Effects of Nanomaterials Reinforced Aggregate on Mechanical Properties and Microstructure of Recycled Brick Aggregate Concrete

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In this paper, the effects of nano-SiO_2 and nano-Al_2O_3 on the brick aggregate properties, compressive strength of brick aggregate concrete at different ages, and microstructure of brick aggregate concrete are investigated. The results show that the water absorption of recycled brick aggregate decreases with the increase of nanomaterials solution concentrations, and the crush index of recycled brick aggregate decreases. Nano-SiO_2 and nano-Al_2O_3 improve the compressive strength of recycled brick aggregate concrete at different ages, and the effect of nanomaterials on the early strength of concrete are more obvious. With the increase of nano-solution concentration, the compressive strength of nano-SiO_2 reinforced brick aggregate concrete increases first and then decreases, while that of nano-Al_2O_3 reinforced brick aggregate concrete increases. The internal structure of nanomaterials reinforced brick aggregate is denser, micro cracks and pores are filled with hydration products, and the interfacial transition zone is more compact.
Keywords: recycled brick aggregate concrete, nano-SiO_2, nano-Al_2O_3, compressive strength, microstructure.

1. INTRODUCTION

With the rapid development of the construction industry, the use of natural aggregates has increased dramatically, and more and more natural resources such as sand and gravel aggregates are consumed [1]. At the same time, a large amount of construction waste is generated in the process of construction and demolition [2], and construction waste accounts for about 40 % of urban waste [3], which is increasingly harmful to the ecological environment [4]. The recycled concrete prepared with the treated construction waste as recycled aggregate can effectively alleviate the above problems. The composition of construction waste is complex, among which waste concrete and waste bricks account for a large proportion [5]. Most of the existing research focuses on recycled coarse aggregate concrete [6], and the research on recycled brick aggregate concrete is relatively less. The research on recycled bricks aggregate concrete is of great significance for the recycling and utilization of construction waste.

Recycled brick aggregate has the disadvantages of high water absorption, many micro-cracks, and large porosity, which leads to the strength of brick aggregate being lower than ordinary coarse aggregate [7], which will inevitably affect the mechanical properties and durability of recycled brick aggregate concrete. Through the study of the mechanical properties of recycled brick aggregate concrete such as compressive strength [8], splitting tensile strength, and flexural strength [9], it was found that when recycled brick aggregate concrete was subjected to load, the interfacial transition zone between brick aggregate and mortar first appeared strain concentration, and the cracks did not develop along the interfacial transition zone, but penetrate the brick aggregate. It can be seen that the weak parts of recycled brick aggregate concrete are quite different from those of recycled coarse aggregate concrete, and there are two weak parts of recycled brick aggregate concrete: interfacial transition zone and recycled brick aggregate [10]. Therefore, to improve the mechanical properties of recycled brick aggregate concrete, it is necessary not only to the bonding strength to strengthen the interfacial transition zone, but also to improve the basic properties of recycled brick aggregate.

Nanomaterials refer to particles with a particle size in the range of 1 to 100 nm, which are characterized by small size and large specific surface area [11, 12]. Nanomaterials can not only fill the gap between cement particles [13], but also improve the structure of the aggregate contact area, and promote the pozzolanic reaction of cement [14], thereby improving the mechanical properties and durability of concrete [15].

There was much research on the application of nano-SiO_2 in concrete [16], the large specific surface area and small size of nano-SiO_2 could promote hydration reaction, consumed a large amount of $Ca(OH)_2$, produced more C-S-H gels to fill the internal pores of concrete [17, 18],

