

Enhanced carbonation curing of cement pastes with dolomite additive

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Abstract: Carbonation curing of cement-based materials has recently received increasing attention as a CO₂ utilization technology. This study aimed at investigating the effects of carbonation curing on the performance of ordinary Portland cement (OPC) pastes with dolomite additive (DPC). The CO₂ uptake capacity, after being normalized to carbonation active components, significantly increased with larger dolomite mixing ratios. For DPC-25% samples under 2.5 MPa curing pressure, the maximum CO₂ uptake capacity reached 23.6 wt%, which was 23% higher than that of pure OPC samples under the same condition. Effects of water to solids (w/s) ratio and temperature on carbonation are two-sided. The optimum w/s ratio for CO₂ uptake capacity of DPC-15% samples was approximately 0.20, while the optimum temperature was equal to 60°C or higher than 60°C. The CO₂ uptake capacity increased with finer particle size and higher CO₂ curing pressures. Compared to large particles, smaller particles are more likely to have a better dilution effect, providing more contact surface for carbonated precipitation. From the pore structure changes perspective, carbonation products filled the interface between the dolomite and amorphous particles. DPC-25% samples with higher dolomite mixing ratios provided more pores and pathways for gas diffusion, and exhibited a more uniform structure, thereby contributing to the highest compressive strength values of DPC-25% samples (63.8 MPa) among all the DPC samples. These findings imply the possible feasibility of dolomite as an additive in carbonation cured building materials. © 2022 Society of Chemical Industry and John Wiley & Sons, Ltd.

Keywords: carbonation curing; CO_2 utilization; compressive strength; dolomite (CaMg(CO_3)₂); microstructure

Introduction

According to the Special Report on Global Warming of 1.5° C published by the Intergovernmental Panel on Climate Change (IPCC),¹ limiting global warming to 1.5° C would reduce the challenging impacts on ecosystems, human health, and well-being, but would require a "deep emissions reductions" and "rapid, far-reaching and unprecedented changes in all aspects of society". Carbon dioxide capture, utilization, and storage (CCUS) is a crucial strategy for achieving these ambitious CO₂ emissions reductions. Among these

reductions, mineral carbonation has been proposed as a promising approach for CO_2 sequestration and utilization. The basic concept of mineral carbonation is to imitate normal processes of chemical weathering of calcium- and magnesium-bearing silicate minerals, such as olivine ((Mg,Fe)SiO₄) and serpentine (Mg₃Si₂O₅(OH)₄).^{2,3} The traditional mineral carbonation process involves two pathways, direct pathways based on high pressure or high temperature and indirect pathways based on mineral leaching and Ca^{2+} and Mg²⁺ precipitation. Although this traditional process has a high CO₂ sequestration potential due to a

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Received December 7, 2021; revised February 6, 2022; accepted February 9, 2022 Published online at Wiley Online Library (wileyonlinelibrary.com). DOI: 10.1002/ghg.2143 large amount of silicates ores and solid wastes,⁴ its application is limited by high operation costs caused by energy and chemical products consumption.^{5,6} Therefore, CO_2 utilization technologies with value-added products and low costs, such as carbonation curing of cement-based materials, will be more applicable in the future.^{7,8}

Ordinary Portland Cement (OPC), as the most commonly used hydrating cement material, can also be used as the raw material for carbonation curing. In the concrete preparation process, OPC first undergoes a hydration process. After the hydration process, the phases in concrete include calcium hydroxide (CH), calcium silicate hydrates (C-S-H), calcium aluminate hydrate, ettringite, unreacted clinker minerals (C₃S, C_2S), and so on.^{9,10} In the carbonation process, these components can react with CO_2 to form carbonates, which leads to a CO₂ uptake of up to 30% of the mass of calcined cement.¹¹ Moreover, carbonation causes numerous changes in cement pastes, including notable changes in pore size distribution, porosity, and strength.^{12,13} Precipitation of CaCO₃ preferentially occurs in pores, resulting in the pore size distribution curves to shift towards smaller pore diameters and reducing the total volume of pores per gram of paste.¹⁴ These changes further lead to the development of compressive strength.⁷

