



A REVIEW: THE EFFECT OF GRAPHENE OXIDE ON THE PROPERTIES OF CEMENT-BASED COMPOSITES

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Abstract: Graphene oxide (GO) is a recently invented 2D nanoplane fiber. GO is typically produced via the chemical oxidation and exfoliation of graphite. It contains active functional groups on its nanoplane surface, and these groups play a major role during the cement hydration process. Preliminarily, the hydration properties of GO–cement composites have been found to result in a higher hydration rate, which affects both the water demand and workability of the composites. Some authors have also reported that reinforcing the cement matrix with GO results in the formation of calcium silicate hydrate (C–S–H) gel in the micropores, thereby enhancing the resultant composite’s mechanical properties. Markedly few studies have examined the durability of GO–cement-based composites. This paper presents a critical review of the functionalities and effects of GO in cement-based composites, including its effects on hydration, workability, transport properties, the evolution of mechanical properties, and durability. This review also covers literature reports related to the life-cycle cost and the carbon footprint of such cement composites.

1 INTRODUCTION

The incorporation of reinforcing materials into concrete has become a common practice to improve its mechanical performance. Microfibers such as steel, glass, polymers, and carbon have been extensively studied for developing fiber-reinforced composites over the past several decades. Although microfibers enhance the ductility and toughness of the concrete matrix, their influences on compressive strength and durability are considered to be limited. The functionalization of carbon and polymer fibers enables them to form covalent bonds with the cement matrix; however, their small specific surface area limits their contribution to the interfacial strength (Wichmann et al. 2008). As such, nanomaterials can provide a better solution than traditional fibers through reinforcing at the nanoscale and allocating a much higher specific surface area for cement matrix interaction. Some nanomaterials even exhibit pozzolanic characteristics by consuming calcium hydroxide to produce calcium silicate hydrate, i.e., C–S–H (Chuah et al. 2014). These characteristics can improve the interfacial structure and internal matrix properties. One such promising nanofibrous material is graphene oxide (GO). It is typically produced from the chemical oxidation and exfoliation of graphite. GO forms as hexagonal 2D sheet layers several nanometers thick and several hundred nanometers long. They are long-plane nanofibers containing ranges of reactive oxygen functional groups, which can actively influence microstructure formation of cementitious materials during hydration.

GO offers several smart properties that can potentially enhance the performance of cement-based materials. Compared to other nanomaterials suitable for cement incorporation, GO has a large specific surface area that contains highly reactive hydroxyl, epoxide, carboxyl, and carbonyl functional groups (Lambert et al. 2009). Although the functionalization of graphene into GO degrades the mechanical properties, GO sheets exhibit a mean elastic modulus of 32 GPa and a tensile strength of 130 MPa, which

are superior to the elastic modulus and tensile strength of cement. The major focus related to investigations of GO in cement is to examine the hydration rate, reinforcing ability of the cement matrix, and formation of the microstructure.

In addition, some challenging issues need to be carefully addressed. The direct mixing of GO with cement can substantially reduce the workability of the cement paste (Pan et al. 2015). This effect is attributable to the adsorption of free water onto the surface of the GO nanostructure. The development of a compatible water-reducing admixture is one approach to mitigating this issue. The agglomeration of nanomaterials adversely affects their ability to improve the properties of concrete. Because GO can easily disperse in water, it is more likely than other nanomaterials to efficiently distribute throughout concrete. However, some authors have argued that achieving a good dispersion of GO in the cement matrix is not guaranteed (Chuah et al. 2014). Another major challenge is to investigate the influence of GO on the cement hydration process and its compatibility with the hydration products. Current understanding of the life-cycle cost and carbon footprint of GO–cement composites is also limited. This review summarizes the state of the art in GO–cement composite research, including investigations related to the production and potential applications, mechanical properties, hardness, microstructure, durability, and life cycle of GO–cement composites.

