# Initial investigation into the carbonation of MgO for soil stabilisation

## Premières investigations sur la carbonatation de MgO utilisé pour la stabilisation des sols

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ABSTRACT: While Portland cement (PC) is the most widely used binder for soil stabilisation, there are significant environmental impacts associated with its production in terms of high energy consumption and $CO_2$ emission. Hence, the development of new low carbon foot-print alternative cements has been encouraged. In this paper, reactive magnesia (MgO) was used as a soil stabilisation binder and the MgO-stabilised soils were carbonated by gaseous $CO_2$ to improve the mechanical properties of the soil and reduce the $CO_2$ emission. The mechanical and microstructural properties of the carbonated MgO stabilised soils were investigated by using unconfined compressive testing, x-ray diffraction (XRD) and scanning electron microscopy (SEM). The results showed that the strength development rates of carbonated MgO-stabilised soils were much faster than those PC- and MgO-stabilised soils, and the unconfined compressive strength of highly carbonated MgO-stabilised soils was close to that of 28-day ambient cured PC-stabilised soils. The XRD and SEM results indicated that nesquehonite ($MgCO_3{\cdot}3H_2O$) was the main product of the carbonated MgO-stabilised soils and responsible for the significant strength development.

RÉSUMÉ : Alors que le ciment Portland est le liant le plus utilisé pour la stabilisation des sols, il y a d'importants impacts environnementaux associés à sa production en termes de consommation d'énergie élevée et d'émission de $CO_2$. Par conséquent, le développement de nouveaux ciments alternatifs à basse teneur en carbone a été encouragé. Dans cet article, la magnésie réactive (MgO) a été utilisée comme liant pour la stabilisation des sols et les sols stabilisés à la magnésie ont été carbonatés par du $CO_2$ gazeux afin d'améliorer les propriétés mécaniques des sols et de réduire les émissions de $CO_2$. Les propriétés mécaniques et microstructurelles des sols stabilisés à la magnésie et carbonatés ont été étudiées en utilisant des essais de compression simples, la diffractométrie de rayons X (DRX), et la microscopie électronique à balayage (MEB). Les résultats montrent que le développement de la résistance des sols stabilisés à la magnésie et carbonatés était beaucoup plus rapide que celui du ciment Portland avec prise à l'air ambiant et celui des sols stabilisés à la magnésie. Ils ont également montré que la résistance à la compression uniaxiale des sols stabilisés à la magnésie et carbonatés était proche de celle des sols stabilisés au ciment Portland avec prise à l'air ambiant pendant 28 jours. Les résultats des DRX et MEB ont indiqués que la nesquehonite ($MgCO_3{\cdot}3H_2O$) était le produit principal des sols stabilisés à la magnésie et carbonatés, et responsable de la forte augmentation de la résistance.

KEYWORDS: soil stabilisation, reactive MgO, carbonation, unconfined compressive strength, microstructure.

## 1 INTRODUCTION

Soil-cement mix technology is one of the most widely used ground improvement methods, with Portland cement (PC) being the most commonly employed binder (Sherwood 1993, Bergado et al. 1996, Al-Tabbaa, 2003). However, there are significant environmental impacts associated with the production of PC in terms of high energy consumption and $CO_2$ emissions (0.85t $CO_2$/t PC), and hence is responsible for 5-8% of anthropogenic $CO_2$ emissions worldwide (WBCSD, 2002; IPCC, 2004).

In order to reduce the usage of PC, new alternative cements have been encouraged. Reactive magnesia (MgO) cements recently emerged as a more sustainable alternative to PC (Harrison 2008). Reactive MgO is generally calcinated from magnesite ($MgCO_3$) at temperatures of ~700-800℃ and should not be confused with dead burned MgO manufactured at a temperature higher than 1400℃, which is known to cause an unsoundness problem in PC due to its delayed hydration behavior (Shand, 2006).

