

Influence of recycled fine aggregate particle size on wet carbonation for producing sustainable aggregate from construction waste

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Abstract

Wet carbonation has emerged as a promising strategy for simultaneously upgrading recycled fine aggregate (RFA), permanently sequestering CO₂, and valorizing construction waste. However, the mechanisms by which particle size regulates ion leaching and carbonation reactions during wet carbonation remain unclear, limiting the effective modification of RFA. This study separately carbonated RFA fractions with different particle sizes to investigate the influence of particle size on wet carbonation and the mechanical properties of mortars prepared with the carbonated RFA. The carbonation behavior of RFA was evaluated through the evolution of solution pH and Ca²⁺/Na⁺ concentrations, while the resulting changes in the physical properties of RFA were subsequently assessed. RFA particles larger than 1.18 mm exhibited slower ion leaching rates and a lower initial solution pH, delaying the formation of a dense surface carbonate layer and thereby facilitating continuous CO₂ diffusion into the aggregate interior, which resulted in more uniform carbonation, pore refinement, and improved aggregate physical properties. RFA particles finer than 1.18 mm underwent rapid ion leaching, which accelerated surface carbonate precipitation, hindered inward CO₂ diffusion, and intensified surface C–S–H decalcification, increasing the total porosity by 18.7% and consequently deteriorating the microstructure and physical properties of the aggregate. After recombining the individually carbonated RFA fractions to match the particle size gradation of natural fine aggregate, mortar prepared with the recombined CRFA exhibited a 25.1% higher 28 d compressive strength than the corresponding mortar prepared with uncarbonated RFA.

Keywords : Recycled fine aggregate; Wet carbonation; Particle size; Ionic concentration; Compressive strength

1. Introduction

Rapid urbanization and large-scale infrastructure renewal have led to a dramatic increase in the generation of construction and demolition (C&D) waste, making its sustainable management an increasingly urgent environmental challenge^[1]. China, the world's largest generator of C&D waste, has seen its annual generation increase from approximately 1.8 billion tonnes in 2017 to 3.5 billion tonnes in 2023, of which waste concrete is estimated to account for approximately 80%^[2,3]. Meanwhile, the growing demand for concrete has substantially

increased the consumption of natural aggregate (NA)^[4], exacerbating the environmental impacts of aggregate extraction, including land degradation, ecosystem destruction, and carbon emissions. Between 2012 and 2022, the consumption of NA increased from 4.59 billion tonnes to 6.63 billion tonnes^[5]. The increasing scarcity of NA^[6], together with the accumulation of waste concrete, has accelerated interest in recycled aggregate production. Recycling waste concrete into recycled aggregate provides an effective strategy to reduce landfill disposal while alleviating the environmental impacts associated with natural aggregate extraction.

Recycled coarse aggregate is more readily utilized because its lower adhered mortar content renders its properties close to those of NA^[7,8], and the corresponding recycling processes are comparatively mature. In contrast, recycled fine aggregate (RFA) typically contains a substantially higher amount of adhered mortar. This residual mortar introduces intrinsic defects, including heterogeneity, high porosity, and numerous micro-cracks, which lead to high water absorption and high crushing value^[9,10]. These drawbacks severely impair the workability and mechanical performance of concrete containing RFA, thereby limiting its widespread application. Accordingly, methods to enhance the performance of RFA have garnered increasing research attention in recent years. Current approaches for improving RFA can generally be classified into three main strategies: (i) removal of adhered old mortar from RFA through mechanical grinding, microwave heating, or acid treatment^[11-13]; (ii) pore-filling using nano-materials to enhance aggregate performance^[14]; and (iii) carbonation treatment, in which CO₂ reacts with calcium-bearing phases in the adhered mortar to form CaCO₃, thereby improving the aggregate microstructure. Mechanical treatments effectively remove adhered mortar but are energy-intensive and may induce additional microcracking. Nano-modification improves surface quality but is often limited by shallow penetration and high material cost. Compared with the aforementioned two methods, appropriately controlled carbonation treatment can simultaneously enhance the intrinsic strength and physical properties of recycled aggregate, avoid secondary damage, and permanently sequester CO₂, making it an effective low-carbon approach for recycled aggregate modification.

Wet carbonation has attracted increasing attention for its rapid reaction kinetics and effectiveness in enhancing the physical performance of RFA, making it an attractive strategy for recycled aggregate modification^[15-17]. During wet carbonation, the carbonation reaction is governed not only by external conditions such as temperature and CO₂ concentration^[18-20], but also by the availability and dissolution of calcium-bearing hydration products in the adhered mortar. These phases, primarily portlandite (Ca(OH)₂), C-S-H gel, and ettringite (AFt), dissolve to release Ca²⁺, which subsequently reacts with dissolved CO₂ to precipitate CaCO₃. Owing to the preferential breakage of the adhered mortar during concrete crushing, finer recycled aggregate fractions generally

contain higher contents of cement paste. For example, the cement paste contents in the 0–4 mm, 4–8 mm, and 8–16 mm fractions were reported to be 28.4%, 18.2%, and 12.6%, respectively^[22]. Together with the larger specific surface area of finer RFA, the increased cement paste content provides more accessible calcium-bearing phases and a larger solid–liquid interfacial area, thereby accelerating ion leaching and resulting in higher concentrations of dissolved Ca^{2+} and Na^+ during wet carbonation.

Although particle size has been recognized as an important factor affecting the wet carbonation of RFA^[23], how particle size regulates ion leaching and the subsequent carbonation reactions remains poorly understood. In particular, particle size may simultaneously influence Ca^{2+} leaching from the aggregate and inward diffusion of carbonate species, but the interaction between these two processes remains unclear. Variations in RFA particle size alter its specific surface area and adhered mortar content, which in turn affect Ca^{2+} leaching from the aggregate into the solution and CO_3^{2-} diffusion into the particle interior. The accelerated leaching of Ca^{2+} is expected to promote rapid surface carbonation, leading to the swift precipitation of carbonation products on the surface layer, a process that may subsequently restrict the inward diffusion of CO_3^{2-} ^[24]. While the CaCO_3 generated by carbonation is generally perceived to densify the microstructure, when the inward diffusion of CO_3^{2-} is hindered, carbonation is primarily confined to the RFA surface; under such conditions, the carbonation of C–S–H gel may simultaneously cause structural degradation of the C–S–H gel and pore coarsening. Therefore, the carbonation behavior of RFA is governed by the dynamic competition between Ca^{2+} dissolution and inward diffusion of CO_3^{2-} . It can thus be inferred that the relationship between RFA carbonation and particle size is not monotonic but exhibits a nonlinear particle size dependence. Existing studies generally evaluate RFA as a bulk material, implicitly assuming that carbonation benefits all particle-size fractions equally, and have not explored the relationship between the ion leaching and carbonation reaction induced by particle size variation^[17,25].

