Research Article

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Effects of nano-SiO₂ modification on rubberised mortar and concrete with recycled coarse aggregates

<https://doi.org/10.1515/ntrev-2022-0024>

received November 10, 2021; accepted December 6, 2021

Abstract: With the recent acceleration of industrialisation and urbanisation, increasing quantities of demolished construction waste and waste tyres are being produced. The production of recycled coarse aggregate (RCA) and rubber particles from this waste, for use as partial or full replacements of normal aggregate in cement concrete, is attracting attention as a solution to the problem of solid waste management. However, the greater incidence of defects in RCA and rubber particles than in normal aggregate limits their application in construction industries. This study evaluated an economic and environmental approach to optimise the performance of rubberised concrete with RCA. Two types of nanomaterials, nano-SiO₂ (NS) solution and NS sol-gel, were used to pretreat RCA and rubber. The effect of the treatment time on the physical properties of the RCA was tested, and the mechanical properties of the rubberised mortar prepared with pretreated rubber were investigated. In addition, a compression test for the rubberised recycled aggregate concrete (RRAC) was designed using the Taguchi method. The effects of four factors (water-cement ratio, rubber content, rubber size, and aggregate treatment) on the stress-strain curve, compressive strength, elastic modulus, specific toughness, and failure patterns of RRAC were also analysed. The results showed that the NS-treated RCA exhibited lower water absorption rate and better mechanical properties. Moreover, the NS-modified rubber enhanced the compressive and flexural strengths

Graphical abstract

of the rubberised mortar by 35 and 17%, respectively. Interestingly, it was found that simultaneous treatment of both RCA and rubber could negatively affect RRAC. Scanning electron microscopy indicated that NS improved the interfacial transition zone separating RCA and rubber from the cement matrix, whereas the pretreated RCA tended to bond with the pretreated rubber in RRAC.

Keywords: recycled coarse aggregate, waste rubber, nano-SiO₂, modification, mechanical properties

1 Introduction

With the recent acceleration of urbanisation, more buildings are being demolished, producing large amounts of demolished construction waste (DCW). It is reported that DCW will reach 27 billion tonnes per year worldwide by 2050 [1]. China produces 2.4 billion tonnes of DCW annually [2]. However, the utilisation rate of DCW is less than 10% in China, implying that the country needs to improve its recycling and reutilisation of DCW. In addition, it is reported that more than one billion tyres are scrapped every year around the world, which further increases the quantity of solid waste [3]. Landfills and

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burning remain the main methods of dealing with waste tyres [4]. DCW and waste tyres are serious ecological threats as they cause soil and water pollution. It is vital to recycle DCW and waste rubber, not only to protect the environment but also to conserve resources.

On the other hand, the consumption of raw materials for concrete production has increased rapidly with the rapid development of construction projects in developing countries, and the demand for more natural aggregates has resulted in a heavy burden on the environment [5]. Using crushed DCW as recycled coarse aggregate (RCA) to produce new environmentally friendly recycled concrete composites and members is of particular interest to maintain natural aggregate and realize waste resources recycling [6–8]. However, due to RCA's high porosity and the presence of aged mortar on its surface, RCA addition degrades the mechanical properties of the concrete [9]. As for waste tyres, they can be crushed into rubber particles and added to concrete as fine aggregates [4,10]. Researchers have suggested that the addition of rubber to concrete improves its energy absorption capacity [11], sound absorption performance [12], and freeze–thaw durability [13]. However, the compressive strength, splitting tensile strength, and bending strength of rubberised concrete decrease, which is attributed to the low strength and high hydrophobicity of the rubber particles [14–16]. To investigate the synergistic effect of RCA and rubber particles in concrete materials and improve the utilisation rate of recycled materials in concrete, rubberised recycled aggregate concrete (RRAC) was developed [17,18]. RRAC is produced by replacing normal coarse aggregates with RCA and partly replacing fine aggregates with rubber. However, RRAC, which contains RCA and rubber, has been reported to exhibit poor static mechanical performance [17,18], which needs to be improved.

It is believed that pretreatment of aggregates plays an important role in improving the properties of RRAC. Many researchers have reported that the presence of aged mortar on the surface of RCA significantly influences the bonding between the RCA and new mortar [19,20]. Some methods, such as heating [21], acid treatment [22], and water washing [23], have been used to remove the attached aged mortar, whereas other methods, such

as cement slurry coating [24], carbonation [25], and chemical solution soaking [26], have focused on strengthening the attached mortar. Moreover, chemical treatment has been shown to modify the surface of rubber to improve the interfacial adhesion between the rubber particles and the cement matrix. The most commonly used chemical agents are NaOH [27], silane coupling agent [28], and acid [29]. Although the abovementioned methods improve RCA and rubber, they suffer from some disadvantages. For instance, some methods involve complicated procedures [30], some introduce harmful ions (Cl^- and SO_4^{2-}) [31], while others produce chemical waste [32], which make them difficult to apply widely in construction engineering.

