

Characterization of Mg components in reactive MgO – Portland cement blends during hydration and carbonation

Runxiao Zhang^a, Nabil Bassim^b, Daman K. Panesar^{a,*}

^a Dept of Civil & Mineral Engineering, University of Toronto, Toronto, Ontario, M5S 1A4, Canada

^b Dept of Materials Science and Engineering, McMaster University, Hamilton, Ontario, L8S 4L8, Canada

ARTICLE INFO

Keywords:

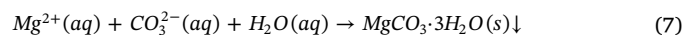
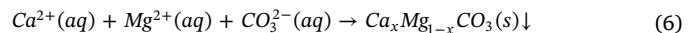
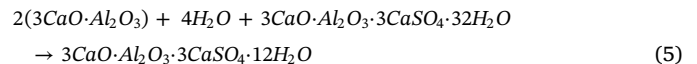
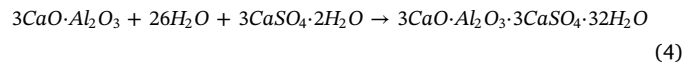
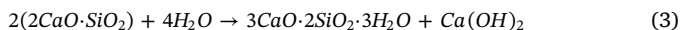
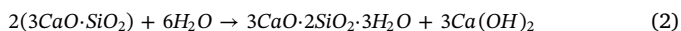
Reactive MgO
Accelerated carbonation
Hydration
Amorphous hydrated Mg carbonate

ABSTRACT

A carbonated binder system containing reactive MgO (r-MgO) and Portland cement (PC) has the potential to provide sufficient mechanical properties and reduce carbon footprint. However, the understanding of the chemistry of hydration and carbonation of the r-MgO-PC system remains unclear. The purpose of this study is to characterize the Mg components that form blends of r-MgO and PC during hydration and carbonation. Paste specimens of r-MgO and PC with r-MgO replacement levels from 10% to 90% were cast, carbonated and examined. Experimental observations reveal that: (1) nesquehonite only occurs in the carbonated r-MgO-PC paste with r-MgO replacement $\geq 60\%$; (2) an amorphous phase of hydrated Mg carbonate was identified in the carbonated paste with 50% r-MgO replacement; and (3) hydrotalcite and magnesium silicate hydrate are major hydration products of r-MgO during the hydration of the paste containing 30% and 40% r-MgO, which are expected to be the precursors of amorphous hydrated Mg carbonate.

1. Introduction

The Portland cement (PC) industry contributes 7% of the global carbon footprint, which is due to its high fossil fuel consumption and CO₂ emission from mineral decomposition [1–5]. Accelerated carbonation of cement-based construction materials has been reported to efficiently enhance the mechanical properties of concrete products and substantially sequester CO₂ [6,7]. Reactive MgO (r-MgO) is a potential construction material that can be used as a Portland cement (PC) replacement to both obtain a satisfactory early age strength and sequester CO₂ at later ages [8–11]. Other than conventional calcination of mined magnesite (MgCO₃) or dolomite (CaMg(CO₃)₂), the technology of recovering Mg²⁺ from seawater and brine water provides a solution to manufacture r-MgO in a more ecofriendly manner [12,13]. The chemical reactions in accelerated carbonation (CO₂ concentration > 50%) cured r-MgO and PC blends in literature [14–19] are shown as Eqs. (1) – (7):



Eqs. (1), (2) and (3) represent the major hydration processes of r-MgO, alite (C₃S – 3CaO·SiO₂) and belite (C₂S – 2CaO·SiO₂), which are stated to occur independently [14–18,20]. When carbonation curing is applied, Ca²⁺ from both Ca(OH)₂ and calcium silicate hydrate (C-S-H) will convert to calcite (CaCO₃) with Mg²⁺ incorporated, as shown in Eq. (6). The incorporation of Mg²⁺ in the crystal lattice of calcite results in the formation of magnesium calcite (Mg-calcite) [21,22]. Mg²⁺ that is not incorporated in calcite will be carbonated independently to form nesquehonite (MgCO₃·3H₂O) (Eq. (7)), which is the most stable Mg carbonate phase in a curing condition of ambient temperature (23 °C) and elevated CO₂ concentration (higher than atmospheric CO₂ concentration) [23–25]. Based on previously published literature [9,26–28], the occurrence of nesquehonite is usually regarded to benefit the strength gain and microstructure densification of paste and mortar.