To elucidate how RFA particle size influences wet carbonation and the resulting modification mechanism, this study adopted a fractionated carbonation strategy. According to the Chinese national standard Sand for Construction (GB/T 14684–2022), the aggregate was classified into five particle size fractions: 0.15–0.30 mm, 0.30–0.60 mm, 0.60–1.18 mm, 1.18–2.36 mm, and 2.36–4.75 mm. Each fraction was then subjected to separate wet carbonation treatments. The individually carbonated fractions were subsequently recombined according to the particle size gradation of natural fine aggregate (NFA) and incorporated into recycled mortar as a replacement for NFA. The influence of particle size on carbonation reactions was evaluated by monitoring solution pH evolution, $\text{Ca}^{2+}/\text{Na}^+$ concentrations, physical properties, pore structure, carbonation products, and mortar mechanical performance, with thermogravimetric analysis (TGA), scanning electron microscopy (SEM), and Brunauer–

Emmett–Teller (BET) analysis employed to elucidate the corresponding phase and microstructural evolution.

2. Materials and methods

2.1. Materials and characteristics

Ordinary Portland cement (P·O 42.5) with a density of 3.12 g/cm^3 was used as the binder, and its main chemical composition is presented in Table 1. Xiangjiang River sand with a fineness modulus of 2.85 and a bulk density of 2650 kg/m^3 was used as the NFA. The particle size distributions of the river sand and the regraded RFA are shown in Fig. 1. CO_2 with a purity exceeding 99% was supplied by Changsha Dingyi Gas Co., Ltd. A polycarboxylate-based superplasticizer (SP) with a water-reducing capacity of approximately 30% was obtained from Shanghai Macklin Biochemical Technology Co., Ltd. Deionized water was used throughout the experiments.

Table 1. Main chemical compositions of cement (wt.%)

Cement	CaO	Fe_2O_3	Al_2O_3	SiO_2	MgO	SO_3	Others	LOI
P·O 42.5	59.8	4.9	4.2	19.2	2.5	3.2	2.1	4.1

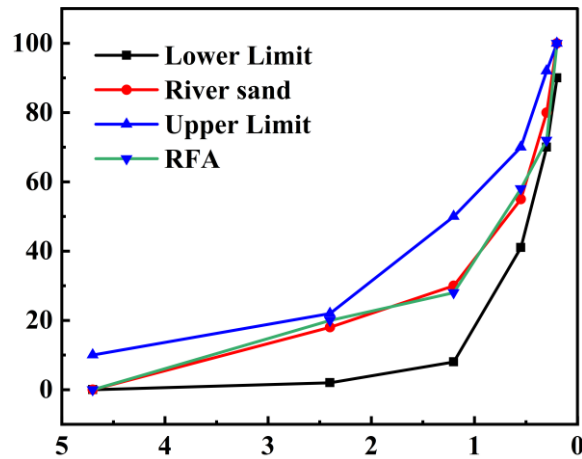


Fig. 1. Grain gradation of river sand and RFA

The mix proportion of the original mortar used for RFA preparation is presented in Table 2. Mortar specimens cured under standard conditions for 1 year were crushed using a jaw crusher, and the resulting RFA was sieved into five particle-size fractions, as shown in Table 3. The combined gradation of these fractions satisfied the continuous grading requirements for natural river sand specified in GB/T 14684–2022. Before wet carbonation, all RFA fractions were conditioned to the saturated surface-dry (SSD) state.

Table 2. Raw mortar mix proportion (kg/m^3)

Sample	Cement	Water	Sand	SP
Raw mortar	682	205	1364	1.4

Table 3. Particle size ranges of different RFA fractions (mm)

Property	RF0.15	RF0.30	RF0.60	RF1.18	RF2.36
Particle size distribution	0.15-0.30	0.30-0.60	0.60-1.18	1.18-2.36	2.36-4.75

2.2. Preparation of recycled mortar

The RFA fractions before and after carbonation were recombined according to the particle size distribution of natural river sand. Mortars were prepared with a water-to-binder ratio of 0.4 and a sand-to-binder mass ratio of 2. Natural river sand was replaced by RFA at replacement ratios of 50%, 70%, and 100%. The mix proportions are detailed in Table 4.

Table 4. Mortar mix proportions with different replacement ratios of RFA (kg/m³)

Sample	Cement	Water	Sand	RFA	SP
N100R0	682	273	1364	0	1.4
N50R50	682	273	682	682	1.4
N30R70	682	273	409.2	954.8	1.4
N0R100	682	273	0	1364	1.4

2.3. Methods

2.3.1. Wet carbonation curing process

The wet carbonation setup used in this study is shown in Fig. 2. RFA was mixed with deionized water at a liquid-to-solid mass ratio of 10:1 and pre-soaked for 15 min. Subsequently, CO₂ was bubbled through the slurry at a constant flow rate of 0.1 L/min for 1 h at 20 ± 2 °C, while the slurry was stirred at 400 rpm. The carbonation duration was selected based on previous studies, which demonstrated that wet carbonation for approximately 1 h is sufficient to achieve the effective modification of recycled aggregate under comparable experimental conditions^[26]. The carbonated RFA (CRFA) was designated according to particle size and carbonation duration; for example, RF0.15-1 represents aggregate with a particle size of 0.15–0.30 mm carbonated for 1 h.

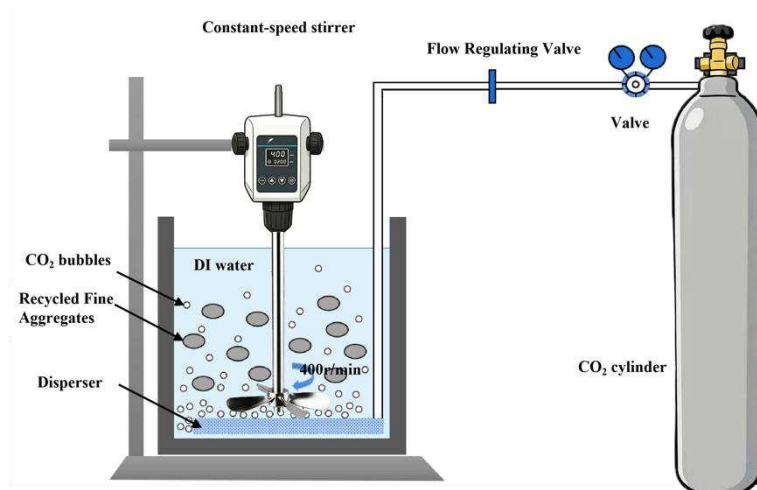


Fig. 2. Wet carbonation device

2.3.2. pH test

Deionized water was boiled to remove dissolved CO₂, and then mixed with the RFA at a 1:10 solid-to-liquid ratio to form a slurry. The slurry was subsequently transferred to a sealed reactor with a gas inlet, and a calibrated pH electrode was immersed into the mixture. At ambient temperature, the suspension was continuously stirred at a constant rate while CO₂ gas was purged through it. The pH was monitored at predefined intervals throughout the experiment.

2.3.3. ICP-OES test

During wet carbonation at $20 \pm 2^\circ\text{C}$, aliquots of the RFA suspension were periodically collected, filtered, and acidified. The concentrations of Ca²⁺ and Na⁺ in the solution were monitored using inductively coupled plasma optical emission spectrometry (ICP-OES).