As green and efficient materials, nanomaterials are widely used in various fields, and their application to cement-based materials has become a popular research topic [33,34]. A number of investigations have been conducted on concrete treated with nanomaterials, among which nano- SiO_2 (NS) is considered the most effective additive [35,36]. Due to its high specific surface area and high reactivity, NS can decrease the porosity of concrete and promote the hydration reaction [37,38]. It has been established that NS addition is also suitable for concrete containing RCA or rubber, as it strengthens the interfacial transition zone (ITZ) between the paste and the aggregates [39,40]. In previous studies, researchers had tended to focus on the effect of NS on the cement matrix, and NS was added to concrete as an additive. It has been reported that RCA and rubber are the weakest parts of RRAC [41]. Effective pretreatment of the aggregates is critical for improving the properties of RRAC, which points to the need for further investigation on direct modification of the aggregates by NS.

The aim of this study is to develop a better understanding of the role of NS in aggregate modification. First, RCA and rubber particles were pretreated with two types of nanomaterial, NS solution and NS sol–gel. The effect of the treatment time on the physical properties of the RCA was tested, and the mechanical properties of the rubberised mortar prepared with the pretreated rubber were investigated using flexural and compression tests. In addition, a cylinder axial compression test for RRAC was designed using the Taguchi method with four factors

Table 1: Properties and chemical composition of OPC

Content						Fineness	Loss on ignition
CaO (%)	SiO ₂ (%)	Fe ₂ O ₃ (%)	Al ₂ O ₃ (%)	SO ₃ (%)	MgO (%)		
62–67	20–24	5–6	4–7	2.12	0.9	1.1	1.7

and three levels. The stress–strain curve, compressive strength, elastic modulus, specific toughness and failure modes of RRAC were analysed. Finally, the microstructures of RCA and the rubber particles were observed using scanning electron microscopy (SEM), and the NS-strengthening mechanism was revealed.

2 Materials

2.1 Cementitious material

Ordinary Portland cement (OPC; P.O.42.5) with a fineness modulus of 1.1 was used as the cementitious material. Its physical properties and chemical composition are shown in Table 1.

2.2 Aggregates

The RCA used was produced by crushed waste concrete, which was obtained from demolished concrete pavement with a compressive strength of 20–30 MPa. It was washed with water to remove impurities and improve its properties. The physical properties of the RCA were tested using the Chinese standard GB/T 14685-2011 [42], and the results are tabulated in Table 2. River sand and rubber particles were used as fine aggregates. Well-graded river sand with a fineness modulus of 2.74 was dried before use. Rubber particles were produced from waste tyres by a factory in Foshan, China. Three rubber sizes – 6 mesh, 60 mesh, and a mixed size ranging from 6 mesh to 60 mesh – similar to the size of river sand were used. The particle size distribution of the aggregates is shown in Figure 1. Figure 2 shows the fine aggregates and their micromorphology.

2.3 Additives

A polycarboxylate superplasticiser (SP) solution was used to adjust the workability of the mixes. The SP solution was diluted to a concentration of 20% from the mother liquor,

Figure 1: Sieve analysis results of aggregates.

which had a solid content of 30.1%. The volume percentage of air in the SP solution was 5%. The properties of the SP solution are shown in Table 3.

2.4 Nanomaterials for modifying aggregates

Two types of nanomaterials, NS solution and NS sol–gel, were used to modify the rubber and RCA. NS solution with 1% NS powder (Nanjing Tansail Co., China) by weight was prepared. The mother liquor of the commercial NS sol–gel (Nanjing Tansail Co., China) had 20% NS by weight. The nanomaterials used are shown in Figure 3, and their properties are shown in Tables 4 and 5.

3 Experimental regime

3.1 Aggregate treatment

Owing to the low solubility and easy agglomeration of NS powder, NS solution with a low concentration of 1% NS

Table 2: Physical properties of aggregates

Aggregates	Size (mm)	Apparent density (kg/m ³)	Bulk density (kg/m ³)	Water absorption rate (%)	Crushing index (%)
RCA	5–20	2,596	1,354	4.82	10.47
River sand	0.075–4.75	2,500	1,780	0.61	—
Rubber	0.25–3.35	1,050	608	—	—

Figure 2: Aggregates used and their micromorphology as revealed by SEM. (a) RCA, (b) river sand (c) 6 mesh rubber, (d) 60 mesh rubber, (e) SEM of sand, (f) SEM of 60 mesh rubber, and (g) SEM of 6 mesh rubber.

powder was manually prepared in this study. In addition, considering the particle size of NS in sol–gel is 5 times bigger than that in the NS powder solution (Tables 4 and 5), the well-dispersed NS sol–gel with a concentration of 20% NS was diluted to a concentration of 5% prior to use.

Figure 4 shows the treatment processes of RCA and rubber particles. Both the RCA and rubber particles were first immersed in NS solution or NS sol–gel, respectively, for 24, 48, and 72 h. Then, they were dried at room temperature before using.

Table 3: Properties of the superplasticiser provided by the manufacturer

Colour	Concentration (%)	Water reduction (%)	Air content (%)	Solid content (%)	Contraction ratio (%)
Light yellow	20	20	3.2	30.1	≥97

Figure 3: Nanomaterials used and SEM of NS powder. (a) NS powder, (b) SEM of NS powder, (c) NS solution, and (d) NS sol-gel.

To investigate the modification effect of RCA by NS treatment, the water absorption rate and crushing index of pretreated RCA were tested and the optimal NS type and treatment time were evaluated, as shown in Figure 4(a).

