

DOE/ARC-2000-008

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March 6-9, 2000

Proceedings of the 25th International Technical Conf. On Coal Utilization & Fuel Systems, Coal
Technology Assoc., Clear Water, FL

by W.K. O'Connor, D.C. Dahlin, D.N. Nilsen, R.P. Walters and P.C. Turner
Albany Research Center
Albany, Oregon



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Albany Research Center
Office of Fossil Energy
U.S. Department of Energy

ABSTRACT

The Albany Research Center (ARC) of the U.S. Dept. of Energy (DOE) has been conducting a series of mineral carbonation tests at its Albany, Oregon, facility over the past 2 years as part of a Mineral Carbonation Study Program within the DOE. Other participants in this Program include the Los Alamos National Laboratory, Arizona State University, Science Applications International Corporation, and the DOE National Energy Technology Laboratory. The ARC tests have focused on ex-situ mineral carbonation in an aqueous system. The process developed at ARC utilizes a slurry of water mixed with a magnesium silicate mineral, olivine [forsterite end member (Mg_2SiO_4)], or serpentine [$Mg_3Si_2O_5(OH)_4$]. This slurry is reacted with supercritical carbon dioxide (CO_2) to produce magnesite ($MgCO_3$). The CO_2 is dissolved in water to form carbonic acid (H_2CO_3), which dissociates to H^+ and HCO_3^- . The H^+ reacts with the mineral, liberating Mg^{2+} cations which react with the bicarbonate to form the solid carbonate. The process is designed to simulate the natural serpentinization reaction of ultramafic minerals, and for this reason, these results may also be applicable to in-situ geological sequestration regimes.

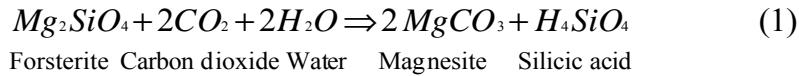
Results of the baseline tests, conducted on ground products of the natural minerals, have been encouraging. Tests conducted at ambient temperature ($22^\circ C$) and subcritical CO_2 pressures (below 73 atm) resulted in very slow conversion to the carbonate. However, when elevated temperatures and pressures are utilized, coupled with continuous stirring of the slurry and gas dispersion within the water column, significant reaction occurs within much shorter reaction times. Extent of reaction, as measured by the stoichiometric conversion of the silicate mineral (olivine) to the carbonate, is roughly 90% within 24 hours, using distilled water, and a reaction temperature of $185^\circ C$ and a partial pressure of CO_2 (P_{CO_2}) of 115 atm. Recent tests using a bicarbonate solution, under identical reaction conditions, have achieved roughly 83% conversion of heat treated serpentine and 84% conversion of olivine to the carbonate in 6 hours. The results from the current studies suggest that reaction kinetics can be improved by pretreatment of the mineral, catalysis of the reaction, or some combination of the two. Future tests are intended to examine a broader pressure/temperature regime, various pretreatment options, as well as other mineral groups.

INTRODUCTION

Carbon dioxide (CO₂) gas is considered the most significant of the greenhouse gases, with emissions estimated at 6 GtC/year from the combustion of fossil fuels alone (Freund and Ormerod, 1997). Means to reduce the CO₂ emissions from the energy and/or process industries has become increasingly emphasized as a primary environmental concern. This can be achieved by efficiency improvements in fossil-fuel-fired power plants, use of alternative energy technologies, sequestration of CO₂, or some combination of these methods. Due to the availability of fossil fuel resources, it is anticipated that they will continue to play a significant role in the world's energy economy well into the future. Thus, emphasis must be placed on improving the efficiency of the fossil-fuel-fired power plant, as well as on methods to sequester the CO₂ emitted from these plants. Sequestration can take many forms, such as: 1) forestation, where CO₂ is removed from the atmosphere by biological activity; 2) aquifer storage, in which the CO₂ is injected into deep brine formations where it is trapped hydrodynamically; 3) deep sea storage, where the CO₂ is injected into the ocean below approximately 3,000 meter depths; and 4) mineral carbonation, in which the CO₂ is reacted with minerals to form solid carbonates. This final form of sequestration is truly a permanent method, because the mineral carbonates are stable over geologic time periods (millions of years), rather than the hundreds to thousands of years of stability expected for the first three forms of sequestration.

Prior discussion of CO₂ storage as mineral carbonates has appeared in the literature (Lackner, et al., 1996; Lackner, et al., 1997). The focus of these discussions was an aqueous process using hydrochloric acid (HCl) to leach serpentine and produce magnesium chloride (MgCl₂). This reaction was well developed by the Tennessee Valley Authority and U.S. Bureau of Mines during World War II as part of an alternative process for the production of magnesium metal (Houston, 1944; Gee, 1946). However, the complexity of this process led the ARC group to consider an alternative mineral carbonation process, the direct carbonic acid route, a new method for which DOE has filed a report of invention (O'Connor, et al., 1999).

