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Review Article

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# Advance on the dispersion treatment of graphene oxide and the graphene oxide modified cement-based materials

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**Abstract:** For the high demand for cement-based materials in buildings, improving the performance of cement-based materials has become the focus of relevant researchers. In recent years, nanomaterials have broad prospects in many fields such as architecture by virtue of their “lightweight, high strength, and strong solidity” characteristics. As a modifier of cement-based materials, it has also become a research hotspot. Graphene oxide (GO) is one of the most representative graphene-based nanomaterials. Because of its extremely high specific surface area and excellent physical properties, it has greatly improved the properties of cement-based materials. GO acts as an enhancer of cement composites that brings people unlimited imagination. The research progress of GO-modified cement-based materials is reviewed. The purpose is to point out the limitations of current research and provide a reference for later research. The dispersion treatment of GO and the properties of its modified cement-based materials are analyzed and summarized. In addition, the further research work that is needed and future development prospect are discussed.

**Keywords:** dispersion treatment, graphene oxide, cement-based materials, development prospects, nanomaterials

## 1 Introduction

With the vigorous development of the construction industry, cement-based materials have become the most widely used civil engineering materials [1]. However, the inherent defects of cement-based materials make them prone to cracks in the service process, which leads to performance degradation and shortened service life [2,3]. Such a situation not only requires high maintenance costs [4,5] but also seriously threatens personal safety. In the context of vigorously advocating high-strength buildings worldwide, various large-scale projects have put forward higher requirements for the performance of cement-based materials. Therefore, the research on the modification of cement-based materials has attracted widespread attention. In recent years, it has been found that traditional enhancement methods [6–8] have mostly unsatisfactory improvements in the performance of cement-based materials. Although new cementitious materials (geopolymers) [9] may also be used as substitutes for cement, they still have the same limitations as cement. With the advancement of nanotechnology, people realized that nanomaterials may be the best “answer” to improve the performance of cement-based materials. Nanomaterials are materials with a size between 1 and 100 nm. Due to its special electrical conductivity, thermal conductivity, optics, magnetism, and other characteristics, they can play an important effect in the fields of electronic information, medical treatment, biotechnology, and industry. [10–15]. In the field of building materials, the excellent effects of

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nano-silica [16] and carbon nanomaterials [17–20] modified cement-based materials have been widely demonstrated by researchers. Nano-reinforced technology is different from previous enhancement methods [21]; nanomaterials have an inhibitory effect on the growth of microcracks in the cement matrix [22,23] and can regulate the aggregation state of hydration products to make them more regular and dense. There is no doubt that nanomaterials can more thoroughly solve the defects of cement-based materials [24–26].

Carbon nanomaterials (CNMs) [27] are one of the most popular nanomaterials at present, and their unique $SP^2$ and $SP^3$ hybrid give them excellent properties [28]. In Figure 1, the crystal structures of different CNMs are shown. Among these materials, graphene and graphene oxide (GO) are the most representative. Graphene is a carbon allotrope in the form of a single-layer two-dimensional honeycomb lattice [29]. Since graphene was discovered by, British physicists, Geim and Novoselov [30,31], it has quickly attracted widespread attention due to its excellent properties. After that, people found more ways to prepare graphene materials [41,42]. Graphene has excellent mechanical, thermal, electrical, and other properties [32–39]. But at the same time, the extremely strong intermolecular force and very few functional groups make its dispersibility and compatibility poor. And due to its higher price, it is very difficult for graphene to achieve industrial applications. Graphene oxide (GO), one of its derivatives, has properties and microstructures very similar to the original graphene and also has extraordinary characteristics. As shown in Figure 1, GO is an intermediate product obtained during the preparation of graphene by the graphite oxide reduction method [38]. Compared with graphene, GO has oxygen-containing groups such as –COOH (on the edge, often higher content), –OH, and –C–O–C (in-plane of GO). These groups can not only reduce the van der Waals force of GO and improve hydrophilicity but also provide a large number of active sites for connecting other functional groups and organic molecules. GO plays a significant role in modifying cement-based materials because of its peculiar layer structure and abundant surface functions. At the same time, the cost of GO is lower than that of graphene [39], which makes it the most widely used graphene-based nanomaterial [40].

Although GO improves cement-based materials, adding nanomaterials to the cement matrix faces many problems that need to be solved urgently. First, we must establish a dispersion standard. What needs to be done to overcome the force between GO sheets and ensure that they can perfectly act on cement-based materials? Second, we need to do a lot of work to make GO compatible with other admixtures. In addition, the process of cement hydration is very complicated, what is the specific working mechanism of GO in the cement matrix? The answers to these questions require a lot of research work.

Based on the great prospects of GO in the field of building materials, this study summarizes and comments

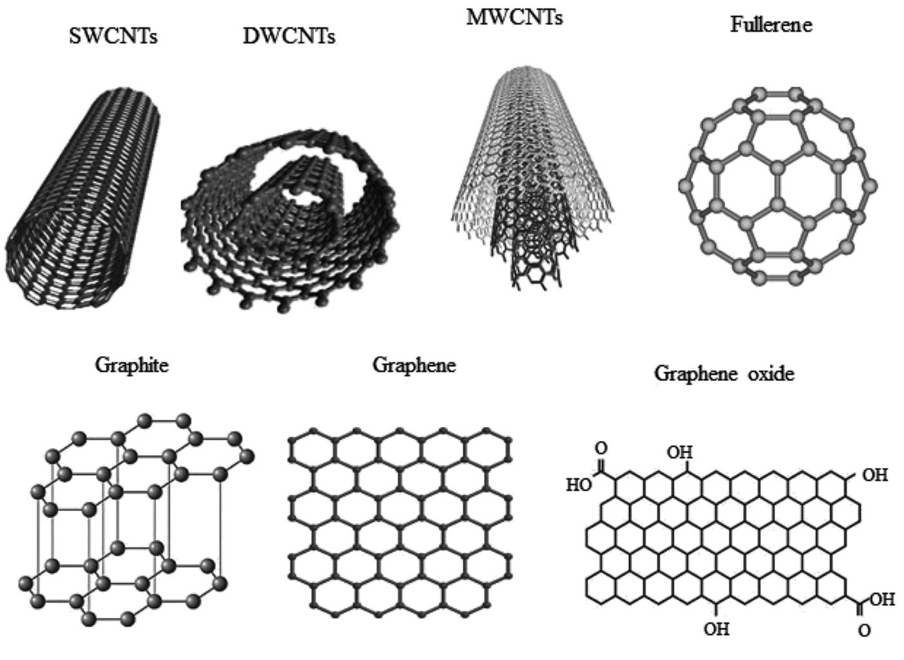

Figure 1: Different structures of carbon nanomaterials (CNMs) [27].

on the research on GO-modified cement-based materials in recent years and discusses the content that has yet to be studied in this field.