To investigate the effect of the modified rubber by NS treatment, rubberised mortar was prepared and the

mechanical properties of the rubberised mortar were tested, as shown in Figure 4(b).

Based on the above results of the physical properties of the modified RCA and mechanical properties of the mortar with modified rubber, three aggregate treatment methods, including rubber modification (RM), RCA

Table 4: Properties of NS powder

Size (nm)	Purity (%)	Specific surface area (m ² /g)	Bulk density (g/cm ³)
20	99	200	0.1

Table 5: Properties of NS sol-gel

SiO ₂ (wt%)	Size (nm)	pH	Viscosity (mPa·s)	Density (g/cm ³)
20	100	6–9	<100	1.21

Figure 4: Treatment methods: (a) Treatment methods of RCA; (b) treatment methods of rubber for preparing rubberised mortar; and (c) treatment methods of RCA and rubber for preparing RRAC. *Combined method denotes simultaneous treatment of rubber and RCA.

Table 6: Mix proportions of the mortar mixtures

Mix proportions (unit weight: kg/m ³)						
Group	Water	Cement	Sand	Rubber	NS type	Treatment time
Reference	225	450	1,305	32	—	—
S24	225	450	1,305	32	NS solution	24 h
S48	225	450	1,305	32	NS solution	48 h
S72	225	450	1,305	32	NS solution	72 h
G24	225	450	1,305	32	NS sol-gel	24 h
G48	225	450	1,305	32	NS sol-gel	48 h
G72	225	450	1,305	32	NS sol-gel	72 h

Note: S24 and G24 denote rubber treated by NS solution and NS sol-gel for 24 h, respectively.

modification (AM), and simultaneous modification of rubber and RCA (RM + AM), were designed for preparing RRAC (Figure 4c).

3.2 Mix proportions

3.2.1 Rubberised mortar

To meet the flowability requirements, seven groups of mortar mixtures with a water-cement (w/c) ratio of 0.5 were designed, as shown in Table 6. The 60 mesh rubber particles were used and treated with the NS solution or NS sol-gel for 24, 48, and 72 h. Following the suggestion of previous study [18], the rubber was used to replace 10% of the river sand by volume.

3.2.2 RRAC

The Taguchi method was used to design the mix proportions of RRAC. Four factors – the w/c ratio, rubber content,

rubber size, and processing method for the aggregates – were considered, and each factor comprised three levels. Therefore, an orthogonal array L₉ (3⁴) developed by Taguchi was used to represent a full factorial experiment. The factors and levels used are shown in Table 7. Three w/c ratios were investigated – 0.3, 0.4, and 0.5 – and rubber was used to replace 5, 10, and 15% of the river sand by volume. Three rubber sizes were used: 6 mesh, 60 mesh, and 6–60 mesh (mixed size). As shown in Figure 4, three aggregate treatment methods were used for preparing RRAC, including RM, AM, and RM + AM. Both RCA and rubber were treated by NS sol-gel for 48 h, which was in accordance with the

Table 7: Factors and levels used in the experiment

Levels	Factor A w/c ratio	Factor B Rubber content (%)	Factor C Rubber size	Factor D Aggregate treatment
1	0.3	5	6 Mesh	RM
2	0.4	10	60 Mesh	AM
3	0.5	15	Mix	RM + AM

Table 8: Mix proportions of RRAC used in the experiment

Mix proportions (unit weight: kg/m ³)							
Group	Orthogonal combination	Water	Cement	Sand	RCA	Rubber	SP
0.3–5%-6-RM	A1B1C1D1	185	617	607.2	958.8	12.9	1.9
0.3–10%-60-AM	A1B2C2D2	185	617	575.3	958.8	25.8	1.9
0.3–15%-M-RM + AM	A1B3C3D3	185	617	543.3	958.8	38.8	1.9
0.4–5%-60-RM + AM	A2B1C2D3	185	463	665.8	1051.2	14.2	1.4
0.4–10%-M-RM	A2B2C3D1	185	463	630.7	1051.2	28.4	1.4
0.4–15%-6-AM	A2B3C1D2	185	463	595.7	1051.2	42.5	1.4
0.5–5%-M-AM	A3B1C3D2	185	370	701.1	1,107	14.9	1.2
0.5–10%-6-RM + AM	A3B2C1D3	185	370	664.2	1,107	29.8	1.2
0.5–15%-60-RM	A3B3C2D1	185	370	627.3	1,107	44.8	1.2

synthetical consideration of the performance improvement and economic cost (immersion time). Nine groups of mix proportions were designed, as shown in Table 8.

3.3 Specimen preparation

3.3.1 Rubberised mortar specimen

To examine the compressive and flexural properties of the rubberised mortar, prismatic specimens with length

of 40 mm, width of 40 mm, and height of 160 mm were prepared and divided into seven groups. Each group consisted of a set of three specimens. The specimens were prepared using a 5 L cement mortar mixer by following the steps shown in Figure 5(a): first, the rubber particles were pretreated with NS solution or NS sol–gel and dried at room temperature; then, the cement, river sand, and rubber particles were mixed under dry conditions for 5 min, after which water was added and mixed with the powder for 5 min. Next the mixture was moulded and vibrated for 1 min. After 24 h, it was demoulded and cured at room temperature for 28 days.

Figure 5: Specimen preparation methods: (a) rubberised mortar and (b) RRAC.