In the direct carbonic acid method, CO₂ is dissolved in a slurry of water and mineral reactant, such as forsterite (Mg₂SiO₄). The CO₂ reacts with water to form carbonic acid (H₂CO₃), which dissociates to H⁺ and HCO₃⁻. Reaction of the carbonic acid with the mineral consumes most of the H⁺ and liberates equivalent amounts of cations (Mg²⁺) and bicarbonate (HCO₃⁻), which react to form the solid carbonate mineral magnesite (MgCO₃). Because this slurry is under high P_{CO₂}, the carbonic acid is continuously regenerated as it is consumed, maintaining the reaction sequence until the mineral reactant is exhausted. The theorized reaction equation follows.



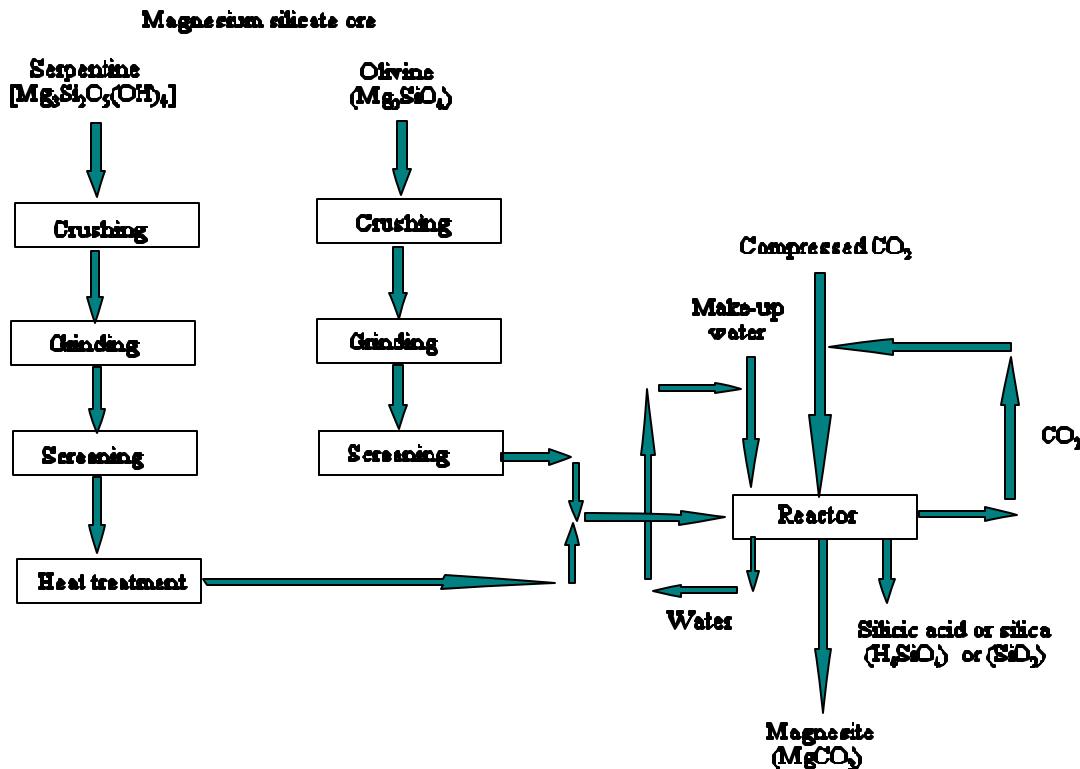


Figure 1.- Process flow diagram for the direct carbonation process.

This method dramatically simplifies the mineral carbonation process flowsheet by combining the mineral dissolution and carbonation reactions in a single unit operation (figure 1). Still, the vast tonnage of mineral reactant necessary for any ex-situ process is significant. The tonnage of silicate mineral necessary to carbonate 100% of the CO_2 emissions from a single 500 MW coal-fired power plant was estimated based on the following assumptions: 1) a mean magnesium oxide (MgO) content in the magnesium silicate ore mineral of 40 weight percent (wt pct); 2) 90% ore recovery; 3) 80% efficiency of the carbonation reaction; and 4) stoichiometry of equation 1. Based on these assumptions, a single 500 MW power plant, generating approximately 10,000 tons/day CO_2 , would require over 30,000 tons/day of magnesium silicate ore. Several ultramafic complexes in North America contain sufficient quantity of magnesium silicate mineral to provide raw materials for the mineral carbonation of all annual CO_2 emissions for many years (Goff, et al., 1997). However, the scale of these operations leads to concern over the environmental impacts and economics of any ex-situ mineral carbonation process. The work conducted at ARC is intended to provide basic understanding of the reaction mechanics, and meaningful engineering data for estimation of industrial scale operating costs.

