



Effects of Graphene Oxide Addition on the Durability of Cementitious Composites

Laísa do Rosário Souza Carneiro¹, Flávia Spitale Jacques Poggiali², Péter Ludvig³

^{1,2,3}Department of Civil Engineering, Federal Center for Technological Education of Minas Gerais - Av. Amazonas, 7675, Nova Gameleira, CEP, 30510-000, Belo Horizonte, Minas Gerais, Brazil
(¹carneirola@hotmail.com, ²ludvig.peter@gmail.com, ³flaviaspitale@gmail.com)

Abstract- In recent years, the demand for more durable concrete structures has steadily increased. To become more durable, concrete should be able of resisting aggressive environmental conditions such as weathering situations, acid attack, carbonation, chloride ions ingress, etc. The increase of the durability of cementitious composites can be achieved through the pore refinement of the material. It seems that the use of nanomaterials is an effective approach in this process, and that the incorporation of small amounts of graphene oxide (GO) has provided improvements in the transport properties of various cementitious matrices. In this paper, a bibliographic survey was carried out, based on publications from indexed international journals, published in the last 10 years. Sixty-one papers constitute the portfolio of this review work, which were used as an object of analysis and investigation of the main durability criteria, namely water absorption, sorptivity, chloride diffusion coefficient, depth of chloride ion penetration, acid attack, porosity and microstructure. To reach greater improvements in the transport properties, the graphene oxide amount should not exceed 0.1% by weight of cement. Due to the presence of oxygenated groups in its surface, graphene oxide acts as a nucleation agent for the cement hydration products, forming flower-like crystals, which are able to fill the pores in the paste. There seems to be an optimal graphene oxide content, above which the Van der Waals attraction forces, present between the oxide layers, overlap the hydrophilic repulsion forces of the oxygenated groups, leading to the agglomeration of the GO leaves. In this case, the material transport properties would be negatively affected. The optimal GO content might be a function of several factors, such as the water/binder ratio, the superplasticizer type and dosage, the dispersion type of the GO leaves, the aggregates' type and fineness and the cementitious composite type, e.g. cement paste, mortar or concrete. In this sense, studies on the individual and combined effect of these factors should be encouraged. Besides, variables such as acid attack and depth of chloride ion penetration should be evaluated in field.

Keywords- Graphene Oxide, Durability, Cementitious Composites

I. INTRODUCTION

Over the years, the demand for more durable concrete structures has progressively increased. To become more durable, concrete must be able to resist aggressive environmental conditions, such as different weather conditions, acid attacks, carbonation, chloride ions, etc. [1]. Thus, the durability of cementitious composites is controlled by their porosity and permeability, which can be improved through the refinement of their pores [2].

A higher number of researches have shown that the addition of small amounts of carbon-based nanomaterials into the cementitious matrix can promote the refinement of pores, especially nanopores. Besides filling empty spaces, such materials create nucleation sites, increasing the degree of cement hydration, by promoting the production of C-S-H and Ca(OH)₂ [3, 4, 5, 6, 7, 8, 9]. The nucleation phenomenon has a substantial impact on increasing mechanical strength and reducing microcrack propagation in the cementitious composite [10].

Among the carbon-based nanomaterials currently used as cementitious additions, graphene oxide (GO) has attracted a great deal of interest in the scientific field, due to its good performance [1]. Such oxide is a two-dimensional layered material, derived from graphene. It has either a hydroxyl (–OH) or epoxy (–O–) group in its basal plane and a carboxyl (–COOH) or a carbonyl (–CO) at the edge of its leaf [11, 12]. The presence of these oxygenated active groups alter the Van der Waals forces that act in the interlayers of the GO sheets, favoring the sheets dispersion in water and increasing their reactivity [13, 14]. Nonetheless, the existence of Ca(OH)₂ in the cementitious medium can lead to agglomeration of the GO sheets [15, 16]. Thus, the use of superplasticizers, such as polycarboxylate [17, 18, 19] and sodium dodecylbenzene sulfonate [20] types is recommended to guarantee the sheets proper dispersion. According to Vallurupalli *et al.* [21], the optimal dosage of superplasticizer is about four times the mass of GO used.

