



Review

Methods for improving the microstructure of recycled concrete aggregate: A review

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HIGHLIGHTS

- Defects in RACs affect their microscopic and macroscopic properties.
- Methods for improving the microstructure of RCA are discussed.
- The advantages and disadvantages of methods are discussed.
- Methods are discussed from the aspects of environment, RCA particle size and application.

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ABSTRACT

Recycled concrete aggregate (RCA) differ from natural aggregate as the former contains attached mortar. Given the recycling process of RCA, that is, by crushing, the microstructure presents several disadvantages, such as porosity, micro-cracks, as well as weak interface transition zones (ITZs), which will also damage the mechanical properties and durability of recycled aggregate concrete (RAC). According to current literature, the microstructure of RAC is mainly affected by ITZs and the porosity of RCA. This paper summarises the principles, advantages and disadvantages of the methods for improving the microstructure of RCA from four aspects, such as removal of attached mortar in the RCA, surface coating on RCA, different mixing methods and CaCO₃ precipitation. Methods are also discussed from the aspects of environment, RCA particle size and application, which provide readers with a reference for improving the microstructure of RCA.

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1. Introduction

Since the beginning of the 21st century, with the rapid development of urbanisation and industrialisation, the construction industry has consumed large amounts of natural resources and produced large amounts of construction and demolition waste [1]. The impact on the environment according to Oikonomou [2] is "Take 50% of raw materials from nature, consume 40% of total energy, and create 50% of total waste"; and construction became the industry with the largest impact on the environment [3].

Concrete is widely used in the construction industry because of its low price, wide application and the ability to take any shape [3,4]. It is a composite material prepared from a certain proportion of cementitious materials, water, aggregates and admixtures [5]. The construction industry produces about 25 billion tons of concrete annually worldwide [6]. Developing countries, such as China and India, which are among the top 10, are the biggest consumers of concrete [3,5].

The massive use of concrete has created a series of problems, such as the massive consumption of natural resources and excessive amount of construction waste, which lead to a shortage in landfill sites; the cost of solving these problems is high [7]. In recent years, an increasing number of experts and scholars have begun to pay attention to the sustainable development of concrete, such as maximising concrete durability, conservation of materials, use of waste and supplementary cementing materials and concrete recycling; thus, recycled aggregate concrete (RAC) has begun to receive increasing attention as a type of concrete recycling [8]. Aggregates generally account for 60%-75% of the concrete volume [9,10]. Natural aggregates (NA) are generally not produced in a short time, and the demolition of buildings produces a large amount of waste concrete; thus, waste concrete is recovered, crushed and mixed in a certain proportion and then used as recycled concrete aggregate (RCA) to replace NA completely or partially to produce RAC [3,11,12]. Research shows that using RCA in concrete preparation does not only satisfy the performance requirements of natural structural concrete, but also solve the landfill shortage problem, which is consistent with the essence of sustainable development, namely, protecting the environment and the rapidly decreasing resources [12-14].

Studies have highlighted the shortcomings of RAC. Given the porosity of RCA, RAC is usually produced by dismantling and crushing waste concrete [5,15]. In comparison with natural aggregate concrete (NAC), the mechanical properties of RAC, such as compressive strength and tensile splitting strength decreased by 25% and 10% respectively, and shrinkage deformation and creep deformation increased by 25% and 30% respectively [5,16,17]. The biggest difference between them is in their absorption capacity [3,18,19]. Given the high porosity of attached mortar, the water absorption capacity of RCA is 2.3-4.6 times that of NA [20]. The water absorption of RCA increased with the decrease of particle size [21,22]. The performance of concrete depends on the properties of the aggregates used. Thus, Shi et al. [9] summarised methods for improving RCA performance by removing or strengthening the attached mortar. The performance of concrete is determined by its microstructure [23,24]. Therefore, improving the microscopic properties of RAC can also improve its macroscopic properties.

2. Microstructure of RAC

NAC is a composite material including NA and cement paste, whereas interfacial transition zone (ITZ) is an interphase in the microstructure between cement paste and NA [3]. RAC is a composite material that replaces the NA in whole or in part with RCA [5,25]. Given that RCA is composed of NA and old cement mortar, so RAC has three ITZs, as follows [26-28]:

1. ITZ between the NA and old cement matrix (Old ITZ),
2. ITZ between the NA and new cement matrix (New ITZ),
3. ITZ between the new and the old cement matrices (New ITZ).

This view is consistent with that suggested for the microstructure of the ultra-high-performance concrete prepared by Zhang et al. [28] using fine recycled concrete aggregate (FRCA: 0-5 mm) and natural fine aggregate (NFA: 0-5 mm), as shown in Fig. 1(a). ITZ₁ lies between the NFA and new cement matrix, ITZ₂ lies between the NFA and old cement matrix and ITZ₃ lies between the new and old cement matrices. Fig. 1(b) presents the ITZ for ultra-high performance concrete prepared by NFA. The microstructure of NAC differs from that of RAC in aggregate and ITZ. Nanoindentation, Scanning electron microscopy (SEM) and Energy-dispersive X-ray spectroscopy were used to study the microstructure of concrete [29]. Nanoindentation method can be used to observe the change in concrete structure at the nanoscale and determine the width and modulus of elasticity of ITZs [30-32]. Almost all SEM instruments are equipped with Energy-dispersive X-ray spectroscopy can be used to measure the concrete hydration

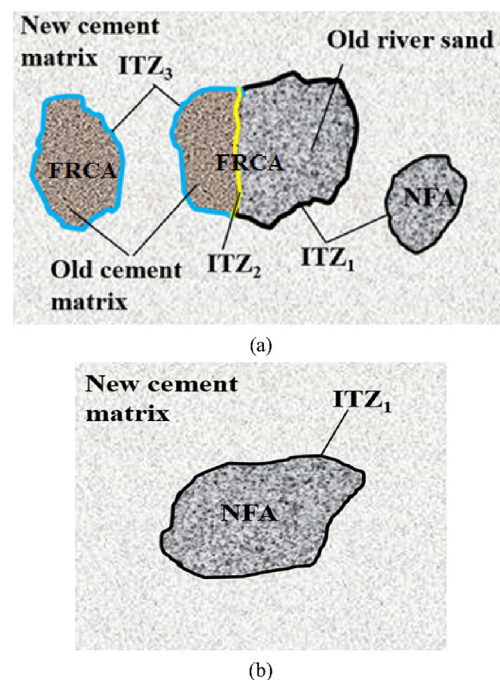


Fig. 1. Different types of ITZs contained in ultra-high-performance concrete prepared with (a) FRCA and NFA; (b) NFA [28].

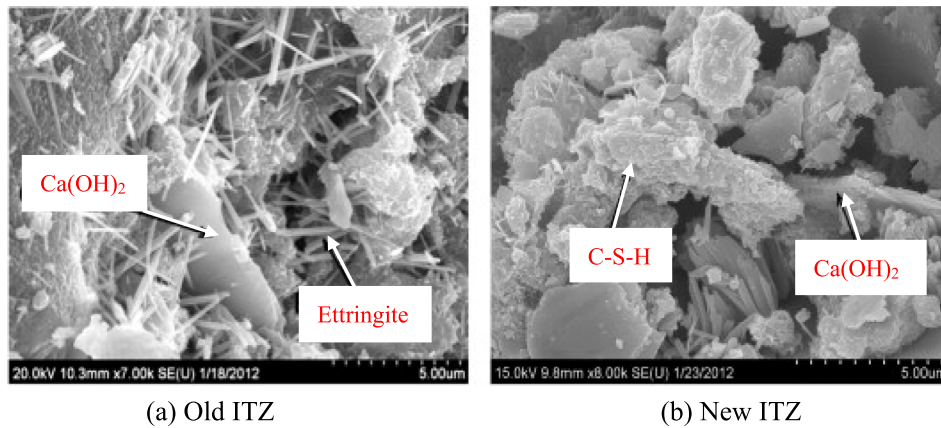


Fig. 2. SEM images of ITZs in RAC [39]. (a) Old ITZ (b) New ITZ.

of cement particles, pore size, shape distribution and content of calcium hydroxide ($\text{Ca}(\text{OH})_2$) and C-S-H on the micron scale [33].

When NAC is subjected to compression or tension, microcracks pass through the porous zone called ITZ near the aggregate, and then spread to the aggregate causing concrete to fail [34,35]. ITZ originated from the “wall effect”, i.e. small cement particles close to a large particle (or aggregates) cannot be packed as dense as the maximum bulk packing density of the small cement particles [36,37]. Li et al. [38] found that ITZs can be perceived to consist of four stages, namely, porosity, $\text{Ca}(\text{OH})_2$, other hydration products (mainly C-S-H gel and ettringite) and unhydrated cement particles. Ettringite usually exists in small amounts in ITZs. C-S-H is smaller than $\text{Ca}(\text{OH})_2$, which easily makes ITZs dense [38]. The volume fraction of C-S-H in the new ITZ exceeds that in the old ITZ, as shown in Fig. 2. The FRCA has higher porosity and water absorption than the coarse recycled concrete aggregate (CRCA), resulting in a higher degree of hydration [39]. The characteristics of ITZs depend on the quality of the attached mortar of the RAC. However, the quantity of the attached mortar has no effect on the ITZ characteristics [40]. The thickness of ITZs depends on the type of aggregate, and its mechanical properties can be reflected by microhardness and elastic modulus [40]. For NAC, the thickness of the ITZ usually ranges from 9 μm to 51 μm . For RAC, the thickness of the ITZs usually ranges from 30 μm to 60 μm [41,42]. In the work of Xiao et al. [32], the thickness of the new ITZ was 55–65 μm and that of the old ITZ was 40–50 μm .

These three ITZs seem to have properties that affect RAC in different ways. The high porosity and cracks of the old ITZ increase the demand for water and reduce the amount of water in the new ITZ [41,43]. The reason is that water is absorbed by the attached cement matrix of the RCA and may weaken the hydration process. However, the absorbed water can accelerate the late hydration process by reducing the self-drying of the new cement matrix, thereby densifying the ITZs [44,45]. Otsuki et al. [40] found that the strength of ITZs depends on their water-binder ratio. Under the condition of low water-binder ratio, the old ITZ affects the properties of RAC. The new ITZ has an effect under a high water-binder ratio. Therefore, under a high water-binder ratio of RAC, if the old ITZ is stronger than the new ITZ, then the strength of RAC is equal to that of NAC. Under a low water-binder ratio, if the old ITZ is weaker than the new ITZ, then the strength of RAC is lower than that of NAC [3,40]. Therefore, it is accepted that ITZs have influences on the strength of RAC [32]. Scholars have two opposing views on ITZ's influence on NAC properties. Nemati et al. [46] and Wong et al. [47] believed that microcracks initiate and expand at ITZ when NAC is subjected to compression load; thus, ITZ is considered the weakest link. Microcracks and high porosity cause decrease in mechanical properties and increase in

permeability of concrete. By contrast, Diamond et al. [48] did not find 30% porosity in ITZ by analysing the binary segmented images of ITZ, which was only slightly higher than that of cement paste. They believed that some pores were filled by $\text{Ca}(\text{OH})_2$ and C-S-H; hence, there had no clear reason to believe that ITZ had apparent negative effects on the permeability and mechanical properties of NAC.