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optimized the internal pore structure. Li et al. [19] analyzed the influence of nano-SiO_2 on the formation law of hydration products and the microstructure of concrete through experiments, and the results showed that nano-SiO_2 promoted the early hydration reaction of concrete. This can not only improve the strength of concrete but also improve its durability of concrete [20, 21]. Wang et al. [22] investigated the synergistic effect of nano-SiO_2 and silica fume on the hydration performance of cement-based materials and found that the composite mixing of nano-SiO_2 and silica fume could make the cement paste more compact in the microstructure. Xiao et al. [23, 24] investigated the effect of fly ash and nano-SiO_2 on the mechanical properties of recycled aggregate concrete and found that mixed fly ash could compensate for the reduction of properties of recycled concrete caused by single-mixed nano-SiO_2. Adding nano-SiO_2 and 30 % fly ash could effectively improve the 28-day splitting tensile strength of recycled aggregate concrete. Meng et al. [25] found that the nano-SiO_2 could significantly improve the density and mechanical properties of the interface transition zone. There are relatively few studies on the application of nano-Al_2O_3 in concrete, nano-Al_2O_3 could improve the mechanical properties, frost resistance, and toughness of concrete [26]. The reason is that nano-Al_2O_3 improves the density of cement hardened paste and increases the interface transition zone of concrete [27]. The results of Li et al. [28] showed that the compressive strength of concrete increased from 15 % to 20 %, and the elastic modulus of concrete increased by more than 40 % after replacing 5 % of cement with nano-Al_2O_3. In addition, nano-Al_2O_3 could reduce the initial setting time of ultra-high-performance concrete, which had a great influence on the performance of concrete [29].

However, there are few studies on recycled brick aggregate concrete, and the effect of recycled brick aggregate on the mechanical properties of concrete at different ages has not been proved. In addition, there are few studies on nanomaterials in brick aggregate concrete, the current research mostly focused on improving the performance of cement mortar and interface transition zone by nanomaterials, while research on improving aggregate performance by nanomaterials was relatively rare. However, improving the properties of brick aggregates is the key to improving the mechanical properties of concrete. Improving the physical properties of recycled brick aggregates through nanomaterials can not only improve the mechanical properties of recycled brick aggregate concrete, but also improve the durability of recycled brick aggregate concrete. This paper investigates the effects of different nanomaterials (nano-SiO_2 and nano-Al_2O_3) and different nanomaterials solution concentrations (1 %, 2 % and 3 %) on the brick aggregate properties (apparent phenomenon, crush index and water absorption), the compressive strength of recycled brick aggregate concrete at different ages (7 d, 14 d and 28 d), and the microstructure of recycled brick aggregate concrete; reveals the strengthening mechanism of nanomaterials on recycled brick aggregate concrete. It can provide a theoretical basis and technical support for the application of recycled brick aggregate.

2. EXPERIMENTAL DETAILS

2.1. Raw materials

1. Cement. P.O 42.5 cement conforming to the Chinese standard GB 175-2007 [30], produced by Zibo Luzhong Cement Co., Ltd. was used in this paper. The properties of cement are listed in Table 1.

Table 1. Performance of cement

Initial setting time, min	Final setting time, min	Compressive strength, MPa		Flexural strength, MPa		Specific surface area, m^2 kg^{-1}
		3-day	28-day	3-day	28-day	
183	248	22.2	47.3	4.9	7.8	342

2. Fine aggregate. The fine aggregate used in this paper is natural river sand with a fineness of 2.7. The properties of natural river sand are listed in Table 2.

Table 2. Properties of natural river sand

Property	Value
Mud content, %	1.7 %
Particle diameter, mm	< 4.75

3. Coarse aggregate. The coarse aggregate used in this paper includes the natural broken stone aggregate (NA) and the recycled brick aggregate (BA), as shown in Fig. 1. The properties of natural broken stone aggregate and recycled brick aggregate are listed in Table 3.

a b

Fig. 1. Coarse aggregate: a – NA; b – BA

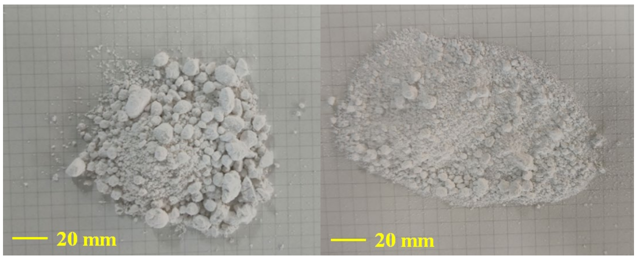
Table 3. Properties of coarse aggregate

Property	Type	
	NA	BA
Size of particles, mm	5 – 26.5	5 – 26.5
Water absorption, %	1.09	12.54
Crushing index, %	4.9	32.9

4. Nanomaterials. The nano-SiO_2 and nano-Al_2O_3 used in this paper are shown in Fig. 2, and the properties of nano-SiO_2 and nano-Al_2O_3 are listed in Table 4.

