



Dispersion of graphene oxide agglomerates in cement paste and its effects on electrical resistivity and flexural strength

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ARTICLE INFO

Keywords:

Graphene oxide
Dispersion
Cement
Electrical resistivity

ABSTRACT

Actual dispersion of graphene oxide (GO) in cement paste was investigated by using both X-ray computed tomography and X-ray photoelectron spectroscopy. It was found that GO nanosheets are mainly agglomerated, as an individual phase, with platelet-like morphology and little GO being absorbed onto surfaces of cement particles and hydration products. By performing an electrical resistivity test, GO agglomerates are found to be more electrically insulative than cement paste. Therefore, it is not possible to develop self-sensing cement composites by incorporating GO directly without resolving its dispersion issue. However, GO agglomerates enhance the flexural strength of cement paste because of their special morphology and intrinsic strength. Results showed that the flexural strength of cement paste was increased by 83% with incorporation of 0.04% GO by weight of cement.

1. Introduction

Graphene oxide (GO) is a two-dimensional nanomaterial derived from graphite by inserting several oxygen functionalities into the parent graphene backbone [1]. GO's superior mechanical properties, high dispersibility in water, and low production cost make it a promising reinforcing material for enhancing the properties of cementitious composites [2–5]. By conducting reactive molecular dynamics simulation, Hou et al. [6] investigated the intrinsic interactions between a GO nanosheet with cement hydration products. It was reported that, if GO could be dispersed in nanosheets form in a cementitious matrix, compared to graphene, GO incorporation endows the cement composite with a higher cohesive force and enhanced plasticity owing to the H-bonds and covalent–ionic bonds. However, it was conjectured that, when the GO nanosheets are introduced into the cementitious matrix, significant agglomerations would occur as the result of chemical cross-linking [7] by the calcium cations that are abundant in cement pore solutions. As reported in prior studies [8–10], GO agglomerates were observed in synthetic pore solutions or cement paste under unusually high GO dosage (0.4% and 0.8% by weight of cement). Under normal dosage of GO (0.02% and 0.04% by weight of cement), direct observation of the actual dispersion of GO agglomerates in cement paste has not been successful owing to the low content and poor contrast

between GO and cement hydrates under scanning electron microscopy (SEM). Therefore, the actual dispersion of GO under normal dosage in cement paste remains unclear.

From a dispersion perspective, GO incorporated into cement paste can be divided into two types: absorbed GO and individual GO. When GO nanosheets are introduced into cement composites, they may be absorbed onto surfaces of reacting cement particles and hydration products by the electrostatic attractive force. They also could exist as individual GO in the form of agglomerates or nanosheets. Individual GO could, theoretically, take the form of agglomerates or a single nanosheet. However, based on prior studies, single-sheet GO cannot exist in cement composites because of the chemical cross-linking of calcium cations [11,12]. Compared to observing GO dispersion by SEM, X-ray computed tomography (CT) is a nondestructive technique for visualizing features in the interior of opaque solid objects. In addition, relatively large three-dimensional (3D) samples can be used in CT to obtain a complete view of GO dispersion in cement composites by observing a large number of two-dimensional (2D) images, whereas, with SEM, only a very small area can be observed at one time. By using CT, dispersion of carbon fiber was quantitatively analyzed based on cross-sectional images of successive layers in cement composites [13,14]. The CT technique was also used to study the microstructure of nanoparticle-incorporated cement paste. The pore structure of cement paste

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incorporated with reduced GO (rGO) was investigated using 3D CT [15]. However, in Ref. [15], the resolution of 100 μm was not high enough to capture rGO dispersion information, except for pores and air voids. It should be noted that, with the CT technique, it is only possible to observe individual GO owing to resolution limitation (normally on the order of a micrometer). Moreover, X-ray photoelectron spectroscopy (XPS) might be helpful in detecting any GO nanosheet absorbed onto surfaces of hydration products or cement particles. Wang et al. [16] reported adsorption characteristics of GO nanosheets on cement particles. However, in Ref. [16], they added small amount of cement (0.09 g) into a GO solution (60 g) to study GO adsorption onto cement particles. The water to cement ratio was ~ 667 (60/0.09), which is much larger than usually adopted values (of < 1). Therefore, their adsorption characteristics might not reflect the actual case in cement paste incorporated with GO for normal water to cement ratio and GO dosage. Based on the above introduction and prior studies, it is viable to investigate the actual dispersion of GO in cement paste by conducting CT and XPS analysis to study individual GO agglomerates and absorbed GO, respectively.

In this study, we used the X-ray micro CT (X-ray μCT) technique with resolution extended to 1 μm to investigate the actual dispersion of GO agglomerates in cement paste. XPS analysis was conducted to investigate GO absorbed onto surfaces of hydration products or cement particles. In addition, GO agglomerates in cement paste were also observed by SEM on cement paste samples after ion polishing. Then, the effects of GO agglomerates on the electrical resistivity and flexural strength of the cement paste were investigated.

