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THE EFFECT OF CARBONATION ON THE MECHANICAL AND MICROSTRUCTURAL PROPERTIES OF LIME AND CEMENT MORTARS

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Abstract: This paper investigates the effects of carbonation on the strength and microstructure of dewatered mortars manufactured from Portland cement (PC), hydrated calcium lime (CL90) and natural hydraulic limes (NHL) of classification 2, 3.5 and 5 with binder/aggregate/water ratios of 1:2:0.78. Dewatering was achieved by placing the mortars on a high porosity brick. Dewatered and non-dewatered mortar specimens were exposed to an atmosphere of nitrogen and nitrogen containing 400 ppm carbon dioxide with a relative humidity of 65%, at 20^{o}C for periods of 14, 28 and 56 days. Following curing compressive strengths were compared to evaluate mechanical performance. Structural morphology was identified using scanning electron microscopy. Results indicated that the CL90 mortars were affected by carbonation but unaffected by dewatering. However, carbonation and dewatering leads to an increase in compressive strength for hydraulic limes and cement mortar mixes. Scanning electron micrographs revealed microstructural differences between the various binders which are associated with compressive strength.

Keywords: *Lime mortar, Portland cement, Dewatering, Carbonation, Hydration.*

1 Introduction

A recent resurgence of lime mortars within the UK building industry has increased the demand for a knowledge and understanding of their properties, with specific attention being paid to the hardening processes. Lime mortars are produced by mixing lime, sand and water, the ratios of which are determined by the application. Limes can be categorised into two fundamentally different types, hydraulic and non-hydraulic. Non-hydraulic limes consist predominately of calcium hydroxide and are manufactured by calcining pure limestone. These set by carbonation, the rate of which is determined by the atmospheric conditions, such as carbon dioxide concentration, relative humidity and temperature (El-Turki et al. 2007, El-Turki et al. 2009, Lawrence 2006). In natural hydraulic limes, dicalciumsilicate (C_2S) is the major hydraulic phase however the phases, C_2AS, C_3S, C_3A and C_4AF were also detected in small amounts (LANAS et al. 2004). The latter are most commonly formed in the manufacture of cement at much higher temperatures.

The ability to determine the extent of carbonation in a lime-based mortar is important in terms of monitoring strength and structural development. Furthermore, the absorption of water from fresh mortar by clay brick substrates depends on the porosity of the substrates.

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The action of dewatering on lime and cement mortars used in the construction industry is believed to have an important influence on the carbonation and hydration reactions and, hence, strength development. However, in practice it is not possible to determine how dewatering affects the strength of a mortar joint on site.

This paper describes the laboratory research in which a number of different mortar mixes has under gone a dewatering process using high porosity brick. Dewatered and non-dewatered mortar specimens were exposed to an atmosphere containing pure nitrogen and 400 ppm carbon dioxide with a relative humidity of 65%, at 20^{o}C for periods of 14, 28 and 56 days. For comparative purposes, high calcium (CL90) lime, hydraulic limes (NHL2, NHL3.5 and NHL5) and Portland cement based mortars were tested.

2 Experimental Method

2.1 Mortar preparation and carbonation

Specimens were manufactured using five different binder types, namely non-hydraulic lime (CL90), three different classification hydraulic limes (NHL2, NHL3.5 and NHL5) and Portland cement (PC) all of which were supplied by Castle Cement Ltd, Clithero. Croxden sand was used to manufacture the samples. Only one mix design was studies by mixing the binder, sand and water in the ratio 1:2:0.78.

Each mortar mix design was cast into cylindrical moulds 18 mm in diameter and 36 mm in length. Half of these specimens were dewatered naturally by placing on a high-porosity Golden Purple brick substrate supplied by Ibstock Brick Limited for 15 minutes while, the other specimens were left in their original condition. Both sets of moulds were then left to harden under laboratory conditions at 25°C and 50% relative humidity for seven days. The non-dewatered and dewatered samples were then demoulded and cured in separate environmental chambers with only nitrogen and nitrogen containing 400 ppm carbon dioxide at a relative humidity of 65% and temperature of 20°C. The average compressive strengths for three dewatered and non-dewatered specimens were measured at 14, 28 and 56 days of curing. Microstructure analysis was performed using a Hitachi S2300 scanning electron microscope.

