

Research Article

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Research on modification of mechanical properties of recycled aggregate concrete by replacing sand with graphite tailings

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Abstract: How to more effectively and comprehensively utilize recycled aggregate resources to prepare a richer new low-carbon concrete material system is the key path to achieve low-carbon development in China's construction field. Based on the material properties of graphite tailings (GT), this article explores the organic combination of GT to replace sand and recycled aggregates (RA) to replace natural stones, in order to propose an efficient recycling path for multiple solid wastes and a new and cost-effective recycled aggregate concrete (RAC). This article focuses on investigating the influences of GT (0–40%) and RA (0–40%) on the workability, water absorption, and mechanical properties of GT reinforced recycled aggregate concrete (GTRAC). Simultaneously, the hydration products, microstructure morphology, key chemical bonds, and pore structure characteristics were analyzed by a combination of microscopic tests such as X-ray diffraction, scanning electron microscope, Fourier transform infrared and mercury intrusion porosimetry to propose the key factors affecting the macroscopic performance of GTRAC. The results show that incorporating 10–30% GT has significant positive effects on water absorption, surface water content, compressive strength, and elastic modulus of RAC. RAC's micro-crack density, hydration, and pore structure distribution can be maximally improved by appropriate GT (20%). However, the high content of GT will lead to the degradation of its mechanical properties. Therefore, 20% GT and 30% recycled aggregate are the optimum combinations of the

concrete material system. However, it might be changed when the ratio of water to binder varies. Finally, this article also comprehensively analyzes and evaluates the performance and price of different types of RACs, which provides theoretical support for evaluating and predicting the practical engineering application value of GTRAC. In summary, the research results in this study provide theoretical guidance for understanding the mechanism of GT on the enhancement of mechanical properties of RAC and exploring new ways of practical engineering application of it.

Keywords: graphite tailings, recycled aggregate, mechanical properties, material properties

1 Introduction

With the development of industrialization and construction industry, the demand for concrete is extremely large. At present, the mining and consumption of natural aggregate materials are relatively large, intensifying the shortage of natural aggregate resources and the difficulty of sustainable development. According to the current estimate, the global demand for construction aggregates is more than 20 billion tons per annum [1]. At the same time, with the rapid development of urbanization in China, 1.5 billion tons of construction and demolition waste are generated every year [2], and the primary disposal method is landfill [3]. Academics believes that an effective way to address the shortage of natural aggregates and waste concrete stockpiles is to recycle concrete demolition waste as recycled aggregate (RA), partially or fully replacing natural aggregates for recycled aggregate concrete (RAC). It has also proven to provide significant environmental and economic benefits [4]. Although the reuse of RA to manufacture RAC has aroused more and more research interest in academia and has been extensively studied, due to the poor quality of RA compared with natural aggregates, the widespread use of RAC in construction projects is limited as before.

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Compared to conventional natural aggregate concrete, RAC has been extensively researched for many properties, including compressive strength [5] and modulus of elasticity [6]. The replacement ratio between the weight of RA and the total weight of coarse aggregate in a concrete mix deems a vital influence on RAC properties [7]. In general, concrete made from RA configurations has a lower compressive strength and modulus of elasticity than conventional concrete. Compared to conventional concrete, the presence of new and old mortar causes the complexity of the interface transition zone (ITZ) of RAC [8], and its strength and compactness become one of the most critical factors affecting the structural and mechanical properties of concrete. Based on the nano-indentation technique, Xiao *et al.* [9] and Lee and Choi [10] found that the average indentation modulus of old ITZ was 70–80% of that of old paste matrix, while the average indentation modulus of new ITZ was 80–90% of that of new paste matrix. The more numerous and weaker ITZs lead to the poor performance of the RAC. Meanwhile, Xiao *et al.* [9] also found obvious voids and $\text{Ca}(\text{OH})_2$ (CH) exist in both old and new ITZs in RAC. It is found that the existence of coarse CH crystals has a certain negative effect on the strength of the ITZ. On the one hand, because of its layered overlapping arrangement, its specific surface area is reduced, and thus, the cohesive force is also reduced accordingly; on the other hand, the directional arrangement of CH makes it more prone to cracking [11]. Thus, researchers are studying how to improve the compressive strength of RAC by incorporating other materials. Existing studies have found that the addition of fly ash and silica fume can mitigate the adverse effects of RAs on the compressive strength of concrete. Kou *et al.* [12] found that 25% fly ash incorporation could maximize the compressive strength of recycled concrete by 17.43%. Abid *et al.* [13] have demonstrated that the use of silica fume can increase the compressive strength of RAC from 22.5 to 28 MPa, an increase of 24.4%. The spherical morphology of fly ash limits the amount of substitution, and silica fume's weak volcanic ash activity limits its early strength. Meanwhile, nano-silica [14] and iron tailing sand [15] have been incorporated, but the cost of nano-silica and the low strength of iron tailing sand recycled concrete limit their widespread use.

At the same time, in 2020, China's development strategy is of achieving “carbon peaking” by 2030 and “carbon neutral” by 2060. It puts forward higher and updated standards and requirements for the green treatment and comprehensive utilization of tailings solid waste in mining enterprises. With the rapid growth of graphite resource consumption, the basic reserves of graphite

deposits in China have accounted for 43% of the world's total. Graphite tailings (GT) is solid wastes produced in large quantities in the process of graphite mining [16], but it is also a resource to be developed. Statistics in recent years have found that the global graphite mine production was approximately 0.21 billion tons [16]. However, mining a 1-ton graphite mine produces 10–15 tons of GT. The large-scale storage of GT takes up a lot of land, pollutes the surrounding environment, and brings serious safety hazards [17]. Furthermore, the scarcity of river sand in the construction industry has become increasingly pronounced in recent years due to the prohibition of river sand exploitation for natural resource-saving and environmental protection [18]. Because the composition of GT is similar to that of river sand, it is possible to use GT as cement materials instead of river sand. The research and development of GT can reduce environmental pollution and effectively recycle resources. At the same time, it can also solve the shortage of building materials and bring significant economic benefits. Liu *et al.* [19] and Kathirvel *et al.* [20] found that GT concrete has good advantages in strength improvement, price, recovery process, etc. They used GT instead of river sand to prepare a new type of concrete and analyzed the material's mechanical properties, durability, and electrical conductivity. The results show that the maximum compressive strength of concrete is 10% GT instead of sand, and the strength improved by 14.9%. Kathirvel *et al.* [21] and Peng *et al.* [22] studied the mechanical characteristics and electrochemical properties of GT-reinforced concrete and mortar with different incorporation amounts. The application of GT shows the feasibility of reusing waste as construction materials, and it also provides a reference to the possibilities for mixing GT into (RA) concrete.

However, research on the modification of RAC by tailings, particularly GT replacement sand, is at a nascent stage. At present, it is necessary to conduct systematic and in-depth research on its material system to support the actual effects of future engineering applications and the realization of double carbon emission reduction. This article uses GT with different dosages to replace sand, and RA was used to replace natural stone (single and mixed method) to prepare graphite tailings recycled coarse aggregate concrete (GTRAC). The compressive strength and elasticity modulus of GTRAC is investigated. The modification mechanism of GT on the mechanical strength of RAC was explained preliminarily through material science experiments such as mesoscopic morphology, compound component, and pore structure distribution. Finally, the optimal substitution rate of GT and RA in concrete was determined to improve the utilization rate of GT and RA and realize the utilization of solid waste.

