Effect of graphene oxide on strength and interfacial transition zone of recycled aggregate concrete

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Abstract
This paper studies the effect of graphene oxide (GO) on the strength and interface transition zone of recycled aggregate concrete (RAC). The results show that the addition of GO enhances the RAC strength, and the compressive strength of the sample containing GO is improved by 7% ~ 20.6% at 28 days, compared with the reference group. Meanwhile, with the addition of GO, the total porosity and the number of harmful pores (> 100 nm) of RAC samples decreased by 8.1% ~ 35.7% and 3% ~ 39.1%, respectively. It is observed from the nano scale characteristics that the addition of GO can significantly reduce pore phase and unhydrated phase content in the matrix, and increase the volume fraction of C–S–H phase, especially the high-density C–S–H phase. In addition, the width of the interface transition zone between old mortar and new mortar containing GO sample is relatively reduced by 25%, but there is no obvious change in the interface transition zone of old aggregate mortar. The strengthening effect of GO on RAC strength is due to the nucleation of GO and the filling effect of micro-aggregate, improving the pore structure and interface transition zone of RAC.

1. Introduction

With the acceleration of economic development and urbanization, the consumption of concrete materials is increasing day by day, and the demand for natural resources such as aggregate, sand and stone is also increasing. At the same time, the exploitation and consumption of natural resources for a long time has led to the depletion of resources and a shortage of natural aggregate [1, 2]. Since 2012, more than three billions tons of construction waste have been generated worldwide, and it shows an increase trend [3]. In order to alleviate environmental pollution and resource shortage, some scholars proposed breaking the waste concrete into recycled aggregate to replace part or all of the natural aggregate to prepare recycled concrete. Unfortunately, RAC has many defects such as complex interface transition zone (ITZ) [4], high porosity and water absorption [5], which lead to unsatisfactory mechanical properties of RAC and seriously restrict its application in engineering buildings. Otsuki et al [6] found that the porosity between old mortar and aggregate was large by scanning electron microscope (SEM), and an ITZ was formed between them. Although the volume proportion of ITZs is small, it is a porous area with a high water-cement ratio. It is a typical weak area of concrete and has a great impact on the mechanical properties of RAC. Poon et al [7] found through SEM that the old ITZ of RAC was mainly composed of some loose hydration products; It is generally accepted that the ITZs, the quality of the old mortar, and the old mortar content of the original concrete influence the properties of RAC [8]. Therefore, attempts to effectively improve the ITZ and microstructure of RC and significantly improve the mechanical properties of RAC have become a global concern.

Recently, with the rapid nanotechnology development, carbon-based nanomaterials have currently become a global concern. Recently, with the rapid nanotechnology development, carbon-based nanomaterials have currently become a global concern. Recently, with the rapid nanotechnology development, carbon-based nanomaterials have currently become a global concern. Recently, with the rapid nanotechnology development, carbon-based nanomaterials have currently become a global concern.
nanomaterials, either in individual or agglomerate forms, provide nucleation sites for cement hydrates deposition and growth [11]. GO, as the single or multilayer of graphene, possess abundant oxygen containing functional groups such as hydroxyl, carboxyl, carbonyl, epoxy etc attaching on hybridized sp2/sp3 aromatic structure through chemical exfoliation. GO owns high aspect ratio ranging from 1500 to 45000, together with large surface area ranging from 700 to 1500 m² g⁻¹ and average thickness of about 0.67 nm [3]. The study showed that the compressive strength and flexural strength of cement mortar increased by 33% and 58% respectively with the addition of 0.05% GO [12]. Mohammed et al concluded that the durability of cement paste was improved in terms of chloride ion penetration and water absorptivity, which may be attribute to the nano-filler effect of GO and further reduces the porosity in GO reinforced-cement matrices, implying higher resistivity of cement matrices to chemical ingress [13]. Previous studies have shown that the incorporation of GO into cement-based composites accelerates the cement hydration. The oxygenated functionalities attached on GO nano-sheets makes GO more comprehensible to the cement particles, consequently allowing the GO-cement-based composites accelerates the cement hydration. The oxygenated functionalities attached on GO nanosheets to boost the reaction of cement with water by acting as nuclei for the cement phases [14]. Other scholars [15–17] reported that GO could strengthen and toughen the cement slurry by regulating cement hydration products to form regular flower or polyhedron. In addition, GO at the interface and its bonding with cement paste could densify the microstructure, reduce the thickness of the interfacial transition zone (ITZ), improve the toughness of the ITZ, and enhance the interfacial strength [18]. As a new mineral admixture, GO is added to RAC. Through the microstructure regulation of GO, the relevant properties of recycled concrete can be improved, which further widens the research, production, and application field of GO.

