite reacted at different temperatures.

The heating rate was 10 K/min in N_2. Reaction conditions: $P = 8.6$ MPa, $t = 6$ h, and $T = 323$ K for curve 1; $T = 353$ K for curve 2.

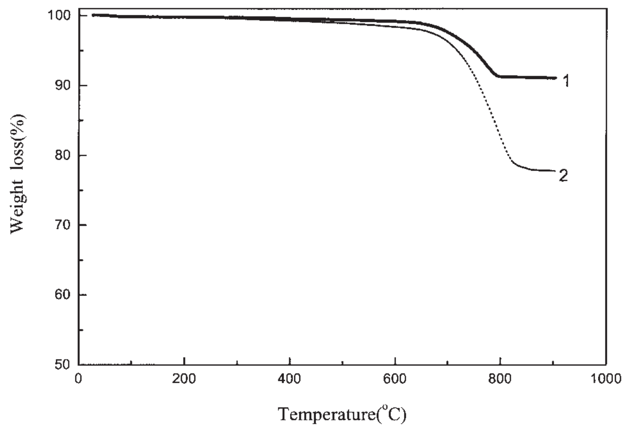

Figure 5. TGA plot of wollastonite reacted in different environments.

The heating rate was 10 K/min in N_2. Reaction conditions: $P = 11.1$ MPa, $T = 373$ K, and $t = 10$ h. Curve 1: wollastonite placed in supercritical CO_2 saturated with water vapor; Curve 2: wollastonite placed in water.

tonite took place in both phases, although it was much more effective in the aqueous phase.

From the experimental results stated above, water is required for the carbonation of rocks to proceed. This means that a reaction takes place in the water phase. A possible reaction mechanism is proposed as follows. When the water absorbs the CO_2 above the aqueous phase, the aqueous phase becomes acidic with the following species existing simultaneously

$$CO_2 + H_2O \leftrightarrows H_2CO_3 \tag{4}$$

$$H_2CO_3 \leftrightarrows H^+ + HCO_3^- \tag{5}$$

$$HCO_3^- \leftrightarrows H^+ + CO_3^{2-} \tag{6}$$

$$H_2O \leftrightarrows H^+ + OH^- \tag{7}$$

Then, the calcium contained in the wollastonite dissolves into the aqueous phase[11] as follows

$$CaSiO_3 + 2H^+ \rightarrow Ca^{2+} + SiO_2 \downarrow + H_2O \tag{8}$$

Finally, the calcium ions react with carbonate ions to form $CaCO_3$ precipitate when the ionic product of Ca^{2+} and CO_3^{2-} exceeds the solubility product of $CaCO_3$ as follows

$$Ca^{2+} + CO_3^{2-} \leftrightarrows CaCO_3 \downarrow \tag{9}$$

The precipitation of $CaCO_3$ has been studied extensively in our laboratory.[12-14] The precipitation rate is related to the CO_3^{2-} concentration, but not to the concentration of other species containing carbonate. The distribution of carbon-containing species based on different pH values has been well established.[15]

The proposed mechanism, which assumes that the precipitation of $CaCO_3$ takes place in the bulk solution, is supported by the SEM microphotographs of unreacted and reacted wol-

