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Adding Graphene Materials On The Mechanical Properties And Durability Of Concrete

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KEYWORDS

Concrete; graphene oxide;

Shell powder ;

Mechanical properties;

Durability

ABSTRACT

Cement concrete, with its advantages of easy molding, good integrity, and significant economic benefits, is the most widely used building material in the world. However, the disadvantages of cement-based composites, including their inherent brittleness, low tensile strength, and susceptibility to corrosive environments, have always been major concerns. Within this framework, the application of nanomaterials and nanotechnology in cement-based materials has become the most attractive solution . Graphene, as a nanomaterial with a unique two-dimensional structure, offers new insights into improving concrete performance through its nano-filling effect and interfacial reinforcement. This study focuses on exploring the effects of different graphene oxide dosages on the workability, mechanical properties, and economic efficiency of concrete. This paper aims to provide a scientific understanding of the application of graphene materials in cement-based materials to promote their future practical application . The results show that the incorporation of graphene oxide effectively improves the compactness of concrete, reduces water absorption and dehydration rates, and enhances the compressive strength, splitting tensile strength, impermeability, resistance to chloride ion attack, and resistance to wet-dry cycles. The overall performance of concrete reaches its optimal state when the graphene oxide dosage is 0.050% . This study provides data support and theoretical basis for the application of graphene oxide in concrete.

INTRODUCTION

Cement concrete is widely used in civil infrastructure construction around the world due to its ease of molding, significant economic benefits, and excellent compressive strength. Currently, global concrete consumption reaches 30 GT annually, making it the most consumed synthetic material in the world [1]. However, due to the inherent quasi-brittleness of concrete and its tendency to deteriorate in harsh environments, it has always been a focus of scientific attention [2-3] . Moreover, the environmental hazards caused by concrete production are not to be underestimated. According to statistics, the energy consumption required for cement production accounts for 7% of global industrial energy consumption, and every ton of cement produced emits 1 ton of CO2 into the atmosphere, accounting for 8% of global anthropogenic carbon dioxide emissions [4-5] . Even more worrying is the unavoidable water consumption caused by concrete production. It is estimated that concrete production consumes approximately 18% of global industrial water consumption annually [6] . Therefore, researchers are committed to improving the performance of cement-based materials to reduce the consumption of concrete components. Different types of fibers (including carbon fiber, jute, steel fiber, etc.) have been used in cement-based materials to improve their mechanical properties and durability by controlling the incubation and propagation of microcracks [7-9] . However, the reinforcement obtained by relying on fibers does not change the structure and toughness of the hardened cement matrix, and brittleness and cracking still occur at the nanoscale [10] .

In recent years, significant progress has been made in the application of nanomaterials in the field of concrete,

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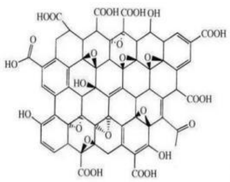
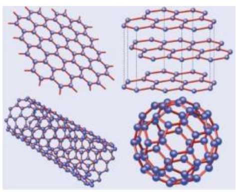
providing new opportunities for improving concrete performance. Graphene's structure can be viewed as a single layer of graphite, with each carbon atom forming covalent bonds with three adjacent carbon atoms through sp^2 hybridization, constituting a hexagonal planar network structure. This structure gives graphene extremely high stability and strength. It is reported that a single layer of graphene is only 0.35 nm thick, yet possesses extremely high mechanical properties (theoretical tensile strength and Young's modulus reach 130 GPa and 1.1 TPa, respectively), thus being considered one of the best nanomaterials for improving the performance of cement-based materials. However, despite the broad application prospects of graphene in concrete, in-depth research is still lacking regarding its specific application in self-compacting concrete with shell powder and its synergistic mechanism with shell powder. Shell powder, as a concrete admixture, can save on concrete costs.[11] Therefore, combining graphene with shell powder and applying it to self-compacting concrete is expected to further improve concrete performance and achieve economic savings. Figure 1 shows graphene.



