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A short discussion on how to effectively use graphene oxide to reinforce cementitious composites

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HIGHLIGHTS

- Addition of PCE can pre-stabilize GO in ACS.
- Interaction between the silica fume and GO in ACS is revealed.
- GO is an effective coating material for fiber treatment.
- Cost of GO-Cement is calculated and compared.

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ABSTRACT

Graphene oxide (GO) with extraordinary properties has been regarded as a good candidate to reinforce cementitious composites. However, direct incorporation of GO as a nano-filler into cementitious composites not only hardly ensures the well dispersion of GO in cement matrix, but also greatly increases the cost of final cementitious products. In this study, three key points on how to effectively use GO to reinforce cementitious composites, including dispersion-effectiveness of GO in cement matrix, interaction-effectiveness of GO with supplementary cementitious materials (SCMs) and cost-effectiveness of GO reinforced cementitious composites, will be comprehensively discussed. Firstly, the mechanism of polycarboxylate ether (PCE) superplasticizer on pre-stabilizing GO in alkaline cementitious solution (ACS) is summarized and how effective of PCE on enhancing the mechanical performance of GO reinforced cementitious composites is analyzed. Secondly, interaction between the silica fume (one of the popular SCMs) and GO in ACS is investigated and the corresponding effect of silica fume on the dispersion of GO in ACS is discussed. Finally, a novel way using GO as a coating material for fiber treatment, such as carbon fiber, polyvinyl alcohol fiber and polyethylene fiber, is proposed, and it reveals that the cost of GO used as a coating material is 11 times lower than that of GO used as a nano-filler to reinforce cementitious composites, but it achieves a similar reinforcement to cementitious composites by strengthening the interfacial bonding between the fiber and cement matrix. In conclusion, the research outcomes give an important summary and guidance on how to effectively use GO to reinforce cementitious composites.

1. Introduction

Nanoscience and nanotechnology have been widely used and shown great potential for developing novel cementitious composites featured with sustainable, multifunctional and intelligent [1,2]. Carbon nanomaterials (CNs), such as carbon nanotubes [3], graphene oxide (GO) [4] and carbon nanofibers [5], have attracted wide spread research since it not only manipulates the structures and properties of cement hydrates at the nano-scale level, but also contributes to the development of multifunctional composites due to their superior properties [6–8]. Among various kinds of CNs, GO has been regarded as the most attractive candidate to reinforce cementitious composites, not only because of its numerous unique properties, such as outstanding tensile strength of 130 GPa, large specific surface area of 2630 (m^2/g) and high thermal conductivity (5300 $W{\cdot}m^{-1}{\cdot}K^{-1}$), but also the well dispersed of GO in aqueous solution due to the oxygen functional groups [9]. However, there is a big difference between the alkaline cementitious solution (ACS) and aqueous solution in terms of calcium (Ca^{2+}) concentration and PH value, and therefore how to ensure the well dispersion of GO in ACS would make a great influence on the properties of GO reinforced cementitious composites. In addition, with the rapid

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development of sustainable concrete, increasing amount of supplementary cementitious materials (SCMs), such as silica fume (SF, as an example discussed in this study) and fly ash, have been incorporated into concrete to replace a moderate amount of cement to reduce the CO_2 emission [10]. However, few investigations have been conducted to reveal the interaction between these SCMs and GO, which is also a key factor determining the performance of GO reinforced cementitious composites. Although some literature claimed that the incorporation of SF benefited to the dispersion of GO in cement matrix using scanning electron microscope and therefore improved the mechanical properties of cement paste [11,12], it still needs to be further investigated to provide a deeper understanding on the interaction between the SF and GO in ACS by characterizing their surface charge, particle size and morphology since only the SEM with high magnification in a very small field of view is insufficient to reflect the overall dispersion of GO in cement matrix. More importantly, although the mechanical enhancement can be easily achieved by adding a small amount of GO as a nano-filler, it also can be done by using others nano/micro-materials with much lower costs, such as nano-silica [13] and metakaolin [14], so the direct incorporation of GO into cement may not be the best way to reinforce cementitious composites due to such a high cost. Therefore, it is of paramount importance in finding a more cost-effective way to use GO to reinforce cementitious composites. To our best knowledge, surface treatment to fiber by GO has a great potential to strengthen the interfacial bonding between the fiber and cement matrix and therefore to reinforce the cementitious composites with enhanced ductility.

In this study, the effect of PCE superplasticizer on pre-stabilizing GO in ACS is firstly revealed, which is then correlated with the mechanical properties of GO reinforced cement paste. Secondly, the interaction between the silica fume and GO in the ACS is deeply understood in terms of dispersion by using zeta potential, scanning electron microscope and particle size analyzer. Finally, a novel way to use GO as a coating material to modify the surface property of fiber, such as carbon fiber, Polyvinyl Alcohol (PVA) fiber and polyethylene (PE) fiber, will be discussed, which can greatly reduce the amount of GO used in cementitious composite but dramatically improve the mechanical behavior by strength the interfacial bonding. In conclusion, the research outcomes give an important summary and guidance on how to effectively use GO to reinforce cementitious composites.