2.2 Amounts of water removed from mortar during dewatering

The degree to which each mortar was dewatered when in contact with the high sorptivity brick substrate was determined by measuring the respective weight change of the mortar and brick substrate before and after dewatering. This was achieved using specially designed cylindrical moulds 18 mm in diameter and 36mm in length which could be removed and weighed after the dewatering process had finished. For the purpose of this study a time of 15 minutes was allowed for dewatering as this was substantially more than the longest dewatering times estimated for the samples.

2.3 Compressive testing

Compressive strength testing was carried out on six samples of each mix; three dewatered and three non-dewatered. The CL90 and NHL mortars were tested using a 10 kN Zwick/Roell compressive testing machine. However the higher strength cement mortars required the use of an INSTRON 600DX Universal Testing machine equipped with a 600 kN load cell. Both machines were fitted with stainless steel platens, one of which allowed movement about a central ball joint to accommodate sample irregularities. Prior to testing the machines were calibrated to ensure the results could be correlated. Measurements were carried out at a constant stroke rate of 5 μms^{-1} with load and displacement recorded automatically on a PC.

2.4 Microstructural analysis

Microstructural analysis was carried out using a Hitachi S2300 scanning electron microscope (SEM) operating at an accelerating voltage of 25 keV. Surface charging of the samples surfaces was reduced by the application of a thin layer of gold using an Edwards Scancoat Six sputter system. Images were obtained from sample fracture surfaces after 14, 28 and 56 days of carbonation.

3 RESULTS AND DISCUSSION

3.1 Amounts of water removed from mortar during dewatering

The water loss from the hydrated calcium lime (CL90), natural hydraulic limes (2, 3.5 and 5) and Portland cement (PC) mortars are shown in figure 1. The figure clearly shows that as the hydraulicity of the binder increases so does the water loss. This is illustrated by comparing the water loss of the non-hydraulic calcium lime which was 11.9% to that of the hydraulic Portland cement mortar which was 32%. A similar trend with greater water loss observed for the more hydraulic binders is observed for the natural hydraulic limes 2, 3.5 and 5.