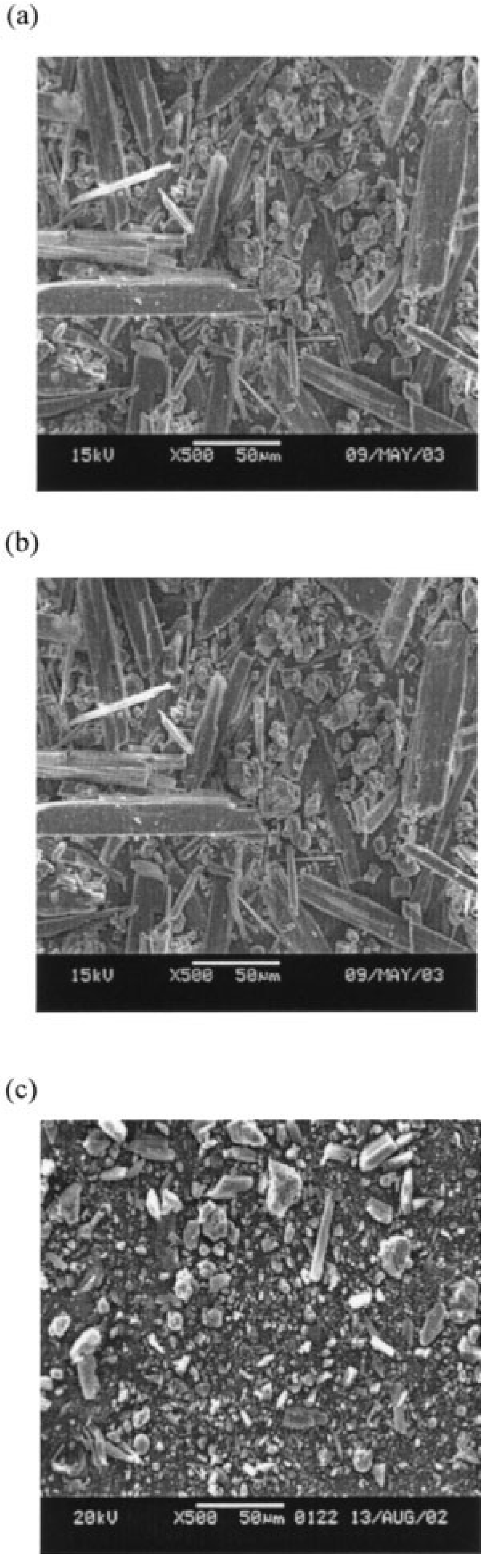

Figure 6. SEM microphotographs of unreacted and reacted wollastonite.

(a) Unreacted; (b) reacted at 6.3 MPa and 383 K for 6 h; (c) reacted at 8.6 MPa and 413 K for 15 h.

lastonite samples. Figure 6 shows SEM microphotographs of an unreacted sample and of samples that reacted for 6 and 15 h. The unreacted sample particles have high aspect ratios, ranging from 5 to 10. After 6 h of reaction time, smaller particles had a somewhat rhombohedron shape, which is a typical morphology of calcite. When the reaction time reached 15 h with a conversion of 95%, most of the acicular particles had disappeared. This means that the carbonation reaction took place in the bulk solution. Meanwhile, the SiO_2 particles, which were more angular in shape compared to the rhombohedral $CaCO_3$ particles shown in Figure 6c, perhaps as a result of attrition of the skeletal grain of wollastonite that reacted under pressure after the calcium in the rocks dissolved into the solution. Therefore, the surface of the wollastonite was first coated with a SiO_2-rich layer when the reaction proceeded. Then, the SiO_2 sloughed off under pressure and agitation, allowing hydrogen ions to further attack the rocks. That is why the supercritical CO_2 was so effective for rock carbonation. This mechanism was confirmed by the X-ray maps and backscatter-electron image of olivine carbonation product taken by O'Connor et al.[7] They distinguished four different particles: original olivine, partially reacted olivine, $CaCO_3$, and SiO_2, in the olivine product.

Based on the above mechanism, the pH value of the solution is the most important factor influencing the rate of rock dissolution (Eq. 8), the rate of CO_2 absorption (Eq. 4), and the rate of $CaCO_3$ nucleation (Eq. 9). A low pH solution will favor the dissolution of wollastonite but hinder the absorption of CO_2. If the pH of the solution is <7, almost no carbonate ions (CO_3^{2-}) exist in the solution; thus, it is difficult for nucleation of $CaCO_3$ to occur.[15] Therefore, an optimal pH exists for the carbonation of wollastonite. Unfortunately, the solution pH could not be measured in this study because a high-pressure pH electrode was not available.[7]

Reproducibility of experimental data

Because this experiment was conducted under high temperature and high pressure, it took a while for the system to heat up, to cool down, and to release pressure. Different procedures for cooling down and releasing pressure were adopted to test the reproducibility of data under the same reaction temperature (110°C), pressure (8.6 MPa), and time (6 h). The results for the different procedures are listed in Table 1. In run 36, in which the slurry temperature was cooled to room temperature before pressure was released, we obtained the lowest conversion of 60.2%. On the contrary, the highest conversion of 94.0% was achieved in run 56, in which the temperature was kept at 110°C for an extra 80 min while the pressure was released. The procedures for reducing temperature and pressure substantially affected the reaction conversion, and it seemed that the temperature was a more important factor and the reaction was suppressed at a temperature $<$ 110°C. Although a higher conversion was achieved in run 56, the procedure followed in run 36 was adopted to show the effects of operating variables more clearly.

To test the reproducibility of experimental data under the same procedures for lowering temperature and pressure, sample b (40.5 μm) was studied in three experiments at two temperatures, 80 and 110°C, while other operating conditions were kept the same. This reproducibility test was necessary because the composition of natural rocks might not be uniform. The results are listed in Table 2. The average conversions were 76.8 and 58.1%, respectively, and the largest deviation from the average value was about 5%, which is acceptable in this type of experiment.