The supplementary cementitious materials (SCMs), with no additional clinkering processes, are commonly used in concrete to partially substitute OPC, which enhances the economics of OPC cement and reduces carbon footprints. Although limestone is widely used as a type of SCM, its resources are limited globally. Due to the increasing demand of construction, as well as the need to protect the environment and prevent soil erosion, limestone available for exploitation will also be reduced. Other carbonate minerals with more reserves, such as dolomite ($CaMg(CO_3)_2$), may be more promising alternatives.¹⁵ Previous studies on dolomite as an SCM focused on natural curing concrete, which mainly involves the hydration process.¹⁵⁻¹⁸ With the increase in dolomite mixing ratios in concrete, a significant decrease in compressive strength has been observed in all ages of the hydration process, which may be due to the fact that dolomite is nonpozzolanic in natural curing conditions.¹⁹ Moreover, dolomite is not stable in the high-alkaline environment of cement. It undergoes the dedolomitization reaction,¹⁵ whereby dolomites react with calcium hydroxide to form calcium carbonate and magnesium hydroxide to fill

pores. In comparison, a limited number of studies have focused on the carbonation process. Yang et al.²⁰ evaluated the early hydration and carbonation (20° C, RH > 60%, 99.5% CO₂ of 1 bar) behaviors of cement paste compacts incorporated with 30% of dolomite powder at low water to cement ratio (0.15) under different curing regimes. They found that dolomite powder increased nucleation points and enhanced the generation of hydration and carbonation products. However, the impacts of dolomite mixing ratios, particle sizes, curing pressures, and temperatures on carbonation have not been elucidated. The behavior of dolomite in carbonation curing concrete should be further investigated.

To evaluate the feasibility of dolomite as an SCM in carbonation curing concrete, this work investigated cement pastes with different dolomite mixing ratios and particle sizes under various water to solid ratios, pressures (0.5-2.5 MPa) and temperatures ($20-60^{\circ}$ C). The effect of dolomite on carbonation and mechanical performance of cement pastes has been systematically studied. To elucidate on the mechanisms of dolomite additive, X-ray diffraction (XRD), mercury intrusion porosimetry (MIP), and scanning electron microscope (SEM) tests were performed to study the microstructure changes before and after carbonation.

Materials and Methods

Materials

The cement used in this study was the Portland cement P.O.42.5 from Shanghai Conch Cement Co., Ltd (Shanghai, China). Dolomite powders were obtained from Lingshou county in Hebei Province in China. Table 1 gives the elemental composition of the cement determined by X-ray Fluorescence spectroscopy (XRF) test. The dolomite powders were ground and sieved into three groups with different particle sizes, small particle size group (average 10 μ m), middle particle size group (average 100 μ m) and large particle size group (average 100 μ m). Middle size dolomite (average 50 μ m) was used unless specified.

Samples preparation

Cement, oven-dried dolomite, and water were mixed in different ratios (Table 2) in a vertical mixer for 1-2 min. The initial water to solids ratio (w/s) referred to the ratio of the mass of water to the mass of solid powders (cement and dolomite). Then, mixtures were poured into steel molds ($20 \times 20 \times 20$ mm) and covered

Table 1. Co	• 1. Composition of materials (wt.%) as determined by XRF analysis.						alysis.			
Materials	CaO	SiO ₂	SO ₃	Fe ₂ O ₃	Al ₂ O ₃	K ₂ O	Ti ₂ O	SrO	MgO	Others
OPC	62.911	21.886	4.649	4.432	4.531	0.934	0.325	0.132	/	0.200
Dolomite	55.290	0.500	0.010	0.030	0.160	0.010	/	/	37.750	6.250

Table 2. Pastes mix designs and carbonation curing conditions.

Samples	Mixing ratios	Initial w/s ratio	Standard carbonation curing condition
DPC-5%	Cement : Dolomite (50 μ m) = 100 : 5	0.4	w/s = 0.175;
DPC-15%	Cement : Dolomite (50 μ m) = 100 : 15	0.4	$T = 40 \pm 2^{\circ} \text{C},$
DPC-25%	Cement : Dolomite (50 μ m) = 100 : 25	0.4	P = 1.5 MPa; RH > 98%
DPC-15%-P10	Cement : Dolomite (10 μ m) = 100 : 15	0.4	
DPC-15%-P25	Cement : Dolomite (25 μ m) = 100 : 15	0.4	
DPC-15%-P50	Cement : Dolomite (50 μ m) = 100 : 15	0.4	
DPC-15%-P100	Cement : Dolomite (100 μ m) = 100 : 15	0.4	