In 1958, Hummer found out that graphite could be oxidized by treating it with potassium permanganate ($KMnO_4$) and sodium nitrate ($NaNO_3$) in a concentrated sulfuric acid (H_2SO_4) medium. Such method of GO production is used to the present day [1, 22, 23, 24], although sustainable and eco-friendly methods have been investigated as well, *e.g.* methods that use water as a liquid medium for the graphite exfoliation process [25]. Graphene oxide can also be partially reduced. In this case, it is called graphene r-oxide or functionalized graphene.

Improving the mechanical properties of cementitious composites by incorporating small amounts of either graphene oxide or reduced graphene oxide has been a widely explored and discussed topic in current literatures [26, 27, 28, 29, 30, 31, 32]. Nonetheless, studies focusing on the durability of such modified composites are still incipient. Consequently, literature reviews that specifically explore the theme of durability are still lacking. Hence, in this work, recent and relevant publication in the area was gathered and an analysis of the main durability criteria, namely water absorption, capillary absorption rate (capillarity), acid attack, depth of penetration, diffusion of chloride ions, porosity and microstructure, was carried out. This work also analyzes the effect of GO coatings on the durability of cementitious composites. This paper aims to synthesize the main discoveries in the area, pointing out research gaps and encouraging the development of new studies.

II. METHODOLOGY

In this paper, a bibliographic survey was carried out, based on publications from indexed international journals, published in the last 9 years. Sixty-one papers constitute the portfolio of this review work, of which 42 specifically explore the durability aspects of graphene oxide modified cementitious composites. Additionally, 19 secondary papers complement this review. Extensive research was carried out in scientific databases, namely Science Direct, Scopus and Scielo from March to May 2021. Results were restricted to papers published from 2013 onwards, except for the secondary papers.

Fig. 1 shows the distribution of the papers, which constitute the portfolio of the present work, over the years. It can be noticed that most of the relevant papers were published in the last three years, which reflects an increasing deal of interest on the part of the scientific community in the subject and justifies the need for this current review.

III. DISCUSSION

A. Microstructure and Porosity

The use of graphene oxide (GO) as a cementitious addition increases the rate of cement hydration [33]. This is due to the high surface energy of GO sheets, which is a reflection of its high specific surface area. Besides, graphene oxide has many oxygenated groups, including hydroxyls ($-OH$) and carboxylic acids ($-COOH$). Such groups react with the anhydrous phases of cement, *i.e.*, C3S, C2S and C3A, turning the surface of GO sheets into nucleation sites for the growth of hydrated crystals [10, 34, 35].

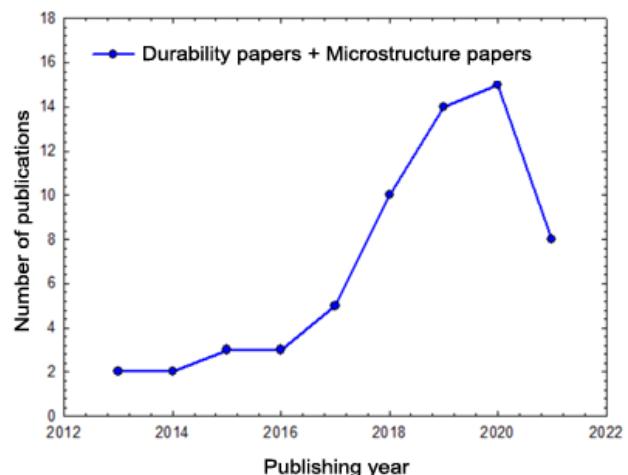


Figure 1. Distribution of the published papers, which constitute the portfolio of the present work, over the years.

This phenomenon is usually called “the seeding effect”. According to Li *et al.* [15], the GO sheets seeding effect depends on the dispersion rate of their nanosheets. Kang *et al.* [7] verified that cement pastes with 0.03%-0.05% GO presented a 20% increase in the degree of hydration at the age of 7 days, when compared to reference ones.

Through the seeding effect, graphene oxide also controls the growth pattern of ettringite (AFt), monosulfate (AFm) and calcium hydroxide (CH) crystals, which, due to their needle and rod shapes, get together in flower-shaped crystals, in the so-called template effect [35, 36, 37, 38]. Such flower-like crystals are deposited inside the paste pores and cracks, expanding themselves and filling such empty spaces.

