



Review

# A Review of Improvement of Interfacial Transition Zone and Adherent Mortar in Recycled Concrete Aggregate

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Abstract: An increasing amount of construction and demolishing waste has made the implementation of recycled concrete aggregate (RCA) a popular topic; RCA can reduce the environmental pollution caused by the construction industry. This paper describes differences in physical, mechanical, and chemical properties between RCA and natural aggregate (NA). For the first time, the methods of interfacial transition zone (ITZ) improvement in RCA, including strengthening through carbonation, incorporation of mineral admixtures, and different mixing approaches, are extensively covered. In addition, the methods used to improve the adherent mortar regions of RCA are covered. This approach makes it possible to demonstrate the impact of different methods on these regions (ITZ and adhered mortar) and overall RCA enhancement. Lastly, a comparison of each of these methods in terms of their effectiveness is presented. The review of several studies concludes that the carbonation treatment method for ITZ and adherent mortar enhancement are the most efficient and sustainable methods compared to others. Summarizing, using RCA instead of NA provides a sustainable solution for the construction industry due to reducing the amount of produced waste and conserving landfill space.

**Keywords:** recycled concrete aggregate; interfacial transition zone (ITZ); recycled concrete; adherent mortar; microstructure

## 1. Introduction

During the last two decades, the awareness of ecofriendly lifestyles has increased, thus changing different policies to meet environmental standards. Increasing urban growth and worldwide consumption of natural resources accelerated the amount of waste and emissions. The construction industry is considered one of the largest carbon dioxide emitters, being responsible for up to 45% of total CO<sub>2</sub> emissions and significantly polluting the environment [1]. Several attempts have been made to raise awareness about the importance of sustainability and particularly the topic of construction waste recycling. This can be explained by the fact that there is a shortage or inconsistencies in construction materials supply, together with rising demand on limited land resources used as disposal places for construction wastes. The issue concerning the massive amount of waste produced during construction and the destruction of buildings led to the beginning of enhanced research and studies regarding recycled aggregate (RA) and its application in concrete production. The implementation of RCA has attracted much attention as an alternative material for conventional concrete from an environmental perspective. However, the properties of recycled concrete aggregate are distinctive from natural aggregates (NA) [2] as it contains two additional components, adhered mortar and an interfacial transition zone (ITZ) between the used natural aggregate and the original cement. Literature shows that the properties of concrete are influenced by RCA [3].

RA has different properties compared to NA that cause different behavior in concrete mixes and in hardened concrete, thus performing differently from conventional concrete [4].



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**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). The properties of concrete made from 100% RA are worse than that for NA, even at the same w/c ratio. For instance, the bulk density of fresh concrete made with NA is approximately 2400 kg/m<sup>3</sup>, while the bulk density of concrete with RA is equal to 2150 kg/m<sup>3</sup>, which is lighter due to the lower bulk-specific density of the RA as well as the increased air content [5]. Therefore, implementing RCA can affect the properties of the new concrete that contains it, which is all related to the components of the RCA.

The three main components of concrete are aggregate, hardened cement paste, and ITZ. The ITZ is the weakest region in the hardened concrete [6]. Mainly, RCA consists of a large quantity of ITZ because NA is covered by the original ITZ, adherent mortar, and new ITZ; the new ITZ is between the adherent mortar and the new mortar [3]. Therefore, to improve the performance of RCA, the enhancement of ITZs together with the adherent mortar region is required. Numerous researchers investigated the use of different approaches and methods for the improvement of concrete characteristics with RCA addition. These include removing the adhered mortar or strengthening RCA using carbonation treatment [7], mineral admixtures [8], calcium carbonate biodeposition [9], sodium silicate solution [10], and water-repellent coat by polymer treatment [11]. Several review papers are available on improving the performance of RCA. For instance, the review paper written by Shaban et al. [12] divided the treatment techniques for eliminating adherent mortar and its quality improvement as was previously performed by Shi et al. [3]. Similarly, Verian et al. [13] compiled different treatment methods that can improve RCA without mentioning which zone of new concrete was improved, making it difficult to understand which method can improve ITZ or adherent mortar or both. Wang et al. [14] also covered different improvement methods of RCA properties that included removal of attached mortar in the RCA, surface coating of RCA, different mixing methods, and incorporation of calcium carbonate. However, the article focuses on improving the microstructure of the RCA without dividing enhanced regions. In comparison to other review papers available on the improvement of RCA, this paper conducts a review of different treatment methods for ITZs and adherent mortar improvement separately. In this way, the effect of each method on the improvement of these regions (ITZs and adherent mortar) is known and which are not previously covered by other review papers.

This article focuses on the difference in the properties between NA and RCA together with different methods for improving the performance of RCA using existing literature and research. Unlike the existing literature, this overview includes a description of the improvement methods aimed explicitly for ITZ and adherent mortar, thus demonstrating how different methods affect them separately and in terms of overall RCA enhancement. The review paper contains a comparative analysis of natural and recycled concrete aggregate properties in terms of physical, mechanical, and chemical properties (Section 2). Comprehensive information and research findings about ITZ improvement by carbonation treatment, incorporation of mineral admixtures, and implementation of different mixing approaches are included in Section 3. Section 4 presents information about the improvement of adherent mortar in RCA using surface solidification (carbonation, pozzolana addition, calcium carbonate biodeposition, and sodium silicate solution) and water-repelling coat by polymer treatment. Finally, the discussion and comparative analysis of all the methods noted above are covered in Section 5.

#### 2. Comparison of NA and RCA Properties

There are differences in the physical, mechanical, and chemical properties of NA and RCA. The quality of parent concrete, curing conditions, and recycled concrete aggregate size, nature, and moisture conditions influence the properties of RCA [15]. About 25 to 60 percent volume of the RCA is the residual mortar, which influences the impairment of original characteristics [15]. This section compares the physical, mechanical, and chemical properties between RCA and NA.

#### 2.1. Physical Properties

Specific gravity, bulk density, size, shape, pore volume, texture, and absorption of RCA are related to the physical properties. Generally, they are inferior in comparison to the natural aggregate. The shape and texture of RCA are mostly angular and rough, whereas natural aggregate has a variety from rounded and smooth to angular. The angular and rough surface results from crushing and demolishing the construction building or waste. According to Safiuddin et al. [16], the specific gravity (SG) and bulk density of RCA are lower because of the porous nature of recycled material. The volume of pores and absorption capacity are significantly higher because of the cracks. Padmini et al. [17] found that the specific gravity of the RCA is significantly less than NA with higher strength of original concrete from which the RCA is produced. Owing to the strong bond between the aggregate and the mortar phase in the high-strength samples, the amount of adhered mortar on the recycled aggregate after crushing is greater than the breaking of lower-strength aggregate. The presence of larger quantities of comparatively lower density parent mortar on RA results in a greater decline in SG. Consequently, water absorption in these types of aggregate is relatively greater than natural aggregate. In the case of crushing of lowerstrength parent concrete, the specific gravity is higher because a larger amount of mortar is detached due to a weak bond. In the experimental study of Lopez-Gayarre et al. [18], the recycled aggregate with 34.2% adhered mortar has lower density and higher absorption rather than a sample with 23% of old mortar. The results of Katz [5] revealed the same pattern of changing properties of a recycled sample with the amount of adhered mortar and additionally showed that characteristics vary marginally with different crushing ages. In summary, the water absorption (WA) of RCA is between 1.93% and 7.77%, the specific gravity of RCA varies from 2.31 to 2.6, and the density of RCA is in the range 1433-2628 kg/m<sup>3</sup>, whereas the WA of natural aggregate is 0.22–1.63%, the SG of natural samples is 2.62–2.86, and the density of NA is 1733–2670 kg/m<sup>3</sup>. A summary of the physical properties of RCA and NA from various studies is presented in Table 1. Overall, the results demonstrate that the properties of NA show superiority to RCA.

<b>Physical Properties</b>	Values RCA	Values NA	Reference
Specific gravity	2.38–2.56	2.8	[1]]
Water absorption	2.2–5%	0.3%	- [1/]
Water absorption	3.47%	0.68%	
SSD specific gravity	2.39	2.86	[19]
Specific gravity oven dried	2.31	2.84	-
Appearance	Rough and porous surface texture	Smooth and round surface texture	
SSD specific gravity	2.51	2.75	[20]
Water absorption	3.92%	1.129%	-
Water absorption	4.2%	1.1%	
SSD density	2445 kg/m <sup>3</sup>	2670 kg/m <sup>3</sup>	[21]
Shape index	21.4%	17.8%	-
Water absorption	2.47-4.61%	0.64–0.74%	[22]
Aggregate density	2284–2585 kg/m <sup>3</sup>	2471–2640 kg/m <sup>3</sup>	- [22]
Dry density	2.36 and 2.2 g/cm <sup>3</sup>	2.68 and 2.69 g/cm <sup>3</sup>	[10]
Water absorption	3.8% and 5%	0.22% and 0.27%	- [18]

Table 1. Comparison of physical properties between RCA and NA.

<b>Physical Properties</b>	Values RCA	Values NA	Reference
Density	$2.5  {\rm g/cm^3}$	2.58 g/cm <sup>3</sup>	[22]
Water absorption	1.93	1.63	[23]
Shape and texture	Angular and irregular particle shape with rough surface	Rounded and smooth appearance	
Bulk density	1458 kg/m <sup>3</sup>	1733 kg/m <sup>3</sup>	[24]
Water absorption	3.98% and 5.72%	1.54%	
SSD specific gravity	2.31	2.66	
Specific gravity oven dried	2.45	2.7	
Specific gravity	$2.36-2.54 \text{ g/cm}^3$	2.6 g/cm <sup>3</sup>	[25]
Water absorption	3.13–7.77%	0.9–1.24%	[23]
Density	2.49–2.58 kg/m <sup>3</sup>	2.62 kg/m <sup>3</sup>	
Water absorption	3.52-4.26%	1.11-1.12%	[2]
Porosity	8.69%	1.62%	
Density	2256–2628 kg/m <sup>3</sup>	_	[26]
Water absorption	1.5-8.7%	_	[20]
Bulk specific gravity	2.55-2.60	_	
Bulk density	1433–1462 kg/m <sup>3</sup>	_	[5]
Water absorption	3.2-3.4%	_	

Table 1. Cont.