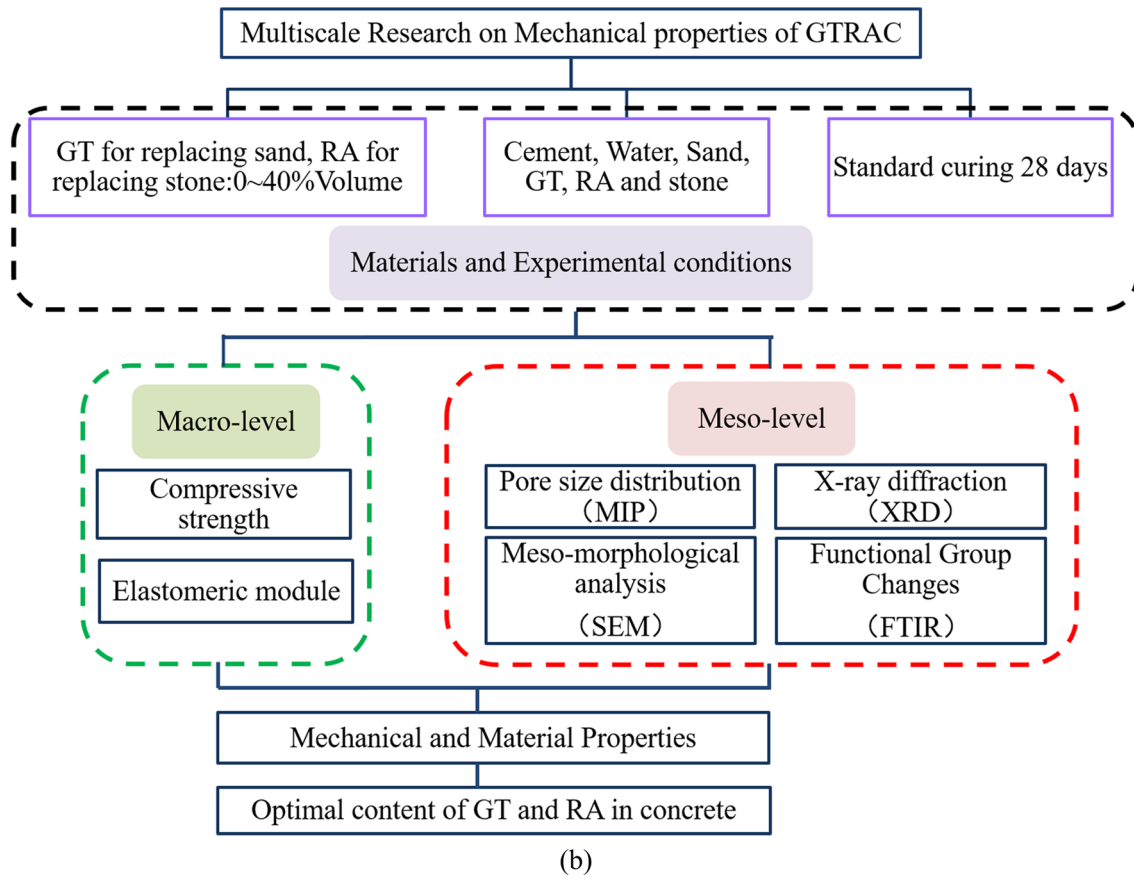
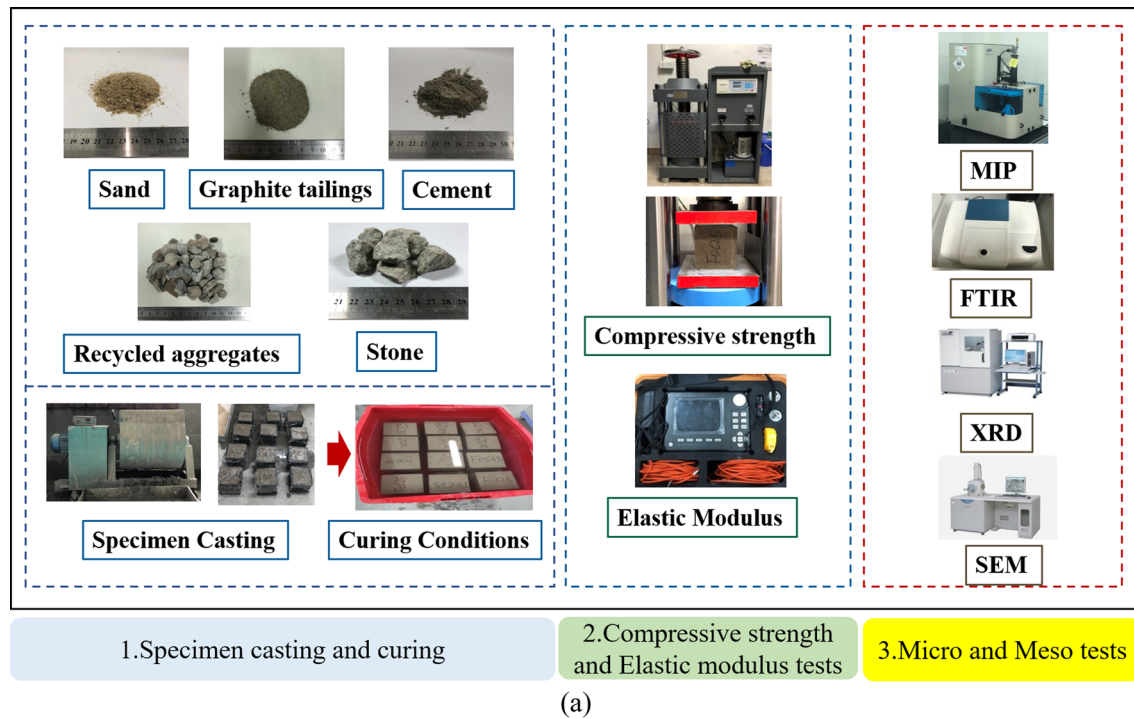


Figure 1: (a) Experimental flowchart; (b) research flowchart.

Table 3: Physical properties of fine aggregate

Fine aggregate	Type	Apparent density ($\text{kg}\cdot\text{m}^{-3}$)	Bulk density ($\text{kg}\cdot\text{m}^{-3}$)	Fineness modulus	24 h water absorption (%)
GT	Fine sand	2,855	1,540	0.90	30.1
Sand	Medium sand	2,460	1,630	2.65	21.9

**Figure 3:** The apparent comparison of natural stone and RA.

150 mm × 150 mm × 150 mm (75 pieces) and 150 mm × 150 mm × 300 mm (75 pieces) were used to determine the water absorption characteristics, compressive strength, and elastic modulus performance at 28-day curing time, respectively. All experimental specimens were placed in the curing room with a temperature of $20 \pm 2^\circ\text{C}$ and relative humidity of 95% for curing for 28 days.

2.2 Experimental methods

After all the test pieces reach the 28-day curing age, the excess water on the surface of the test pieces was wiped off. It was placed at room temperature for 24 h, and the mechanical properties were tested after waiting for it to dry naturally.

2.2.1 Macro performance testing

2.2.1.1 Workability properties test

The slump bucket is used to measure the slump height of fresh concrete under specified conditions, and the workability

is measured by slump according to the Chinese standard GB/T50080-2016 [29].

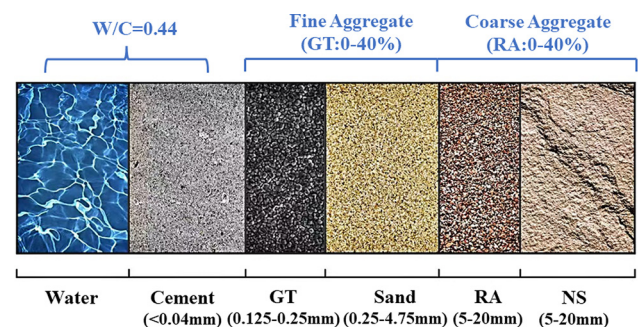
2.2.1.2 Water absorption and surface moisture content

According to the Chinese standard JGJ/T70-2009 [28], the GTRAC specimens are dried at $105 \pm 5^\circ\text{C}$ for 48 ± 0.5 h and weighed to obtain the mass m_0 . The sample is immersed completely in water for 48 ± 0.5 h and removed; the free water on the surface of the specimens is wiped off with a rag to measure the mass m_1 . The arithmetic average of the measured values of the three specimens is used as the water absorption of the concrete and is accurate to 1%. The water absorption of concrete is calculated using equation (1):

$$W_x = 100 \times (m_1 - m_0)/m_0, \quad (1)$$

where W_x is the water absorption of concrete (%), m_1 is the mass of a specimen after water absorption (g), and m_0 is the mass of a dried specimen (g).

In addition, the surface moisture content of GTRAC was measured in this article. GTRAC specimens were taken out of the curing room, and the surface was wiped dry.

**Figure 4:** The substitution methods for RA and GT.**Table 4:** Physical properties of coarse aggregate

Coarse aggregate	Size (mm)	Apparent density ($\text{kg}\cdot\text{m}^{-3}$)	24 h water absorption (%)	Crush indicators (%)
NS	5–20	2,550	1.21	14
RA	5–20	2,360	8.02	27.6

Table 5: Mix proportion design of RA concrete from graphite tailings

W/C = 0.44	Mix proportion design of concrete (kg·m ⁻³)							Replacement rate of GT (%)	Replacement rate of RA (%)
	Water	Cement	Sand	GT	NS	RA	Water reducing agent		
NC	2.19	4.97	6.51	0.00	15.18	0.00	0.0497	0	0
GT1RA0	2.19	4.97	5.86	0.65	15.18	0.00	0.0497	10	0
GT2RA0	2.19	4.97	5.20	1.30	15.18	0.00	0.0497	20	0
GT3RA0	2.19	4.97	4.55	1.95	15.18	0.00	0.0497	30	0
GT4RA0	2.19	4.97	3.90	2.32	15.18	0.00	0.0497	40	0
GT0RA1	2.19	4.97	6.51	0.00	13.66	1.52	0.0497	0	10
GT1RA1	2.19	4.97	5.86	0.65	13.66	1.52	0.0497	10	10
GT2RA1	2.19	4.97	5.20	1.30	13.66	1.52	0.0497	20	10
GT3RA1	2.19	4.97	4.55	1.95	13.66	1.52	0.0497	30	10
GT4RA1	2.19	4.97	3.90	2.32	13.66	1.52	0.0497	40	10
GT0RA2	2.19	4.97	6.51	0.00	12.14	3.04	0.0497	0	20
GT1RA2	2.19	4.97	5.86	0.65	12.14	3.04	0.0497	10	20
GT2RA2	2.19	4.97	5.20	1.30	12.14	3.04	0.0497	20	20
GT3RA2	2.19	4.97	4.55	1.95	12.14	3.04	0.0497	30	20
GT4RA2	2.19	4.97	3.90	2.32	12.14	3.04	0.0497	40	20
GT0RA3	2.19	4.97	6.51	0.00	10.63	4.55	0.0497	0	30
GT1RA3	2.19	4.97	5.86	0.65	10.63	4.55	0.0497	10	30
GT2RA3	2.19	4.97	5.20	1.30	10.63	4.55	0.0497	20	30
GT3RA3	2.19	4.97	4.55	1.95	10.63	4.55	0.0497	30	30
GT4RA3	2.19	4.97	3.90	2.32	10.63	4.55	0.0497	40	30
GT0RA4	2.19	4.97	6.51	0.00	9.11	6.07	0.0497	0	40
GT1RA4	2.19	4.97	5.86	0.65	9.11	6.07	0.0497	10	40
GT2RA4	2.19	4.97	5.20	1.30	9.11	6.07	0.0497	20	40
GT3RA4	2.19	4.97	4.55	1.95	9.11	6.07	0.0497	30	40
GT4RA4	2.19	4.97	3.90	2.32	9.11	6.07	0.0497	40	40

Notes: NC is the natural aggregate concrete as standard group. GT is the graphite tailings used to replace river sand and RA is used to replace natural aggregate. GT1RA1 is the graphite tailing – RA concrete in which 10% GT + 10% RA.

