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Incorporation of graphene oxide and silica fume into cement paste: A study of dispersion and compressive strength

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HIGHLIGHTS

- Dispersions of graphene oxide in pore solution and cement paste was studied.
- Mechanism of GO aggregation in cement paste is cross-linking of calcium cations.
- Silica fume is effective to improve GO dispersion in cement paste.
- Appropriate amount of silica fume improves GO dispersion and compressive strength.

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ABSTRACT

As a 2D nanomaterial, graphene oxide (GO) has attracted considerable attentions for reinforcing cementitious materials. However, special attention must be paid to the dispersion of GO in the matrix, as the quality of nanomaterial dispersion correlates directly with its effectiveness for improving mechanical and other properties. In this study, the dispersion of graphene oxide (GO) in simulated pore solution and cement paste was investigated. It was found that severe GO aggregation occurred in presence of divalent calcium ions in both pore solution and cement paste. However, the GO aggregates were not stable under shear mixing. After vigorous mixing, the massive GO aggregates split into medium-sized particles, ranging from few to several 100 μm. To improve the GO dispersion in cement paste, silica fume was used to mechanically separate individual GO nanosheets. The dispersion was then investigated using microstructure analysis and mechanical properties. The results showed that, with the addition of silica fume, the dispersion of GO nanosheets was greatly improved.

1. Introduction

Cement is the principal binder material in the production of concrete, which is the most widely used construction material in the world. However, the major limitation of concrete and cement based materials is their quasi-brittle nature, which is blamed for its poor resistance to crack formation, low tensile strength and strain capacities.

Recently discovered graphene oxide (GO) is a layered nanomaterial consisting of hydrophilic oxygenated graphene sheets, bearing hydroxyl and epoxide functional groups on their basal planes and having carbonyl and carboxyl groups located at the sheet edges [1]. Due to the presence of these oxygen-containing functional groups, GO can readily yield stable dispersions in water, consisting mostly of 1-nm-thick sheets [2]. In terms of mechanical properties, the elastic modulus and tensile strength of GO is around 32 GPa and 130 MPa [3,4], respectively. Most importantly, it can be synthesized in large quantities from inexpensive graphite powder. As such, GO has attracted much attention in various applications [3,5,6]. One of those applications is to reinforce cement based materials. Successful incorporation of GO into cement based materials can address the issues of cement based materials by virtue of its excellent mechanical properties, large specific surface area, large aspect ratio, and functional groups favorable for cement hydration products.

A few studies have investigated effects of GO nanosheets on mechanical and transport properties of cement based materials,

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including cement paste and cement mortar. Gong et al. [7] reported that the introduction of 0.03% by weight GO nanosheets into cement paste increased the compressive strength and tensile strength by more than 40%. Zhu et al. [8] reported that the introduction of 0.05% by weight of GO into cement paste increased compressive strength by 15–33% and flexural strength by 41–59%. Lv et al. [9,10] showed that mechanical properties were improved by the incorporation of GO in cement paste and mortar, respectively. To take advantage of the high surface area of GO, the transport properties of GO incorporated cement mortar were studied [11]. The findings revealed enhancements in water sorptivity and chloride penetration. It should be noted that these studies paid little attention to dispersion of GO nanosheets in the cement based materials.

Although the incorporation of GO improves the properties of cement based materials, its dispersion in cement based materials still remains unknown. Park et al. [12] reported that GO nanosheets can be chemically cross-linked by divalent cations to produce GO paper with much larger dimensions than that of original GO nanosheets. It is foreseeable that dispersion of GO would not be good due to abundance of calcium cations in fresh cement paste. Therefore, it is imperative to study the nature of GO dispersion and how to improve that dispersion, as the quality of nanomaterial dispersion in the matrix correlates directly with its effectiveness for improving mechanical and other properties.

In this study, the dispersion of GO in both simulated pore solution (saturated $Ca(OH)_2$ solution) and cement paste was first investigated. It was found that the stability of GO was greatly influenced by divalent calcium ions. Severe aggregation of GO occurred when it was introduced into the simulated pore solution (saturated $Ca(OH)_2$ solution) and cement paste. Therefore, GO dispersion must be improved to more effectively enhance both mechanical and other properties of cement based materials.