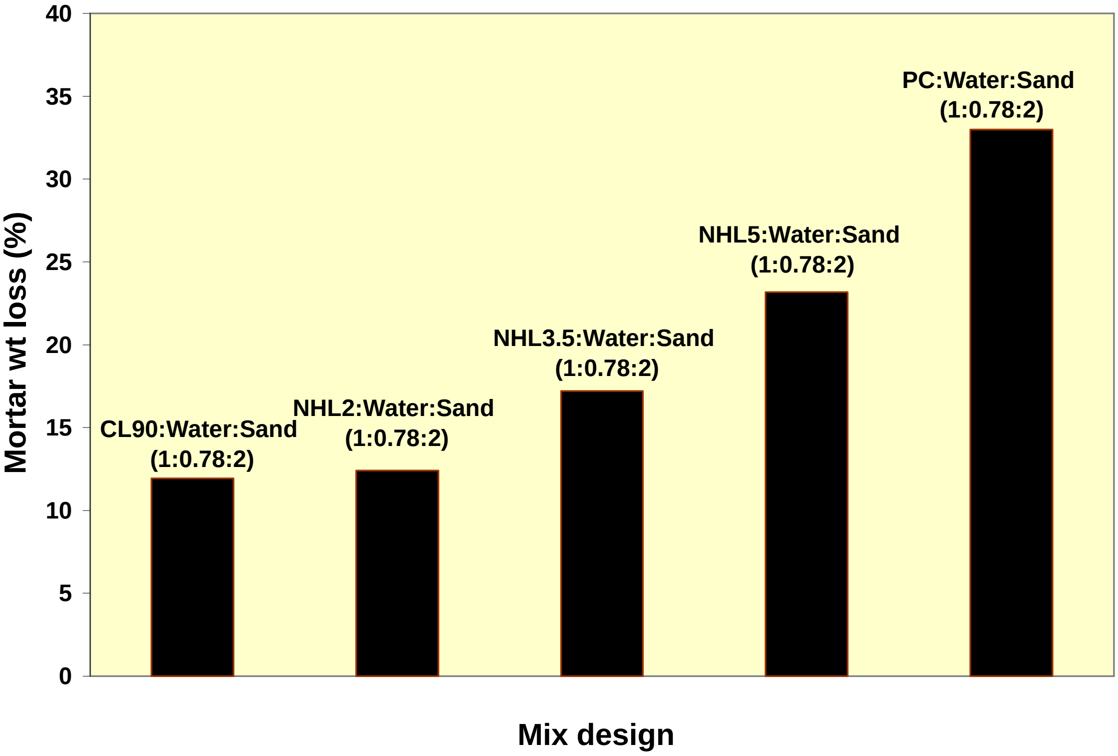

Figure 1: Water loss from each mortar.

3.2 Compressive strengths

Figure 2(a–e) shows the average compressive strengths of both non-dewatered and dewatered mortars at 14, 28 and 56 days of curing in N_2 and N_2 containing 400 ppm CO_2, a relative humidity of 65% and a temperature of 20°C. The results presented show that, for all mixes, the failure stress increased when the mortars cured in the environment containing 400 ppm carbon dioxide. Furthermore, the strength increased with curing time as well as with binder hydraulicity. The increase in compressive strength of CL90 mortars with time is attributed solely to carbonation, which is the transformation of portlandite to calcite.