MATERIALS

Several magnesium silicate minerals are suitable for the ex-situ carbonation reaction, including serpentine [$\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$], enstatite (MgSiO_3), and olivine, which actually represents a mineral group encompassing the solid-solution series between magnesium-rich (forsterite, Mg_2SiO_4) and iron-rich (fayalite, Fe_2SiO_4) end members. Each of these minerals occurs in ultramafic complexes, several of which are well documented in North America. Serpentine is an alteration product of olivine and other magnesium silicates, and occurs in huge deposits on both the east and west coasts of North America (Goff, et al., 1997). Olivine, particularly the forsterite end member, is also found in huge deposits on both coasts (Hunter, 1941; IGCP, 1977), although not to the extent of serpentine. For the purpose of the initial test series, simplification of the reaction sequence was desired. This meant selecting a magnesium silicate mineral of high purity, low water content, and high MgO concentration. Because serpentine is a hydrated mineral and contains a lower molar concentration of MgO than olivine, olivine was selected for the initial series of direct carbonation tests. Following the initial carbonation test series, during which basic reaction parameters were established, serpentine was included in the study. The chemical compositions of the olivine and serpentine head samples are included in table 1.

The MgO concentration of the two head samples was of most interest, at 49.7 wt pct in the olivine and 45.8 wt pct in the serpentine. The MgO content of the serpentine is actually greater than the theoretical stoichiometric amount of 43.0 wt pct. This is likely an indication of remnant olivine mixed with the serpentine, although this was not identified in the X-ray diffraction (XRD) analyses (table 2). The XRD analyses confirm the presence of forsterite as the primary phase in the olivine head sample, with a trace of enstatite. The XRD analyses of the serpentine head sample were less definitive, however. Like olivine, serpentine actually represents a mineral group, including lizardite, antigorite, and chrysotile. More detailed analysis is necessary to better define the mineralogy of the serpentine head sample used for this study. However, observation in hand sample indicates that the sample is composed primarily of antigorite, with minor amounts of chrysotile. The presence of chrysotile, the asbestosiform variety of serpentine, would likely require additional safety measures during mining and milling to minimize air-borne contamination. The bench-scale crushing operations for the current study were conducted dry, with bag filtration of the fugitive dust. The bench-scale grinding operations were conducted wet, to prevent dusting. Similar procedures would likely be necessary at the industrial scale.

EXPERIMENTAL METHODS

The initial proof-of-concept tests were conducted in an autoclave with continuous agitation or stirring, while process parameters were monitored and recorded in real time. The stirred-tank-reactor (STR) was prepared for operation by the following procedure:

- 1) 100 g of olivine weighed and placed in the STR;
- 2) 400 ml of distilled water poured into the STR;
- 3) the STR sealed, evacuated, weighed (tare weight), and placed into an ice bath;

Table 1.- Chemical composition of the olivine and serpentine feed and products (wt pct).

Oxide	SC-25 Olivine head	SC-25 carbonation product	SC-28 carbonation product	SC-40 carbonation product	Serpentine head	Serpentine heat treated in air	Serpentine heat treated in CO ₂	SC-34 carbonation product	SC-39 carbonation product
Al ₂ O ₃	0.202	0.171	0.210	0.132	0.156	0.293	0.261	0.246	0.196
CaO	0.178	0.230	0.088	0.231	0.028	0.214	0.433	0.140	0.254
Cr ₂ O ₃	0.044				0.037				
FeO	5.966	5.271	5.169	2.019	4.333	0.913	2.649	0.797	0.386
Fe ₂ O ₃	2.558	0.429	<0.010	4.058	2.629	7.345	3.915	5.730	4.973
MgO	49.677	35.507	34.007	36.841	45.843	50.343	46.676	39.008	33.673
K ₂ O	0.007			0.005	<0.002				0.007
SiO ₂	41.357	26.786	25.929	26.357	37.500	40.929	41.571	31.929	28.071
Na ₂ O	0.099			0.144	<0.002				0.414
Volatiles ¹	0.380	32.548	33.604	30.700	11.500	1.230	2.270	22.900	30.500
Total	100.466	100.942	99.006	100.487	102.026	101.267	97.775	100.749	98.474
¹ Volatile constituents listed below.									
CO ₂	<0.300	32.306	33.500	29.800	0.380	0.090	0.340	20.300	28.200
C, fixed	0.021	0.242	0.104	0.213	0.027	0.010	0.059	0.074	0.160
H ₂ O, dehyd. ²					0.150	0.230	0.210		
H ₂ O, bonded ³	0.359			0.687	10.943	0.901	1.661	2.526	2.140
Total	0.380	32.548	33.604	30.700	11.500	1.230	2.270	22.900	30.500
LOI	0.380			30.700	11.500	1.230	2.270	22.900	30.500
Total Fe	6.430	4.400	4.020	4.410	5.210	5.850	4.800	4.630	3.780

² Water of dehydration (free moisture), weight loss measured after 1 hour at 105°C, in air.

³ Chemically-bonded (interstitial) water, measured as the difference between the LOI and all other volatiles.

LOI: Loss on Ignition, weight loss measured after 1 hour at 1,000°C, in argon.

Table 2.- X-ray diffraction analyses of the feed and products.