## 2 Research on the dispersion of GO

Considering a strong van der Waals force between the nanoparticles, particle agglomeration is prone to occur during the preparation and use process, which limits its application in cement-based materials. Many reports have pointed out [43–49] that $Ca^{2+}$ and $OH^-$ in the cement paste have great damage to the stability of GO dispersion. The complexation of $Ca^{2+}$ and the rapid deoxygenation reaction in an alkaline environment are the main causes of GO agglomeration. In order to explore the main reasons affecting the dispersion of GO, Zhao *et al.* [50] studied the dispersion behavior of GO in $Ca(OH)_2$, $CaCl_2$, and NaOH solutions. The results showed that a small amount of calcium (2.2 mM) is enough to destabilize the GO suspension and quickly agglomerate, and the alkaline environment is the secondary cause of GO agglomeration. It is the most important task that improving the dispersion of GO in the cement matrix to make full use of the excellent properties of GO. And all these years, people have been working on the dispersion of nano-materials and made major discoveries.

As listed in Table 1, physical dispersion and chemical modification are the main ways to improve the dispersion of nanomaterials. Unlike general nanomaterials, GO has a larger specific surface area, which makes it more difficult to disperse. Li *et al.* [43] proposed that only relying on physical dispersion can only improve the dispersibility of GO in water, but it will still reunite when exposed to $Ca^{2+}$. In contrast, through a lot of research, people found that chemical modification [86] is more effective in improving the dispersion of GO in the cement matrix. Hu *et al.* [57] synthesized triethanolamine-graphene oxide (TEA-GO) to improve the dispersion of GO in cement-based materials. After TEA modification, the –C–O–C group of GO was removed, and the –COOH group was replaced by –OH in TEA. Experiments showed that TEA-GO has better dispersibility in cement (Figure 2).

Similarly, Wang *et al.* [58] obtained modified GO (P–S–GO) through complex chemical reactions. By comparison, it is found that GO has obvious agglomeration in saturated lime water, while P–S–GO has better dispersibility and the amount of aggregation is negligible. In the study by Wang *et al.* [59], GO was reacted with rare earth elements to generate new functional groups, which

**Table 1:** Summary of dispersion for selected nanomaterials

| Nanomaterials | Dispersion method | Highlights | Disadvantages | Ref. |
|---|---|---|---|---|
| CNT | Combination of ultrasonic dispersion and surfactant | The best dispersion of carbon nanotube requires the combined action of ultrasonic dispersion and surfactants | The test operation is more complicated and needs to consider compatibility with surfactants | [22] |
| Nano $ZrO_2$ and Nano $Al_2O_3$ | 20 kHz ultrasonic frequency | Larger ultrasonic power can effectively destroy the intermolecular forces of nanomaterials | The method is complicated, and the cost is relatively high | [51] |
| Nano $TiO_2$ | Salicylic acid (SA) and arginine (ARG) were decorated into nano $TiO_2$ particle surface | When the aqueous phase was pH = 3–5, and when the concentration of NaCl 0.05–0.2 mol/L, the stability of the emulsion was allowed to stand best | The stability of the emulsion is greatly affected by temperature | [52] |
| Nano $CaCO_3$ | By incorporating dispersant (sodium dodecylbenzene sulfonate) and ultrasonic dispersion together | Sodium dodecylbenzene sulfonate can improve the electrostatic repulsion and steric hindrance between particles | The method is complicated to operate | [53] |
| Fullerene | By incorporating natural organic matter (NOM) | Natural organic matter (NOM) affects fullerene aggregation through a steric effect | This method is cumbersome and has special requirements for the type and nature of NOM | [54] |
| MWCNT | Microwave accelerated reaction | The CNT wrapped by polyvinylpyrrolidone (PVP) was somewhat less prone to agglomeration | Long-term stability of MWCNT–PVP lower due to potential partial unwrapping of PVP layer | [55] |
| Nano $SiO_2$ | Ultrasonic dispersion | When the mass fraction of nano $SiO_2$ in concrete is 1%, the most effective dispersion condition is that the ultrasonic frequency is 59 kHz, the power is 135 W, and the action time is 5 min | The conclusion of the experiment is affected by the mode of action of ultrasonic equipment and related parameters | [56] |

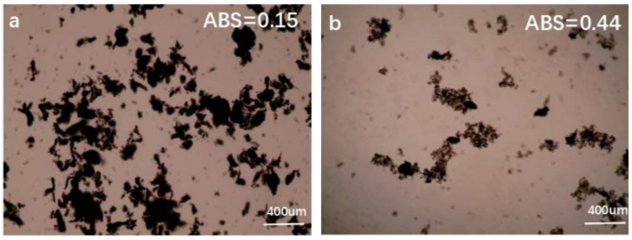

**Figure 2:** Dispersion state of GO (a) and TEA-GO (b) in pore solution [57].

reduced the interface energy and surface energy of GO and successfully enhanced its dispersion. Although the above reports have confirmed that the preparation of copolymers is an effective means to improve the dispersion of GO, the cumbersome process is complicated and time-consuming. In addition, it is also necessary to consider issues such as the compatibility of chemical reagents with the hydration system [49] and the cost of equipment [60], which has led people to pursue simpler and economical modification methods. As the properties of Superplasticizer products on the market become more and more excellent, the polycarboxylate water-reducing agent (PC), which can be well compatible with the cement system, has become the choice of many scientific researchers due to its efficient performance and simple usage [61–66,71]. The reason why PC modifies GO is to form a “protective shell” outside GO. The –COOH of PC will adsorb $Ca^{2+}$ near GO to avoid direct contact between GO and $Ca^{2+}$ (Figure 3). At the same time, the steric hindrance effect of PC [65] and hydrogen bonding effect [59] also help GO maintain a stable dispersion. With the further in-depth research on PC-modified GO, Qin *et al.* [67] found that when PCE has a larger charge density and a longer side chain, the steric hindrance effect is more significant. This means that PCE with specific properties may be more beneficial to improve the dispersion of GO in the cement matrix.

