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Carbonate binders: Reaction kinetics, strength and microstructure

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Abstract

This study focussed on the synthesis of calcium carbonate binders, in situ, from the reaction between hydrated lime and carbon dioxide (CO_2). The aim was to establish the characteristics of the calcium carbonate binders that are associated with its strength, which was considered as an indicator of binder performance. The role of the parameters that are known to play an important part in the kinetics of hydrated lime carbonation processes, in changing the strength of a binder was examined in detail.

The parameters identified were CO_2 gas pressure, exposure time and the initial degree of compaction of raw material. All hydrated lime mixtures were prepared at a constant water/solid ratio of 0.25. The hydrated lime compacts made at a range of compaction water/solid ratio (W/S) of 0.25. The hydrated lime compacts made at a range of compaction pressures (0.65–6.0 MPa) were exposed to different CO_2 gas pressures (ambient to 2 MPa) for different periods of time. The resulting products were tested for the amount of $Ca(OH)_2$ that had converted to carbonate, and for compressive strength. A microstructural analysis of the products was carried out using scanning electron microscopy.

The rate of $Ca(OH)_2$ conversion to carbonate seemed to be enhanced with increasing gas pressure, but it decreased with increasing compaction of the initial mixture. It was revealed that the crystalline state and the morphology of the carbonate formed, rather than the degree of conversion of calcium hydroxide into carbonate, is highly critical to the strength of the binder. The study concluded that in the development of calcium carbonate binder, it is important to meet the experimental conditions that favour the crystallisation of calcium carbonate.

Keywords: Kinetics; Microstructure; Mechanical properties; $CaCO_3$; Composite

1. Introduction

There has been no use of carbonate binders in the recent history of building and construction. This is possibly due to the rapid development of Portland cement technology. However, because of the proven mechanical properties associated with naturally occurring carbonate binders [1], and also the consumption of carbon dioxide (CO_2) in their formation, the development of carbonate binders artificially can be expected to have a great impact on the sustainable growth of the construction industry in the future.

In the development of any binder in situ, it is necessary to understand the underlying chemistry of the processes involved, the characteristics that are associated with the performance of the binder, and the factors, if any, which control these characteristics. The establishment of these criteria is important in developing carbonate binders that are suitable for any industrial application.

Some knowledge of the key characteristics associated with the carbonate binders to be developed, can be drawn from the geological evolution of carbonate binders which occurs in nature. Limestone, beachrock, dolomite, magnesite and marble are a few of the naturally occurring calcium and/or magnesium carbonate binders. Although these formations occur over thousands of years, these products are proven to possess superior chemical and physical

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properties. The superior qualities of these geological materials are attributed to their compacted, interlocked and well-developed crystalline calcium and/or magnesium carbonate microstructures. Therefore, in developing carbonate binders in situ, obtaining microstructures that replicate or to a certain extent are similar to those of nature, can be one of the main tasks faced.

1.1. Calcium carbonate binders from carbonation of hydrated lime

One of the ways to obtain calcium carbonate artificially is by a simple reaction between calcium hydroxide and carbon dioxide, a concept used in the building industry since ancient times. Though not much progress has occurred in the recent past, on the formation of carbonate binders directly from hydrated lime, the concept is presently indirectly used in a number of other applications. Carbonation of cement, where calcium carbonate is formed from the reaction between cement hydrates (calcium hydroxide and calcium silicate hydrates) and CO_2, is found to be beneficial in strengthening the long-term durability of glass fibre concrete [2,3] and in the production of cement-bonded particleboards and fibreboards [4].

Although there has not been much progress in industrial applications of carbonate binders obtained directly from hydrated lime carbonation, the kinetics of this process has been discussed in the literature [5–8].

The chemical reaction for the carbonation of hydrated lime is

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O + 74\,KJ$$

The overall reaction is exothermic and proceeds in two stages: the CO_2 diffusion process, followed by the formation of calcium carbonate.

The process occurs through a dissolution–precipitation mechanism and therefore the presence of water is an important requirement. However, the diffusion of CO_2 in water is much slower than in air, and therefore the reaction can be retarded if the material is saturated with water. The formation of carbonate itself is associated with an accumulation of water and an increase in solid volume. The newly formed $CaCO_3$ tends to be accommodated in the internal pores, thus reducing permeability. Therefore, the amount of water contained and the initial porosity of the system undoubtedly has an impact on the rate of the CO_2 diffusion process. The progress of the second step (the formation of carbonate) depends mainly on the amount of $Ca(OH)_2$ available and its solubility, which depends on particle size and crystal shape (8) and the amount of CO_2 dissolved in water, which is dependent on the initial CO_2 concentration and pressure (5, 7). In general, the dissolution of CO_2 in water increases with increasing CO_2 concentration and pressure. Solubility is also a function of temperature. Increasing temperature decreases the solubility of both $Ca(OH)_2$ and CO_2 in water. Therefore, the rate of the overall hydrated lime carbonation process depends on several factors such as CO_2 concentration and pressure, moisture content, temperature, relative humidity and thickness of the specimen. The mechanism of cementation, as outlined by Moorehead [5], involves the precipitation of calcium carbonate as amorphous or microcrystalline structures form first in smaller capillaries, with subsequent expansion into the larger pores and reducing permeability of the compact.