Extensive research has been conducted at the University of Cambridge since 2004 into the reactive MgO cements, as detailed in Al-Tabbaa (2013). Reactive MgO hydrates, much faster than dead burned MgO, to form brucite ($Mg(OH)_2$). Although brucite has a very limited binding ability, it could carbonate to form one or more of the hydrated magnesium carbonates, namely nesquehonite ($MgCO_3{\cdot}3H_2O$), hydromagnesite ($Mg_5(CO_3)_4(OH)_2{\cdot}5H_2O$) and/or dypingite ($Mg_5(CO_3)_4(OH)_2{\cdot}4H_2O$). The hydration of MgO and carbonation of brucite both are expansive reactions, which significantly fill available pores. These hydrated magnesium carbonates also form well ramified networks of massive crystals with a very effective binding ability resulting in substantial and rapid strength increase. For example, in porous construction blocks, they significantly outperformed corresponding PC blocks with strengths of 200-300% higher (Liska 2009; Unluer 2012). High levels of carbonation have been achieved in full-scale porous blocks trial production (Liska et al. 2012a and b), reabsorbing most of the $CO_2$ generated during the decomposition of the magnesite.

In this paper, reactive MgO was initially used as a soil stabilisation binder and the MgO-soil samples were carbonated by $CO_2$ gas to improve the mechanical properties of the soil and reduce the $CO_2$ emission. This initial work is thereafter complemented by investigating the impact of relevant variables including soil type, soil water content, binder content and carbonation method (Yi et al., 2012), as well as by using a laboratory-scale auger to model the installation of carbonated soil-MgO deep mixed columns (Yi et al. 2013).

## 2 MATERIALS AND METHODS

### 2.1 *Binders, soils and sample preparations*

A model soil was used, namely a slightly clayey silty sand, consisting of 90% sharp sand, 5% kaolin clay and 5% silica flour. The sharp sand (obtained from Ridgeons, Cambridge, UK), had a D50 of 0.8 mm and coefficient of uniformity of 4.3, and the kaolin clay (obtained from Richard Baker Harrison, Ilford, UK) had a liquid limit of 51% and plastic limit of 30%. The silica flour was obtained from David Ball Group, Cambridge. The water content of the soil was 10%. Reactive MgO (obtained from Richard Baker Harrison, Ilford, UK) and PC (obtained from Castle Cement, UK) were applied at 13% dry content to the soil. The reactive MgO had the following oxide composition: MgO: 97.2%, CaO: 1.2%, $SiO_2$: 1.2%, $Al_2O_3$: 0.2% and $Fe_2O_3$: 0.2%, while the PC had: CaO: 63.6%, $SiO_2$: 13.6%, $Al_2O_3$: 10.2%, $Fe_2O_3$: 2.7%, $SO_3$: 6.9%, MgO: 0.6% and $K_2O$: 0.9%.

The sharp sand, kaolin clay and silica flour were initially mixed and homogenised for 5 minutes in a bench-top food mixer after which water was added and the mixing continued for further 5 minutes. The MgO was then applied to the model soil and the entire system was then mixed twice for 5 minutes with an inspection inbetween the two intervals. The homogenised mix was then placed in cylindrical moulds, with 50 mm diameter and 100 mm height, applying consistent moderate compaction in three layers by rodding and eliminating any trapped air pockets. The samples were demoulded ~1 hour later, then placed in the triaxial apparatus and subjected to the carbonation procedures detailed below. A subset of MgO and PC stabilised samples was also cured in their moulds under “ambient” conditions, of 20±2℃ and 95±3% relative humidity, for 1, 7, 28 and 90 days.