#### 2.2. Mechanical Properties

Fundamental mechanical properties such as abrasion value, crushing value, and impact value of recycled concrete aggregate are inferior to natural aggregate [16]. They are taken into account in this part of the review to make a comparison of mechanical properties between RCA and NA. The maximum allowable abrasion value is 50% of the aggregate weight, and it is valid for both types, recycled and normal aggregate [16]. The magnitude of abrasion value for RCA, which was mostly determined by the Los Angeles abrasion test in the majority of studies, is in the optimal range from 29% to 48%. This value is less than the maximum acceptable value (50%). However, the test results are higher compared to NA. The ability to withstand compressive strength, named the crush resistance of aggregate, is determined by aggregate crushing value. Recycled aggregate has a relatively higher value due to weak ITZ and adhered mortar, which is not observed in natural aggregate [27]. High value indicates the weakness of aggregate. The ability to withstand the dynamic load is evaluated based on the aggregate impact value. Generally, the RCA has a greater percentage of loss in mechanical experiments than NA because weak ITZ of RCA leads to breakoff due to residual mortar. The lower the value, the stronger is the aggregate. The mechanical properties of RCA and NA from various papers are enlisted in Table 2. The literature clearly shows RCA is inferior to those of NA in terms of physical and mechanical properties.

Table 2. Comparison of mechanical properties between RCA and NA.

Properties	Values RCA	Values NA	Reference
Crushing value	23–32%	22–25%	
Impact value	21–38%	17–18%	[17]
LA abrasion value	29–48%	26%	

Properties	Values RCA	Values NA	Reference	
LA abrasion value	38.8%	21.56%		
Impact value	35.81%	17.37%	[20]	
Crushing strength	115.3 kN	231.3 kN		
LA abrasion value	33.1–37.2%	24-26.4%	[18]	
Micro-Deval abrasion loss	15.1 and 22.1%	11.9%	[24]	
Crushing value	23.1 and 26%	18.2%	— [24]	
Crushing value	22.5-23.9%	21.7%	[25]	
LA abrasion value	15–34%	-	[26]	
LA abrasion value	20–45%	11.5–38.9%	[13]	
LA abrasion value	28–33%	24%	[28]	
LA abrasion value	33.5%	19.8%	[29]	
Crushing test	24%	13%	[4]	
LA abrasion value	32%	11%	[1]	

Table 2. Cont.

## 2.3. Chemical Properties

The amount of chemical compounds in recycled concrete aggregate is mainly influenced by the nature of the parent concrete and the amount of the adhered mortar. The comparison of chemical properties in this section considers chloride, sulfate, and potassium content. As investigated by Verian et al. [13], the potassium content of RCA is almost eight times greater in comparison to NA, which may lead to a high-risk alkali–silica reaction (ASR). The obvious increase in chloride content in the RCA is due to the nature of parent concrete. The concrete that was demolished for the production of RCA, described in Rahal [19], was obtained from the subtropic climate zone, Hawally area, Kuwait, while chloride-based deicers were applied to the sample in Verian et al. [13]. These are the main reasons for an increase in the percentage of chloride relative to natural aggregate. Consequently, the probability for steel corrosion rises considerably. However, sulfate content has an absolutely opposite relationship in the recycled aggregate. The amount of sulfate ions is lower in contrast to natural aggregate. Table 3 presents the distinction in the chemical content of NA and RCA.

Chemical Properties	Values RCA	Values NA	Reference
Chloride content	0.3%	0.14%	[19]
Potassium ion ppm	239 ppm	30–32 ppm	
Chloride ion ppm	851 ppm	377–395 ppm	[13]
Sulfate ion ppm	39 ppm	106–120 ppm	
Chloride content	0.0012-0.0016%	-	[20]
Sulfate content	nt 0.0025 -		[30]
Sulfate content	0.01–0.03	0.02	[31]

Table 3. Comparison of chemical properties between RCA and NA.

In general, it can be stated that the physical and mechanical properties of NA are superior to those of RCA. It can be explained by the microstructure of the RCA, the presence of a weak ITZ, and adherent mortar. Nevertheless, the chemical properties of RCA are directly dependent on the content of the parent concrete and the adherent mortar which is left on the aggregate. Hence, it is difficult to determine the exact chemical properties of RCA. Yet, the overall properties of RCA can be improved by enhancing the properties of ITZ and adherent mortar through different improvement methods.

## 3. Improvement of ITZ of RCA

The presence of pores in the old ITZ and cracks that remain in RA due to the crushing process makes old ITZ weak. According to published literature, the new ITZ, which connects RCA with the new mortar, is also a weak link because of the presence of areas with high water content averting the formation of a strong bond between RCA and the new cement paste [32]. In the case of the new ITZ, the regions that affect the high water absorption and porosity of RCA are composed of loose hydrated particles [33]. Improving the performance of ITZ enhances the overall performance of RCA. It can be accomplished by filling in minute pores and microcracks in the ITZ. This section discusses the improvement of old and new ITZ through carbonation, incorporating mineral admixtures, and different mixing approaches.

#### 3.1. Carbonation Treatment

Carbonation is one of the methods which can be used to decrease the porosity and enhance the performance of ITZ. In this process, CO<sub>2</sub> can penetrate into the pores of the RCA and further react with CH and C–S–H to fill fissures [34]. The treatment can be conducted in different ways, including pretreatment, changing pressure, etc.

Li et al. [6] analyzed the influence of the carbonation treatment by comparing the properties of RCAs before and after treatment. For this purpose, the RCA was obtained from a parent concrete with w/c equal to 0.36. The mixture proportion contained water, cement, sand, and natural aggregate with 161, 445, 702, and 1098 kg/m<sup>3</sup>, respectively. The percentage of air-entraining agent was about 0.03, while compressive strength at 28 days was equal to 55 MPa.

The samples of RCA were preconditioned in a drying chamber to remove excessive moisture content before the carbonation process. Afterward, the carbonation of the samples was carried out in a standard carbonation chamber. A 1% phenolphthalein alcohol solution was sprayed every 12 h on carbonated recycled concrete aggregates (CRCA) to determine if a reaction had occurred. The physical properties of RCA and CRCA are presented in Table 4. In the study, curing of samples was performed for 3 days at 70 ± 5% relative humidity and the laboratory-maintained 23 ± 2 °C temperature. The Vickers microhardness and SEM analysis were conducted to evaluate the effect of carbonation treatment.

Samples	Size (mm)	Apparent Density (kg/m <sup>3</sup> )	Water Absorption (%)	Porosity (%)
	20–30	2635	4.67	16.52
RCA	30–40	2627	4.60	16.35
	>40	2610	4.21	15.72
	20–30	2677	3.14	9.45
CRCA	30–40	2651	3.42	9.52
	>40	2623	3.80	9.74

Table 4. Properties of RCA and CRCA [6]. Reproduced with permission from [6], Elsevier, 2019.

During crushing pattern examination, some differences between RCA and CRCA were identified. In total, four different stages in the load–displacement curve were observed. During stage 1, when a load was applied to the original RCA, the surface was crushed, while in stage 2, the ITZ collapsed, causing the separation of mortar and NA. In stage 3, the mortar was crushed; finally, in stage 4, the aggregate was crushed with an increase in loading. CRCA samples also showed four stages in the load–displacement curve. Stage 1 was similar to that of RCA, where the surface crushed. However, the difference was during

stage 2, where the mortar connected to the RA was crushed first. Later, in stage 3, ITZ was crushed with further crushing of the NA during stage 4. The difference between the crushing patterns could be explained by the enhancement of ITZ after the carbonation treatment that increased density and crushing stress.

The results of microhardness for RCA and CRCA are presented in Figure 1. The microhardness of the recycled aggregate increased from 88.6 to 108.7 after carbonation treatment. Consequently, Figure 1 shows that the microhardness of the ITZ around CRCA was higher than the ITZ around RCA, which also explains the difference between crushing patterns. The measured microhardness of the treated samples (CRCA) in ITZ was 31% higher in comparison to the original RCA, with an average of value equal to 71.8 for CRCA and 56.7 for original RCA. It can be referred to as the lower WA of the CRCA sample resulting in a relatively lower w/c ratio around the CRCA that helps to improve the ITZ. Similarly, the microhardness of CRCA also increased in the adhered mortar. However, the increase was only 17.4%, which is significantly lower compared to the improvement in ITZ. It can be concluded that the improvement in ITZ was greater than the adhered mortar after the carbonation treatment. Thus, compared to the adhered mortar on the surface, the carbonation treatment significantly strengthened the ITZ.



Figure 1. Vickers microhardness around RCA and carbonated RCA [6]. Reproduced with permission from [6], Elsevier, 2019.

In the same research, the SEM of the attached mortar and ITZs in the RCA and CRCA were compared. Needle-like ettringite can be observed in the adherent mortar of RCA, while no crystals were observed after carbonation due to their complete disappearance. In addition, numerous micro cracks, voids, and loose hydrated products resulted in poor bonding between the attached mortar and NA (Figure 2a). In contrast, Figure 2b reveals densified ITZ with hydration products such as CaCO<sub>3</sub>, which improved its microhardness. Thus, SEM and Vickers microhardness tests confirm the improvement of ITZ in CRCA. Overall, the main finding was that the carbonation treatment improved the original ITZ together with the adherent mortar in the RCA.

Zhang et al. [35] aimed to improve the properties of recycled gravel concrete aggregate (G-RCA) and recycled crushed stone concrete aggregate (C-RCA). G-RCA and C-RCA were derived from beams having compressive strength equal to 30 and 50 MPa, respectively. Carbonation treatment was performed at relative humidity, temperature, and  $CO_2$  concentration of  $60 \pm 5\%$ ,  $20 \pm 2$  °C, and  $20 \pm 2\%$ , respectively. In the experiment, no pretreatment procedure was adopted. However, after a certain period of time, the speci-



mens were sprayed with a 1% alcoholic solution of phenolphthalein to distinguish between carbonated and noncarbonated portions.

**Figure 2.** SEM images of (**a**) attached mortar and (**b**) ITZ in RCA before and after treatment [6]. Reproduced with permission from [6], Elsevier, 2019.

The reaction between CO<sub>2</sub> with the hydration products of cement and unhydrated cement particles, both recycled samples became denser by 4.7–5.6%. Carbonation treatment improved the properties of G-RCA and C-RCA, thus decreasing water absorption by 7.6–9.6% and crushing value by 22.6–28.3%. Consequently, the percentage of mechanical crushing value on average declined by 20%, and the proportion of water absorption lowered by 23–28%. Decreased water absorption value reduced the thickness of the water film, thus enhancing the original ITZ of RCA. SEM images demonstrated that the carbonated sample had a denser ITZ, and fewer large pores between the aggregate and cement paste compared to the porous noncarbonated aggregate (Figure 3). Overall, the original ITZ and the newly formed ITZ in mortar were improved, as determined by SEM examination.