Table 1. Conversion of Wollastonite (Sample b) with Different Experimental Procedures*

Run No.	Procedure	Conversion (%)
36	Heater turned off, slurry cooled down to room temperature in 5 h, and pressure then released to atmospheric pressure in 1 h.	60.2
55	Heater turned off, pressure released and reduced to atmospheric pressure in 80 min, and temperature decreased to 95°C, and then to room temperature in 4 h.	87.8
56	Pressure released and reduced to atmospheric pressure in 80 min, temperature kept in 110°C, and then slurry cooled down to room temperature in 5 h.	94.0

*Reaction conditions: $P = 8.6$ MPa, $T = 383$ K, $t = 6$ h.

Effect of the reaction time on conversion

The effect of the reaction time on conversion is shown in Figure 7. The conversion increased as the reaction time increased from 3 to 15 h. The conversion jumped from 41 to 75% as the reaction time increased from 3 to 6 h, and it then increased gradually to 95% as the reaction time increased to 15 h and the reaction approached completion. This is a typical result for a solid–fluid reaction, indicating the decrease of the reacting surface area or the thickening of the product layer with the extent of reaction of the solid. Because the product layer on the wollastonite particle would slough off constantly, as mentioned before, the decrease in the carbonation rate arises from the decrease of the reacting surface area of the sample. In subsequent experiments performed to study the effects of operating variables, we adopted 6 h as the reaction time because it is more suitable with respect to the magnitude of conversion and the length of the reaction time.

Effect of the particle size on conversion

Three different sizes of wollastonite were sieved to investigate the size effect in this study. The sizes were 88–105 μm (sample a), 53–63 μm (sample b), and 37–44 μm (sample c).

Table 2. Reproducibility of Experimental Data of Sample b Reacted for 6 h at 8.6 MPa

Temperature (°C)	Conversion (%)	Average Value of Conversion (%)	Deviation from Average Value (%)
80	73.5	76.8	3.3
	75.2		1.6
	81.6		4.8
110	60.2	58.1	2.1
	55.6		2.5
	58.5		0.4

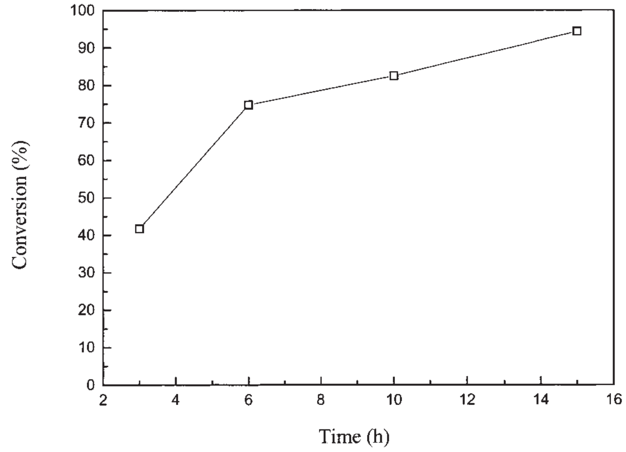

Figure 7. Effect of the reaction time on the carbonation conversion of wollastonite.

$T = 413$ K and $P = 8.6$ MPa.

The conversions for these samples at two reaction temperatures, 323 and 353 K, are shown in Figure 8. The conversion of wollastonite increased with a decrease in particle size from sample a to sample b; however, there was no difference in conversion between the two smaller sizes. The first step of rock carbonation was believed to be the dissolution of rock into the bulk solution, and the smaller sample, which had a higher surface area, should have undergone higher conversion. The results for sample a and sample b were consistent with the proposed mechanism; however, the results for the two smaller sizes were not. The particle sizes of the samples were then checked with a laser light analyzer (Coulter LS230). The mean sizes of the three samples were 147.6, 40.5, and 34.1 μm. It was clear that the sizes of the two smaller samples were quite close; thus, there was little difference in conversion.

Effect of pressure on conversion

The pressure range investigated in this study was between 8.6 and 13.9 MPa, whereas the critical pressure of CO_2 is 7.48 MPa. The reaction temperature and time were set at 413 K and 6 h, respectively. The pressure effect on the conversion of wollastonite through carbonation is shown in Figure 9, which shows that all the conversions were around 75%. Vorholz et al.[16] studied the equilibrium of the CO_2–H_2O binary system, and some of their results are shown in Table 3, which shows that the mole fraction of CO_2 in water at 393.15 K is rather independent of pressure in the range between 8 and 15 MPa. It is reasonable to assume that the amount of CO_2 dissolved in water at 413 K is also independent of pressure; thus, pressure has no effect on conversion. In addition, there was no pressure effect in the pressure range between 8.6 and 13.9 MPa with respect to attrition of the SiO_2 layer on the skeletal grain of reacted wollastonite.