At present, there is still a lack of applied studies on the effect of GO on different ITZs in RAC. Therefore, the effect of GO on the RAC strength and ITZs were studied in this paper. The influence of GO on the hydration characteristics of slurry and the microstructure of RAC, especially the ITZ was analyzed by ¹H low-field nuclear magnetic resonance (LF-NMR) technology, mercury injection technology (MIP) and nano indentation technology. It plays an important role in revealing the enhancement mechanism of GO in RAC.

2. Experimental

2.1. Materials
The physical properties of cement and GO are shown in table 1. The cement used is P·O 42.5 ordinary Portland cement, and its chemical composition is shown in table 1. The transmission electron microscopic (TEM) image (figure 1) that GO sheets are highly transparent and tend to curl at the edges. The fine aggregate was river sand with fineness modulus of 2.45. Recycle coarse aggregate (RCA) is the crushed aggregate of waste building concrete, with crushing index of 14.21 and particle size of 5 ~ 25 mm. A polycarboxylate-based superplasticizer (PS) was adopted to improve the flowability of fresh mixtures. The mixing water is tap water.

2.2. Mix design and synthesis
In order to study the effect of GO on RAC strength and ITZs, the selected four designs of RAC in this study are shown in table 3. All the pastes were formulated by mixing at the constant water / binder (w/b) ratio of 0.55 under the fixed procedure. The mass content of GO is 0%, 0.01%, 0.03% and 0.05% of cement, which are respectively labeled as C0, GO1, GO3 and GO5. Briefly, GO and PS were added into mixing water under ultrasonic dispersion for 2h, then GO dispersion was mixed with cement to form cement slurry. Then the cement slurry and RCA were mixed and stirred for 10 min, and fine aggregate was added. After stirring, the concrete was injected into the 150 mm × 150 mm × 150 mm cubic test molds. The sealed specimens were demoulded after 24 h curing at room temperature, and aged at a standard environment (T = 20 ± 2 °C, relative humidity >90%) until 7 days and 28 days for compressive strength test.

| Sample | CaO | SiO₂ | Al₂O₃ | Fe₂O₃ | MgO | SO₃ | Na₂O | MnO | K₂O | P₂O₅ |
|--------|-----|------|-------|-------|-----|-----|------|-----|-----|------|
| Cement | 57.8| 20.2 | 9.2   | 8.35  | 1.89| 2.79| 0.13 | /   | 0.82| /    |

| Purity | Thickness | Flake diameter | Carbon content | Oxygen content | Dispersant |
|--------|------------|----------------|---------------|---------------|-------------|
| 98%    | ~1 nm      | 0.3–12 μm      | 46%           | 53%           | Water       |
2.3. Test methods

The setting time of cement paste was measured in accordance with the standard GB/T 1346–2001. The hydration process was studied by 1H low field nuclear magnetic resonance (LF-NMR). The ingredients were mixed with water in each group, and then were put into the cylindrical mold with size of Φ 5 mm × 20 mm. A LF-NMR spectrometer (micro-mr20, niuma/Co. Ltd, China) was used, the fixed magnetic field was 0.28 T, the 25 mm magnet coil worked at 11.9 MHz and the temperature was maintained at 30 ± 0.01 °C. The compressive strength test was carried out according to the standard GB/T 50081–2019. Quantachrome Poremaster GT-60 instrument mercury injection method (MIP) was used to test the pore structure of RC sample at 28d. The tested sample was obtained from the interior of RAC. The fragments with a diameter of about 5 mm were soaked in ethanol to stop further hydration. The MIP test was carried out after drying in an oven at 60 °C for 48 h. The sample preparation process of nano indentation is detailed in the published literature [19]. The matrix area and ITZs area including aggregate old mortar (ITZ-I) and old mortar new mortar (ITZ-II) are selected in the internal area of RAC, as shown in figure 2. The indentation spacing of matrix and ITZs area is 5 μm in both transverse and vertical directions. The ITZs area size is 10 μm × 25 μm, respectively. For each interface transition region, the randomly selected regions were tested by nano indentation, and the distribution of elastic modulus was obtained. Calculation formula of elastic modulus $E$ of nano indentation is referred to other reports [20, 21].