2 THE PRODUCTION OF GRAPHENE OXIDE AND ITS APPLICATION IN CEMENT-BASED COMPOSITES

Two suitable industrial scale-up routes for GO production with minimal environmental impacts have been developed: the *chemical reduction route* (CRR) and the *ultrasonication route* (USR) (Arvidsson et al. 2014). GO is commonly synthesized from the oxidation of natural graphite via the modified Hummers method (Park & Ruoff 2009). Briefly, KMnO_4 , graphite flake, concentrated H_2SO_4 , and orthophosphoric acid are mixed and then stirred for 24 h at 50 °C. The resultant mixture is added to H_2O_2 (30%) and centrifuged. The separated product is finally washed with water, HCl, and ethanol at pH 7, then maintained at 70 °C for 12 h.

To prepare the GO–cement composites, GO is suspended in distilled water and sonicated for 3 h to obtain a homogeneous solution; cement (such as ordinary Portland cement, OPC) is then added to the mixture while the desired water-to-cement (w/c) ratio is maintained. Sand and aggregate are added to the GO–cement mix thereafter. In addition to the production of GO via the chemical reduction method, it can also be produced through ultrasonication. An example of a GO–cement-based composite production method is schematically presented in Figure 1.

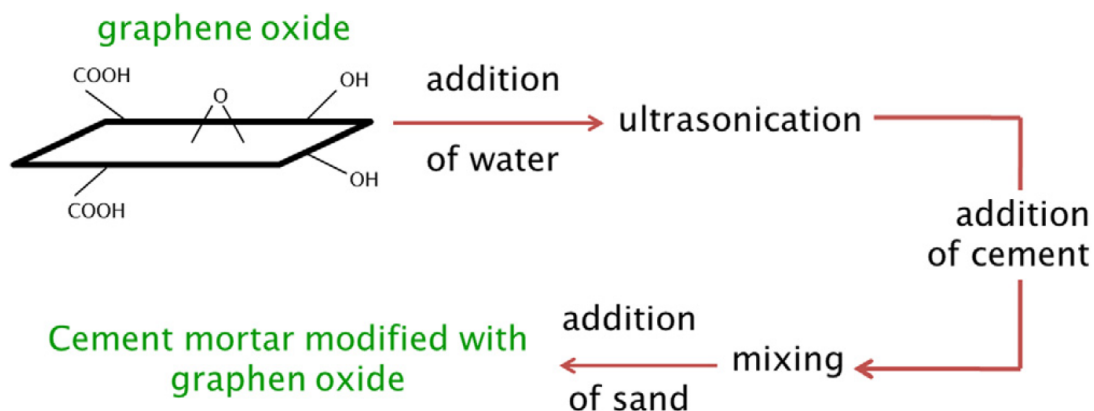


Figure 1: Schematic of cement–GO composite formation (Horszczaruk et al. 2015)

GO is used in the cement composite to serve both as a fibrous material and as a performance-enhancing additive. Different types of fibers are typically incorporated into concrete composites to improve their mechanical properties. In this sense, natural fibers such as asbestos were used initially (Aziz et al. 1981), followed by the use of glass fibers (Majumdar & Ryder 1968), steel fibers (Brandt 2008), and synthetic

fibers (Aulia & Rinaldi 2015) over time. The incorporation of fiber into concrete adds a new dimension to its properties by substantially improving its tensile strength, toughness, and the ductility. The extent to which fibers improve the mechanical properties of concrete differ with the size, shape, and type of fiber. However, various supplementary cementitious materials such as fly ash, silica fume, metakaolin, slag, and MgO have been used to improve the specific performance of concrete over the past few decades (Bastos et al. 2016; Qureshi et al. 2016).

The production scale of concrete with fibers has progressively increased because of the increasing demand of highly ductile construction materials. Table 1 presents the physical properties of common fibers used in concrete, along with recently developed graphene-based fibers.