However, Yan *et al.* [46] confirmed that ultrasonic pretreatment combined with the use of water reducing agents for a certain period can make GO dispersion better. And it is shown in Figure 4 that the dispersion of GO is better after 30 minutes of ultrasonic pretreatment.

In addition to the type of water-reducing agent and the ultrasound time, the amount of water-reducing agent [66,75] and the power of ultrasound [117–119] are also research hotspots. However, the current conclusions on these two aspects are quite different. Zhao *et al.* [75] suggested that the mass ratio of PC and GO is 10:1, while Lu *et al.* [66] believed that when the mass of PC is 15 wt% of GO, it is most conducive to dispersion. The power of ultrasound has a great influence on the dispersion state of GO. Too small power will lead to a poor dispersion effect and too much power may damage the structure of GO. Li *et al.* [117] pointed out that when the ultrasonic energy is 15 Wh/L, the dispersion of GO/PVA composites is better. Liu *et al.* [118] found that when the ultrasonic power is 100 W, the dispersion state of GO/nanosilica composite is the best. Gao *et al.* [119] pointed out that the mechanical properties of GO/CNT-OPC slurry are the best when the ultrasonic time is 15 min and the power is between 81 and 94 W. Considering that the composite materials they studied are different and the parameters of ultrasonic equipment and corresponding treatment methods are different,

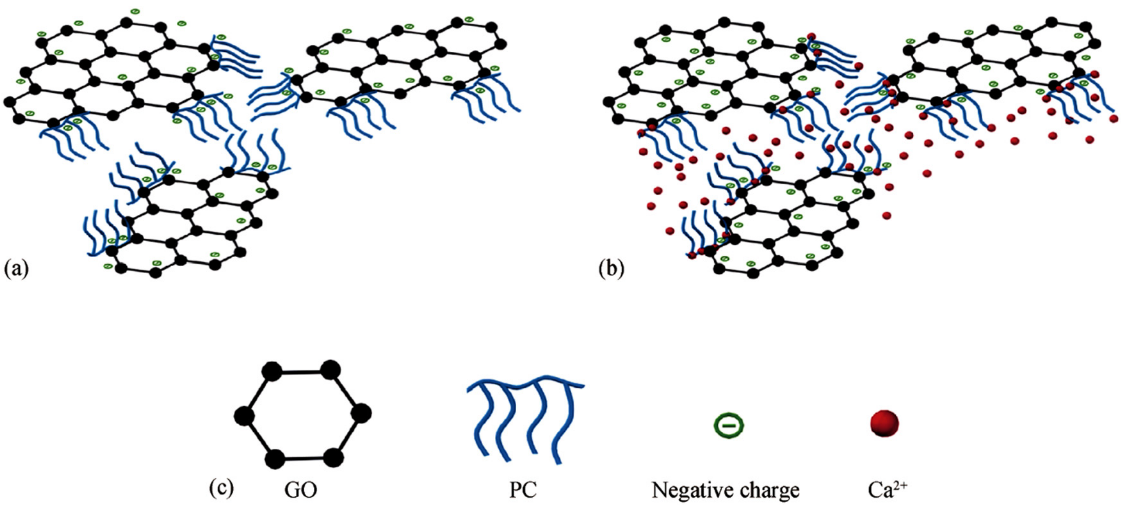

**Figure 3:** Dispersion mechanism of PC-GO [62]. (a) Dispersion mechanism in water, (b) dispersion mechanism in cement paste, (c) simulated structures of the GO, PC, negative charge and $Ca^{2+}$.

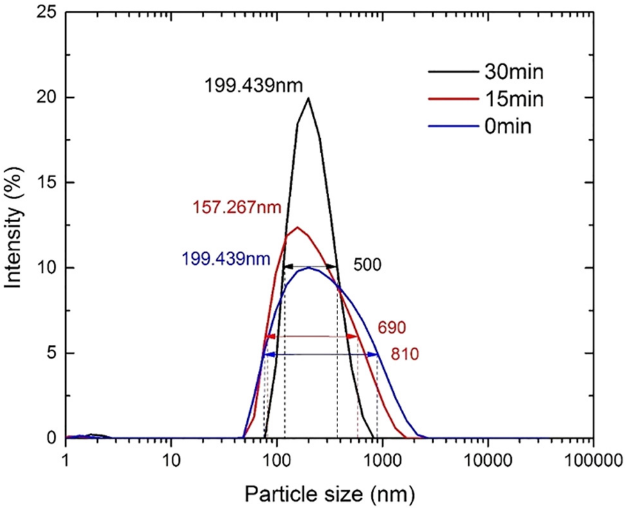

**Figure 4:** Size distributions of GO particles under different dispersing processes [46].

it is impossible to generalize universality rules from their research. The ultrasonic power and ultrasonic time still need to be determined according to each person's research object. On the premise of passing a lot of experiments, the best dispersion plan can be found.

In fact, the above-mentioned various dispersion methods are either too costly or cumbersome to operate and ultimately difficult to adapt to actual projects. At present, there are many kinds of PC in the market, which improves the workability of cement composites. The subtle differences in the molecular structure of PC will eventually play different roles. As a result, there are not many types of PC that can improve the dispersion of GO. It can be inferred that preparing a dispersant suitable for GO may improve the dispersion efficiency in the future.