3. Results and discussion

3.1. Setting time and hydration process

The setting time change of cement paste with GO added is shown in the figure 3. It can be seen that the addition of GO significantly shortens the setting time of cement slurry, and this phenomenon becomes more obvious with the increase of GO content. Compared with the reference group, the initial setting time of sample containing GO is shortened by 22% ~38%. This is consistent with previous research results [22], in which the rapid physical adsorption and chemical interaction between the GO and cement at the beginning of mixing have a significant shortening on the setting time of cement paste at an early age.
The hydration process of concrete paste is monitored by LF-NMR technology. With the hydration progress, the physical bound water in the paste is gradually consumed and transformed into chemical bound water. Its signal cannot be detected, but the cement hydration process can be evaluated by the change of overall signal strength. As shown in figure 4, the change trend of total signal strength in all slurries is very similar, and a critical value exists. Beyond this critical value, the overall signal strength decreases rapidly. Therefore, this critical point can be regarded as the transition point from the dormant period to the accelerated period of hydration. Compared with C0, the signal intensity of the slurry decreases with the increase of GO content, and the critical point also presents a trend of advance. This phenomenon further confirms the promotion effect of GO on cement hydration. The same conclusion is obtained in the previous literature that GO surface contains a large number of active oxygen-containing groups (–OH, –COOH, –O–) to provide nucleation sites for cement hydration [23, 24]. GO acts as a nucleating matrix for C–S–H, which preferentially forms on the GO rather than solely on the surface of the adjacent cement grains [25]. The nucleation behavior of hydrates on the nanoparticles accelerates the hydration of cement [23]. Lu et al [26] confirmed that GO sheets promoted cement hydration since the water molecules on the GO surface provided a reservoir of water and transport channels for further hydration.

3.2. Compressive strength

The effect of GO content on the compressive strength of RAC is shown in figure 5. It can be concluded that the incorporation of GO improves the compressive strength of RAC. Compared with C0, the compressive strength of GO1, GO3 and GO5 samples at 7d is increased by 10.6%, 22.6% and 24.4%, respectively. The strengthening effect of GO on RAC decreases with the increase of curing age, and the 28-days compressive strength of samples containing GO increases by 7%, 18% and 20.6% respectively, similar to the previous reports [27]. This enhancement is mainly attributed to the nanoscale size of GO, which fills the gap between cement particles, thereby reducing the harmful porosity in the matrix [28]. It is confirmed by MIP test. Meanwhile, the nucleation
of GO transforms rod-shaped hydrated crystals into columnar and fully open petal-like hydrated crystals, making the microstructure denser and delaying the further expansion of cracks [29].

3.3. Pore structure
Figure 6 shows the pore structure evolution of RAC sample for 28d with different GO dosage. In figure 6(a), the pores of RACs are mainly between 20 nm and 200 nm. With the addition of GO, the peak position of pore size distribution curve gradually moved to the left gradually, suggesting that the presence of GO leads to the decrease of pore size in RAC matrix. According to the study of Zhan et al [30], the pores are classified according to the pore size, as shown in figure 6(b). It can be found that the total porosity of RAC matrix shows a decreasing trend with an increase of GO content. Compared with C0, the total porosity of GO1, GO3 and GO5 samples decreases by 8.1%, 18% and 35.7% respectively. More notably, the number of harmful pores (>100 nm) in the samples with GO respectively decreases by 3%, 34.9 and 39.1% compared with C0 (figure 6(c)). It is well known that pore sizes greater than 100 nm have a negative effect on concrete strength [31]. At the same time, the number of harmless holes and less harmful holes in samples containing GO is significantly greater than that in the reference group. This shows that the pore structure of RAC is modified by the incorporation of GO, which further explains why the RAC sample containing GO has better mechanical properties. As previous literature reported [32–34], one factor leading to the refinement of the pore structure comes from the pore-filling effect of the nano-sized
GO sheets. The other relates to the seeding effects of GO that facilitate more hydration products to fill pore space.

### 3.4. Nanoindentation analysis

The type and content of cement hydration products in the matrix have a significant impact on the RAC macro performance. The nano indentation elastic modulus \( E \) of C0 and GO5 matrix was measured by nano indentation test, as shown in figure 7. According to the different elastic modulus, the volume fraction of each phase in the matrix can be obtained: pore phase (\( E < 12 \) GPa); Low density C–S–H gel (LD C–S–H, \( 12 \leq E < 22 \) GPa); High density C–S–H gel (HD C–S–H, \( 22 \leq E < 32 \) GPa); CH (32 \( \leq E < 40 \) GPa); Unhydrated particles (\( \geq 40 \) GPa) [30, 31]. The volume fraction of each phase in the matrix area is shown in table 4. Compared with C0, the contents of pore phase and CH phase in GO5 matrix are significantly reduced, and the content of unhydrated phase is also reduced. At the same time, the content of C–S–H in GO5 matrix is increased by 29.6%, and that of HD C–S–H is increased by 30%. It can be concluded that the addition of GO can increase the total content of C–S–H in RAC matrix, especially HD C–S–H. This is mainly due to the nucleation effect of GO.