3.3.2 RRAC specimen

A 30 L horizontal mixer was used to prepare 27 RRAC cylinder specimens with diameter of 100 mm and height of 200 mm. Figure 5(b) shows the preparation procedure: first, the rubber particles and RCA were pretreated with NS sol-gel for 48 h, and dried at room temperature. Next cement, river sand, and the rubber particles were mixed under dry conditions for 5 min, after which SP and half of total water were added to the mixture and mixed for 3 min. Finally, RCA and the remaining water were added, and the mixing was continued for 5 min. After mixing, the mixture was moulded and vibrated for 2 min. After 24 h, it was demoulded and cured at room temperature for 28 days.

3.4 Experimental methods

3.4.1 Tests for RCA properties

The water absorption rate and crushing index of RCA after treatment were tested in accordance with the Chinese standard GB/T 14865-2011 [42]. The water absorption rate of RCA was calculated using equation (1):

$$W = \frac{G_1 - G_2}{G_2}, \quad (1)$$

where W is the water absorption rate of RCA; G_1 is the mass of the aggregate sample under the saturated surface dry condition; and G_2 is the mass of the dried RCA.

The crushing index of RCA was calculated using equation (2):

$$Q_e = \frac{M_1 - M_2}{M_1}, \quad (2)$$

where Q_e is the crushing index of RCA; M_1 is the mass of the aggregate sample; and M_2 is the residual mass of the RCA after crushing.

3.4.2 Prism flexural and compression tests

The prism flexural test of the mortar specimens was conducted in accordance with the Chinese standard GB/T 17671-1999 [43], using a YAW-300C machine with the loading speed set to 50 N/s, as shown in Figure 6. After the flexural test, the compression test was conducted on a lateral section of the broken prism with a compression area of 40 mm × 40 mm. The compression test was also

Figure 6: Prism flexural and compression test machine.

conducted using the YAW-300C machine with the loading speed set to 2,400 N/s.

3.4.3 Cylinder axial compression test

The cylinder axial compression test was conducted by the Chinese standard GB/T 50081-2019 [44] using a Matest

Figure 7: Cylinder compression test machine.

Figure 8: Effect of treatment time on the water absorption rate of RCA.

material testing machine, which has a capacity of 4,000 kN. The loading speed was set to 0.12 mm/min. Loading was stopped when the stress decreased to 10% of the peak stress. Two linear variable displacement transducers were used to measure the axial strain of the specimen. In addition, two strain gauges were attached to the middle of the cylinder to measure the hoop strains of the specimen, as shown in Figure 7. Before testing, plaster with a compressive strength of 800 MPa was used to level the surface of the cylinder to eliminate load eccentricity. Both the load data and strain data were collected by a high-speed acquisition system at a frequency of 1 Hz.

Figure 9: Effect of treatment time on the crushing index of RCA.

Figure 10: Effect of treatment time on compressive strength of rubberised mortar.

3.4.4 SEM test

To observe the microstructure of the specimen after the compression test, SEM test was performed using an S3400N machine with a working distance of 4 mm and an accelerating voltage of 15 kV. The samples, which were obtained from the fracture surfaces of the specimens, were less than 10 mm × 10 mm × 10 mm in size. The samples were gold coated and tested under high vacuum.

Figure 11: Effect of treatment time on flexural strength of rubberised mortar.

Table 9: Compression test results of RRAC

Group	w/c ratio	Rubber content (%)	Rubber size (mesh)	Aggregate treatment	Compressive strength (MPa)	Elastic modulus (GPa)	Specific toughness (%)
0.3-5%-6-RM	0.3	5	6	RM	51.91	49.10	0.37
0.3-10%-60-AM	0.3	10	60	AM	46.76	40.03	0.45
0.3-15%-M-RM + AM	0.3	15	Mix	RM + AM	32.21	30.38	0.43
0.4-5%-60-RM + AM	0.4	5	60	RM + AM	31.88	30.00	0.44
0.4-10%-M-RM	0.4	10	Mix	RM	18.68	30.42	0.70
0.4-15%-6-AM	0.4	15	6	AM	31.32	46.35	0.38
0.5-5%-M-AM	0.5	5	Mix	AM	23.19	24.61	0.82
0.5-10%-6-RM + AM	0.5	10	6	RM + AM	17.75	29.06	0.68
0.5-15%-60-RM	0.5	15	60	RM	18.38	20.45	0.65

4 Result and discussion

4.1 Effect of treatment on water absorption rate and crushing index of RCA

Figure 8 shows the effect of the treatment time on the water absorption rate of RCA. Increasing the treatment time significantly reduced the water absorption rate of RCA. The water absorption rate of RCA treated with NS solution decreased linearly as the treatment time increased. With a treatment time of 72 h, the water absorption rate of RCA decreased by over 30%. This result can be explained by the pozzolanic reaction between the NS particles and $\text{Ca}(\text{OH})_2$ and the filling effect of the NS particles, which densified the microstructure of RCA. The water absorption rate of RCA treated with NS sol-gel was much lower than that treated with NS solution. For instance, when RCA was treated with NS sol-gel for 24, 48 and 72 h, the water absorption rate was decreased by 28, 32, and 56%, respectively. This is because a layer of silica gel from the well-dispersed NS sol-gel was formed homogeneously on the surface of RCA, leading to the attachment of considerable number of agglomerated NS particles [39,45].