The microstructure of the cement matrix consists of two structural units: phenograins and groundmass. Phenograins include unhydrated particles, partially hydrated particles, fully hydrated particles and empty shell hydrated particles., and the groundmass consists of small particles embedded in the pores, and a network of groundmass and pores confines the partial porosity of the cement paste [23]. When the content of calcium silicate and calcium aluminate in cement is relatively high, additional $\text{Ca}(\text{OH})_2$ and ettringite crystals will be generated at the initial stage of hydration to adhere to the surface of RCA. This process increases the porosity of the RCA surface and makes the microstructure of RAC loose and porous, thus affecting RAC properties. In the later stage of hydration, numerous C-S-H gels and $\text{Ca}(\text{OH})_2$ crystals were generated, and the number of ettringite crystals and pores decreased, thereby making the microstructure of RAC uniform and compact [49].

FRCA has an irregular shape, high water absorption rate and impurity content and RAC prepared using FRCA has large shrinkage and permeability, but the problems of CRCA are small [27,39]. RCA has lower density, higher porosity and higher water absorption capacity than NA due to the existence of attached mortar, whereas the number of attached mortars increases with the decrease in RCA particle size [18,39,50–52]. Figs. 3 and 4 showed the surfaces of NA and RCA, respectively. The surface of NA is smooth and has few pores, whereas the surface of RCA has many irregular pores and loose particles, which may participate in or affect the hydration reaction of cement and the formation of $\text{Ca}(\text{OH})_2$ and ettringite, thus affecting the properties of RAC [4,22,53]. Guedes et al. [4] found that the $\text{Ca}(\text{OH})_2$ and ettringite content and porosity of FRCA, compared with NFA, increased. However, the hydration products can reduce the amount of unhydrated cement in ITZs and fill the pores of ITZs [4,54]. Khalaf et al. [55] indicated the size of cracks and pores on the surface of and inside the CRCA is different, the cement paste cannot penetrate the aggregate due to its viscosity, and the water is easily absorbed by CRCA; thus, the bonding between CRCA and the cement paste is poor. Moreover, water enters into the pores of CRCA, and large $\text{Ca}(\text{OH})_2$ crystals easily form on the CRCA surface and pores, resulting in lower mechanical properties of RAC than NAC [15]. On the contrary, Boudali et al. [54] showed that with the increase in hydration time, water and cement paste can enter into the CRCA for hydration reaction,

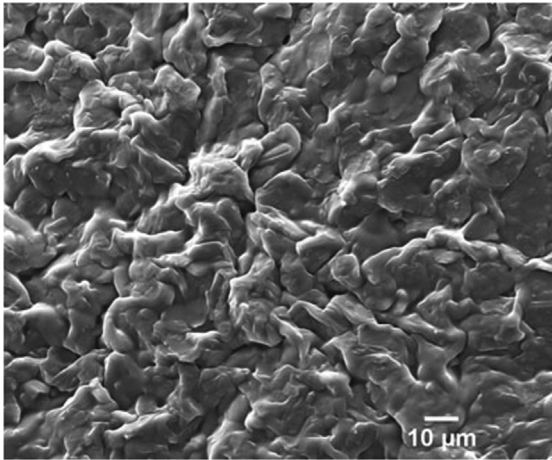


Fig. 3. SEM micrograph of NA surface [4].

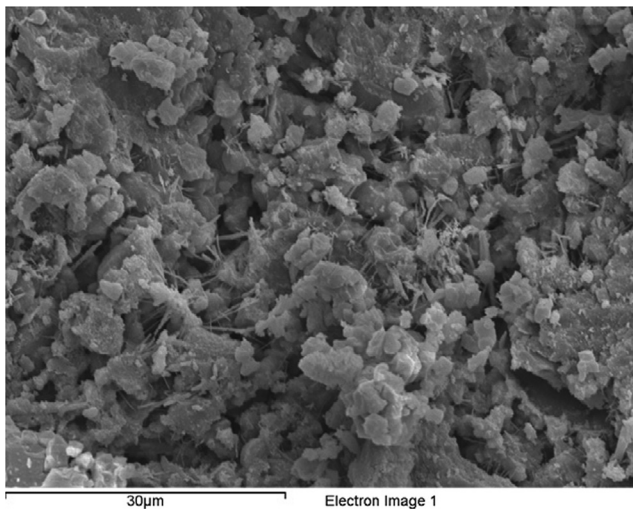


Fig. 4. SEM micrograph of RCA surface [53].

thereby filling partial pores of the ITZs and improving the mechanical properties of RAC. The mineral composition and surface structure of the RCA will affect the growth of hydration products and the bonding between the new and old cement matrices, which will affect the mechanical properties of RAC.

3. Defects in RAC microstructure

The defects of the microstructure of RAC are manifested in the weak physical and chemical bonding between RCA and cement paste, and the presence of cracks and pores, thereby weakening the mechanical properties and durability [56]. In addition, attached mortar also is the weak part in RAC [55]. The presence of attached mortar makes the porosity of RAC higher than that of NAC, especially FRAC, and the porosity increases with the increase in the replacement level and w/c of RCA [52,55]. The weakening of the physical and chemical bonding between RCA and cement paste can be attributed to two reasons. On the one hand, the surface of RCA has loose particles, as shown in Fig. 4. On the other hand, a water film is formed near the aggregate, and a porous $\text{Ca}(\text{OH})_2$ rich layer is formed between the RCA and cement paste perpendicular to the aggregate surface due to the high water absorption of RCA [53,54]. Compared with ITZ of NAC prepared with NFA (0–5 mm) as is shown in Fig. 5(a), there are more micro-cracks and pores in the old ITZ of RAC prepared with 100% FRCA (0.15–5 mm) as is shown in Fig. 5(b) and (c), which affect the ultimate strength of RAC [15,57]. As the fracture of the original concrete reduces the fracture density of the old ITZ and introduces negligible new fractures, the microstructure of attached mortar becomes more fragile due to the increase in porosity and fractures, which may affect the microstructure of RAC [3,35]. When the replacement levels of CRCA and FRCA are less than 30%, the effect on the strength of RAC is small [1,11].

The failure of RAC is the result of the joint action of ITZ failure and the internal cracking of RCA, whereas the failure of NAC is the result of the expansion of cracks along the ITZ [35]. As the RCA has micro-cracks, when it is cast into the new cement paste, the old ITZ merges with the new ITZ, thereby forming a weak bond of the cement matrix and RCA, which in turn causes the failure of RAC [34]. Li et al. [58] studied the development of cracks in the microstructure of RAC under uniaxial compression using digital image technology. They found that cracks develop towards the loading direction. Firstly, micro-cracks were generated around the RCA, and the crack width increased. The cracks gradually developed from the ITZs to the cement mortar, eventually leading to RAC cracking. On the basis of the idea that cracks initiated and propagate at low-strength ITZ, when the strength of the new ITZ is lower than that of the old ITZ, the first observable crack appears in the new ITZ area. However, when the strength of the new ITZ is higher than that of the old ITZ, the first micro-crack occurs in the old ITZ. This finding is similar to that obtained by Ryu [59], that is, the strength of RAC depends on the relative quality of ITZs, and the

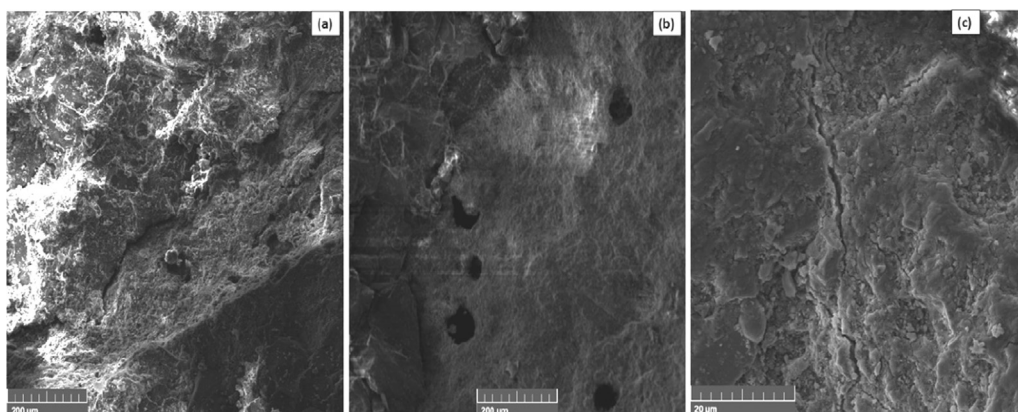


Fig. 5. SEM images of ITZ in: (a) NAC; (b) Recycled fine aggregate concrete (100% FRCA replacement level); (c) Recycled fine aggregate concrete at high magnification [57].

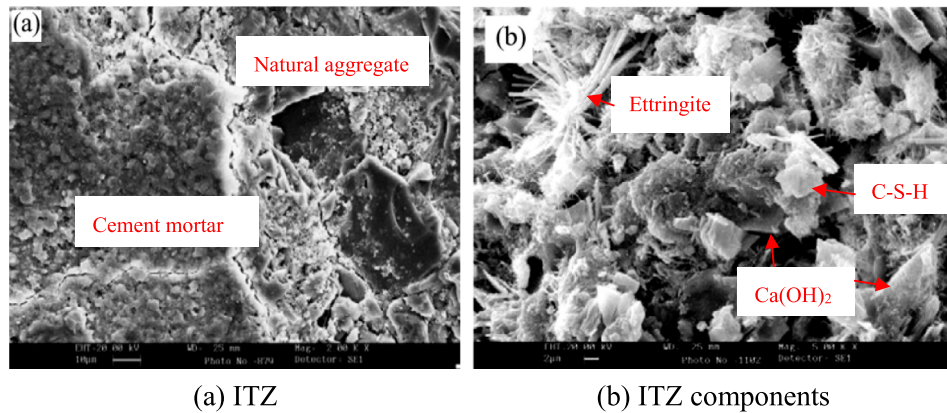


Fig. 6. Microstructure of NA-cement interface [18]. (a) ITZ (b) ITZ components.

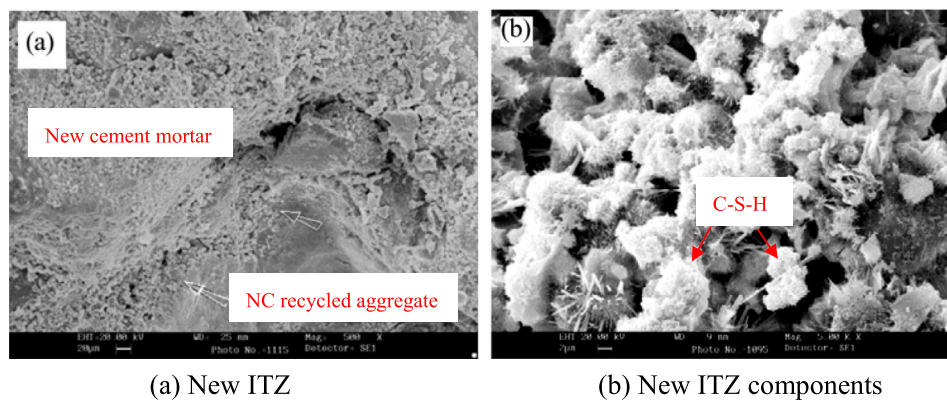


Fig. 7. Microstructure of NC-cement interface [18]. (a) New ITZ (b) New ITZ components.