Shi et al. [36] evaluated the influence of carbonation on the densification of new and old ITZ. The experiment was carried out at relative humidity, temperature, and  $CO_2$ 

concentration of 70  $\pm$  5%, 20  $\pm$  3%, and 20  $\pm$  2 °C, respectively. The surface cleaning and spraying with 1% phenolphthalein solution were used to measure the carbonation depth. The carbonation-treated RCA resulted in the formation of both fiber-like and cotton-like materials, which further filled voids and densified ITZ between RA and adhered mortar. The densification of ITZ after carbonation treatment is shown in the micrographs presented in Figure 4.



Figure 3. SEM image of (a) uncarbonated RCA; (b) carbonated RCA [35]. Reproduced with permission from [35], Elsevier, 2015.

The microhardness of different samples was also investigated, including a reference sample (R), RCA after carbonation treatment (R-C), RCA treated by fly ash (R-F), silica fume (R-S), and nano-SiO<sub>2</sub> slurries (R-N-S). The microhardness results shown in Figure 5 indicate five different regions: aggregate, old ITZ, old matrix, new ITZ, and new matrix. The reference sample showed the lowest microhardness values compared to the ones treated with pozzolana slurry and  $CO_2$ . Overall, the findings of the research indicated that RCA treatment strengthened and resulted in denser new and old ITZs, leading to improved mechanical properties and durability.



**Figure 4.** SEM images of (**a**) old ITZ in RCA; (**b**) carbonated RCA [36]. Reproduced with permission from [36], Elsevier, 2018.



Figure 5. Microhardness of new and old ITZ [36]. Reproduced with permission from [36], Elsevier, 2018.

Xuan et al. [37] conducted another study in which the authors compared and evaluated the performance of RCA extracted from a designed concrete (NRCAs) and from an old type of RCA (ORCA) obtained from demolished old buildings. The original NCRA mix proportion included 460 kg/m<sup>3</sup> cement, 205 kg/m<sup>3</sup> water, w/c ratio equal to 0.45, 430 kg/m<sup>3</sup> of 5–10 mm natural aggregate, 530 kg/m<sup>3</sup> of 10–20 mm natural aggregate, 700 kg/m<sup>3</sup> of sand, and 3.47 kg/m<sup>3</sup> of superplasticizer. The NRCA concrete was crushed at a construction waste recycling plant after a six-month external curing period. Before carbonation treatment, the recycled samples was preconditioning in a drying chamber at  $25 \pm 3$  °C and  $50 \pm 5\%$  relative humidity to remove excess moisture content. This pretreatment procedure controls the optimal moisture of samples in the range 40–70%, hence accelerating the carbonation process. The carbonation treatment of aggregates was carried out in a chamber for 24 h with a carbon dioxide concentration of approximately 100% at a pressure of 0.1 and 5 bar. In the study, the w/c ratio for the concrete with RCA including NRCA and ORCA was equal to 0.55, while the cement content for the mix proportion was  $325 \text{ kg/m}^3$ . The dosage of the superplasticizer was increased with the increment of RCA to maintain the slump value from 150 to 200 mm.

The carbonation results for the sample with NRCA, carried out at 5 bar pressure, showed a slight increase in density, crushing value, and CO<sub>2</sub> uptake compared to 0.1 bar. WA percentage decreased by 1% and 1.1% at 0.1 and 5 bar, respectively. Thereby, the crushing value of the RA changed from 27.8% to 21.9% at 0.1 bar pressure and to 20.6% at 5 bar. However, the carbonated ORCA demonstrated less improvement on the properties. The microhardness results in Figure 6 showed that the microhardness of carbonized NRCA was equal to 97  $\pm$  9 and higher than that of noncarbonated NRCA (87  $\pm$  17). As a result of the lower WA, the microhardness of new ITZ in the carbonated NRCAs was higher than that of the uncarbonated sample. Hence, the physical, as well as the mechanical properties of the concrete sample with treated NRCAs, were improved. Overall, carbonation treatment with different pressure implementation had a lesser impact on the enhancement of RCA when subjected to a longer duration of treatment (i.e., 24 h), but generally, carbonated NRCAs demonstrated higher microhardness of the new ITZ than the one around the noncarbonated sample.



**Figure 6.** Vickers microhardness of new ITZs before and after treatment of NRCAs [37]. Reproduced with permission from [37], Elsevier, 2016.

The effect of carbonation treatment on new ITZ of RCA was also investigated in detail by Zhan et al. [38]. For this purpose, the authors used samples with new cement mortar against the carbonated hydrated cement paste (HCP). To prepare different types of RCA, the w/c ratios 0.25 and 0.40 for HCP were used. Cubic samples of HCP were placed into a  $CO_2$  curing chamber under internal pressure of 1 bar with no temperature regulation but a relative humidity maintained at about  $54 \pm 5\%$ . Afterward, a prepared new cement mortar having a design strength of 50 MPa was used to fill the space between RCA and the interior wall of the mold with the help of mechanical vibration to ensure complete filling. Mostly, the formation of ITZ is caused by the wall effect and an aqueous film that forms around the aggregate during the fresh state. Consequently, this leads to a less efficient filling of the space with hydration products near the aggregate, and hence there is a higher chance in developing porosity in this area. Therefore, in this research, the authors focused on the porosity profiles within the ITZ.

Figure 7 demonstrates the results of the porosity profile of ITZ against the distance from the aggregate surface. The average detectable pore contents were found to be approximately 25% in the bulk pastes. For RCA at nearly 10  $\mu$ m, the average porosity was up to 49.1% in RCA040, while in RCA025 it was around 47.3%. When comparing the porosity for the ITZs of carbonated RCA and noncarbonated RCA, it was found that both values are similar, except that the porosity of treated RCA at the innermost 10  $\mu$ m was slightly lower compared to the untreated sample. This phenomenon indicates that RCA carbonation seems unable to enhance the microstructure properties in terms of porosity.



**Figure 7.** Porosity of ITZ at different distances from the aggregate [38]: (**a**) RCA040 and carbonated RCA040; (**b**) RCA025 and carbonated RCA025. Reproduced with permission from [38], Elsevier, 2020.

Yue et al. [39] analyzed the effect of carbonation treatment by using Vickers hardness values for the ITZ to measure variations in the width and microhardness of old slurry–new slurry (LJ–XJ). During the experiment, the authors used cement with 50.7 MPa compressive strength, a w/c ratio of 0.35, and cylindrical samples 20 mm in depth.

In order to analyze the sample and easily identify the regions, the beat method (matrix point) was used. To conduct the experiment, nine regions were chosen with a  $4 \times 5$  lattice. To avoid the discrepancy in the microhardness of the ITZ and the mortar zone caused by the surface roughness, the unhydrated cement clinker, and the distribution of fine aggregates, researchers used a box diagram. With its help, the outliers in the RBI data can be eliminated, thus allowing for standard hardness identification of the mortar zone.

Figure 8 demonstrates results from the microhardness test of the LG–XJ sample. It can be observed that the microhardness of ITZ increased while the width was reduced after 28 days. Before carbonation, the microhardness of ITZ was around 125–171 MPa and the width was equal to 70–80  $\mu$ m. After carbonation, however, microhardness was increased to 144–182 MPa, and width was decreased by 20  $\mu$ m. It can be said that RCA without carbonation has a looser ITZ and, additionally, its concrete has a greater number of pores and cracks [40]. As a result of carbonation treatment, it can be concluded that the reaction enhances the production of CaCO<sub>3</sub> and thus improves the microstructure of ITZ and decreases its porosity [40].





Figure 8. Effect of carbonation treatment on the microhardness of ITZ [39]. Reproduced with permission from [39], Hindawi, 2018.

To summarize, during the carbonation procedure, the reaction products not only densified the surfaces of RA, but also penetrated into RCA and improved the ITZ. Consequently, RCA itself becomes stronger by having higher microhardness and enhanced properties, which suggests that the carbonation treatment improves the properties of RCA. Carbonation treatment is an environmentally friendly method that can produce a large amount of CaCO<sub>3</sub> and enhances the ITZ region.

#### 3.2. Incorporation of Mineral Admixtures

The incorporation of mineral admixtures is one of the alternative methods to improve ITZ of RCA. The enhancement proceeds through the pozzolanic reaction that forms new C–S–H products and densifies ITZ by filling voids [3]. According to Katz [33], admixtures work as microfillers in the ITZ between the aggregate surface and the matrix. In this section, the role of mineral admixtures in improving the performance of ITZ is discussed.

One of the mineral admixtures that can be used to enhance the performance of ITZ is silica fume. It reacts with calcium hydroxide and forms a denser layer that covers the surface of the aggregate and strengthens ITZ. In a study conducted by Katz [33], RCAs were produced from three types of concrete with w/c ratio of 0.77, 0.53, and 0.35, which represented three strength levels: low, medium, and high, respectively. These samples were then crushed to obtain RCA particles. In this experiment, the raw silica fume was used with a superplasticizer to treat RCA samples, where the superplasticizer took the role of a regulation tool to disperse 1 kg of silica fume in 10 L of water. Test results showed that after presoaking for 24 h in a solution and oven-drying, the weight of RCA increased by 0.5–0.8%. The saturated silica fume considerably decreased the number of loose crumbs on the RCA and made it denser, as revealed from the obtained SEM image (refer to Figures 2 and 4 in Reference [33]).

The incorporation of silica fume into RCA helps to enter silica fume particles in loose layers of RCA and into the cracks. Owing to the filler effect, this layer enhances the ITZ during the concrete hardening process. Additionally, the improvement of the weakened RCA region is due to a pozzolanic reaction between silica fume and portlandite. This improved RCA region is further distributed from the NA and the old cement paste to the new cement zone. Impregnation of silica fume particles provides a stronger impact at an early age in comparison to slowly developing pozzolanic reactions. However, the products of the pozzolanic reaction, which were formed inside the aggregate, to some extent, strengthened the new ITZ between RCA and the new cement matrix, and improved the properties of the old paste. The covered layer decreased the WA capacity and increased the density of the sample by penetration of C–S–H into the new ITZ, making it denser.