Subsequently, the PW-2 multifunctional surface material moisture meter is used to measure the moisture content of the three surfaces of each sample under the condition of ensuring that the test block test environment is similar, and the average value of the test is the measured value of the surface moisture content.

2.2.1.3 Mechanical properties test

This article tests and determines the compressive strength and elastic modulus according to GB/T 50081-2002 [27]. The compressive strength is tested on the RFP-03 intelligent force tester. The elastic modulus is tested on an electrohydraulic servo universal compression testing machine. The specific operation steps of the elastic modulus are as follows:

The micrometer was first fixed to the micro-deformation measuring frame at a distance of 150 mm and then placed on the testing machine for loading. At a reference stress of 0.5 MPa, the load is applied to the initial load

value F_0 held for 60 s and the deformation ε_0 is recorded. Immediately, the load is applied continuously and uniformly for 60 s at one-third of the axial compressive strength to the load value F_a , and the deformation ε_a is recorded. Finally, the load is unloaded in the same manner as the loading rate. The value of the modulus of elasticity shall be calculated using equations (2) and (3):

$$E_c = (F_a - F_0) \times L / (A \times \Delta n), \quad (2)$$

$$\Delta n = \varepsilon_a - \varepsilon_0, \quad (3)$$

where E_c is the modulus of elasticity of concrete in static compression (MPa); F_a is the load at a stress of one-third axial compressive strength (N); F_0 is the initial load at a stress of 0.5 MPa (N); A is the compressive area of the specimen (mm²); L is the measurement distance (mm); Δn is the average value of the deformation on both sides of the specimen at the last loading from F_0 to F_a (mm); ε_a is the average value of the deformation on both sides of the specimen at F_a (mm); ε_0 is the average value of the deformation on both sides of the specimen at F_0 (mm).

The values for compressive strength and modulus of elasticity are the arithmetic mean of each set of three specimens.

2.2.2 Materials characterization tests

Several material experiments are conducted to further explore the effect of the GT and RA on the mechanical performance of concrete. Samples are subjected to mercury intrusion porosimetry (MIP), scanning electron microscopy (SEM), X-ray diffraction (XRD), and Fourier-transform infrared spectroscopy (FTIR) to analyze the changes in the pore structure distribution, mesomorphology, hydration products, and chemical bonding or molecules in the concrete specimens. The preparation and drying conditions of these samples are in accordance with Chinese regulations GB/T 21650-2011 [30], GB/T 16594-2008 [31], GB/T 30904-2014 [32], and ISO 19618-2017 [33].

3 Results and discussion

3.1 Macro-properties of GTRAC

3.1.1 Water phase change and workability of GTRAC

The water absorption and surface water content of cement-based materials such as concrete or cement mortar are

closely related to hydration degree. They are one of the most significant influencing factors of concrete's mechanical and durability performance. In this study, the water absorption and surface water content of GTRAC are shown in Figure 5. Due to RA's water absorption characteristics, the water absorption rate of RAC is often larger than that of ordinary concrete. Studies have shown that RAC with a water absorption rate of less than 50% replacement rate has better performance when the water absorption rate is between 1.5 and 4.5% [34,35]. The water absorption rate of GTRAC is all within this range. GT has an inhibitory effect on the water absorption of GTRAC [20]. A certain amount of GT can reduce the water absorption of GTRAC by a maximum of 3.67%. In addition, a certain amount of GT can increase the surface water content of GTRAC (3.68% at greatest). Because GT contains higher active minerals than natural sand, it promotes the interface bonding between the aggregates in the concrete so that the old mortar attached to the surface of the RA and the mortar of the newly mixed concrete are better combined. A certain amount of GT class improves the compactness of the overall structure of concrete and reduces the water absorption rate. The internal moisture content affects the change of the surface moisture content. On the one hand, the denser the surface and the whole of the concrete (the lower the internal moisture content), the more difficult it is for water molecules to pass through the connected pores under the action of osmotic pressure and capillary surface tension. Most of the water molecules stay on and near the surface of the concrete test block causing an increase in

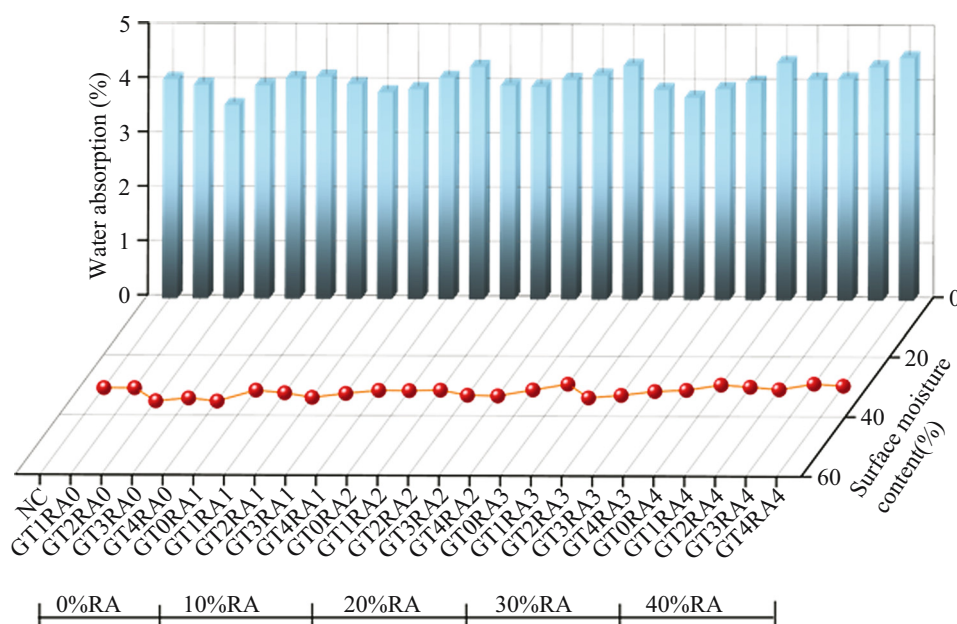


Figure 5: Surface moisture content and water absorption of GTRAC (GT1RA1 is the graphite tailing – RA concrete in which 10% GT + 10% RA).

surface water content. In addition, the more fully hydrated the concrete, the more the hydration products (decrease in internal moisture content), the increase in internal water demand, which promotes the diffusion of water molecules to the surface of the concrete through connected pores, increasing surface water content.

However, GT's changes in GTRAC's compactness have certain limitations, whether based on filling efficiency or promoting the hydration process. Due to the property limitation of GT's own active tailings material, its high-water absorption determines the increase in internal water content and the decrease in surface water content (30–40% replacement rate) after excessive mixing. At the same time, under the dual influence of GT and RA on the water phase, the workability performance of GTRAC also shows a specific correlation change law as shown in Figure 6. The results show that, because of the small particle size and large specific surface area of GT, it needs to absorb a large amount of water to wet the surface. Therefore, the initial slump of fresh concrete mixed with GT and RA gradually decreases.

3.1.2 Compressive strength performance of GTRAC

The mechanical properties of concrete are related to cement strength, water–cement ratio, and other factors. In order to investigate the effect of different admixtures of GT and RA on the mechanical properties of concrete, the method of varying the amount of water-reducing agent added was used to make fresh concrete achieving the same slump level (about 150 mm). The effect of GT and RA on concrete's compressive strength is illustrated in

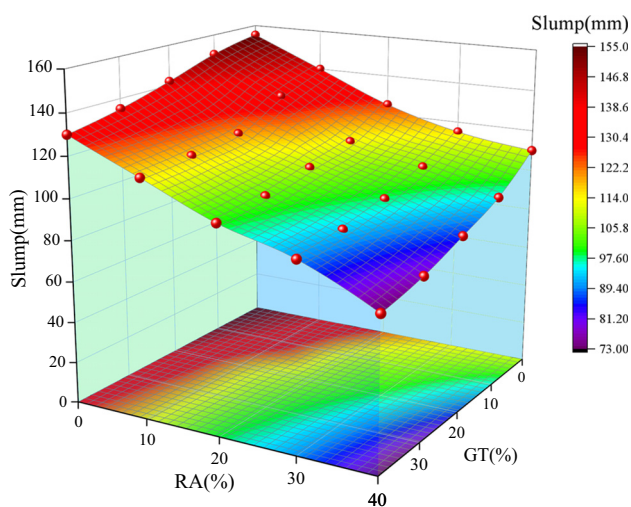


Figure 6: Initial slump of GTRAC.