This study aimed to establish the conditions required to develop high strength calcium carbonate binders using hydrated lime and carbon dioxide. It also aimed to relate the performance of the binder, as reflected by compressive strength, to the kinetics of the carbonation process and the morphological changes of calcium carbonate. The relevance of the three parameters (CO_2 gas pressure, exposure time and the degree of initial compaction) that control the overall kinetics of the carbonation process to the strength of the binder was examined in detail. The initial degree of compaction, which is linked to the permeability of the compact, was changed by applying different compaction pressures during the preparation of the hydrated lime compacts.

2. Materials and methods

Hydrated lime, was used as the $Ca(OH)_2$ source. The samples were kept in a sealed container to minimise carbonation in storage. The chemical analysis of the hydrated lime, obtained by X-ray fluorescence and expressed as oxides, is shown in Table 1. Industrial grade CO_2 gas was used throughout the work.

The water/solid ratio (W/S) by mass of the initial compacts was kept constant at 0.25. The parameters used in the

Table 1
Chemical composition (%) of hydrated lime

Al_2O_3	CaO	Fe_2O_3	K_2O	MgO	Na_2O	SiO_2	Other oxides	Loss on ignition
0.57	71.3	0.23	0.22	0.59	0.08	1.3	n.d	25.4

Table 2
Mix formulations

Experiment (mix formulation)	Parameters			Variable parameter
	Compaction pressure (MPa)	CO_2 pressure (MPa)	CO_2 exposure time[a] (h)	
1	0.65	0.1,1,2	0, 0.5, 1, 2	Gas pressure and time
2	2	0.1,1,2	0, 0.5, 1, 2, 6	Gas pressure and time
3	0.65, 2, 6	2	0, 0.5, 1, 2, 6	Compaction pressure and time

[a] The compacts with zero exposure time are considered as controls in the experiments.

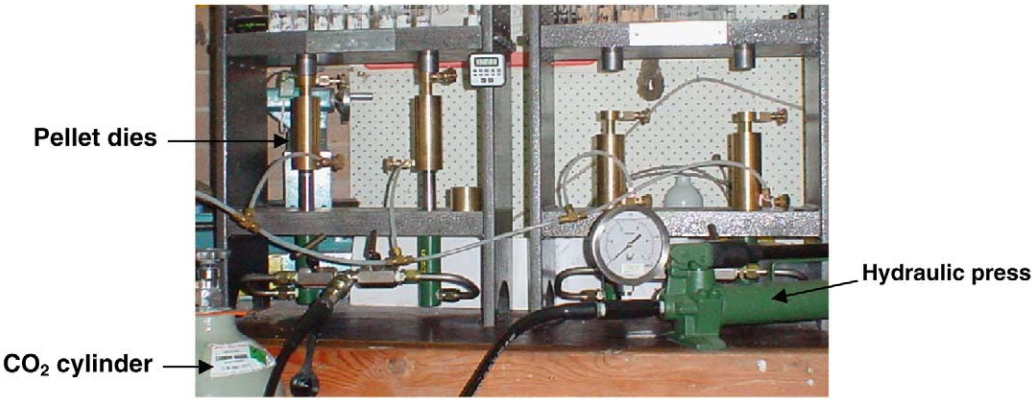

Fig. 1. The equipment used to prepare compacts and to expose them to CO_2 pressure.

study were compaction pressure, CO_2 gas pressure and exposure time. The experiments (different mix formulations) were designed to have different variables, as shown in Table 2. All experiments were carried out at room temperature (23 ± 1 °C).

The equipment used to prepare samples at the required compaction pressures and to subsequently treat them with pressured CO_2 is shown in Fig. 1. (The design of the equipment was based on an earlier model developed by Moorehead [9].) It consists of four cells (pellet dies) with internal diameters of 16 mm and a hand-operated hydraulic press. CO_2 gas at the required pressure can be delivered to the die from a gas cylinder.

The hydrated lime was hand mixed with distilled water at a W/S of 0.25, and a known weight of this mixture was introduced into the pellet die and subsequently compacted at the respective mechanical pressure for 2 min. (The amount of sample was chosen so that all the compacts had a similar height of 25 mm after compaction. Therefore, the densities of the initial compacts at 0.65, 2 and 6 MPa were 990, 1390 and 1590 kg/m^3, respectively.) The mechanical pressure was released before exposing the sample to CO_2 gas, which was delivered from the cylinder at a controlled pressure. Each compact was flushed with CO_2 for about 1 min before closing the top outlet of the cell to ensure a 100% CO_2 environment in the system throughout the reaction time. After exposing the compacts for different times, they were taken out and kept at 23 ± 2 °C and 50 ± 5% RH for 24 h. With each mix formulation, a few samples were also prepared and were kept in the above conditions but without exposure to the CO_2 pressures studied. These samples were used as controls. In the results, the controls are presented as the compacts exposed to zero time at the relevant gas pressures.