with plastic wrap to prepare cubic samples. After a 24 h hydration process at ambient conditions $(25 \pm 2^{\circ}C, 80\%-90\% \text{ RH})$, the pastes were demolded and dried in the oven at 60°C for 2–5 h to the certain w/s to provide diffusion pathways. The w/s of the samples was controlled to 0.175, except for the tests with different w/s (from 0.12–0.364). It should be noted that small size samples were used here considering the size of the reactor. This may lead to a greater CO₂ uptake capacity and compressive strength compared to larger size samples due to the diffusion depth of the carbonation curing process,²¹ but it does not affect the intercomparison of the dolomite additive Portland cement (DPC) samples.

Carbonation curing

The standard carbonation curing condition is shown in Table 2. To investigate the effects of pressure and temperature, samples were cured at different pressures (0.5, 1.5, and 2.5 MPa) and temperatures (20, 30, 40, 50, and 60° C). Setup and operating procedures were as previously described.²² The CO₂ uptake ratio ω was defined as the ratio of the mass of reacted CO₂ to the mass of the whole sample before carbonation curing, which can be expressed as:

$$\omega = \frac{m_{\rm CO_2}}{m_{\rm initial}} \times 100\%,\tag{1}$$

whereby m_{CO_2} is the mass of absorbed CO₂ calculated through the manometric method using pressure and

temperature data from the experimental setup for carbonation curing,¹³ while m_{initial} is the initial mass of the whole sample before carbonation curing. For comparisons, reference groups were placed at ambient conditions for the same duration.

The CO₂ uptake ratio after normalization $\omega_{\text{normalized}}$ was expressed as shown in Equation (2):

$$\omega_{\text{normalized}} = \frac{\omega}{m_{\text{cement}}} \times (m_{\text{cement}} + m_{\text{dolomite}}), \quad (2)$$

whereby m_{cement} and m_{dolomite} are the masses of cement and dolomite in raw materials, respectively.

Characterization

Mercury intrusion porosimetry (MIP) tests were performed on a Micromeritics AutoPore IV 9510 analyzer. Slices (2-5 mm in diameter) were obtained from the surface of the paste and dried in the oven at 105°C to completely remove the water from the slices before mercury intrusion. XRD was performed on a PANalytical X-ray Diffractometer using CuKa-radiation at 45 kV and 40 mA. Patterns were obtained by step scanning from 10 to 70° 2θ . The Jade 6.0 software and Powder Diffraction File 2004 package were used for analysis. Powders for tests were obtained from the surface of the samples where the carbonation reaction was completely executed after which they were ground into fine powders. Morphology and microstructure of different samples were analyzed by field emission scanning electron microscopy (SU-8010,

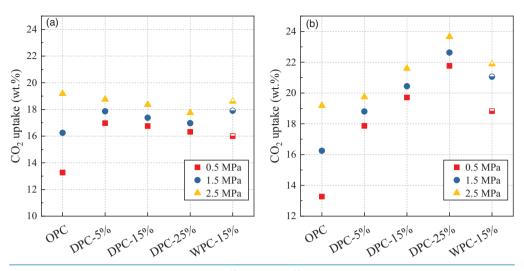


Figure 1. CO_2 uptake ratio of DPC, OPC,¹³ and WPC²² samples (a) before normalization and (b) after normalization to cement mass in the sample

Hitachi). The slices of samples were coated by a thin gold layer for 60 s before SEM examination. Compressive strength test results were measured after carbonation curing or natural curing at the standard ages of 7 days. The compressive strength testing machine (60 tons) was obtained from Jinan MTS Test Technology Co., Ltd (Shandong, China). Each result is presented as the average value of three samples in the same mixing and curing conditions.

