



## Early-age interaction mechanism between the graphene oxide and cement hydrates



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### HIGHLIGHTS

- Early-age interaction between the GO and cement hydrates was revealed.
- Ca<sup>2+</sup> cations can be captured by the GO at the very beginning of mixing.
- GO accelerates the cement dissolution into aqueous solution.
- GO can restrict the drying shrinkage by densifying matrix and self-curing effect.

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### ABSTRACT

The remarkable properties of graphene oxide (GO) make it as an ideal candidate for developing high performance cementitious composites. In this study, it is the first time to investigate the early-age physical adsorption and chemical interactions between the GO and cement hydrates, which have a great influence on the hydration development, microstructures and drying shrinkage of the GO modified cement paste. The experimental results indicated that the addition of GO led to the decreased fluidity of cement paste, resulting from the rapid interaction between the GO and divalent metal cations released from cement hydrates, characterized by Ultraviolet-visible spectroscopy and X-ray photoelectron spectroscopy. It was found that 0.08 wt% GO improved the cement hydration rate/heat by accelerating the cement dissolution, providing nucleation site and regulating the microstructure of cement hydrates. The GO covered cement hydrates at the hydration of 10 min was characterized by scanning electron microscopy. Moreover, the addition of GO restricted the 28-day drying shrinkage of cement paste due to the more compacted microstructures and self-curing effect at the early age. In conclusion, the research finding gives a thorough understanding on early-age interaction mechanism when the GO is immediately mixed with cement particles, and provides a valuable guidance to the sustainable design of GO modified cementitious materials for construction use.

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## 1. Introduction

Ordinary Portland cement is one of the major components in conventional concrete, which is the most widely used construction building materials for various types of civil infrastructure. The brittleness and lack of tensile strength are the two major shortcomings of cement-based materials [1–4]. Many attempts have been conducted to improve the toughness and tensile properties of cement-based materials by adding macro fibers and steels

[5,6]. However, most of commonly used macro fibers are organic-based materials, which not only have a weak bonding to inorganic cementitious materials, but also cannot restrict the propagation of the nano- or micro-cracks in cement matrix due to the fiber size effect [7]. Recently, extensive research has investigated the effect of nanomaterials and nanotechnology on macro-properties of cement-based materials. For example, nanomaterials [8,9], such as nano-silica [10,11] and carbon nanotubes (CNTs) [12,13], can significantly enhance the mechanical properties of cement-based materials due to their nano-filling, nucleation effects as well as greater chemical reactivity, and the defects or cracks in nanoscale level can be restricted by adding these novel nanomaterials.

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Parveen et al. [14] indicated that flexural and compressive strength of functionalized CNTs reinforced cement mortar increased up to 17% and 23% at 56 days, respectively, and with a moderate amount of surfactants, the CNTs reinforced cement mortar exhibited higher stiffness, fracture energy and ductility as compared to plain cement mortar. In addition, Hou et al. [15] showed that 5.0 wt% nano-TiO<sub>2</sub> not only accelerated cement hydration process in the first 2 h, but also improved the water retention property and volume stability of hardened cement paste.

Since 2004, graphene-based materials [16], including graphene [17–19], graphene oxide (GO) [20] and reduced graphene oxide [21], have been rapidly developed due to its numerous unique properties, including outstanding tensile strength of 130 GPa, excellent Young's modulus of 0.5 TPa and large specific surface area of 2630 (m<sup>2</sup>/g). The oxygen-containing functional groups of GO, such as hydroxyl, epoxy and carboxyl, make it more easier to be dispersed in aqueous solutions, which is an important prerequisite for better dispersion of GO in composite materials. Therefore, GO has widely been used in energy storage materials [22], photocatalyst materials [23], semiconductors [24] and biological composites [25]. Recently, many studies have addressed that the addition of GO contributed to the mechanical enhancement of cement-based materials. Li et al. [26] indicated that incorporation of 0.04 wt% GO increased the tensile strength by 67% compared to that of plain cement paste. Lu et al. [27] demonstrated that 11.1% and 16.2% increase in compressive and flexural strength of cement paste can be achieved by incorporating 0.05 wt% GO, and it also showed the effect of the hybrid GO/CNTs composites on the mechanical behavior of cement paste. Li et al. [28] showed that with the help of silica fume, a more obvious mechanical enhancement of GO reinforced cement paste can be obtained due to the better dispersion of silica fume covered GO in cement matrix. It should be pointed out that compared with CNTs, GO shows more obvious and stable mechanical reinforcement to cementitious materials due to the better mechanical properties itself and good dispersion in aqueous solutions. Therefore, it is clearly seen that GO plays an important role in enhancing the mechanical behavior of cement-based materials by regulating the microstructure and hydration process effectively.