Shi et al. [36] used silica fume (SF) slurry, fly ash (FA) slurry, and nano-SiO<sub>2</sub> (NSi) slurry with a water-to-solid ratio of 10:1, 10:1, and 20:1, respectively, to enhance the ITZ of RCA. Ultrasonication was implemented for mixing the slurries to uniformly disperse the particles. RCA was derived from concrete specimens having compressive strength equal to 30 MPa and a particle size less than 4.75 mm. For treatment, RCA was dried for 48 h at 60 °C and then combined with slurry for 0.5 h and soaked for 1 h. After the treatment procedure, all samples were dried for 120 min at 100 °C for further analysis. The investigation of specimens showed that aggregate with the incorporation of fly ash had the largest amount of absorbed pozzolanic products. This was due to dynamic viscosity for admixtures, which was the lowest for fly ash and the highest for nano-SiO<sub>2</sub>. After treatment, the microhardness and density of RCA increased. The increased magnitudes of microhardness in the old ITZ were 67.7%, 55.6%, 32.2% by silica fume, nano-SiO<sub>2</sub>, and fly ash, respectively. Additionally, the strengthening effect was more evident in new ITZ, and a higher increase in microhardness percentage was identified. Therefore, the impregnation of RCA by silica fume was more beneficial because it showed a higher increase in mortar microhardness and densified ITZs.

The ITZ can also be improved by using different combinations of mineral admixtures. The enhancement in ITZ performance depends on the type and amount of mineral admixture used and their relative ratios with respect to each other. Wang et al. [41] evaluated the performance of phosphorus slag (PHS), ground granulated blast-furnace slag (GGBS), and FA in various combinations, i.e., 20% PHS (P2), 10% PHS + 10% FA (P1F1), and 10% PHS + 10% GGBS (P1S1). For this experiment, RCA was obtained from the debris of a cement pavement from the town of Hangzhou with a particle size gradation from 0.005 to 0.02 m and WA equal to 3.63% by mass. The concrete cores obtained from the pavement had a compressive strength of 30 MPa. According to the test results, all the treated samples showed improved performance. The mixture of PHS and GGBS was found to be most effective. The improved performance of PHS and GGBS combination can be associated with their simultaneous reaction, which results in a more compacted aggregate with denser new ITZ. In comparison to PHS and FA, GGBS has a higher surface area and it reacts quickly to form pozzolanic products. For the results of the microstructure of RCA treated and untreated samples after 90 days of curing, please refer to Figures 4–7 in Reference [41]. There are many cracks and voids inside the old ITZ (refer to Figure 4 in Reference [41]); however, specimens made from P2, P1F1, and P1S1 showed significant improvement. In particular, the sample with the addition of GGBS showed the greatest effectiveness in enhancing the microstructure of the old ITZ (P1S1) compared to P2 and P1F1. It can be seen that the formation of C–S–H and the additional ettringite fills the empty space, thereby increasing compaction [41].

Summarizing the abovementioned results obtained from the incorporation of single and multiple mineral admixtures, it can be stated that for better impregnation, superplasticizer and/or ultrasonication should be used. Among conventional mineral admixtures, silica fume showed the best performance in improving ITZ and making it denser. In the case of multiple admixtures, the mix of 10% PHS and GGBS was the most effective among different combinations of mineral admixtures (PHS, FA, and GGBS). Therefore implementation of mineral admixtures contributes to the filling of pores and voids and the formation of C–S–H gel, which leads to strengthening of the ITZ region. It should be noted that the incorporation of mineral admixtures has a lower effect in improving the microhardness of treated RCA than carbonation treatment.

#### 3.3. Mixing Approaches

One of the ways to improve the ITZ is to adopt different mixing strategies. Improving ITZ by adopting different mixing strategies means that the conventional mixing steps are modified to affect the RCA during or at the start of mixing to improve its performance through better microstructure. The mixing approaches are named according to the number of steps in relation to normal/conventional mixing. There are mainly three types of mixing

for RCA concrete. These are the normal-mixing method (NM), double-mixing method (DM) or two-step mixing (TSMA), and triple-mixing method (TM).

Tam et al. [42] investigated the effect of the two-step mixing approach on the microstructure of RCA. The difference between normal and two-step mixing was in the proportionally splinted required water and sequence of materials utilized during mixing. Figure 9 represents the detailed steps of both mixing approaches used during the experiment.



**Figure 9.** Mixing procedure of the (i) normal mixing and (ii) two-stage mixing approach [42]. Reproduced with permission from [42], Elsevier, 2005.

In the two-stage mixing approach, half of the water is firstly used to form a thin-coated layer of cement slurry on the RCA [42], which reduces the high absorption capacity of RCA and fills the minute pores and cracks in RCA. Thereafter, the remaining water was used to finish the mix procedure of the concrete. Figure 10b shows a clear densification of the old ITZ after implementation of TSMA, whereas there are noticeable gaps in the microstructure RCA concrete made with normal mixing (NMA) (Figure 10a). The experiment using the two-stage mixing approach was found to be an effective approach to enhance the mechanical properties by developing a stronger and denser old ITZ.

The SEM images of new ITZ obtained after TSMA and NMA presented in Figure 11 shows that both new and old ITZ were improved. The SEM image of TSMA treatment concrete demonstrated an improvement in the new ITZ (Figure 11a), which is stronger and denser compared to NMA. Thus, the authors pointed out that a two-step mixing approach can improve both new and old RCA ITZ.

Another two-step mixing approach developed by Kisku et al. [43] followed the same concept of dividing normal mixing into two main stages. However, the authors added silica fume slurry during the first steps of mixing. The method of mixing design was based on equal mortar volume (EMV), which means that the required amount of mortar in two-stage mixing was identical to the amount in the normal mixing process. Details of the TSMA procedure is shown schematically in Figure 12.

The RCA was derived from a 45-year-old demolished building located in Dhanbad, India. The water absorption, crushing value, and moisture content were equal to 2.54%, 34.8%, and 1.3, respectively. To demonstrate the influence of the treatment on the ITZ of the RCA, backscattered secondary electron (BSE) image analysis was used. All common characteristics such as voids, cracks, and unhydrated residue inherent in recycled concrete aggregates are visible in Figure 13a. The slurry made of cement and SF added during the initial stage of mixing was able to penetrate into the RCA. Consequently, the ITZ was densified by the interaction of the clinker compound hydration and the deposition of hydrated products. Figure 13b demonstrates denser texture of the old ITZ. Therefore, the EMV method of mix design and application of TSMA resulted in an improved and densified ITZ between NA and adhered mortar in RCA.





Kong et al. [44] compared three mixing approaches, presented in Figure 14. For the experimental investigation, natural river sand with water absorption of 1.6%, coarse RA derived from concrete having a compressive strength of 40 MPa, and FA with finely ground slag (S) as pozzolanic materials were used. The procedure for DM was adopted from the research conducted by Tam et al. [42]. For TM, wet aggregates were first obtained by mixing coarse and fine aggregates with a certain amount of water for 15 s. Then, the admixture was added and mixed for 15 s so that the surfaces of the aggregate are coated with the admixture. During the next 30 s, cement was added to the mix. Lastly, a superplasticizer with the rest of the water was added, and mixing was continued for 60 s. Cubic shapes of 100 mm were prepared for experimental tests. By applying TM, coating the RCA with a thin layer of pozzolanic particles in the first mixing stage was achieved. Figure 15 shows the admixtures not only filled cracks and pores of the RA with fine particles, but also formed new hydration products by reacting with accumulated CH at the curing age of 1 and 3 d, thus improving the properties of the ITZ. Additionally, the surface treatment could affect the density increase in the cement matrix around the RA.







Figure 12. Steps of TSMA mixing [43]. Reproduced with permission from [43], Elsevier, 2020.







**Figure 13.** (a) Porous old ITZ and normal mixing; (b) dense old ITZ after TSMA [43]. Reproduced with permission from [43], Elsevier, 2020.

Similarly, the authors identified the method that enhanced the compressive strength of the samples. It is known that the pores and cracks inside the ITZ of RA significantly affect the ultimate strength of RCA. The high water content in the new ITZ of RCA is caused by high WA of these minute pores and cracks. Large crystals of CH accumulate in the pores of RA and on the surface of the old concrete, which reduces the compressive strength of the RCA compared to NAC, as shown in Figure 17. By implementing DM on RCA, the strength was found to increase in comparison to NM. The compressive strength of the samples rose due to the application of TM at various curing ages.



(b) Double mixing method (DM), coating with cement and admixture



(c) Triple mixing method (TM), coating with admixture

**Figure 14.** Mixing methods: (a) NM; (b) DM; (c) TM [44]. Reproduced with permission from [44], Elsevier, 2010.



**Figure 15.** Surface microstructure of the concrete coated with fly ash at the curing age of 1 and 3 days [44]. Reproduced with permission from [44], Elsevier, 2010.



**Figure 16.** Surface microstructure of the concrete with slag at the curing age of: (**a**) 1 d; (**b**) 28 d [44]. Reproduced with permission from [44], Elsevier, 2010.

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**Figure 17.** Compressive strength of RCA for different mixing methods [44]. Reproduced with permission from [44], Elsevier, 2010.

Overall, two improvement methods (DM and TM), enhanced ITZ by filling pores and microcracks. Based on different experiments, it can be said that mixing approaches help to improve the mechanical properties and also enable RCA to be applied on a wider scale. However, it would be better to use TM because it demonstrated higher effectiveness in improving the RCA performance compared to DM. Table 5 summarizes the details of improving old and new ITZ through carbonation, incorporating mineral admixtures, and using different mixing approaches.

Table 5. Improvement methods for ITZ of RCA.