Table 4. Properties of nanomaterials

Type	Particle size, nm	Specific surface area, m^2 g^{-1}	Bulk density, g m^{-3}
Nano-SiO_2	20	240	0.06
Nano-Al_2O_3	30	100	1.5

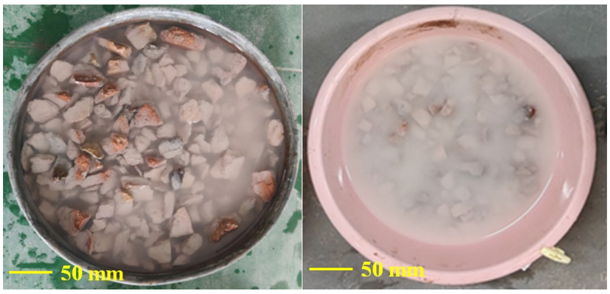


a b

Fig. 2. Nanomaterials: a – nano-SiO_2; b – nano-Al_2O_3

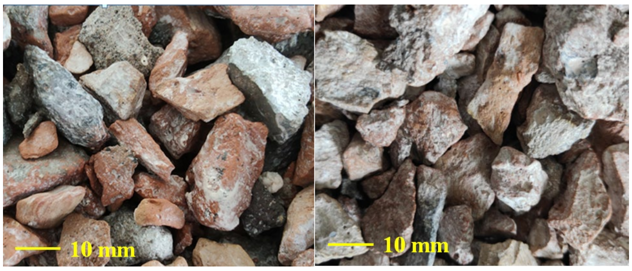
2.2. Nanomaterial reinforced brick aggregate method

The nano-powder was added to the solution and mechanically stirred for 5 min. Then, the nano-solution was dispersed by an ultrasonic dispersion instrument to prevent the agglomeration of particles. The brick aggregate was soaked in different nano-solutions (nano-SiO_2 and nano-Al_2O_3) with different solution concentrations (1 %, 2 %, and 3 %) for 2 days, as shown in Fig. 3. The apparent phenomenon of brick aggregate after soaking in different nano-solutions is shown in Fig. 4. Natural broken stone aggregate were not reinforced by nanomaterials.



a b

Fig. 3. Aggregate immersion: a – nano-SiO_2; b – nano-Al_2O_3



a b

Fig. 4. Apparent phenomenon of BA: a – nano-SiO_2; b – nano-Al_2O_3

2.3. Mix proportions

The mix proportions of natural aggregate concrete (NAC) and brick aggregate concrete (RBC) are listed in Table 5.

Table 5. Mix proportions (kg m-3)

Type	Water	Cement	Natural aggregate	Sand	Brick aggregate *
NAC	208	320	1120	700	0
RBC	208	320	784	700	336
* Brick aggregate reached the surface-dry moisture state.					

2.4. Test method

2.4.1. Water absorption

The aggregates were placed in the container with water, and the water surface was about 5 mm higher than the aggregate surface. After 24 hours, the aggregates were taken out, and the water on the aggregate surface was wiped dry to reach the saturated surface dry state, and the total mass of the sample and the shallow plate was weighed (m_2). The saturated surface dry aggregates were dried in a drying oven at 105 °C ± 5 °C and cooled for more than 1 h. The total mass of the aggregates and the shallow plate (m_1) and the mass of the shallow plate (m_3) were weighed. Water absorption (ω_{wa}) is calculated as follows:

$$\omega_{wa} = \frac{m_2 - m_1}{m_1 - m_3} \times 100\% \cdot \quad (1)$$

2.4.2. Crush index

The aggregates were divided into two layers and loaded into the crushing index tester, and 200 kN was uniformly loaded at a speed of 1 kN/s and stabilized for 5 s, and then unloaded. The aggregates were poured out and their mass (m_0) was weighed. The crushed fine particles with particle size less than 2.5 mm were screened out and the remaining aggregate mass (m_1) was weighed. The crush index (δ) is calculated as follows:

$$\delta = \frac{m_0 - m_1}{m_0} \times 100\% \cdot \quad (2)$$

2.4.3. Compressive strength

The compressive strength test was carried out with a computer-controlled electronic universal hydraulic testing machine, and the control stress was 0.1 MPa/s until the test specimen was damaged.

2.4.4. Microscopic analysis

The specimen was cut into a suitable size by the cutting machine to ensure that the observation surface was flat. After drying in a drying oven at 60 °C, gold plating and vacuum treatment were performed, and scanning electron microscopy was used for observation.