The compacts were tested for compressive strength and the percentage of $Ca(OH)_2$ conversion. For each compressive strength measurement, the average value of three or four compacts was considered. The morphology, microstructure and elemental composition of the resulting products were examined using scanning electron microscopy (SEM) in conjunction with electron dispersive X-ray analysis (EDAX). The amounts of $Ca(OH)_2$ that had reacted with CO_2 in the mixtures at different exposure times were determined by a gravimetric method [10]. (After exposure to CO_2, the compacts were dried under vacuum, weighed and then placed at 500 °C for a few hours until a constant weight was attained. The weight loss at 500 °C was used to calculate the amount of $Ca(OH)_2$ left in the compacts. A calibration test carried out with mixtures of known quantities of $Ca(OH)_2$ and $CaCO_3$ has shown that this method can be used to determine amount of $Ca(OH)_2$ left in the compacts with a high level of accuracy.)

3. Results

3.1. $Ca(OH)_2$ conversion to carbonate and compressive strength development

The $Ca(OH)_2$ conversion and compressive strength development, with respect to CO_2 exposure time, of mix formulation 1 (W/S 0.25, 0.65 MPa) at 0.1, 1 and 2 MPa gas pressure, are shown in Fig. 2. It can be seen that the time taken to convert $Ca(OH)_2$ decreased with increasing CO_2 pressure. In other words, the rate of hydrated lime conversion increased with increasing CO_2 pressure. At 2 MPa gas pressure, 80% of conversion had occurred within 30 min of exposure and remained nearly constant thereafter irrespective of the exposure time. For the same amount of conversion, the time taken at 1 MPa was about 60 min. At 0.1 MPa gas pressure, only 60% conversion was achieved, even after 2 h of exposure. Compared to the controls, there was a slight increase (about 2 times) in strength at 0.1 MPa with exposure time, but no strength improvement at the other gas pressures.

Fig. 3 shows the $Ca(OH)_2$ conversion and compressive strength development, with respect to CO_2 exposure time, of mix formulation 2 (W/S 0.25, 2 MPa) at 0.1, 1 and 2 MPa gas pressure. These results also show that increasing CO_2 pressure increased the amount of $Ca(OH)_2$ conversion at a given time. However, compared with 0.65 MPa compaction (Fig. 2), at each CO_2 pressure, the time taken for similar $Ca(OH)_2$ conversion was longer at 2 MPa compaction.

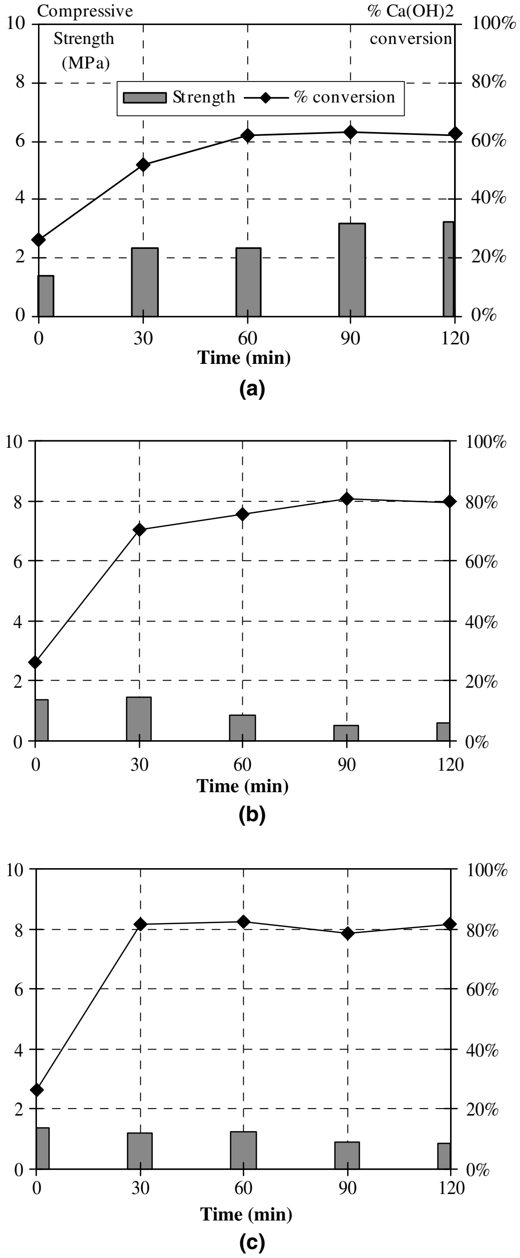

Fig. 2. $Ca(OH)_2$ conversion and compressive strength development of compacts made at W/S 0.25 and 0.65 MPa compaction pressure: (a) 0.1 MPa CO_2 pressure, (b) 1 MPa CO_2 pressure and (c) 2 MPa CO_2 pressure.