As shown in Figure 9, NS also greatly influenced the crushing index of RCA. After treatment with either NS solution or NS sol-gel, the crushing index decreased approximately linearly with increasing treatment time. After treatment with NS sol-gel, the crushing index of RCA decreased by approximately 10% more than that after treatment with NS solution at same treatment time on average. The improvement in the crushing index of RCA treated with NS sol-gel is attributed to the reaction of NS particles with aged

Figure 12: Stress-strain curve of RRAC.

mortar to form C–S–H gel, which increased the strength of the RCA. Previous studies also reported that the average microhardness of the aged mortar at the surface layer of RCA can be enhanced after NS treatment [39,45].

4.2 Effect of treatment on compressive and flexural strength of rubberised mortar

The effect of NS treatment on the compressive strength of the rubberised mortar is shown in Figure 10. The compressive strength of the rubberised mortar increased with the treatment time. For instance, after treatment with NS solution for 72 h, the compressive strength of the rubberised mortar was 23.93 MPa, an increase of 35% compared with that of untreated mortar. This can be attributed to the consumption

and refinement of $\text{Ca}(\text{OH})_2$ crystals by NS, making the rubber particles and cement paste connect closely with the C–S–H crystal to form a more compact concrete structure [46,47].

A similar improvement was also found in the rubberised mortar treated with NS sol–gel. Moreover, treatment of rubber with NS sol–gel produced a slight increase in the compressive strength of the rubberised mortar compared with that in the NS solution treatment. A possible reason is that NS sol–gel attaches more evenly on the rubber surface. Another interesting finding is that when the treatment time was changed from 48 to 72 h, the increase in the compressive strength slowed down, demonstrating that 48 h treatment is sufficient to improve its compressive strength if time cost needs to be considered.

Figure 11 demonstrates the effect of NS treatment on the flexural strength of the rubberised mortar. With

Figure 13: Stress–strain curve of RRAC with different w/c ratios of: (a) 0.3, (b) 0.4, and (c) 0.5.

treatment times of 24, 48, and 72 h, the flexural strength of the rubberised mortar increased by 12, 14, and 17% after treatment with NS solution, respectively. It has been reported that the addition of rubber can improve the toughness of the mortar. However, due to the hydrophobicity of the rubber particles, a substantial amount of air enters the mortar. As a result, the number of pores increases, and the bonding between rubber and the cement matrix is weakened [46,47]. The NS attached to the surface of the rubber significantly improved the hydrophilic property.

The type of NS material showed only a slight influence on the flexural strength of the rubberised mortar: NS sol-gel performed a little better than NS solution. For instance, with a treatment time of 24 h, there was no difference between the flexural strengths of rubberised mortar

treated with NS sol-gel and with NS solution. A possible reason for the indistinguishable effects of the two treatments at 24 h is that treatment time is too short for NS sol-gel to adhere to the rubber particles effectively. In addition, for the treatment time of 48 and 72 h, there is no change in the flexural strength of NS sol-gel-treated rubberized mortar. This could be attributed to the reason that the rubber was covered by a layer of silica gel, which prevented nanoparticles from continuing to adhere to the rubber particles, thus limiting the improvement in flexural strength. In general, the mechanical properties of the rubberised mortar improved dramatically after rubber was treated with NS sol-gel. However, a turning point was reached (48 h for NS sol-gel) beyond which the improvement in the properties of rubberised mortar with increasing treatment time slowed down. Therefore, considering time

Figure 14: Stress–strain curve of RRAC with different rubber contents of: (a) 5%, (b) 10%, and (c) 15%.

cost, 48 h was regarded as the optimum time, and it was the treatment time used with RRAC. However, for NS solution, the turning point of the treatment time occurred at 24 h, which is mainly attributed to the lower concentration and poorer dispersion of NS solution compared with NS sol-gel.

4.3 Effect of orthogonal factors on RRAC

The compressive properties of the tested RRAC specimens are listed in Table 9, including compressive strength, elastic modulus, and specific toughness. It is noted that these results in Table 9 are the average values of three same specimens.

4.3.1 Stress-strain curve

The axial compressive stress-strain relations of the RRAC specimens are shown in Figure 12. It is observed that similar to the stress-strain curve of OPC concrete, the curve of RRAC mainly consists of an elastic ascending section and an inelastic descending section. The incorporation of rubber led to a flatter curve in the ascending section compared with the curve of recycled concrete in a previous study [48].

Figure 13 shows that the w/c ratio greatly influences the stress-strain curve of RRAC. Expectedly, as the w/c ratio decreased, the curve of the ascending section became steeper, and the peak stress increased, indicating that a lower w/c ratio can significantly improve the compressive

Figure 15: Stress-strain curve of RRAC with different rubber sizes: (a) 6 mesh rubber, (b) 60 mesh rubber, and (c) mixed-size rubber.

strength and stiffness of RRAC, but at the expense of greater brittleness. The descending section of the curve showed a flatter trend with increasing w/c ratio. A possible explanation for this is that a softer cement matrix makes it easier for the toughness of rubber to manifest itself.