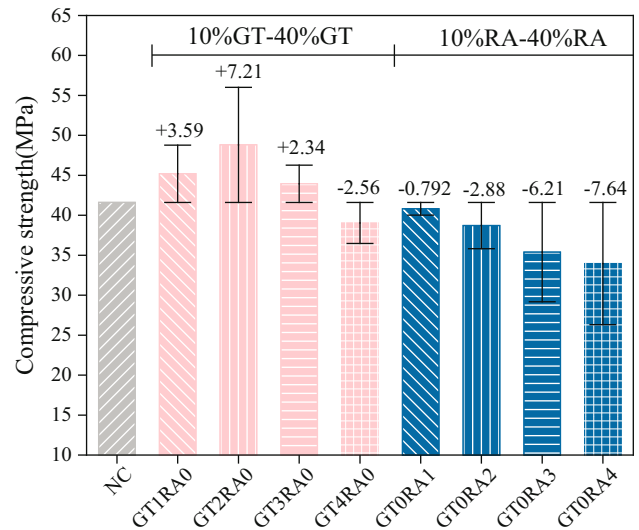


Figure 7: Compressive strength of concrete from GT and RA.

Figures 7–9. As shown in the figures, for the concrete material system mixed with GT or RA alone, the compressive strength first increases and decreases with the rise of the amount of GT and decreases significantly with the increase of RA. GT alone can increase the compressive strength by up to 17.45%, due to the fine grain size of the GT improving the internal structure and thus the compressive strength, which is consistent with the conclusions obtained by Wang et al. [16] and Lee et al. [20]. At the same time, the presence of initial cracks in the RA [9] reduces the compressive strength by up to 17.17%. It is worth mentioning that, under the influence of GT and RA, the compressive strength properties of GTRAC have been improved to a certain extent. When the same amount of RA is incorporated, the compressive strength improves

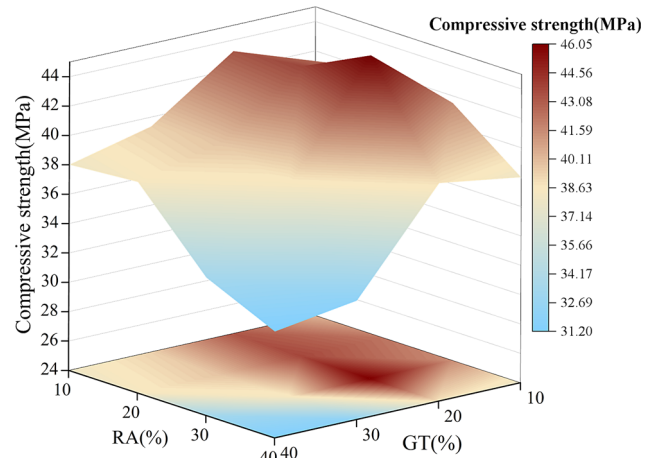


Figure 8: Compressive strength of GTRAC.

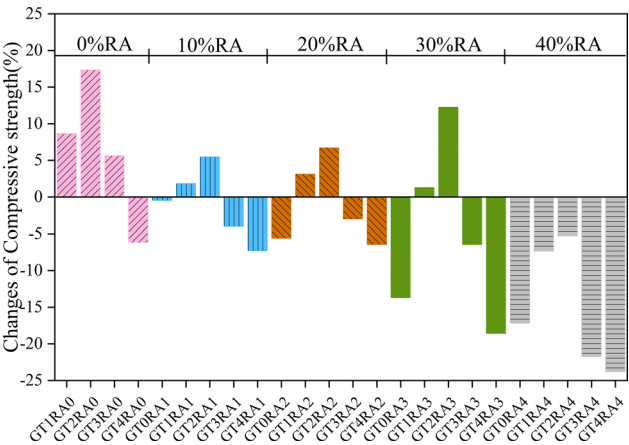


Figure 9: Changes in compressive strength of GTRAC.

as the GT replacement rate increases (not more than 30%) significantly. When the GT content is 20% and the RA content is 30%, the compressive strength of GTRAC is at the maximum rate of improvement of 12.24%. However, the significant factor that changes the compressive strength of GTRAC continues to be the rate of RA substitution. When the RA replacement rate is 40%, GT does not have any gain

effect on improving GTRAC compressive strength performance and even has a negative effect.

This is due to the characteristics of the properties of GT and RA raw materials leading to different effects on changes in compressive strength. When the replacement rate of GT and RA does not exceed 30%, due to the small aggregate fineness of GT, the concrete can be evenly wrapped during the concrete forming process, which has the effect of filling and repairing micro-cracks or micro-defects [17], thereby improving GTRAC Strength of. However, due to the influence of water absorption, GT and RA’s replacement rate exceeds 30%, and GT and RA jointly absorb the free water that participates in the curing reaction of cement-based materials, resulting in a decrease in the compressive strength of GTRAC.

In addition, the above-mentioned related conclusions can also be obtained from the elevation view of the compression failure form of the experiment (shown in Figure 10); 20% GT is introduced to improve the stability of aggregates and the bonding ability between aggregates in GTRAC. However, as the replacement rate of GT and RA continued to increase, the binding capacity between aggregate aggregates decreased significantly,

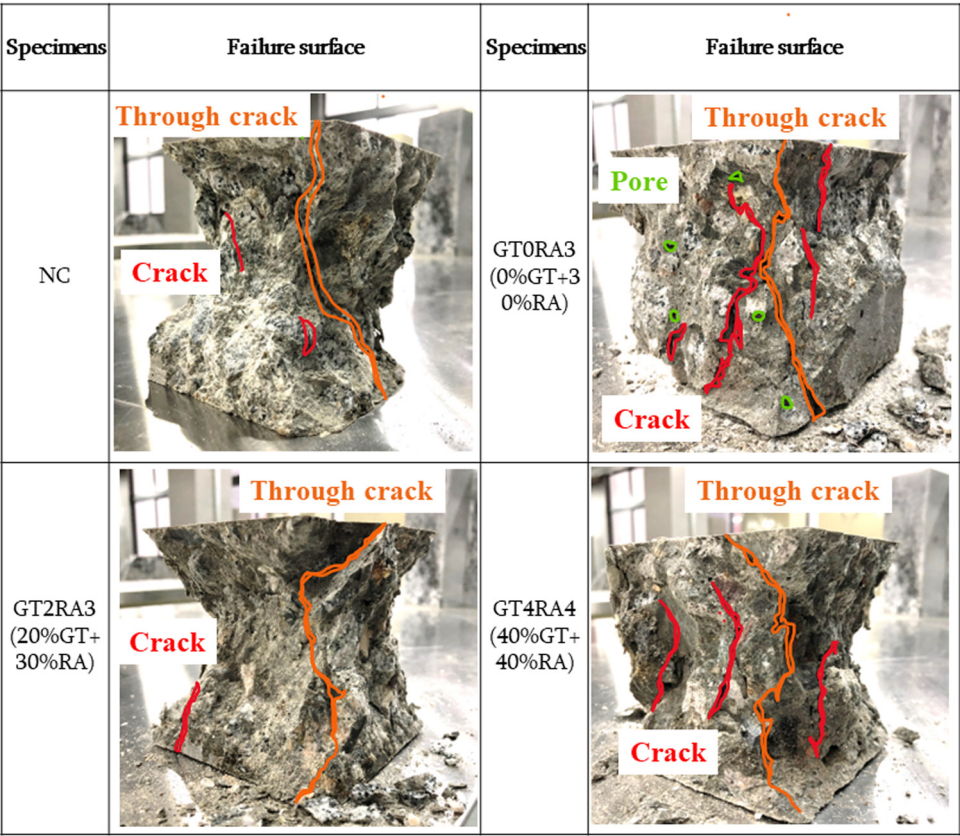


Figure 10: Failure surface of GTRAC.

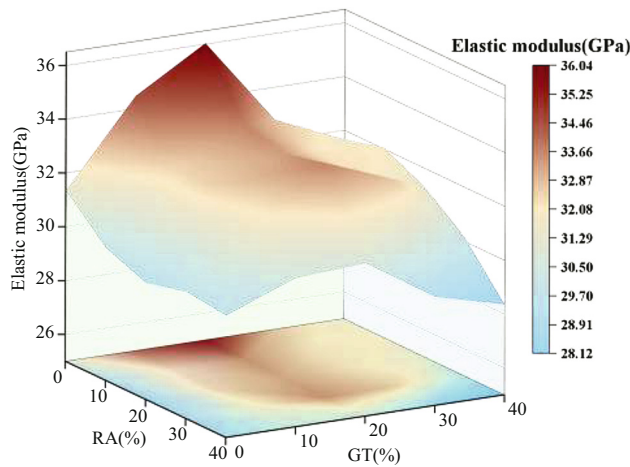


Figure 11: Elastic modulus of GTRAC.

and a more significant proportion of side peeling occurred. It is because RA produced more internal micro-cracks during the crushing process of abandoned buildings [5,7]. In the loading process, the development speed of internal cracks in RA is more significant than that between cement-based interfaces. The fragmentation of concrete is the most unfavorable mode caused by aggregate damage.