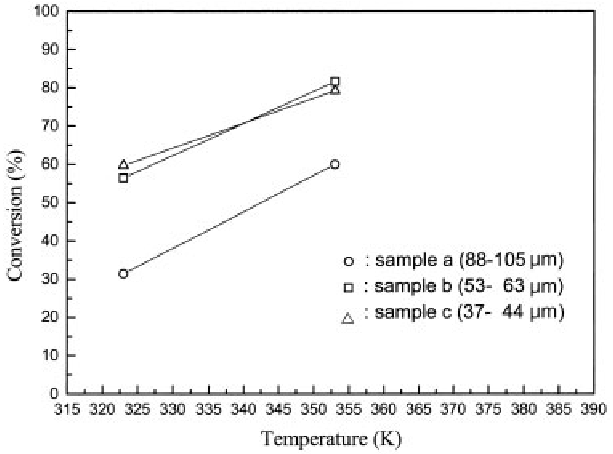

Figure 8. Conversion of wollastonite carbonation for various particle sizes at 323 and 353 K.

$P = 8.6$ MPa and $t = 6$ h.

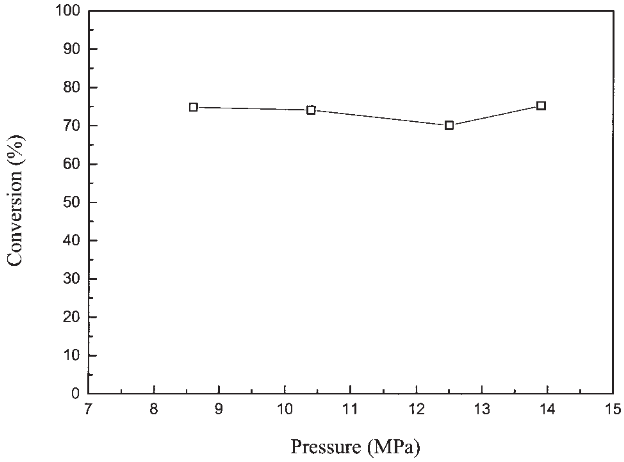

Figure 9. Effect of pressure on the carbonation conversion of wollastonite reacted at 413 K for 6 h.

Effect of temperature on conversion

Figure 10 shows the temperature effect on the conversion of wollastonite through carbonation at $P = 8.6$ MPa and $t = 6$ h for three different samples. For sample a with a size of 147.6 μm, which was the largest size studied, the conversion increased from 30 to 75% as the temperature increased from 323 to 413 K. For sample b (40.5 μm) and sample c (34.1 μm), on the other hand, the maximum conversion was achieved at 353 K. A possible explanation is based on the assumption that the controlling step of wollastonite carbonation in this study was the dissolution rate of wollastonite. When the temperature increased, the dissolution rate of silicate rocks increased.[17] On

Table 3. Mole Fraction of CO_2 in Aqueous Phase (X_{CO_2}) and Supercritical Phase (Y_{CO_2}) at 393.15 K under Various Pressures*

Pressure (MPa)	X_{CO_2}	Y_{CO_2}
8.0	0.0136	0.9658
8.0	0.0105	0.9641
10.0	0.0121	0.9694
10.0	0.0126	0.9706
10.0	0.0113	0.9669
12.5	0.0132	0.9750
15.0	0.0142	0.9747

*From Vorholz et al.[16]

the other hand, a high temperature led to low solubility of CO_2,[16] which resulted in a higher pH and, thus, a lower dissolution rate. The two opposite effects of temperature competed with each other and produced different results for various sizes of wollastonite rock.

Effect of the solution composition

To study olivine carbonation, O'Connor et al.[7] added $NaHCO_3$ and NaCl to the slurry solution to improve the reaction rate dramatically. In our experiments, the effect of the solution chemistry on wollastonite carbonation was also investigated. Figure 11 compares the results for two different types of liquid that made up the reaction slurry: one was distilled water and the other was 1 M $NaHCO_3$ solution. The operating conditions were the same as those shown in Figure 10, that is, $P = 8.6$ MPa, $t = 6$ h, and $T = 323$, 353, and 383 K. When the temperatures were 323 and 353 K, the difference in conversion was minimal; however, the difference was about 20% when the temperature increased to 383 K. It should be noted that the conversion fell in the distilled water run but rose continuously in the $NaHCO_3$ solution run, when the temperature increased from 353 to 383 K. In their olivine carbonation study, O'Connor et al.[7] postulated that the addition of $NaHCO_3$ (0.45–0.64 M) had a buffering effect that kept the solution pH within a certain range, which favored olivine carbonation and caused the $NaHCO_3$ to act as a "catalyst." Although the pH value of the solution was not monitored in this study during experimental runs because of the lack of a high-pressure pH sensor, we believe that the buffering effect existed in our system. It should be noted that, as stated earlier, an optimal pH exists for wollastonite conversion.