Results and Discussion

CO₂ uptake capacity

Effects of mixing ratios and pressures

Figure 1(a) shows the effects of different dolomite mixing ratios and pressures of DPC samples after carbonation curing. For better comparisons, results of pure OPC pastes and wollastonite-Portland cement (WPC) pastes (85 wt.% OPC and 15 wt.% wollastonite [CaSiO₃]) under similar carbonation curing condition $(40^{\circ}\text{C}, 0.16 \text{ w/b}, 0.5-2.5 \text{ MPa})$ in our previous study were also included.²² Compared to the pure OPC without dolomite, all DPC samples with different mixing ratios under the curing pressure of 2.5 MPa exhibited a lower CO₂ uptake capacity. When more dolomite was mixed in the pastes, the CO₂ uptake ratio exhibited a downward trend, with DPC-5%, DPC-15%, and DPC-25% samples dropping to 18.8%, 18.4%, and 17.7%, respectively. Since dolomite is an inert mineral, which cannot react with CO₂, increasing dolomite proportions may result in reduced absolute CO₂ uptake capacity. However, under 0.5 and 1.5 MPa, the CO₂ uptake capacities of DPC samples were higher than those of pure OPC samples, although pure OPC samples contained more carbonation active components. This was quite different from the situation under 2.5 MPa. It is possibly because the enhancement of dolomite on gas diffusion was more obvious under lower pressures than under high pressures. According to Vincent Dutzer et al. on Fick's second law,²³ gas diffusion was not only associated with the pressures, but was also associated with gaseous pathways and the microstructure of cement paste. Inert additives, such as dolomite, provide extra gaseous pathways for CO₂ diffusion¹³ and the effects of additives on gas diffusion enhancement were more significant under lower pressures. This will be discussed in detail below.

To compare the enhancement of different mixing ratios of additives on cement carbonation and eliminate the effects of inert dolomite powders, values of CO_2 uptake ratios were normalized to the cement mass as shown in Figure 1(b). Here, the dolomite, whose main component is $CaMg(CO_3)_2$, is considered to be an inert powder. To simplify the calculation, unevenness of the quality of water combined with cement powder and dolomite powder were not taken into consideration. Therefore, the ratio of different powders in the initial dry raw materials was used for normalization (as shown in Equation (2)). Although the wollastonite in WPC samples can also react with CO_2 , which has been illustrated in our previous study,²² in this study, the WPC samples were normalized to the cement mass for better comparisons with OPC and DPC samples. However, it should be noted that because wollastonite can also react with CO_2 , the normalized CO_2 uptake value of WPC is overestimated.

Unlike the decreasing trend before normalization, the CO₂ uptake ratios after normalization exhibited an obvious increasing trend with increasing dolomite mixing ratios, reaching 23.6 wt.% in DPC-25% samples under 2.5 MPa curing pressures. Although this value is smaller than the theoretical CO₂ uptake capacity (47.7 wt.%) estimated by the Steinour formula, 24 it is significantly higher than the CO₂ uptake value of pure OPC samples (19.2 wt.%). Moreover, compared to pure OPC samples, the CO₂ uptake ratios of DPC-25% after normalization increased by 63.9% under 0.5 MPa and only increased by 22.9% under 2.5 MPa. As described above, dolomite can provide extra gaseous pathways by supporting internal microchannels, allowing more active components to participate in the carbonation reaction. However, it is worth noting that in the case of this experimental study, the two effects on gas diffusion, dolomite addition, and CO₂ pressure are interrelated. The promotion effect of dolomite addition was more significant at lower pressure. It is possibly because higher pressure makes the CO₂ diffusion region closer to the entire sample cross-section, that is, the carbonation reaction depth is closer to the maximum depth of the sample, and therefore the promotion effect of dolomite addition is diminished. Moreover, although the additives of dolomite and wollastonite exhibited similar enhancement effects on CO_2 uptake, they may have different mechanisms. Wollastonite particles can directly react with CO₂, which can increase the degree of reaction.^{22,25} Dolomite cannot react with CO₂ but possibly could act as a seed crystal like calcite in the carbonation process to guide the precipitation of carbonation products on specific crystal surface.²⁶ The final apparent CO₂ uptake value was a comprehensive result of different promotion effects.

Effects of temperature and water to solids ratio

Figure 2 shows CO_2 uptake of DPC-15% samples during 2 h of carbonation at different temperatures. About 80% of the total CO_2 mineralization occurred within the first 30–40 min regardless of curing temperatures. Moreover, the total CO_2 capacity increased with increasing temperatures. As curing

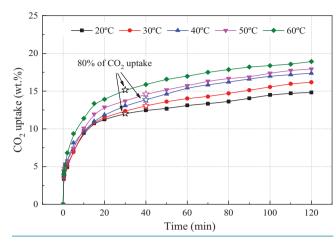


Figure 2. CO₂ uptake capacity of DPC-15% samples under different temperatures

temperatures increased from 20 to 60° C, the total CO₂ uptake for DPC-15% samples increased from 14.8 to 18.9 wt.%. It is possible that higher temperatures promote CO₂ diffusion rates and chemical reaction rates, especially in the initial stage as the product layer of carbonation reaction is not yet fully developed.