Table 1: Physical properties of common fibers

Fiber type	Tensile strength (GPa)	Elastic modulus (GPa)	Elongation at break (%)	Diameter/ thickness (μm)	Aspect ratio	Information source
Steel	1.5	200	3.2	500	20	Reproduced from Bastos et al. 2016
Glass (E-glass)	3.45	72	4.8	5–10	600–1500	
Glass (AR-glass)	1.8–3.5	70–76	2	12	600–1500	
Polypropylene	0.1–0.8	8	8.1	100	150	
Polyvinyl alcohol	0.8	29–36	5.7	14–650	430–860	
Carbon	2.5	240	1.4	7	710	Chuah et al. 2014
Graphene	~130	1000	0.8	~0.08	6000–600,000	
CNTs	11–63	950	12	15–40	1000–10,000	
GO	~0.13	23–42	0.6	~0.67	1500–45,000	

*GO: graphene oxide, and CNTs: carbon nanotubes

Notably, the functionalization of graphene to GO results in a material with diminished mechanical properties. However, the functional groups of GO render it hydrophilic and highly dispersible in water. This distinctive property of GO enables its efficient incorporation into cement to form composites with the potential for enhanced mechanical properties.

3 PROPERTIES AND PERFORMANCE OF GO–CEMENT COMPOSITES

The microstructural material characteristics of GO can modify the physiochemical properties of cementitious materials. Table 2 lists the physical performance characteristics of recently reported GO–composites. This section presents the influence of GO on the hydration, workability, mechanical properties, microstructure, transport properties, and durability of cement-based composites.

3.1 Cement Hydration Process and Workability

Few studies have been conducted to elucidate the fundamental influence of GO in the cement hydration process. Horszczaruk et al. (2015) recently suggested that the kinetics of the cement hydration process is not strongly influenced by GO. They found that the microstructure morphology of the GO composite was similar to that of the fresh cement mortar, indicating that the GO was likely homogeneously distributed in the composite matrix. However, other recent research contradicts the homogeneous distribution of GO, claiming that GO regulates the formation of flower-like hydration crystals (Figure 2) that substantially improve the mechanical strength of the composite (Lv et al. 2013). The flower-like crystals continue to grow and become denser over time. GO has also been reported to accelerate the degree of hydration of Portland cement (PC) paste systems (Gong et al. 2015); this acceleration of hydration was confirmed on the basis of the increase in the non-evaporable water content and the calcium hydroxide content upon the addition of GO to the composite.

Table 2: The performance of GO in cement-based composites (compared to control (no GO))

Cement and GO proportion (wt%)	Compressive strength improvement (%)	Tensile strength Improvement (%)	Elastic modulus improvement (%)	Flexural strength improvement (%)	Pore properties	Durability increase	Source
0.01-0.06	--	--	--	--	Increase small gel pores	Reduce chloride ingress	Mohammed et al. 2015
0.05	15.0 -33.0	--	~6.5	41.0-59.0	Gel pore vol. increased	Yes	Pan et al. 2015
0.03	40.0	40.0	--	--	Reduce 13.5% total porosity	Yes	Gong et al. 2015
3.00	--	--	200.0-500.0	--	Reduce pores	Yes	Horszczaruk et al. 2015
1.5+0.5 wt% SP	--	48.0	--	--			Babak et al. 2014
0.05	24.4	--	--	70.5	Reduce pores	Yes	Wang et al. 2015
GO+CNT	--	--	--	72.7	Reduce pores	Yes	Li et al. 2015
GO	--	--	--	51.2			
CNT	--	--	--	26.3			
0.01	13.4	28.0	--	51.7	Increase small gel pores	Yes	Lv et al. 2013
0.03	38.9	51.0	--	60.7			
0.05	47.9	24.2	--	30.2			
FA+GO	--	--	200.0	48.0	Increase small gel pores	Yes	Saafi et al. 2015
0.01	--	--	376.0	130.0			
0.35	--	--	380.0	134.0			
0.50	--	--					