(a) Chemical bonds in graphene oxide



(b) Graphene structure

Fig.1. graphene

1.Literature Review

At present, graphene nanosheets (GNPs), graphene oxide (GO) and reduced graphene oxide (rGO) are the three main forms that can be used in industrial applications [12]. Related studies have shown that adding a small amount of GNPs can accelerate ion exchange, promote cement hydration reaction, and produce more hydration products; on the other hand, GNPs with high specific surface area can inhibit crack propagation and incubation, so that the cement matrix can obtain better mechanical properties and durability [13]. Wang et al. [14] proved that uniformly distributed GNPs can effectively improve the mechanical properties of cement composites by reducing the stress under external load. Qureshi et al. [15] analyzed and compared the mechanical properties of cement paste with 0.01% to 0.16% of GNPs by weight of cement, and found that 0.02 wt% of GNPs had the best effect on improving the compressive and flexural strength of cement paste, but when the concentration of GNPs in the cement matrix exceeded 0.02 wt%, the improvement effect of cement paste on compressive and flexural strength gradually decreased. Ho et al. [16] studied the effects of different dosages (0.01%, 0.03%, 0.05%, 0.07%, 0.1% and 0.3%) of ultra-large (56±12 mm) GNPs exfoliated by electrochemical peeling on the compressive and tensile strength of cement mortar. The results showed that adding 0.07% GNPs was the optimal dosage, and the compressive and tensile strength of cement-based composites increased by 34.3% and 26.9% respectively after 28 days. This enhancement is mainly attributed to: on the one hand, the incorporation of GNPs increased the hydration degree of cement paste, resulting in the generation of more hydrated calcium silicate gel; on the other hand, the unique barrier effect and filling effect of GNPs increased the penetration path of harmful substances such as water molecules and chloride ions into the cement matrix, which improved the durability of cement-based materials [17-18] . However, due to the strong van der Waals forces between GNPs layers and their own hydrophobicity, GNPs are prone to agglomeration and it is difficult to achieve uniform dispersion in aqueous solutions. Non-uniformly dispersed GNPs not only limit the excellent performance of GNPs, but also cause the deterioration of the performance of cement-based materials.

As a derivative of graphene, graphene oxide (GO) has abundant oxygen groups (hydroxyl, epoxy, etc.) on its basal surface, which makes GO highly hydrophilic. In addition, the high density of oxygen-containing functional groups can promote the activation reaction of mineral components in cement and accelerate the hydration of cement [19]. Studies have shown that the nucleation effect of GO can promote the early hydration reaction of cement, generate more CSH to provide a dense microstructure, and improve the mechanical properties and durability of cement matrix to varying degrees [20-24] . In order to understand the reasons for the improvement of mechanical properties of GO and the strengthening mechanism, researchers have devoted themselves to the study of the hydration process, microstructure and composition of GO-based cement composites. Zhao et al. [25] showed that the compressive strength of GO-reinforced cement mortar increased by 34.1%, 26.9% and 22.6% after 3 days, 7 days and 28 days of hydration, respectively. In addition to the early stage of cement hydration, contradictory microstructure characterization results and various possible strengthening mechanisms of GO in cement composites have also been reported in the literature. The aforementioned scientists' research on graphene materials shows that, when graphene is incorporated into concrete mixtures, it can improve the mechanical properties and durability of concrete, inhibit crack propagation, and significantly reduce the porosity of the cement skeleton.

2.Experimental Design

2.1.Experimental Materials

Cement: P·O R45 cement, main chemical composition shown in Table 1, physical and mechanical properties shown in Table 2; Water: laboratory tap water; Sand: natural river sand, meeting the standard for construction sand, bulk density less than 1.5 g/m3, fineness modulus of 1.9, moisture content less than 1%; Stone: gravel , bulk density not less than 2.6 g/m3, particle size between 5 mm and 7 mm; the graphene oxide used in the research has a spacing of less than 1 nm , as shown in Figure 2. Shell: calcined and crushed, particle size less than 0.5 mm, bulk density less than 2.9 g/m3, fineness modulus of 2.9, moisture content less than 1%, as shown in Figure 3. The water-reducing agent selected is a polycarboxylate-based high-efficiency water-reducing agent that meets the requirements of "Concrete Admixtures" (GB 8076-2008) .