### 2.2 *Carbonation procedure and testing*

A triaxial apparatus was used to permeate pressurised gaseous $CO_2$ through the MgO-treated soil as shown in Figure 1. The samples were subjected to 400 kPa confining pressure and then followed by upward permeation of the gaseous $CO_2$. First, the $CO_2$ outflow tap was open during the carbonation process, and the inflow $CO_2$ pressure was maintained at a low value of 20 kPa to reduce leakage. However, the $CO_2$ leakage was still serious, and hence only four carbonation periods were conducted using this method, which were 0.75, 1.5, 3 and 6 hours. Thereafter, the carbonation process was modified: the $CO_2$ outflow tap was closed two minutes after turning on the $CO_2$ inlet while keeping the inlet open therefore maintaining the $CO_2$ pressure at the 200 kPa level for the designated carbonation periods: 0.75, 1.5, 3, 6, 12, 24, 48 and 96 hours. Besides, a subset of samples carbonated for 24 hours using this method was then cured under ambient conditions for 7, 28 and 90 days.

Figure 1.The triaxial cell used for MgO-stablised soil carbonation.

In addition, an incubator, with 20% $CO_2$ concentration (1 bar) at 20±2℃ and relative humidity of 95±3%, was also used to carbonate MgO-stabilised soils for comparison purposes. The carbonation periods were 12, 24, 48, 96 and 168 hours (7 days). A subset of samples carbonated for 7 days using this method were then cured under ambient conditions for 28 and 90 days.

All the samples were tested in triplicate for their unconfined compressive strength (UCS) at a constant displacement rate of 1.14 mm/min. X-ray diffraction analysis (XRD) and scanning electron microscopy (SEM) were conducted for selected mixes.

## 3 RESULTS AND DISCUSSIONS

### *3.1 Unconfined compressive strength*

Figure 2 shows the UCS of MgO-stabilised soils carbonated using different carbonation methods. The MgO-stabilised soils carbonated in an incubator, which is generally used to carbonate the MgO-based construction blocks (Liska 2009; Unluer 2012), took ~96 hours of carbonation to reach a maximum UCS value (~8 MPa). While those carbonated in triaxial cell with 200 kPa $CO_2$ stationary showed similar strength development behavior but with much faster rate, whereby after ~3 hours the stabilised soil also reached a similar maximum UCS value. The UCS of the MgO-stabilised soils carbonated in triaxial cell with 20 kPa $CO_2$ flow through for 45 minutes was much lower than those with 200 kPa $CO_2$ stationary, mainly due to the lower $CO_2$ pressure of the former (Yi et al. 2012). However, the attained strength is approximately the same regardless of the $CO_2$ pressure applied and its concentration. Figure 2 indicates that there is no need to keep the $CO_2$ flowing through the sample during the carbonation process, which causes $CO_2$ leakage.

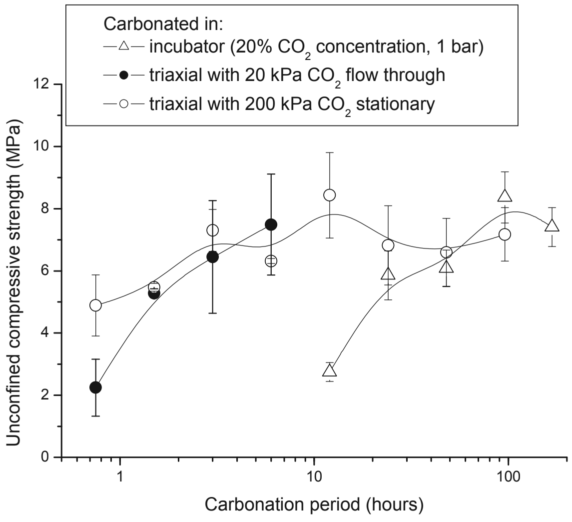


Figure 2. UCS of MgO-stabilised soils carbonated in incubator, triaxial cell with 20 kPa $CO_2$ flow through and 200 kPa $CO_2$ stationary.