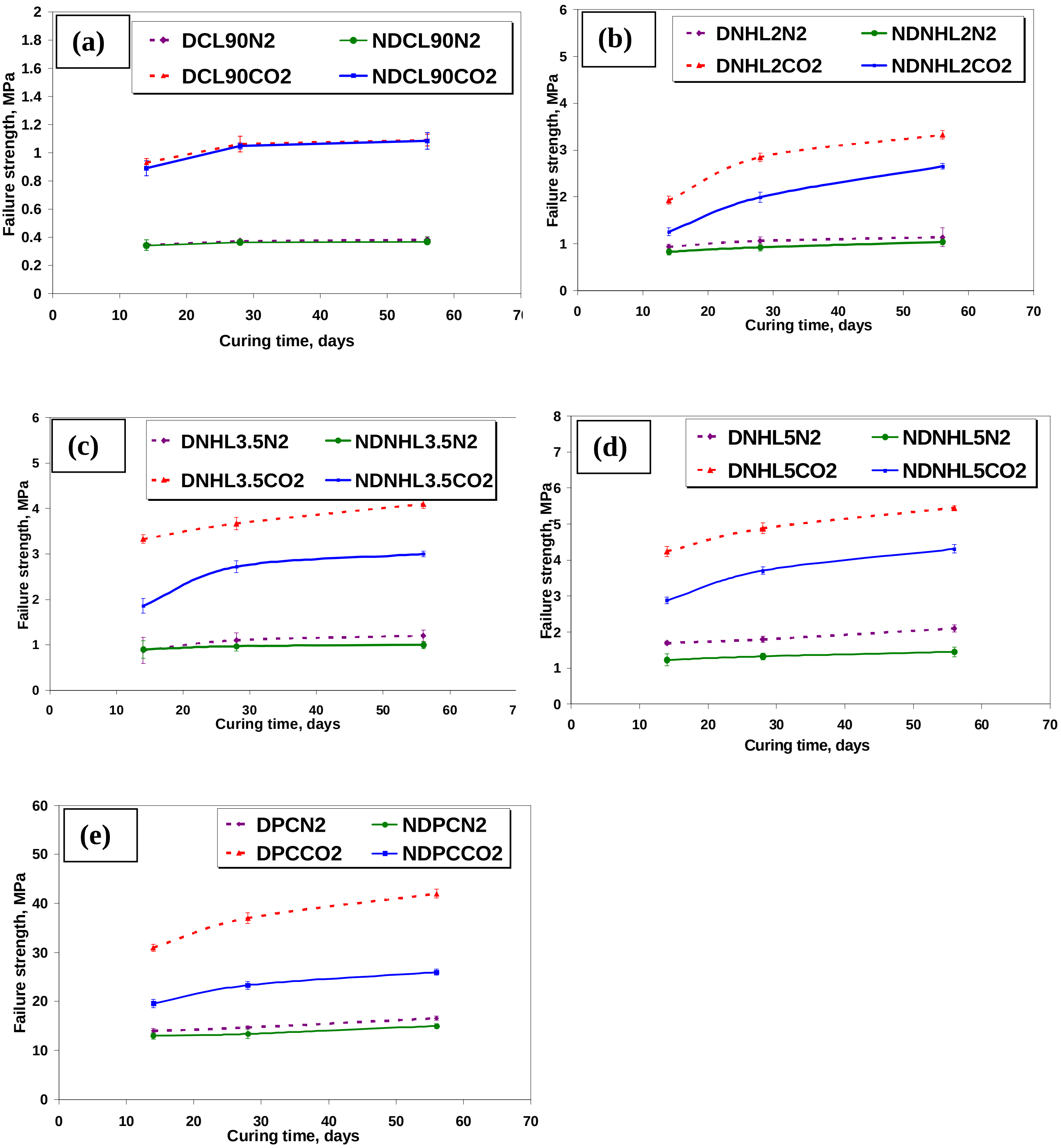

Figure 2: Compressive strength of dewatered and non-dewatered mortar versus time after curing in N_2 and N_2 containing 400ppm CO_2. CL90, (b) NHL2, (c) NHL3.5, (d) NHL5, (e) PC. (ND= Non-dewatered, D= Dewatered)

Previous results indicate that 28 days is sufficient for the carbonation process to have initiated throughout the entire volume of the sample (El-Turki et al. 2006). With the exception of the CL90 mortars (Figure 2a), where dewatering had little effect on strength, all the hydraulic lime mortars showed a clear increase in strength at all curing times for dewatered mortar over the non-dewatered equivalent (Figure 2b-d). This observation can be extended to the cement mortars (Figure 2e). Dewatered samples containing Portland cement binders showed the greatest increase in strength with smaller increases in strength observed for the samples containing hydraulic lime binders.

3.3 Microstructure and composition

Scanning electron micrographs of fracture surfaces from the non-dewatered and dewatered mortars manufactured from CL90 after 56 days of curing in N_2 and N_2 containing 400 ppm CO_2 are shown in figure 3(a-b) and figure 3(c-d) respectively. The surface of both the non-dewatered and dewatered samples cured in N_2 consists predominantly of hexagonal calcium hydroxide crystals (portlandite), with small amounts of calcium carbonate crystals (Figures 3a and 3c). This observation in a good agreement with the strength results of the corresponding samples (Figure 2a).