3. RESULTS AND DISCUSSION

3.1. Nano-reinforced aggregate properties

3.1.1. Water absorption

The water absorption of brick aggregate after soaking in different nano-solutions is shown in Fig. 5. Whether in nano-SiO_2 solution or nano-Al_2O_3 solution, the water absorption of recycled brick aggregate decreased with the increase of solution concentrations. The reason is that the nano-material can penetrate inside the micro-cracks of aggregate and fill the internal pores, thereby reducing the water absorption of the aggregate [31].

In addition, when the nano-solution concentration was the same, the water absorption of recycled brick aggregate soaked in the nano-SiO_2 solution was lower than that soaked in the nano-Al_2O_3 solution. When the nano-SiO_2 solution concentration was 1 %, 2 % and 3 %, the water absorption of recycled brick aggregate decreased by 17.6 %, 21.4 %, and 27.9 %, respectively. When the the nano-Al_2O_3 solution concentration was 1 %, 2 % and 3 %, the water absorption of recycled brick aggregate decreased by 5.9 %, 16.3 %, and 24.2 %, respectively.

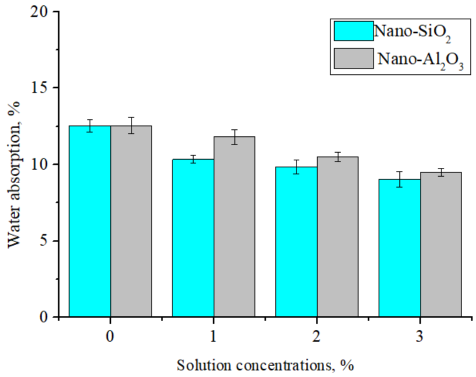


Fig. 5. Water absorption of brick aggregate after soaking in nano-solutions

3.1.2. Crush index

The crush index of brick aggregate after soaking in different nano-solutions is shown in Fig. 6.

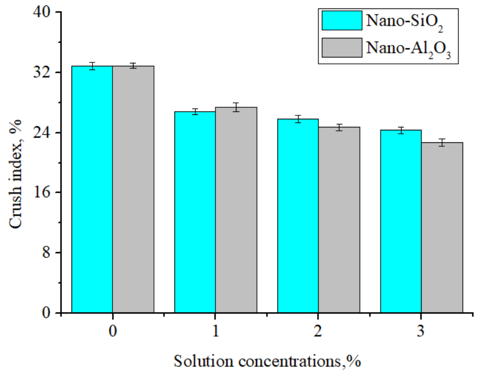


Fig. 6. Crush index of brick aggregate after soaking in nano-solutions

Whether in nano-SiO_2 solution or nano-Al_2O_3 solution, the crush index of recycled brick aggregate decreased with the increase of solution concentrations. The reason is that the nanomaterials filled the pores of the internal micro-cracks to increase the aggregate strength, resulting in the reduction of the crush index.

In addition, when the nano-solution concentration was 1 %, the crush index of recycled brick aggregate soaked in nano-SiO_2 solution was lower than that soaked in nano-Al_2O_3 solution; when the nano-solution concentrations were 2 % and 3 %, the crush index of recycled brick aggregate soaked by nano-SiO_2 solution was higher than that soaked by nano-Al_2O_3 solution. The reason is that the solubility of nano-SiO_2 is higher than that of nano-Al_2O_3, when the nano-solution concentration is low, nano-SiO_2 is easier to enter the pores inside the aggregate and fill the pores. With the increase of nano-solution concentration, more agglomeration products appeared on the aggregate surface soaked in nano-SiO_2 solution (as shown in Fig. 7). The agglomeration product prevents water from entering the aggregate, reduces the water absorption of the aggregate, but slows down the improvement of the aggregate strength.

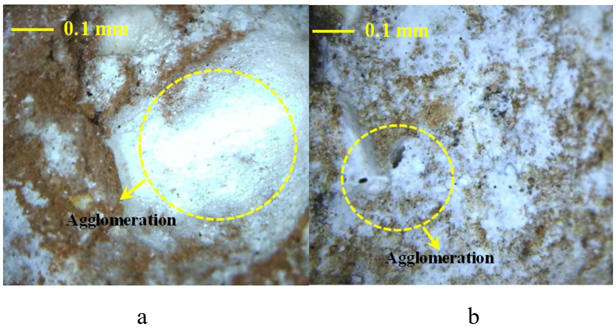


Fig. 7. Surface magnification of aggregate: a – 3 % nano-SiO_2; b – 3 % nano-Al_2O_3

Therefore, when the nano-solution concentration exceeded 2 %, the crushing index of the aggregate after soaking in nano-SiO_2 solution was higher than that after soaking in nano-Al_2O_3 solution.