2.3.4. Physical property tests of RFA

The water absorption, apparent density, and crushing value of RFA were determined in accordance with GB/T 14684-2022. Water absorption was determined from the mass difference between oven-dried and SSD aggregates after water immersion. Apparent density was measured using the pycnometer method based on water displacement. The crushing value was determined by subjecting a standard aggregate sample to compressive loading using a hydraulic compression testing machine, and was calculated as the percentage of crushed material passing the specified sieve. Each test was performed in triplicate, and the average value was reported.

2.3.5. TG/DTG test

The samples were immersed in isopropanol to terminate further hydration and carbonation reactions, then ground to a powder using an agate mortar. The samples were dried at 40 °C, and approximately 10 mg of powder was tested under a nitrogen flow of 50 mL/min. The temperature was increased from 35 °C to 950 °C at a heating rate of 10 °C/min, and the mass change with temperature was recorded.

2.3.6. FTIR test

Fourier-transform infrared spectroscopy (FTIR) was used to analyze the functional group characteristics of the samples, employing a Nicolet iS20 spectrometer (Thermo Fisher Scientific). The samples were immersed in isopropanol to terminate hydration, then ground into powder, and vacuum-dried to constant weight. Spectral data were collected in the range of 2000–500 cm⁻¹.

2.3.7. SEM test

Vacuum-dried RFA samples were sputter-coated with a thin layer of gold to improve surface conductivity, and their microstructural characteristics were examined using a scanning electron microscope (TESCAN MIRA LMS) operated at an accelerating voltage of 20 kV under high vacuum conditions, with observations conducted at magnifications ranging from 500 to 5000.

2.3.8. BET test

The specific surface area, pore volume, and pore size distribution were analyzed by nitrogen adsorption-desorption isotherms at 77 K. Prior to testing, all samples were vacuum-dried at 40 °C for 48 h. The Barrett-Joyner-Halenda (BJH) model was applied to the desorption branch to determine the pore size distribution.

2.3.9. Compressive strength test

Compressive strength was tested in accordance with the Chinese standard GB/T 17671-2021 (Method of testing cements—Determination of strength). Specimens with dimensions of 40 × 40 × 160 mm were loaded at a rate of 2.4 kN/s after 28 d of curing. Three replicate specimens were tested for each group, and the average value was reported as the compressive strength.

3. Results

3.1. Effect of particle size on pH and ion concentrations of the solution

3.1.1. pH

Fig. 3 presents the evolution of solution pH during wet carbonation of RFA with different particle sizes. The initially alkaline solution resulted from the dissolution of unhydrated cement phases and alkaline hydration products in the adhered mortar, which released alkaline species into the aqueous phase^[27]. At the initial stage of CO₂ injection, the solution pH in all RFA fractions exhibited a rapid initial decline. This can be attributed to the dissolution of gaseous CO₂ in the solution, which generated carbonate species (CO₃²⁻ and HCO₃⁻) and decreased the solution pH^[28]. Previous studies have shown that at pH > 10.5, CO₃²⁻ is the predominant carbonate species, with a much higher concentration than HCO₃⁻, which favors the formation of large amounts of CaCO₃. In contrast, at pH < 7, CO₃²⁻ almost completely disappears, thereby preventing the effective precipitation of Ca²⁺^[29]. Although all samples followed the same initial carbonation pathway, the subsequent evolution of pH exhibited a pronounced dependence on particle size. The pH of the RF0.15 suspension decreased more slowly than that of RF2.36 and stabilized only after approximately 40 min. This behavior is attributed to the larger specific surface area and higher

adhered mortar content of finer RFA, both of which promoted continuous ion leaching into the solution. The sustained release of alkaline species maintained a higher solution alkalinity, thereby buffering the decrease in pH during carbonation^[28]. These results demonstrate that particle size influences the evolution of solution alkalinity by regulating ion leaching during wet carbonation.

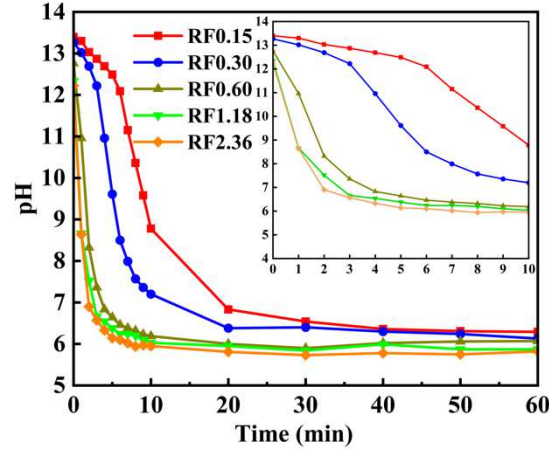


Fig. 3. Evolution of solution pH during wet carbonation of RFA with different particle sizes

3.1.2. Ion concentration

The Ca^{2+} concentration reflects the availability of reactive calcium for carbonation, whereas Na^+ is mainly released from the dissolution of C–S–H gel^[30]. Accordingly, the concentrations of Ca^{2+} and Na^+ in the solution during carbonation were monitored. Fig. 4 shows the evolution of Ca^{2+} and Na^+ concentrations in the solution during carbonation for RFA of different particle sizes. The initial concentrations of Ca^{2+} and Na^+ increased markedly with decreasing particle size, indicating enhanced dissolution kinetics in the finer fractions. This trend is attributed to the larger specific surface area and higher adhered mortar content of the finer fractions, both of which facilitate ion release.

Following CO_2 injection, Ca^{2+} concentration decreased sharply because Ca^{2+} rapidly reacted with dissolved carbonate species to precipitate CaCO_3 under the initially alkaline conditions. As carbonation proceeded, the continuous decrease in pH promoted dissolution of calcium-bearing phases, leading to a gradual recovery of the Ca^{2+} concentration until equilibrium was reached. The extent of this rebound became more pronounced in the finer fractions, suggesting that particle size affected the dissolution behavior of calcium-bearing phases during wet carbonation. In contrast, the Na^+ concentration increased continuously throughout the carbonation, reflecting the progressive decalcification and dissolution of C–S–H gel as carbonation proceeded^[27]. The substantially greater increase in Na^+ concentration observed for RF0.15 indicates that the dissolution of C–S–H gel became increasingly pronounced with decreasing particle size.