3.1.3 Elastic modulus changes of GTRAC

The elasticity modulus of GTRAC is shown in Figures 11 and 12. The modulus of elasticity follows approximately the same trend as the compressive strength. Several micro-cracks on the cement paste and aggregate surface are observed due to the adherence of the old mortar on the RA surface, resulting in a weakening of the transition zone at the old interface and thus a reduction in the

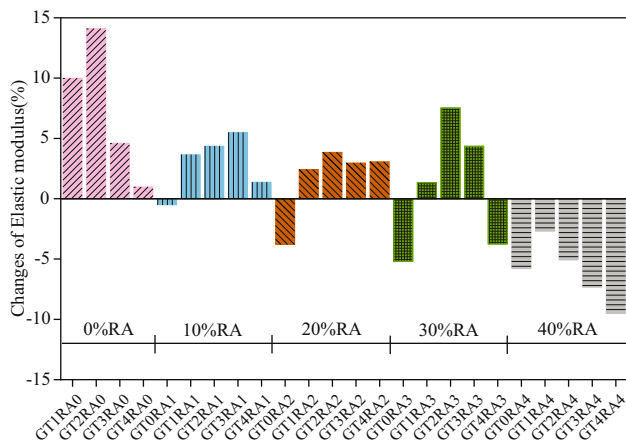


Figure 12: Changes of elastic modulus of GTRAC.

modulus of elasticity [6]. However, GT has a promoting effect on ensuring the elastic modulus of GTRAC. When the replacement rates of GT and RA are 20 and 30%, respectively (under the dual doping system), GTRAC has the best elastic modulus (which is 7.14% greater than the elastic modulus of NC). Furthermore, when the GT substitution rate exceeds 30%, the continued increase of GT does not significantly improve the elastic modulus of GTRAC. Because there are ITZ or internal micro-cracks (without orientation) between aggregate and cement, coarse aggregate and cement in concrete, these factors determine the elastic modulus of concrete. Therefore, the reasons for the change of the elastic modulus of GT and RA are as follows: (1) The presence of micro-cracks and excessive ITZ in RA can cause the bond between the aggregates to fail prematurely under load, giving the concrete less stiffness and more significant lateral deformation. (2) Due to the filling effect of GT's ultrafine aggregate, the structure and distribution of the internal aggregates and hardened gel of GTRAC are changed. (3) RA itself has the characteristics of many edges and corners, a rough surface, and easy adhesion [7]. When combined with the gel, it can produce a solid physical bond. These three factors produce coupling effects and interact during the GTRAC stress period, leading to changes in the overall stiffness.

In addition, the relationship between compressive strength and modulus of elasticity of GTRAC is shown in Figure 13. According to the linear relationship between the modulus of elasticity and compressive strength, the fitting equation for the relationship between elastic modulus and compressive strength is in equation (4) [36]:

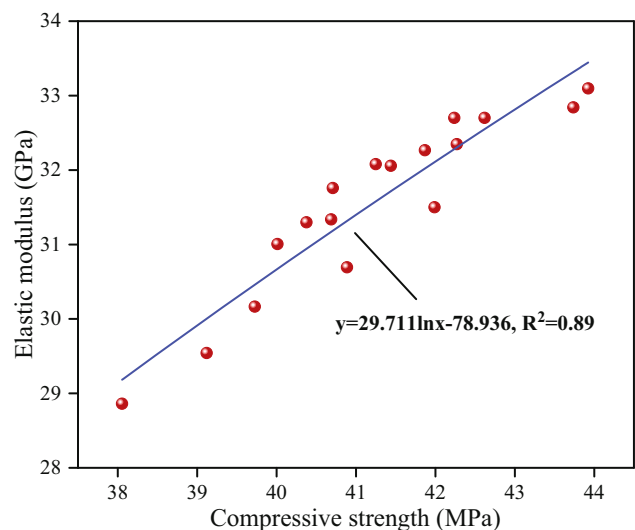


Figure 13: Relationship between compressive strength and modulus of elasticity of GTRAC.

$$E = B \times \ln(f_c) + A. \quad (4)$$

In this article, a well-determined coefficient between compressive strength and modulus of elasticity can be determined in equation (5):

$$E = 29.711 \ln(f_c) - 78.936, R^2 = 0.89, \quad (5)$$

where E is the modulus of elasticity and f_c is the compressive strength. R^2 is the correlation coefficient, which is used to evaluate the coincidence degree between compressive strength and elastic modulus. The closer R^2 value is to 1, the higher the coincidence degree is. The result shows that the R^2 of the regression equation was close to 0.9, indicating that the compressive strength and modulus of elasticity of GTRAC were well correlated and the regression equation was fitted successfully.

3.2 Material characteristic analysis on GTRAC

To further explore the mechanical improvement mechanism of GTRAC, a series of meso-tests were conducted to analyze the changes of meso-structure and hydrate formation based on the influence of the addition of GT.

3.2.1 Material morphology analysis on GTRAC

In this article, the microstructure of GTRAC was investigated using SEM in terms of morphology, densification, and hydration products. It can be obtained from Figure 14 that the amount of GT substituted in 20% (Figure 14b) increases the compactness between the cementitious materials compared to concrete without GT. Similarly, the addition of 30% RA (Figure 14c) results in more cracks and larger pores between the gelling materials than without RA, all of which are consistent with the findings of previous scholarly studies [9]. When GT is substituted at 20% (Figure 14d), the hydration products are partially calcified, such as CH and other substances. They are filled into the ITZ of the aggregate and cement mortar together with the dense and fibrous C–S–H gel, further improving the strength of the ITZ. At the same time, the pore structure and internal crack space are filled with visible new crystals and covered by many hydration products, which ultimately results in a denser concrete structure.

However, GT promotes the increase in the formation of CH and C–S–H to a limit. The addition of 40% GT (Figure 14e) will not lead to the formation of many alkaline hydrates such as CH but will increase the pore

structure and the number or width of cracks. It depends on whether the water phase is evenly distributed during the hydration process. Due to higher water absorption and lower fineness of GT, an increase in GT replacement rate will decrease the degree of hydration. When the RA substitution rate increases, the initial micro-cracks or micro-defects in the concrete increase. Finally, the combination of substances reduces the bonding ability. Likewise, these changes in the pore structure can be demonstrated in Figure 17.

3.2.2 Mineral composition changes of GTRAC

The compound composition analysis results of GTRAC hydration products are shown in Figure 15. The results showed that the RAC without GT addition (GT0RA3) had higher peaks of C_3S and C_2S due to many water-free and inactive cement clinker particles attached to the RA surface, which were not susceptible to secondary hydration reactions. Meanwhile, the addition of 20% GT (GT2RA3) to the RAC had a significant positive effect on improving the XRD peak intensities and peak areas of C–S–H and CH as well as the consumption of C_3S and C_2S compared to the RAC without GT. As can be seen from the figure, compared to GT0RA3, the CH and C–S–H peak areas of GT2RA3 increased by 62.5 and 55.1%. The peak area of C_3S decreased by 54.5%, respectively, which is reasonable because the addition of GT consumed a large amount of CH produced by the hydration of C_2S and C_3S in the cement and combined with it to produce a dense C–S–H gel. The gel adheres to ITZ or cracks and enhances ITZ adhesion, which in turn makes the matrix structure tight.

However, GT did not lead to the production of many hydration products. Compared with GT0RA3, GT4RA3 added with 40% GT consumed 40% of the C_3S peak area but contributed little to promoting CH and C–S–H production. Even the peak area of C–S–H decreased by 2%. It indicates that 40% GT addition did not play a role in the reaction, which may be due to the agglomeration effect of GT, which was also pointed out in the study of Liu et al. [17]. Many GT particles agglomerated to become larger, weakening the volcanic ash reaction. At the same time, most free water would be consumed during the agglomeration process, leading to the uneven distribution of the aqueous phase. C_2S and C_3S are difficult to react with water, thereby affecting the process of hydration reaction. Moreover, a large amount of GT introduces more weak interfaces, leading to a decrease in adhesion between gel systems.