**Table 1**  
Chemical composition of cement.

CaO	SiO <sub>2</sub>	SO <sub>4</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	K <sub>2</sub> O	TiO <sub>2</sub>
65.40	19.47	5.71	3.86	3.0	1.58	0.49	0.26

However, most of earlier works have focused on the microstructures and mechanical behavior of GO reinforced cement-based materials at the later age (14 or 28 days), few studies investigated how is the interaction between the GO and cement hydrates, such as the physical adsorption and chemical interactions, when they are mixed together at beginning. A vital issue on 'how does the early-age interaction between the GO and cement hydrates influence the later-age properties of cement paste' has not been settled. In this study, the early-age physical adsorption and chemical interactions between the GO and cement hydrates were firstly investigated by an adsorption test, Ultraviolet-visible (UV-vis) spectroscopy and X-ray photoelectron spectroscopy (XPS). The microstructures of GO covered cement particles at the very beginning (6 min) of cement hydration were observed using scanning electron microscopy (SEM). Then, the real-time monitoring of hydration process of the GO modified cement paste was conducted by isothermal calorimetry. Finally, the effect of GO on the 28-day drying shrinkage was investigated using a Demac gage. This paper aims to give a thorough understanding on early-age interaction mechanism between the GO and cement hydrates, and provides a valuable guidance to the sustainable design of GO reinforced cementitious materials for construction use.

## 2. Materials and methods

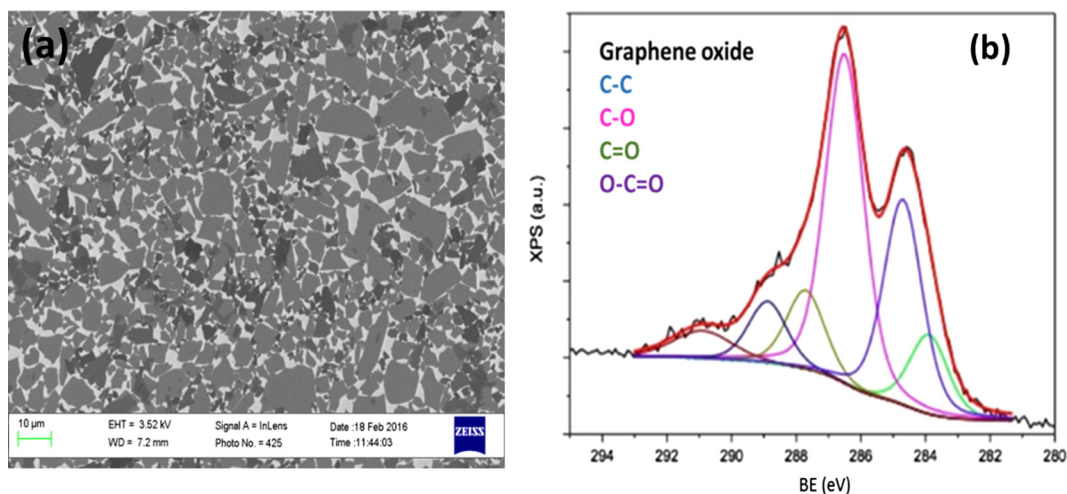
### 2.1. Materials

Ordinary Portland cement (Type 52.5, Green Island, HK) was used in this study and the chemical composition measured by X-ray fluorescence is listed in Table 1. The GO (C/O ratio of 1–1.4) aqueous solution with 4 mg/mL was purchased from Graphenea<sup>®</sup>, Spain. Fig. 1 shows the morphology and oxidation degree of GO characterized by SEM and XPS. The GO aqueous solution was ultra-sonicated by a Sonics Vibra-Cell vcx-500 ultrasonic processor (Vibra-Cell-Sonics & Materials, Inc.) with the amplitude of 30% for 15 min before use to ensure the uniform dispersion of GO.

### 2.2. Methods

#### 2.2.1. Sample preparation

Table 2 lists four mix proportions of the GO modified cement paste with w/c ratio of 0.35. The dosage of GO varies from 0.02, 0.04 to 0.08% by the weight of cement. The aqueous solution of GO was firstly ultra-sonicated for 15 min and then gradually mixed with cement powders in high mixing speed for 8 min before cast



**Fig. 1.** (a) SEM and (b) XPS results of the GO (<http://www.graphenea.com>).

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