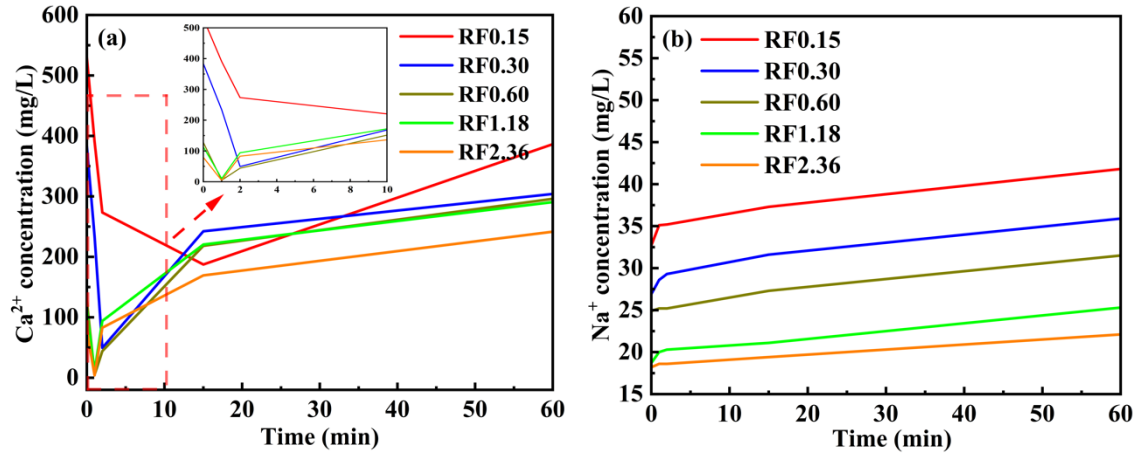


Fig. 4. Evolution of Ca^{2+} and Na^{+} concentrations during wet carbonation of RFA with different particle sizes

3.2. Effect of particle size on the physical properties of CRFA

3.2.1. Water absorption

Fig. 5 presents the water absorption of RFA with different particle sizes before and after wet carbonation. Before carbonation, water absorption decreased with increasing particle size, and the same trend was maintained after carbonation. This is mainly attributed to the fact that the surface of the coarser RFA has less adhered mortar content, which results in lower initial porosity and consequently lower water absorption^[31]. The effect of carbonation exhibited a clear transition at approximately 1.18 mm. For RFA fractions in the 0.15–1.18 mm range, water absorption increased by 6.1%, 4.8%, and 5.6% after carbonation, suggesting a deterioration in the physical performance of RFA finer than 1.18 mm. For RFA fractions in the 1.18–4.75 mm range, water absorption decreased by 0.9% and 9.0% following carbonation. This divergence may be attributed to the dissolution of calcium-bearing phases in the RFA finer than 1.18 mm, which likely increased internal porosity during carbonation, together with the structural deterioration induced by prolonged wet carbonation^[28]. Under the same carbonation duration, RFA larger than 1.18 mm underwent a more moderate carbonation process, which may have promoted the deposition of carbonation products within the pore structure and contributed to reduced water absorption.

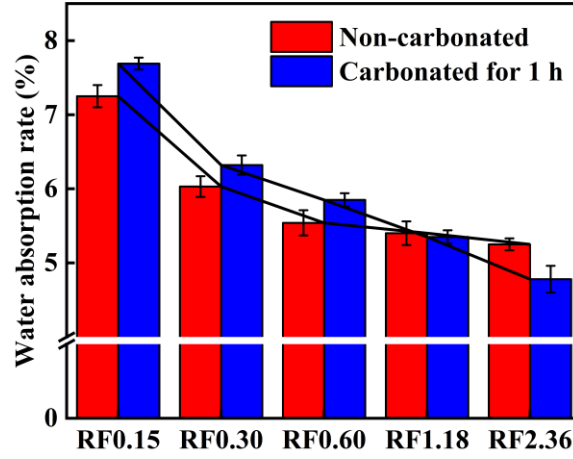


Fig. 5. Water absorption of RFA before and after wet carbonation

3.2.2. Apparent density

Fig. 6 illustrates the variation in the apparent density of RFA with different particle sizes before and after wet carbonation. Before carbonation, apparent density increased with increasing particle size, and the same trend was maintained after carbonation. However, the effect of carbonation on apparent density exhibited a distinct transition at a critical particle size of 1.18 mm. For RFA fractions finer than 1.18 mm, wet carbonation led to a decrease in apparent density. Specifically, after carbonation, the apparent densities of RF0.15, RF0.30, and RF0.60 decreased by 1.6%, 1.2%, and 1.6%, respectively. Conversely, for RFA fractions larger than 1.18 mm, the apparent density increased after carbonation, with RF1.18 and RF2.36 exhibiting increases of 0.4% and 2.3%, respectively. Combined with the water absorption results, these findings indicate that wet carbonation increased the porosity of RFA fractions finer than 1.18 mm. This deterioration may be attributed to the larger specific surface area of finer RFA, which facilitates more extensive interaction between the aggregate surface and dissolved CO_2 species during wet carbonation, potentially leading to microstructural deterioration. In contrast, for RFA fractions larger than 1.18 mm, the formation of carbonation products may have contributed to improved aggregate compactness and reduced water absorption, as reflected by the increased apparent density after carbonation^[27].

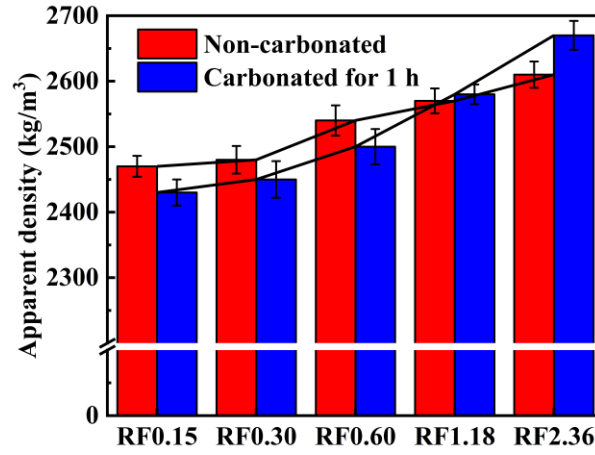


Fig. 6. Apparent density of RFA before and after wet carbonation

3.2.3. Crushing value

Fig. 7 presents the crushing values of RFA with different particle sizes before and after wet carbonation. The effect of wet carbonation on crushing value exhibited a clear transition at approximately 1.18 mm. For RFA finer than 1.18 mm, the crushing value increased by 4.5%–8.0% after carbonation, indicating a deterioration in mechanical resistance. This deterioration is attributed to the decalcification of surface C–S–H gel during carbonation, which weakened the adhered mortar^[32]. In contrast, for RFA larger than 1.18 mm, the crushing value decreased by 5.5%–8.8%, suggesting that the precipitation of carbonation products enhanced the mechanical integrity of the adhered mortar. These results are consistent with the water absorption and apparent density measurements, further confirming that wet carbonation exerts opposite effects on the physical properties of fine and coarse RFA fractions.