Improvement Method	Conditions/Materials	Main Findings	Reference
	Relative humidity at 70 $\pm$ 5%, temperature at 23 $\pm$ 2 °C, 20% CO <sub>2</sub> concentration	<ul> <li>The microhardness of ITZ in the treated samples (CRCA) was 31% higher in comparison to the original RCA.</li> <li>SEM images showed densified ITZ with hydration products.</li> </ul>	[6]
	Relative humidity at 60 $\pm$ 5%, temperature at 20 $\pm$ 2 °C, 20 $\pm$ 2% CO_2 concentration	<ul> <li>Percentage of mechanical crushing value on average declined by 20%.</li> <li>Proportion of water absorption lowered by 23–28%.</li> <li>SEM images demonstrated that the carbonated sample had a denser ITZ and fewer large pores.</li> </ul>	[35]
	Relative humidity at 70 $\pm$ 5%, temperature at 20 $\pm$ 2 °C, 20 $\pm$ 3% CO2 concentration	• SEM images revealed filled voids and densified ITZ of treated RCA with the formation of both fiber-like and cotton-like materials.	[36]
Carbonation treatment	CO <sub>2</sub> concentration of approximately 100% at a pressure level of 0.1 bar and 5 bar	<ul> <li>Water absorption percentage decreased by 1 and 1.1 at 0.1 and 5 bar, respectively.</li> <li>Crushing value of the RA has changed from 27.8% to 21.9% at 0.1 bar pressure and to 20.6% at 5 bar.</li> <li>Microhardness of carbonized NRCA was equal to 97 ± 9, which was higher than the noncarbonated NRCA (87 ± 17).</li> <li>Microhardness of new ITZ in the carbonated NRCAs was somewhat higher than that of the uncarbonated sample.</li> </ul>	[37]
	Internal pressure of 1 bar with no temperature regulation, but relative humidity maintained at $54 \pm 5\%$	• Porosity of ITZ in treated RCA at the innermost 10 μm was slightly lower compared to the untreated sample.	[38]
	Rapid carbonation was based on the GB/T 50082-2009	<ul> <li>Before carbonation, the microhardness of ITZ was around 125–171 MPa, and width was equal to 70–80 μm.</li> <li>After carbonation, microhardness of ITZ increased to 144–182 MPa, and the width decreased by 20 μm.</li> </ul>	[39]

Improvement Method	Conditions/Materials	Main Findings	Reference
Incorporation of mineral admixtures	Raw silica fume was used with a superplasticizer to treat RCA samples	• SEM images demonstrated that a pozzolanic reaction, which formed inside the aggregate, strengthened the new ITZ.	[33]
	RCA was dried for 48 h at 60 °C and then combined with slurry for 0.5 h and soaked for 1 h	<ul> <li>The increased magnitudes of microhardness in the old ITZ were 67.7%, 55.6%, and 32.2% by silica fume, nano-SiO<sub>2</sub>, and fly ash, respectively.</li> <li>Strengthening effect was more evident in the new ITZ and higher increase in the percentage of microhardness was identified.</li> </ul>	[36]
	Performance of phosphorus slag (PHS), ground granulated blast-furnace slag (GGBS), and fly ash (FA) in various combinations, i.e., 20% PHS (P2), 10% PHS+10% FA (P1F1), and 10% PHS+10% GGBS (P1S1) was tested	<ul> <li>Mixture of PHS and GGBS was found to be most effective.</li> <li>Sample with the addition of GGBS showed the greatest effectiveness in enhancing the microstructure of the old ITZ (P1S1) compared to P2 and P1F1.</li> </ul>	[41]
	Two-stage mixing approach	<ul> <li>SEM images revealed clear densification of the old ITZ after implementation of TSMA.</li> <li>SEM images of TSMA-treatment concrete demonstrated an improvement in the new ITZ, which was stronger and denser compared to the NMA.</li> </ul>	[42]
Mixing approaches	Two-stage mixing approach	• Backscattered secondary electron (BSE) image demonstrated denser texture of the old ITZ.	[43]
	Double-mixing method (DM) and triple-mixing method (TM)	<ul> <li>SEM images revealed formation of new hydration products, thus improving properties of the ITZ.</li> <li>Compressive strength of the samples increased due to the application of TM at various curing ages.</li> </ul>	[44]

# Table 5. Cont.

## 4. Improvement of Adhered Mortar of RCA

To improve the performance of RCA so that they are comparable with NA, the performance of adhered mortar of RCA should be improved. This section focuses on the improvement of adhered mortar by surface solidification through carbonation, slurry coating, biodeposition, sodium silicate solution, and polymer treatment. The following improvement methods focus on increasing density, decreasing porosity, and solving the adherent mortar's high water absorption capacity.

#### 4.1. Surface Solidification

The densification of the surface of RCA can be achieved in different ways: carbonation, cement slurry or mineral admixtures slurry, biodeposition, and sodium silicate solution. These methods improve the porous surface of the adhered mortar, thereby making the surface of RCA stronger and impermeable. In this section, these treatment methods along with their enhancement mechanism to improve properties of RCA are described in detail.

## 4.1.1. Carbonation Treatment

The adhered mortar contains a significant amount of pores, which in turn, significantly increases the porosity of the RCA. During the carbonation process, the  $CO_2$  can enter into the pores of adhered mortar and react with the main hydration products of the adhered mortar, i.e., Ca(OH)<sub>2</sub> and C–S–H, as demonstrated in Equation (1) below, thus producing CaCO<sub>3</sub> and silica gel (SiO<sub>2</sub>) [7].

$$Ca(OH)_2 + CO_2 \Rightarrow CaCO_3 + H_2O$$
  

$$C - S - H + CO_2 \Rightarrow CaCO_3 + SiO_2 + nH_2O$$
(1)

These reaction products fill the pores and lower sponginess in adhered mortar. Carbonation treatment can be conducted in various ways; however, the final result is the densification of the adherent mortar in the RCA [3].

Li et al. [45] evaluated the influence of carbonation treatment on RCA and the mechanical properties of the concrete. For this purpose, a special idealized model of real RCA was used, which is termed as model recycled aggregate concrete (MRAC). The model consists of nine specimens of RCAs (named as MRCA) molded in circular disc surrounded by cement mortar matrix with specific distribution. Two different types of old cement mortar matrix having compressive strength equal to 55.6 MPa (M1) and 37.5 MPa (M2) were used. The water-to-cement ratio in M2 was higher than in M1, thus the porosity of M1 was lower in comparison to M2. The concrete samples prepared with M1 and M2 cement mortar were

A-M1 and MRCA-M2. The MRCA samples is shown in Figure 18.



Figure 18. Modeled aggregate MRCA [45]. Reproduced with permission from [45], Elsevier, 2018.

The CO<sub>2</sub> curing of the MRCAs was performed by placing them inside the curing chamber with a constant pressure maintained at 0.1 bar. Anhydrous silica gel was placed in the chamber to mitigate the negative influence of high humidity by removing evaporated water. Thereafter, MRCAs samples were soaked in water and air-dried. Figure 19 shows the geometric dimensions of MRAC specimens. The carbonated concrete samples prepared with M1 and M2 cement mortar were designated as MRCA-CM1 and MRCA-CM2, respectively.



**Figure 19.** Geometric dimensions of MRAC: (**a**) front face; (**b**) side face [45]. Reproduced with permission from [45], Elsevier, 2018.

The enhancement in the properties of RCA due to carbonation treatment was determined using a microhardness test. Considering the common width of the ITZ equal to 50 um, the distance tested in the M1 sample was set equal to 100 um. As shown in Figure 20, the microhardness of the cement paste increased in the carbonated sample. The average microhardness of carbonated MRCA-CM1 and MRCA-CM2 samples was around 34% and 12% higher compared to MRCA-M1 and MRCA-M2, respectively. The microstructure of the cement paste with a higher water-to-cement ratio improved noticeably, thus the increase in microhardness of MRCA-M2 was higher than in MRCA-M1.



Figure 20. Microhardness test [45]. Reproduced with permission from [45], Elsevier, 2018.

The compressive strength of the MRAC specimens was also compared. Test results demonstrated that the compressive strength of carbonated MRAC-CM1 and MRAC-CM2 specimens were 6.07% and 7.69% higher compared to uncarbonated specimens MRAC-M1 and MRAC-M2, respectively. It can be explained by a reduction in porosity, which caused the enhancement in the strength of the old ITZ and old mortar. Overall, the increase in compressive strength was found to be significant for mortar having a higher w/c ratio. Additionally, the obtained values suggested an enhancement in microhardness of the old ITZ and the old mortar in the MRCA specimen. However, the improvement of the old ITZ was more considerable compared to the old mortar.

Li et al. [6] evaluated the influence of carbonation treatment on the adherent mortar. The experimental details of carbonation treatment were described previously in the ITZ improvement. It was demonstrated that pores of the adhered mortar were occupied by products of the carbonation reaction. The relationship between attached mortar content and WA with different particle sizes is presented in Figure 21, which shows that a linear relation exists between the attached mortar content and WA in both RCA and carbonated RCA with particle sizes 20–30, 30–40, and >40 mm. It can be observed that the trend line of RCA is higher than CRCA, suggesting that CRCA is much denser and less absorbent compared to RCA. Carbonation treatment of RCA samples resulted in lower water absorption owing to the reason that medium-size pores were occupied by calcium carbonates.

Overall, studies conducted by Xuan et al. [37] and Li et al. [6] revealed that reaction products after carbonation treatment accumulate in the pores of adhered mortar, hence increasing the density and enhancing the physical properties of RCA by reducing porosity and water absorption. In addition, no hazardous byproducts are produced during carbonation, making it the most efficient and environmentally friendly treatment method for improving RCA.



**Figure 21.** Relation between attached mortar content and WA with particle sizes (**a**) 20–30 mm; (**b**) 30–40 mm; (**c**) >40 mm [6]. Reproduced with permission from [6], Elsevier, 2019.

## 4.1.2. Mineral Admixture

Another way to improve the performance of adherent mortar is to incorporate pozzolanic materials such as fly ash, silica fume, nanosilica, rice husk ash (RHA), and ground and granulated blast furnace slag (GGBS). The use of pozzolanic material is suggested as the most effective method for improving the properties of adhered mortar. The phenomenon can be explained by the reaction between the pozzolanic materials and CH remaining in the adhered mortar, resulting in the formation of a C–S–H gel, which sufficiently improves the bonding characteristics of RCA and yields improved mechanical properties.

Kong et al. [44] evaluated the role of mineral admixture incorporation during the double- and triple-mixing approach. The details of the mixing approach can be found in the previous section describing the improvement of ITZ. As a result of double mixing, a thin layer of low w/c ratio slurry was formed on the surface of RCA. The slurry encased the recycled aggregate specimen and accumulated inside the pores, which in turn densified the surface and resolved the friability issue, as shown in Figure 22. It was suggested that there was an improvement in the adherent mortar because coated pozzalanic particles consumed CH, which was inside of the pores and on top of the surface of the attached mortar.





**Figure 22.** SEM image (i) in pore and (ii) on the surface [44]. Reproduced with permission from [44], Elsevier, 2010.

Through the triple-mixing approach, the loose surface of RCA was densified by incorporating the mineral admixtures: finely ground slag and fly ash. Both admixtures showed evident results in filling the voids and forming new hydration products by consumption of CH. Figure 23 shows active densification of the surface, which strengthened the recycled aggregates.