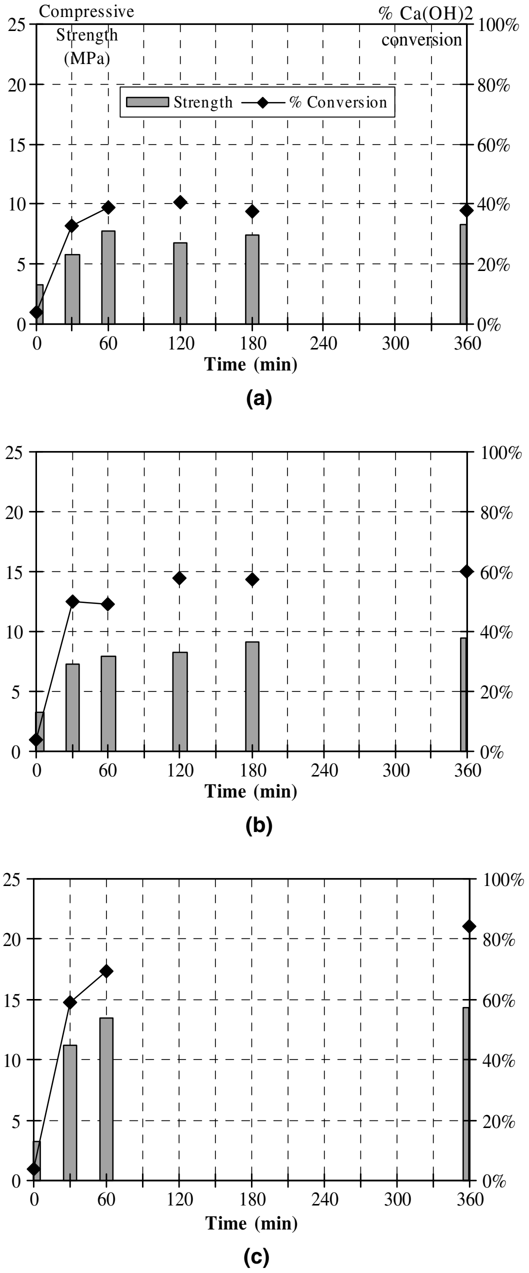

Fig. 3. $Ca(OH)_2$ conversion and compressive strength development of compacts made at W/S 0.25 and 2 MPa compaction pressure: (a) 0.1 MPa CO_2 pressure, (b) 1 MPa CO_2 pressure and (c) 2 MPa CO_2 pressure.

For example, it took about 6 h of exposure, even at 2 MPa gas pressure, to reach 80% conversion, implying the effect of increasing compaction pressure in decreasing the rate of $Ca(OH)_2$ conversion. The conversion in 6 h of the compacts studied at 0.1 and 1 MPa were 40% and 60%, respectively. Therefore, increasing compaction pressure from 0.65 to 2 MPa decreased the rate of $Ca(OH)_2$ conversion to calcium carbonate.

Unlike the samples compacted at 0.65 MPa (Fig. 2), the samples prepared at 2 MPa compaction showed a considerable improvement in strengths at all CO_2 pressures studied (Fig. 3). After 6 h of exposure, the strength increase compared with the controls was about 2.5, 3 and 5 times for the 0.1, 1 and 2 MPa gas pressures respectively. It has to be noted that this kind of strength improvement was not achieved at 0.65 MPa compaction even with 80% conversion of $Ca(OH)_2$, irrespective of the CO_2 pressure. Therefore, it is apparent that strength characteristics of the carbonate binder are not only related to the amount of calcium carbonate formed, but also some other factor(s) that will be discussed in the next section.

The $Ca(OH)_2$ conversion and compressive strength development, with respect to CO_2 exposure time, for the compact prepared at 6 MPa and exposed to 2 MPa gas

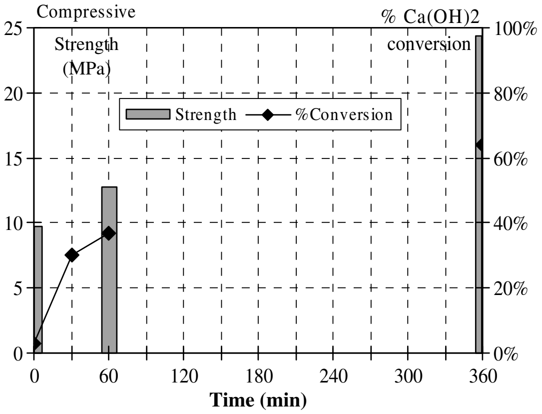

Fig. 4. $Ca(OH)_2$ conversion and compressive strength development of compacts made at W/S 0.25, 6 MPa and exposed to 2 MPa CO_2 pressure.