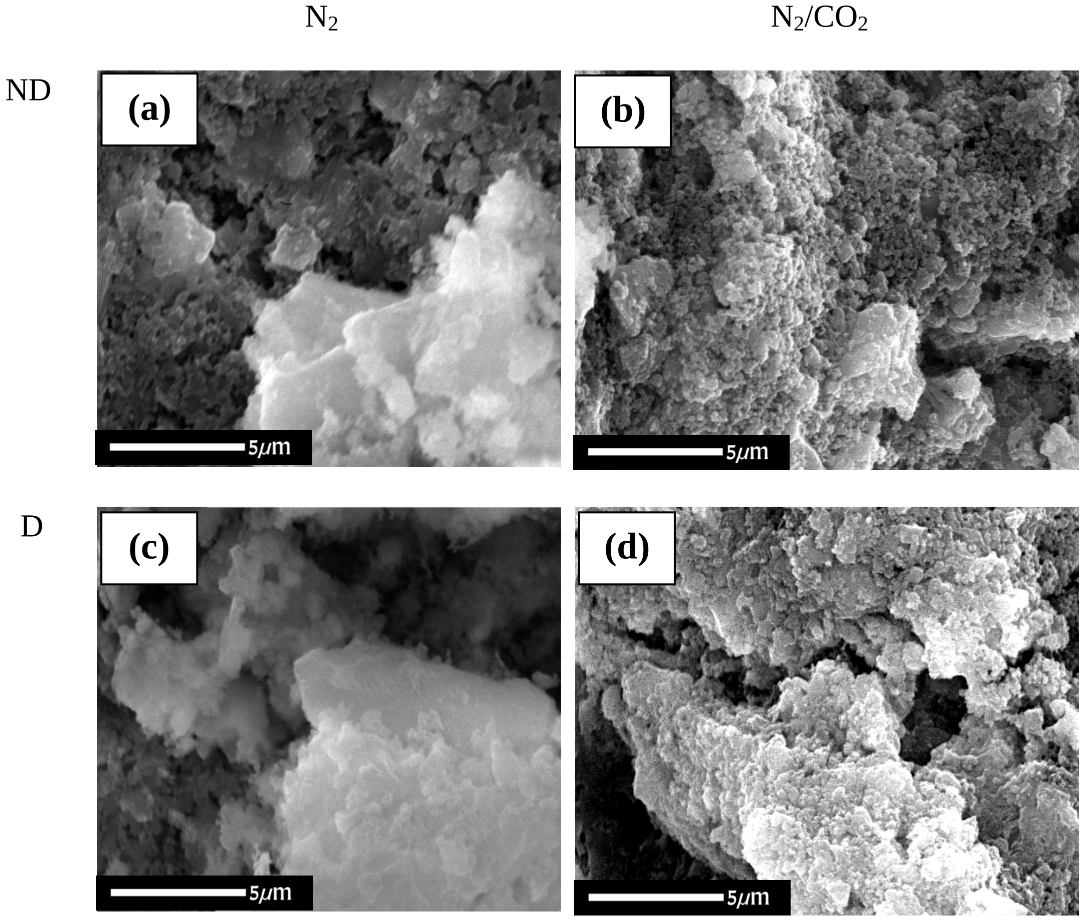

Figure 3: Secondary electron micrographs from the fractured surfaces of non-dewatered and dewatered CL90 mortars after curing for 56 days; (a) ND Mortar cured in N_2 at RH 65% and 20^oC; (b) ND mortar cured in N_2 containing 400 ppm CO_2 at RH 65% and 20^oC; (c) D Mortar cured to N_2 at RH 65% and 20^oC; (d) D mortar cured to N_2 containing 400 ppm CO_2 at RH 65% and 20^oC. (ND= Non-dewatered, D= Dewatered)

Exposure of non-dewatered and dewatered CL90 mortars to an atmosphere of N_2 containing 400 ppm CO_2 produces a significant change in the morphology, as shown in figure 3b and figure 3d respectively. It is likely that the calcium carbonate crystals have grown to cover the entire surface after 56 days of curing. No hexagonal calcium hydroxide crystals were observed indicating complete carbonation. As previously mentioned, the increase in compressive strength of CL90 mortars with time is attributed exclusively to carbonation, which is the transformation of portlandite to calcite.

Scanning electron micrographs recorded from a fracture surface, after 56 days curing, of non-dewatered and dewatered NHL5 mortar samples in N_2 and N_2 containing 400ppm CO_2 are shown in figure 4(a-b) and figure 4(c-d) respectively. The images of non-dewatered and dewatered mortars cured in N_2 revealed morphology consisted of hexagonal portlandite and needle-like crystals. In comparison with the images obtained from the similar mortar samples after 56 days of carbonation, these images showed higher proportion of high aspect ratio needle shaped crystals (Figure 4b and Figure 4d). These needles are the major hydraulic phase in the natural hydraulic limes and contribute to the mortar strength (El-Turki et al. 2007, LANAS et al. 2004). A small number of calcium carbonate crystals, were also observed indicating the onset of carbonation.