Figure 3 presents the UCS of uncarbonated MgO- and PC-stabilised soils and carbonated MgO-stabilised soils cured under ambient conditions. It is evident that the UCS values of the uncarbonated MgO-stabilised soil are much lower than those PC-stabilised soils, and both of the two mixes took ~28 days to finish most of their strength development. Comparing of Figure 2 and Figure 3 indicates that the carbonation significantly increased the UCS of MgO-stabilised soils in a very short time, and the UCS of highly carbonated MgO-stabilised soils was close to that of the 28-day PC-stabilised soils, which was ~10 times that of 28-day uncarbonated MgO-stabilised soils. However, the ambient curing period did not affect the strength of carbonated MgO-stabilised soil significantly, indicating the carbonated MgO-stabilised soil could be used to support a structure just after the completion of the carbonation procedure.

This is confirmed by the results of laboratory-scale auger installed soil-MgO deep mixed columns (Yi et al. 2013).

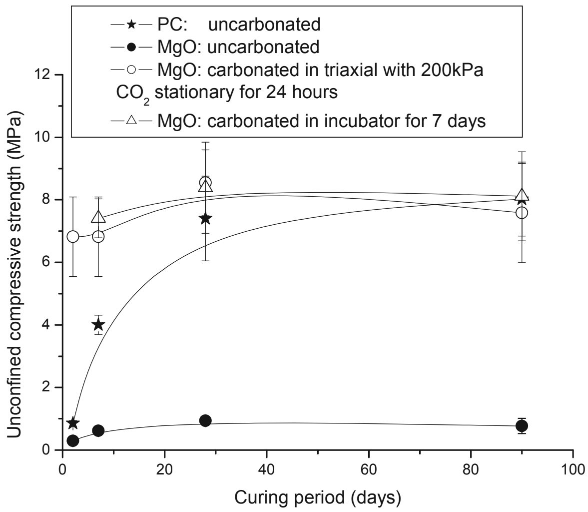
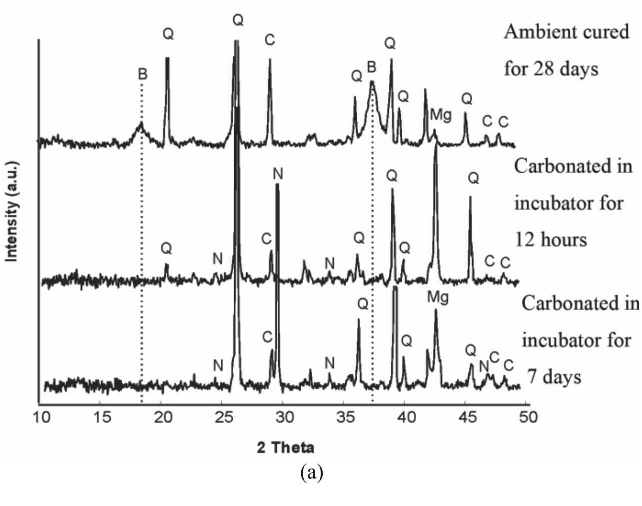


Figure 3. UCS of uncarbonated MgO- and PC-stabilised soils and carbonated MgO-stablised soils versus ambient curing period.