*GO: graphene oxide; CNT: carbon nanotube; FA: fly ash; and SP: superplasticizer

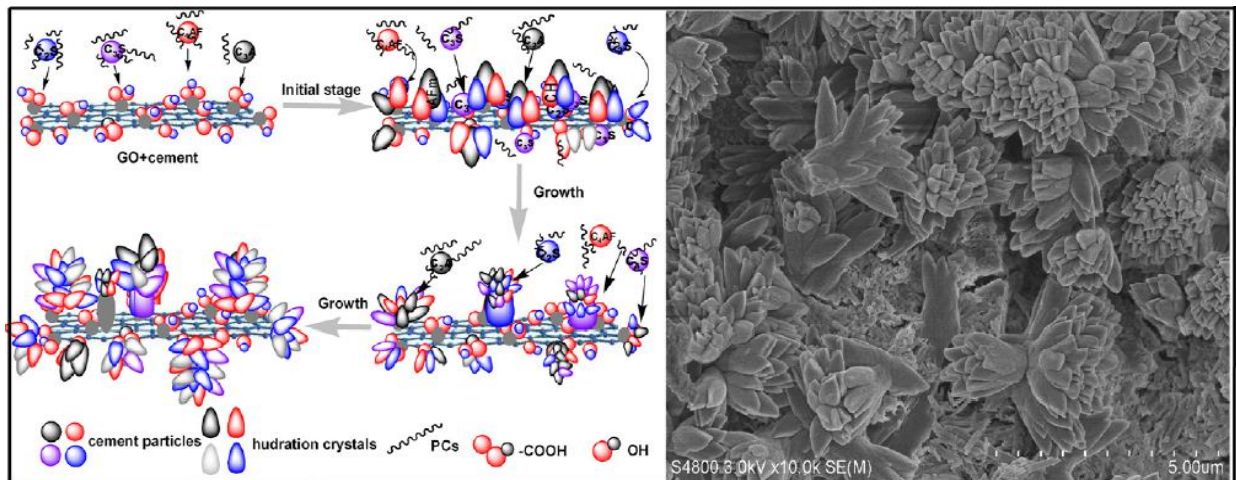


Figure 2: Schematic and scanning electron microscopy image of crystal-flower formation in the cement hydration process in 0.03% GO-cement (Lv et al. 2013)

Although some researchers have suggested that GO results in an increase in the number of small pores that contain C-S-H gel during the hydration process (Horszczaruk et al. 2015), the influence of GO in the

cement hydration process is poorly understood, with different researchers expressing different opinions. The surface of the GO 2D plane plays a major role in the cement hydration process. In a recent investigation, Lv et al. (2013) explained the contribution of GO as occurring through the growth of nanocrystals of flower shapes. The functional reactive groups of GO contain -OH, -COOH, and -SO₃H, which react with C₂S, C₃S, and C₃A during the growth stage, where the hydration reaction is temporarily retarded in cement. Different rod-shaped crystals start to form and grow into flower-like shapes over time on the GO surface after this retarding phase. This specific hydration phase and products are controlled by GO via a template effect. Column-shaped crystals grow in the pores and cracks and lose structure; they continue to grow apart, eventually forming the fully bloomed flower-like crystals. These hydration products reinforce the matrix at the nano/microstructure level and densify the matrix, which improves the mechanical properties and durability of the resulting GO-concrete composites. However, the recent findings of Cui et al. (2017) contradict the work of Lv et al. (2013), suggesting that there might be a pitfall in the scanning electron microscopy (SEM) sample preparation method because these flower-like crystals are the carbonation products of the cementitious hydrates. Although Lv et al. (2013) attempted to explain the contribution of GO during the cement hydration and growth stage, an explicit investigation is still required to elucidate the fundamental influence of GO on the complete cement hydration kinetics. This includes the heat release and phase characteristics of GO in the diffusion, dispersion, nucleation, and growth stages.

The workability of concrete and cementitious materials is an important factor for gauging the transport and placement properties of fresh materials. Typically, the greater specific surface area of nanoparticles requires extra water, which reduces the amount of free water essential for overall lubrication at a particular w/c ratio. This adverse effect on workability has been observed through nanosilica formation. The literature on the effect of GO on cement hydration suggests that the 2D sheets of GO act as a double-edged sword that promotes C-S-H nucleation at the expense of workability (Chuah et al. 2014). An initial mini-slump test by Gong et al. (2015) showed a 50% decrease in workability. Additional tests by the same authors suggested that the workability is reduced by 34.6% upon the addition of 0.03 wt.% GO. Similarly, Pan et al. (2015) have reported an approximately 42% reduction in workability through a mini-slump test in a GO-cement composite containing 0.05 wt.% GO. Although GO easily disperses in water, its nanosheet layers consume extra water, thereby increasing frictional resistance among the cement particles. This resistance decreases the ample lubrication properties of GO-cement composite.