Material	Primary phase	Secondary phase	Minor phase	Trace phase
Olivine head	forsterite (Mg ₂ SiO ₄)	ND	ND	enstatite (MgSiO ₃)
SC-25 carbonation product	magnesite (MgCO ₃) ferroan magnesite [(Mg,Fe)CO ₃]	ND	ND	forsterite enstatite
SC-28 carbonation product	magnesite ferroan magnesite	ND	ND	forsterite enstatite
Serpentine head	serpentine [(Mg ₃ Si ₂ O ₅ (OH) ₄] (one or more serp. minerals)	ND	ND	magnetite (Fe ₃ O ₄)
Serpentine, heat treated in air	forsterite	ND	ND	hematite (Fe ₂ O ₃)
SC-34 carbonation product	magnesite	ND	forsterite	ND
Primary phase: 40 to 100% Trace phase: 1 to 10%	Secondary phase: 20 to 60% ND: not detected	Minor phase: 5 to 30%		

- 4) liquid CO₂ introduced into the STR;
- 5) the STR placed back on the scale, weight of CO₂ determined
- 6) pressure bled off while STR on scale until the proper weight of CO₂ remained;
- 7) STR heated to final process temperature and pressure.

The proper weight of CO₂ was calculated in advance using the compressibility factor equation of state, $PV = zRT$, where V represents the molar volume, and z represents the compressibility factor for CO₂ at the desired test conditions. The solubility of CO₂ in water at those same conditions was also included. A detailed description of the procedure is included in the report of invention (O'Connor et al., 1999).

Following the initial test series, a new autoclave system was developed including a CO₂ gas booster pump controlled by a pressure switch. The new test procedure is as follows:

- 1) a slurry of 167 g of olivine and 950 ml distilled water poured into the STR;
- 2) the STR sealed, purged with CO₂, stirrer speed set at 1,000 rpm;
- 3) the STR heated to final operating temperature, stirrer speed set at 2,000 rpm;
- 4) CO₂ injected to final operating pressure of 126 atm ($P_{CO_2} = 115$ atm).

All tests conducted in the new system followed this simplified procedure. The inclusion of the gas booster pump permitted operation under relatively constant P_{CO_2} , which was not possible in the former system. The impact of this improvement is discussed in the following section.

EXPERIMENTAL RESULTS AND DISCUSSION

Olivine Carbonation

The initial series of olivine carbonation tests included no pretreatment options, other than size reduction to the desired particle size. These tests, exploratory in nature, were intended to produce a baseline set of test parameters. The test summaries for a selection of the olivine carbonation tests are included in table 3. Tests SC-4 through SC-14 were conducted by the first experimental procedure described above. This system included continuous agitation, but no gas dispersion in the water column. The carbonic acid process was successfully demonstrated during a series of tests conducted under identical conditions ($T = 185^{\circ}C$, $P_{CO_2} = 115$ atm, in distilled water) for 3, 6, 12, 24, and 48 hours. One limitation of the initial system was the inability to add CO₂ to the vessel as the carbonation reaction progressed. Thus, for longer test times, the P_{CO_2} decreased with increased extent of reaction, which tended to retard the reaction rate. This led to the development of an improved autoclave system, and all tests conducted after SC-14 followed the second experimental procedure described above.

Table 3.- Test summary for a selection of olivine carbonation tests.

Test	Particle size, um	Heat pretreatment	Autoclave pretreatment	Time, h	Carbonation Temp., C	PCO ₂ , atm	Carbonation starting solution	Product solids CO ₂ , wt pct	Percent stoich. conversion
SC-4	106x75	none	none	24	185	115	distilled H ₂ O	18.30	51.8
SC-10	53x37	none	none	6	185	115	distilled H ₂ O	1.25	3.5
SC-11	53x37	none	none	48	185	115	distilled H ₂ O	19.80	56.1
SC-12	53x37	none	none	3	185	115	distilled H ₂ O	0.33	0.9
SC-13	53x37	none	none	12	185	115	distilled H ₂ O	6.58	18.6
SC-14	53x37	none	none	144	150	51	distilled H ₂ O	6.21	17.6
SC-25 ¹	-37	none	none	24	185	115	distilled H ₂ O	32.30	91.5
SC-28	-37	none	supercritical H ₂ O	24	185	115	distilled H ₂ O	33.50	94.9
SC-31	150x106	none	none	24	185	115	distilled H ₂ O	3.74	10.6
SC-40	-37	none	none	6	185	115	0.5 M NaHCO ₃ , 1 M NaCl	29.80	84.4
SC-42	-37	none	none	6	115	80	0.5 M NaHCO ₃ , 1 M NaCl	0.00	0.0
SC-43	-37	none	none	6	185	80	0.5 M NaHCO ₃ , 1 M NaCl	24.09	68.2

¹ Test SC-25 and all subsequent tests conducted in new autoclave system with gas dispersion.

Table 4.- Test summary for a selection of serpentine carbonation tests (conducted in new autoclave system with gas dispersion).

