



# Nanocomposite of cement/graphene oxide – Impact on hydration kinetics and Young's modulus



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## HIGHLIGHTS

- Graphene oxide(GO)/cement matrix composites have been synthesized.
- Graphene oxide additive in cement 3 (wt%) significantly enhanced Young's modulus.
- The kinetics of the hydration is not strongly altered in GO/cement composite.
- The morphology of the GO/cement composites is almost the same as the reference.
- The homogeneous distribution of the GO flakes in a cement matrix was observed.

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## ABSTRACT

The application of nanomaterials in construction is a new alternative to enhance the mechanical properties of the traditional materials, e.g., cement mortars and concretes. One of the most interesting nanomaterials which still requires detailed investigation is graphene and graphene oxide. The study presented in this paper aims at assessing how 3 wt% of graphene oxide incorporated into the cement can affect the microstructure and physical–mechanical properties of the cement composite. Therefore, here we present study on early age mechanical response of the cement mortar modified with graphene oxide using atomic force microscopy (AFM). The kinetics of the hydration process was investigated by Infrared, Raman, X-ray diffraction (XRD) techniques. The morphology of the nanocomposite was revealed by the scanning electron microscopy (SEM).

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## 1. Introduction

Cement based composites are the most important and widely used building material at present [1]. Progress in the field of nanomaterials presents an invaluable opportunities to further raise the performance bar with the inclusion nanomaterials in cementitious composites [2,3]. There have been many recent studies on newly produced nanomaterials in cement composites such as nanosilica [4–6], nanotitanium oxide [7], carbon nanotubes (CNTs) [8–12]. Nano-reinforcements in cementitious matrix materials are more effective than conventional reinforcements at millimeter

scale because they can control nano-size cracks before they develop into micro-size cracks [13]. In the past years, graphene and its derivatives have been investigated extensively. This is due its exceptional mechanical, electrical, thermal and optical properties, high surface area-to-volume ratio, and unique atomic structure [14]. Therefore, many applications are expected, for instance, electronic devices [15–18], composites [19] and nano-medicine [20,21]. The superlative properties of graphene oxide has a promising future when combined with ordinary Portland cement (OPC), forming a nanocomposite. The 2D GO nanosheets provides an extra dimension to interact with cement and concrete matrix [22,23]. The Young's modulus of graphene oxide is estimated to be about 1 TPa [24]. In order to design plain, reinforced, and prestressed concrete structures, the elastic modulus  $E$  is a fundamental parameter that needs to be defined. Study on the modulus of elasticity of cement is necessary for further analysis

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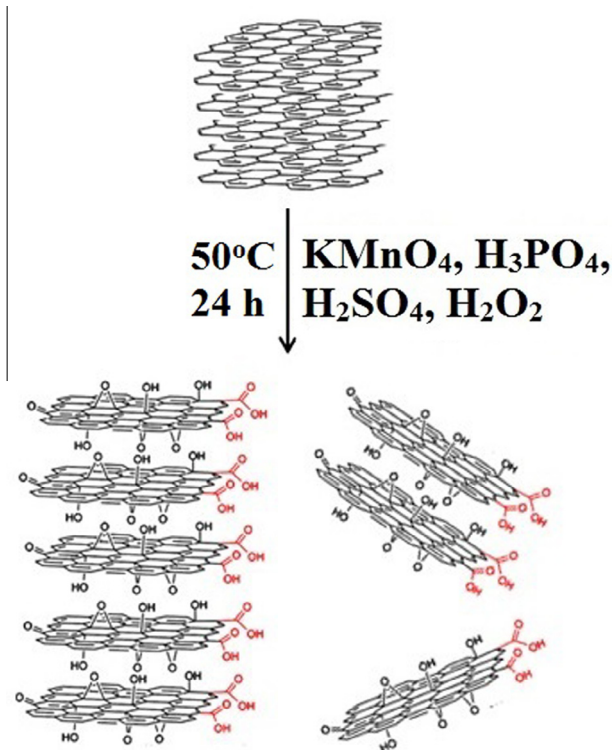


Fig. 1. Schematic representation of the graphite oxidation.