Qureshi and Panesar [39] observed that pores of $\sim 10\mu m$ were completely filled by flower-like (or flake-like) crystals in samples with 0.02% of GO and 0.04% of GO. Li *et al.* [15] verified that, in samples with 0.02% of GO, the amount of macropores was significantly reduced, by 40%, while in samples with 0.04% of GO the reduction was of 10%. Long *et al.* [40] observed that micro and sub-micrometric cracks were inhibited in hardened cement pastes, due to the addition of GO sheets. Likewise, Muthu *et al.* [41] verified that GO sheets resist to shrinkage cracks of sizes between 10 nm and 10 μm in cement pastes. Thus, graphene oxide is capable of turning the porous microstructure of cementitious composites into denser ones [35], by reducing its mesopores [42].

By acting as a nucleation agent and promoting the cement hydration, graphene oxide also affects the morphology and the distribution of cement hydration products, redefining the paste microstructure [30]. By analyzing Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM) images, Tong *et al.* [43] found that C-S-H gel phases formed around the graphene oxide nanosheets were larger than those formed in the control samples. This indicates that a greater amount of C-S-H gel has precipitated around the nanosheets. The authors also verified that no apparent transition zone was set between the GO nanosheets and the C-S-H gel, suggesting that a good interfacial bond was established between them, which resulted

in a more compact microstructure. According to Indukuri and Nerella [1], this bond is favored by the high surface area of the GO sheets and by its wrinkled morphology. In this sense, an efficient dispersion of the GO sheets, combined with a high amount of oxygenated groups, leads to a greater production of C-S-H and Ca(OH)2, with a uniform distribution, which results in a denser microstructure [1, 39].

Once the transport properties of cementitious materials are highly dependent on their pore size distribution, total porosity, connectivity between pores and tortuosity [44], it is expected that the addition of graphene oxide into cement matrix also modify such properties.

B. Water Absorption

The water absorption test is generally used to assess the porosity of cementitious composites, since these pores influence the water absorption, due to capillary action [45]. Fig. 2 presents the relative reduction in water absorption of various cementitious samples, evaluated by five different authors. In a study with cement pastes, carried out by Indukuri and Nerella [1], and in other ones with cement mortars, conducted by Prabavathy *et al.* [45] and Roy *et al.* [46], the water absorption reduction increases as the GO amount gets higher, until it reaches an optimum value, and starts decreasing from there on. According to Indukuri and Nerella [1], when added in high amounts, GO sheets or flakes agglomerate due to Van der Waals attraction forces, present between the oxide layers. These forces overlap the hydrophilic repulsion forces, generated by GO oxygenated functional groups. In such scenario, the GO sheets absorb a high amount of water molecules, which results in a non-uniform w/c ratio and in a non-homogeneous distribution of the hydration products. Consequently, both mechanical and transport properties of the cementitious composites might be compromised [1, 17, 47, 48].

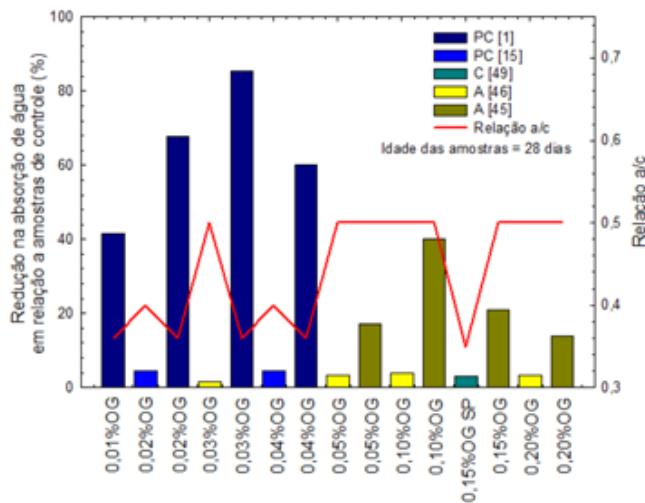


Figure 2. Relative reduction in the water absorption of cement pastes (CP), Concretes (C) and Mortars (M) samples with the addition of varied graphene oxide amounts, compared to control samples, according to different authors. "SP" indicates the use of superplasticizer.