# 3 Research progress of GO-modified cement-based materials

Based on the above description, people have realized that the dispersion treatment of GO is the key work of whether it can be used as a modifier for cement-based materials. The extremely large surface energy of GO makes it difficult to disperse evenly in the cement matrix. In this case, GO cement composites are difficult to meet the requirements of actual projects. It is unrealistic to talk about its application prospects in the construction field. Researchers have proposed many methods to disperse GO and confirmed that the performance of cement-based materials has been significantly improved when GO is uniformly dispersed [61–75]. There is no doubt that it has important reference significance for the development of building materials in the future.

## 3.1 The influence of GO on the fluidity of cement-based materials

The fluidity affects the construction performance of cement-based materials. Meanwhile, the low fluidity of cement paste can result in large pores and, thus, adversely affecting the mechanical properties. Many researchers have reached a consensus that GO is not conducive to liquidity. Generally, cement-based materials tend to have faster coagulation speed, increased slurry viscosity, difficulty in compaction, and more pores after adding GO. The current explanations for this phenomenon are mainly as follows: (1) the large specific surface area [68,69] and functional groups of GO promote hydration, accelerate the aggregation of cement particles, and adsorb more free water in the cement slurry; (2) some researchers believe that the formation of GO agglomerates will trap the free water in the system to a higher degree, resulting in a decrease in the fluidity of cement-based materials [44,71]; (3) the van der Waals force between GO flakes makes cement particles attract each other [76], resulting in a decrease in the fluidity of cement composites; (4) the nano-size effect of GO affects the interaction between cement particles and water-reducing agent, resulting in weakening of the repulsive force between cement particles and reducing the fluidity of cement paste [70]. In response to this problem, researchers have proposed various methods to compensate for the negative effects caused by GO, in which water-reducing agent is the more commonly used method [59,71–73]. Taking PC as an example, under the dual effects of steric hindrance [65] and electrostatic repulsion [66], it can disperse cement particles and release the accumulated water in the cement flocculation process to compensate for the negative reduction of free water caused by GO influences. In addition, some scholars pointed out that the incorporation of silica fume (SF) [45] and fly ash (FA) [85] can also improve the negative effects caused by GO. After SF is encapsulated by GO, higher fluidity and lower rheological parameters can be obtained, compared with samples under the same SF dosage. Also, FA has a "ball effect" due to its spherical glass structure, which can effectively reduce the resistance during relative sliding between particles, thereby promoting the fluidity of the slurry.

## 3.2 The microstructure and mechanical properties of GO-modified cement-based materials

Mechanical properties are the most basic and most valued performance indicators of cement-based materials in engineering applications. In recent years, many studies have pointed out the potential of GO as a cement-based material enhancer [76–100]. The research of Zeng *et al.* [70] showed that the flexural strength and compressive strength of cement-based composites can be greatly improved when the content of GO is 0.1 wt%. This is because GO has a significant promotion effect on cement hydration behavior, has a regulating effect on cement hydration products, and can develop cement hydration products in an orderly and regular manner and reduce microscopic defects. Long *et al.* [79] studied the influence of GO nanosheets on the static and dynamic mechanical properties and microstructure of cement slurry. The results show that the flexural strength and compressive strength of cement slurries with a GO content of 0.05 and 0.2 wt% after hardening for 28 days are increased by 12–26% and 2–21%, respectively, when compared with unmodified slurry. This is because the addition of GO has a positive effect on the hydration process, which in turn affects the mechanical properties of the material. SEM analysis shows that the added GO can promote cement hydration, refine the capillary pore structure, reduce the pore content, and increase the density of cement slurry. Figure 5(a–c) shows that when the cement stone was cured for 7 days, the blank group of specimens contained multiple micropores and microcracks in the relatively low-density C–S–H region. In the specimens containing GO, GO presents a unique two-dimensional structure in the slurry, which can effectively deflect or tilt and twist the cracks around the sheet, thereby hindering the formation of fine cracks and preventing the cracks from penetrating and extending (Figure 5(d–f)). This is similar to the conclusion of Pan *et al.* [90].

Wang *et al.* [85] studied the influence of GO on the hydration heat evolution and hydration degree of FA-cement composites. The results show that GO can have a synergistic effect with FA. GO regulates the microstructure by controlling the orientation of hydrated crystals, accelerating the secondary hydration of FA, reducing the total pore volume, etc., so that FA-GO-cement composites have higher mechanical strength. Yang *et al.* [86] found that the 3 and 7 days compressive strengths of cement-based composites increased by 42.3 and 35.7%, respectively, when the content of GO was 0.2 wt%. Moreover, GO has a more obvious increase in the compressive strength of cement-based materials in the early stage. The higher the GO content in the cement, the slower the increase in the later strength. Lv *et al.* [88] found that a lower content (0.01–0.03 wt%) of GO will significantly enhance the strength of cement mortar. Especially when the amount of GO is 0.03 wt%, the 28-day tensile strength increases by 78.6%, and the flexural strength and compressive strength increase by 60.7 and 38.9%, respectively. It is pointed out that this is because the GO sheet has a template function, which forms a denser structure by controlling the aggregation state and growth of cement hydration products. However, when the GO content is large, the cement hydrate crystals will flocculate due to GO, resulting in a decrease in strength. The conclusion of Li *et al.* [87] is similar. They proposed that when the GO content exceeds 0.04 wt%, the growth rate of the mortar's flexural strength begins to decrease. Yuan *et al.* [91] showed that the growth of GO cement hydrated crystals has a template regulation effect. Under the regulation of the GO crystal template, C–S–H gel can grow on GO sheets regularly and densely, which improves the density of cement-based materials. New building materials have always been a topic of great concern to people. Deng *et al.* [111] conducted an in-depth study on the heat resistance of recycled concrete and obtained the law of strength change of recycled concrete at different temperatures and recycled aggregate replacement rates. It is worth mentioning that recycled concrete has positive significance for the effective use of construction waste. However, due to the inherent defects of recycled aggregate, its performance is poor [9]. Guo *et al.* [110] confirmed that GO can significantly improve the micromechanical properties and microstructure of the transition zone of the recycled concrete interface. But they said that GO does not participate in hydration but has a certain coagulation effect during the phase distribution process, which increases the C–S–H gel contact points and increases the bulk density. The remaining research on the improvement effect of GO on the mechanical properties of cement-based materials is listed in Table 2.