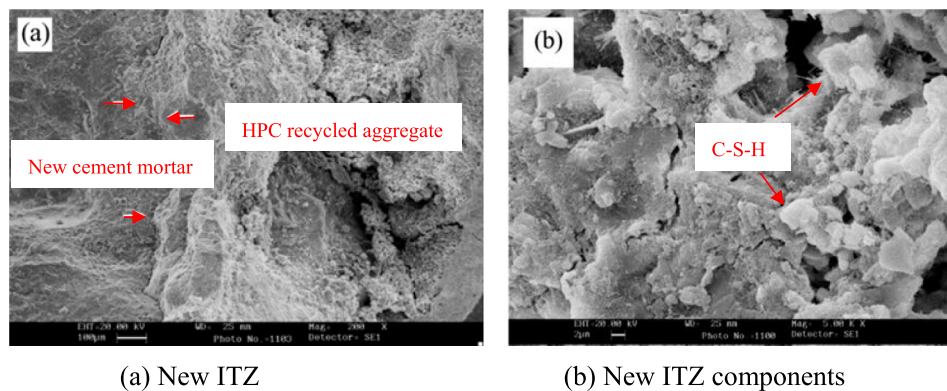


Fig. 8. Microstructure of HPC-cement interface [18]. (a) New ITZ (b) New ITZ components.

microhardness of the ITZs represents their quality. When the microhardness of the new ITZ is high, the strength of the RAC depends on the quality of the old ITZ. When the microhardness of the old ITZ is high, the strength of RAC depends on the quality of the new ITZ.

The higher the strength of the original concrete, the lower the porosity in the new ITZ of RAC [18,60]. Poon et al. [18] observed SEM images of three new ITZs of different strength aggregates, namely recycled normal-strength concrete (NC) aggregate, recycled high-performance concrete (HPC) aggregate and NA. Among

them, granite is NA. Fig. 6 showed well-crystallised Ca(OH)_2 crystals, ettringite and C-S-H at the NA-cement interface. Fig. 7 presented the NC-cement interface consists primarily of granular C-S-H, and the porosity of the NC-cement interface is similar to the NA-cement interface. Fig. 8 showed that the HPC-cement interface is mainly composed of C-S-H gel, which is considerably less than the porosity of the previous two. The microstructure of RAC is related to ITZs and the porosity of RCA, so the microstructure and properties of RAC can be improved by changing the microstructure of RCA and ITZs.

Table 1
Enhancement treatments for the microstructure of RCA.

Enhancement methods	Principle	Advantages	Disadvantages	References	
Removal of attached mortar in RCA	Heat treatment	Remove the attached mortar by generating thermal stress difference	<ul style="list-style-type: none"> • Reduce the width and length of cracks in the old ITZ and the pore size of attached mortar • Enhance the bonding between RCA and the new cement paste 	<ul style="list-style-type: none"> • The pore size of RCA increases after 350 °C • Emissions of CO₂ 	[61–67]
	Ultrasonic cleaning	Remove weak attached mortar on the RCA surface and weak attached mortar with water	<ul style="list-style-type: none"> • Enhance the bonding between RCA and the new cement paste • Increase compressive strength of RAC 	<ul style="list-style-type: none"> • Energy consumption and water waste • Hard to remove strong attached mortar 	[9,22]
	Pre-soaking in acid	Remove the attached mortar on the RCA surface and attached mortar	<ul style="list-style-type: none"> • Reduce the width and length of cracks in the old ITZ and the pore size of attached mortar • Enhance the compactness of new ITZ • Waste solution of CH₃COOH can store CO₂ 	<ul style="list-style-type: none"> • High concentrations (0.8 mol) of acid increase the porosity of RCA • Waste solution of HCl and H₂SO₄ causes environmental problems 	[68–74]
Surface coating on RCA	Pozzolanic materials	Pozzolan reaction with Ca(OH) ₂ to form new hydration products	<ul style="list-style-type: none"> • Reduce the porosity of RCA and make ITZs dense and strong • Improve the durability and mechanical properties of RAC • Conductive to waste disposal 	<ul style="list-style-type: none"> • Nanoparticles cannot reduce the width of old ITZ 	[15,22,41,58,75–83]
	Polymer emulsion	The polymer emulsion coats the surface of RCA and fills the pores	<ul style="list-style-type: none"> • Fill the pores in the RCA and dense the microstructure • PVA can enhance the bonding between the new cement paste and CRCA 	<ul style="list-style-type: none"> • Silicon-based polymers weak the bonding between CRCA and the new cement paste • Waste solution causes environmental problems 	[9,42,84–90]
Different mixing methods CaCO ₃ precipitation	Different mixing methods	Pre-mix RCA with a coating on its surface	<ul style="list-style-type: none"> • Fill the pores in the RCA and dense the microstructure of RAC • Make ITZs dense and strong • Improve the mechanical properties and durability of RAC 	<ul style="list-style-type: none"> • Limited properties improvement 	[3,15,38,40,43,59,79,91–97]
	Calcium carbonate biodeposition	Bacteria precipitate CaCO ₃ in the pores of RAC, causing chemical reactions	<ul style="list-style-type: none"> • CaCO₃ is used to fill pores of RCA • Enhance the bonding between RCA and new cement paste • Improve the quality and strength of RAC 	<ul style="list-style-type: none"> • The quality of CaCO₃ is uncertain • Increase the number of small pores 	[9,34,98–101]
Different mixing methods	Carbonation	CO ₂ reacts with hydration products to form CaCO ₃	<ul style="list-style-type: none"> • CaCO₃ is used to fill pores of RCA • Make ITZs dense and strong • Improve mechanical properties and durability of RAC • Protect environment 	<ul style="list-style-type: none"> • Ca(OH)₂ reaction is incomplete • Increase the number of small pores 	[40,42,60,89,93,102–126]

4. Enhancement treatments

The main improvements proposed in this paper are from four aspects, as shown in Table 1, including the following principles, advantages and disadvantages: (1) removal of attached mortar in the RCA, (2) surface coating on RCA, (3) different mixing methods and (4) CaCO₃ precipitation.

- (1) Removal of attached mortar can improve the bonding between the cement paste and RCA and dense ITZs. These methods are more suitable for CRCA because CRCA contains a low proportion of attached mortar [61–74]. However, ultrasonic cleaning method cannot remove the attached strong mortar on the surface of RCA [9]. Excessive acid solution concentration and high temperature for heating RCA are not good for reducing the porosity of RCA [66,67]. Ultrasonic cleaning and heating RCA require energy consumption, and the waste liquid generated by HCl and H₂SO₄ is unfavourable to the environment [9,62,71].
- (2) Coating on the surface of RCA by using pozzolanic materials or polymer emulsions can reduce the porosity of RCA [75–90]. Pozzolanic materials, such as fly ash (FA), silica

fume (SF), blast-furnace slag and nanosilica (NS), can also improve ITZs [22,41,58]. However, nanolimestone (NL) is not obvious for reducing cracks of ITZs due to the poor dispersion [58]. A hydrophobic film formed on the surface of the RCA after the reaction of silicon-based polymer and C-S-H, which weakens the bonding between the RCA and the new cement paste [89]. In addition, the waste liquid produced by polymer emulsions is not good for the environment [42].

- (3) Different improved mixing methods, such as two-stage mixing method (TSMA), double mixing method (DM), triple-mixing method (TM), mortar mixing approach (MMA), sand enveloped mixing approach (SEMA), and optimized triple mixing method (OTM), which can form a low w/c cement coating or pozzolanic coating on the surface of RCA by pre-mixing RCA, thereby reducing the porosity of RCA and improving ITZs [15,40,43,91–97].
- (4) Calcium carbonate (CaCO₃) is produced by biodeposition on the surface of RCA or carbonation of RCA, which fills the pores of RCA and improves ITZs, thereby improving the microstructure of RCA [98–121]. However, CaCO₃ is not conducive to the filling of small pores [100,121].

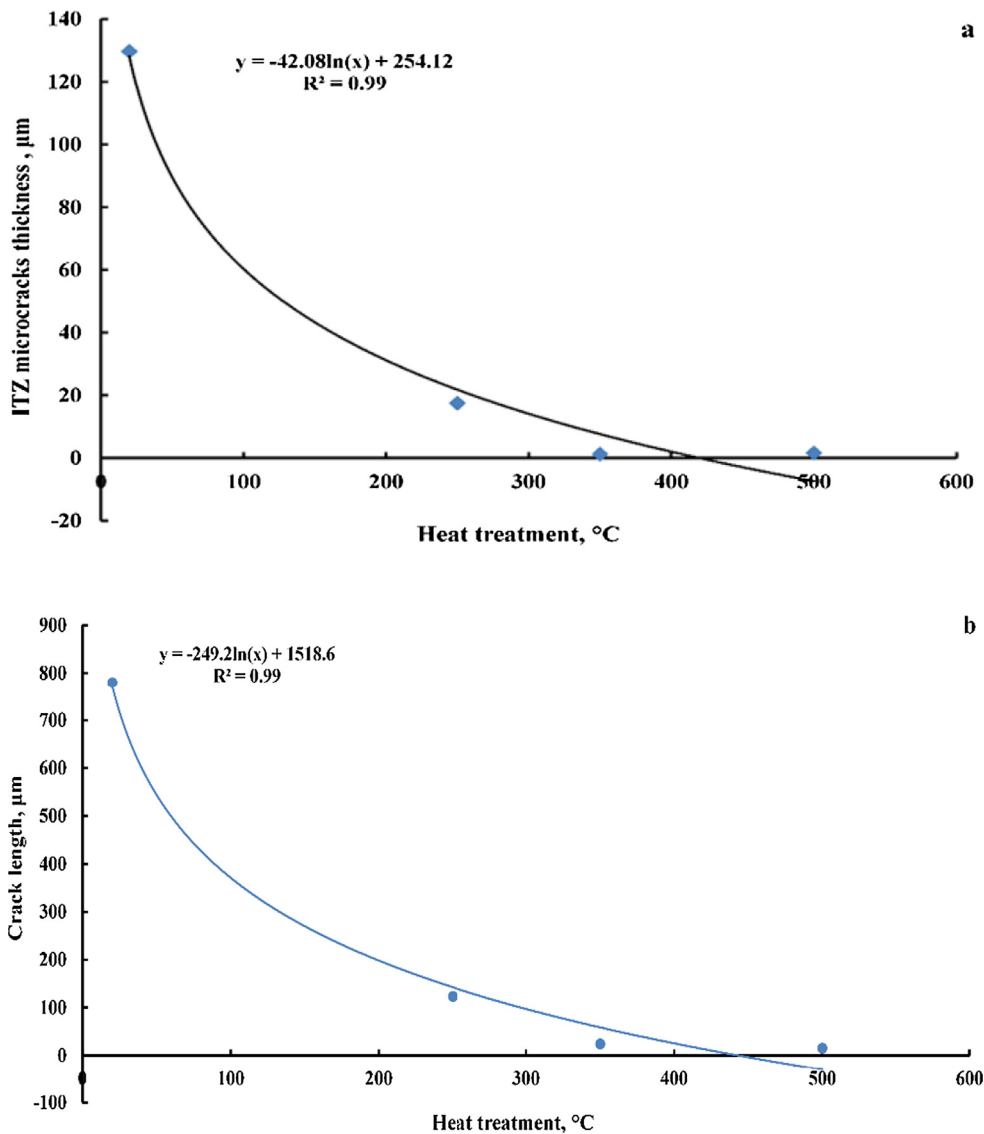


Fig. 9. Behavior of old ITZ microcracks through heat treatment in: (a) width; (b) length [67].