**Figure 23.** Surface microstructure of the old concrete after fly ash implementation: (i) 1 day; (ii) 3 days [44]. Reproduced with permission from [44], Elsevier, 2010.

Lastly, it was noted that the incorporation of finely ground slag demonstrated a better improvement over fly ash due to better enhancement in the microstructure. Therefore, the triple-mixing approach with the application of mineral admixtures, primarily FGS, was found to be more effective.

Another material having the ability to fill the pores in the mortar and can be used as pozzolana is nanosilica. Hosseini et al. [8] evaluated the impact of nanosilica particle addition to improve the mechanical properties of RCA. Coarse RAs with SG of 2.42, WA of 4.8%, and size of 20 mm were used. To fabricate the concrete made of RCA, materials such as cement, sand, and coarse aggregate were mixed in a concrete rotary mixer, and then nanoparticles, water, and water-reducing agent were added for better workability. For the compressive strength testing, cubic specimens with dimensions  $10 \times 10 \times 10$  cm<sup>3</sup> were used. During the experiment, the compressive strength of the specimen improved compared to the specimen without incorporated nanosilica. The filler effect and higher pozzolanic reactivity imparted by nanosilica affected the improvement in strength. It was found that the strength development was mainly due to the reaction between CH and nanosilica, which resulted in the development of additional C-S-H gel. Overall, higher compressive strength of RCA was observed after its treatment with nanosilica due to its high specific surface area, which enables to the creation of denser microstructure in the mortar [12]. Finally, the authors emphasized that as the overall performance of the specimens improved, the properties of the adherent mortar were also enhanced. However, one of the drawbacks of using nanosilica is that it requires a large amount of water, which may result in the need to add an admixture for water reduction.

In summary, different studies were performed incorporating pozzolanic material that showed improvement in the properties of RCA. However, most experiments such as [2,46,47] compared the influence of the treatment on the overall performance of the RCA without analyzing the effect on the adherent mortar in particular.

#### 4.1.3. Calcium Carbonate Biodeposition

Calcium carbonate biodeposition is one of the methods to improve the properties of RCA. This method works by decreasing the WA ability in the adherent mortar. Bacteria, which has the ability to accelerate the formation of calcium carbonate on the surface of the aggregate, plays a crucial role in calcium carbonate biodeposition due to the presence of adequate negative zeta potential on the surface of the cell wall [48]. The biomineralized precipitation has a good cementing and filling effect on microcracks and adhered mortar (Figure 24), thus having the potential to intensify the use of RCA [9].



## After

**Figure 24.** RA improved by biodeposition of calcium carbonate [9]. Reproduced with permission from [9], MDPI, 2021.

Compared to other methods, calcium carbonate biodeposition is a natural method that has a lower environmental impact due to naturally occurring substrates for seeding [49]. Bacteria called Sporosarcina pasteurii was adopted for the biodeposition technique by Grabiec et al. [48]. Inoculation was performed with a liquid culture medium obtained from urea. The reaction is shown in Equation (2):

$$Sp. cell + Ca^{2+} \Rightarrow Sp. cell - Ca^{2+}$$

$$CO(NH) + 2I spocel 2N fla^{2+} \oplus OSp. cell - Ca^{2+}$$

$$CO(NH_2)_2 + 2H_2O \Rightarrow 2NH_4^+ + CO_3^{2-}$$

$$Sp. cell - Ca^{2+}_2 + CO_3^{2-} = Sp. cell - CaCO_3$$

$$(2)$$

In this reaction, Sp.cell (bacteria cell-wall surface) attracts  $Ca^{2+}$  (calcium ion), which further reacts with  $CaCO_3$  (calcium carbonate ions) that originates from urea hydrolysis. Then, by increasing the pH of the medium through NH<sub>4</sub><sup>+</sup> (ammonia ions), the precipitation efficiency of the calcium carbonate improved. In order to precipitate calcium carbonate, firstly, the growing substance containing bacteria was kept at 25 or 35 °C for 72 h [12]. After 3 days of precipitation, RCA was submerged in this substance for an additional 72 h. From the results, it was detected that a reduction in WA by up to 21% was obtained [12]. In addition, improved bonding in the surface of the aggregate was achieved due to filling of voids in adherent mortar by precipitated  $CaCO_3$ .

Feng et al. [50] compared the performance of the modified recycled fine aggregate (RFA) in mortar to nonmodified RFA and to mortar specimens made of fine natural aggregate (NFA). RFA was obtained from Tongxiang Tongde Wall Building Materials Co., Ltd., China, and had an apparent density of 2530 kg/m<sup>3</sup> and a WA of 24.5%. Before modification, RFA was oven-dried at 105 °C and then placed for 24 h in a proliferation culture media that contained Sporosarcina pasteurii previously activated by ultrapure water. Afterward, RFA

was placed into precipitation media consisting of Tryptone (3 g/L), NH<sub>4</sub>Cl (10 g/L), Urea (20 g/L), Na<sub>2</sub>CO<sub>3</sub> (2.12 g/L), and an indefinite content of CaCl<sub>2</sub>.

Figure 25 demonstrates RFA before and after treatment, showing modified RFA covered with white calcium carbonate. Most of the precipitation is located at the upper surface of M-mRFA, which can be explained by dependence on the distribution of bacteria. defects.



**Figure 25.** (a) Untreated RFA and (b) modified RFA [50]. Reproduced with permission from [50], Elsevier, 2020.

According to Figure 26, the water absorption of the M-mRFA decreased in comparison to RFA without modification. The reduction after the treatment was 22.0%, 27.3%, 28.6%, and 31.4% at 3, 7, 10, and 14 days, respectively. Similarly, the compressive and flexural strength of the modified recycled mortar was enhanced by 14.3% and 84.5%, respectively, compared with mortars cast with nonmodified RFA. In other words, the calcium carbonate treatment contributed to improving the properties of the specimen related to flexural and compressive performance, thus demonstrating the potential application of microbial-modified treatment for casting mortar and concrete.



**Figure 26.** Effect of treatment on water absorption [50]. Reproduced with permission from [50], Elsevier, 2020.

The implementation of biodeposition of calcium carbonate requiring concrete modifications and microbiology approaches is a promising method to improve the quality of RCA because it results in a considerable reduction in water absorption. Along with this, calcium carbonate biodeposition uses bacteria that occur in nature, which makes the treatment without any toxic waste and is therefore environmentally friendly. However, this treatment is still little known, and its practical application requires additional research.

## 4.1.4. Sodium Silicate Solution

Sodium silicate solution is another approach used to strengthen adherent mortar by forming C–S–H gel [51]. The reaction occurs between sodium silicate solution and calcium hydroxide, and is described by Equation (3):

$$Na_2SiO_3 + Ca(OH)_2 + H_2O \Rightarrow C - S - H + NaOH$$
(3)

Ali [10] studied the effect of sodium silicate addition to a recycled concrete aggregate containing mortar (RAM). For this purpose, sodium silicate was used in the amounts of 0%, 1%, 2%, 3%, 4%, and 5% by weight of cement in RAM and the performance of the RAM was compared with the performance of the natural aggregate mortar (NAM). The higher compressive strength and improved durability properties of RAM at 2% NS and 3% NS compared to NAM were reported. The study results show that with 2% and 3% sodium silicate, the compressive strength of the RAM sample increased by 25–28% compared to NAM, refer to Figure 3 in Reference [10]. It can be explained by the presence of CH from the old concrete in the RCA and its further reaction with sodium silicate, which resulted in the formation of C–S–H gel, thus improving the compressive strength of the RAM specimen. At 3% sodium silicate, the highest value of the compressive strength was observed, which was about 14% larger compared to NAM. Further increase in the sodium silicate concentration (beyond 3%) contributed to the gradual reduction in compressive strength. Hence, 3% sodium silicate by weight of cement in RAM was suggested as an optimal value for improving the compressive strength.

It should be mentioned that higher concentrations of sodium silicate could influence the alkalinity level of mortar. The reaction between sodium silicate and CH releases NaOH, which reduces compressive strength, as reported by Shaban et al. [12]. Overall, the implementation of sodium silicate solution reduces the WA of RCA by filling pores, voids, and cracks inside the microstructure by forming C–S–H gel. However, treatment by sodium silicate solution can increase the possibility of alkali–silica reaction by introducing alkalis. Similarly, this treatment method does not work at its full potential at high temperatures, so it is suggested first to investigate which treatment method is most effective depending on the final goal.

#### 4.2. Water-Repellent Coat by Polymer Treatment

The alternative way to improve the characteristic of RCA is to apply a water-repellent coat by polymer treatment on the surface of RCA. The reason is to improve the loose adhered mortar. The water-repellent coat works not only as a filler but also as a repellent layer to decrease the water absorption of RCA. The water-repellent coat forms when the polymer penetrates the RCA and alters the surface from hydrophilic to hydrophobic [52].

The polymer dosage is an important factor that directly affects the final result of treatment. Kou and Poon [11] evaluated the performance of polyvinyl alcohol (PVA) treatment and its influence on the compressive and tensile strength, water absorption, shrinkage, and chloride penetrability. For the experiment, crushed granite was used as the NA, while crushed old concrete rubble derived from reinforced concrete buildings in Hong Kong were used as recycled aggregates. For PVA treatment, RCAs were firstly placed in desiccators with a vacuum pump at 920 mbar pressure, while PVA solution was prepared by separately adding PVA powder into boiled water. After that, 6, 8, 10, and 12% PVA solutions were added into the desiccators to soak aggregates in the polymer solutions for 24 h. For the concrete mix, the w/c ratio was taken as 0.5, and samples of 100 mm cubes,  $75 \times 75 \times 285$  mm<sup>3</sup> prisms, and  $200 \times 100$  mm<sup>2</sup> diameter cylinders were cast. Final measurements showed that 10% of PVA addition was the optimal dose because the change in water absorption magnitude was significant with the addition of 6% to 10% of PVA, whereas 12% PVA had minor changes compared to the initial water absorption value. The increase in density was around 2% of the initial density of RCA. The most significant effect was on the water absorption value relative to density because the value dropped from 6.23% to 2.39% and 1.62% at oven-dried and air-dried options, respectively. Summarizing,

the filling ability of PVA improved not only the overall performance of the RCA, but also the adherent mortar as well.