Table.1. Main components of cement (%)

Ca O	SiO_2	Al2O3	Fe2O3	SO3	Na 2 O	K_2O	M gO
6 4.12	2 2.31	6.42	4.37	1.1	0.75	0.56	0.37

Table.2 . Physical and mechanical properties of cement

Standard consistency water requirement /%	Specific surface area / (m² / kg)	Condensation time/min		Compressive strength / MPa		Flexural strength / MPa	
		Initial condensation	Final Condensation	3 d	28 days	3 d	28 days
2 8	3 60	1 75	2 35	2 7.5	4 9.0	5.5	8.0

Fig.2. Graphene used in the experiment

2.2.Experimental Proportions

To explore the effects of graphene oxide (GO) on concrete, this study designed a concrete mix proportion as follows : cement :shell powder: water : fine aggregate : coarse aggregate : water-reducing agent = 400 : 100 : 180 : 820 : 1020 : 6 (kg/m³) . In the mix design, the cementitious material consisted of cement and mineral powder, with the mineral powder serving as a partial substitute for cement, representing 25% of the total cementitious material. To investigate the effect of GO on concrete performance, four mix proportions were designed with GO dosages of 0% , 0.025% , 0.050% , and 0.075 % of the total cementitious material, corresponding to dosages of 0 , 0.125 , 0.250 , and 0.375 kg/m³ , respectively defined as GO-00 , GO-25 , GO-50 , and GO-70 . In addition, a fixed water-to-cement ratio of 0.36 is used to achieve the best balance between strength and workability.

2.3.Test Methods

To determine the practical building performance and mechanical properties of graphene oxide concrete, we need to conduct slump, mechanical properties , and durability tests on the specimens to obtain relevant data and analyze its related mechanical properties.

The workability of the concrete mixture is determined according to the "Technical Specification for Application of Self-Compacting Concrete" (JGJ/T 283-2012), with slump flow used to characterize rheology. The operating procedure is as follows: Fresh concrete is poured into a standard slump cone; after vertically lifting the cone, the maximum flow diameter of the static concrete is measured (accurate to 5 mm); the self-compacting grade is determined based on the diameter value (>650 mm is the highest grade, 550-650 mm is suitable for pumping).

For mechanical property testing, compressive strength and splitting tensile strength were conducted according to the "Standard for Test Methods of Physical and Mechanical Properties of Ordinary Concrete" (GB/T50081-2019) .

Compressive strength testing used 100mm × 100mm × 100mm cube specimens, while splitting tensile strength testing used ⌀ 100mm × 200mm cylindrical specimens. After curing under standard conditions for 7, 28 , and 90 days, the specimens were subjected to loading tests using a hydraulic universal testing machine, and the corresponding strength values were recorded.

The water absorption-dehydration test was conducted according to the "Standard for Test Methods of Long-Term Performance and Durability of Ordinary Concrete" (GB/T50082-2009) . After curing, the concrete specimens were sealed with paraffin wax on their top and bottom surfaces, with the sides serving as reserved water absorption surfaces. The specimens were placed in a humid environment and weighed at regular intervals (e.g., 0.5-48 hours) to record the changes in water absorption mass. After the water absorption test, a dehydration test was conducted in a drying oven to observe the changes in mass of the specimens during the drying process. In the wet-dry cycle test, the specimens were placed in a 3% sodium chloride solution and underwent alternating drying and wetting processes. After the wet-dry cycle was completed, the ultrasonic pulse velocity (UPV) test was performed, and the UPV change rate was calculated. The calculation is shown in formula (1) . After the wet-dry cycle was completed , a 0.10 % concentration silver nitrate solution was sprayed onto the surface of the specimens. The silver chloride generation area (white) and the unreacted area (brown) formed a clear boundary line. The penetration depth of chloride ions was determined by measuring the depth of the white area. The rapid chloride ion penetration test adopts the ASTM C1202-12 standard , using a cylindrical specimen of ⌀ 100mm × 50mm , with an applied voltage of 60V for 6 hours . The resistance of concrete to chloride ion erosion is evaluated by measuring the total amount of electricity applied (coulomb value).

$$\text{UPV change rate} = \frac{\text{Initial UPV value} - \text{UPV value after cycle}}{\text{Initial UPV value}} \tag{1}$$

Corrosion resistance testing was conducted using accelerated corrosion testing. The specimen was a 100mm × 200mm cylinder with embedded reinforcing steel bars, immersed in a 3% NaCl solution, and subjected to a 30V DC current. The time it took for the concrete to crack due to steel bar corrosion was recorded. In addition, to assess the concrete's resistance to chloride ion attack, the penetration depth of chloride ions in the concrete was measured by spraying a 0.10 equivalent concentration of silver nitrate solution. Throughout the corrosion process, the half-cell potential of the specimen was measured every 24 hours for further analysis of its corrosion resistance.