In addition, the trend of CO₂ uptake ratio for DPC samples was a little different from that of pure OPC pastes investigated in our previous study.¹³ OPC samples exhibited the highest CO₂ uptake capacity under optimum temperature of 40°C, while DPC samples exhibited a continuous increase in CO₂ uptake capacity as temperature increased in the range of 60°C. The effect of temperature is usually two-sided.^{13,27} On one hand, higher temperatures promote CO₂ diffusion and ion leaching, while on the other hand, higher temperatures are not conducive to the dissolution of CO_2 gas in the pore solution due to water evaporation and thermodynamics. Higher optimum w/s of DPC samples, as described in the next paragraph, may result in a higher optimum temperature for CO₂ uptake by DPC samples.

Water to solids ratio (w/s) plays a vital role in carbonation process (Figure 3). DPC-15% samples had an optimum w/s ratio of 0.20 for best CO₂ uptake capacity, which was higher than that of pure OPC samples (w/s = 0.14).¹³ The optimal w/s value is the balance of the two-side effect of pore water.^{22,28} On one hand, internal pore water provided the necessary pore solution environment for CO₂ solubility, ion leaching, and carbonation products precipitation. On the other hand, excess pore water would block CO₂ diffusion. The difference in optimum w/s ratio between DPC and

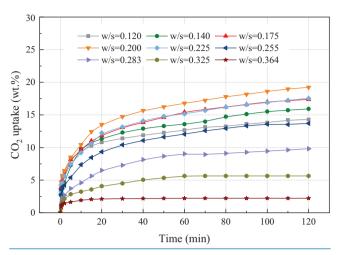
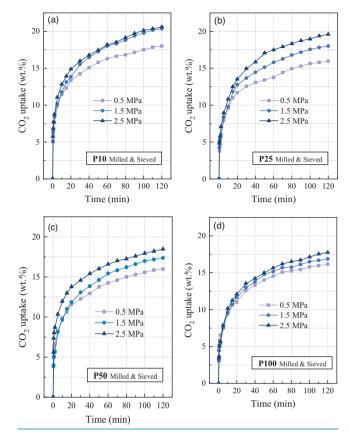


Figure 3. CO_2 uptake of DPC-15% samples with different w/s ratios

OPC is possibly due to improved pore volumes after the dolomite mixing, which provided additional space for solution environment and CO_2 diffusion. Therefore, the balance moves to the higher w/s value.

Effect of dolomite particle sizes

As shown in Figure 4, after 2 h of carbonation curing, DPC samples exhibited decreased CO_2 uptake capacity, from 20.6 to 17.8 wt.%, as dolomite particle sizes increased from 10 to 100 μ m. Although smaller dolomite particles could densely fill the space and lead to reduced pore volumes, it resulted in the highest CO_2 uptake ratio of 20.6 wt.%, even higher than pure OPC samples of 19.2 wt.% at 2.5 MPa pressure. This can possibly be explained by the dilution effect. Dolomite with an average particle size of 10 μ m is similar to





OPC cement particles with an average particle size of about 7 μ m and therefore has a better dilution effect and provides more contact surface for carbonated precipitation, as shown in Figure 5. Another possible

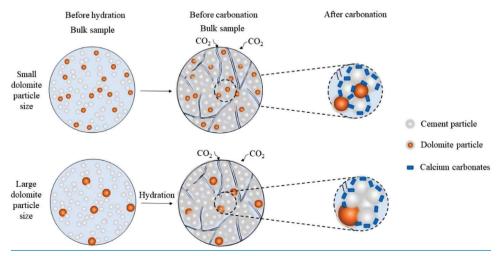


Figure 5. The schematic diagram of the effect of particle size on the carbonation curing

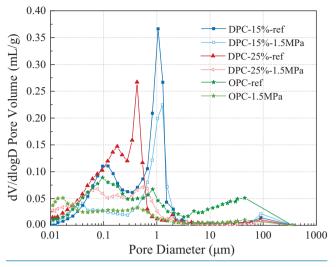


Figure 6. Changes in pore size distributions of OPC and DPC samples assessed by MIP

reason is that dolomite provides nucleation seeds by forming calcite through the dedolomitization reactions, and smaller particles provide more nucleation sites for carbonates. However, these hypotheses need to be tested.