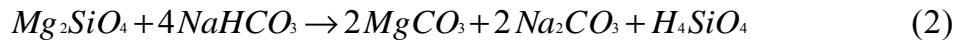
Test	Particle size, um	Heat pretreatment ¹	Autoclave pretreatment	Time, h	Carbonation Temp., C	PCO ₂ , atm	Carbonation starting solution	Product solids CO ₂ , wt pct	Percent stoich. conversion
SC-32	-37	none	none	24	185	115	distilled H ₂ O	11.40	34.2
SC-33	-37	none	none	24	185	115	distilled H ₂ O	11.50	34.5
SC-34	-37	in air	none	24	185	115	distilled H ₂ O	20.30	57.0
SC-35	-37	none	supercritical H ₂ O	24	185	115	distilled H ₂ O	6.93	20.8
SC-37	-37	in air	none	24	185	115	0.5 M NaHCO ₃	22.60	63.5
SC-38	-37	in air	none	24	185	115	0.5 M NaHCO ₃ , 1 M NaCl	24.70	69.4
SC-39	-37	in CO ₂	none	6	185	115	0.5 M NaHCO ₃ , 1 M NaCl	28.20	83.2
SC-41	-37	in CO ₂	none	24	185	115	distilled H ₂ O	20.17	59.5
SC-44	-37	none	none	6	185	115	0.5 M NaHCO ₃ , 1 M NaCl	2.28	6.8
SC-45	-37	in air	none	6	185	115	0.5 M NaHCO ₃ , 1 M NaCl	14.45	40.6

¹ Heat treatments conducted for 2 hours at 600-650°C.

The effects of particle size, temperature, and P_{CO_2} on the olivine carbonation reaction were examined by completion of the tests included in table 3. Particle size was recognized as a major factor determining reaction rate and extent of reaction, because most mineral dissolution reactions are surface controlled. This effect is evident in the extent of reactions depicted in figure 2. Extent of reaction, as a measure of the stoichiometric conversion of the silicate to the carbonate, increased dramatically with decreasing particle size, to over 90% for the test conducted on -37 micron olivine feed material (SC-25). Based on this result, all subsequent tests were conducted on material ground to this particle size.

A second series of tests, conducted at a complimentary sequence of increasing temperature and P_{CO_2} , helped to define the lower limits for these parameters (figure 3). At 115°C and $P_{CO_2} = 80$ atm, no discernable reaction had occurred after 6 hours. However, increasing the temperature to 185°C while holding P_{CO_2} constant resulted in over 65% extent of reaction in 6 hours. Extent of reaction was improved further, to nearly 85%, by increasing the P_{CO_2} to 115 atm while holding the temperature constant at 185°C. This test series, coupled with an earlier test conducted at 150°C and subcritical P_{CO_2} (SC-14), strongly suggests that supercritical P_{CO_2} improves reaction rate and extent of reaction. In addition, temperatures above 150°C are likely necessary to achieve effective mineral dissolution rates.

This latter test series was conducted in a modified solution chemistry, 0.5 M $NaHCO_3$, 1 M $NaCl$, which dramatically improved the reaction rate. The sodium bicarbonate ($NaHCO_3$) is a more effective CO_2 carrier, demonstrated by product solution concentrations of nearly 20 g/liter CO_2 , compared to roughly 0.5-1.0 g/liter CO_2 in the tests using distilled water. The salt addition provides Cl^- which is thought to help activate the mineral. The modified solution is very stable, with nearly constant pre- and post-test pH (~7.8), and relatively constant CO_2 concentrations (~20 g/liter). This suggests that as the



$NaHCO_3$ is consumed by reaction with the mineral, it is regenerated by reaction with carbonic acid, which is also continuously regenerated due to the high P_{CO_2} in the system. The following equations represent a possible reaction sequence in this modified solution chemistry.

Pretreatment of the olivine for 1 hour at supercritical water conditions (385°C, $P_{H_2O} = 272$ atm), was conducted immediately prior to carbonation in distilled water for 24 hours at 185°C and P_{CO_2} of 115 atm (SC-28). This resulted in the highest extent of reaction achieved, although the modest gain, from 91% for test SC-25 to 95% for test SC-28, would not likely justify the additional energy expended. Maintaining the lowest possible reaction temperature and pressure is desired for the technical and economic feasibility of the direct carbonation process.

Serpentine Pretreatment and Carbonation

Initial carbonation tests conducted on serpentine ground to -37 microns resulted in much lower extent of reaction than identical tests conducted on olivine (table 4). The hydrated state of the serpentine was expected to have a negative impact on the carbonation results, so thermal gravimetric analysis (TGA) and differential thermal analysis (DTA) were conducted on the serpentine head sample to determine the appropriate temperature for removal of all water (figure 4). The endotherms on the plot signify three separate stages of water removal. Desorption of adsorbed water was complete at 160°C, while evolution of water of crystallization (dehydration) was accomplished at 374°C. However, dehydroxylation (evolution of constitutional water, or hydroxyl groups) required a temperature of 614°C. Subsequent heat treatments were conducted for 2 hours at 650°C, resulting in a consistent weight loss of 11 wt pct.

It was recognized that oxidation of the magnetite in the serpentine during heat treatment could potentially form a passive layer of hematite on the mineral surface, inhibiting the carbonation reaction. Thus, heat treatments were conducted in oxidizing (air) and non-oxidizing (CO_2) atmospheres to investigate this possibility. Analyses of the heat-treated products confirm that oxidation to hematite did occur in heat treatments conducted in air, while oxidation was limited during heat treatments conducted in CO_2 (tables 1-2). XRD analysis also confirms that the serpentine was transformed to forsterite as a result of the heat pretreatment (table 2).