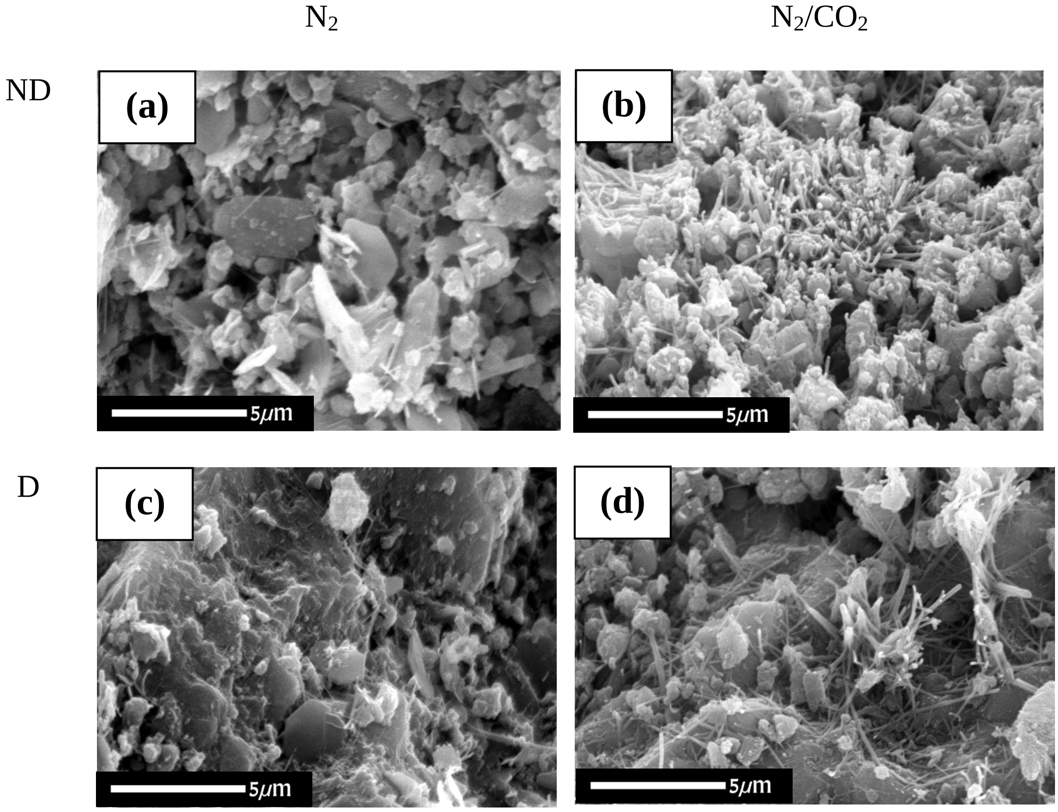

Figure 4: Secondary electron micrographs from the fractured surfaces of non-dewatered and dewatered NHL5 mortars after curing for 56 days; (a) ND Mortar cured in N_2 at RH 65% and 20^oC; (b) ND mortar cured in N_2 containing 400 ppm CO_2 at RH 65% and 20^oC; (c) D Mortar cured in N_2 at RH 65% and 20^oC; (d) D mortar cured in N_2 containing 400 ppm CO_2 at RH 65% and 20^oC. (ND= Non-dewatered, D= Dewatered)

Figures 5a and 5c show the scanning electron micrographs obtained from non-dewatered and dewatered Portland cement mortars after 56 days of curing in N_2. The microstructure is totally different to that of the lime mortars examined. The images consists almost entirely of interconnected high aspect ratio needle shaped crystals and hexagonal $Ca(OH)_2$ plates. PC, hardens by the hydration of cement clinkers, namely C_3S, C_2S, C_3A and C_4AF, all of which hydrate at different rates, thus influencing important factors such as setting time, workability and mechanical strength. The microstructures of both non-dewatered and dewatered mortars after 56 days of carbonation are shown in figure 5b and figure 5d respectively. They consist almost entirely of interconnected high aspect ratio crystals. These crystals may includes various hydration products such as calcium silicate hydrate, calcium hydroxide, ettringite and monosulpahte, but carbonated pastes may contain calcite in place of calcium hydroxide, and possibly gypsum from decomposition of the ettringite and monosulphate phases (BROCKEN et al. 2000).