In summary, it is generally accepted that a lower amount of GO can significantly improve the mechanical properties of cement-based materials, and the root cause is focused on the impact of GO on the microstructure [72–85,88–100]. However, people's current understanding of the enhancement mechanism is still divided. The enhancement mechanisms proposed in the reports can be summarized as follows:

(1) A new chemical bond is formed between C–S–H gel and GO [75,83], which could improve the load

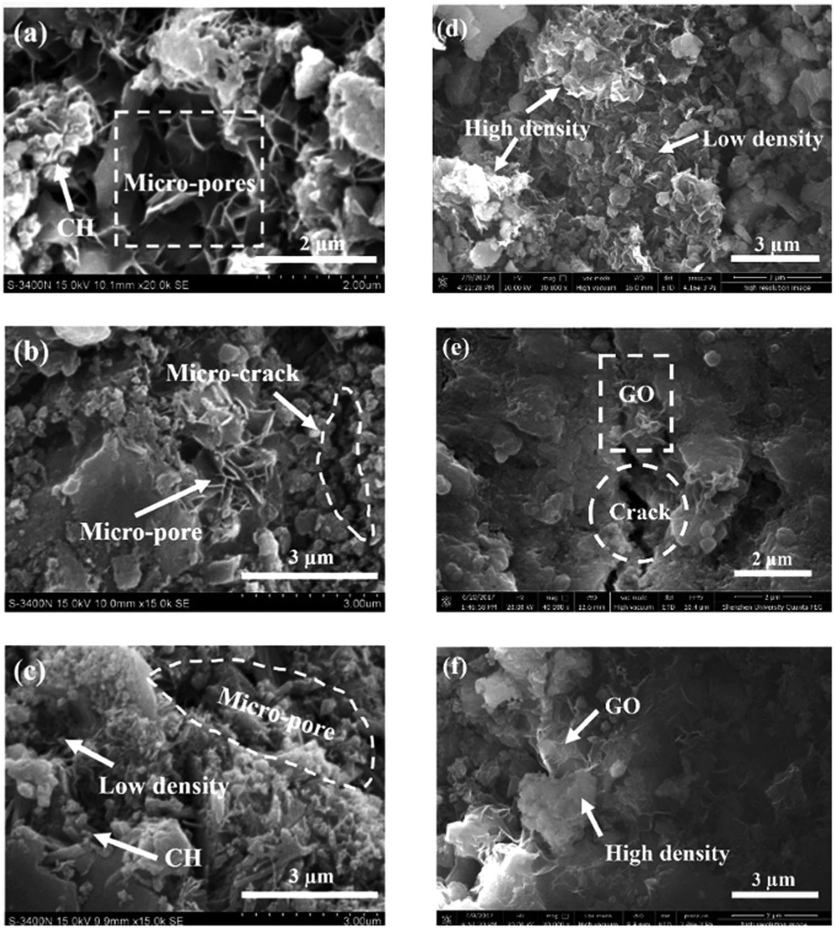

**Figure 5:** SEM images of crystal morphology at different magnifications in plain-cement paste (a–c) and in GO-cement paste (d–f) after 7 days of curing [79].

capacity. Zhao *et al.* [75] proposed that C–SH has a layered sandwich structure. As the GO sheet is inserted into the C–S–H layer, –COOH and $Ca^{2+}$ form a bond to form a denser C–S–H gel (as shown in Figure 6), thereby enhancing the increase in tough cement-based composite material.

(2) Lv *et al.* proposed that GO has a template effect [88,89,108], it makes the originally scattered hydration products gradually become compact and regular. With the increase of curing time, the hydration products grow compactly on the GO lamellae and eventually become flower-like crystals (as shown in Figure 7(a)–(f)), which enhances the density of cement-based materials.

(3) The GO lamellas are connected in vertical and horizontal directions to form a three-dimensional network structure. –COOH and $Ca^{2+}$ at the edge of GO form a COO–Ca–OOC structure to connect the three-dimensional network structure. As the hydration products are further inserted into the three-dimensional structure, a denser microstructure is formed [82] to realize the enhancement and toughness of cement-based composites (as shown in Figure 8).

(4) GO could promote cement hydration [46,66,79,80,85–87,100], refine the pores, and then achieve performance improvement of cement-based materials.

Unlike most studies, Yang *et al.* [86] concluded that although GO improves the performance of cement-based materials, it does not affect the structure of C–S–H. Cui *et al.* [109] also pointed out that the experiment of Lv *et al.* [108] has defects, that is, the possibility of carbonization of cement samples used for SEM analysis needs to be considered, which will cause the main component of flower-like crystals to be calcium carbonate instead of C–S–H. As for the current explanations of the enhancement mechanism, it is quite convincing to discuss each under the current research progress, but the inconsistency of the conclusions in the literature also shows that the current behavior of GO in the hydration process