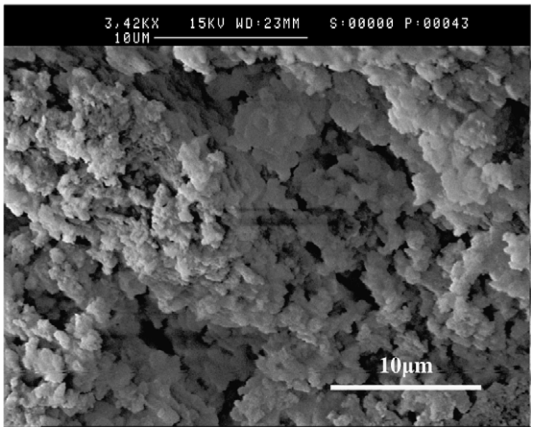

Fig. 5. SEM of the compact made at W/S 0.25, 0.65 MPa and exposed to 2 MPa CO_2 for 0.5 h.

pressure is shown in Fig. 4. For this compact, at 6 h the strength improvement was about 2.5 times that of the control and the $Ca(OH)_2$ conversion was 60%. In comparison, for the samples made at 2 MPa compaction (Fig. 3(c)), the strength improvement was 5 times that of the control, with 80% conversion. The increase in compaction pressure from 2 to 6 MPa lowered the rate of hydrated lime conversion.

These results indicate that it is the combined effect of compaction pressure and CO_2 gas pressure that aids in the achievement of a carbonate binder with optimum properties with respect to both $Ca(OH)_2$ conversion and strength. For the samples studied, the maximum strength improvement (5 times), combined with maximum $Ca(OH)_2$ conversion (80%), was obtained in a shorter time (6 h) using 2 MPa compaction pressure and 2 MPa CO_2 pressure. The density of the respective initial hydrated lime compact was 1390 kg/m^3.

3.2. Microstructure and morphology

In addition to the microstructure of the products, the SEM study was also intended to investigate the role of parameters on the depth of carbonation. The carbonation process of hydrated lime, in general, first takes place at the surface and progresses towards the middle of the compacts. Therefore, the SEM results presented are from different depths of compacts (two depths, 2 mm and 6 mm from the curved surface, were compared in this study).

Different morphologies and crystal development of the formed $CaCO_3$ were obtained, depending on the degree of compaction of the raw material. SEM results also showed that increasing CO_2 pressure enhances the depth of carbonation of hydrated lime compacts.

3.2.1. Compacts prepared at 0.65 MPa compaction pressure

The microstructure of the compacts prepared at 0.65 MPa compaction pressure consisted of uniformly arranged massive globes of microcrystalline $CaCO_3$, very much of amorphous appearance (Fig. 5). At this degree of compaction, this pattern of microstructure was consistent for all gas pressures, exposure times and depth of the compact. As stated previously, with the exception of samples exposed to 0.1 MPa CO_2 pressure, no strength improvements were obtained at 0.65 MPa compaction pressure even at 80% $Ca(OH)_2$ conversion.

3.2.2. Compacts prepared at 2 MPa compaction pressure

With 2 MPa compaction pressure, compared with 0.65 MPa compaction, well-developed crystalline morphologies of calcium carbonate could easily be identified, irrespective of the gas pressure. There were some indications that the size of the crystals changed with the depth of the compact in accordance to the gas pressure and exposure time.

SEMs of the compacts exposed to CO_2 pressure at 0.1 MPa for 6 h and at different depths, are shown in Fig. 6. At a depth of 2 mm (Fig. 6(a)), many developed $CaCO_3$ crystals, even packed in layers, were evident. The conversion of $Ca(OH)_2$ was 40% at this stage, and the strength improvement was 2.5 times that of the control. The microstructure at 6 mm consisted mainly of $Ca(OH)_2$ (Fig. 6(b)) (as analysed by EDAX), implying that the $Ca(OH)_2$ conversion reaction had taken place, predominantly closer to the surface rather than in the middle of the compacts.

Figs. 7(a)–(c) show the microstructure of the compacts exposed to 2 MPa CO_2 pressure, at different exposure times and at different depths. At 0.5 h, the percentage of $Ca(OH)_2$ conversion in these compacts was about 60%, and the microstructure at a depth of 2 mm at this exposure time (Fig. 7(a)) consisted mainly of $CaCO_3$ of clearly well-developed crystalline structures. The crystals were about 2–3 µm in size.