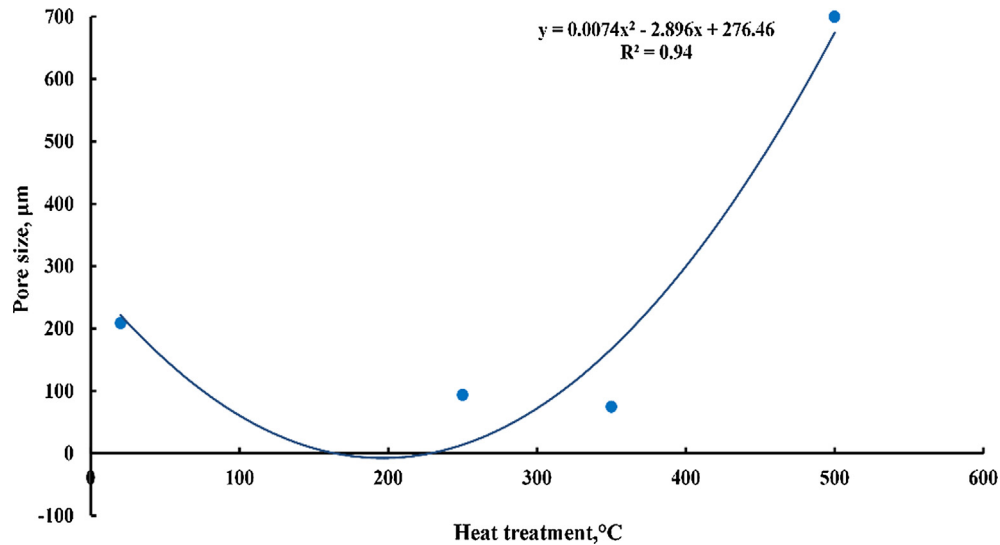


Fig. 10. Pore size of CRCA attached mortar behavior through heat treatment [67].

4.1. Removal of attached mortar in RCA

4.1.1. Heat treatment

The thermal stress difference between NA and the attached mortar can be generated by heat treatment to remove the attached mortar [61]. This process enhances the bonding between RCA and the new cement paste, and reduces the width and length of cracks in the old ITZ and the pore size of attached mortar, thereby effectively recovering RCA [62–64]. However, carbon dioxide (CO₂) emissions from the aggregate recovery process are between 1.5 kg CO₂/t and 4.5 kg CO₂/t, resulting from energy consumption [65,66]. Al-Bayati et al. [67] heat-treated CRCA (4.75–19 mm) at 250 °C, 350 °C and 500 °C in a conventional electric oven for 1 h. Figs. 9 and 10 showed that at 0 °C–35 °C, the width and length of micro-cracks in the old ITZ and the pore size of CRCA decrease sharply with the increase in temperature. However, after 350 °C, the width and length of micro-cracks in the old ITZ decrease slightly, but the pore size of CRCA attached mortar increased dramatically. It was suggested that the optimum processing temperatures of ITZs and mortar are 250 °C and 350 °C, respectively [66].

4.1.2. Ultrasonic cleaning

Ultrasonic cleaning of RCA removes the weak attached mortar of RCA and enhances the bonding between RCA and the new cement paste, but this method will cause energy consumption and water waste [9,22]. Katz [22] immersed the CRCA (4.75–19 mm) in an ultrasonic bath and washed its surface with water for 10 min, repeating this operation until clear water was obtained. The result showed that the attached mortar on the surface of CRCA are reduced; thus, the bonding of RCA to the new cement paste were improved. The increase in compressive strength of RAC was 3% at 7 days and 7% at 28 days possibly because the method cannot remove the strong attached mortar [9].

4.1.3. Pre-soaking in acid

The attached mortar is removed by pre-soaking RCA in the acid solution through the reaction between the acid and the hydration products in the attached mortar, which can reduce the width and length of cracks in the old ITZ and the pore size of attached mortar, enhance the compactness of new ITZ, and improve the quality of RCA [68–70]. HCl and H₂SO₄ can be used in strong acids, and CH₃-

COOH can be used in weak acids [68]. The waste solution of these two strong acids will produce certain environmental problems. However, Ca²⁺ in the waste solution of CH₃COOH can store CO₂, and reduce carbon emissions [71].

Tam et al. [68] proposed a pre-soaking method, that is, the CRCA (10 mm and 20 mm), which was first immersed in HCl solutions with a concentration of 0.1 mol for 24 h at 20 °C. CRCA was then watered with distilled water to remove the acidic solvent. Two untreated CRCA were separately immersed in the other two acidic solutions with a concentration of 0.1 mol by the above method, namely, H₂SO₄ and H₃PO₄, respectively. The SEM showed that the new ITZ after treatment (Figs. 12–14) is denser than the new ITZ without treatment (Fig. 11) because of the removal of the attached mortar. The result is consistent with the conclusion of Kazemian et al. [72], they soaked CRCA (4.75–19 mm) in 0.1 mol HCl solution by the pre-soaking method. Moreover, the surface porosity of RCA does not increase due to the low concentration of HCl. Kim et al. [73] pre-soaked CRCA (5–20 mm) in HCl solution for 48 h with aggregate-to-solution ratio of 1:4.5. The reduction of CRCA poros-

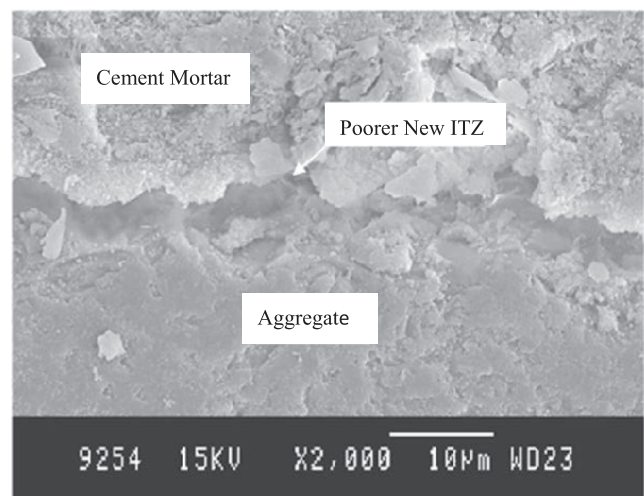


Fig. 11. Poorer new ITZ for NMA without pre-soaking treatments [68].

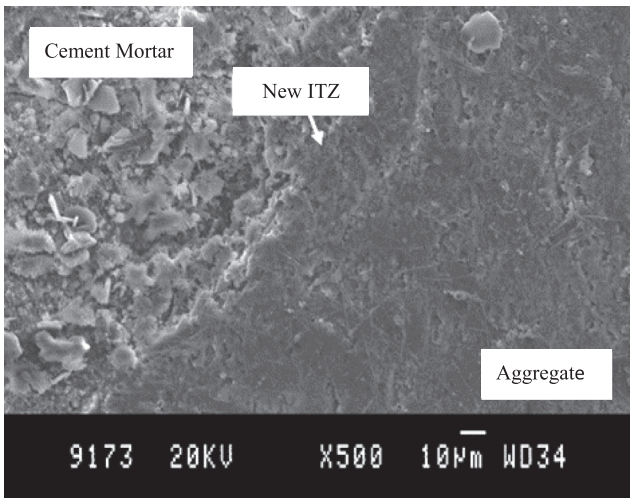


Fig. 12. New ITZ with HCl treatment [68].

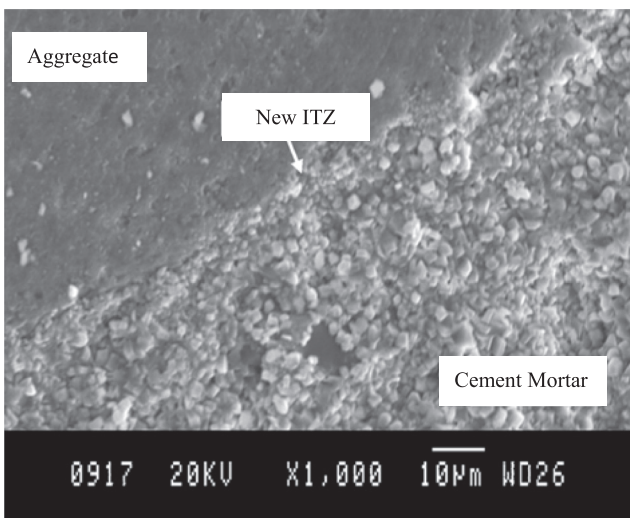


Fig. 13. New ITZ with H₂SO₄ treatment [68].

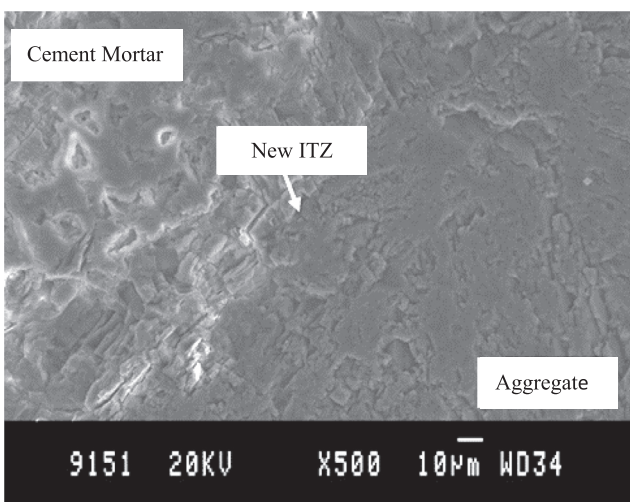


Fig. 14. New ITZ with H₃PO₄ treatment [68].

Table 2

Microcrack width and length in old ITZ and pore size in mortar surface before and after acid treatments [67].

CRCA treatment/ property	Microcrack width (μm)	Microcrack length (μm)	Pore size (μm)
CRCA untreated	129.70	780	208.9
CRCA soaking in HCl	5.29	209.1	139.9
CRCA soaking in CH ₃ COOH	1.93	28.8	156.1

ity improved the compressive strength and durability of RAC, and treated CRCA was suitable for structural concrete.