Table 2: Effect and mechanism of GO on performance improvement of cement-based materials

| Optimal dose of GO (wt%) | Curing age (days) | Intensity growth rate (%) | | | Highlights | Ref. |
|---|---|---|---|---|---|---|
| | | Tensile str. | Flexural str. | Compressive str. | | |
| 0.07 | 7/28 | — | 200/85.7 | 31.3/21.9 | SEM showed that GO makes the hydrated crystals more regular and finally forms a dense structure | [72] |
| 0.03 | 28 | 65.5 | 60.7 | 38.9 | XRD and SEM analysis showed that GO changed the morphology and arrangement of hydrated crystals | [74] |
| 0.16 | 14 | — | 11.62 | 3.21 | GO connects nano cracks and locks cement hydration products | [78] |
| 0.05 | 28 | — | 12–26 | 2–20 | SEM showed that the incorporation of GO can increase the density of the C–S–H phase and inhibit the propagation of cracks | [79] |
| 0.03 | 28 | — | 55.8 | 31.9 | XRD and EDS analysis showed that GO can adjust cement hydration products into standardized hydration crystals through template effect | [87] |
| 0.03 | 3/28 | 51.0/78.6 | 70.7/60.7 | 45.1/38.9 | FT-IR, XRD, and SEM analysis showed that GO can effectively adjust the microstructure of hydrated crystals | [88] |
| 0.04 | 28 | — | 14.2 | 37.0 | GO can densify cement slurry and reduce porosity | [89] |
| 0.05 | 28 | — | 41–59 | 15–33 | SEM analysis showed that GO suppresses the occurrence of cracks | [90] |
| 0.1 | 28 | — | — | 10.2 | GO nanosheets (GONPs) have an obvious remodeling effect on the microstructure of cement paste. AFM scanning and SEM images showed that a better interface bond is formed between GONPs and the C–S–H gel around them | [92] |
| 1.6 | 28 | — | — | 20 | Good workability (w/c = 0.6) is conducive to the uniform dispersion of GONPs. They act as fillers and reactants to enhance the microstructure of the cement paste | [94] |
| 0.06 | 28 | — | 138.44 | 23.89 | SEM showed that GO makes the growth of hydrated crystals more regular, and the micropores and cracks tend to be reduced and refined | [97] |
| 0.03 | 28 | — | 23.83 | 10.85 | GO optimized the morphology and distribution of hydration products; at the same time, the filling effect of GO made the hardened slurry more uniform and dense | [98] |
| 0.03 | 28 | — | — | 9 | GO provided nucleation sites to promote cement hydration | [99] |
| 0.04 | 7 | — | 83 | — | GO agglomerates and cement matrix have good adhesion | [100] |

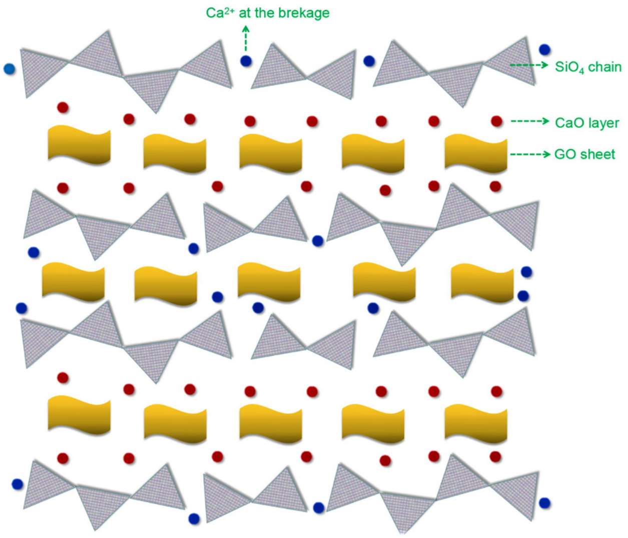

**Figure 6:** Schematic diagram of the mechanism of GO regulating C–S–H proposed by Zhao *et al.* [75].

is not clear. In addition to the different microscopic models established, the researchers' conclusions on the optimal dosage and modification effect of GO are also quite different, which can also be seen from Table 2. Therefore, deep exploration of the micro-control mechanism of GO on cement-based materials is still the focus of the next work.

In addition, compared with conventional cement-based concrete, the research on GO in recycled concrete, ultra-high performance concrete, and geopolymers is not rich enough. Expanding the scope of research is the next important work.