3.2. Failure mode

The compressive failure mode of natural aggregate concrete and brick aggregate concrete after 28 days of curing is shown in Fig. 8. The compressive failure of natural aggregate concrete occurred in the interface transition between aggregate and mortar, and the natural aggregate was generally complete. For the brick aggregate concrete, the damaged cracks penetrated through the brick aggregate, and the brick aggregate was destroyed. For the nanomaterials reinforced brick aggregate concrete, the number of concrete cracks became less, and the specimen was more complete after failure. Although the brick aggregate strength was improved, the brick aggregate was still destroyed or crushed after failure.

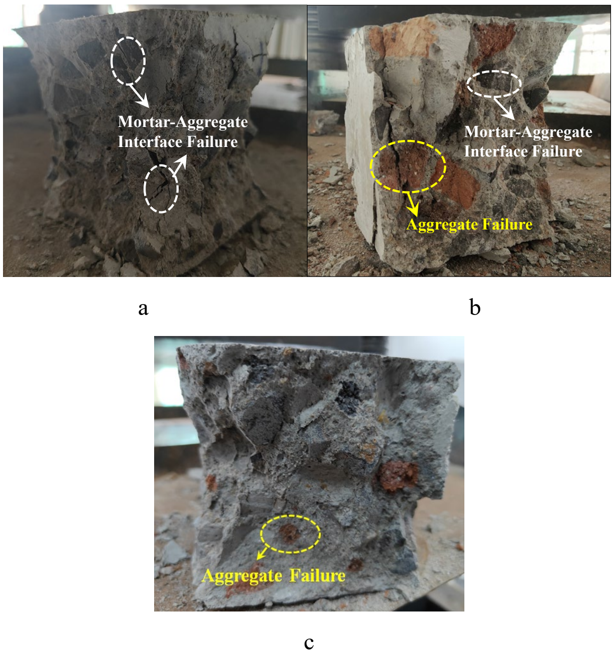


Fig. 8. Compressive failure mode of concrete: a – natural aggregate concrete; b – brick aggregate concrete; c – nanomaterials reinforced brick aggregate concrete

3.3. Compressive strength

3.3.1. Effect of nano-SiO_2 on compressive strength

The compressive strength of nano-SiO_2 reinforced brick aggregate concrete at different ages is shown in Fig. 9. Nano-SiO_2 improved the compressive strength of recycled brick aggregate concrete at different ages, and the effect of nano-SiO_2 on the 14-day compressive strength was more obvious, resulting in the compressive strength of recycled brick aggregate concrete exceeding that of natural aggregate concrete.

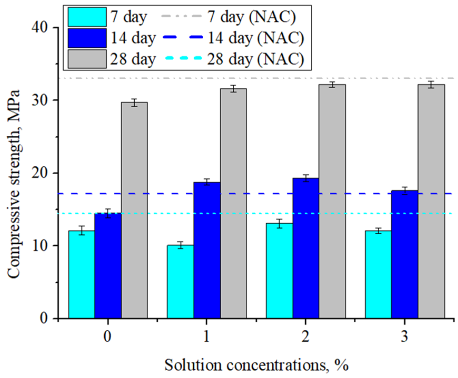


Fig. 9. Compressive strength of nano-SiO_2 reinforced brick aggregate concrete

The reasons for the improvement of concrete compressive strength caused by nano-SiO_2 are as follows: on the one hand, recycled brick aggregate strength increases after soaking in nano-SiO_2 solution, and the compressive strength of recycled aggregate concrete is improved; on the other hand, the nano-SiO_2 attached to the surface of recycled brick aggregate reacts with the hydration product $Ca(OH)_2$ of cement, forming C-S-H that has a certain contribution to the concrete strength, promoting the hydration of cement, improving the binding strength between cement mortar and aggregate, and thus improving the concrete strength [32, 33].