Figure 14 shows the effect of the rubber content on the stress–strain response of RRAC. In general, in the ascending section of the curve, which corresponds to greater rubber content, RRAC exhibited a lower peak stress, implying that the addition of rubber decreased the strength of RRAC. Comparing Figure 14(a) and (c), it is clear that with increasing rubber content, the descending section of the stress–strain curve of RRAC became flatter. This result is consistent with the characteristics of rubberised concrete with and without the

recycled aggregate [49,50], which are caused by the greater toughness of rubber compared with river sand. The figure also shows that the hoop strain of RRAC increased with higher rubber content, proving that the weaker aggregate tended to produce more ductile concrete than the stronger aggregate.

Figure 15 shows the stress–strain curve of RRAC with different rubber sizes. In the ascending part of the curve, with the same unit stress increase, the strain developed in RRAC with 60 mesh rubber was higher than that developed in RRAC with 6 mesh rubber. That is, smaller rubber particles decreased the elastic modulus of RRAC. However, no significant change was found in the descending stage of the stress–strain curves of RRAC with 6 mesh and 60 mesh rubber. In addition, the inclusion of mixed-size rubber

Figure 16: Stress–strain curves of RRAC with different aggregate treatment methods: (a) RM, (b) AM, and (c) RM and AM.

particles lowered the peak stress and flattened the descending section of the stress–strain curve. These results indicate that well-graded rubber particles decreased the strength of RRAC, but absorbed energy more effectively after the peak stress.

The type of aggregate treatment method is also an important factor for the stress–strain curve of RRAC, as shown in Figure 16. A comparison of Figure 16(a) and (b) indicates that RM improved the toughness of RRAC and AM improved its strength, which is consistent with the results of earlier studies on RCA concrete and rubberised concrete [51,52]. However, the curves of RRAC with RM and AM simultaneously display neither of the abovementioned features, as shown in Figure 16(c).

4.3.2 Compressive strength

To investigate the effect of orthogonal factors on the compressive strength of RRAC, Figure 17 presents the average compressive strength of the specimens with the same level of orthogonal factor. Similar analysis is used hereinafter for the elastic modulus and specific toughness of the RRAC.

Figure 17(a) shows that the compressive strength of RRAC decreased rapidly as the w/c ratio was increased, which has also been reported for rubberised concrete without RCA [53]. The compressive strength of RRAC decreased by 37 and 55%, respectively, as the w/c ratio was increased from 0.3 to 0.4 and 0.5. It is believed that

Figure 17: Effects of orthogonal factors on compressive strength of RRAC. (a) Water–cement ratio, (b) rubber content, (c) rubber particle size, and (d) treatment method.

RRAC is sensitive to the influence of the w/c ratio, which is attributed to the high water absorption rate of RCA [54].

Figure 17(b) shows that the rubber content decreased the compressive strength of RRAC. This reduction was mainly caused by the low strength and stiffness of the rubber particles, and the weak bonding between rubber and the cement paste. However, the compressive strength of RRAC did not decrease linearly with the increasing rubber content. Changing the rubber content from 10% to 15% had little effect on the compressive strength. This is in line with the results obtained from the rubberised concrete without RCA [14,55].

Figure 17(c) shows the influence of the rubber size on the compressive strength of RRAC. For single-size rubber

particles, the compressive strength of RRAC decreased slightly as the rubber size was decreased. This is mainly because the rubber particles act as a solid hydrophobic air-entraining agent, and smaller rubber particles with a larger specific surface area have better gas adsorption capacity [56]. However, the compressive strength of RRAC with mixed-size rubber particles was significantly lower than that of RRAC with single-size rubber particles, possibly because well-graded rubber particles tend to aggregate and form defects in concrete.

Figure 17(d) shows that among the three aggregate treatment methods, AM has the best effect on compressive strength. However, simultaneous treatments with RM and AM decreased the compressive strength. This could be because

Figure 18: Effect of orthogonal factors on elastic modulus of RRAC. (a) Water–cement ratio, (b) rubber content, (c) rubber size, and (d) aggregate treatment.

Figure 19: Effect of orthogonal factors on specific toughness of RRAC. (a) Water-cement ratio, (b) rubber content, (c) rubber size, and (d) aggregate treatment.

the NS molecules remaining on the surface of the rubber and RCA tended to aggregate, which made it easier for rubber particles to adhere to the surface of RCA. Thus, weak areas were produced, which reduced the bonding between the aggregates and mortar.

4.3.3 Elastic modulus

The effects of the orthogonal factors on the elastic modulus of RRAC are shown in Figure 18. According to the Chinese standard GB/T 50081-2019 [44], the elastic modulus of the tested

Table 10: Standard error of three levels of each factor

Factors	w/c ratio	Rubber content	Rubber size	Aggregate treatment
Compressive strength	11.63	4.72	4.85	3.59
Rank	1	3	2	4
Elastic modulus	7.80	1.10	7.09	3.59
Rank	1	4	2	3
Specific toughness	0.15	0.06	0.09	0.03
Rank	1	3	2	4

Table 11: Optimal level of the orthogonal factors

Factors	w/c ratio	Rubber content (%)	Rubber size	Aggregate treatment
Compressive strength	0.3	5	6 Mesh	AM
Elastic modulus	0.3	5	6 Mesh	AM
Specific toughness	0.5	10	Mixed size	RM

specimen was defined as the secant modulus of its stress–strain curve at the point of the 1/3 peak stress. As shown in Figure 18(a), expectedly, the elastic modulus of RRAC decreased by 10 and 38% as the w/c ratio was changed from 0.3 to 0.4 and 0.5, respectively, which is consistent with a previous investigation on the rubberised concrete [53]. The mortar matrix became softer because it contained more water, resulting in a relative incompact system.