The effect of the heat treatment atmosphere on the serpentine carbonation reaction was minimal for the tests conducted in distilled water (figure 5), although heat treatment in general improved the extent of reaction from 34% to over 57%. However, the effect of the heat treatment atmosphere was significant for the carbonation tests conducted in the modified solution chemistry (0.5 M NaHCO_3 , 1 M NaCl). Extent of reaction increased to over 83% for the serpentine heat treated in CO_2 , compared to 41% extent of reaction for the serpentine heat treated in air; both of these results were achieved in just 6 hours. In fact, heat treatment of the serpentine in CO_2 resulted in an extent of reaction which virtually duplicated that achieved for olivine in the modified solution chemistry (serpentine test SC-39, olivine test SC-40).

The results from these studies confirm that olivine and serpentine are equally amenable to the direct carbonation process, although serpentine requires heat pretreatment. Additional studies are underway to determine whether chemical pretreatment of serpentine can replace the heat treatment, and whether chemical pretreatment can improve the reaction rate for both olivine and serpentine. Investigations of a broader temperature/pressure regime, modified carbonation solutions, catalysts, and pretreatment options are underway.

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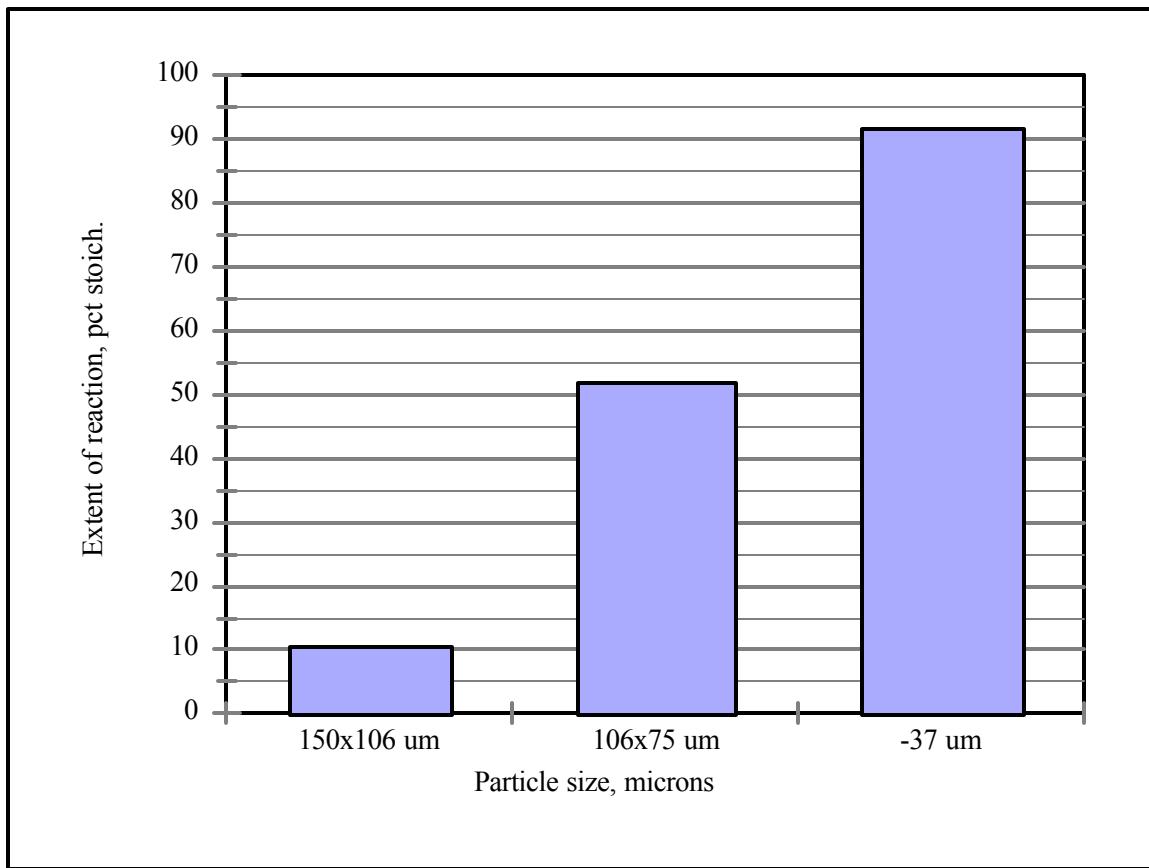


Figure 2.- Effect of particle size on the olivine carbonation reaction.
Test conditions: 24 h, $T = 185^{\circ}\text{C}$, $P_{\text{CO}_2} = 115$ atm, in distilled water.

