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Development of reactive MgO-based Engineered Cementitious Composite (ECC) through accelerated carbonation curing

Hao-Liang Wu [a,1], Duo Zhang [b], Brian R. Ellis [b], Victor C. Li [b,*]

[a] Jiangsu Key Laboratory of Urban Underground Engineering & Environmental Safety, Institute of Geotechnical Engineering, Southeast University, Nanjing 210096, China
[b] Department of Civil and Environmental Engineering, University of Michigan, Ann Arbor, MI 48109, USA

HIGHLIGHTS

- A green ECC is developed based on MgO and fly ash through accelerated carbonation.
- Compression, tension, and multiple cracking characters are evaluated.
- CO_2 uptake and materials sustainability of the newly developed ECC are assessed.
- ECC with 1-d carbonation achieves both environmental and technical benefits.

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ABSTRACT

The use of reactive magnesium oxide (MgO) is widely recognized in carbonated concrete formulations associated with permanent sequestration of CO_2. Engineered Cementitious Composite (ECC) is an advanced fiber reinforced cement-based composite with high tensile ductility and intrinsically tight crack width. In this paper, we investigate an alternative binary binding system for ECC: reactive MgO and fly ash cured with an accelerated carbonation process. Compressive strength, density, carbonation depth, tensile performance and crack pattern of the carbonated reactive MgO-based ECC were investigated at various curing ages. In addition, the CO_2 uptake and materials sustainability, in terms of energy consumption, net CO_2 emission and cost of the newly developed ECC were assessed. The objective of this research is to further advance the application of reactive MgO and utilization of CO_2 in the construction industry through novel ECC material. It was observed that carbonation curing densifies the binding system, thus leading to an increase in both compressive and first cracking tensile strengths of ECC. The tensile strain capacity of the carbonated reactive MgO-based ECC achieved up to 6% with an average crack width below 60 μm after 1-day carbonation. Compared to conventional ECC (M45) and concrete, the 1-day carbonated reactive MgO-based ECC could reduce the net CO_2 emission by 65% and 45%, respectively. It is concluded that environmental and technical benefits could be simultaneously achieved for the 1-day carbonated reactive MgO-based ECC incorporated with 50% fly ash. The findings of this research shed light on further applications of reactive MgO cement in the precast industry.

1. Introduction

As the cement and concrete industries are facing an increasing pressure of CO_2 emission reduction, intensive efforts have been devoted to developing low-carbon alternative binders [1,2] and manufacturing approaches [3,4]. Among emerging alternative binders, reactive MgO has demonstrated an outstanding ability to sequester CO_2 as compared to Portland cement (PC) [5,6]. A relatively lower temperature is needed for the calcination process of the reactive MgO (i.e., 700–1000 °C for reactive MgO vs. 1450 °C for PC), thus allowing the use of alternative fuels with relativity low heating values (e.g., refuse-derived fuel and hybrid) [7]. The reactive MgO forms the binding property through reacting with H_2O and CO_2 to sequester CO_2 and gain strength [8–12]. Eq. (1) shows the hydration of MgO while Eqs. (2)–(4) represent the carbonation reactions. The hydration product of MgO, i.e., brucite, contributes little to no bonding capability, hence demanding a sufficient carbonation degree to form desirable strengths. To do so requires an intentional carbonation process to allow

* Corresponding author.
E-mail addresses: duzhang@umich.edu (D. Zhang), brellis@umich.edu (B.R. Ellis), vcli@umich.edu (V.C. Li).
[1] Joint Ph.D. student at the University of Michigan, USA.

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transformation of MgO to a variety of hydrated magnesium carbonates, mainly including nesquehonite, hydromagnesites and dypingite. The ability of fixing gaseous CO_2 through MgO carbonation has spurred interest in utilizing CO_2 to develop carbonated reactive MgO as a binder for building materials.

$$MgO + H_2O \rightarrow Mg(OH)_2\,(\text{brucite}) \quad (1)$$

$$Mg(OH)_2 + CO_2 + 2H_2O \rightarrow MgCO_3 \cdot 3H_2O\,(\text{nesquehonite}) \quad (2)$$

$$5Mg(OH)_2 + 4CO_2 \rightarrow 4MgCO_3 \cdot Mg(OH)_2 \cdot 4H_2O\,(\text{hydromagnesites}) \quad (3)$$

$$5Mg(OH)_2 + 4CO_2 + H_2O \rightarrow 4MgCO_3 \cdot Mg(OH)_2 \cdot 5H_2O\,(\text{dypingite}) \quad (4)$$

The use of carbonated reactive MgO as a binder faces two intrinsic barriers: (1) the strength development largely relies on carbonation, thus a porous microstructure with small thickness is preferred in facilitating CO_2 diffusion; and (2) reduction in pH of pore solution increases the risk of steel depassivation, hence limiting the application of steel reinforcements [6,9]. These two barriers restrict the application of the carbonated reactive MgO to producing non-structural porous materials at small scale, which are of limited usage in the precast industry.

The needs of further exploring applications of reactive MgO binder with high CO_2 sequestration capacity and production efficiency motivated this study. Engineered Cementitious Composite (ECC) could be a promising candidate for this purpose [13]. ECC represents a class of high tensile ductility materials with strain-hardening and multiple-cracking behavior [14,15]. The tensile strain capacity of ECC is normally over 3%, nearly 300 times that of conventional concrete [16]. The high tensile ductility could reduce the necessities of incorporating steel reinforcements in certain applications, offering opportunities for low-pH binders such as the carbonated reactive MgO [14,15]. Another unique feature of ECC is that the width of the multiple cracks can be controlled to below 60 μm [17], contributing to a low permeability and enhanced durability in the cracked condition. This tight crack width control is autogenous, without dependence on steel reinforcement. A number of structural applications have highlighted substantial technical benefits of using ECC, e.g., impact resistance, structural rehabilitation and seepage proofing [15,18].