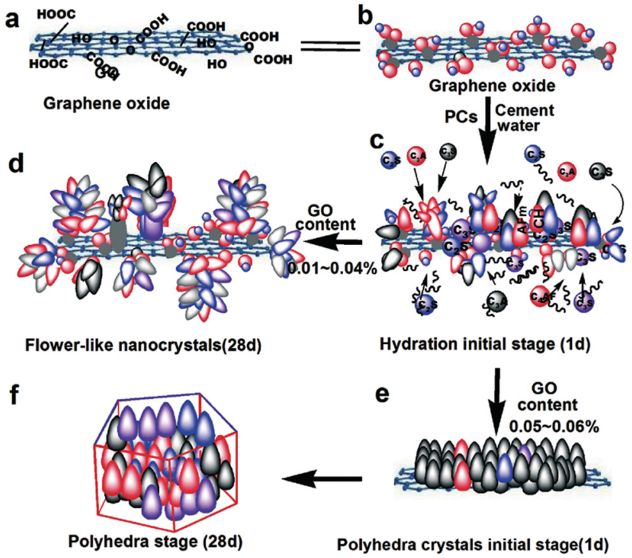

**Figure 7:** Schematic diagram of GO regulating the hydration crystal of cement proposed by Lv *et al.* [89].

## 3.3 The durability of GO-modified cement-based materials

Durability refers to the characteristics of cement-based materials that could withstand various harsh environments [101]. Due to its structural defects, cement-based materials are susceptible to corrosion by factors such as carbonization, alkali-silica reaction (ASR), chloride corrosion, freeze-thaw cycles, fire, thermal cracking, and bacteria [102,105] during service, which will have a significant impact on its mechanical properties and service life. Improving the durability of cement-based materials is an important task in the construction industry. At present, people have confirmed the conclusion that a dense and regular structure is beneficial to the durability of cement-based materials. GO participates in regulating the crystal structure of cement hydration products, improving the internal porosity of cement mortar and the weak area of interface transition, and regulating the hydration products to form a regular microstructure. The densification of the internal microstructure slows down and hinders the erosion of $Cl^-$, $SO_4^{2-}$, and other corrosive ions and improves the impermeability and durability of cement-based composites [96]. The literature [103] pointed out that the special lamella structure of GO will form a sponge-like structure in the cement mortar, limiting the penetration depth of $Cl^-$, and then improving the performance of the cement mortar against $Cl^-$ penetration. The study also shows that GO can also improve the carbonization resistance and frost resistance of cement-based materials. Lv *et al.* [104] proposed that GO greatly reduces the number of pores and cracks by regulating the growth of hydration products so that cement-based materials can better resist external corrosion factors. Yang *et al.* [96] studied the effect of GO on the corrosion resistance of cement mortar in composite salt solutions. Through SEM and energy spectrum analysis, the reference specimen was compared with specimens with different GO content, and it was found that the internal structure of the reference specimen was severely damaged after corrosion. The overall look is messy and sparse, and more corrosive ions have penetrated into the specimen. When 0.03 wt% GO is added, the internal structure of the cement mortar is compact and regular, without obvious corrosion marks. It is proved that GO improves the corrosion resistance of the cement matrix

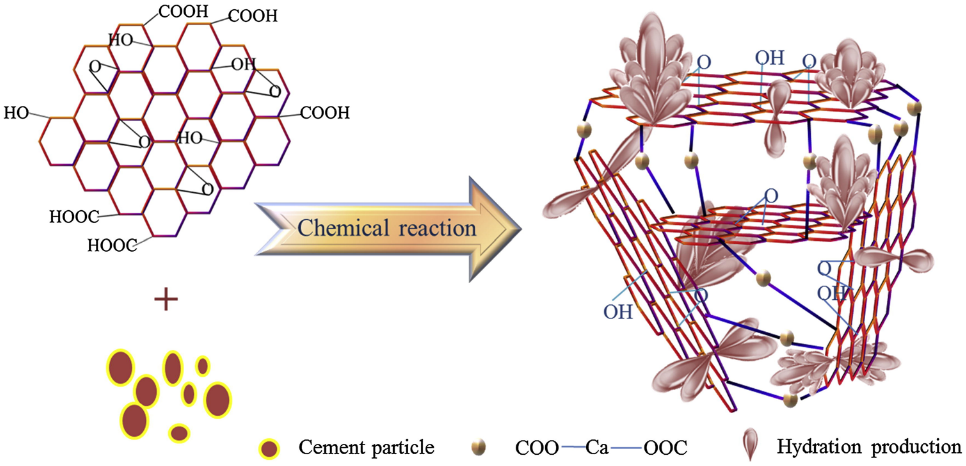

**Figure 8:** Schematic diagram of the mechanism of GO regulating hydration crystals proposed by Wang *et al.* [82].

by adjusting the density of the internal structure and reducing the pore volume. Mohammed *et al.* [106] studied the heat resistance of GO mixed with ordinary and high-strength concrete. The results showed that GO keeps the specimens with higher residual strength and better crack resistance after being exposed to high temperatures. This is because GO is incorporated into the cement matrix to form nanometer and micrometer channels, which helps release the vapor pressure and prevent a large amount of peeling. Gao *et al.* [107] studied the influence of graphene oxide/multi-walled carbon nanotubes (GO/MWCNT) composites on the impermeability of cement-based materials and found that GO/MWCNT materials can be used as nucleation sites for the growth of hydration products, promoting hydration reaction. The well-dispersed MWCNT has the crack bridging ability and can inhibit the propagation of nano-scale cracks. The interfacial adhesion between the GO sheet and the hydrated product between the micropores may significantly reduce the porosity and average pore size of the sample (Figure 9).

Guo *et al.* [112] found that GO can significantly reduce the gas permeability coefficient of recycled concrete at different curing ages. The reason is that the nucleation effect of GO can adjust the structure of hydrated crystals and improve the microscopic cracks of recycled concrete. Zhang *et al.* [113] stated that GO reduces the voids in the cement-based self-leveling microstructure by adjusting the structure of cement hydration products, thereby effectively inhibiting the intrusion of $Cl^-$. At the same time, due to the enhanced compactness of the cement matrix, its wear resistance can be effectively improved. Li *et al.* [114] also proved that the synergistic effect of GO and PVA fibers can significantly improve the pore structure of cement-based materials, reduce porosity, improve the resistance of cement-based materials to $Cl^-$ penetration, and reduce the shrinkage of cement-based materials. Interestingly, in addition to improving the microstructure of cement-based materials, GO can also improve durability in other methods. Yu *et al.* [115] prepared a GO-epoxy resin (EP) composite coating. The results showed that the GO-EP composite coating significantly blocked water molecules and ions in the solution, which can greatly improve the impermeability of concrete. They prepared a GO-epoxy resin (EP) composite coating. The results showed that the GO-EP composite coating blocked water molecules and ions in the solution. It is expected that this technology will be introduced into the construction industry to improve the impermeability of concrete. Zhang *et al.* [116] used GO to modify isobutyltriethoxysilane. A sol-gel method was used to prepare GO/isobutyltriethoxysilane composite emulsion. SEM and EDS showed that the composite emulsion can form a dense hydrophobic layer on the surface of the concrete, thereby improving the impermeability of the concrete.

These studies have proved that GO could significantly improve the durability of cement-based materials. It is of great significance to maintain the performance of cement-based materials in response to acid–base corrosion, high temperature, freeze-thaw, and other influencing factors. These characteristics of GO have important applications in some coastal areas or heavy saline areas.

## 4 Discussions and future trends

Although GO-cement-based materials have shown great application value and potential, the current research has not yet gone out of the laboratory, and the wide application of it in practical engineering also need to solve the following challenges:

(1) To solve the dispersion problem: many studies have shown that the agglomeration of GO has a serious impact on the mechanical properties and durability of cement-based materials. Although many researchers have proposed methods such as ultrasonic dispersion, copolymer modification, and addition of water-reducing agents, the feasibility of these methods has not yet been uncertain in large-scale engineering applications. On the one hand, the high cost needs to be considered; on the other hand, it is undoubtedly more difficult to maintain the dispersion effect in large-scale projects.