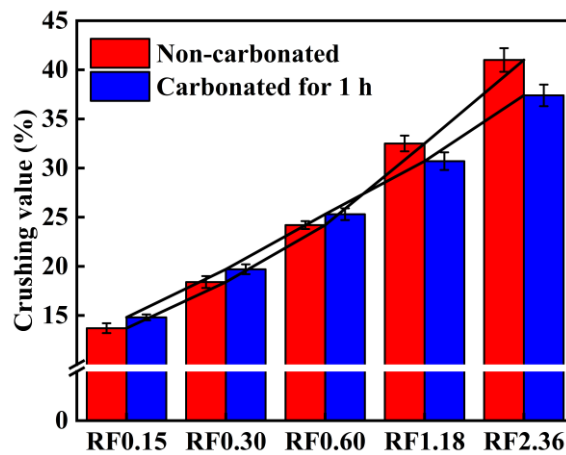


Fig. 7. Crushing value of RFA before and after wet carbonation

3.3. Effects of particle size on the phase composition of CRFA

3.3.1. TG/DTG analysis

The phase evolution of CRFA was evaluated from the mass loss characteristics over different temperature ranges to investigate the influence of particle size on carbonation products. As shown in Fig. 8, the TG/DTG curves display three major mass loss peaks, corresponding to distinct processes: the mass loss between 50–200 °C is associated with the evaporation of bound water; that between 400–500 °C with the thermal decomposition of Ca(OH)_2 ; and that between 550–800 °C with the decomposition of CaCO_3 ^[33,34].

Prior to wet carbonation, the RFA had experienced a certain degree of natural carbonation during storage, leading to variations in the contents of Ca(OH)_2 and CaCO_3 among different particle-size fractions. As shown in Fig. 8(a) and Fig. 8(b), the Ca(OH)_2 peak intensity of RF0.15-0 was lower than that of RF2.36-0, whereas its CaCO_3 peak intensity was higher, suggesting that finer RFA (RF0.15) experienced a higher degree of natural carbonation. After wet carbonation, the Ca(OH)_2 content in RF0.15 decreased markedly, while the CaCO_3 content increased substantially. By comparison, the changes in RF2.36 were much less pronounced, suggesting a relatively limited phase transformation during carbonation. Interestingly, the markedly different physical-property evolution of RF0.15 and RF2.36 cannot be explained solely by the extent of CaCO_3 formation, suggesting that the distribution and reaction pathway of the carbonation products should also be considered.

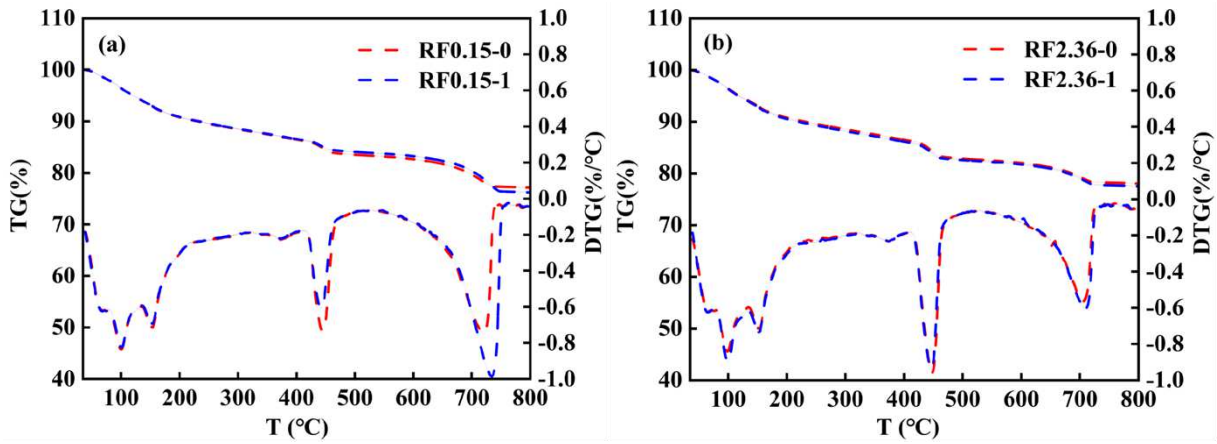


Fig. 8. TG and DTG curves of CRFA with different particle sizes

Based on the analysis of the TG curves, the contents of Ca(OH)_2 and CaCO_3 in CRFA with different particle sizes are presented in Fig. 9. After wet carbonation, the CaCO_3 content of RF0.15 increased from 14.5% to 17.9%, while that of RF2.36 increased only from 11.0% to 11.5%. However, this increase in CaCO_3 content did not translate into improved physical properties for the finer RFA fractions. This may be attributed to the extensive surface carbonation occurring locally on finer RFA fractions, thereby degrading their microstructure. For RF0.15, the Ca(OH)_2 content decreased by 14.7%, whereas the CaCO_3 content increased by 23.1%. Because the increase in CaCO_3 exceeded the decrease in Ca(OH)_2 , the results suggest that CaCO_3 was produced not only from Ca(OH)_2 carbonation but also from the decalcification and carbonation of C–S–H gel^[28].

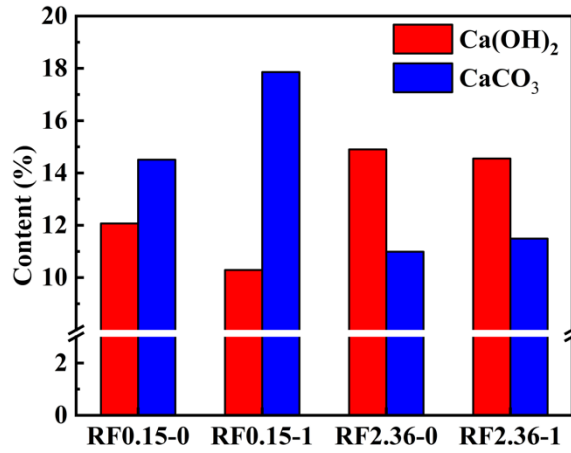


Fig. 9. Ca(OH)₂ and CaCO₃ content of CRFA with different particle sizes

3.3.2. FTIR analysis

Fig. 10 presents the FTIR spectra of RF0.15 and RF2.36 after wet carbonation, together with that of the suspended powder recovered from the carbonation solution of RF0.15 (denoted as RF0.15-F) to further identify the products generated during carbonation. The absorption peaks at approximately 710 cm⁻¹, 873 cm⁻¹, and 1420 cm⁻¹ are assigned to the C–O bond vibrations of CaCO₃^[35]. Compared with RF2.36, RF0.15 exhibited more pronounced peaks at around 873 cm⁻¹ and 1420 cm⁻¹, indicating a higher CaCO₃ content. This finding is consistent with the TG analysis results. The absorption peaks located between 900 cm⁻¹ and 1200 cm⁻¹ are attributed to the Si–O bond vibrations^[34]. Specifically, the peak at 963 cm⁻¹ corresponds to the Q² units in the C–S–H gel^[36]. Compared with RF2.36, RF0.15 exhibited a lower intensity of the 963 cm⁻¹ band, suggesting pronounced structural modification of C–S–H during carbonation, likely associated with enhanced decalcification of C–S–H in the finer fraction. Furthermore, RF0.15-F exhibited pronounced absorption peaks near 710 cm⁻¹, 870 cm⁻¹, and 1400 cm⁻¹, indicating the presence of a substantial amount of CaCO₃. The broadening and shift of the FTIR peaks of RF0.15-F from the 900–1200 cm⁻¹ region to approximately 1030 cm⁻¹ suggest that the C–S–H gel underwent decalcification–polymerization, generating abundant highly polymerized silica gel^[37], thus confirming the extensive carbonation of C–S–H gel in RF0.15.

