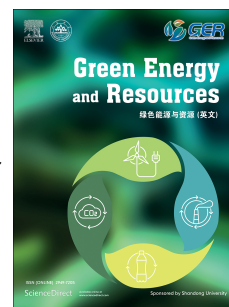


Journal Pre-proof

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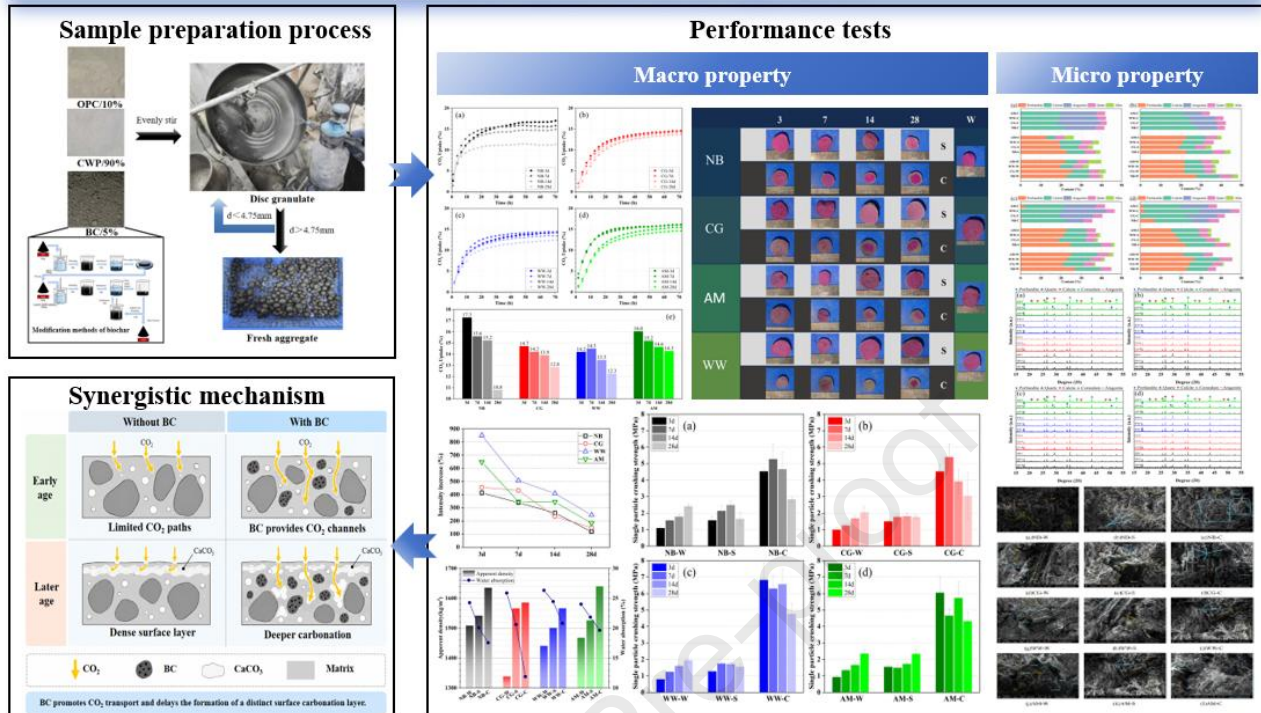
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Research on the carbon sequestration potential and performance of modified biochar promoting carbon capture artificial aggregates



Research on the carbon sequestration potential and performance of modified biochar promoting carbon capture artificial aggregates

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Abstract: Currently, due to the dense nature of the concrete matrix and the formation of a dense calcium carbonate layer during carbonation that hinders further CO₂ diffusion, the carbonation process of concrete takes a long time. Biochar (BC), characterized by its porous structure and chemical stability, has shown potential in improving the mechanical properties and carbon sequestration capacity of materials. However,

the influence mechanism of BC obtained by different modification methods on material performance remains unclear. To address this, this study developed concrete waste-biochar aggregate (CWBA) using construction waste powder (CWP) and BC, and investigated the effect of different BC modification processes on the performance of CWBA. In this study, a BC dosage of 5% was adopted, and BC was treated by water washing and alkali modification, respectively, while an unmodified group was used as a control. The physical properties, single particle compressive strength, and CO₂ uptake of the prepared CWBA were evaluated through macro-experiments, and the mechanism was analyzed based on micro-experiments. Among all the tested groups, AM-C (alkali-modified BC, carbonation curing) and NB-C (non-biochar, carbonation curing) exhibited the highest aggregate densities. At 3 days, the WW (water-washed BC) group exhibited the greatest carbonation gain, while at 28 days, the AM group showed the greatest carbonation gain and the NB group the smallest; the strength could be increased by up to 851% after carbonation. These results demonstrate that BC modified by alkali treatment may destroy the biomass tar layer on its surface and release the pore structure, thereby better offsetting the reduction in CO₂ uptake caused by matrix densification during curing, and providing greater carbonation gain under long-term service conditions.