Ismail et al. [69] compared the effect of HCl solution concentration on CRCA (10–14 mm) surface. On the basis of the method proposed by Tam et al. [68], they designed the concentrations of HCl solutions to be 0.1, 0.5 and 0.8 mol and then soaked CRCA in three solutions for 1 day, respectively. It was concluded that three HCl solutions could reduce the attached mortar on the RCA surface, but 0.8 mol HCl solution eroded the surface of CRCA because of its high concentration, which increased the porosity of RCA; thus, 0.5 mol was recommended. When the concentration of HCl solution was 0.5 mol and the replacement level of RCA was 60%, improvements in ITZs lead to increased durability of RAC. Such an improvement was encouraged for the production of structural concrete without corrosive reinforcement because of the corrosion of Cl⁻ [74]. Al-bayati et al. [67] studied the effect of acidity of solution on the microstructure of CRCA (4.75–19 mm) by using the pre-soaking method. The concentration of acid solution was 0.1 mol, which was composed of HCl (37%) and CH₃COOH (99.7%). The test results indicated that both acids can reduce the width and length of microcracks in the old ITZ, as well as the pore sizes in the mortar surface (Table 2), and CH₃COOH better reduced the width and length of the microcracks, while HCl better reduced the pore size of the mortar.

4.2. Surface coating on RCA

4.2.1. Pozzolanic materials

RCA is immersed in or sprayed with the pozzolanic slurry can form a coating on the surface of RCA. Pozzolanic materials, such as FA, blast furnace slag, SF and metakaolin can react with Ca(OH)₂ to generate C-S-H, effectively reduce the porosity of RCA and make ITZs dense, thus improving the microstructure and mechanical properties of RAC [15,22,75–77]. Pozzolanic materials are industrial by-products, and their use facilitate waste disposal problems [78].

Katz [22] studied the effect of SF coating on the microstructure of RAC and presoaked the CRCA in a 10% heavy SF solution to form an SF coating on the CRCA (4.75–19 mm) surface. The result showed that SF reacted with Ca(OH)₂ to promote cement hydration and filled pores as a microfilmer, which reduced weak attached mortar on the surface of CRCA caused by the crushing of the old concrete and enhanced the bonding between CRCA and new cement paste. This finding agreed with the conclusion of Tam et al. [79], who used the two-stage mixing method (TSMA) to coat the surface of RCA (≤ 20 mm) with SF (2% by weight of cement), thereby reducing the microcracks of RAC and densifying the ITZs. Li et al. [41] found that the application of FA, SF and blast furnace slag pozzolanic materials to the CRCA (5–31.5 mm) in different mixing manners can make the new ITZ structure dense. The minimum thickness of the coating covering the entire new ITZ is 0.5 mm, and the mixing of the three is more effective for the compactness of the new ITZ because of the high packing density.

Nanoparticles can not only act as a filler to a dense microstructure due to high reactivity but also further promote cement

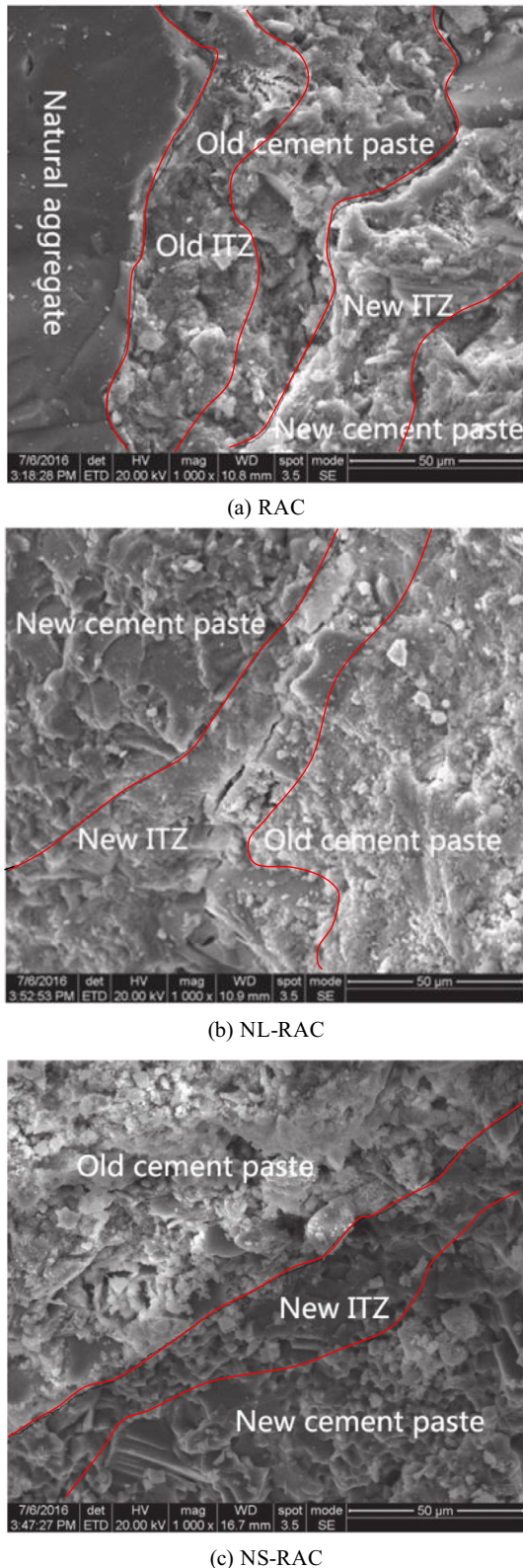


Fig. 15. Microstructure of ITZs in RACs (RAC: RAC prepared with untreated CRCA; NL-RAC: RAC prepared with NL modified CRCA, and NS-RAC: RAC prepared with NS modified CRCA) [58]. (a) RAC (b) NL-RAC (c) NS-RAC.

hydration. NS and NL are the most commonly used nanoparticles. The NS size is 1–50 nm, which can be used for RCA pores filling and pozzolanic reaction, and the NL size is 15–80 nm, which is mainly used for RCA pores filling [58,80,81]. Li et al. [58] covered

the surface of CRCA (5–26.5 mm CRCA of 30% replacement level) with NS slurry accounting for 1%–2% by weight of cement (8–15 nm) or NL slurry accounting for 1%–2% by weight of cement (15–40 nm) via TSMA. RACs prepared with CRCA modified by two kinds of nanoparticles were named as NS-RAC and NL-RAC respectively. A group of RAC without nanoparticles was used as a control. ITZs of the three RACs were shown in Fig. 15. Fig. 15(a) showed obvious cracks in ITZs. The nanoparticles could densify the microstructure and reduce the micro-cracks. Fig. 15(b) showed that few cracks remained in the new ITZ, and Fig. 15(c) showed no cracks in the new ITZ, indicating that NS is more effective than NL. Colloidal NS can enter new ITZ more easily than powdered NL, thereby causing pozzolanic reaction, producing more secondary C-S-H, filling cracks and pores of CRCA and improving the microstructure and mechanical performance of RAC. Appropriate superplasticiser could be added to increase the dispersion of NL, which is further conducive for entering the RCA [58].

Moreover, nanoparticles can reduce the width of new ITZ. Zhang et al. [82] soaked CRCA (5–35 mm) in a solution with a concentration of 5% NS and 3% NL for 45 min, then compared the ITZs width of RAC prepared by CRCA at 3 days and 28 days. They found the old ITZ width (40 μm) does not change, while the new ITZ width reduced from 60 μm to 50 μm. It was because the nanoparticles that they used could not penetrate into the old ITZ. Shaikh et al. [83] used two methods to added NS (25 nm) into RAC (100% CRCA replacement level), that is, pre-soaking CRCA in the 2% NS solution or directly adding the 2% NS solution into RAC. The result showed that the porosity of RAC using pre-soaking method is about 30% lower than that of direct incorporation method. The reason is that the use of pre-soaking method can lead to the formation of pozzolanic reaction of NS with hydration products in advance.

4.2.2. Polymer emulsion

Two typical polymer emulsions include polyvinyl alcohol (PVA) emulsions and silane-based polymers [9]. By coating the RCA surface, the porosity of RCA is reduced, and the microstructure of RAC is densified. However, the waste solution of polymer emulsion causes environmental pollution [42]. PVA is a water-soluble polymer that can be used to reduce the w/c and increase the bonding of cement paste to RCA [9]. The added PVA can be adsorbed on the surface of RCA, and the inactivation of Ca(OH)₂ site prevents the growth of Ca(OH)₂ and may be replaced by C-S-H [84]. Kou et al. [85] proposed immersing CRCA (10 and 20 mm) in the PVA solution for 24 h. They found that PVA can fill the pores of attached mortar and seal the CRCA surface, thereby reducing the porosity of CRCA and enhancing the adhesion of new cement paste to CRCA. PVA improved CRCA was encouraged to produce structural concrete similar to the durability of NAC.

Silicon-based polymers include silane, siloxane and both (Fig. 16). The silicon-based polymer can fill the pores of RCA and react with C-S-H to form a hydrophobic film on the surface of RCA [86]. The addition of sodium silicate to the silane and siloxane solutions facilitated hydrolysis and further compacted the microstructure of RAC [8,87,88]. Zhu et al. [89] treated CRCA (31.4% 10 mm CRCA and 62.9% 20 mm CRCA) with silane paste (100 g/m² and 200 g/m²) or mixed RAC with silane solution (1%

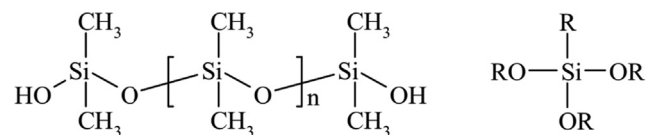


Fig. 16. Siloxane and silane [85].

and 2% by cement weight). It was concluded that the bonding between CRCA and the new cement paste is weakened, possibly because of the formation of hydrophobic film on the surface of CRCA. Santos et al. [90] observed that when CRCA (4.75–9.52 mm) was immersed in 20% silane emulsion for 24 h, the result showed that silane reduce the number of pores under 100 nm, but the overall porosity of CRCA decreased slightly.

4.3. Different mixing methods

Compared with the early normal mixing approach (NMA), Ca (OH)₂ hydration is incomplete due to insufficient water. Many cracks and pores remain in the RCA, and the ITZs are weak. The improved mixing methods proposed later can form a low w/c cement coating on the RCA surface, effectively reducing the cracks and pores in the RCA, improving the ITZs and thus improving the mechanical properties and durability of the RAC [3,91,92]. However, the properties improvement of RAC is limited by the improved mixing method without pozzolanic materials [93]. Therefore, researchers have proposed TSMA [43], TM [15], MMA [96], SEMA [96], and OTM [97].