According to SEM images, analyzed by Lv *et al.* [34], the addition of up to 0.03% of GO leads to the formation of increasingly larger flower-like crystals. Nonetheless, an addition of 0.04% of GO leads to the agglomeration of the sheets, which assume the form of polyhedral crystals. Finally, an addition of 0.05% of GO results in a large number of polyhedral crystals, which suggests a non-uniform distribution. This study corroborates with the hypothesis that GO sheets agglomerate when added in high amounts inside cementitious medium. It is also in agreement with the studies of Indukuri and Nerella [1] and Prabavathy *et al.* [45]. Therefore, when it comes to cementitious materials, there seems to be an optimal amount of graphene oxide, which varies from material to material, *i.e.*, cement paste, cement mortar or concrete, and acts as a nucleation agent for hydration products, producing large flower-like crystals, which fill larger pores, transforming them into smaller ones. This fact directly affects the water absorption of cementitious materials.

C. Capillary Absorption

Sorptivity is the property that porous materials have of absorbing and conducting water via capillary pores, due to capillary action [1]. Such parameter can be experimentally determined, being the ratio between the distance traveled by the water, in millimeters, and the time raised to the square root, in seconds ($\text{mm}/\text{s}^{1/2}$). Fig. 3 shows the relative reduction in capillarity absorption of cementitious composites due to the addition of graphene oxide. Small additions of up to 0.16% of GO area able to reduce the total capillary absorption of cement pastes, cement mortars and concretes. Except for the study of Li *et al.* [15], the addition of up to 0.16% of GO has led to a reduction of, at least, 10% in the capillary absorption of various cementitious samples, when compared to control ones. This can been verified in the studies of Indukuri *et al.* [35], Roy *et al.* [46], Prabavathy *et al.* [45] and Akarsh *et al.* [49].

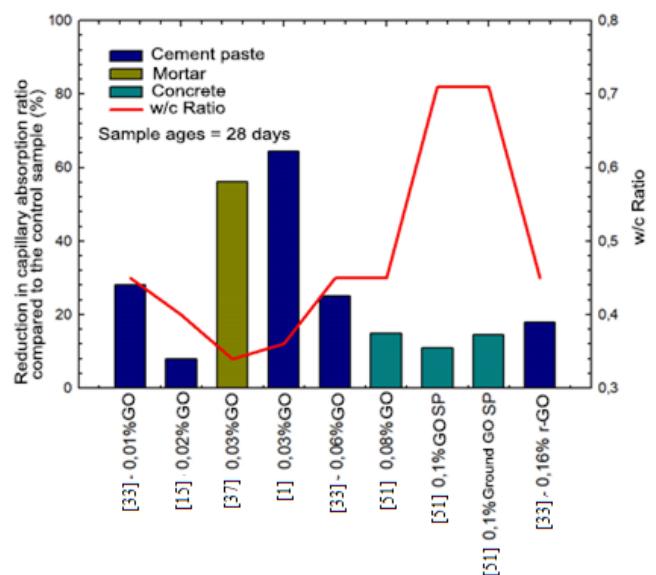


Figure 3. Relative reduction in capillary absorption of Cement Pastes (CP), Concretes (C) and Mortars (M) with the addition of graphene oxide in different amounts, compared to control samples, according to different authors. "SP" indicates the use of superplasticizer.

In general, for cement pastes and mortars, good capillary results might be obtained by using up to 0.06% of GO only. Furthermore, with only 0.03% of GO, Indukuri and Nerella [1] and Mohammed *et al.* [37] were able to reduce the capillarity of cement pastes and mortars by 64% and 56%, respectively.

D. Penetration Resistance and Chloride Diffusion

The ingress of chloride ions in concrete, through its pores network and cracks, might also compromise the durability of reinforced concrete elements, since it enables the local corrosion of the reinforcement. In this sense, special attention must be given to structures located in marine environments, in which chloride ions are abundant.

Recent researches have found that the addition of small amounts of graphene oxide into cement matrices could significantly reduce the chloride ions diffusion, as well as their penetration depth [1, 9, 37, 52, 53]. Fig. 4 presents the relative reduction in the depth of penetration of chloride ions in cementitious samples, evaluated by four different authors. The graphic shows that small amounts of graphene oxide, up to 0.2% of GO by cement mass, significantly reduce the chloride ions diffusion in cement pastes and mortars. By the addition of only 0.01% of GO, Mohammed *et al.* [37] were able to reduce the chloride ions penetration in mortars by 80.5% when compared to control samples. Similarly, by adding only 0.03% of GO, Indukuri and Nerella [1] reduced the chloride penetration by 55% in cement pastes, when compared to control samples. The four authors found that the addition of up to 0.1% of GO was able to reduce the ions penetration depth in, at least, 20%, when compared to control samples. Nonetheless, for high GO amounts, the reduction of the penetration depth of chloride ions was less expressive, due to the nanomaterial agglomeration. This is observed in both cement pastes and mortars samples.