3.Results and Discussion

3.1.Performance

The slump spread test results of concrete with different graphene oxide (GO) dosages are shown in Table 1. With increasing GO dosage, the flowability of the concrete initially increased slightly and then gradually decreased. The flowability reached its maximum value (approximately 720 mm) when the GO dosage was 0.050%, indicating that an appropriate amount of GO can improve the rheological properties of the paste in synergy with water-reducing agents. This is mainly attributed to the dispersing effect of oxygen-containing functional groups on water molecules and cement particles on the GO surface. When the dosage increased to 0.075%, the flowability decreased due to enhanced interactions between GO layers and increased paste viscosity, but it still met the construction requirements for self-compacting concrete (>550 mm).

Group	GO parameters (%)	Collapse spread (mm)	Liquidity rating
G0-00	0	680	SF2
G0-25	0.025	700	SF2
G0-50	**0.050**	**720**	**SF3**
G0-75	0.075	650	SF2

Table.3. Workability of concrete with different GO content

3.2.Mechanical Properties

Figure 3 and Table 2 shows the changes in compressive strength and splitting tensile strength of concrete at 7 days, 28 days, and 90 days under different GO dosages. Overall, the addition of GO significantly improved the early and long-term strength of concrete. When the GO dosage was 0.050%, the 28-day compressive strength increased by approximately 18.6% compared to the control group (GO-00), and the splitting tensile strength increased by

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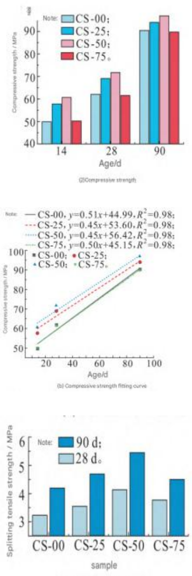
approximately 22.4%. The main mechanisms of strength improvement include: filling effect: GO nanosheets can fill the micropores in cement paste, improving matrix density; nucleation effect: the abundant functional groups on the GO surface promote heterogeneous nucleation of hydration products (CSH), accelerating the hydration process; bridging and crack-inhibiting effect: GO forms a two-dimensional network between microcracks, inhibiting crack propagation and enhancing the toughness of the material. It is worth noting that when the GO dosage exceeds 0.050%, the strength growth trend slows down and even slightly declines, which may be related to local defects caused by GO agglomeration.



(a)Compressive strength
(b) Compressive strength fitting curve
(c) Splitting tensile strength

Group	GO content (%)	Compressive strength (MPa)			
		7 days	28 days	90 days	Remark
GO-00	0.000	32.5	45.0	48.2	
GO- 25	0.0 25	35.2	49.8	52.1	
GO- 50	0.0 50	**38.1**	**53.4**	**55.8**	
GO- 75	0.075	37.2	51.9	54.3	

Group	GO content (%)	Splitting compressive strength (MPa)			
		7 days	28 days	90 days	Remark
GO-00	0.000	2.8	3.5	3.7	
GO- 25	0.0 25	3.1	3.9	4.0	
GO- 50	0.0 50	**3.4**	**4.3**	**4.4**	
GO- 75	0.075	3.2	4.1	4.2	

Note: Bold data represents the optimal values (GO-50 group).

Table.4. Effect of different GO dosages on the mechanical properties of concrete

3.3.Durability

3.3.1.Water absorption and dehydration behavior

Table 3. shows the water absorption rate of concrete with different GO admixtures over time . With increasing GO admixture, both the water absorption rate and saturated water absorption rate of the concrete decreased significantly. The 48-h water absorption rate of the GO-50 group was approximately 34% lower than that of the control group, indicating that GO effectively filled the capillary channels and hindered water transport. Dehydration tests further confirmed that GO-modified concrete also experienced a slower water loss rate, which is beneficial for maintaining stable internal humidity in alternating wet and dry environments and reducing the risk of shrinkage cracking.

Time (h)	GO-00 (0%)	GO-25 (0.025%)	GO- 50 (0.050 %)	GO- 7 5 (0.0 7 5%)
0.5	1.2	1.0	0.8	0.9
1	1.8	1.5	1.1	1.3
2	2.5	2.0	1.5	1.8
4	3.2	2.6	1.9	2.2
8	4.0	3.2	2.3	2.7
twenty four	5.1	4.0	2.9	3.4
48	**5.8**	**4.5**	**3.4**	**3.9**

Note: 48-hour data show that the water absorption rate of the GO-50 group was reduced by approximately 41.4% compared to the control group (GO-00).