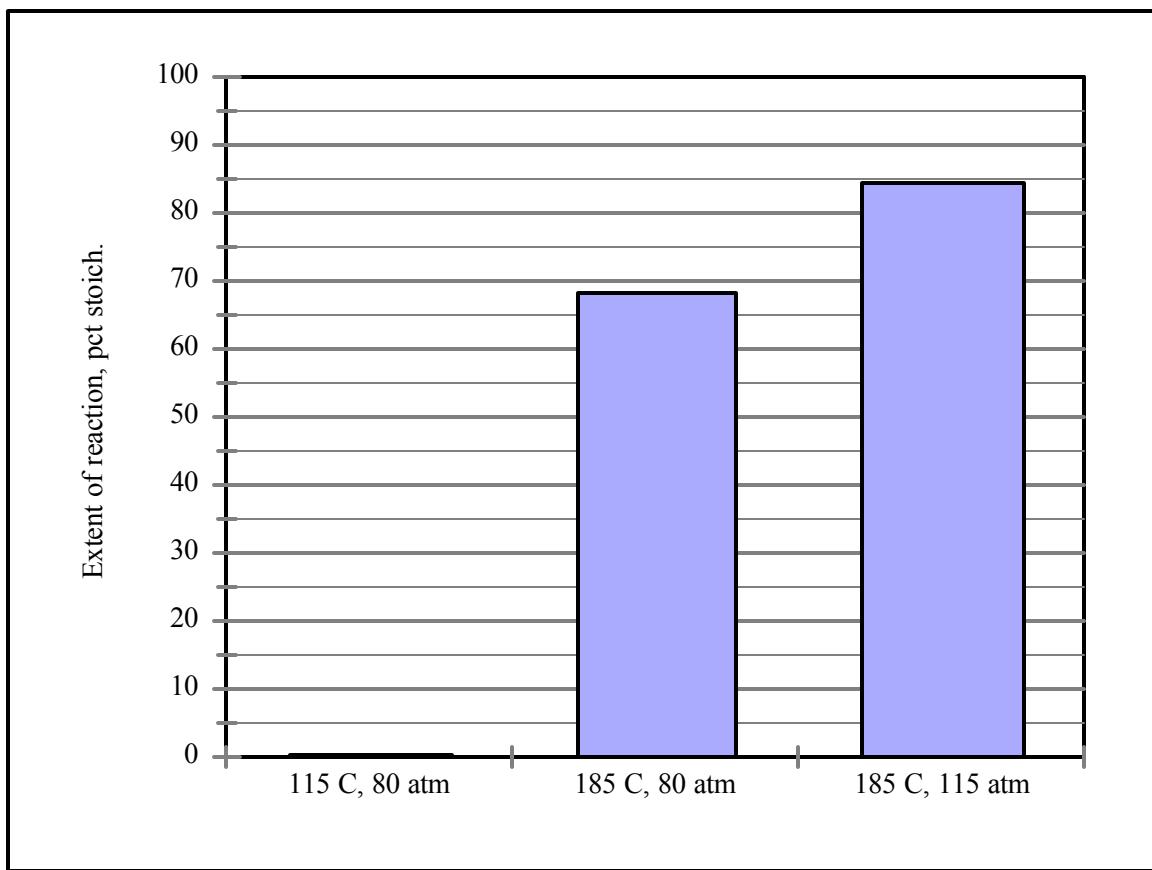


Figure 3.- Effect of temperature and P_{CO_2} on the olivine carbonation reaction.
Test conditions: 6 h, in 0.5 M $NaHCO_3$, 1 M $NaCl$.