Figure 18(b) shows the effect of rubber content on the elastic modulus of RRAC. The addition of rubber slightly decreased the elastic modulus of RRAC, as rubber particles have a lower elastic modulus than sand. This makes the elastic modulus of rubber particles and mortar different, which facilitates crack formation [57].

Figure 18(c) shows that, as with the compressive strength, the elastic modulus of RRAC decreased with smaller rubber size. However, rubber size had a greater influence on the elastic modulus than on the compressive strength. Moreover, as with the compressive strength, the elastic modulus of RRAC with mixed-size rubber particles was lower than that with single-size rubber particles. This can be explained by the aggregation of rubber particles, which was also the main reason for the decreased compressive strength with mixed-size rubber particles described earlier.

Figure 18(d) shows the influence of aggregate treatment methods on the elastic modulus of RRAC. RRAC with AM exhibited the highest elastic modulus, indicating that RCA could play an important role in determining the elastic modulus of RRAC. However, simultaneous treatment of RRAC with RM and AM decreased the elastic modulus. This decrease is attributed to the formation of weak zones, as mentioned earlier when discussing the compressive strength.

4.3.4 Specific toughness

Toughness is generally used to characterise the energy dissipation capacity of concrete under uniaxial compression, which is calculated as the area surrounding the stress–strain curve of a concrete specimen [58]. Furthermore, the specific toughness is defined as the ratio of the

area under the stress–strain curve to the compressive strength. Table 9 and Figure 19 show the calculation results for the specific toughness of RRAC.

As shown in Figure 19(a), the w/c ratio clearly influences the specific toughness of RRAC. RRAC specimens with w/c ratios of 0.4 and 0.5 had 21% and 72% higher specific toughness values, respectively, than RRAC with a w/c ratio of 0.3. These observations are explained by the increased brittleness of RRAC at lower w/c ratios, which weakens its deformation capacity.

Figure 19(b) shows that as the rubber content was increased, the specific toughness of RRAC first increased and then decreased, which agrees with results obtained from previous studies [49]. The main reason for the increase in the specific toughness is the excellent energy absorption capacity of the rubber particles. However, high rubber content reduces the density of RRAC and leads to hole formation, which decreases its energy absorption capacity [57].

Figure 19(c) shows the effect of rubber size on the specific toughness of RRAC. Rubber size had only a slight influence on the specific toughness of RRAC. However, RRAC with mixed-size rubber had a higher specific toughness than RRAC with single-size rubber, possibly because mixed-size rubber particles have greater potential to absorb energy.

Figure 19(d) shows that among the three aggregate treatment methods, RM produced the highest specific toughness, indicating that rubber plays an important role in the toughness of RRAC. However, simultaneous treatment of RRAC with RM and AM resulted in a lower specific toughness.

4.3.5 Comparison of orthogonal factors on RRAC

The standard error of three levels was used to characterise the influence of the orthogonal factors on the properties of RRAC. The calculated standard errors of three levels of each factor are shown in Table 10. The w/c ratio is the most influential factor for the mechanical properties of RRAC, followed by rubber size. Rubber content has greater influence on the compressive strength and

Figure 20: Failure patterns of RRAC. (a) 0.3–5%–6-RM, (b) 0.3–10%–60-AM, (c) 0.3–15%–M-RM + AM, (d) 0.4–5%–60-RM + AM, (e) 0.4–10%–M-RM, (f) 0.4–15%–6-AM, (g) 0.5–5%–M-AM, (h) 0.5–10%–6-RM + AM, and (i) 0.5–15%–60-RM.

Figure 21: Comparison of microstructures of RRAC with and without rubber modification treatment: (a and b) with treatment and (c and d) without treatment.

specific toughness of RRAC than aggregate treatment; however, aggregate treatment has greater influence on the elastic modulus.

The optimal level of the factors is shown in Table 11. It can be seen that the orthogonal combination of 0.3–5%–6-AM is the optimal combination for the compressive strength and elastic modulus of RRAC. In addition, RRAC with a w/c of 0.5 and a rubber content of 10% has the highest specific toughness, and mixed-size rubber particles and RM can also improve the specific toughness of RRAC.

4.3.6 Failure modes

The failure patterns of the specimens are shown in Figure 20. With low w/c ratios, RRAC tended to break into large pieces, whereas under high w/c ratios, a number of small pieces peeled off from RRAC. The greater stiffness of RRAC at lower w/c ratios caused more brittle failures. In addition, it is believed that RCA content is responsible for an important effect on the failure pattern of RRAC. As the rubber content

was increased, multiple thin cracks were observed on the surfaces of the specimens, which were mainly caused by the low elastic modulus of rubber. Rubber particles enhance the capacity for deformation before cracking and inhibit the propagation of micro-cracks [49]. No obvious differences in RRAC behaviour were observed with single-size rubber. However, RRAC with mixed-size rubber tended to produce long, wide cracks that penetrated the specimen. Another interesting observation was that some rubber particles connected closely with the matrix on the failure surface of RRAC with RM treatment, whereas rubber particles were found to peel off the failure surface of RRAC without RM treatment. This finding has evidenced that rubber treatment by NS can effectively improve the ITZ zone of rubber and the cement matrix.