In addition, with the increase of nano-SiO_2 solution concentration, the compressive strength of concrete at different ages increased first and then decreased. When the concentration of the nano-SiO_2 solution was 1 %, 2 %, and 3 %, the 14-day compressive strength of recycled brick aggregate increased by 29.7 %, 33.1 %, and 21.4 %, respectively. The reason is that when the nano-SiO_2 solution concentration exceeds 2 %, the agglomeration of nano-SiO_2 on the aggregate surface is more serious, which leads to the incomplete reaction between nano-SiO_2 and cement hydration products. The isolation of agglomerated nano-SiO_2 leads to a new interface transition zone (ITZ) in concrete, as shown in Fig. 10.

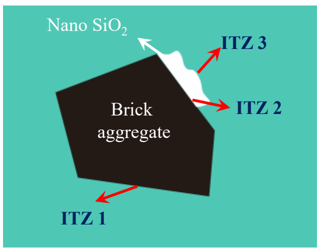


Fig. 10. Schematic diagram of ITZ

ITZ 1 is the mortar-brick aggregate interface transition zone; ITZ 2 is the nano-SiO_2-brick aggregate interface transition zone; ITZ 3 is the nano-SiO_2-mortar interface transition zone. ITZ is the weak zone of concrete [34, 35], which leads to a decrease in concrete strength.

3.3.2. Effect of nano-Al_2O_3 on compressive strength

The compressive strength of nano-Al_2O_3 reinforced brick aggregate concrete at different ages is shown in Fig. 11. The compressive strength of nano-Al_2O_3 reinforced brick aggregate concrete at different ages was larger than that of brick aggregate concrete, and the compressive strength of nano-Al_2O_3 reinforced brick aggregate concrete at different ages increased with the increase of nano-Al_2O_3 solution concentration. The reasons for the improvement of concrete compressive strength caused by nano-Al_2O_3 are as follows: on the one hand, nano-Al_2O_3 improves the microscopic pore structure and interface transition zone of concrete, reduces aggregate cracks, and improves the brick aggregates strength; on the other hand, the secondary hydration reaction between nano-Al_2O_3 and cement hydration products forms a new dense network structure [36], which refines the harmful calcium hydroxide crystals in the interfacial transition zone, increases the content of hydration product C-S-H in the weak interfacial transition zone, thereby significantly improving the strength of brick aggregate concrete [37].

In addition, the effect of nano-Al_2O_3 on the early strength of concrete was more obvious, which was consistent with the results obtained by Xie [38]. When the nano-Al_2O_3 solution concentration increased from 0 % to 3 %, the 7-day compressive strength, 14-day compressive strength, and 28-day compressive strength increased by 19.8 %, 43.4 %, and 8.4 %, respectively. This is because nano-Al_2O_3 particles accelerate the formation of hydration products [39].

It can also be seen from Fig. 9 and Fig. 11 that when the solution concentration is 1 %, the 7-day compressive strength of nano-SiO_2 (nano-Al_2O_3) reinforced recycled brick aggregate concrete was lower than that of unreinforced recycled brick aggregate concrete.

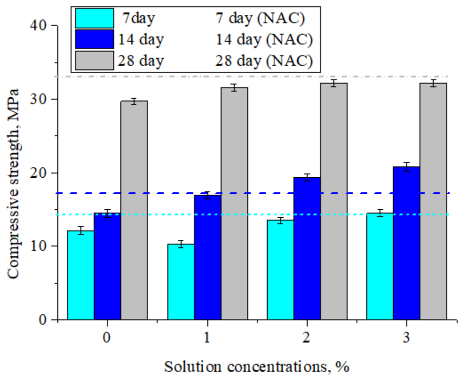
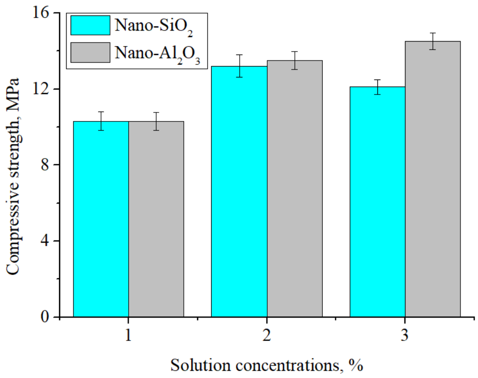


Fig. 11. Compressive strength of nano-Al_2O_3 reinforced brick aggregate concrete

The reason is that when the nano-solution concentration is low, there are less nano-particles attached to the brick aggregate surface, and its hydration reaction with cement is slow. At the age of 7 days, the degree of hydration reaction between nanomaterials and cement is relatively low. In addition, nanomaterials that do not participate in hydration reaction hinder the improvement of bond strength between aggregate and cement mortar to some extent.