Silica fume is very fine amorphous silica produced by electric arc furnace as a by-product of the production of metallic silicon or ferrosilicon alloys [13]. It is a powder with particle size 100 times finer than that of Portland cement particles. It is also pozzolanic. Silica fume has been used in cement mortar and concrete and significant effects on the properties of the resulting materials have been identified [14,15]. These effects pertain to properties such as strength [16], permeability [17], and bonding strength with aggregates [18].

A few attempts have been made to improve the dispersion of carbon nanofibers (CNFs) and carbon nanotubes (CNTs) in cement paste by the incorporation of silica fume. Sanchez et al. [19] reported that silica fume facilitated the dispersion of CNFs by acting as wedges to mechanically separate the carbon nanofibers. Kim et al. [20] found that the dispersion of CNTs was improved by the incorporation of silica fume in cement paste, leading to enhancements in mechanical and electrical properties. Yazdanbakhsh et al. [21] reported that silica fume, if used in sufficient proportions, largely prevented the reagglomeration of CNFs in fresh cement paste.

In this study, a new mixing procedure was designed that used silica fume to pre-disperse GO nanosheets before mixing with cement. The GO dispersion was then investigated by both microstructure analysis and mechanical testing using scanning electron microscopy (SEM) and compressive strength, respectively.

2. Materials and sample preparations

2.1. GO

GO solution was purchased from Graphenea®. In the solution, GO nanosheets were dispersed in water at the concentration of 4 mg/ml. A SEM image of GO is shown in Fig. 1. The mean size of the GO nanosheets was around 1 μm. The major functional groups on the GO surface were found to be –OH and –COOH. Elemental analysis of the GO is shown in Table 1.

2.2. Cement powder

General purpose ordinary Portland cement (OPC) conforming to the requirements of Type I – Normal Cement, as defined by ASTM C 150, was used throughout the study. The chemical composition of the cement powder as determined by X-ray fluorescence is shown in Table 2.

2.3. Silica fume

The chemical composition of the silica fume as determined by X-ray fluorescence is shown in Table 3. The particle size distribution of silica fume as measured by a laser particle size analyzer (Cilas 1190) is shown in Fig. 2. The mean particle diameter is 0.37 μm, the surface area of the silica fume particles was around 20 m^2/g.

2.4. Cement paste preparation

A high-speed shear mixer (CTE Model 7000) was employed to prepare cement paste with GO and silica fume. Mixing procedures similar to those in ASTM C1738-11a were adopted:

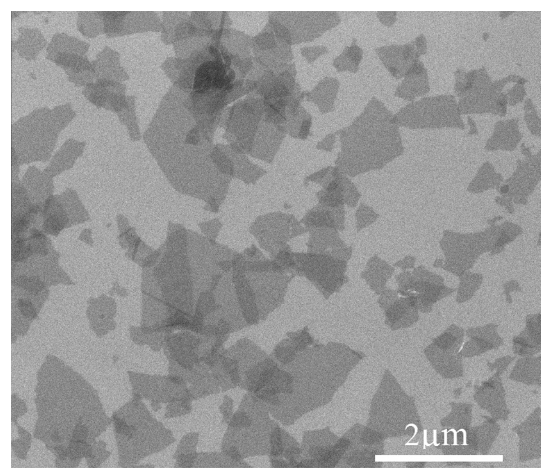

Fig. 1. SEM image of GO.

Table 1
Elemental analysis of GO.

Element	Carbon	Hydrogen	Nitrogen	Sulphur	Oxygen
%	49–56	0–1	0–1	0–2	41–50

Table 2
Chemical composition of cement powder.

Al_2O_3	SiO_2	CaO	Fe_2O_3	K_2O	MgO	Na_2O	SO_3	LOI
4.7	19.9	63.9	3.4	0.5	1.3	0.2	2.6	3.0

Table 3
Chemical composition of silica fume.

Al_2O_3	SiO_2	CaO	Fe_2O_3	K_2O	MgO	Na_2O	SO_3
–	96.43	0.78	0.073	1.28	0.70	–	0.38