ECC can be tailored in terms of chemical formulations and micromechanics to accommodate different types of raw ingredients [15]. In conventional ECC, PC and fly ash are the main binding ingredients while synthetic fibers such as PVA, PP and PE fibers serve as reinforcing elements. Driven by interest in developing more sustainable materials, prior investigations have examined a number of mineral admixtures and alternative binders, e.g., ground granulated blast-furnace slag (GGBFS), alkali-activated binders, to develop low-carbon ECC [19–21]. More recently, the carbonated reactive MgO-based binder was introduced to ECC, achieving a tensile strain capacity of up to ~4.0% and an ultimate tensile strength of ~4.1 MPa after 28-day carbonation [13]. Despite the desirable mechanical performance, the carbonation duration was long (7–28 days) which lessened application appeals in the precast industry, for which efficient manufacturing is economically critical. Additionally, the environmental and economic impacts associated with the carbonated reactive MgO-based ECC remain uncertain.

In further pushing the application of reactive MgO and improving the production efficiency, an accelerated carbonation curing process that was previously adopted in conventional precast concretes [3,9,22–23] is developed here for a reactive MgO-based ECC. The carbonation duration can be shortened to 1 day at an elevated CO_2 pressure. Effects of the accelerated carbonation curing on mechanical properties, cracking behavior and microstructure of the reactive MgO-based ECC are examined. Sustainability and cost of this new version of ECC are also investigated.

2. Experimental program

2.1. Materials and mix proportion

Light-burnt MgO (MAGOX® premium grade, Premier Magnesia, LLC) and fly ash (class F, Headwaters Resources Inc) were used as binders. 99.5% of the MgO can pass the standard #325 mesh and 100% MgO can pass the #100 mesh, as per ASTM E11 [24]. Table 1 lists the chemical compositions of the MgO and fly ash. Polyvinyl alcohol (PVA) fibers with 1.2% oil content supplied by Kuraray Ltd. (Japan) were used for the ECC. Mechanical properties of the PVA fiber are listed in Table 2. Tap water was used for mixing. To achieve a sufficient workability of the fresh mixture, sodium hexametaphosphate (SHMP, chemical formula: $Na(PO_3)_6$, Alfa Aesar™) [25] and superplasticizer (SP, W.R. Grace & Co.) were applied.

Two mix proportions were studied with 70% and 50% MgO by the binder's mass, respectively. Table 3 shows the details of the mix proportions. The two mixtures were denoted as C# and N#, where # referred to MgO content by binder mass (i.e., 0.7 or 0.5). The C# referred to the mixtures subjected to the accelerated carbonation curing, and the N# referred to the non-carbonated reference that was cured in air (T = 20 ± 2 °C, Relative humidity (RH) = 65 ± 2%).

2.2. Sample preparation

Both mixtures were prepared using a 4-liter mortar mixer. The solid ingredients, including reactive MgO and fly ash, were dry-mixed at 100 rpm for 2 min. A SHMP-SP solution was prepared using the mixing water. SHMP was first added into the mixing water and was stirred for 5 min using a magnetic stirrer at a rate of 240 rad/min and a temperature of 85 °C. After mixing, SP was added to the solution and was stirred for another 1 min. The SHMP-SP solution was added to the dry ingredients in the mortar mixer and was mixed at 150 rpm for 3 min to produce a uniform matrix. The PVA fibers were then added and were mixed at the same rate for an additional 5 min. The fresh mixtures were cast into cubes (50 mm × 50 mm × 50 mm) and dogbone-shaped molds (Fig. 1) followed by 3-min vibration. These specimens were covered with a plastic sheet to prevent moisture loss and set in air (temperature = 20 ± 2 °C and RH = 65 ± 2%), and were demolded at 21 h after casting.

The reactive MgO-based ECC specimens after demolding were split into two groups, i.e., carbonation-cured ECC (C#) and non-carbonated ECC (N#). To facilitate CO_2 diffusion in ECC, the demolded specimens were placed on a grid plate for a 3-hour de-mold conditioning at temperature of 20 ± 2 °C and RH of 65 ± 2%. The purpose of the de-mold conditioning was to remove excess moisture in the porous space of ECC to provide interconnected pathways for CO_2 diffusion. The reactive MgO-based ECC carbonation was performed in a pressure chamber depicted in Fig. 2. The carbonation chamber was continuously supplied with CO_2 gas of 99.8% purity and 2.0 bar pressure (2×10^5 Pa). A tray of water (~100 g) was placed inside the chamber to prevent sample water evaporation during the carbonation process. The cube and dogbone-shaped ECC specimens were carbonated for 1 day, 3 days and 7 days. The specimens' mass changes induced by carbonation curing were recorded.

2.3. Performance evaluation

The compression test was performed on the cube specimens following ASTM C109/C109M [26]. The loading rate was set at 0.34 ± 0.07 MPa/s. Triplicate specimens for each batch were prepared and tested. Phenolphthalein indicator was used to determine the depth of carbonation on the cross section of cubes.

Table 1
Chemical compositions of MgO and fly ash determined by XRF.

Chemical component[a]	MgO (%)	FA (%)
SiO_2	0.31	52.19
Al_2O_3	0.2	22.23
SO_3	–	2.16
MgO	95.76	0.93
P_2O_5	–	0.11
K_2O	–	2.56
TiO_2	0.01	1.01
Fe_2O_3	0.13	13.49
CaO	0.81	3.40
Cl	0.03	0.01
Loss on ignition (at 950 °C)	2.75	1.01

Note: [a]Chemical component is analyzed by X-ray fluorescence method using ARL9800XP + XRF spectrometry.