2. Materials

GO suspension with 4 mg/mL was purchased from Graphenea®, Spain. The PCE superplasticizer (ADVA 189) with a concentration of 28–30 wt% was purchased from Grace Concrete Admixture Products, Hong Kong. 52.5R Portland cement (Green Island, HK) and silica fume (Microsilica 920U) were supplied by Elkem Co., Ltd in Norway. Particle size distribution of SF dispersed in aqueous solution was shown in Fig. 1. PVA fiber (KURALON K-II REC 15), carbon fiber (Jiaxing Newtex Composites Co., Ltd., China) and PE fiber (Spectra® 1000, Honeywell) were used as the substrates for GO coating.

3. Experiment and characterization

Part 1): The pre-stabilizing effect of GO in ACS by PCE was investigated by comparing the Ca^{2+} concentration in the two suspensions of (GO + PCE + Cement + Water) and (GO + Cement + Water). The first suspension was fabricated by adding 2 mL of GO (4 mg/mL), 4 mL of PCE (solid conc. wt% = 28–30 wt%), 1 g of cement particle into 15 mL of distilled water in order. The similar dosage was used for preparing the second suspension. The concentration of Ca^{2+} was measured following the Standard Method [15] using an atomic absorption spectrophotometer (AAS, VARIAN 200FS). The corresponding flexural properties of the GO pre-stabilized by PCE reinforced cement paste (150 × 30 × 10 mm) were studied using 3-point bending test, following the procedure prescribed by ASTM C78/C78M-10 and measured with a span of 90 mm and a stroke control at a loading rate of 0.15 mm/min. Three specimens of each mix proportions were tested for obtaining the mean flexural strength value, together with the standard deviation.

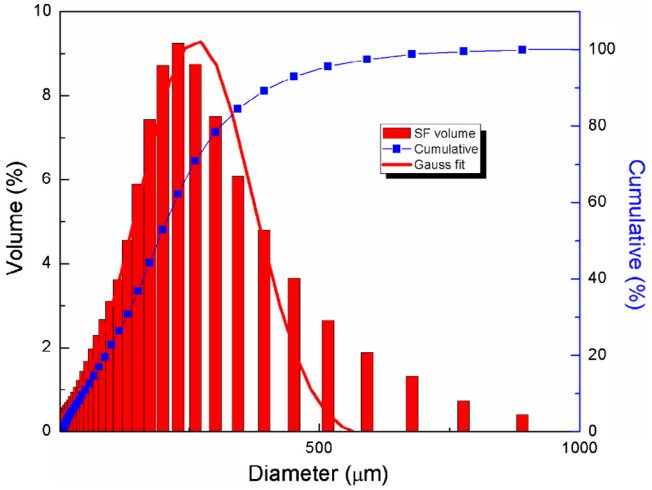

Fig. 1. Particle size distribution of SF in aqueous solution.

Part 2): Based on the experienced dosage of SF and GO to cement, 10.0 wt% and 0.05 wt%, the weight ratio of SF to GO in the current study is fixed at 200 for various characterization. Zeta potential was used to characterize the interactions between the SF and GO in ACS processing a PH value of 12.57, which was adjusted with a KOH buffer solution. Each test was run for 5 cycles and the average value was used to indicate the zeta potential of the testing suspensions. Dispersion morphology and particle size distribution of SF and SF/GO in ACS were measured by a laser particle size analyzer (90Plus/BI-MAS, Brookhaven) and SEM (JSM-6390, Jeol). A droplet of suspension was deposited on a monocrystalline silicon substrate and a thin layer (5 nm) of gold was sprayed after the evaporation of liquid in the suspension before the SEM characterization.

Part 3): Firstly, GO coated PVA fiber was fabricated by immersing PVA fiber into a uniform distributed GO solution, and followed by an ultrasonication mixing. The chemical bonding between the PVA fiber and GO was measured by using FTIR (Bio-Rad FTS 6000). For uniaxial tensile test, typical dog-bone shaped specimens were adopted and the dimensions of the sample are shown in Fig. 2. Both ends of the sample were closely attached in the machine and then loaded under displacement-control at a loading speed of 0.1 mm/min. Three specimens of each mix proportions were tested for obtaining the mean mechanical strength value. Composite was characterized by SEM (JEM-6390). Secondly, for GO coated carbon fiber, GO can be deposited on the surface of carbon fiber by electrophoretic depositing method [16]. The ITZ between the GO coated fiber and cement matrix was characterized by SEM. Surface wettability of the carbon fiber and GO/carbon fiber was measured by contact angle meter (DIGIDROP, GBX). For the 14-day mechanical properties measurement, specimens with dimensions of 40 × 40 × 40 mm were used for compressive test and 150 × 30 × 10 mm for three-point bending test. The compressive test was conducted using a MTS system at a loading of 2 kN/s. Four specimens of each mix were tested for obtaining the mean