2. Materials and sample preparation

2.1. GO

Aqueous GO suspensions with concentration of 4 mg/ml were produced by Graphenea[®] (Spain). In the as-received GO suspension, no surfactant was used to stably disperse the GO owing to its hydrophilicity. Elemental analysis of GO showed a carbon to oxygen ratio of ~ 1.1 , suggesting a relatively high oxidation degree. The hydrodynamic size of GO nanosheets was measured by using a dynamic laser scattering method following a well-established procedure [17]. GO sizes were Gaussian distributed and the mean diameter of GO nanosheets was ~ 847.3 nm.

2.2. Cement powder

General purpose ordinary Portland cement (OPC) conforming to ASTM C150 was used throughout the study. The chemical composition measured by X-ray fluorescence (XRF) of the cement is listed in Table 1.

2.3. Preparations of cement paste samples

Cement paste samples were fabricated by using a high-speed shear mixer (CTE Model 7000). The mixing procedure similar to that in ASTM C1738-11a was adopted. Before mixing with cement powder, the correct amount of diluted GO solution was ultrasonicated for 5 min for improving the dispersion of GO nanosheets. Then, the solution was placed in the mixing bowl and mixing was started at 4000 rpm while the correct amount of cement powder was added to the GO solution over a 30 s period. Mixing was continued at 4000 rpm for another 30 s then switched to 12000 rpm for another 30 s of mixing. The mixer was

Table 1
Composition of cement powder.

Oxide	Al ₂ O ₃	SiO ₂	CaO	Fe ₂ O ₃	K ₂ O	MgO	Na ₂ O	SO ₃	LOI
%	4.7	19.9	63.9	3.4	0.5	1.3	0.2	2.6	3.0

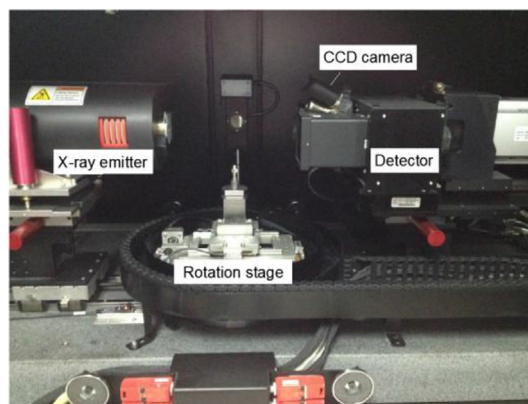


Fig. 1. Internal features of XRadia Micro XCT-400.

then switched off and the mixture was left to stand for 150 s before any material collected on the side of the bowl was scraped down. Final mixing for 30 s was done at 12000 rpm.

The water to cement ratio for all cement paste samples was fixed at 0.4. According to different GO dosage, the samples were denoted as CG_n, in which n refers to GO dosage. Four different GO dosages were applied in this study, 0.01%, 0.02%, 0.03%, and 0.04% by weight of cement. For example, CG4 refers to cement paste samples containing 0.04% GO by weight of cement. The reference sample without adding GO was denoted as C.

3. Experiments

3.1. X-ray μCT

Dispersion of GO agglomerates in cement paste were investigated by using an X-ray μCT system (XRadia Micro XCT-400) located at Shenzhen University (China). As shown in Fig. 1, the X-ray μCT system comprised a micro-focus X-ray emitter, a rotation stage that allows for 360° imaging, an image intensifier detector with three multiple charge-coupled device (CCD) cameras, and an image processing unit. The X-ray tube used in this study was equipped with a small focal spot (micro-focus X-ray-tube) and optical magnification enabled a high resolution. By means of X-ray μCT , the resolution of volumetric picture elements (voxels) was extended to 1 μm . In addition, the working distances between source, sample, and detector were typically ~ 100 mm in this system, which means full tomography can be achieved even for larger samples.

Cement paste samples C and CG4 were investigated by using X-ray μCT to study dispersion of GO agglomerates in CG4. Cylindrical C and CG4 samples were prepared by pouring a fresh C and CG4 mixture into a plastic tube with an inner diameter of 5 mm. Then the samples were vibrated to ensure good compaction. The samples were observed by X-ray μCT after curing for 7 days at a temperature of 20 °C and a relative humidity of 95%.

In this study, a 1024 × 1024 pixel X-ray camera was used while considering four elemental parameters. The applied X-ray source excitation was 80 keV and the current was 101 μA , as selected according to sample geometry and material compositions, while the magnification was 4×. The reconstructed image matrix has a volume of 1024 × 1024 × 1000, corresponding to 1000 slices of 1024 × 1024 pixels each, while respective pixel size and voxel size were 4.5413 μm and 4.5413 × 4.5413 × 4.5413 μm^3 , respectively. The X-ray absorption of each voxel, represented by the material-specific X-ray absorption coefficient, was normalized to 8-bit gray values.

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