Table.5.showing the change in water absorption rate of concrete with different GO admixtures over

3.3.2.Resistance to chloride ion corrosion

The total charge flux measured by the rapid chloride ion penetration test (ASTM C1202) is shown in Table 4. The addition of GO significantly reduced chloride ion permeability; the charge flux of the GO-50 group was only about 42% of that of the control group. The silver nitrate spray test also showed that the chloride ion penetration depth decreased significantly with increasing GO dosage. This indicates that the dense microstructure and hydrophobic barrier formed by GO effectively prolong the penetration path of the corrosive medium.

Group	GO content (%)	Total power consumption (coulombs)	Standard deviation
GO-00	0.000	3250	±120
GO- 25	0.0 25	2100	±95
GO- 50	**0.0 50**	**1350**	**±80**
GO- 75	0.075	1800	±90

*Note: The GO-50 group (0.050%) had the lowest total charge, which was about 58.5% lower than the control group (GO-00).

Table.6. Results of rapid chloride ion penetration test (total electrical charge) for concrete with different GO admixtur

3.3.3.Resistance to wet and dry cycling

After 30 wet-dry cycles, the change rate of ultrasonic pulse velocity (UPV) of each group of specimens is shown in Table 5. The GO-50 group had the lowest UPV change rate (only about 1.8%), indicating that its internal structure suffered the least damage during the cycle. Scanning electron microscopy (SEM) observation (Table 6.) revealed that the cracks in the GO-modified specimens were smaller and more dispersed, further confirming the inhibitory effect of GO on the development of microcracks.

Group	GO content (%)	UPV change rate (%)
GO-00	0.000	4.2
GO- 25	0.0 25	2.8
GO- 50	**0.0 50**	**1.8**
GO- 75	0.075	2.5

*Note: The GO-50 group had the lowest UPV change rate (1.8%), indicating that it had the best resistance to wet-dry cycling.

Table.7. Change rate of ultrasonic pulse velocity (UPV) after wet-dry cycles in concrete with different GO admixtures

Group	GO content (%)	Image description
GO-00	0.000	Numerous capillary pores are visible, indicating a loose CSH gel structure with microcracks.
GO- 25	0.0 25	The number of pores is reduced, the CSH gel is more dense, and a small number of GO sheets are visible dispersed within it.
GO- 50	0.0 50	The structure is extremely dense with very few pores. GO sheets are evenly distributed and bridged between hydration products, forming a continuous network.
GO- 75	0.075	The structure is relatively dense, but slight GO aggregation is visible in some areas, forming a few weak interfaces.

Table.8.comparison of scanning electron microscope (SEM) images of concrete.

3.3.4.Resistance to wet and dry cycling

Accelerated corrosion tests showed that the addition of GO significantly prolonged the corrosion induction period of steel reinforcement. The cracking time in the GO-50 group specimens was delayed by approximately 2.3 times compared to the control group. Half-cell potential monitoring revealed that the corrosion potential of the steel reinforcement in GO-modified concrete remained consistently at a positive level, indicating a more stable passivation film.

3.4.Economic and Environmental Benefits Analysis

Although the addition of GO increases material costs, its extremely low dosage (0.050% produces a significant strengthening effect) and ability to partially replace cement and improve concrete durability extend the service life of structures and reduce maintenance costs. The synergistic use of shell powder further reduces cement usage, aligning with resource conservation and low-carbon development principles. Preliminary life-cycle assessments indicate that GO-shell powder composite modified concrete offers superior economic and environmental benefits over a 30-year service life.

Conclusion

This study systematically investigated the effects of graphene oxide (GO) combined with shell powder on the workability, mechanical properties, and durability of self-compacting concrete. The main conclusions are as follows: 1. The incorporation of GO significantly improves the compactness and early strength development of concrete. When the dosage is 0.050%, the 28-day compressive strength and splitting tensile strength of concrete increase by 18.6% and 22.4%, respectively, while still meeting the self-compacting requirements . 2. GO effectively inhibits the initiation and propagation of microcracks through filling, nucleation, and bridging effects, thereby improving the concrete's impermeability, resistance to chloride ion attack, and resistance to wet-dry cycles . 3. The synergistic effect of GO and shell powder not only enhances the mechanical and durability properties of concrete but also reduces cement usage, demonstrating good resource conservation and environmental friendliness . Considering both performance improvement and economic feasibility, a GO dosage of 0.050% is the optimal choice, suitable for marine environments, saline soil areas, and critical infrastructure projects where high durability is required.

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