(2) To improve the problem of decreased fluidity: due to the higher surface energy and the rich hydrophilic groups and other factors, GO will consume more free water in the system, resulting in the reduced fluidity of cement slurry, and thus affecting the working performance of cement-based materials. Especially when the coarse aggregate is added to prepare concrete, the frictional resistance of the system is further increased, which is not conducive to the workability of concrete. Although some people have proposed methods to compensate for the decline in liquidity, the incorporation of GO still exacerbates the loss of liquidity over time and increases the difficulty of construction.

(3) Expanding the depth and breadth of research: at this stage, researchers have constructed different models of the regulation mechanism of GO on the mechanical properties of cement-based materials and made corresponding explanations at the micro-level. However, the diversification of the mechanism also shows that people are not clear about the exact behavior of GO in the hydration process, and the experimental results of different researchers are quite different. Second, the current research is more focused on ordinary Portland cement-based materials, while other researches such as recycled concrete, ultra-high performance concrete (UHPC), and geopolymers materials are still scarce.

(4) Expanding durability research: although scholars have studied the role of GO in cement-based materials in terms of freeze-thaw resistance, carbonization resistance, high-temperature resistance, and chloride salt penetration, there are few other durability studies such as shrinkage test, early crack resistance test, and alkali-aggregate reaction test. In addition, the time span of durability research must be longer.

(5) Cost reduction: another factor restricting the application of GO in engineering is its higher price. Although the cost of GO is lower than that of original graphene, it still does not have advantages compared with traditional modification methods.

(6) Research on nanomaterials needs to be further deepened: at present, most studies still focus on the performance of nanomaterials themselves. The structural design and optimization of nanomaterials themselves need to be further explored. Improving the richness and controllability of the original

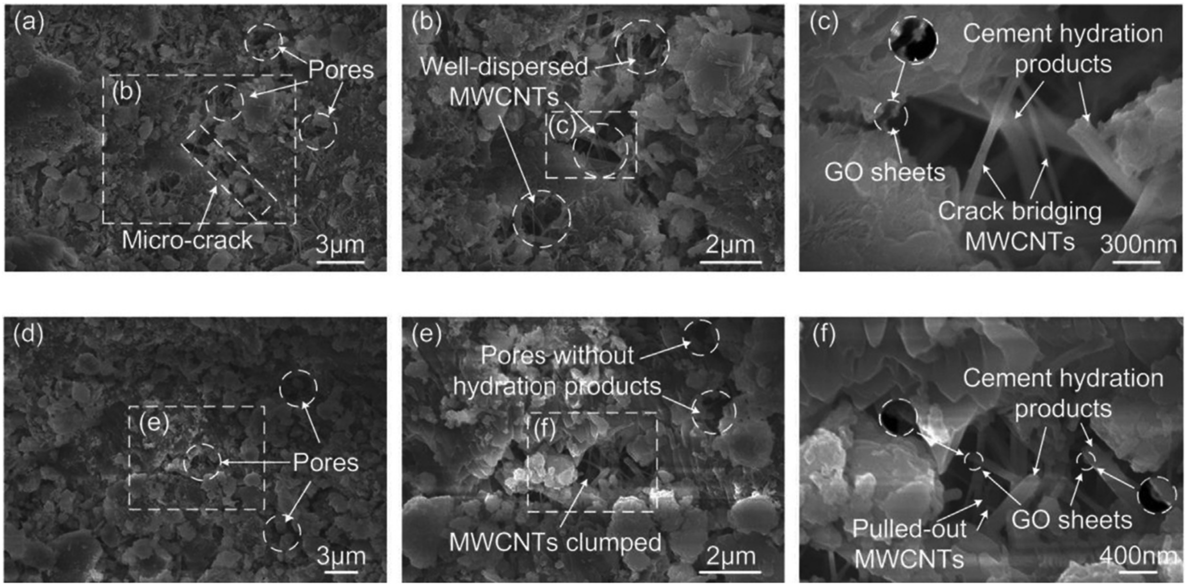

**Figure 9:** SEM images of GO/MWCNT-OPC pastes: (a–c) Sample G/M-1; (d–f) Sample G/M-3 [107].

nanomaterials may be the main research direction in the future.

## 5 Conclusions and prospects

Reviewing previous studies, one can get the following conclusions:

(1) The advantage of GO is its unique layer structure and excellent physical properties. Compared with other reinforcing materials, the interface adhesion between GO and cement-based materials is stronger. The nano-filling effect and nucleation effect of GO not only can effectively reduce the pore volume but also can adsorb hydration products to the surface and regulate them into the dense and regular structure. GO is phenomenal in improving the mechanical properties and durability of cement-based materials, which is of great significance for the research and development of high-performance cement-based products.

(2) At this stage, researchers have understood the influence of GO content ratio on the mechanical properties of cement-based materials, established a related hydration model, and have a preliminary understanding of the enhancement mechanism. These studies not only have great reference value for those who are just beginning to understand GO-cement-based materials, but also lay a solid theoretical and experimental foundation for the next stage of research on cement-based composite materials.

(3) The synergistic effect of GO with other materials such as surfactants, SF, FA, and PVA fibers has been proven by many researchers. The high activity of GO makes it extremely malleable, and new features can be obtained by functionalizing it, this will depend on the design requirements for concrete.

(4) The limitation of the current research is mainly that there are still various shortcomings in the dispersion method of GO, and a method that combines cost-effectiveness and dispersion efficiency has not been found. In addition, people's understanding of nanomaterials is not thorough enough, and there are still many differences in the working mechanism of GO. As shown in Table 2, in the current different studies, the modification effects of GO are quite different, which makes people worry about the stability of GO in cement composites.

(5) Cement-based materials are currently the most mainstream building materials, and their performance improvement is of great significance to the technological development and innovation in the construction field. The current research proves that GO provides a very potential way to improve the performance of cement-based materials. However, it should be pointed out that more research work should be done to solve the current problems in the future. This is the premise for GO to be widely used in the construction industry.

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