The comparison of the strengthening effects of nano-Al_2O_3 and nano-SiO_2 is shown in Fig. 12.



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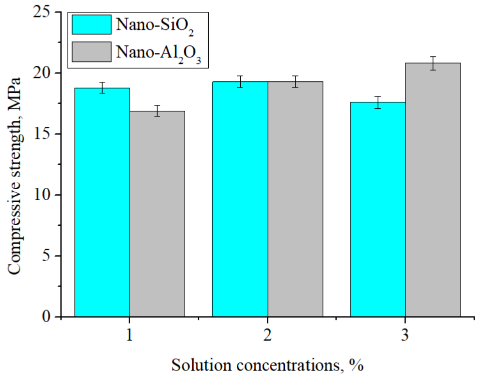

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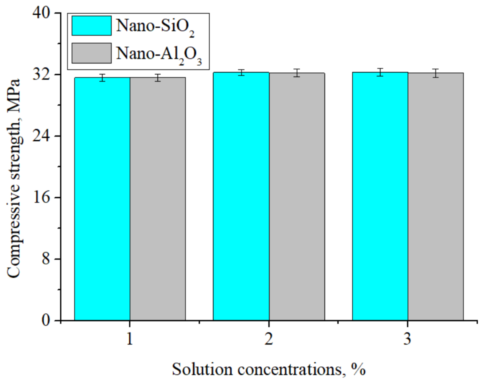


c

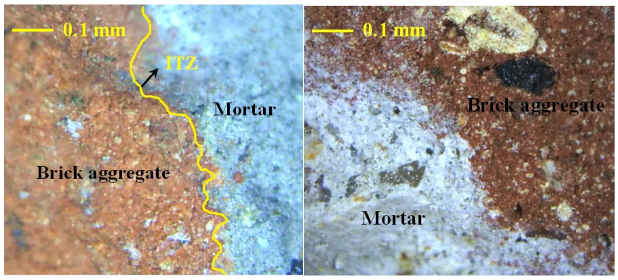
Fig. 12. Comparison of strengthening effects of nano-Al_2O_3 and nano-SiO_2: a – 7 d; b – 14 d; c – 28 d

When the nanomaterials solution concentration was 1 %, the compressive strength of nano-SiO_2 reinforced brick aggregate concrete was slightly higher than that of nano-Al_2O_3 reinforced brick aggregate concrete, especially the 14-day compressive strength; when the nanomaterials solution concentration was 2 %, the compressive strength of nano-SiO_2 reinforced brick aggregate concrete was the same as that of nano-Al_2O_3 reinforced brick aggregate concrete; when the nanomaterials solution concentration was 3 %, the compressive strength of nano-SiO_2 reinforced brick aggregate concrete was slightly lower than that of nano-Al_2O_3 reinforced brick aggregate concrete.

The reasons are as follows: the solubility of nano-SiO_2 is higher than that of nano-Al_2O_3, nano-SiO_2 is easier to enter the pores inside the aggregate, and fills the pores and micro-cracks to improve the aggregate strength. In addition, nano-SiO_2 is smaller than nano-Al_2O_3, with a large specific surface area and a more complete hydration reaction with cement, resulting in higher concrete strength [40]. However, with the increase in solution concentration, nano-SiO_2 is more prone to agglomeration. The agglomeration phenomenon hinders the nanoparticles from entering the aggregate, and at the same time, it also generates a new interfacial transition zone due to an incomplete hydration reaction, thereby reducing the concrete strength.

3.4. Microstructure

The electronic microscope produced by Jiangsu Leyes Technology Co., Ltd. was used to observe the internal structure of brick aggregate concrete as shown in Fig. 13 It can be seen that there were micro-cracks and pores in the brick aggregate concrete, and the interface transition zone between aggregate and mortar was obvious. The internal structure of nanomaterials reinforced brick aggregate was denser, micro-cracks and pores were filled by hydration products, and the interface transition zone was more compact.