Figure 8 shows the contour map of elastic modulus distribution in ITZ-I of RAC sample at 28d. The dark blue area in the figure has low elastic modulus, representing a relatively weak area in RAC. The red and yellow areas represent aggregates. Green and cyan areas represent unhydrated cement particles and CH crystals. From the color change trend in the figure, the dark blue area between aggregate and old mortar is regarded as ITZ-I of RAC. The ITZ-I width distribution of RAC sample is shown in figure 9. The elastic modulus of ITZ-I and old mortar shows increase or decrease non-uniformly, which may be due to the non-uniformity of RAC and the concentration of pores and CH crystals in ITZs. The thickness of ITZ-1 in C0 and GO5 samples is about 40 \( \mu \text{m} \), which is similar to the results of previous study [31]. This also implies an insignificant effect of GO incorporation on RAC sample ITZ-I.
As shown in figure 10, the uneven distribution of elastic modulus in the ITZ-II and old mortar zones may be related to the existence of some pores and CH crystals in the old mortar and ITZ zones. In terms of width (figure 11), the thickness of ITZ-II is in the range of 45 to 60 μm, 5 to 20 μm wider than the that of ITZ-I. Compared with C0 sample, the width of ITZ-II in RAC decreases by 25% after GO adding, and the elastic modulus of ITZ-II and new mortar are increased. This may be due to the covalent bond formed between the oxygen-containing functional groups on the surface of GO and the cement hydration product C–S–H, which improves ITZs [35]. In addition, Ca$^{2+}$ and Al$^{3+}$ near the C–S–H surface usually bridge oxygen atoms in the silicate chain and the hydroxyl group in GO, enhancing the covalent bond between the cement matrix [27] and playing a bridging effect in the nanoscale cracks in the matrix [36].
Figure 9. The widths of ITZ-I of sample (a) C0, (b) GO5.

Figure 10. Contour map of elastic modulus distribution in ITZ-II of sample (a) C0, (b) GO5.

Figure 11. The widths of ITZ-II of sample (a) C0, (b) GO5.
4. Conclusions

The following conclusions can be obtained according to the above research in this paper.

(1) The addition of GO reduced the setting time of cement slurry. With the increase of GO content, the LF-NMR total signal intensity of slurry decreased, and the critical point showed a trend of advance. Compared with C0, the 7 days compressive strength of GO1, GO3 and GO5 specimens increased by 10.6%, 22.6% and 24.4%, respectively. The enhancement effect of GO on RAC decreased slightly with the increase of curing age.

(2) The pore structure of RAC samples at 28 days was refined by the nucleation effect of GO and the filling of microaggregates. Compared with C0, the total porosity and the content of harmful pores (> 100 nm) of the samples containing GO decreased by 8.1% ~35.7% and 3% ~39.1%, respectively.

(3) From the nanoscale characteristics, it can be found that, compared with the reference group, the content of pore phase and CH phase in the matrix with GO sample was significantly reduced, the content of C–S–H was increased by 29.6%, and the content of HD C–S–H was especially increased by 30%. In terms of ITZ, the ITZ-I interfacial width of recycled concrete is about 40 μm, and the effect of GO addition on ITZ-I of RAC sample is not significant. In addition, the addition of GO reduced the ITZ-II width of the RAC sample by 25% compared with the reference group, and the elastic modulus of the ITZ-II and the new mortar were significantly enhanced.

(4) The mechanical properties enhancement mechanism of GO on RAC are mainly concluded: promotion of cement hydration, refinement of pore structure, enhancement of microstructure density, and the improvement of ITZ in RAC.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Author contributions

Conceptualization, Investigation (Qidong Wang); Data curation, Writing—original draft and Funding acquisition (Changshun Zhou); Visualization, Writing—review and editing (Xudong Wang); Supervision and Resources (Zixuan An); Methodology and Project administration (Yeke Luo). All authors have read and agreed to the published version of the manuscript.

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Conflicts of interest

The authors declare that they have no competing financial interest conflicts in this paper.

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