Structure and mineral phase evolutions MIP tests

Pore size distributions of DPC samples with different mixing ratios are shown in Figure 6. Compared to OPC samples, dolomite mixing significantly increased the pores (<2 μ m) of DPC reference samples. OPC reference samples exhibited a gentle distribution with several small peaks, implying a broader distribution of pore sizes. However, large holes (10–100 microns) in OPC reference samples was possibly because a small number of samples taken from the OPC reference sample for the MIP test contained large pores (10–100 micron) generated during mixing. Since the pores

associated with carbonation are concentrated in the range of 0.1–10 μ m, this did not affect the analysis of the pore changes during carbonation. DPC reference samples had one or two sharp peaks, and most of the increased pores were mainly distributed within certain pore size ranges. DPC-15% samples significantly increased pore volumes in the 0.7–1.1 μ m range, while DPC-25% samples exhibited an increase in the 0.2–0.6 μ m range. These findings imply that dolomite mixing provided more micro pores and pathways for CO₂ gaseous transport, as discussed in section 3.1. In addition, it is interesting to note that as dolomite ratio increased from 15% to 25%, peak pore sizes of the sample before carbonation moved toward smaller pore sizes, which is possibly associated with structural change of hydration gel system. The higher mixing ratios may bring a more significant dilution effect and reduce the interface pores between inert mineral and cement matrix. Meanwhile, carbonation process significantly reduced porosity and pore volumes, and increased bulk density in all carbonated samples (Table 3). This is in line with the common understanding that the carbonation reaction has a densification effect.

Mineral phase evolution

Figure 7(a) shows mineral phase changes of DPC samples under different pressures before and after carbonation. The peaks of calcium hydroxide, nonhydrated C_3S , C_2S , and little ettringite existed in the DPC-15% reference samples. The dolomite CaMg(CO₃)₂, whose peak at around 31°, was also observed in the reference samples. After 2 h of carbonation, peaks of calcium hydroxide disappeared, implying that almost all calcium hydroxide had been consumed in the carbonation process. The peaks of C_3S and C_2S were slightly decreased. At around 29°, peak of calcite (CaCO₃) increased significantly after

Table 3. MIP test results of OPC, DPC-15%, and DPC-25% samples.							
	Porosity	Total intrusion volume (ml/g)	Bulk density (g/ml)				
OPC-ref	38.43%	0.2581	1.4887				
OPC-1.5	20.90%	0.1065	1.9625				
DPC-15%-ref	35.13%	0.2149	1.6350				
DPC-15%-1.5MPa	25.50%	0.1386	1.8398				
DPC-25%-ref	38.31%	0.2085	1.8377				
DPC-25%-1.5MPa	24.62%	0.1305	1.8865				



 CaMg(CO₃)₂ ♦ C₃S - Ca₃SiO₉ A C2S - Ca2SiO4 + CH - Ca(OH), Calcite- CaCO₃
 Ettringite Ouartz - SiO 2.5MPa 1.5MPa 0.5MPa Ref 20 30 40 50 60 2θ (°)

Figure 7. Mineral phase of DPC-15% samples under different pressures (0–2.5 MPa, 40°C, 0.175 w/s)

carbonation, and it increased with increasing curing pressure. The major carbonation product in carbonated DPC-15% samples was calcite. Unreacted C_2S and C_3S still existed after carbonation at around 32° and 34°. It should be noted that intensities of CaMg(CO₃)₂ peaks remained almost the same as those of reference samples, which verified that dolomite almost did not react with CO_2 in carbonation process.

SEM characterization of microstructure changes

As shown in Figure 8a and c, for DPC-15% and DPC-25% samples before carbonation curing, dolomite particles could be easily found with the smooth surface and dense structure, which was hardly hydrated. There is a clear interface between dolomite particles and hydration products before carbonation, with a lot of pores in the interface area. Therefore, the CO_2 gas could more easily diffuse into the interior of the samples through these pathways. Some needle-like ettringite smaller than 2 μ m could also be observed in reference samples, which grew in the gap between the particles. However, for the DPC samples after the carbonation curing process (Figure 8b and d), ettringite disappeared, while a large amount of calcite and silica gel were formed, making the microstructure denser. It should be noted that carbonation products, such as calcite, precipitated on the pores and filled the interface between dolomite and amorphous particles. In addition, DPC-25% samples possessed more