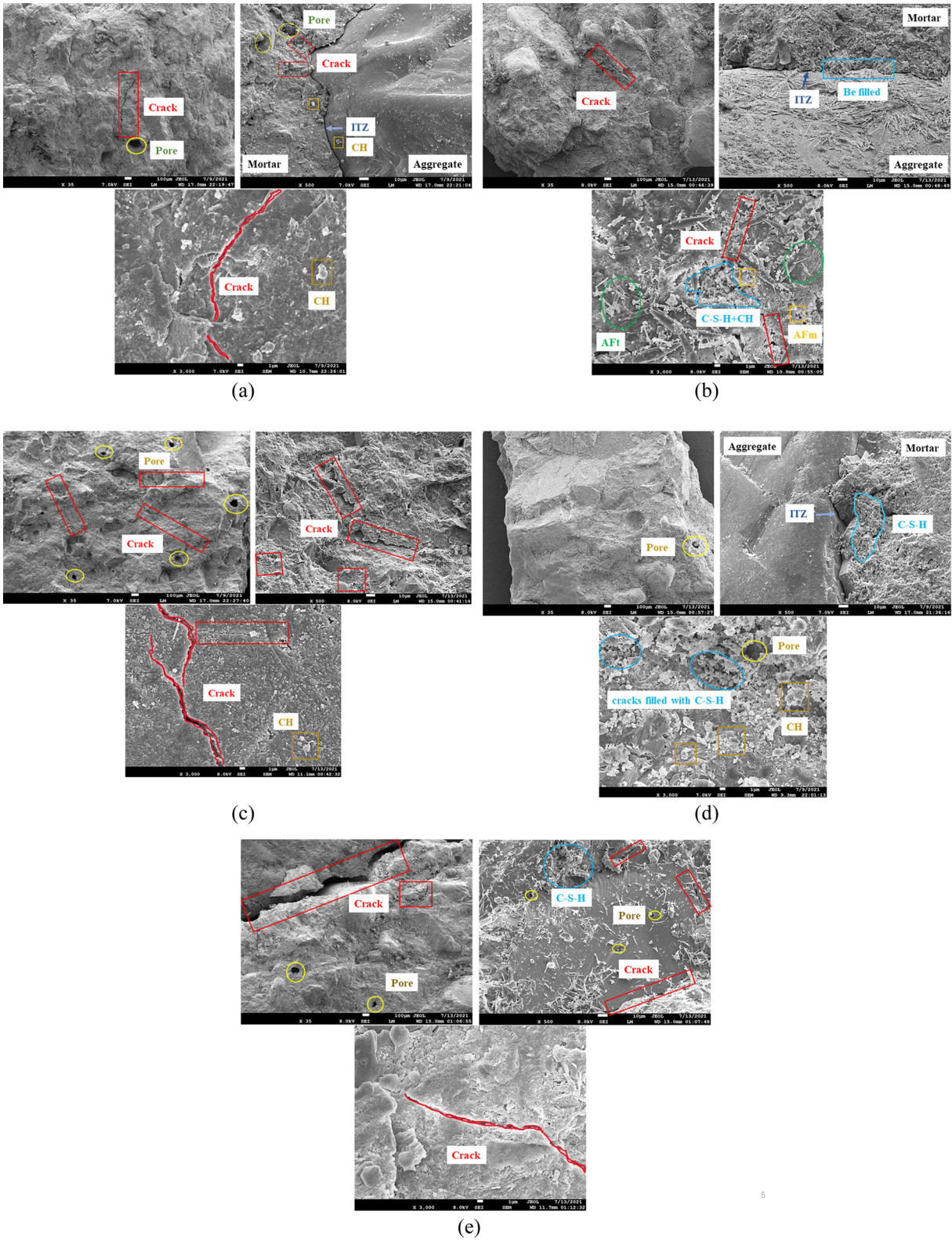


Figure 14: Mesomorphological and mineral crystals analysis on GTRAC: (a) NC, (b) GT2RA0, (c) GT0RA3, (d) GT2RA3, and (e) GT4RA3.

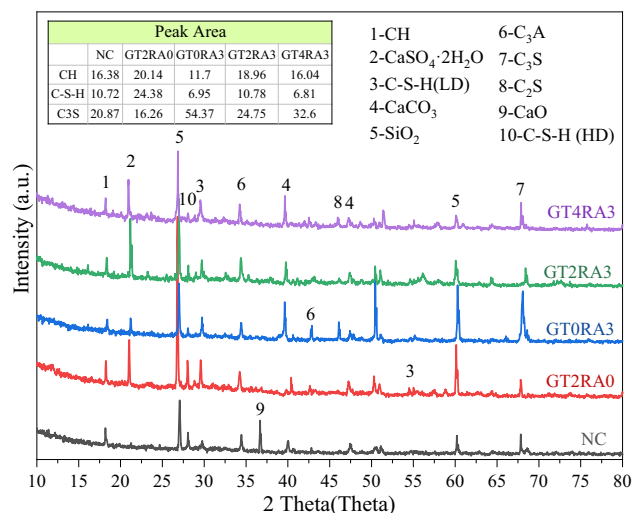


Figure 15: Hydration product analysis of GTRAC.

3.2.3 Functional group changes in the GTRAC

The changes in the functional groups of the GTRAC are determined based on the FTIR experiment and shown in Figure 16. The CO₃²⁻ bending vibration absorption peak (875) and the H-O-H bending vibration absorption peak (1,640) are characteristic peaks for CaCO₃ and H₂O, respectively. It is because changes in the amount of GT and RA blended lead to changes in the amount of calcium carbonate produced and the amount of internal bound water in GTRAC, which causes fluctuations in the two peaks. Meanwhile, the OH vibration (3,640) in Ca(OH)₂ was enhanced in GT2RA3 mixed with 20% GT compared with GT0RA3, which had the effect of increasing the amount of Ca(OH)₂ production from hydration products. Meanwhile, the changes in the Si-O tensile vibration absorption peak (1,090), the Si-O out-of-plane bending vibration absorption peak (525), and the Si-O in-plane bending vibration absorption peak (455) in Q3 tetrahedra are obvious. It indicates that the addition of GT promotes the polymerization of the gelling material to gradually increase with the hydration process and finally form C-S-H(II) gels. Simultaneously, it indicates that the addition of GT effectively promotes the key hydration products in GTRAC, and the hydration process is effectively facilitated. The changes brought about by GT are also found in the studies by Yang et al. [17] and Kathirvel et al. [20]. These hydration products increase the contact area with other cementitious materials and further improve the compactness of the recycled concrete, which is the basis for the great compressive strength that GT2RA3 can achieve. However, after adding 40% GT, the various absorption peaks of the hydrated product did not change significantly. In summary, a proper combination of

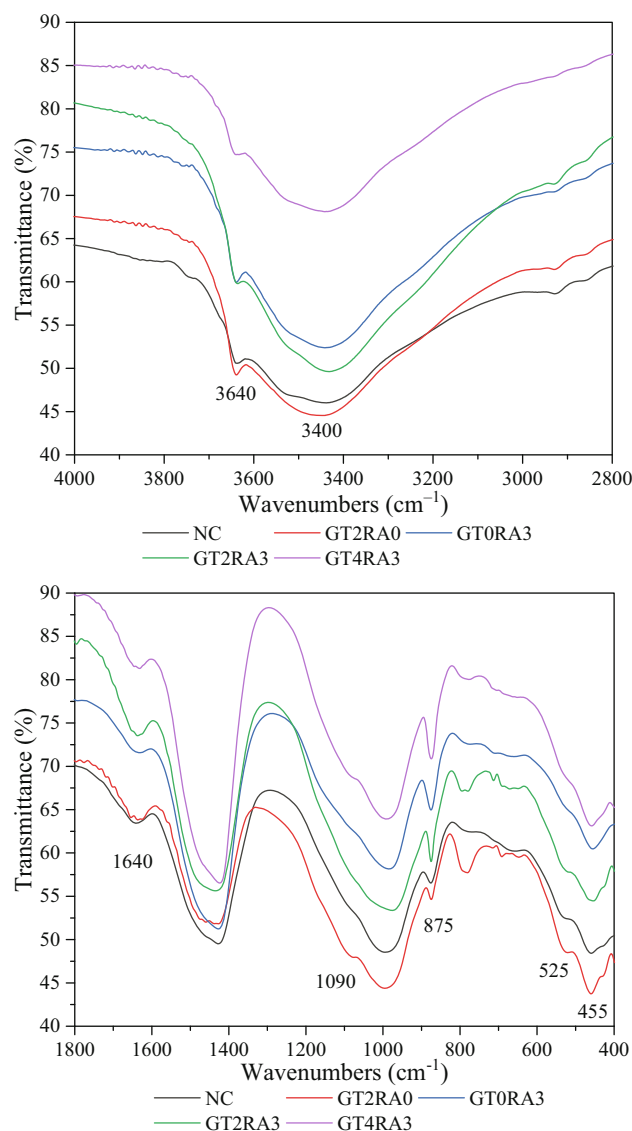


Figure 16: Functional group analysis on GTRAC.

GT and RA can change the vibration of the O-H active group in calcium hydroxide and the Si-O functional group in hydration products, thereby promoting the improvement of the mechanical properties of GTRAC by changing the hydration process.