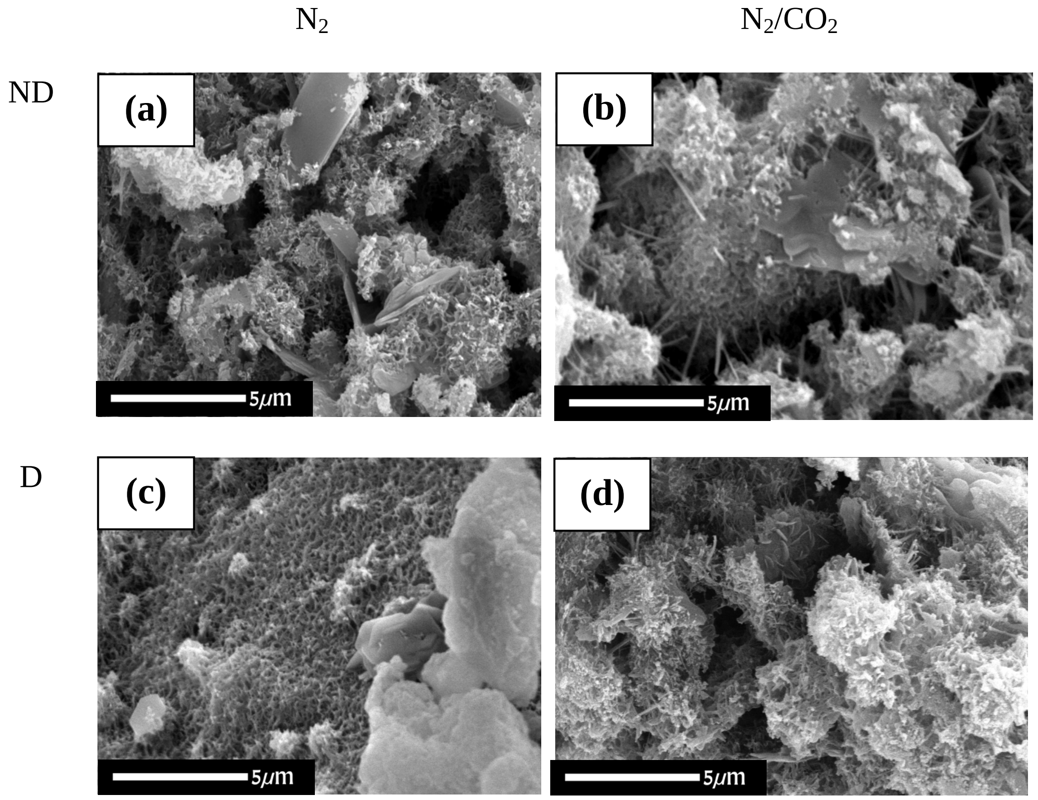

Figure 5: Secondary electron micrographs from the fracture surfaces of non-dewatered and dewatered PC mortars after curing for 56 days; (a) ND Mortar cured in N_2 at RH 65% and 20°C; (b) ND mortar cured in N_2 containing 400 ppm CO_2 at RH 65% and 20°C; (c) D Mortar cured in pure N_2 at RH 65% and 20°C; (d) D mortar cured in N_2 containing 400 ppm CO_2 at RH 65% and 20°C. (ND= Non-dewatered, D= Dewatered)

4 CONCLUSIONS

The following conclusions can be drawn from the results:

a) The carbonation had clear effect on the strength of both non-dewatered and dewatered mortars manufactured from hydrated calcium lime (CL90) and natural hydraulic limes (NHL) of classification 2, 3.5 and 5 and Portland cement (PC).

b) The dewatering process resulted in a dramatic increase in the strength of the hydraulic mortars when comparisons were made at 14, 28 and 56 days with non-dewatered equivalent samples. The dewatering process had no effect on the CL90 mortars where the hydration reaction plays no part in the hardening process

c) It is suggested that dewatering of mortars prepared with hydraulic binders (PC and natural hydraulic limes) increases the rate of crystallisation of hydrated silicate phases within the matrix, having a significant influence on the mechanical strength. Moreover, dewatering reduces the w/c ratio, giving a denser microstructure and hence higher strength. This view is supported by secondary electron micrographs where the micrographs indicate that growth of C-S-H crystals between sand grains could account for the increased strength of dewatered hydraulic mortars.

5 Acknowledgements

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