The polymer treatment (PT) can be carried out by one polymer or by combination of various polymers. Spaeth and Tegguer [53] examined RCA treatment by soluble sodium silicate (SS) and five silicon-based (SB) polymers to determine type and concentration of PT that has more effect on the overall RCA improvement. These combinations were P1-Sodium silicate, P2- Octyl/methyl methoxy co-oligomeric siloxane/silane, P3- Octyl triethoxy silane, P4- Siloxane/propyl trimethoxy silane, P5- Siloxane/propyl triethoxy silane, and P6- Siloxane/alkylalkoxysilane. For this purpose, two different approaches were adopted: soaking in polymer solution from P1 to P6 for 5 min and then drying (simple combination) or soaking in P1 for 3 min following by drying and then again impregnating in polymer solution P2 to P6 for 5 min followed by drying (double combination).

The main reason for using double soaking in two different polymer solutions was to increase the hydrophobic properties of treated RCA. The results showed that double impregnations were not always efficient as in most cases; it did not demonstrate additional effect in improvement of properties of treated RCA. However, the combination 30% of P1 and 30% of P2 had the lowest value of water absorption coefficient (0.7%). The reason for the effectiveness of 30% P1+ 30% P2 is the formation of co-polymeric film after the reaction of silicate and silanol films inside the pores of RCA. Therefore, it was concluded that among the six types of polymers, the most influencing the adherent mortar was 30% sodium silicate and 30% octyl/methyl methoxy co-oligomeric siloxane/silane.

Overall, the polymer treatment enhanced the properties of RCA. However, most studies did not investigate in detail its impact on the adherent mortar and related improvement to the overall enhancement of RCA. Therefore, investigation on the adherent mortar part improvement requires additional research. Table 6 summarizes the various improvement methods described in this section and their key findings.

The main aim of different methods of improving RCA is to enhance its overall properties. However, most of the experimental investigations did not explicitly mention which part, i.e., ITZs or adherent mortar, was improved. Hence, this part presents conclusions drawn from various studies in which the properties and performance of RCA were improved without specifying the improvement region. The improvement results obtained by adopting different approaches such as biodeposition, incorporating mineral admixtures, carbonation treatment, polymer treatment, different mixing approaches, and coating with pozzolanic powder are listed in Table 7. The biodeposition approach was used by Nele et al. It was concluded that this approach reduced WA and improved the compressive strength of concrete made from treated RCA [54]. Similarly, studies on the mineral admixture incorporation demonstrated an increase in the values of compressive strength and reduction in WA when compared to the control sample [46,55-59]. Some experimental studies compared the addition of different silica fume content during the mineral admixture treatment procedure. Most studies suggested that incorporating 10% of SF significantly improved compressive and tensile strength in RCA samples. Some researchers evaluated the influence of carbonation treatment in improving RCA [35,60–63]. These researchers compared compressive strength, water absorption, and SEM images of carbonation-treated samples with the reference samples. They found that carbonation-treated samples performed better than the reference samples, hence, testifying to the effectiveness of the carbonation treatment. Polymer treatment was used by one of the research teams to show that it effectively decreased WA of the treated specimen [64]. Another method used for the improvement in RCA properties was the implementation of different mixing approaches. Liang et al. [65] investigated the implementation of the mortar-mixing approach (MMA) and the sand-enveloped-mixing approach (SEMA) as alternative ways to improve the properties of RCA. Overall, the improvement in the compressive strength values was identified by using both mixing approaches and their potential application in future studies for RCA enhancement was suggested. Tam and Tam [30] evaluated the application of the two-stage mixing approach (TSMA). The reduction in water absorption and chloride penetration was

identified, thus proposing this mixing approach for future application. Another method that has the potential in practical application is coating with pozzolanic powder suggested by Li et al. [47]. This treatment method has been observed to increase the values of compressive strength and reduces the chloride permeability in treated samples. Conclusively, it can be deduced that carbonation treatment is the most popular approach based on its efficiency in improving the mechanical properties together with the durability performance.

Table 6. Improvement methods for adherent mortar of RCA.

Imp	rovement Method	Conditions/Materials	Main Findings	Reference
	Carbonation	Constant pressure of 0.1 bar and addition of anhydrous silica gel	<ul> <li>Carbonated sample had increased values in the microhardness of the cement paste with 34% and 12% increase from original values.</li> <li>Enhancement in microhardness of the old ITZ and the old mortar was observed, but the improvement of the old ITZ was more considerable.</li> <li>Compressive strength of carbonated specimens was 6.07% and 7.69% higher in comparison to uncarbonated.</li> </ul>	[45]
		Relative humidity at 70 $\pm$ 5%, temperature at 23 $\pm$ 2 °C, and 20% CO <sub>2</sub> concentration	<ul> <li>Pores of the adhered mortar were occupied by products of the carbonation reaction.</li> <li>Linear relation can be noticed between attached mortar content and water absorption.</li> <li>CRCA is much denser, and less absorbent compared to RCA.</li> </ul>	[6]
surface solidification	Pozzolana Addition	Mineral admixture incorporation during double-mixing method (DM) and triple-mixing method (TM)	<ul> <li>Slurry enveloped the recycled aggregate sample and accumulated inside the pores.</li> <li>Images showed active densification of the surface that resulted in strengthening the recycled aggregates.</li> <li>Incorporation of finely ground slag (FGS) demonstrated a better improvement over fly ash with better enhancement in the microstructure.</li> </ul>	[44]
01		Addition of nanosilica particles	<ul> <li>Compressive strength of the specimen improved compared to the sample without incorporated nanosilica.</li> </ul>	[8]
	Calcium Carbonate Biodeposition	Sporosarcina pasteurii previously activated by ultrapure water	<ul> <li>Water absorption reduction after the treatment was up to 31.4% after 14 days.</li> <li>Compressive and flexural strength of the recycled mortar were enhanced by 14.3% and 84.5%, respectively compared with mortars cast by RFA.</li> </ul>	[50]
	Sodium Silicate Solution	Sodium silicate was used in the amount of 0%, 1%, 2%, 3%, 4%, and 5% by weight of cement	<ul> <li>At 3% of sodium silicate, the highest value for the compressive strength was observed, which was about 14% larger compared to NAM.</li> <li>3% by weight of cement in RAM of sodium silicate could be considered as optimum value for the compressive strength improvement.</li> </ul>	[10]
Wate	r-Repellent Coat by	Polyvinyl alcohol (PVA) treatment	<ul> <li>10% of PVA was the optimal dose.</li> <li>Increase in density was around 2% of the initial density of RCA.</li> </ul>	[11]
Ро	lymer Treatment	Soluble sodium silicate (SS) and five silicon based (SB) polymers	<ul> <li>Double impregnations were not always efficient.</li> <li>Combination 30% of P1 and 30% of P2 had the lowest value of water absorption coefficient, which was 0.7%.</li> </ul>	[53]

Improvement Method	Main Findings	Reference
Biodeposition	<ul> <li>Has a positive impact in enhancing the properties of concrete.</li> <li>Water absorption of RCA concrete was decreased by 1%.</li> <li>Compressive strength of RCA concrete was increased by 25%.</li> </ul>	[54]
	<ul> <li>Addition of 10% SF content in the mixture has a positive effect on properties of RCA, especially on the compressive strength at the age of 28 days.</li> <li>Water absorption (WA) of RAC specimens decreased when more than 5% SF content was added.</li> </ul>	[55]
	<ul> <li>Effect of mineral admixtures, especially SF, significantly improved the durability properties such as sorptivity and chloride permeability.</li> <li>Use of FA and microsilica had almost the same compressive strength at 28 and 56 days.</li> </ul>	[56]
Mineral Admixture	<ul> <li>Incorporation of 10% SF into RCA increased compressive and tensile strengths in comparison to control concrete mix without SF.</li> <li>Addition of SF decreased the porosity of RCA.</li> </ul>	[57]
	<ul> <li>Compressive strength of RAC concrete mixture with SF concentration of 5% and 10% increased by 19% and 21%, compared to the control mixture without SF.</li> <li>WA of concrete containing RAC with SF decreased, especially at later ages.</li> </ul>	[58]
	<ul> <li>Reference mixture (REF) had a value of compressive strength equal to 33.9 MPa, the mixture with RAC without fly ash (REC) had 31.1 MPa compressive strength, and the compressive strength of RCA sample treated with FA addition was 34.1 MPa.</li> <li>Highest value of compressive strength was observed from a sample with FA addition.</li> </ul>	[46]
	<ul> <li>Addition of SF or metakaolin (MK) at a content of 10% the weight of portland cement (PC), decreased the strength reduction to 8% and 3%, respectively.</li> <li>MK was relatively more effective in controlling water penetration and decreasing strength reduction compared to SF.</li> </ul>	[59]
	• Accelerated RCA carbonation decreased WA of RCA by filling pores by forming calcium carbonate.	[60]
	<ul> <li>Has a positive impact on the properties of RCA.</li> <li>Compressive strength of CRAC increased by 21.05% in comparison to the untreated RCA sample.</li> <li>Depth of water penetration of RCA reduced by 31.5 mm as compared to untreated RCA.</li> </ul>	[61]
Carbonation	• After treatment, a considerable reduction in WA and porosity of the RCA was noted.	[62]
Carbonation	<ul> <li>Carbonation treatment significantly improved the physical properties of RCA.</li> <li>Apparent density increased by 4.7–5.6%.</li> <li>WA decreased by 7.6–9.6%.</li> <li>Crushing value decreased by 22.6–28.3%.</li> </ul>	[35]
	<ul> <li>Improvement of RCA using accelerated carbonation proved to be effective.</li> <li>WA of RCA decreased by 25.8%.</li> <li>Density of treated RCA was much higher in comparison to the untreated RCA sample.</li> </ul>	[63]
Polymer treatment	<ul> <li>Polymer treatment with water-repellent-based emulsions with siloxane polymers had a positive impact on the properties of RCA.</li> <li>Treated RCA absorbed less than 20% of water compared to the initial WA value of the control mix using untreated RCA.</li> </ul>	[64]

 Table 7. Methods for improving properties of recycled aggregate concrete.