3.2.4 Evolution of pore structure on GTRAC

The pore structure is a key factor in determining the mechanical properties of concrete, which can be characterized by an MIP test. Figure 17 and Table 6 show the effect of GT and RA on the pore size distribution, porosity, total pore volume, and total pore area of the GTRAC. The cumulative pore volume percentages are shown in Figure 18. According

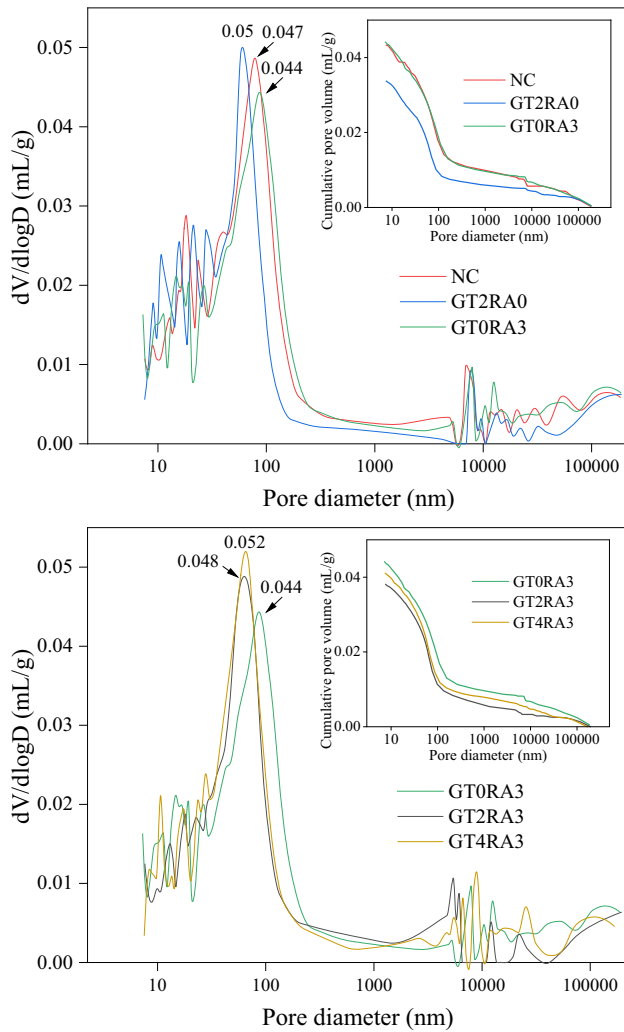


Figure 17: Pore size distribution of GTRAC.

to the classification of the pore, a pore size <20 nm is harmless, 20–100 nm is less harmful, 100–1,000 nm is harmful, and a pore size greater than 1,000 nm is more harmful. The results showed that the main peak height of GT2RA0 increased from 0.047 to 0.050 $\text{mL}\cdot\text{g}^{-1}$ and shifted to the left by 16.22 nm, the pore envelope area decreased, and the porosity decreased by 1.33% compared with that of NC. At

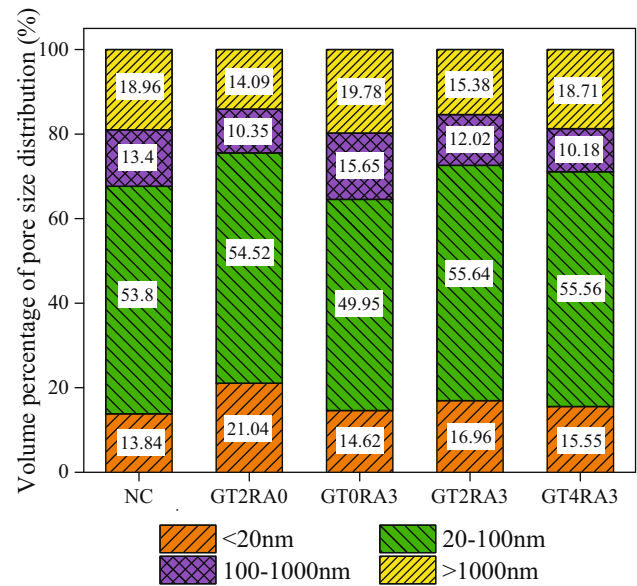


Figure 18: Pore volume percentage of GTRAC.

the same time, the total pore volume of GTRAC decreased, and the percentage of small pores increased by 52.0%. The percentage of significant pores decreased, indicating that the addition of 20% GT had good pore structure and densification properties, which improved the strength [20]. Furthermore, due to the addition of RA in GT0RA3, the main peak height decreased by $0.003 \text{ mL}\cdot\text{g}^{-1}$ and shifted to the right by 8.8 nm, and the harmful pore envelope region (>100 nm) was 3.07% larger than that of NC. It is because there are a lot of holes in the ITZ of RAC [9], and the increase in its dosage mainly affects the larger pore structure and the distribution of internal pores in GTRAC, leading to the decrease in strength. It quantitatively explains why GT and RA change the mechanical properties of concrete. Notably, comparing the pore structure distribution curves of GT0RA3 and GT2RA3, it can be found that GT2RA3 can promote the peak of the pore size distribution curve in GTRAC to shift to the left and rise by $0.004 \text{ mL}\cdot\text{g}^{-1}$, and $1.33 \text{ m}^2\cdot\text{g}^{-1}$ reduced the total accumulated pore area. Meanwhile, the porosity decreases by 1.34%, from 15.88 to

Table 6: Porosity, summary of pore size statistics, total pore size, and area

Specimen number	Porosity (%)	Mean pore diameter (nm)	Mode pore diameter (nm)	Median pore diameter (nm)	Total intrusion volume ($\text{mL}\cdot\text{g}^{-1}$)	Total pore area ($\text{m}^2\cdot\text{g}^{-1}$)
NC	14.61	36.24	74.99	65.13	0.0433	5.30
GT2RA0	13.27	35.36	58.77	58.77	0.0337	4.31
GT0RA3	15.88	42.13	83.79	81.83	0.0441	5.65
GT2RA3	14.54	40.55	64.39	63.23	0.0381	4.32
GT4RA3	14.92	42.09	70.22	65.99	0.0410	4.37

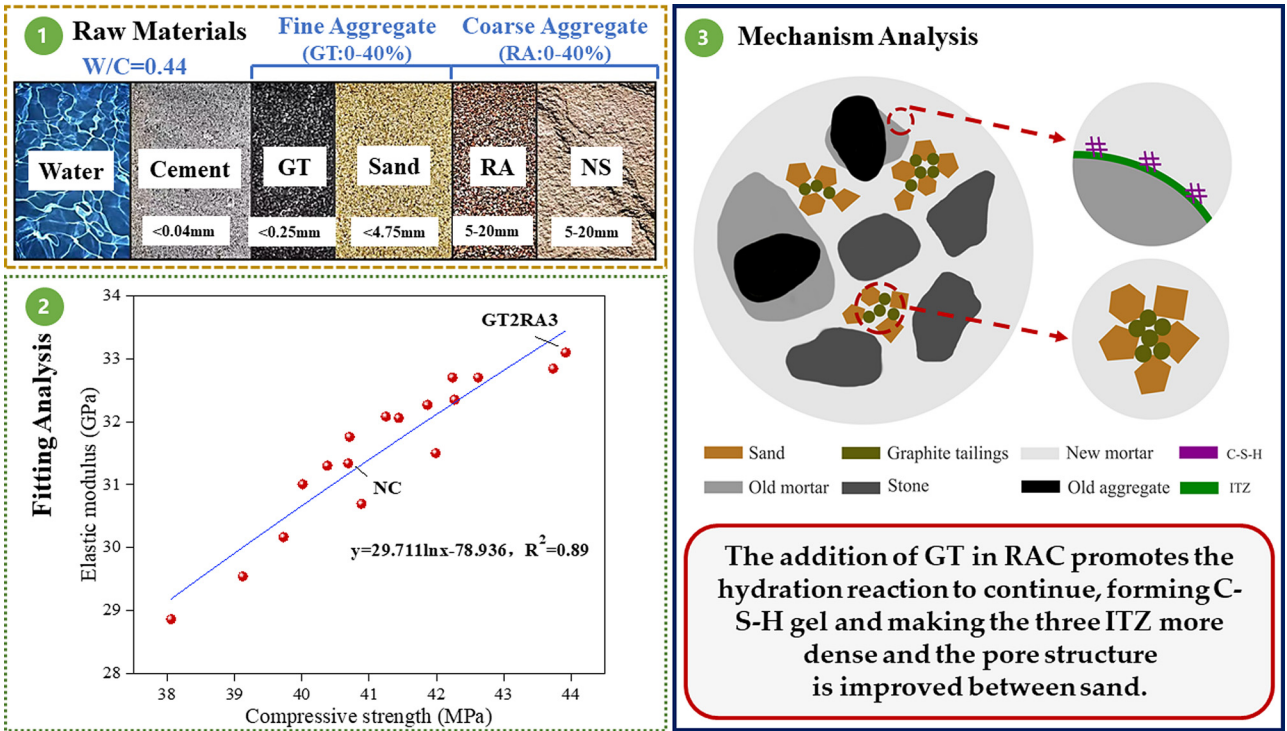


Figure 19: Effect mechanism diagram of GTRAC.

Table 7: Unit price of raw materials

Raw materials	Water	Cement	Natural sand	Natural stone	RA	FA	SF	BP	IT	GT
Unit price (USD·ton ⁻¹)	0.42	72.6	21.31	16.85	8.92	32.69	67.50	0	0	0

Notes: The price of water is the price of domestic water for residents.