Further exposure to 2 MPa CO_2 pressure remarkably improved the morphology of $CaCO_3$ at a depth of 2 mm (Fig. 7(b)). It consisted of blocks of $CaCO_3$ crystals (about 4 µm in size) that were packed together giving a layered configuration, to a certain extent similar to natural

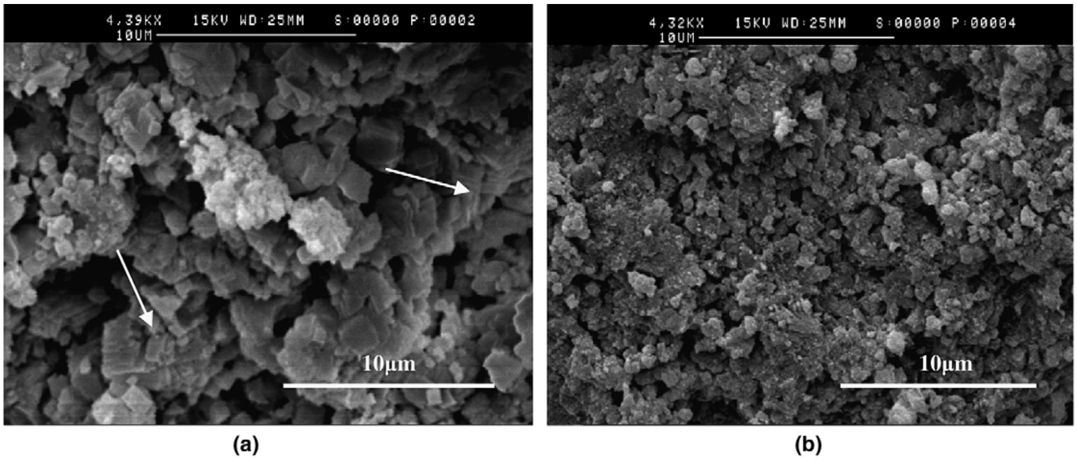

Fig. 6. SEMs of the compact made at W/S 0.25, 2 MPa and exposed to 0.1 MPa CO_2 for 6 h: (a) at 2 mm depth (arrows indicate $CaCO_3$ morphologies); and (b) at 6 mm depth.

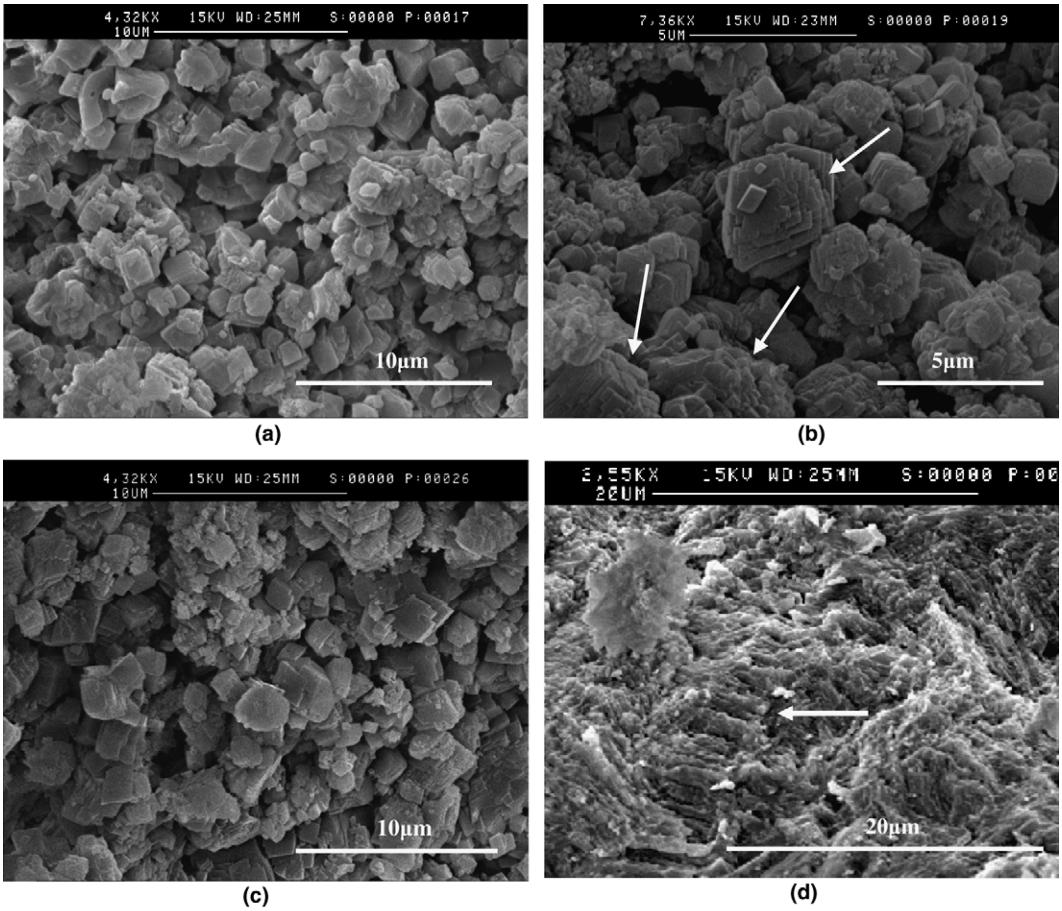

Fig. 7. SEMs of the compact made at W/S 0.25, 2 MPa and exposed to 2 MPa CO_2: (a) 2 mm depth at 0.5 h exposure; (b) 2 mm depth at 6 h exposure (arrows indicate $CaCO_3$ morphologies); (c) 6 mm depth at 6 h exposure; and (d) microstructure of beachrock (arrow indicates layered $CaCO_3$ morphology).

carbonate structures. Fig. 7(d) shows the microstructure of a beachrock sample examined in the laboratory.