Improvement Method	Main Findings	Reference
	<ul> <li>Implementation of the mortar-mixing approach (MMA) improved properties of RCA.</li> <li>MMA improved compressive strength of RAC without any surface pretreatment making the compressive strength equal to 31.7 MPa.</li> </ul>	[65]
Approaches	• The sand-enveloped-mixing approach (SEMA) is an alternative way to enhance the compressive strength of RCA.	
	<ul> <li>Two-stage mixing approach (TSMA) enhanced the performance of untreated RCA.</li> <li>Water permeability value improved by 35.41% compared to untreated sample.</li> <li>Chloride permeability value improved by 29.98% compared to untreated sample.</li> </ul>	[30]
Coating with pozzolanic powder	<ul> <li>Implementation of new mixing technique—coating using a pozzolanic powder such as FA, SF, blast furnace slag (BS), or their combination—improved the compressive and flexural strength of RCA.</li> <li>Highest value of compressive strength was equal to 47.6 MPa after 28 days, while the flexural strength value was around 7.31 MPa after 28 days.</li> <li>Chloride permeability value improved by 29.98% compared to an untreated sample.</li> </ul>	[47]

## 5. Application of RCA in Concrete

Various pilot projects have been carried out in countries such as the USA, France, Germany, and Portugal to use RCA for concrete [66,67]. Nevertheless, the application of RCA in concrete and its practical implementation is not fully developed owing to the weak mechanical characteristics of RA in comparison to NA.

One research experiment was conducted to identify certain criteria for RCA to be implemented for reinforced concrete [68]. For the experiment, the authors used four specimens that included one control mix and three mixes with different amounts of adherent mortar, water absorption, and relative density of the aggregates. The study's results suggested that the maximum adherent mortar content should be 50%; the percentage of water absorption should be equal to 3%, while the relative aggregate density can be 2.3 or higher. In addition to the mentioned properties, the presence of chloride should be addressed. There is always a risk of contamination caused by RCA [66]. This can be explained by the origin of the RCA, which is mainly debris of concrete elements. It has been established that the content of RCA in concrete can affect the resistance to chloride permeation resistance compared to NA [69]. The reduction in porosity can help improve chloride resistance. Similarly, the alkali–silica reaction can be introduced into new concrete with RCA when implementing RCA from concrete debris exposed to the alkali–silica reaction [66].

Overall, the number of studies related to the practical implementation of RCA in concrete is very limited, so it is recommended to study this aspect in more detail, and identify standardized tests in order to define specific regulations for using RCA in practical application.

#### 6. Discussion

In this paper, numerous treatment methods to improve the properties of ITZ and adhered mortar have been described and explained in detail. The comparison of plans was made according to the region of improvement of the RCA.

Several methods, such as carbonation, mineral admixture addition, and variation in mixing approaches, were discussed to improve the properties of ITZ in RCA. The first treatment method that was investigated was a carbonation treatment. The majority of research studies used a Vickers microhardness test and SEM to evaluate the effectiveness of a treatment on the ITZ enhancement. It was found that treatment produces a large amount of CaCO<sub>3</sub>, which results in the improvement in the microstructure of the ITZ, thus suggesting the effectiveness of this method for practical application. Another way to

improve the properties of the ITZ and its performance is to add mineral admixtures. Results from the analysis demonstrated that the incorporation of single and multiple mineral admixtures can be used to treat and enhance the performance of ITZ. Among conventional mineral admixtures, silica fume showed the best performance in strengthening and making ITZ denser. However, implementing mineral admixtures has a lower effect in improving the microhardness values when compared with the carbonation treatment approach. Different mixing approaches were also evaluated to enhance the properties of the ITZ, even though their improvement in properties is limited. The study findings demonstrate that two mixing methods such as DM and TM were found to be effective in filling pores and microcracks by forming new hydration products and reducing the amount of accumulated CH crystals, thus further improving strength and durability of RCA concrete. It should be noted that the TM approach demonstrated a considerable increase in compressive strength of the RCA in comparison to DM. Overall, it can be said that all the methods as mentioned earlier could be used to improve ITZ's performance, but, depending on the final goal, the treatment method should be selected accordingly.

Based on previous studies on adherent mortar improvement, implementation of pozzolana addition such as FA, SF, and NSi can modify the surface of RCA, enhance the properties of adherent mortar by filling pores and voids, and produce C-S-H gel. Hence, it can be considered as an effective treatment due to its ability to improve the bond between cement paste and RCA. The efficiency of pozzolana addition depends on the content of  $Ca(OH)_2$  in adherent mortar, the particle size of pozzolanic materials, and on the products of the reaction. Thus, compared to other pozzolana materials, the utilization of nanosilica is the most effective way to enhance the properties of adherent mortar because of its high reaction rate and high specific area. However, even if this method is effective in enhancing properties, the cost of using nanosilica is high, thus making it hard to adopt in practical application. Polymer treatment is another method to treat the adherent mortar that can improve workability and durability, serve as filler, and create a water-repellent layer. However, most studies examined the effect of polymer treatment on the overall improvement of RCA without a detailed investigation of its effect on adherent mortar, or only by indirectly mentioning the possibility of the enhancement in adherent mortar region due to overall improvement of RCA properties. Therefore, more research should be carried out to determine its potential in practical application.

Using calcium carbonate biodeposition to treat RCA is also an effective method to enhance the quality of adherent mortar. This method is natural and has the lowest environmental impact due to the implementation of naturally occurring substrates for seeding. The calcium carbonate biodeposition method uses special bacteria that can accelerate the formation of calcium carbonate on the surface of RCA, thus reducing the WA ability in the adherent mortar. However, the impact on the compressive strength is not significant, the treatment is expensive and time-consuming, and its practical application requires more tests and investigations before actual application. Implementation of sodium silicate solution is another treatment method for strengthening adherent mortar because it reduces water absorption by filling pores and voids through C–S–H gel formation. Despite these advantages, sodium silicate solution can increase the possibility of an alkali-silica reaction that leads to a decrease in compressive strength. Compared to the aforementioned methods, carbonation treatment is not only an effective method for enhancing the properties of adherent mortar but is also environmentally friendly. It increases the density and improves the physical properties by reducing porosity and water absorption. Hence, this method is considered to be an effective and sustainable treatment for improving the properties and performance of adherent mortar, but its drawback is that carbonation is time-consuming. Table 8 summarizes the common advantages and disadvantages of each treatment method for ITZ and adherent mortar that were mentioned by different researchers.

	Advantages	Disadvantages
Carbonation treatment	<ul> <li>This method is environmentally friendly;</li> <li>It improves physical properties by reducing porosity and water absorption;</li> <li>It produces a large amount of CaCO<sub>3</sub> and increases the density;</li> <li>It improves microstructure of ITZ and properties of adherent mortar.</li> </ul>	• Treatment is time-consuming.
Mineral Admixture	<ul> <li>It improves the bond between cement paste and RCA;</li> <li>It fills pores and voids and produces C-S-H gel;</li> <li>Silica fume showed the best performance among conventional mineral admixtures.</li> <li>It improves microstructure of ITZ and properties of adherent mortar.</li> </ul>	<ul> <li>Treatment is time-consuming;</li> <li>Uneconomical when nanosilica is used as an admixture.</li> </ul>
Mixing approaches	<ul> <li>This method forms new hydration products;</li> <li>It reduces the amount of accumulated CH crystals;</li> <li>TM approach demonstrated a considerable increase in compressive strength of the RCA;</li> <li>It improves the properties of the ITZ.</li> </ul>	Limited improvement of properties.
Calcium Carbonate Biodeposition	<ul> <li>This method is natural and has low environmental impact due to naturally occurring substrates;</li> <li>It produces a large amount of CaCO<sub>3</sub> and reduces water absorption;</li> <li>It improves properties of adherent mortar.</li> </ul>	<ul> <li>Insignificant impact on the compressive strength;</li> <li>Treatment is expensive and time-consuming;</li> <li>Its practical application requires more tests and investigations.</li> </ul>
Sodium Silicate Solution	<ul> <li>This method reduces water absorption;</li> <li>It fills pores and voids and produces C–S–H gel;</li> <li>It improves properties of adherent mortar.</li> </ul>	<ul> <li>Treatment is expensive;</li> <li>Its implementation can increase the possibility of an alkali–silica reaction;</li> <li>This method does not work to its full potential at high temperatures.</li> </ul>
Water-Repellent Coat by Polymer Treatment	<ul> <li>This method improves workability and durability;</li> <li>It serves as filler and can create a water-repellent layer;</li> <li>It improves properties of adherent mortar.</li> </ul>	<ul> <li>Treatment is expensive and time-consuming;</li> <li>Produces waste that requires disposal;</li> <li>This method is impractical on a large scale.</li> </ul>

Table 8. Advantages and disadvantages of improvement methods for ITZ and adherent mortar.

## 7. Conclusions and Recommendations

The implementation of RCA for concrete production has become popular in the construction industry. However, characteristics of RCA are different compared to NA due to the presence of two components, an adhered mortar and an ITZ between used NA and the original cement. This paper provides an overview of previous research on comparing the physical, mechanical, and chemical properties of RCA together with different treatment methods for improving the ITZ and adherent mortar separately.

(a) It was determined that the properties of RCA are worse than the properties of NA mainly because of the porous microstructure and weak interfacial transition zone. Physical characteristics of RCA such as density, specific gravity, and bulk density are much lower, while water absorption is higher compared to NA due to the porous nature of RCA. In terms of mechanical properties, RCA is also inferior to NA, because the crushing value and impact value of RCA is relatively high while crushing strength is lower due to weak ITZ and adhered mortar. However, these properties can be improved by using different enhancement methods.

- (b) The studies from different researchers indicate that the main methods for enhancing the ITZ properties of RCA are carbonation, incorporation of mineral admixtures, and different mixing approaches. Each of these methods for ITZ improvement has various outcomes depending on the desired final properties. Nevertheless, every treatment method has its own drawbacks in practical application. Compared to these methods, the carbonation treatment method has great potential in the application for improving the properties and performance of ITZ.
- (c) The methods for adherent mortar strengthening include surface solidification through carbonation, slurry coating, calcium carbonate biodeposition, and water-repelling coat using polymer treatment. Carbonation treatment is considered to be effective and environmentally friendly for the improvement of adherent mortar in RCA, densifying the microstructure, and enhancing durability and mechanical properties.

However, in order to practically apply RCA in concrete manufacturing, more research and investigation on the impact of improvement methods is suggested, not only on a small scale under laboratory conditions but in actual projects. Similarly, the practical application of RCA in concrete should be further explored in detail, thereby guiding the rules for its implementation in actual projects. In order to apply RCA implementation to large-scale commercial production, further detailed examination and investigation are required.

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