



Microstructure development and mechanism of hardened cement paste incorporating graphene oxide during carbonation

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ABSTRACT

In this work, the carbonation mechanism of cement paste containing graphene oxide (GO) was examined by evaluating its electrochemical, kinetic, and microstructural characteristics under accelerated carbonation, corresponding to a 20% concentration of CO₂, a temperature of (30 ± 1)°C, and a relative humidity of 65%–70%. Transport properties of the composites were studied using a non-destructive electrochemical impedance spectroscopy (EIS) technique, while their carbonation kinetics was investigated via thermogravimetric analysis (TGA). The obtained EIS results indicated that the ion diffusion and transport resistance increased after the incorporation of GO, while TGA results revealed that the carbonation of portlandite (CH) and calcium-silicate-hydrate (C-S-H) was significantly inhibited during early ages of carbonation due to the increased degree of hydration. In addition, the hybrid GO/hydration products from the carbonation process were characterized, and the formation of a hydrated phase coated with a carbonated layer was observed via scanning electron microscopy and energy dispersive spectroscopy. Porosity variations of the studied materials during carbonation were also evaluated using a mercury intrusion porosimetry method. The porosity of the OPC decreased more significantly during the initial carbonation period as compared to the effect observed for the GO cement-based material.

1. Introduction

Cement-based materials are extensively used worldwide in civil infrastructure, which is often exposed to harsh environments and severe loading conditions. The durability of structures in aggressive environments is a key parameter that strongly affects their service lives and maintenance costs. In particular, carbonation of cement-based materials represents a serious problem that reduces their durability, and it is commonly considered a neutralization phenomenon inside hardened cement-based materials. Carbonation involves the reaction between the carbon dioxide and calcium-containing phases, which reduces the pH of the pore solution [1–3], thus leading to de-passivation of reinforcing bars and corrosion in the presence of moisture and oxygen [4–7].

The hydration products interacting with dissolved CO₂ mainly consist of portlandite (CH) and calcium-silicate-hydrate (C-S-H). Although CH is expected to be more susceptible to carbonation from a thermodynamic point of view, the carbonation processes of CH and C-S-H occur simultaneously [8–10]. In previous studies, the morphology of calcium carbonate products (including calcite, vaterite, and aragonite

minerals [11–13]) and amorphous calcium carbonate formed under various conditions has been investigated [14–17]. The obtained results demonstrate that vaterite and aragonite are usually formed at larger CO₂ concentrations, low pH of the cement matrix, or low C/S ratio of the C-S-H exposed to CO₂ gas. It has also been found that carbonation produces significant changes in the microstructure of cement-based materials. According to the results of a number of studies [18–20], the capillary pores with sizes ranging between 10 and 300 nm can be reduced by carbonation, causing a systematic reduction in total porosity. It has been found [15] that the observed decrease in total porosity of cement-based materials results not only from the CH carbonation, but also from the decalcification and polymerization of C-S-H and formation of amorphous silica gel.

Recently, several researchers have attempted to apply nanotechnology to cement-based materials to improve their mechanical performance and durability [21,22]. It is known that nanoparticles can fill the voids in the cement matrix and enhance the suppression of the ion transport, and thus improve its durability, including the resistance of carbonation [23–29]. Among various nanomaterials, graphene oxide

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(GO) [30–34] and graphene nanoplatelets (GNPs) [28,35,36] represent unique two-dimensional (2D) nanomaterials with a sheet-like structure that can effectively improve the strength and durability of cement-based materials and ensure a relatively large contact area with the host matrix [37,38]. Furthermore, the carboxylic functional groups of GO can form strong covalent bonds between the GO and C-S-H or CH [33,39].

Several studies have been conducted to investigate the microstructural and transport properties of cement-based materials containing 2D nanoparticles. According to Liu et al. [40], Du et al. [28,41], and Mohammed et al. [27], the addition of GNPs can reduce ingress of chloride ions in the cement matrix and, therefore, improve its corrosion resistance. According to Mohammed et al. [42], GO acted as air-entraining admixture by forming small pores in the cement matrix, which can improve the frost resistance of cement-based materials. By performing atomic modeling, Teng et al. [43] found that the migration of water molecules in the C-S-H interlayer pores of cement-based materials can be much faster than that in the pores of GO cement-based materials, while the cyclic loading of the former materials was much higher during freeze-thaw cycles. From these results, it can be concluded that the sheet-like structure of GO and GNP can inhibit the diffusion and migration of ions in the cement paste. However, studies in which 2D nanoparticles are used to improve the transport property of cement-based material remain quite limited. In particular, few efforts have been focused on the effect produced by GO addition on carbonation.