* Corresponding author at: Dept. of Civil & Mineral Engineering, University of Toronto, 35 St. George St., Toronto, Ontario, M5S 1A4, Canada.
E-mail address: d.panesar@utoronto.ca (D.K. Panesar).

However, the current understanding of the reaction mechanism during the hydration and accelerated carbonation processes of the r-MgO-PC system is insufficient in two aspects: (1) the role of Al-containing components in influencing the hydration and carbonation of r-MgO; and (2) the threshold for the formation of nesquehonite. In the hydration of PC, tricalcium aluminate (C₃A – 3CaO·Al₂O₃) as the major Al-containing component mostly reacts with gypsum (CaSO₄) and water forming ettringite (3CaO·Al₂O₃·3CaSO₄·32H₂O) and monosulfoaluminate (3CaO·Al₂O₃·3CaSO₄·12H₂O) as shown in Eqs. (4) and (5) [19,29,30]. With the presence of MgO in PC, hydrotalcite (Mg₆Al₂CO₃(OH)₁₆·4(H₂O)) has been reported during the hydration of a system containing PC and MgO/SiO₂ [31]. Hydrotalcite-like Mg-Al layered double hydroxides are often identified in the alkali-activated slag cement with moderate to high MgO content (> 5 wt.%) [32,33]. But in the r-MgO-PC system, there are no reports of hydrotalcite formation. In terms of the formation of nesquehonite, nesquehonite does not occur in mixes with ≤ 40% r-MgO replacement and only high Mg-calcite was reported as the Mg carbonate in the literature [9,26]. High Mg-calcite refers to calcite with MgCO₃ mole% ≥ 10% [21,22], which was also reported to form in the carbonated PC paste with a 20% r-MgO replacement [17]. However, it has been demonstrated that the formation of high Mg-calcite in hardened paste relies on the high degree of supersaturation with respect to the MgCO₃ established due to the existence of Mg carbonates, for example nesquehonite [34]. Therefore, in the carbonated r-MgO-PC blends where high Mg-calcite forms but no nesquehonite forms (for example carbonated mixes with ≤ 40% r-MgO replacement in [26,27]), some unreported Mg carbonates (undetectable by X-ray diffraction (XRD)) are expected to provide the high degree of MgCO₃ saturation for the formation of high Mg-calcite. In this circumstance, the existence of this/these Mg components will improve our understanding on the carbonate composition (mainly Mg calcite and nesquehonite) of the r-MgO-PC system. Further characterization of the phase change in the r-MgO-PC system during hydration and carbonation will facilitate the application of r-MgO as a greener (lower carbon footprint) construction material compared to PC.

The objectives of this study are two-fold: (1) to characterize the quantity change of Mg-calcite and nesquehonite as the r-MgO replacement level ranges from 10% to 90%; and (2) to explore the Mg components that form during the hydration and carbonation of a r-MgO-PC system. Blends of r-MgO and PC with r-MgO weight percentages from 10% to 90% were cast and carbonated to examine the phase change during hydration and carbonation. r-MgO replacement levels were selected to identify the threshold r-MgO replacement for nesquehonite formation. Thermogravimetric and differential thermal analysis (TG/DTA), XRD, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were utilized to identify and describe the Mg components that form at different stages of the reaction.

2. Experimental program

2.1. Material

Type GU (general use) PC, manufactured by Holcim Canada (now CRH), and r-MgO with a reactivity of 68 s (determined by the accelerated reactivity test from [15]), which was supplied by Liyang Special Materials Company in China, were utilized as the binder for the casting of r-MgO-PC paste. r-MgO with a mode particle size ≤ 22 μm and a specific surface area of 22.2 m²/g, is a calcinate of MgCO₃ at approximately 800 °C. The chemical compositions of r-MgO and PC are presented in Table 1. The major components of PC include C₃S, C₂S, C₃A and tetracalcium aluminaoferrite (C₄AF – 4CaO·Al₂O₃·Fe₂O₃), while r-MgO consists of MgO and some minor quantities of quartz (SiO₂) and anhydrite (CaSO₄), as indicated in Fig. 1.