3.2 Mechanical Properties

The addition of GO can improve the mechanical properties of cement-based materials. Specifically, GO prevents cracks, whereas microfibers restrict crack propagation after the failure has already initiated. The carboxyl, hydroxyl, and epoxy functional groups of GO adversely affect the electrical properties of graphene (Palermo et al. 2016). However, these functional groups increase GO solubility in aqueous cement matrices, where they serve as a nucleation agent for C-S-H crystals (Babak et al. 2014). The addition of approximately 0.05 wt% of GO to PC paste increased the compressive strength by 15–33%, increased the flexural strength by 41–59%, and slightly increased the elastic modulus to 3.7 GPa from 3.48 GPa (Pan et al. 2015). Gong et al. (2015) also found that the addition of 0.03 wt% of GO to a composite increased both the compressive strength and the tensile strength by approximately 40% compared to those of plain cement. In a similar study, the addition of 3 wt% of GO was found to improve the Young's modulus for a plain cement mix considerably, from 1–10 GPa for plain cement mix to 5–20 GPa (Horszczaruk et al. 2015). Wang et al. (2015) found that a GO content of only 0.05% wt%, where the GO was dispersed with a superplasticizer, resulted in 24.4% and 70.5% improvements in the compressive and flexural strengths of cement, respectively. However, the optimum mix proportions of GO were found to be approximately 0.03 wt% on the basis of the tensile strength and flexural strength development over 28 days (Lv et al. 2013). Another recent study has suggested that the combination of GO and carbon nanotubes (CNTs) increases the flexural strength of the cement matrix by 72.7%, compared to increases of 51.2% when GO was used alone and 26.3% when CNTs were used alone (Li et al. 2015). Nevertheless, these improvements are sensitive to the specific grades of GO, and the optimization of its mix proportions should be studied more explicitly. In principle, GO in cement mixes acts as a nanofiber and forms a barrier to crack propagation,

which results in an increase in the mechanical properties of the resulting composite compared to that of the plain cement. These effects were observed in SEM images collected after the hydration of cement paste mixes (Bastos et al. 2016). Whereas cracks in fresh cement paste samples pass across the dense hydration products, fine cracks with occasional discontinuous branches are observed in the hydrated surface of GO–cement composites.

Geopolymers in the form of in situ reduced graphene oxide (rGO) have recently been used to improve the mechanical properties of fly-ash-based alternative cementitious materials (Saafi et al. 2015). Geopolymers with GO concentrations of 0.35 wt% rGO exhibited 134%, 376%, and 56% increases in flexural strength, Young's modulus, and flexural toughness, respectively. This work opens new prospects for the utilization of GO to improve alkaline-activated alternative binder materials.

3.3 Microstructural Integrity

The long 2D nanosheet structure of GO functions as a reinforcing nanofiber to strengthen the brittle cement matrix. The homogeneous dispersion of oxygen-bearing functional groups on GO may influence the nucleation of C–S–H gel and or produce flower-like crystals that densify the microstructure, which may make it durable. The GO flakes also show a nucleation effect during cement hydration, forming stronger bonding between the GO flakes and the cement hydration products (Figure 3). The hydrophilic groups and higher surface energy of GO encourage the deposition of cement hydration products, and the GO surface acts as a nucleation site during the hydration process (Babak et al. 2014). This effect further accelerates the overall cement hydration.

An in-depth study is still needed to elucidate its stability, the fundamental mechanism of the behavior of GO during cement hydration, and the interaction of GO with the various phases of cement for improving various properties.