Tam et al. [43] proposed the TSMA and compared the ITZs that mixed RAC using NMA and TSMA. The NMA involves adding all

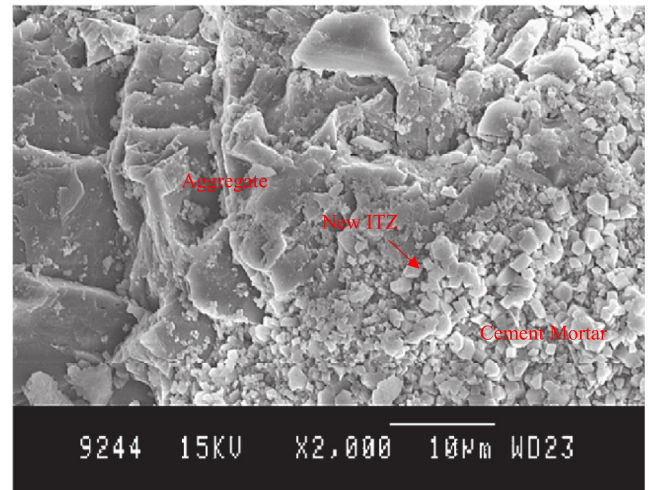
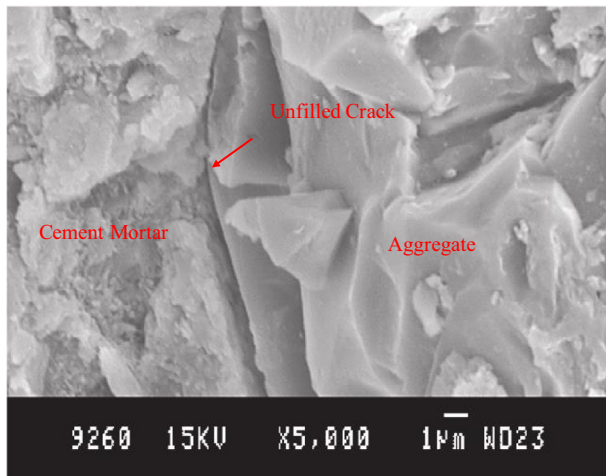


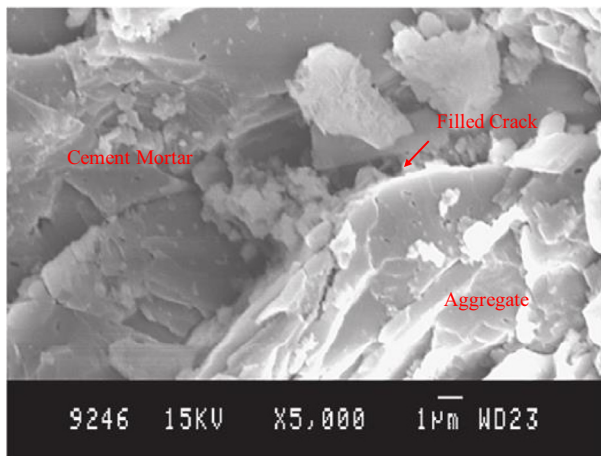
Fig. 18. New ITZ in RCA using TSMA [43].

materials at once and then mixing them. Given the porosity of RCA, unfilled cracks remain in the RCA, as shown in Fig. 17(a). The TSMA produces RAC in two stages. In the first stage, a certain portion of water and cement were mixed with RCA (20% 0–5 mm FRCA, 40% 10 mm CRCA and 40% 20 mm CRCA), which will form a thin layer of cement mortar on the surface of RCA, thereby filling the pores and cracks of RCA, as shown in Fig. 17(b). The new and old ITZ were further strong and dense, as shown in Figs. 11, 18 and 19. When using TSMA, the RCA replacement level was 25%–40%, which is most effective for improving the microstructure [94]. Li et al. [38] studied the effect of TSMA and NMA on the performance of ITZs of RAC prepared with a replacement level of 100% CRCA. In comparison with NMA, TSMA decreased the porosity of ITZs, the volume fraction of Ca (OH)₂ and ettringite, increased the volume fraction of C-S-H gel and densified the ITZs. The compressive strength of the RAC was improved by enhancing the microstructure of ITZs. Otsuki et al. [40] used the double mixing method (DM) that similar to TSMA to coat the high water–binder ratio (0.55) CRCA with a low water–binder cement paste. In comparison with single mixing method, the micro-hardness of the new ITZ increased significantly. Similarly, Ryu [59] previously treated CRCA with DM and found that the thickness of the new ITZ decreased, the w/c of RAC decreased, and the durability of RAC increased.

The addition of pozzolanic materials in the first stage of TSMA can form a layer of pozzolanic coating on the surface of RCA, thereby filling the pores and cracks in the ITZs, generating C-S-H gel with unhydrated Ca(OH)₂, densifying the ITZs and increasing the influence of ITZs on the microstructure of RAC. Tam et al. [79] added SF to replace 2% of cement in the premixing stage and proposed two methods based on TSMA, namely, TSMA_s and TSMA_{sc}, to improve the TSMA. TSMA_s only adds SF in the premixing stage. TSMA_{sc} adds SF and a certain proportion of cement in the premixing stage. The latter is more cement coated than the former in the premixing process, as shown in Fig. 20, thereby further densifying the ITZs. Rajhans et al. [95] proposed a similar method based on TSMA, namely, TSMA_{sf}, in which SF (replace 7% FA) was added in the pre-mixing stage to form a SF coating on the surface of CRCA (5–20 mm), and FA (accounted for 34.78% of the cement weight) was added in the second stage. In comparison with TSMA, ITZs were further densified, and the improvement of ITZs increased the strength of the resulting self-compacting concrete. These results are consistent with those of Liang et al. [96], who mixed cement mortar and a silica solution to pre-treat the surface

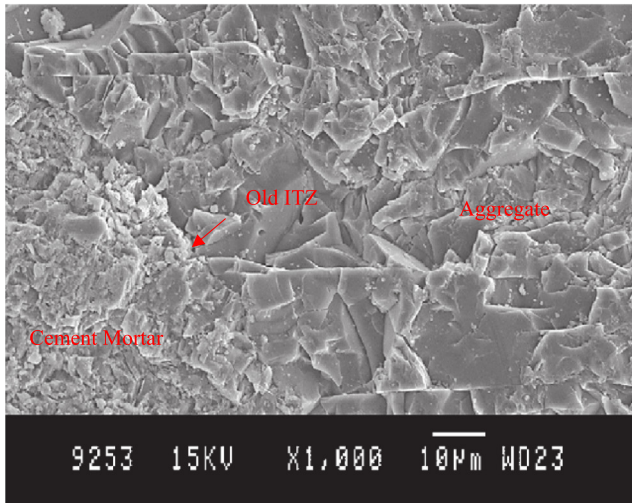


(a) Unfilled crack in RCA using NMA

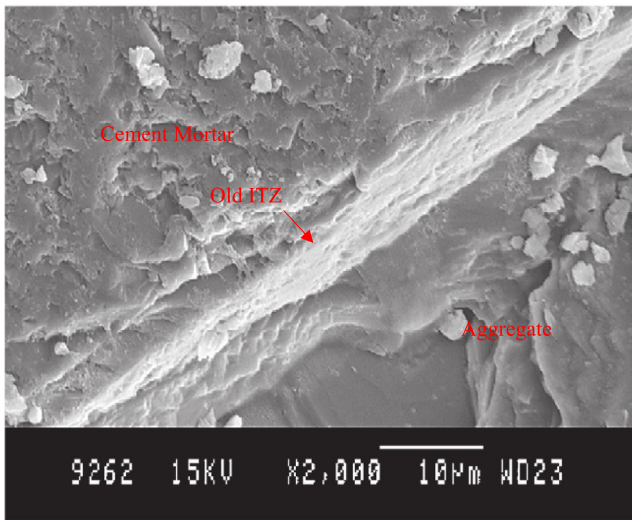


(b) Filled crack in RCA using TSMA

Fig. 17. Cracks in RCA using NMA and TSMA [43]. (a) Unfilled crack in RCA using NMA (b) Filled crack in RCA using TSMA.



(a) Old ITZ in RCA using TSMA



(b) Old ITZ in RCA using NMA

Fig. 19. Old ITZ in RCA using TSMA and NMA [42]. (a) Old ITZ in RCA using TSMA (b) Old ITZ in RCA using NMA.

of CRCA by using MMA and SEMA and produced a good bonding between CRCA and the new cement paste.

When using the TSMA or DM, the mixed particles in the pore and ITZs are limited, and the pozzolanic reaction is incomplete. On the basis of the above mentioned methods, Kong et al. [15]

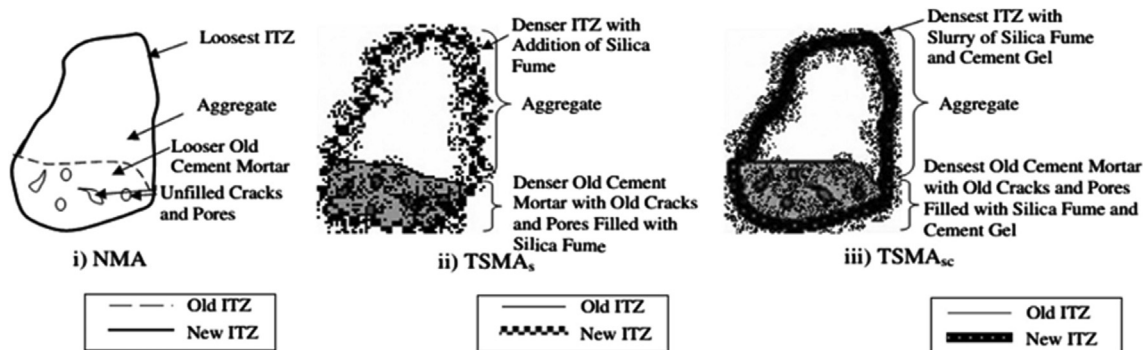


Fig. 20. Microstructure of RCA after adopting: i) NMA, ii) TSMA_s, iii) TSMA_{sc} [79].

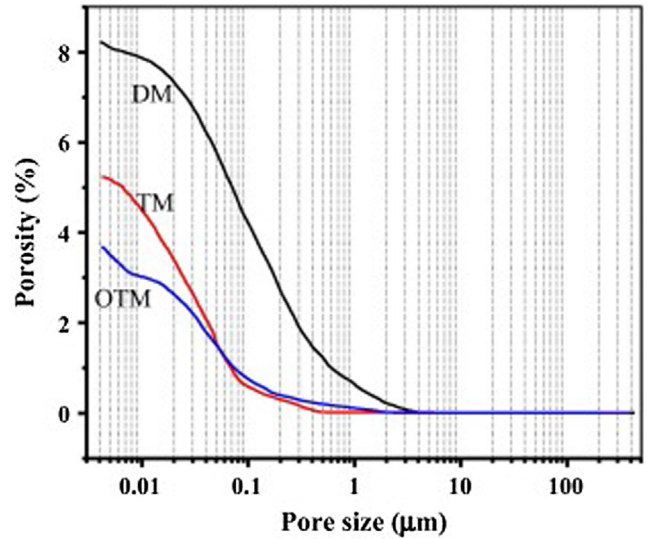


Fig. 21. Porosity of ITZs in RAC prepared by different mixing methods (DM: Double mixing method, TM: Triple mixing method, and OTM: Optimized triple mixing method) [97].

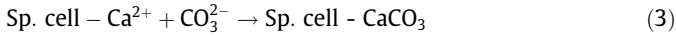
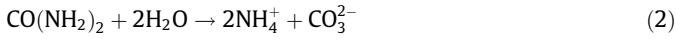
proposed TM that a FA coating or slag coating was formed on the surface of CRCA (5–20 mm) and then a cement coating was formed in surface-coated aggregate. Compared with TSMA and DM, TM further improved the filling effect and pozzolanic reaction, and improved ITZs. Zhang et al. [97] proposed an OTM to prepare RAC (100% CRCA + 30% FRCA). The difference between this method and DM and TM is the order in which pozzolanic materials (SF and slag), water and superplasticiser are added. As shown in Fig. 21, the porosity of ITZs prepared by OTM method is the lowest because this method can disperse the pozzolanic material well and the content of Ca(OH)₂ and ettringite is further reduced, thus reducing the porosity of RAC.