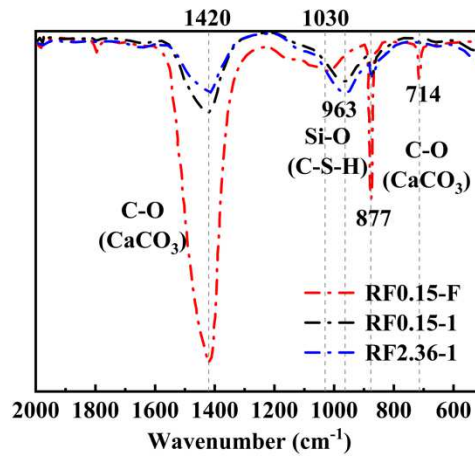


Fig. 10. FTIR spectra of CRFA with different particle sizes

3.4. Effects of particle size on the microstructure of CRFA

3.4.1. Micromorphology

Fig. 11(a) and Fig. 11(b) present the surface morphologies of CRFA with different particle sizes observed by SEM. All CRFA fractions were covered by newly precipitated CaCO_3 , this surface precipitation is attributed to the dissolution of Ca^{2+} from the recycled aggregate into the solution and its subsequent reaction to form CaCO_3 . As shown in the Fig. 11(a), for the RF0.15, the surface was covered by numerous fine CaCO_3 particles with poor crystallinity, indicating that rapid nucleation dominated over crystal growth during carbonation^[38]. Furthermore, these fine carbonate particles were loosely interconnected by silica gel generated from C–S–H decalcification^[27], resulting in a porous and discontinuous surface layer. Such a microstructure is consistent with the increased pore volume and deterioration of the physical properties observed for the finer fractions. For the RF2.36, fewer but substantially larger and well-crystallized CaCO_3 crystals were observed, suggesting that slower nucleation favored subsequent crystal growth. These crystals formed a continuous and dense carbonate layer capable of filling surface pores and micro-cracks, thereby contributing to pore refinement and the improved physical properties of the coarse RFA.

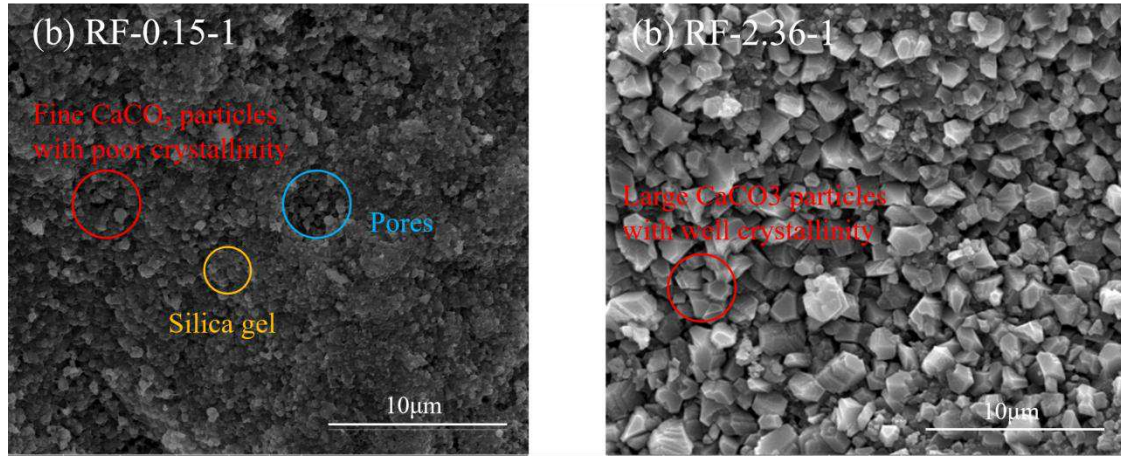


Fig. 11. SEM images of CRFA with different particle sizes

3.4.2. Pore structure

The pore structure of CRFA with different particle sizes was characterized by BET analysis, and the corresponding total pore volumes are summarized in Table 5. After wet carbonation, the total pore volume of RF0.15 increased by 18.7%, indicating a substantial increase in pore development. In contrast, RF2.36 exhibited a slight reduction in total pore volume of 2.6% after carbonation. These results are consistent with the macroscopic physical property changes observed for the respective fractions. Fig. 12(a) and Fig. 12(b) illustrate the variations in pore structure of RFA with different particle sizes before and after wet carbonation. After carbonation, RF0.15 exhibited an increase in pores within the 1–3 nm range and around 50 nm, leading to a higher total pore volume. This pore development is consistent with the accelerated Ca^{2+} leaching observed in the ion concentration analysis and the decalcification of C–S–H gel revealed by FTIR, indicating that accelerated calcium release and C–S–H decalcification contributed to the deterioration of the pore structure and the generation of additional pores. This pore coarsening is consistent with the deterioration of the physical properties observed for the finer fraction. In contrast, for RF2.36, the volume of less harmful pores was relatively reduced after wet carbonation^[39,40], accompanied by a slight leftward shift of the BET peak in the 30–60 nm range, indicating pore structure refinement. This refined pore structure agrees with the reduced water absorption, increased apparent density and lower crushing value of RF2.36 after carbonation.

Table 5. Total pore volume of CRFA with different particle sizes (cc/g)

Property	RF0.15-0	RF0.15-1	RF2.36-0	RF2.36-1
Total pore volume	0.03923	0.04657	0.03138	0.03056