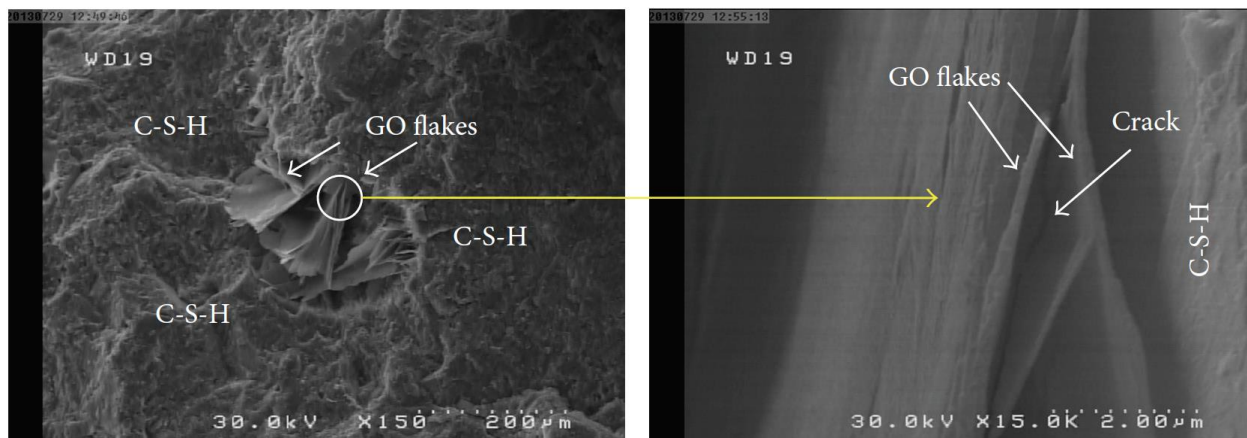


Figure 3: The SEM image showing the microstructural integration of GO flakes in cement mortar matrix (Babak et al. 2014)

3.4 Transport and Durability Properties

A recent study suggested that the direct mixing of GO in cement mortar systems (0.01–0.06% by weight) modifies the pore size and distribution and effectively hinders the ingress of chloride ions (Mohammed et al. 2015). The addition of GO increases the size of small pores. Typically, those smaller than 15 nm, can increase to pore sizes in the range from 100 to 1000 nm. In contrast, GO decreases larger-sized pores, and the capillary pores are also reduced with increasing GO fraction. These effects on the cement composite matrix differs from those of other nanoparticles. For example, nano silica particles reduce the

to the CRR (Arvidsson et al. 2014). Although no “winner” was clear in terms of all impact categories, the sensitivity analysis suggested that recovering diethyl ether during the USR led to substantially lower impacts in all categories compared to the CRR. Similarly, the recovery of acid solvents in the Hummers process via CRR could considerably reduce the blue water footprint.

Nevertheless, few studies have been reported on the life-cycle assessment and carbon footprint analysis of GO nanomaterials, mainly because of a lack of understanding of characterization factors (CFs) for the life-cycle impact assessment. In their recent study, Deng et al. (2017) derived a CF based on three factors pertaining to GO in the aquatic environment: the toxic effect factor, fate factor, and the exposure factor. The analysis resulted in a CF of 777.5 potentially affected species (PAF) day m³ kg⁻¹ for GO, a fate factor of 27.2 days, and an exposure factor of 0.93. Their sensitivity analysis predicted the uncertainty of a variable CF value between 1 and 10³ PAF day m³ kg⁻¹. Furthermore, a comparison between GO and CNT suggests that the CF baseline of GO is less than that of CNTs and that functionalized GO results in a much lower value compared to the original GO.

5 CONCLUSION

Experiments with GO have demonstrated good reinforcing and microstructural features stemming from its active functional groups and much higher specific surface area compared to that of 0-D nanoparticles. GO has been shown to influence the cement hydration properties at the molecular level. The direct mixing of GO in cement increases the mechanical properties of the resultant composites as a result of modification to the pore size distribution, production of gel materials in the pores, and the corresponding ability to effectively hinder ions ingress. To date, no known study has reported the behavior of GO–cement composites under decarbonation, acid resistance, sulfate resistance, and radioactive resistivity. The nano reinforcing and water-dispersion properties of GO could be further investigated for developing self-healing concrete and other self-repairing materials. The production of GO is a highly environmentally friendly compared to cement and other cement supplementary nanomaterials. Hence, an in-depth life-cycle cost assessment and carbon footprint analysis should be carried out on GO–cement-based composite systems. Advances in GO production and GO–cement composite materials may bring efficiency gains in the construction industry and mitigate the overall carbon footprint on Earth.

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