$CaCO_3$ crystals smaller in size (2–3 μm), not as developed as above, could be clearly seen in this compact, even at a depth of 6 mm (Fig. 7(c)), in contrast to the samples exposed to 0.1 MPa gas pressure for 6 h. The amount of $Ca(OH)_2$ converted in these compacts was 80% at 2 MPa gas exposure and, as is evident from the microstructural

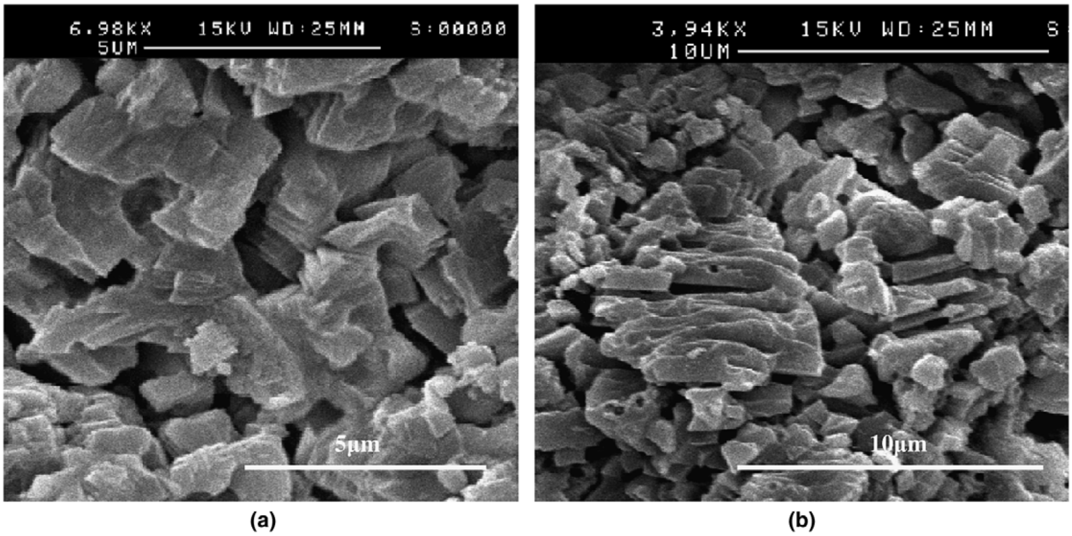

Fig. 8. SEMs of the compact made at W/S 0.25, 6 MPa and exposed to 2 MPa CO_2 for 6 h (at a depth of 2 mm from the surface).

data, the diffusion of CO_2, and hence conversion reaction had taken place even in the middle of the compact, to a considerable extent.

These compacts showed the maximum strength increase, possibly implying that strength is mostly related to the morphology of $CaCO_3$ formed.

3.2.3. Compacts made at 6 MPa compaction pressure

Fig. 8 shows SEMs of the compacts prepared at 6 MPa and exposed to 2 MPa CO_2 pressure. Higher compaction pressure certainly improved the morphology of $CaCO_3$. Not only were there well-developed crystals, but these were also connected and formed a network-like structure (Fig. 8(a)). These compacts also showed elongated calcium carbonate crystal morphologies (Fig. 8(b)). These results also support the argument that although increasing compaction pressure reduces the rate of $Ca(OH)_2$ conversion, it facilitates the formation of well-developed calcium carbonate crystal morphologies.

X-ray diffraction (XRD) analysis of above samples revealed that the type of calcium carbonate polymorph present in these samples is calcite.

4. Discussion

The results presented give a detailed insight into the process of the development of carbonate binders from the carbonation of hydrated lime, with respect to both the rate of $Ca(OH)_2$ conversion and the strength of the binder.

The maximum conversion of $Ca(OH)_2$ obtained in this work was 80%, i.e. full conversion was never attained. This could be due to two reasons. As the reaction proceeds, due to the water produced by the carbonation process itself and the deposition of newly formed $CaCO_3$, the permeability of the compact is reduced, and this in turn has an impact on the diffusion of CO_2. Therefore, reaction is terminated prematurely before all the $Ca(OH)_2$ is converted to $CaCO_3$.