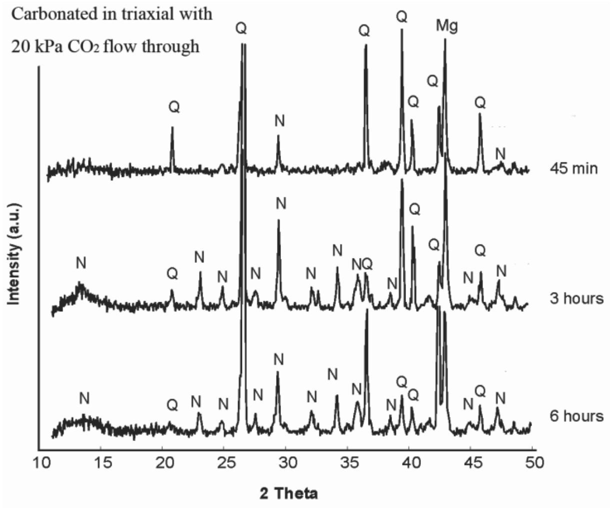
*3.2 X -ray diffraction*

Figure 4 shows the XRD diffractograms of the MgO-stabilised soils ambient cured for 28 days (uncarbonated) and carbonated in incubator for 12 hours and 7 days, the MgO-stabilised soils carbonated in triaxial cell with 20 kPa $CO_2$ flow through for 45 minutes, 3 hours and 6 hours, as well as the MgO-stabilised soils carbonated in triaxial cell with 200 kPa $CO_2$ stationary for 45 minutes, 3 hours and 4 days. Two strong brucite ($Mg(OH)_2$) peaks and a weak MgO peak are detected for the 28-day ambient cured MgO-stabilised soil (Figure 4a). For the carbonated samples, the disappearance of the brucite peak together with the gradually weakening of the MgO peak suggest the carbonation of the MgO resulting in the formation of nesquehonite ($MgCO_3{\cdot}3H_2O$). With no other changes in the mineralogy, this correlates with the rapid and significant strength development of carbonated MgO-stabilised soils.

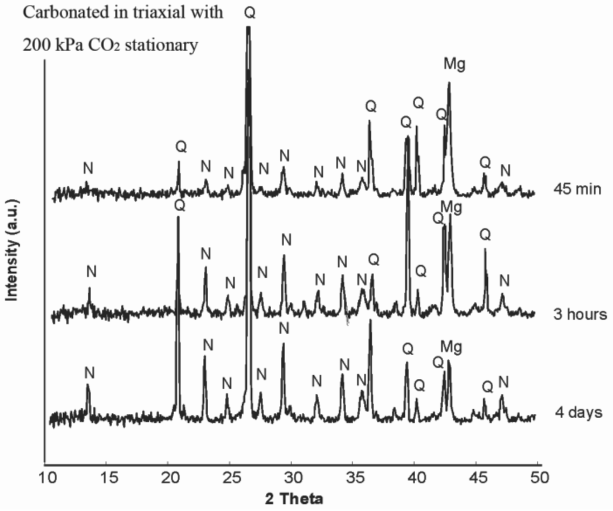
Additionally, the MgO peak was detected in all the carbonated MgO-stabilised soils, including those carbonated in an incubator for 7 days and in a triaxial cell with 200 kPa $CO_2$ stationary for 4 days, which were much stronger than that in the 28-day ambient cured sample. These results indicated that the water content (10%) was not enough for all the MgO (13%) to carbonate to form nesquehonite. Hence the MgO content should be linked to the water content of the soil, as well as its porosity.



(a)



(b)



