



Combined impact of silicate-amorphicity and MgO-reactivity on the performance of Mg-silicate cement

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HIGHLIGHTS

- MgO, ceramic and glass wastes have been used for preparing Mg-silicate cement.
- The reactivity of MgO produced at 800 °C is higher than that produced at 1200 °C.
- Glass waste showed amorphous content higher than ceramic waste.
- Different methods were used to evaluate the performance of the prepared cement.
- The performance of Mg-silicate cement mainly depends on silicate and MgO reactivity.

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ABSTRACT

This work investigated the synergistic impact of silicate- and MgO-reactivity on the performance of Mg-silicate-based cement. Ceramic waste (CW) and glass waste (GW) as two silicate sources were used. Different decarbonation temperatures (800 and 1200 °C) were applied on magnesium carbonate to yield MgOs with different reactivities (MgO₈₀₀ and MgO₁₂₀₀). Highest strength and shortest setting times associated with an enhancement in silicate dissolution rate and the formation of large magnesium silicate hydrate (MSH) were recorded in case of GW-MgO₈₀₀-H₂O system. The use of MgO₁₂₀₀ resulted in the retardation of MSH-formation rate in both of GW-MgO-H₂O and CW-MgO-H₂O systems. Comparing with GW-MgOs set, the CW-MgOs one showed the lowest performance at all curing ages. Due to high alumina content in CW, secondary hydroxalcalite-like phases were detected in CW-MgOs alongside MSH-phase.

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1. Introduction

Portland cement (PC) is the dominant form of cements used worldwide, with approximately 4100 billion tones produced in 2017 [1]. PC was manufactured by the calcination of a homogeneous mixture of limestone and clay at elevated temperature (~1450 °C) in rotary kiln, yielding solidified clinker, followed by crushing with 3–5 mass% gypsum. PC manufacturing is considered as one of the main industries which contributes in environmental pollution; where one tonne of cement generated approximately one tonne of CO₂, SO_x and NO_x as well as 60 kg of cement kiln dust [2,3]. Magnesia-based cements take a great interest due to their low production energy (temperature below 1000 °C) and ability to develop strength via carbonation [4–8]. Moreover, it can be beneficially used in different applications ranging from refractory

stable [9,10] to waste encapsulation [11]. Magnesium oxychloride [12–15], magnesium oxysulfate [16–19], magnesium phosphate [20–24], and magnesium silicate hydrate [25–31], are the common binders of Mg-based cements. Magnesium silicate cement regarded as an economic cement, which can be prepared by the activation of amorphous silicates (silica fume, fly ash, and ground granulated blast furnace slag) by reactive MgO at room temperature [32–34].

The mechanism of MSH-formation includes dissolution and precipitation processes [35]. The dissolution of amorphous silica was done at pH >9, resulted from the hydration of MgO {brucite, Mg(OH)₂}, yielding activated silicate species which interact with magnesium ion to produce MSH-binder. Therefore, the rate of MSH-formation strongly depends on the availability of Mg²⁺ and OH⁻ [26]. The performance of Mg-silicate based cement mainly depends on the curing conditions [30,36] and Mg/Si ratio [37,39]. Sodium hexametaphosphate (Na₆MP), as water reducer agent

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and fluidity enhancer to Mg-based cement, have been previously used [39,40].

The novel contribution of this work is to study the synergistic impact of silicate amorphicity and MgO-reactivity on the performance of magnesium silicate cement. Glass waste and ceramic waste, as silicate materials with different amorphicity have been individually blended with MgOs with different reactivities in presence of sodium hexametaphosphate. The performances of Mg-silicate cements were assessed by setting times and compressive strength testing at different hydration ages. The hydration process was evaluated by X-ray diffraction (XRD), thermogravimetric analysis (TG/DTG) and Fourier transform infrared (FTIR) spectroscopy. Meanwhile, the cement microstructure was investigated via field emission scanning electron microscopy (FESEM).