Compaction pressure plays an important role in the rate of $Ca(OH)_2$ conversion. Increasing compaction pressure reduces the permeability of the compact to the gas, and hence the amount of $Ca(OH)_2$ converted into $CaCO_3$ at a given time. As revealed in the results, CO_2 gas pressure also has an effect on the rate of the hydrated lime carbonation process. Increasing gas pressure not only increases the rate of carbonation, but it also facilitates the depth of carbonation.

The main factor revealed in the data is that compressive strength is not necessarily dependent on the degree of $Ca(OH)_2$ transformed to $CaCO_3$. From our SEM observations, it was clear that the morphology or the crystalline state of the $CaCO_3$ formed plays an important role in determining the strength of the binder. The experimental conditions that favour the optimum conversion (80%) did not necessarily guarantee the best binder strength. It is apparent that the conditions, which are a combination of gas pressure and compaction pressure that favour crystallisation of $CaCO_3$ improve the strength of the binder. This is one of the significant outcomes of this study, which has not shown in the past, and a key factor to consider in the development of carbonate binders.

Crystallisation of a compound from a solution is a two step process; nucleation and crystal growth. When a solution reaches the saturation state with respect to relevant ions, nucleation of compounds occur; in the case of $CaCO_3$ the ions are Ca^{2+} and CO_3^{-2}. These nuclei will grow into larger crystals (crystal growth) depending on the availability of ions in the solution. The morphology of the crystals can vary dramatically with the rate of above two processes. Rapid crystallisation processes can often result amorphous-appearing products with no defined morphology [11].

In the lime carbonation process, one factor determining the morphology of calcium carbonate is the rate of its formation. The slower the rate of formation the more defined morphologies will result. This is controlled by both gas and compaction pressures. Increasing gas pressure increases CO_2 diffusion, rate of $Ca(OH)_2$ conversion and thereby calcium carbonate is formed at an increased rate. Increasing compaction pressure decreases the porosity of the hydrated lime compact thereby reducing the rate of diffusion of CO_2 and hence the rate of carbonate formation. This eventually results more defined crystalline morphologies of calcium carbonate. The impact of porosity and pore size distribution of the initial hydrated lime compacts on the crystallisation of calcium carbonate is currently being investigated.

Although the conclusions drawn from this work are limited to a certain sample size, they certainly give some indication of the effects of different experimental parameters on the development of carbonate binders from hydrated lime carbonation, and the information evaluated in this study will undoubtedly help the further development of carbonate binders. The development of building products such as flat sheets can be identified as one area of carbonate binder application.

5. Conclusions

The following conclusions can be drawn from this study:

- In the lime carbonation process, it is not the amount of $Ca(OH)_2$ converted (carbonate formed) that is the main factor controlling the strength of the binder, but the morphology of the carbonate. Well-developed crystalline structures and crystal habits of carbonate give better binder performances.
- The problems associated with low permeability of the hydrated lime compact towards CO_2 diffusion can be overcome using high CO_2 pressure, which facilitates the reaction even in the middle of the compact, resulting in near-maximum conversion of $Ca(OH)_2$ to calcium carbonate.
- Calcium carbonate microstructures giving high strengths can be obtained in situ, from the lime carbonation process, under controlled experimental conditions, within reasonably shorter times (hours). For the size of the sample investigated in this study, the maximum strength improvement (5 times), combined with maximum $Ca(OH)_2$ conversion (80%), was obtained in 6 h using 2 MPa compaction pressure and 20 atm CO_2 pressure.

References

[1] Meldrum FC. Calcium carbonate in biomineralisation and biomimetic chemistry. Int Mater Rev 2003;48(3):187–224.

[2] Jones RH, Jr, Cement treated with high pressure CO_2, United States Patent 5,650,562, 1997.

[3] Knopf FC, Roy A, Samrow HA, Dooley KM. High pressure molding and carbonation of cementitious materials. Ind Eng Chem Res 1999;38(7):2641–9.

[4] Soroushian P, Ostovari K. Accelerated processing of cement-bonded particleboard and fibreboard, United States Patent 5,744,078, 1998.

[5] Moorehead DR. Cementation by the carbonation of hydrated lime. Cem Concr Res 1986;16:700–8.

[6] Korneev VT, Tsvang AS. Study of carbonation of lime quartz mixtures. Translated from Zh Prikl Khim 1986;59(1):165–8.

[7] Van Balen K, Gemert Van D. Modelling lime mortar carbonation. Mater Struct 1994;27:393–8.

[8] Cazalla O, Rodriguez–Navarro C, Sebastian E, Cultrone G. Aging of lime putty: effects on traditional lime mortar carbonation. J Am Ceram Soc 2000;83(5):1070–6.

[9] Moorhead D. Personnel Communication.

[10] De Silva P. Chemical and microstructural developments of cements with metakaolin, PhD thesis, University of Aberdeen, UK, 1992.

[11] Myerson AS. Hand book of industrial crystallisation. 2nd ed. Boston: Butterworth Heinemann; 2002.