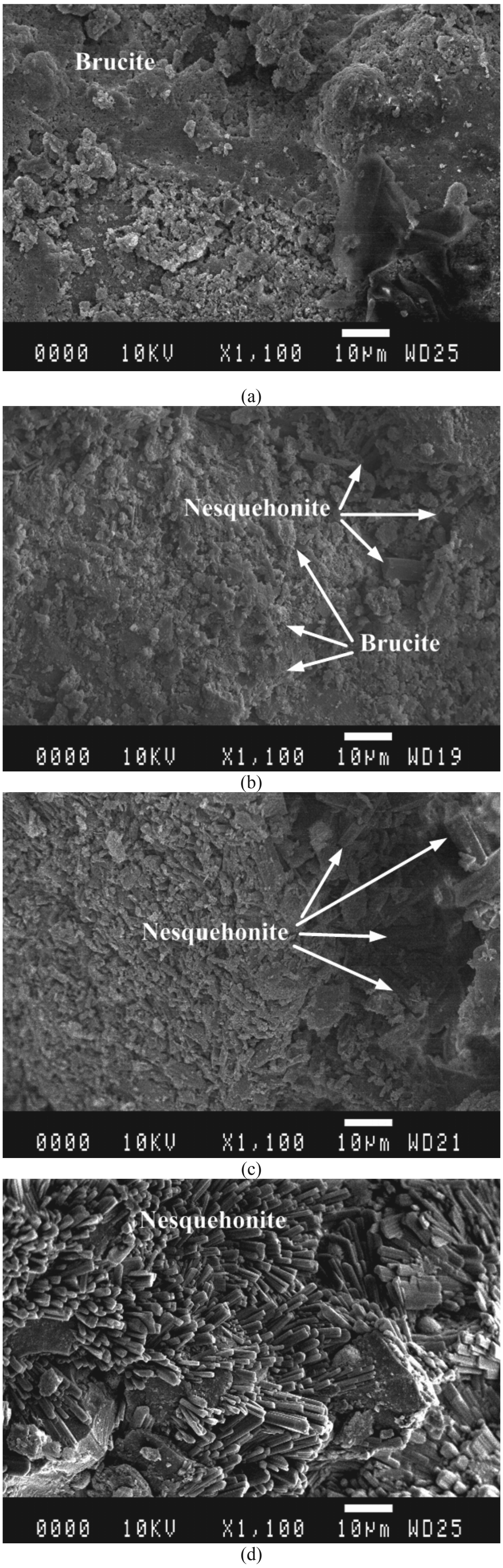
(c)

Figure 4.XRD diffractograms of MgO-stabilised soils: (a) ambient cured for 28 days and carbonated in incubator; (b)carbonated in triaxial cell with 20 kPa $CO_2$ flow through; and (c) carbonated in triaxial cell with 200 kPa$CO_2$ stationary. The abbreviations stand for: B-Brucite, C-Calcite, Mg-MgO, N-Nesquehonite, Q-Quartz.

*3.3 Scanning electron microscopy*

Typical SEM micrographs of the MgO-stabilised soils ambient cured for 28 days (uncarbonated), carbonated in triaxial cell with 200 kPa $CO_2$ stationary for 45 minutes and 4 days, as well as that carbonated in an incubator for 7 days are shown in Figure 5.

The 28-day ambient cured sample (Figure 5a) exhibits the presence of brucite, which has a very limited binding ability due to its poorly interconnected structure (Liska 2009; Unluer 2012). All the carbonated MgO-stabilised soils (Figure 5b, c and d) display formation of nesquehonite, consistent with the XRD results. Besides, brucite was also observed in Figure 5b confirming that there was only partial carbonation at 45 minutes, and which then disappeared at 4 days (Figure 5c) confirming that the carbonation proceeded to a high degree.



(a)

(b)

(c)

(d)

Figure 5.Scanning electron micrographs of the MgO-stabilised soils: (a) ambient cured for 28 days (uncarbonated); carbonated in triaxial cell with 200 kPa $CO_2$ pressure for (b) 0.75 hours and (c) 4 days; (d) carbonated in incubator for 7 days.

## 4 CONCLUSION

The results showed that the carbonation significantly increased the strength of MgO stabilised soil, and the UCS of highly carbonated MgO stabilised soil was close to that of the 28-day ambient cured PC-stabilised soil, which was ~10 times that of 28-day uncarbonated ambient cured MgO-stabilised soil. The carbonation of MgO stabilised soil in the triaxial cell with high $CO_2$ pressure happened much faster than in the incubator, the former could be finished in a few hours while the latter took a few days. Dissimilar to the uncarbonated ambient cured MgO- or PC-stabilised soil, the UCS of highly carbonated MgO stabilised soil did not increase with ambient curing period. The XRD and SEM results indicated that nesquehonite, one of the hydrated magnesium carbonates, was the main product of the carbonated MgO in soil.

## 5 ACKNOWLEDGEMENTS

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