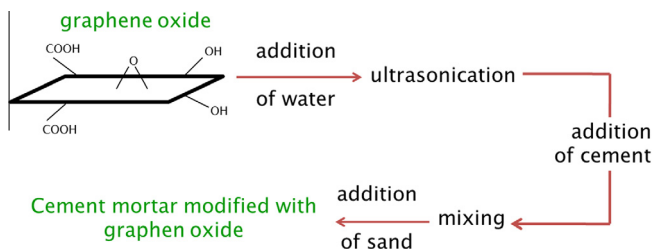


Fig. 2. Schematic representation of the cement-GO nanocomposite formation.

of research on cementitious composites. Nowadays, different methods are widely used to measure  $E$  values for cement materials. One of the techniques is nanoindentation measured by an atomic force microscopy probe (AFM) [25]. With the concurrent benefit of excellent mechanical performance, graphene will improve the

toughness and strength of cementitious matrix materials. The hydration of cement and the accompanying phenomena such as heat generation, strength development and shrinkage are the results of interrelated chemical, physical and mechanical processes. All these processes of hydration kinetics plays an important role on microstructural development and final properties of hydrated cement pastes [26,27]. The nanometer scale structure of the cementitious matrix is composed of porous calcium-silicate-hydrate (CSH) gel phase. Its structure is strongly related to the mechanical properties of the formed cement paste. Additionally, the morphology of CSH influences the hydration process. However, the control of the hydration is difficult, especially during the early stages of the reaction. This is due to its amorphous and fragile structure. Therefore, the formation of high quality concrete is strongly related to the understanding of the processes taking place even in the first 10 min of the cement hydration [11,28–30]. Throughout the duration of the process water molecules penetrate the cement grains constantly. This reaction is accompanied by a heat release. Two products are formed during the process: calcium hydroxide  $\text{Ca}(\text{OH})_2$  (portlandite) and calcium silicate CSH gel. Portland cement modified with nanomaterials has become a new alternative to traditional OPCs. It also has a chance to be an alternative for the common cement additives such as silica fume or fly ash [4,5]. In the field of cement nanocomposites, oxidized form of graphene – graphene oxide (GO) is widely investigated [22,23]. However, the impact on the structure of the cement hydration process has not been fully revealed yet. Planar structure of graphene sheets which creates significant contact area with the cement material. Mechanical properties and shape of graphene oxide are the main reason of significant improvements in compressive and flexural strength of the cement materials. For carbon nanostructures to be fully utilized within a material, they must first be properly dispersed. Studies have shown that graphene oxide shows better adhesion properties than graphene [31,32]. GO is a form of graphene with the oxygen containing functional groups such as carboxyl, hydroxyl and epoxy [33,34]. The presence of oxygen containing functional groups significantly alters the Van der Waals forces between the graphitic sheets. Therefore, their water-based dispersion is much more stable and homogeneous than the water dispersion of pristine graphene [31,32,35]. Moreover, the oxygen functional groups provides many potential sites for advantageous chemical or physical interactions. They can act as a binder between the GO flakes and cement paste to obtain greater uniformity of the cement matrix [36,37].

The study presented in this paper aims at assessing how 3 wt% of graphene oxide incorporated into the cement can affect the microstructure and physical-mechanical properties of the cement composite. The kinetics of hydration process will be also studied

**Table 1**  
Chemical composition of OPC (CEM I 42.5R).

Type of ordinary Portland cement	Content [wt%]									
	Loss on ignition	$\text{SiO}_2$	$\text{Al}_2\text{O}_3$	$\text{Fe}_2\text{O}_3$	$\text{CaO}$	$\text{MgO}$	$\text{SO}_3$	$\text{Na}_2\text{O}$	$\text{K}_2\text{O}$	$\text{Cl}^-$
Cement CEM I 42,5R	3.5	19.5	4.9	2.9	63.3	1.3	2.8	0.1	0.9	0.06

**Table 2**  
Compositions of cement mortars.

Type of sample	Cement [g]	Water [g]	Sand [g]	Graphene oxide [g]	Nanoparticles, cement mass, %
Reference	17.57	10	54.5	0	0
GO3	17.57	10	54.5	0.527	3

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