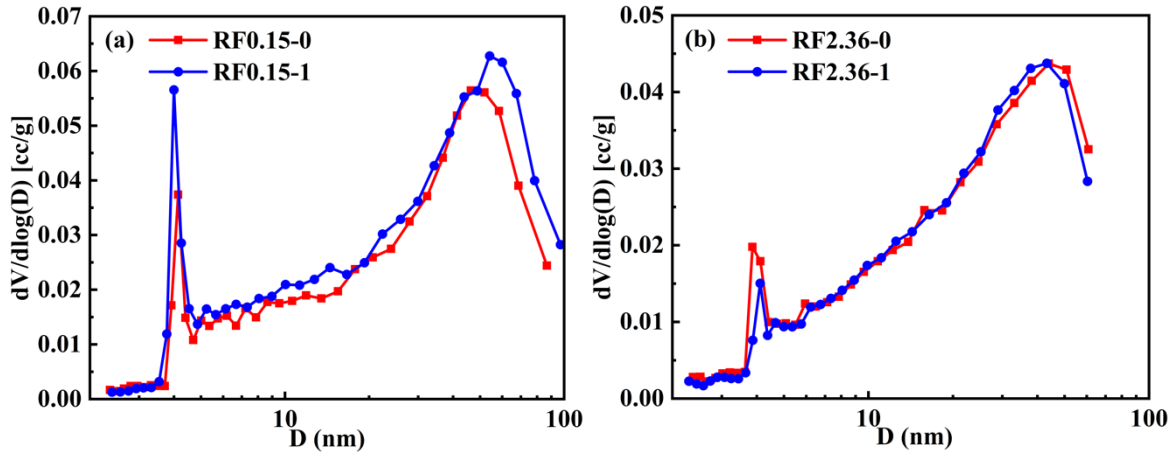


Fig. 12. Pore size distribution curve of CRFA with different particle sizes

Combined SEM and BET analyses show that rapid surface precipitation of fine CaCO_3 crystals in RF0.15 was accompanied by C–S–H decalcification and pore generation, resulting in an overall increase in porosity. The dense surface precipitation likely restricted the inward diffusion of dissolved carbonate species, thereby limiting carbonation within the particle interior. In contrast, RF2.36 developed larger and well-crystallized CaCO_3 particles that preferentially filled the internal pore network, suggesting that the slower surface reaction provided sufficient time for dissolved CO_3^{2-} to diffuse into the aggregate interior before precipitation. These results suggest that particle size plays an important role in governing the carbonation pathway of RFA by regulating ion leaching behavior and the formation characteristics of carbonation products. Accordingly, the finer fraction tended to undergo surface-dominated carbonation accompanied by C–S–H decalcification and pore coarsening, whereas the coarser fraction exhibited more pronounced pore refinement, likely associated with the deposition of carbonation products within the pore network.

3.5. Effects of CRFA on the compressive strength of recycled mortar

Fig. 13 presents the 28 d compressive strengths of recycled mortars incorporating uncarbonated RFA and CRFA. For mortars incorporating uncarbonated RFA, compressive strength decreased with increasing aggregate replacement ratio, which is attributable to the higher defect content and inferior physical properties of porous RFA. Compared with uncarbonated RFA, the use of CRFA increased the compressive strength of recycled mortars, with the strengthening effect becoming more pronounced as the aggregate replacement ratio increased. Specifically, when the replacement ratio of RFA reached 100%, the compressive strength of the recycled mortar prepared with CRFA increased by 25.1% compared with the mortar prepared with uncarbonated RFA.

Despite the varying effects of carbonation on different RFA particle-size fractions, recycled mortar prepared with the recombined CRFA still exhibited an overall increase in 28 d compressive strength. This improvement is

primarily attributed to the improved physical properties of the 1.18-4.75 mm CRFA fractions, where pore filling by well-crystallized CaCO_3 effectively reduced the inherent defects of the recycled aggregate. Meanwhile, the silica gel generated by extensive carbonation of the 0.15-1.18mm fractions reacts with $\text{Ca}(\text{OH})_2$ released during cement hydration to generate secondary C–S–H gel, which may contribute to the densification of the interfacial transition zone^[17,41]. Therefore, the overall strength enhancement is likely attributable to the combined effects of improved physical properties of the 1.18-4.75 mm fractions and the potential secondary hydration associated with the 0.15-1.18 mm fractions.

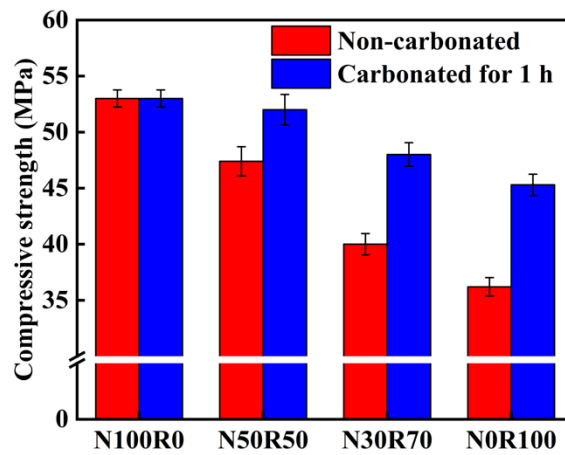


Fig. 13. 28 d compressive strength of recycled mortars with different RFA replacement ratios

4. Discussion

During the wet carbonation of RFA, a pronounced particle-size-dependent divergence in modification performance was observed. Despite their greater pH buffering capacity and faster ion leaching rates, RFA particles with sizes finer than 1.18 mm exhibited a certain deterioration in physical properties, as revealed by physical and microstructural characterization. In contrast, RFA particles with sizes larger than 1.18 mm exhibited more pronounced improvements in physical properties upon carbonation. This phenomenon may be attributable to the carbonation of RFA being governed not only by Ca^{2+} leaching but also by the extent of CO_3^{2-} diffusion.

Unlike gas–solid carbonation, where CO_2 penetration is primarily governed by gaseous diffusion through the pore network of aggregates, wet carbonation involves the transport of dissolved carbonate species in the liquid phase. Therefore, CO_3^{2-} diffusion during wet carbonation is controlled not only by the pore structure but also by the precipitation of surface carbonation products. Based on the experimental results, the mechanisms of wet carbonation for RFA of different particle sizes are illustrated in Fig. 14. For RFA particles finer than 1.18 mm, the relatively large specific surface area markedly accelerated Ca^{2+} leaching, which, coupled with the dissolution of

certain hydration products, led to an increase in the original porosity of the RFA. Under these conditions, the elevated Ca^{2+} concentration promoted rapid precipitation of fine CaCO_3 crystals on the particle surface. The resulting carbonate layer may have restricted further inward diffusion of CO_3^{2-} , thereby confining carbonation mainly to the surface region and leaving the interior pores insufficiently filled. Mechanistically, it is speculated that the concurrent decalcification of C–S–H gel in the RFA surface layer to form silica gel may, to some extent, impair its capacity as the primary load-bearing phase to bind solid constituents such as aggregate, unhydrated cement particles, and hydration products, thereby resulting in a loosened RFA skeleton^[42]. It is thus speculated that the increased porosity induced by the dissolution of calcium-bearing phases in RFA particles finer than 1.18 mm, together with the reduced C–S–H content and the loosened RFA skeleton resulting from C–S–H gel decalcification, may be the primary causes of the deterioration in physical properties. Similarly, Jiang et al. reported that for RFA with particle sizes of 0.15–0.30 mm, the amount of silica gel formed reached as high as 69.5%, considerably higher than that in other size fractions, and this was accompanied by pore coarsening and carbonation products exhibiting a loosely packed, cluster-like morphology^[28]. Conversely, for RFA particles larger than 1.18 mm, the smaller specific surface area limited the extensive leaching of Ca^{2+} , thereby keeping the Ca^{2+} concentration in the solution at a relatively low level during the early stage of carbonation. The slow and sustained carbonation reaction was insufficient to rapidly consume CO_3^{2-} at the particle surface, thereby providing a time window for CO_3^{2-} to diffuse deeper into the interior pores of the RFA. Consequently, CO_3^{2-} could diffuse further into the interior pore network of the RFA before reacting with dissolved Ca^{2+} , which is presumed to promote the in-situ precipitation of CaCO_3 within the pores. This process may contribute to pore refinement and matrix densification, while the slower carbonation kinetics may also favor the formation of larger, well-crystallized CaCO_3 particles on the aggregate surface.