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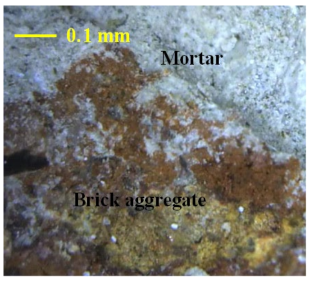

c

Fig. 13. Internal structure of brick aggregate concrete: a – RBC is brick aggregate concrete; b – nano-SiO_2-RBC is the nano-SiO_2 reinforced brick aggregate concrete; c – nano-Al_2O_3-RBC is the nano-Al_2O_3 reinforced brick aggregate concrete

Fig. 14 shows the microstructure of brick aggregate concrete. There were more pores and micro-cracks in the brick aggregate and its interface transition zone with cement mortar. Although there were hydration products C-S-H and AFt, the accumulation was relatively concentrated, and the pores and micro-cracks could not be filled. In addition, the aggregate interface had a smooth interface, and the hydration products were not attached, resulting in poor bonding performance at the interface zone.

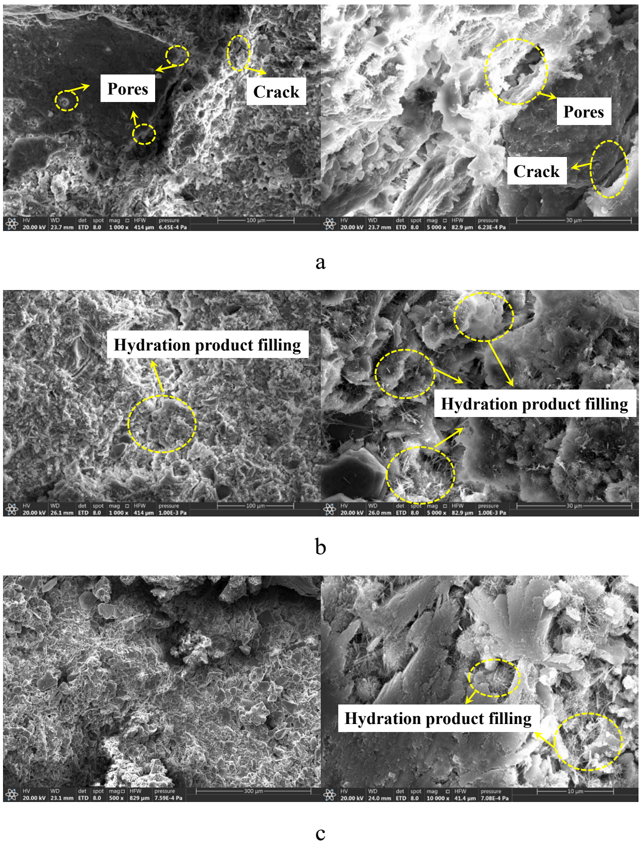

Fig. 14. Microstructure of brick aggregate concrete: a – the microstructure of brick aggregate concrete; b – the microstructure of nano-SiO_2 reinforced brick aggregate concrete; c – the microstructure of nano-Al_2O_3 reinforced brick aggregate concrete

In the nano-SiO_2 reinforced brick aggregate concrete, a large number of needle-shaped AFt and flocculent C-S-H were generated in the cracks and pores of the brick aggregate and its interface transition zone with cement mortar, which has a good filling effect on cracks and pores, and the interface transition zone structure was more compact. The properties of aggregate and interface transition zone were improved, thus improving the compressive strength of brick aggregate concrete.

The principle of nano-Al_2O_3 reinforced brick aggregate concrete was similar to that of nano-SiO_2, which filled the cracks and pores in the brick aggregate and the interface transition zone between brick aggregate with cement mortar; In addition, nano-Al_2O_3 reacted with cement hydration products to generate more types of hydration products, forming a dense network structure, improving the arrangement of hydration products at the interface, thereby enhancing the compressive strength of brick aggregate concrete [40].

4. CONCLUSIONS

1. The water absorption of recycled brick aggregate decreased with the increase of nano-SiO_2 and nano-Al_2O_3 solution concentrations, and the crush index decreased.
2. Nano-SiO_2 and nano-Al_2O_3 improved the compressive strength of recycled brick aggregate concrete at different ages, the effect of nano-material on the early strength of concrete was more obvious.
3. With the increase of nano-solution concentration, the compressive strength of nano-SiO_2 reinforced brick aggregate concrete at different ages increased first and then decreased, while that of nano-Al_2O_3 reinforced brick aggregate concrete increased.
4. The internal structure of nanomaterials reinforced brick aggregate was denser, micro cracks and pores were filled with hydration products, and the interfacial transition zone was more compact.

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