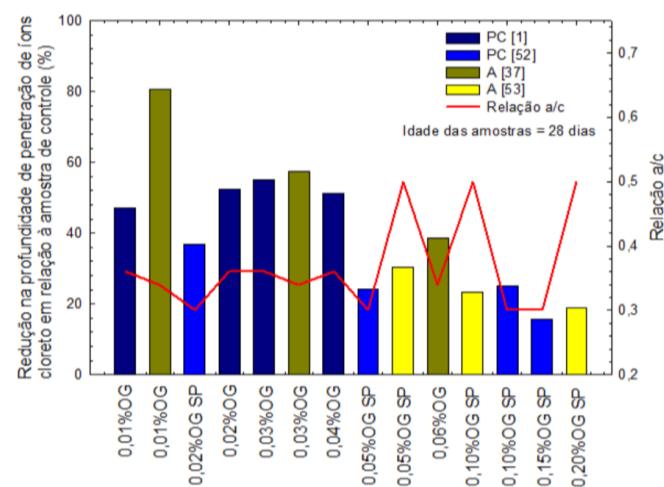


Figure 4. Relative reduction in the penetration depth of chloride ions in Cement Pastes (PC) and Mortars (M) samples with the addition of various graphene oxide amounts, when compared to control samples, according to different authors. "SP" indicates the use of a superplasticizer.

The chloride diffusion resistance was also analyzed by other authors, by determining the chloride migration coefficient. Yu and Wu [9] verified that the migration coefficient in ultra-high performance concretes (UHPC) with recycled aggregates was reduced with the addition of graphene oxide, reaching a minimum value by introducing 0.06% of GO. For Indukuri and Nerella [1], this maximum reduction was achieved by introducing 0.03% of GO in cement pastes. In this case, the modified pastes presented a migration coefficient of $12.6 \times 10^{-12} \text{ m}^2/\text{s}$, against $18.9 \times 10^{-12} \text{ m}^2/\text{s}$ of control pastes.

E. Resistance to Acid Attack

The resistance to acid attack is also an important parameter to analyze when evaluating the durability of concrete structures, especially when it comes to chemical deposits, cooling towers, wastewater clarifiers, septic tanks and radioactive waste storages [54, 55].

Table 1 presents the effect of graphene oxide addition on cementitious samples subjected to different acid attacks, when compared to control samples. In all the analyzed cases, the addition of GO favored the sample conservation, by reducing the mass loss caused by the chemical attack and increasing the residual compression strength, when compared to the control samples. By using 0.15% of GO (by cement mass), Akarsh *et al.* [49] reduced the mass loss of concrete samples, submitted to a 5% sulfuric acid solution, in more than 70%, at 56 and 90 days. By introducing only 0.03% of GO, Indukuri and Nerella [1] were able to reduce the mass loss of cement pastes samples submitted to either hydrochloric acid and sulfuric acid solutions (5% concentration) in more than 20%, at 28 days.

Devi and Khan [56] found that the residual compressive strength and the mass loss of concrete samples containing recycled aggregate with 0.1% of GO were significantly improved, when compared to the control samples. The authors attribute this improvement to the filler effect of graphene oxide at the Nano level. Tong *et al.* [43] evaluated the residual strength of mortars with 0.1% of GO, submitted to ammonium nitrate solution, with a concentration of 15%.

After 5 months, it was found that the graphene oxide samples had lost 22% of their strength, against 30% by the control samples. This suggests that graphene oxide has the potential to slow down the deterioration process induced by the acid solution, increasing the corrosion resistance of cementitious materials.

F. Graphene Oxide Based Hydrophobic Coatings

Graphene oxide can be used in the synthesis of hydrophobic coatings, which are deposited on the surface of existing or precast concrete elements, in the form of aqueous solutions, via spraying or submersion methods [57, 58]. Such coatings can also be applied along with epoxy resin [59, 60, 61].