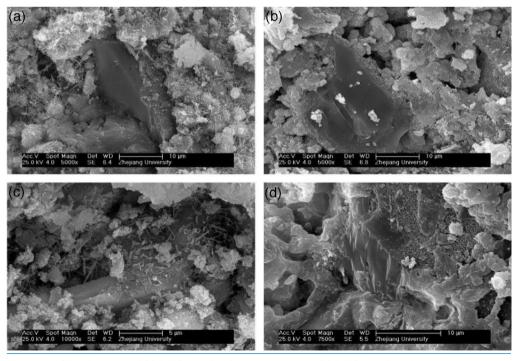


Figure 8. SEM images of (a) DPC-15% reference samples (b) DPC-15% carbonated samples (c) DPC-25% reference samples, and (d) DPC-25% carbonated samples (40°C, 0.175 w/s, 1.5 MPa)

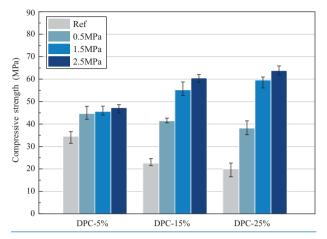


Figure 9. Compressive strength test results of DPC-5%, 15%, and 25% samples under different curing pressures (40° C, 0.175 w/s)

uniform structures than DPC-15% samples, which may have resulted in higher compressive strength.

Mechanical properties

Figure 9 shows compressive strength values of DPC samples with different mixing ratios. For the reference groups, compressive strengths decreased sharply as more dolomite was mixed. The average compressive strength of DPC-25% reference samples was only about 20 MPa, much lower than that of the pure OPC reference samples (about 46 MPa in the previous study).¹³ However, the trend of carbonated DPC samples after 2-h curing exhibited exactly the opposite trend. Among all the DPC samples, DPC-25% carbonated samples exhibited the highest compressive strength among, which could reach 63.8 at 2.5 MPa curing pressure, over three times higher than that of the reference sample. DPC-25% samples had the highest compressive strength which is attributed to higher carbonation conversion degrees (as shown in Figure 1b) and a denser structure (as shown in Figure 8).

Regarding the effects of curing pressures, compressive strength values increased at higher curing pressures. In addition, the highest mixing ratio (25%) resulted in the most significant improvement from reference to 2.5 MPa. As the curing pressure increased from 0.5 to 2.5 MPa, compressive strength of DPC-25% increased by 67.0% while that of DPC-15% increased by 46.1% and DPC-5% increased by 5.8%, only. This is probably because the densification effect of carbonation products for DPC-15% samples was relatively insignificant compared to that of DPC-25% samples with high porosities (as shown in Figure 6).

Conclusions

In this study, the effects of reaction conditions on performance of cement pastes have been comprehensively investigated, including mixing ratios, water to solids ratios (w/b), temperatures, CO_2 pressures, and particle sizes. After normalization to carbonation active components, CO₂ uptake capacity significantly increased with increasing dolomite mixing ratios, and the maximum value reached 23.6 wt.%, which was 23% higher than that of pure OPC samples under the same condition. The optimum w/s ratio for CO₂ uptake capacity of DPC-15% samples was about 0.20, while the optimum temperature was equal to 60°C or higher than 60°C. In addition, CO₂ uptake capacity of DPC samples increased with higher CO₂ curing pressures and finer particle sizes. Although smaller dolomite particles could densely fill the space and lead to reduced pore volumes, it resulted in the highest CO₂ uptake ratio of 20.6 wt.%, even higher than pure OPC samples of 19.2 wt.% at 2.5 MPa pressure. Dolomite with an average particle size of 10 μ m is similar to OPC cement particles with an average particle size of about 7 μ m and therefore has a better dilution effect. The intrinsic mechanism of the influence of the addition of dolomite lies in the dilution effect on the cement hydration products and the provision of contact surface. Although the compressive strength of the DPC reference samples was lower than that of the pure OPC samples, the carbonation precipitation allowed the DPC sample to exhibit a more uniform structure, further strengthening the compressive strength of the DPC sample, thereby contributing to the highest compressive strength values (63.8 MPa) of DPC-25% samples, over three times higher than that of DPC-25% reference sample. These findings imply the possible feasibility of dolomite as an additive in carbonation cured building materials.

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