Table 1

Chemical composition of r-MgO and PC (weight%).

	MgO	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Na ₂ O	K ₂ O	SO ₃	Loss on ignition
r-MgO	88.52	3.18	2.76	0.42	0.26	0.06	0.03	0.61	3.62
PC	2.33	61.48	19.19	5.35	2.38	0.23	1.14	4.07	2.46

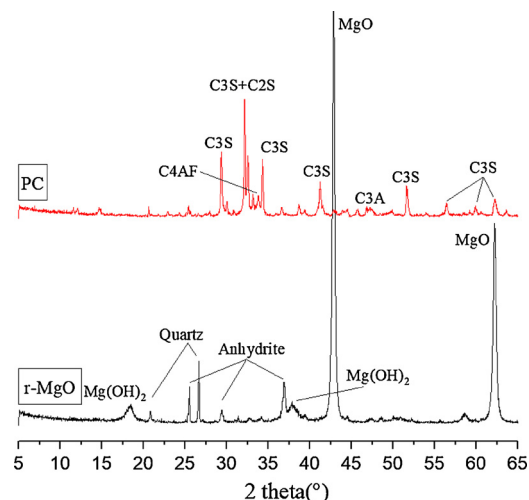


Fig. 1. XRD patterns of PC and r-MgO.

2.2. Sample preparation and curing condition

Nine r-MgO and PC blends with r-MgO replacement levels from 10% to 90% were designed to characterize the Mg component change during the hydration and carbonation in the r-MgO-PC blends. As presented in Table 2, the water-to-binder (w/b) ratio varies, because the high fineness of r-MgO particles increases the water-demand of the mixes [35]. Precisely weighed r-MgO and PC powders were mixed for two minutes. After the powders were homogeneously blended, water was gradually added. Meanwhile, the paste was manually mixed by the beater of the mixer. After all the water was added, the beater was applied on the mixer. After all the water was added, the beater was applied on the mixer. After all the water was added, the beater was applied on the mixer. Fresh paste was cast into 25 × 25 × 25 mm cube moulds (3 cubes for each mix), covered with plastic films and cured in a plastic box with frequently sprayed water. Taking the paste with a 50% r-MgO replacement as an example, 40 g of r-MgO, 40 g of PC and 40 g of water were used for the sample preparation. At 24 h of age, the cubes were demoulded and left in an ambient lab environment (temperature 20 ± 3 °C and relative humidity 50 ± 5%) for one day to dry. Eventually, all the mixes were subjected to a CO₂ curing chamber. Only one curing condition (95% relative humidity and 99% CO₂ concentration) was chosen to accelerate the carbonation rate, since a combination of high relative humidity and CO₂ concentration was found to facilitate the carbonation of r-MgO system [27,34,36]. Samples were tested after 14, 28 and 56 days of exposure to the carbonation environment. For the r-MgO-PC blends without carbonation curing, samples were cured with plastic films covering in an environment with 95% relative humidity until the 16 days.

At each testing age, half of the cubes of each mix were removed from the curing chamber and crushed into small pieces with a maximum diameter of 5 mm. The small pieces were then submerged into 20 ml of isopropanol in a sealed container with the isopropanol replaced twice in 3 h. The samples were then taken out of the isopropanol and dried in a vacuum oven (35 °C) until constant mass achieved. Dried samples were then ground to powders for XRD and TG/DTA analysis with a maximum diameter less than 38 μm. Since each powder sample was taken from half of a cube (cut, dried and ground into powder) of

Download English Version:

<https://daneshyari.com/en/article/10999962>

Download Persian Version:

<https://daneshyari.com/article/10999962>

[Daneshyari.com](https://daneshyari.com)