The objective of this study was to investigate the influence of the GO addition (with contents of up to 0.2 wt%) on the carbonation mechanism of cement-based materials. The diffusion and transport of ions in the GO-containing composites under accelerated carbonation conditions were evaluated using an electrochemical impedance spectroscopy (EIS) technique. Since the pore structure of cement-based materials can significantly affect their transport properties, and hence carbonation, mercury intrusion porosimetry (MIP) was employed to evaluate the pore structure. In addition, the effect of the GO addition on the carbonation of C-S-H and CH was examined using a thermogravimetric analysis (TGA) technique. The barrier effect of the resulting hybrid hydration products on the carbonation of the studied materials was characterized using a scanning electron microscopy (SEM) instrument equipped with an energy dispersive spectroscopy (EDS) module. By synergistically performing the electrochemical, chemical, and microstructural experiments, systematic analysis results describing the carbonation behavior of the GO-containing cement-based materials were obtained.

2. Materials and methods

2.1. Materials

Ordinary Portland cement (PI 42.5R) conforming to the requirements of Chinese Standard GB175 [44] was employed. The chemical composition and physical properties of the cement are listed in Table 1. The particle-size distribution of the cement was determined using a laser particle-size analyzer, as shown in Table 2.

Graphite oxide powder was used for the preparation of a GO nanosheet solution. The properties of the graphite oxide are given in Table 3. A combination of the polycarboxylate-based, high-range,

Table 1
Chemical compositions and physical properties of cement.

Chemical compositions	Ingredient Content (mass %)	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	SO ₃	K ₂ O	Na ₂ O	LOI
		64.42	20.52	5.62	3.78	2.11	2.10	0.28	0.20	0.87
Physical properties	Specific surface area (m ² /g)	ρ_0 (g/cm ³)	Setting time (min)		Flexural strength (MPa)		Compressive strength (MPa)			
	0.581	3.00	Initial	Final	3 d	28 d	3 d	28 d		
			112	145	6.50	9.20	34.80	58.00		

Table 2
Particle-size distribution of cement.

Materials	Cement (mass %)	
Sieve opening (μm)	Passing (%)	Accumulation (%)
0.100–0.211	0.52	0.52
0.211–0.498	2.58	3.10
0.498–1.054	5.08	8.18
1.054–2.003	4.99	13.17
2.003–5.251	13.16	26.33
5.251–9.983	15.16	41.49
9.983–21.12	26.87	68.36
21.12–40.15	23.23	91.59
40.15–84.95	8.37	99.96
84.95–161.40	0.04	100

Table 3
Properties of graphite oxide.

Appearance	Solid content (mass %)	pH	Viscosity (Pa-s)	Absorbance ratio A230/A600	Carbon (mass %)	Molar ratio (O/C)
Brown paste	43 \pm 1	≥ 1.2	≥ 2	≥ 45	47 \pm 5	0.6 \pm 1

water-reducing admixture (PC) was used to improve the fluidity of the fresh GO/cement composites. The PC-based superplasticizer conforms to the requirements of Chinese Standard JG/T223 [45]. The rates of maximum water reduction of the PC-based superplasticizer are in the range 30%–35%. The characteristics of the chemical admixtures are shown in Table 4.

2.2. Methods

2.2.1. Preparation and characterization of GO nanosheets

To prepare GO nanosheets, a specified amount of graphite oxide powder was mixed with deionized water for 30 min using a magnetic stirrer. The resulting aqueous suspension of graphite oxide with a concentration of 5 g/L was sonicated with an ultrasonic homogenizer for 2 h. The ultrasonicator was operated for 2 s with 4-s intervals between cycles to avoid suspension overheating. After ultrasonication, the morphology of the obtained GO nanosheets was verified using atomic-force-microscopy (AFM) and transmission-electron-microscopy (TEM) techniques, while their chemical-bonding characteristics were examined using a Fourier-transform infrared (FTIR) spectrometer.

2.2.2. Mixture proportioning and specimen preparation

During the fabrication of GO/cement-paste composites, different PC-to-GO mass ratios were used to ensure similar fluidity of all samples. First, a mixture of PC with half of the aqueous dispersion was thoroughly stirred. After that, cement was added to the PC/aqueous dispersion and mixed using a mixer for 1 min. This was followed by the addition of the remaining half-portion of the aqueous dispersion and mixing for another 1 min. Finally, the obtained GO/cement paste was cast into two molds with dimensions of 10 \times 10 \times 10 mm³ and 30 \times 30 \times 30 mm³. After demolding, the specimens were cured in a moisture room at 20 °C and 95% relative humidity for 28 d. The

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