14.54%, reducing the envelope area of harmful pores (>100 nm) by 8.03%. Nevertheless, this change is insignificant for the harmless pore area (<20 nm). It is because the fineness modulus of GT is very low. When acting together with RA, the old mortar pores on the RA surface are repaired, although the initial cracks cannot be filled to improve the mechanical properties.

However, when the content of GT was further increased to 40%, the effect of improving the pore distribution was not obvious under the negative effect of easy agglomeration, and the percentage of each accumulated pore showed a decrease. The porosity increased to 14.92% and was greater than that of NC (14.60%). It indicates that the high-water absorption of GT leads to its limited ability to repair the concrete by improving the pore structure space. In conclusion, the appropriate combination of GT and RA materials can improve the pore size distribution of GTRAC to increase its compactness and improve its mechanical properties.

3.2.5 Optimal combined dosage of GT and RA and mechanical improvement mechanism

Based on the above research results, this article analyzes the relationship between compressive strength, elastic modulus, and water absorption. It proposes the best combination of GT and RA based on linear fitting (Figure 19). It can find that the compressive strength and elastic modulus are negatively correlated with water absorption, which indicates that the addition of GT can increase the compactness of GTRAC, thereby improving the mechanical properties. By comparing the point areas on the two fitting curves, the group with the best mechanical properties is obtained as GT2RA3. At the same time, this article analyzes the mechanism of how GT improves the mechanical performance of GTRAC. According to the analysis of the material characterization results of GTRAC under the combined action of GT and RA, (1) the particle size of GT is smaller than that of the natural river sand.

Table 8: Evaluation of the comprehensive performance of GTRAC with other mineral admixtures RAC

Material system (W/C = 0.44)	Optimal replacement value (%)		Mechanical properties (Compared with NC)	Maximum of compressive strength (MPa)	Cost (USD·m ⁻³)	Comprehensive evaluation			
	Mineral	RA				Optimum substitution	Reaction mechanism	Performance improvement	Value engineering
RAC only	0	25	—	36.0–40.0 [24,25,37]	63.09	☆	☆	☆	☆
RAC + Fly ash (FARAC)	10	25	↑	42.4 [12]	64.58	★	★	★	☆
RAC + Silica fume (SFRAC)	5	30	↑	42.0–45.0 [38,39]	63.77–65.45	★	★	★	☆
RAC + Brick powder (BPRAC)	10	25	↑						
	5	30	↑	33.5 [40]	62.05	☆	☆	★	☆
RAC + Iron tailings (ITRAC)	20	30	—	40.4–41.2 [41,42]	59.32	★	☆	☆	★
RAC + GT (GTRAC) (This study)	20	30	↑	45.0	59.32	★	★	★	★

Notes: Calculate the cost according to 1 cubic meter of C30 concrete. Among them, water:cement:sand:stone = 0.38:1:1.11:2.72, and various mixing amounts are calculated according to the substitution ratio of mass.

The right amount of GT (20%) has a good physical filling effect and can replace part of sand as aggregate to form a reasonable gradation to improve the mechanical properties. (2) GT has a promoting effect on the hydration process. The existence of GT has improved the defect of poor bonding between the new concrete grout interface and the old concrete interface. This is because many calcium hydroxide and calcium alumina crystals are enriched at the interface between the old slurry and the new slurry. The active minerals in GT will react with these crystals to promote the formation of C–S–H gel, thereby improving the mechanical properties of GTRAC.

3.2.6 Comprehensive performance evaluation of GTRAC and other types of RAC

The unit price of each raw material is shown in Table 7. All prices are from the average market price of raw materials announced in China in April 2022 and converted to USD (1 USD = 6.69 CNY). The all-around performance of GTRAC was evaluated in comparison with that of other mineral dopant RAC at optimum mix ratios and is shown in Table 8. A comprehensive evaluation of the mechanical properties of these novel RAC systems was performed based on the same water–cement ratio ($W/C = 0.44$) and standard cube strength C30, mainly including the following four aspects. The performance was evaluated by comparing the optimum substitution, reaction mechanism, mechanical properties, and value engineering, and the degrees of advantages and disadvantages were indicated by asterisks.

3.2.7 RA and mineral utilization

With regard to the optimum substitution rates for RA and minerals in Table 8, the substitution rate of BPRAC for brick is only 5%, which is a very low utilization rate compared to other mineral RACs. Also, the compressive strength is only 33.51 MPa [40]. These two deficiencies are key to the lack of widespread utilization of BPRAC.

3.2.8 Reaction mechanism

The main mechanisms of action of mineral dopants to improve the mechanical properties of RAC are the morphological effect, the filling effect, and the volcanic ash effect. The studies show that FARAC [12] and SFRAC [38,39] have all three of these effects. However, for BPRAC

and ITRAC, only the filling effect and the morphological effect are reflected in the compressive strength. That is why they only have 33.51 [37] and 40.4 MPa [41]. The good volcanic ash activity is the advantage of GTRAC over other tailings and is the key to its ability to replace natural sand and increase the strength of RAC, and be widely utilized.

3.2.9 Performance improvement

From Table 8, the addition of reactive mineral admixtures to RAC can improve the overall performance. Fly ash and silica fume can improve the workability and strength of RAC better. However, compared with GT, brick power, iron tailings, and steel slag of RAC show a weaker performance improvement and have little application value. From Table 8, the addition of reactive mineral admixtures to RAC can improve the overall performance. Kou et al. [12] and Dilbas et al. [38] showed that fly ash and silica fume could better improve the workability and strength of RACs. However, macroscopic and microscopic studies of the mechanical properties of BPRAC [40] and ITRAC [41,42] show that although BP and IT can improve the pore structure of RAC to increase the compressive strength, the increase in compressive strength is not significant and has little application value.

Although fly ash can improve the workability of mixing materials [12], the massive application of fly ash will lead to a too large slump, and fly ash particles are easy to float and bleed, which is not conducive to the practical application of engineering. Meanwhile, the contribution of silica fume to RAC is mainly before 28 days. Therefore, SFRAC is generally considered inferior to pure cement concrete or fly ash concrete in terms of long-term strength growth. Their low early strength and substitution amount make it challenging to obtain practical engineering applications. GTRAC has an advantage over FARAC [12] and SFRAC [38,39] in reusing waste material for almost the same compressive strength. At the same time, GT enhanced RAC performance and offers advantages in all four areas while providing good workability. It is essential to GTRAC's advantage over other mineral RAC materials.

3.2.10 Value engineering

Based on a comprehensive survey of the raw material usage, types, and unit prices of ready-mixed concrete in China (Table 7), C30 ordinary concrete with a standard

ratio [23] (water:cement:sand:stone = 0.38:1:1.11:2.72) was selected as the economic evaluation index. Since BP, IT, and GT are solid waste, their prices are calculated at 0. The calculation results of the total price of one cubic meter of concrete are shown in Table 8. After calculation, the price of C30 standard concrete is 65.55 USD per cubic meter, and the price of recycled concrete that replaces 25% RA is 63.09 USD per cubic meter. From the cost calculation results in Table 8, due to the high price of SF itself, there is no advantage in reducing the price after incorporation. The amount of BP added is relatively small, and its own low-price advantage is difficult to exert. The cost per cubic meter of FARAC, ITRAC, and GTRAC was reduced by 2.376, 4.24, and 4.24%, respectively, compared with RAC incorporating 25% RA. However, considering the performance of other performances, GTRAC has wider application prospects than FARAC and ITRAC.

In summary, compared to other mineral RAC systems, GTRAC in this study is a new product that improves the overall performance of RA concrete by considering the renewable use of solid waste resources. GTRAC has a good balance between good workability and mechanical properties and has good prospects for engineering applications.

4 Conclusions

As the waste of graphite mining and abandoned construction waste, GT and RA have already caused considerable damage to the natural ecological environment. Similarly, solving the massive accumulation of dumped construction waste is a critical issue. Incorporating GT and RA into concrete is an effective and sustainable development method. In this article, GT was added to RAC instead of river sand, and the mechanical properties and material characterization of GTRAC were studied. The following conclusions can be drawn from this study.

- GT significantly inhibited the workability of RAC. The maximum compressive strength of concrete is 20% of the replacement rate of GT and 30% of the replacement rate of RA, and the strength is increased by 15.01%. The elastic modulus change is linearly related to the compressive strength, which also increases by 7.14%. In addition, the optimal material system reduces the water absorption of GTRAC, and the variation of surface water content was opposite to that of water absorption.
- The microscopic analysis combined with SEM, XRD, FTIR, and MIP showed that the homogeneously dispersed GT had a good filling effect. GT effectively enhanced the

bond between the aggregate and the cement matrix by changing the microscopic morphology, improving the pore size structure distribution, and acting as a densifier. In addition, the addition of GT changed the hydration products of RAC, the functional groups, or bonding positions of key hydration compounds to influence the mechanical properties of the RAC material system.

- Compared with other solid waste RAC, GTRAC has great advantages in optimum substitution, reaction mechanism, mechanical properties, and value engineering. In conclusion, 20% GT content to 30% RA replacement ratio are the optimal admixture of GTRAC.

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Conflict of interest: We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of the manuscript.

Data availability statement: Data available on request due to constraints required for subsequent corresponding work. Readers in need can ask and we can provide. Thank you very much for your understanding.

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