4.3.7 SEM test

Figure 21(a)–(d) shows the microstructures of RRAC with and without RM treatment. A comparison of these figures

Figure 22: Comparison of microstructures of RRAC with and without RCA modification treatment: (a and b) with AM treatment and (c and d) without AM treatment.

shows that rubber particles without treatment bonded poorly with the cement matrix, and partially or completely peeled off. Cracks extended along the ITZ, which was the main reason for failure of the specimen. After RM treatment, the rubber particles connected closely with the cement matrix, resulting in a denser RRAC structure. The improvement in the ITZ between the rubber and mortar improved the mechanical properties of RRAC.

The microstructures of RRAC with and without AM treatment are shown in Figure 22(a)–(d). The images show that without AM treatment, the structure of RRAC is loose. In addition, numerous wide cracks appeared around the ITZ. NS treatment improved the ITZ of RCA, and the cracks tended to occur in the cement matrix. Further, the surface of RCA became smoother with fewer pores, indicating that NS filled the holes of RCA effectively.

Figure 23: Microstructures of RRAC with simultaneous RCA modification and rubber modification treatments (a and b).

Figure 23(a) and (b) shows the microstructures of RRAC treated simultaneously with AM and RM. Rubber particles and RCA tended to aggregate, which significantly reduced the strength of RRAC. Cracks extended along the interface between RCA and rubber, finally penetrating the cement matrix and connecting them, which is attributed to the degraded mechanical properties of RRAC due to the simultaneous AM and RM.

5 Conclusion

An experimental study was conducted to investigate the effects of nano-SiO₂ modification on RCA and rubber, aiming to improve the performance of the rubberised concrete with RCA. Two types of nanomaterials, NS solution and NS sol-gel, were used to pretreat RCA and rubber by immersion. The physical properties of RCA were tested. Mechanical and microstructural tests were conducted on rubberised mortar and RRAC modified with three methods, and the following are the conclusions from the test results:

- 1) NS treatment significantly improved the physical properties of RCA. The water absorption rate and crushing index of RCA decreased after treatment with NS solution and NS sol-gel, with NS sol-gel exhibiting a stronger effect than NS solution. With 48 h treatment, water absorption rate decreased by 32 and 23%, and the crushing index decreased by 39 and 29% for NS sol-gel and NS solution, respectively.
- 2) NS treatment of rubber improved the compressive strength and flexural strength of the rubberised mortar. This is attributed to the consumption and refinement of Ca(OH)₂ crystals by NS attached to the surface of the rubber, leading to a close connection between the rubber and the matrix. The NS treatment time of rubber did not linearly contribute to the mechanical properties of the rubberised mortar. It is suggested that based on a synthetical consideration of the performance improvement and economic cost, rubber treated with NS sol-gel for 48 h is the optimal method for improving the compressive strength and flexural strength of the rubberised mortar.
- 3) Treatment with NS modified the ITZ between RCA, rubber, and the matrix effectively. The rubber particles and RCA combined closely with the cement matrix, leading to a compact RRAC microstructure. However, simultaneous treatment of rubber and RCA created a weak zone, where rubber and RCA tended to aggregate. Cracks extended along the interface of RCA and rubber, penetrating the cement matrix.
- 4) The addition of rubber might significantly reduce the mechanical properties of RRAC. The results indicate that RRAC with a modified rubber content no more than 10% could display good compressive behaviour. In addition, single-size rubber particles rather than mixed-size rubber particles are suggested to be used for maintaining the compressive strength of RRAC, and bigger rubber size is beneficial to the mechanical properties of RRAC.
- 5) Among the three aggregate treatment methods of RRAC, RM increased the toughness of RRAC, whereas AM increased its compressive strength and elastic modulus. However, simultaneous application of RM and AM treatments degraded the mechanical properties of RRAC, mainly because the NS molecules remaining on the surface of rubber and RCA tended to aggregate. Thus, NS modification with simultaneous application of RM and AM cannot be recommended.
- 6) It is suggested that for the rubberised concrete without RCA, rubber modification with NS sol-gel can effectively improve the mechanical properties of the concrete. As for the rubberised concrete with RCA, only RCA modification with NS sol-gel was the recommended treatment method for increasing the mechanical properties of the concrete.

As the cost of nanomaterials continue to drop with the progress of nanotechnology, NS modification has great potential for use in aggregate modification, as it is an efficient and environment-friendly method. However, the effect on the aggregate depends on the type of nanomaterial used, and more investigations on various nanomaterials and aggregate modification methods will be conducted in the future.

Acknowledgements: The authors gratefully acknowledge the financial support provided by the National Natural Science Foundation of China and the Guangdong Basic and Applied Basic Research Foundation.

Funding information: This study was funded by the National Natural Science Foundation of China (Nos. 12072078 and 11672076) and the Guangdong Basic and Applied Basic Research Foundation (No. 2019B151502004).

Author contributions: All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Conflict of interest: The authors state no conflict of interest.

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