2. Experimental program

The materials used in this investigation were magnesium oxide (MgO), ceramic waste (CW), glass waste (GW), and sodium hexametaphosphate (Na₆MP). MgOs with different reactivities were prepared by decarbonation of magnesium carbonate (MC) at temperatures of 800 and 1200 °C for 2 h, which labeled as MgO₈₀₀ and MgO₁₂₀₀. Magnesite was obtained from El-Gomhouria Company, Cairo, Egypt. CW was collected from Gloria Company for ceramic industry, Beni-Suef, Egypt. GW was obtained from Sphinx Glass Ltd Company, Giza, Egypt. Na₆MP with purity of 99.99% was purchased from LOBA Company, India. The chemical oxide analyses of starting materials which conducted by X-ray fluorescence spectrometer (Xios, PW1400) are given in Table 1. The X-ray diffractograms (Fig. 1) proved that the crystallinity degree of MgO increased with decarbonation temperature. Acetic acid neutralization test [41], was carried out to assess the MgO reactivity. In this test, 5 g of MgO was dispersed in 100 ml of 0.25 M acetic acid; phenolphthalein was added as a pH indicator. The reactivity was determined by the recording time at which the solution changed to pink color. The longer the time, the low MgO reactivity is. The test was conducted in duplicate for each MgO and the average value was calculated. Solution color was changed in 41 sec in case of MgO₈₀₀; the longer time (136 s) was recorded when MgO₁₂₀₀ dispersed in acetic acid solution. This is an indication of the higher MgO₈₀₀ reactivity comparing with MgO₁₂₀₀. As shown in Fig. 2, CW-diffractogram showed the aluminosilicate amorphous structure with the formation of crystalline phase affiliated to quartz and albite minerals. Meanwhile, a completely vitreous structure with hump at 25–35 2θ° was observed in case GW-pattern. The Rietveld analysis of XRD [42] demonstrates amorphicity percentages of 31 and 100% in case of CW and GW, respectively. Particle size distribution (Fig. 3) indicated that the mode particle sizes of MgO₈₀₀ and MgO₁₂₀₀ are ~10 and ~18 μm. This shows the effect of temperatures rising on MgO-sintering.

Before the preparation of cement paste, GW and CW were individually ground to fine powder to pass through 75 μm sieve. Where, the surface areas of CW and GW powder were 3410 and 3440 cm²/g, respectively. After crushing, GW- and CW-powders were individually blended with MgOs (MgO₈₀₀ or MgO₁₂₀₀) at weight ratio of 50:50. Na₆MP with 1 wt, % of solid content was

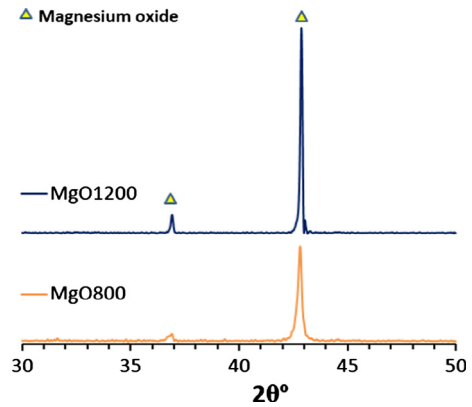


Fig. 1. XRD-patterns of MgOs prepared from the magnesite calcination at 800 and 1200 °C.

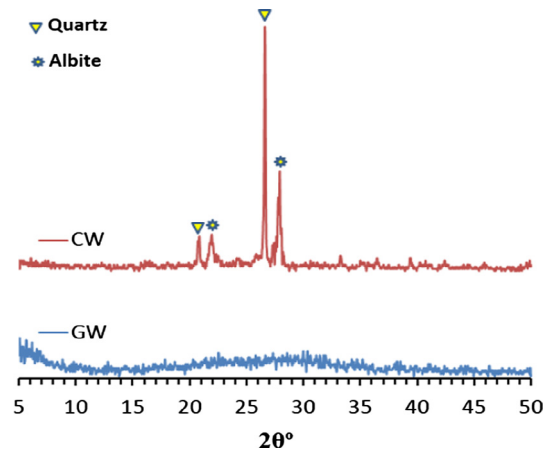


Fig. 2. XRD-patterns of ceramic and glass wastes (CW&GW).

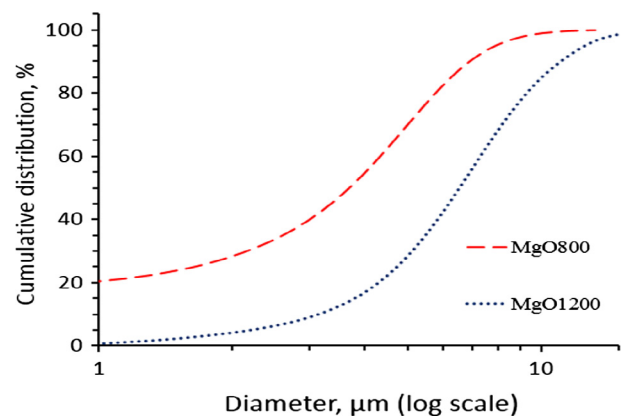


Fig. 3. Particle size distribution of MgO₈₀₀ and MgO₁₂₀₀.

Table 1

Chemical oxide analyses of starting materials (wt, %).

Samples	Oxides								
	SiO ₂	CaO	MgO	Fe ₂ O ₃	Al ₂ O ₃	SO ₃	Na ₂ O	K ₂ O	loss
GW	88.42	2.14	0.63	1.29	0.81	0.59	4.28	0.27	0.34
CW	65.13	0.89	1.16	4.26	21.15	2.35	1.29	1.39	0.72
MC	0.04	0.02	47.25	0.02	0.06	0.01	61.78	0.12	51.45

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