Conclusion

A carbon dioxide disposal method was proven to be feasible in this study, in which supercritical carbon dioxide reacted with a slurry of wollastonite, which converted to $CaCO_3$ and SiO_2. Several operating variables were investigated, including reaction time, reaction pressure, the particle size of wollastonite, reaction temperature, and solution composition. Among them, reaction temperature and solution composition were found to be the most important factors. A high conversion of 90% was achieved by using a 1 M $NaHCO_3$ solution to prepare wollastonite slurry and conducting carbonation reaction at 8.6 MPa and 383 K for 6 h. According to the evidence shown in SEM microphotographs of the reaction products and the experience gathered in our laboratory with respect to calcium carbonate precipitation, a reaction mechanism—in which attrition of the SiO_2-rich layer that occurs after the calcium in the rocks dissolves into the solution allows further chemical attacks on the rocks—has been proposed to explain the effects of operating variables.

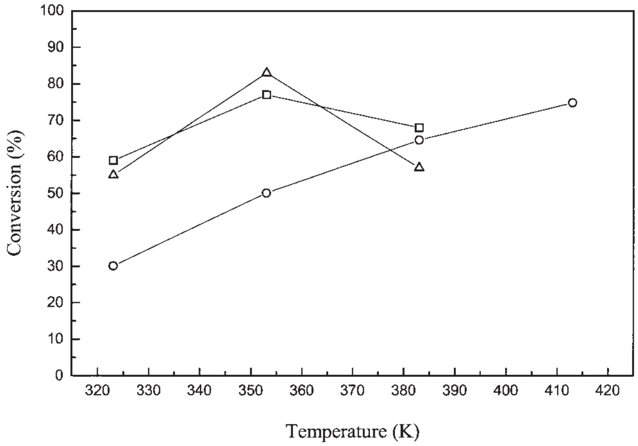

Figure 10. Effect of temperature on the carbonation conversion of wollastonite.

Operating conditions: $P = 8.6$ MPa and $t = 6$ h. ○, sample a; △, sample b; □, sample c.

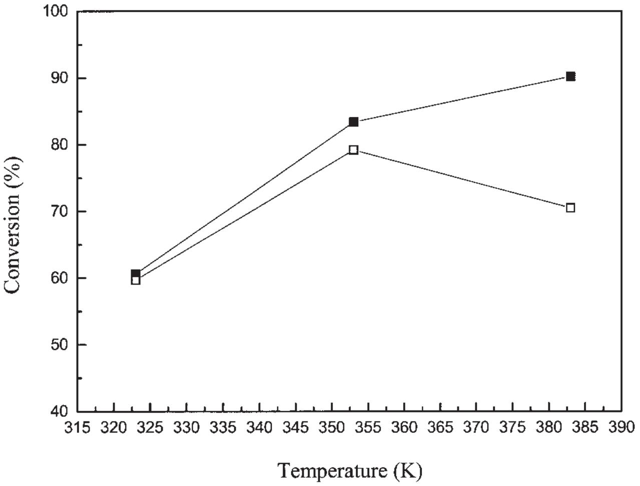

Figure 11. Effect of the solution chemistry on the carbonation of wollastonite (sample c).

Operating conditions: $P = 8.6$ MPa and $t = 6$ h. □, distilled water; ■, 1 M $NaHCO_3$ solution.

Notation

C_E = weight of calcium equivalent to one liter of EDTA solution, g
M = weight of wollastonite containing one g-mole of calcium, g
P = pressure, MPa
T = temperature, K
t = reaction time, h
V_E = titration volume of EDTA solution, L
W = weight of wollastonite, g
W_R = weight of sample remained at 900°C in TGA analysis, g
X = conversion of wollastonite carbonation
X_{CO2} = mole fraction of CO2 in aqueous phase
Y_{CO2} = mole fraction of CO2 in supercritical phase
ΔW = weight loss of sample between 600 and 900°C in TGA analysis, g

Acknowledgments

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