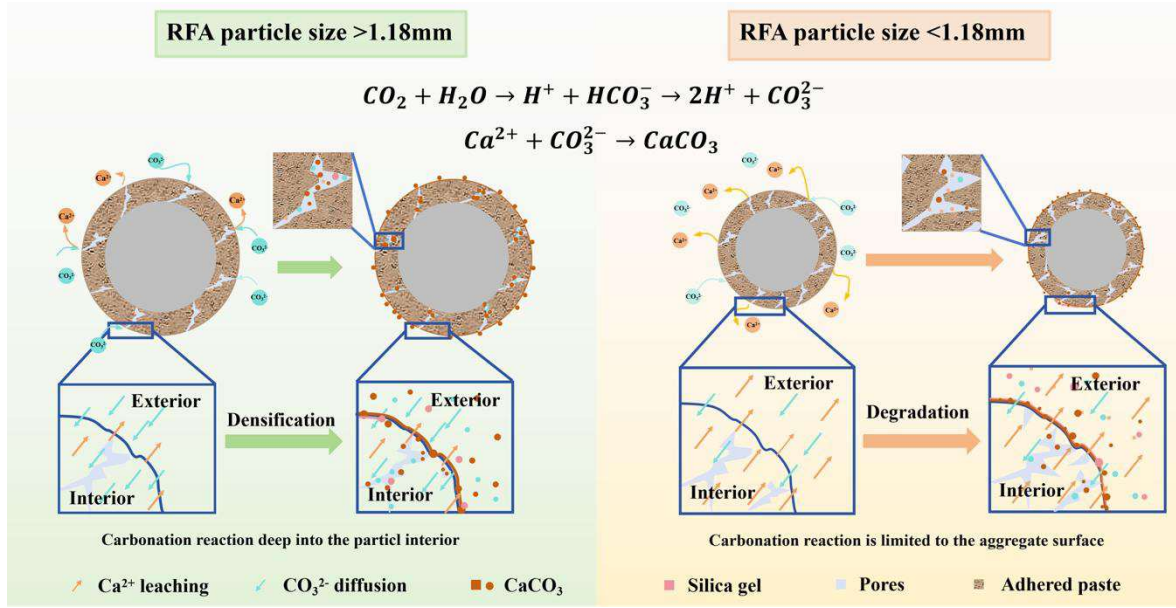


Fig. 14. Mechanisms of product distribution and microstructural evolution in wet carbonation of RFA with different particle sizes.

Fang et al. demonstrated that a 10-minute wet carbonation treatment significantly improved the overall performance of RFA, highlighting that short-term wet carbonation is highly effective for the rapid modification of RFA^[43]. However, in this study, when the carbonation time was extended to 1 h, it was found that this densification, caused by the excessively rapid surface carbonation of RFA, became the dominant factor responsible for the over-carbonation of particles finer than 1.18 mm. The CaCO₃ crystal layer on the RFA surface formed rapidly after the onset of carbonation, thereby hindering the inward diffusion of CO₃²⁻. Concurrently, the silica gel produced by C–S–H decalcification adversely affected the microstructure, ultimately resulting in pore structure deterioration. For RFA particles larger than 1.18 mm, appropriately extending the carbonation time facilitated more thorough inward diffusion of CO₃²⁻, thereby enabling in-situ filling of the interior pores within the RFA. These results suggest that distinct optimal carbonation duration windows exist for different particle size fractions: short-term rapid carbonation is suitable for the surface modification of fine RFA particles (finer than 1.18 mm), whereas relatively longer carbonation is more favorable for the thorough internal carbonation of coarser RFA particles (larger than 1.18 mm). Therefore, establishing recovery processes tailored to different particle size fractions of RFA can better facilitate its resource utilization.

Despite the deterioration in the physical properties of carbonated fine RFA particles finer than 1.18 mm, the mortar incorporating recombined CRFA exhibited a 25.1% increase in 28 d compressive strength compared with that prepared using uncarbonated RFA, indicating that the beneficial effects of the carbonated 1.18–4.75 mm fractions, together with the potential secondary hydration associated with the 0.15–1.18 mm fractions, outweighed

the adverse effects associated with the deterioration of the 0.15-1.18 mm fractions. After wet carbonation, RFA particles larger than 1.18 mm developed a densified matrix, with their interior pores filled and a well-crystallized CaCO_3 crystal layer formed on the surface, which enhanced the aggregate's physical properties and consequently further improved the load-bearing capacity of the mortar. Although RFA particles finer than 1.18 mm exhibited deteriorated physical properties, the silica gel formed on their surfaces through C–S–H decalcification retains pozzolanic activity and could react with $\text{Ca}(\text{OH})_2$ released during cement hydration to form additional C–S–H gel, potentially contributing to matrix densification and improving interfacial bonding^[44]. In addition, the nano- CaCO_3 generated during carbonation may serve as nucleation sites for cement hydration products, thereby promoting cement hydration and contributing to the densification of the recycled mortar matrix. Collectively, these mechanisms are likely to partially offset the adverse influence of the deteriorated physical properties of the 0.15-1.18 mm fractions.

5. Conclusions

This study investigated the influence of particle size on the wet carbonation of RFA by separately carbonating different particle-size fractions, followed by evaluation of the physical properties of the resulting CRFA and the mechanical performance of mortars prepared with the recombined aggregates. The main conclusions are as follows:

(1) The 0.15-1.18 mm RFA fractions exhibited higher initial solution pH and accelerated dissolution of calcium-bearing phases, resulting in higher Ca^{2+} and Na^+ concentrations during wet carbonation. Nevertheless, the enhanced ion leaching did not improve the effectiveness of carbonation modification and was instead accompanied by deterioration of the physical properties of the 0.15-1.18 mm fractions.

(2) Wet carbonation exerted distinct effects on the physical properties of different RFA particle-size fractions, with 1.18 mm identified as the critical particle size. For the 0.15-1.18 mm RFA fractions, water absorption and crushing value increased, while apparent density decreased. In contrast, the 1.18-4.75 mm RFA fractions exhibited reduced water absorption and crushing value together with increased apparent density, indicating an improvement in aggregate physical properties.

(3) Carbonation of 0.15-0.30 mm RFA fractions generate CaCO_3 particles with finer particle size and lower crystallinity, accompanied by C–S–H carbonation to form silica gel. This generates numerous new pores in the approximately 50 nm range, thereby increasing the total pore volume by 18.7% and resulting in structural degradation. The 2.36-4.75 mm RFA fractions, in contrast, form larger and more crystalline CaCO_3 on their surface,

effectively filling the internal pores and micro-cracks, thereby reducing total pore volume and significantly refining the pore structure.

(4) Recombining the individually carbonated RFA fractions to match the particle-size gradation of NFA improved the mechanical performance of recycled mortars. At a 100% RFA replacement ratio, the 28 d compressive strength increased by 25.1% compared with mortar prepared using uncarbonated RFA.

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