4.4. CaCO₃ precipitation.

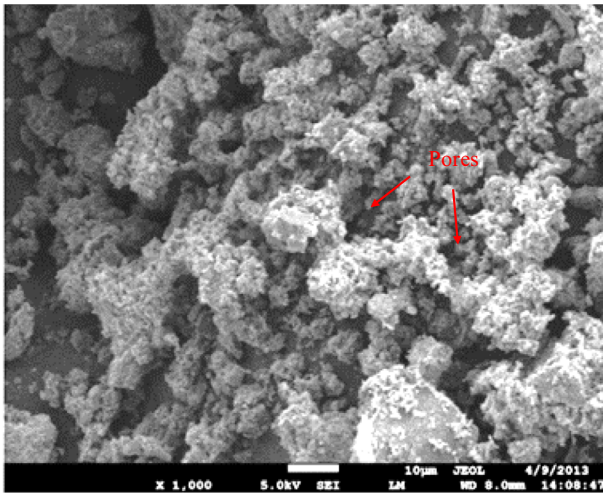
4.4.1. CaCO₃ biodeposition

CaCO₃ biodeposition means that bacteria can precipitate CaCO₃ on the surface of RCA [9,98]. The method occurs naturally in the environment and in principle does not harm the environment [99]. By using *S. pasteurii* as an example, Grabiec et al. [99] inoculated *S. pasteurii* in a liquid culture medium contained urea. They then placed the liquid culture medium in an incubator at 30 °C for 24 h. The culture solution containing bacteria was then poured into a container containing dried CRCA (12–16 mm) at 78 °C for 24 h for biodeposition. *S. pasteurii* attracted Ca²⁺ and then reacted

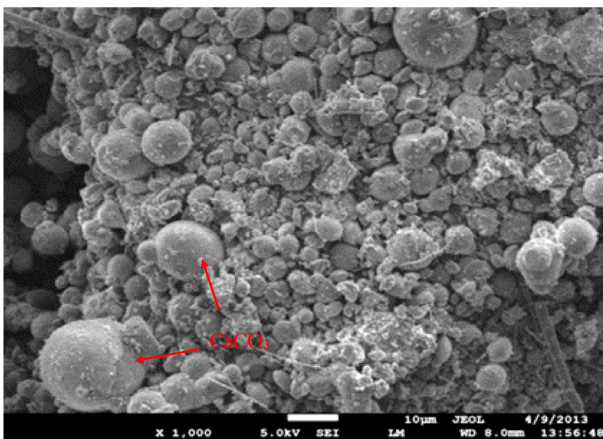
with CO_3^{2-} to form CaCO_3 , which filled the pores of CRCA. The biodeposition process was as follows:



This finding is consistent with the result of Qiu et al. [34], who diluted *S. pasteurii* to 10^8 cells/ml by inoculating it in a liquid culture medium contained CaCl_2 (16.8 g/l) and urea (20 g/l). Sodium hydroxide (1 N solution) was used to adjust the pH = 9.5 and then immersed CRCA (15–20 mm) under SSD conditions in the medium for biodeposition for 3 days at 35 °C. As shown in Fig. 22(a), the surface of untreated CRCA has many pores, whereas Fig. 22(b) presented the surface of bacterial precipitation-treated CRCA has many spherical crystals. Table 3 showed that the surface crystal tends to be composed of bacteria-precipitated CaCO_3 that are used to fill the pores. In addition, they also studied the effects of four variables on CaCO_3 precipitation on the surface of CRCA. The four variables were PH (7.0–10.5) of culture medium, Ca^{2+} concentration (5.6–50.4 g/l of CaCl_2), *S. pasteurii* concentration (10^6 – 10^8 cells/ml) of culture medium, and temperature (25 and 35). The amount of CaCO_3 reached its maximum at pH = 9.5



(a) Untreated CRCA



(b) Bacterial precipitation-treated CRCA

Fig. 22. SEM photos of CRCA before and after bacterial precipitation [34]. (a) Untreated CRCA (b) Bacterial precipitation-treated CRCA.

Table 3
EDX results of untreated RCA and MCP-treated RCA [34].

Element	Weight (%)	
	Untreated RCA	MCP-treated RCA
Ca	3.41	29.37
Si	28.92	0.13
C	15.65	17.19
O	47.36	51.09
Others	3.66	2.22
Total	100.00	100.00

and increased with the concentration of Ca^{2+} , the concentration of *S. pasteurii* and temperature, giving a reference for the research conditions of CaCO_3 biodeposition. Wang et al. [100] using bacteria to treat CRCA (6.3–20 mm) and MA. MA is a mixed recycled aggregate composed of crushed concrete, masonry, glass, and wood. They first cultured *bacillus sphaericus* on sterile medium containing yeast and urea for 24 h, and then cultured the two aggregates on two media for 24 h. CaCO_3 acts as a binder to enhance the bonding between CRCA and the new cement paste, thus increase the strength of the RAC. As is shown in Fig. 23, CaCO_3 also reduced the porosity and pore volume of RAC. However, the number of small pores was increased due to the fact that the pore size became smaller and CaCO_3 could not fill small pores.

CaCO_3 precipitated by bacteria also has an improved effect on new ITZ. Zeng et al. [101] activated *S. pasteurii* in a culture medium for 3 days. They then placed dried CRCA (10–20 mm) into another medium that contained *S. pasteurii* and stored the media in an incubator at 30 °C for 1 day, and then CRCA was grown in a different culture medium without *S. pasteurii* at 30 °C for 6 days. The microhardness of new ITZ₁ between NA and the new cement paste increased by 0.01%, while microhardness of ITZ₂ between the attached mortar and the new cement paste increased by 17.39%. Thus, most of the bacteria are adsorbed between the attached mortar and the new cement paste. CaCO_3 biodeposition is a promising technology in improving the microstructure and strength of RAC [102]. It is not widely used in RCA at present, and the quality of the produced CaCO_3 can be further studied [99,100].

4.4.2. Carbonation

Natural carbonation can occur during the process of concrete removal and recycling. The recovery of one ton of concrete can absorb 11 kg of CO_2 [102]. Carbonation of concrete can reduce CO_2 emissions by nearly 3% per year, which is good for the environment [103]. In natural conditions, carbonation of concrete is a very slow process and its rate depends on many factors, such as concrete mix design, environmental conditions, geometry of the concrete elements and so on. Thus, accelerated conditions are

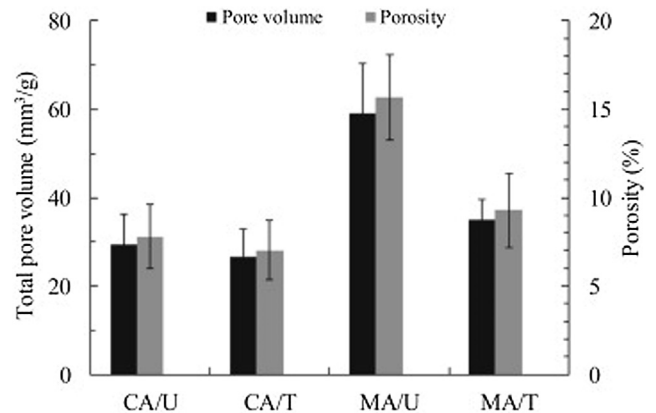
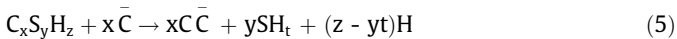


Fig. 23. Porosity changes of CRCA and MA after the bio-treatment (CA/U: untreated CRCA; CA/T: treated CRCA; MA/U: untreated MA; MA/T: treated MA) [100].

usually applied to measure rate of concrete carbonation and predict rate of carbonation in natural conditions [104]. Thiery et al. [105] carbonised cement paste until its mass increase was stable. They found that when the concentration of CO₂ in accelerated carbonation is 50%, the absorption capacity of cement can reach 65%. When CO₂ concentration is 50% under accelerated condition and 0.04% under natural condition, the carbonation degree of C-S-H and Ca(OH)₂ is similar. Xuan et al. [93] carbonised CRCA in an airtight 100-L steel-cylindrical chamber for 24 h with a CO₂ concentration of 100% and pressure levels of 0.1 and 5.0 bar. The particle size of CRCA is 5–10 mm and 10–20 mm respectively, and its content is 554 kg/m³ respectively. Before carbonation, CRCA should be preconditioned in a drying chamber with a temperature of approximately 25 °C to bring the water content close to the optimal relative humidity (RH: 40–70%) for accelerating carbonation, the result showed the 5–20 mm CRCA can absorb 7.5 kg of CO₂ per ton on average. As shown in Table 4, 5–10 mm CRCA has the highest CO₂ uptake at a pressure of 5.0 bar, which can be explained that CRCA with a smaller particle size has a better ability to absorb CO₂ because of its large specific surface area [106]. The same is true for FRCA, Zhang et al. [60] found that the smaller the particle size of FRCA is, the faster the carbonation rate is, and the carbonation reaction occurred rapidly in the first 20 min as shown in Fig. 24.

The working principle of the carbonation reaction is that CO₂ enters the pores of the attached mortar and chemically reacts with its main hydration product, as follows [107]:



where C stands for CaO, H stands for H₂O, S stands for SiO₂, and \bar{C} stands for CO₂. The x, y, z and t are the stoichiometric coefficients of Eq. (5) and they are not exactly defined [107]. SiO₂ can react with Ca(OH)₂ to form secondary C-S-H. Hence, the reduction of Ca(OH)₂ promotes further hydration of cement [108]. In addition, unhydrated calcium trisilicate, calcium disilicate, ettringite and monosulphate hydrate can also react with CO₂ [9]. Slegers et al. [109] and Shah et al. [110] believe that the carbonation of these two hydration products occurs simultaneously. However, some researchers [111,112] believe that the carbonation of Ca(OH)₂ occurs first in the first stage, and the carbonation rate of C-S-H will exceed that of Ca(OH)₂ in the later stage and unhydrated Ca(OH)₂ will form a thin layer on the surface of the RCA, resulting in incomplete carbonation.

Many researchers use accelerated carbonation technology to study the influence on the microstructure of RAC. This technology can not only reduce the porosity of RAC by precipitating CaCO₃, but also improve ITZs and attached mortar. Xuan et al. [93] found that the microhardness of new ITZ increased after carbonation of CRCA (5–20 mm) using the conditions described above (Fig. 25). Li et al. [42] carbonised CRCA (>20 mm) in the standard carbonation chamber for 3 days at atmospheric pressure, 20 °C, approximately 20% CO₂ concentration and 70 ± 5% RH, they found that CaCO₃ produced by carbonation fill the pores and cracks of the old ITZ and attached mortar. Moreover, the microhardness of old ITZ and attached mortar increased by 31% and 17.4%, respectively (Fig. 26). Li et al. [113]

Table 4
CO₂ uptake of CRCA at different particle sizes and pressures [93].