Korayem *et al.* [56] verified that GO based coatings, with a concentration of 0.015 mg/mL, can reduce the water absorption and capillarity of concrete by up to 40% and 57%, respectively. He *et al.* [55] found that a concentration of 1 g/L leads to a greater reduction in gas permeability in both wet and dry cured mortar samples.

TABLE I. EFFECT OF GRAPHENE OXIDE ADDITION IN CEMENTITIOUS MATERIALS EXPOSED TO DIFFERENT TYPES OF ACID ATTACK

Graphene Oxide (% weight of cement)	Matrix	A/C	Type of Exposure	Improvements when compared to the control sample (%)	References
0,03	Cement Paste	0,3	Nitric acid	17% of reduction in mass loss	[41]
				25% of reduction in cross-sectional area loss	
0,03	Cement Paste	0,36	5% Hydrochloric acid	28,4% of reduction in mass loss at 28 days	[1]
			5% Sulfuric acid	22,8% of reduction in mass loss at 28 days	
0,15	Concrete	0,35	5% Sulfuric acid	76,1% of reduction in mass loss at 56 days	[49]
				71,4% of reduction in mass loss at 90 days	
0,1	Mortar	0,5	5% Hydrochloric acid	The residual compressive strength of the modified sample is 311% higher than the control one	[45]
			5% Sulfuric acid	The residual compressive strength of the modified sample is 219% higher than the control one	

According to Zheng *et al.* [60], coatings made by the combination of 0.3% of graphene oxide and epoxy resin present greater Water Contact Angles (WCA) than those made of pure resin. In addition, the coated concrete samples present lower water absorption and chloride diffusion coefficient values, when compared to the samples coated with epoxy only and to those not coated at all. This is due to the chemical reaction of GO hydroxyl groups with the epoxy resin, which establishes covalent bonds between both products. This fact allows an uniform dispersion of the GO sheets in the resin medium, which enhances its barrier effects and increases the hydrophobic properties of the coating. Similarly, Guo *et al.* [61] found that the waterproofing and the chloride ions resistance were significantly improved by the use of epoxy resin-based coatings with 0.5% TiO₂-graphene oxide. According to Yu *et al.* [59], GO-epoxy resin based hydrophobic coatings are able to reduce the water transportation in the C-S-H pores in a significant way. Furthermore, the coatings also reduce the mobility of sodium (Na⁺) and chloride (Cl⁻) ions.

IV. CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK

From the literature review carried out in this work, the following conclusions can be made:

- The introduction of small amounts of graphene oxide (generally up to 0.1% cement weight) into cementitious composites, *i.e.* concrete, mortar and cement pastes, improves the transport properties of such materials, as well as their durability.
- Due to the oxygenated groups on their surface, the graphene oxide sheets acts as nucleation sites for cement hydration products, forming flower-like crystals, which are able to fill the paste pores, specially the macropores. In addition, GO sheets favors a better distribution of the hydration products in the cement matrix, which leads a more compact structure. Consequently, both water absorption and harmful ions entrance (*e.g.* chloride ions)

are reduced, as verified by several authors in the recent literature.

- There seems to be an optimal content of GO sheets, which would act as nucleation sites for hydration products, leading to the formation of flower-like crystals of maximum and optimized size. Such crystals would fill the largest number of large pores, turning them into smaller ones. Nonetheless, for higher amounts of GO, the Van der Waals attraction forces, that act between the oxide layers, would overcome the hydrophilic repulsion forces of the oxygenated groups, resulting in the agglomeration of the GO sheets. In this scenario, the transport properties would be negatively affected.

The following proposals are made, as suggestions for future work:

- The optimal content of GO depends on several factors, such as the water/cement ratio, the superplasticizer type and dosage, the dispersion type of the GO sheets, the aggregates type and fineness, as well as the cementitious substrate (cement paste, mortar or concrete). In this sense, studies on the individual and combined effect of such factors must be further explored.
- Once the codependency between the optimal GO sheets content and the several factors listed above is further understood, dosage studies of concretes and mortars with GO sheets might be developed.
- Up to this date, studies on acid attack and penetration of chloride ions are restricted to the laboratory environment. To evaluate the potential and limitations of GO modified cementitious composites, such variables should also be investigated in the field, *e. g.* marine environment.

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