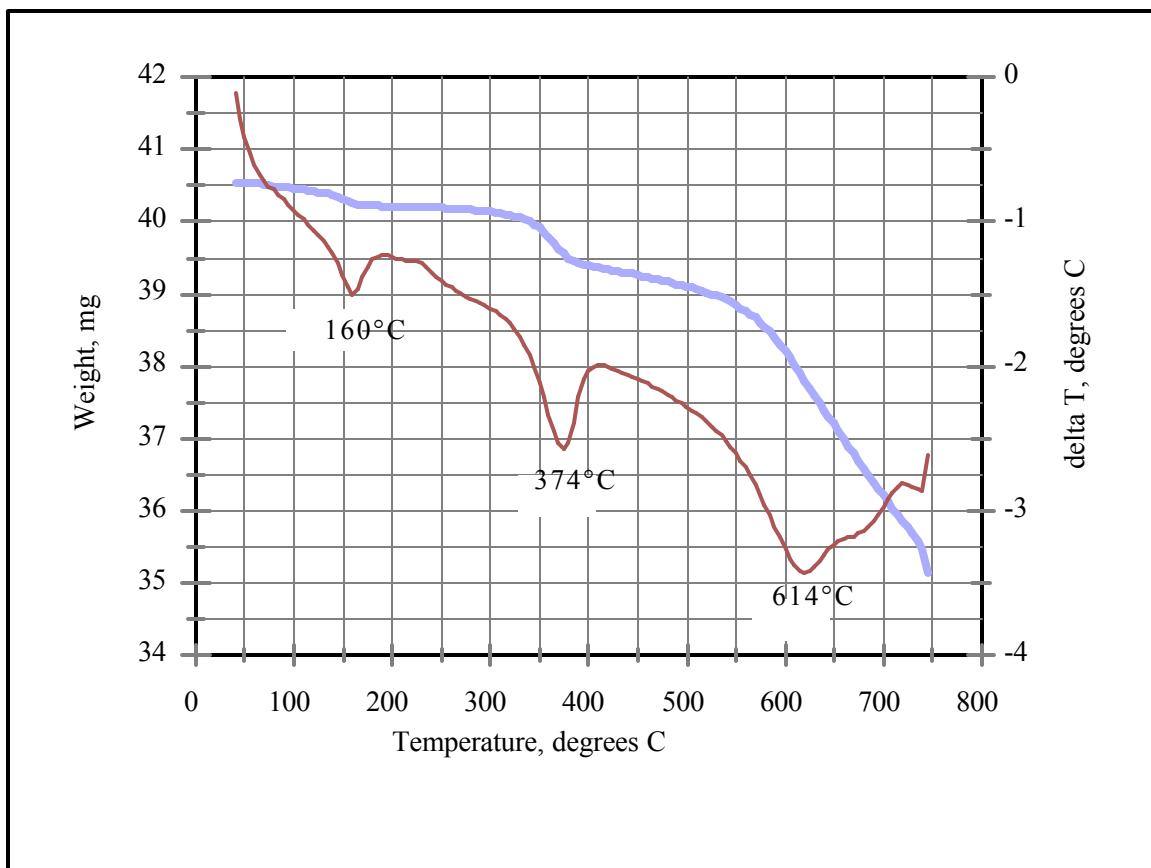


Figure 4.- Thermal gravimetric analysis (heavy line) and differential analysis (light line) for the serpentine head sample.

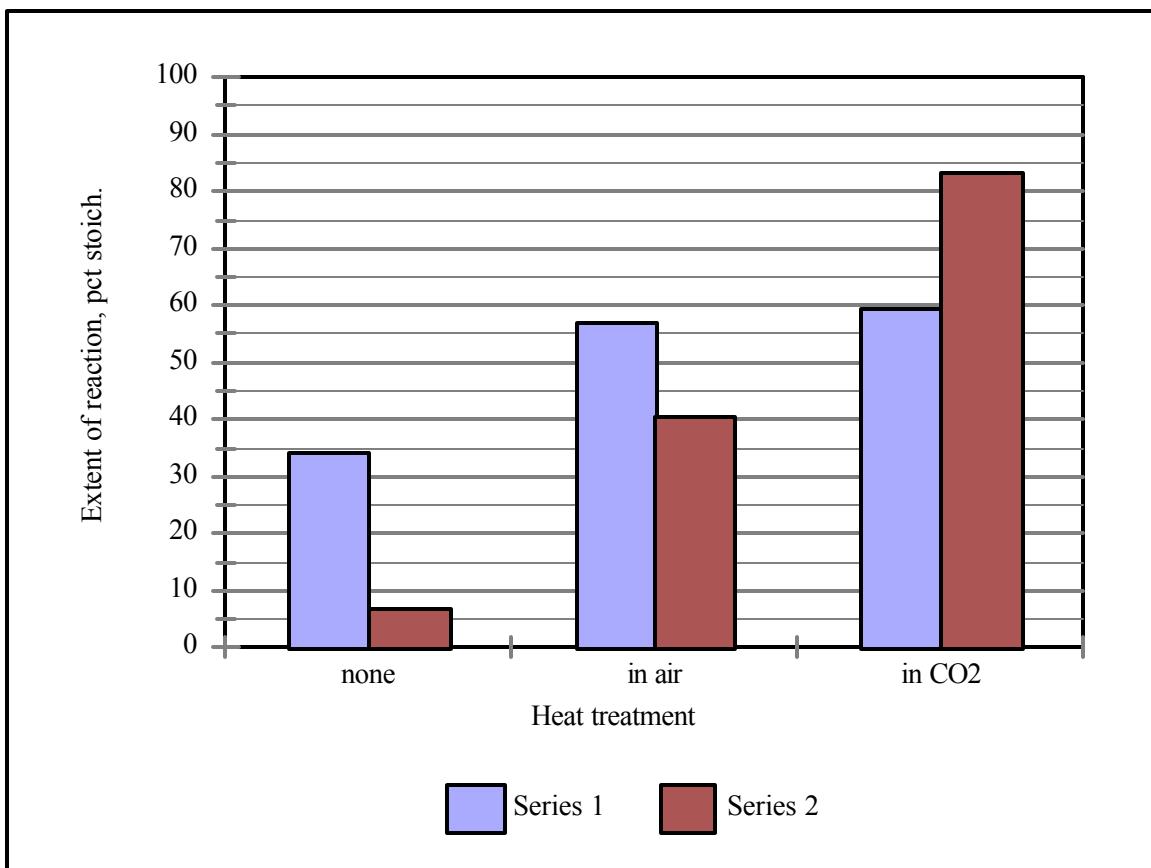


Figure 5.- Effect of heat treatment on the serpentine carbonation reaction, in two series:
(1) 24 h, T = 185°C, P_{CO_2} = 115 atm, in distilled water; and (2) 6 h, T = 185°C, P_{CO_2} = 115 atm, in 0.5 M NaHCO₃, 1 M NaCl.