Aggregate	Size (mm)	CO ₂ uptake (%)	
CRCA	Carb. under 0.1 bar	5–10	0.74
		10–20	0.65
	Carb. under 5.0 bar	5–10	0.81
		10–20	0.66

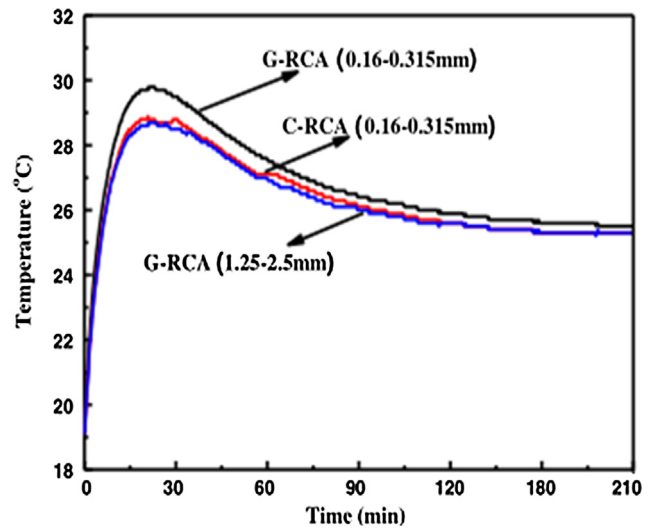


Fig. 24. Carbonation of RCAs (G-RCA: recycled gravel concrete aggregate; C-RCA: recycled crushed stone concrete aggregate) [60].

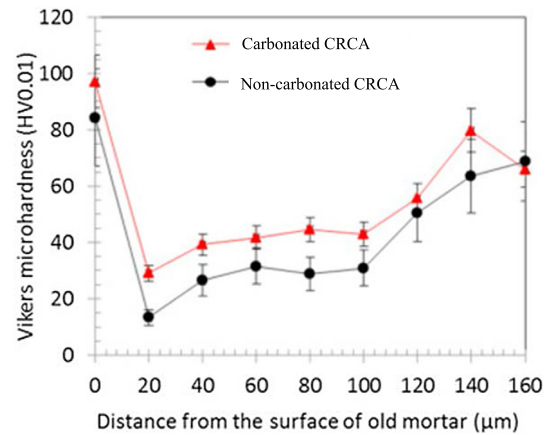


Fig. 25. Vickers microhardness developments at new ITZ around carbonated and non-carbonated RCAs [93].

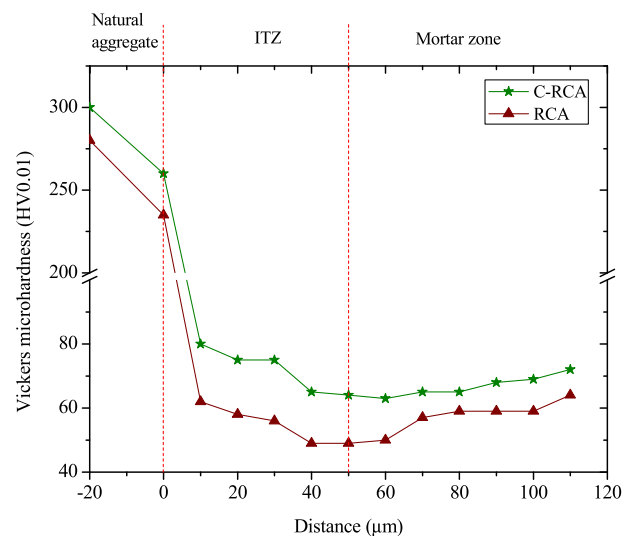


Fig. 26. Vickers microhardness developments at old ITZ around carbonated RCA (C-RCA) and non-carbonated RCA [42].

treated CRCA (5–20 mm) in a 100% CO₂ environment under a moderate pressure (0.1 bar above atmospheric) for 3 weeks. They found that the degree of microhardness improvement of the old ITZ is more noticeable than that of the attached mortar. However, Shi et al. [114] carbonised FRCA (≤ 4.75 mm) in a carbonation chamber for 3 days with a CO₂ concentration of $20 \pm 2\%$ and pressure level of atmospheric pressure. They found that the degree of microhardness improvement of ITZs is more noticeable than that of the attached mortar. Yue et al. [115] studied ITZs microstructure of RAC prepared with CRCA (5–25 mm) after carbonation of 28 days, it was found that carbonation decrease the width of the new ITZ by approximately 10–20 μm , but the reduction in the width of the old ITZ is less obvious. Wang et al. [116] carbonised modeled CRCA in an accelerated carbonation test chamber for 28 days at 30 ± 3 °C, $20 \pm 2\%$ CO₂ concentration and $70 \pm 5\%$ RH. Three modeled CRCA were then mixed with three new cement mortars with w/c of 0.68, 0.45, and 0.37. The result showed that the strength of ITZs increased by 22%, 10% and 3%, respectively. Moreover, changes in microstructure will cause changes in durability of RAC. The porosity of RAC is linear to the water absorption of RAC, and the water absorption can be used to characterise the durability of RAC [117]. Xuan et al. [117] carbonised CRCA (5–20 mm) in an airtight 100-L steel-cylindrical chamber for 24 h with a CO₂ concentration of 100% and pressure levels of 0.1 and 5.0 bar. The result showed that the porosity of RAC decreases with the increase of CO₂ pressure, resulting in the dense RCA microstructure and improved durability of RAC. Zhan et al. [118] found that CRCA (5–10 mm) carbonized under the accelerated condition can only reduce the porosity of the surface of the attached mortar. However, it will enhance the impermeability of the attached mortar, thereby improving the corrosion resistance of RAC and encouraging the application of carbonated CRCA in reinforced concrete.

Given insufficient carbonatable compounds in RCA, some researchers recommend pre-soaking RCA in a solution containing Ca(OH)₂ to produce more CaCO₃ before RCA pre-drying [119]. Pan et al. [119] soaked FRCA (less than 4.75 mm) in Ca(OH)₂ solution and set three variables: Ca(OH)₂ solution concentration, CO₂ concentration and water content. The result showed that the best conditions for carbonising FRCA were 0.01–0.05 mol/kg Ca(OH)₂ solution, 70% CO₂ concentration and 5% water content, and the porosity of RFCA decreased the most. Zhan et al. [120] soaked the simulated CRCA attached mortar in a saturated limewater solution for 3 days and accelerated the carbonation reaction. They found that the porosity of the attached mortar is reduced by approximately 33% compared with that before the treatment; thus, the microstructure of the mortar was dense. Although carbonation reduces the porosity of RAC, the volume of small pores (< 0.1 μm) increased after carbonation. The reason for this may be CaCO₃ refinement pore structure and cannot fill small pores [9,121]. Thierry et al. [105] believed that the change in porosity of RCA caused by carbonation depends largely on the type of cement paste. Cement paste with high cementitious materials (FA, SF, etc) content may cause an increase in the porosity of RCA after carbonation which is probably because of the low content of Ca(OH)₂.

RAC has a higher carbonation depth than NAC. The carbonation depth of RAC is related to many factors, such as RCA replacement level, original concrete grade, mixing method, w/c, the incorporation of mineral additives, polymer emulsions and ITZ on the diffusivity of CO₂. The carbonation depth increases with the replacement level of FRCA [122,123]. When the CRCA replacement level is less than 70%, the carbonation depth of RAC increases with the replacement level. When the CRCA replacement level is greater than 70%, the opposite is true; the carbonation depth of RAC decreases with the increase in original concrete grade [124]. The depth of carbonation decreases with w/c (the w/c can be reduced

using a superplasticiser) decrease [40,125]. The addition of mineral additives increases the carbonation depth by lowering alkaline content that can be carbonized [123]. The carbonation depth is significantly reduced with the amount of silane, and the carbonation depth of the silane surface-treated RCA is less than the overall treatment [89]. The carbonation depth increases with ITZ on the diffusivity of CO₂ [126].

5. Conclusions

This paper summarises the characteristics, defects of RAC microstructure and improvement methods of RCA microstructure on the basis of existing literature.

- (1) Compared with the microstructure of NAC, the microstructure of RAC is more porous due to the existence of attached mortar, resulting in mechanical properties and durability of RAC inferior to that of NAC.
- (2) ITZ consists of pores, Ca(OH)₂, other hydration products (mainly C-S-H gel and ettringite) and unhydrated cement particles. The volume of C-S-H in new ITZ is larger than that in the old ITZ, which due to the high porosity and cracks of the old ITZ, the water content of the new ITZ is reduced, and the initial hydration process is weakened, which may weaken the microstructure of the RAC.
- (3) The microstructure of RAC is related to the porosity of RCA and ITZs, so the microstructure and properties of RAC can be improved by changing the microstructure of RCA and ITZs. The four main approaches enhancing RAC microstructure include removing the attached mortar in the RCA, surface coating on RCA, different mixing methods, and CaCO₃ precipitation.
- (4) By removing the attached mortar of RCA, the microstructure of RCA can be improved and good quality RCA can be recycled. RCA pre-soaked in HCl solution can be used for the production of structural concrete without corrosive reinforcement, but the concentration of HCl and the environmental impact of the waste liquid it produced should be considered.
- (5) Sealing polymer emulsions on the surface of RCA reduces the porosity of RCA, but the hydrophobic film produced by the reaction of silicon-based polymer and C-S-H is not conducive to the bonding between RCA and the new cement paste. RCA soaked in PVA solution is encouraged to produce structural concrete, but the adverse effect of the waste liquid it generates should be considered.
- (6) Pozzolanic materials, such as FA, SF, blast-furnace slag, and NS, fill the surface of RCA and react with Ca(OH)₂ to form C-S-H by coating the pozzolanic materials on the surface of RCA, which can reduce the porosity of RCA and improve ITZs. Pozzolanic materials are readily available as industrial by-products and are environmentally friendly.
- (7) Compared with NMA, different improved mixing methods, such as TSMA, SEMA, MMA, TM and OTM, pre-mix RCA to form a low w/c cement coating or pozzolanic coating on the surface of the RCA, which can better fill the pores of RCA and dense ITZs. Using TSMA to coat SF on the surface of RCA is beneficial to the production of self compacting concrete.
- (8) CaCO₃ precipitated by bacteria on the surface of RCA reduces the porosity of RCA and improves new ITZ. CaCO₃ biodeposition is not harmful to the environment, and the resulting CaCO₃ on the microstructure of RCA is expected to be further studied.

- (9) RCA is usually carbonized under the accelerated carbonation condition. CO_2 reacts with $\text{Ca}(\text{OH})_2$ and C-S-H to produce CaCO_3 that reduces the porosity of RCA and improves ITZs. Carbonated RCA is encouraged to be used for reinforced concrete, and carbonation is beneficial to environment because it can absorb CO_2 .
- (10) Methods of removing the attached mortar are suitable for CRCA, because the proportion of attached mortar of CRCA is lower than that of FRCA. Methods of using polymer emulsions and CaCO_3 biodeposition are currently only found that the improvement of CRCA microstructure, while three methods of using pozzolanic materials, such as SF, FA and slag, different mixing methods and accelerated carbonation can improve the microstructure of FRCA and CRCA.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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