Keywords: Construction waste powder; Biochar; Modified BC; Artificial aggregate; CO₂ uptake.

1. Introduction

Against the backdrop of accelerated global urbanization, the construction industry has consumed more resources and emitted more CO₂ (Barcelo et al., 2014; Yee, 2001). Moreover, the amount of construction waste generated during construction activities is also rising (Akhtar and Sarmah, 2018; Wang et al., 2026). Substantial CO₂ emissions contribute to global warming and a range of other climate-related issues (Marescaux et al., 2018). Consequently, China has proposed carbon neutrality, a strategy that can effectively mitigate the excessive CO₂ emissions from industrial activities and thus reduce the atmospheric concentration

of CO₂ (Weng et al., 2021). To achieve this goal, it is of great significance to develop methods for capturing and sequestering CO₂ from the air (Meng et al., 2022).

Construction waste (CW) originates from waste materials generated by urban building demolition. According to relevant data, the output of CW in China is gradually increasing; however, the utilization rate of CW in China remains around 40%, whereas in developed countries, the utilization rate in this field can exceed 80% (Aslam et al., 2020). Therefore, in the face of increasingly severe global environmental challenges, it is urgent to enhance the comprehensive utilization level of CW. Studies have shown that it is feasible to produce artificial aggregates from construction waste (Ding et al., 2025; Niu et al., 2026; Zheng et al., 2025). Research indicates that during the process of mechanically crushing waste CW to produce recycled aggregate (RA) as a substitute for natural aggregate (NA), the resulting concrete waste powder (CWP) may cause environmental issues related to water, air, and soil (Rakhimova and Shi, 2024), yet it still possesses a certain carbon sequestration capacity. CWP is rich in high-calcium components, creating a highly alkaline environment. When concrete materials are exposed to the atmosphere, CO₂ can penetrate through pores and chemically react with alkaline substances such as Ca(OH)₂ inside the concrete, thereby forming stable carbonates like CaCO₃, which achieves carbon sequestration (Gomez Hoyos and Vazquez, 2013; Kaliyavaradhan et al., 2020). However, due to the dense nature of the concrete matrix and the formation of a dense calcium carbonate layer during carbonation that hinders further CO₂ diffusion, the carbonation process of concrete in service is very slow (Wang and Fan, 2025; Xuan et al., 2016). Therefore, seeking an effective method to improve the carbonation rate of CWP is crucial for enhancing the resource utilization level and efficiency of CW.

The key to improving the carbonation efficiency of concrete lies in regulating and optimizing the microstructure of concrete. Biochar (BC) is a product obtained by pyrolyzing common biomass raw materials,

characterized by wide availability of raw materials, low processing difficulty, and the ability to digest and absorb biomass waste. It possesses a rich pore structure and high chemical stability (Chen et al., 2022), which can increase the pore structure of concrete, thereby enhancing carbonation efficiency. Many studies have shown that incorporating BC into cementitious materials can improve the mechanical properties and carbon sequestration capacity of cement-based materials (Chen et al., 2025; Gimbun et al., 2025; Gupta and Kashani, 2021; Mirabrishami et al., 2026; Roy and Nassiri, 2026; S. Song et al., 2023). Furthermore, Yang et al. (Yang et al., 2025) conducted experiments by adding different dosages of rice husk biochar, and the results showed that adding an appropriate amount of rice husk biochar under standard curing conditions significantly improved the mechanical properties of the specimens. Maljaee et al. (Maljaee et al., 2021) investigated the effect of waste-derived biochar on the performance of cement mortar and found that the compressive strength increased with biochar content up to an optimal proportion, beyond which the compressive strength decreased. Liu et al. (Liu et al., 2024) found that incorporating excessive BC could increase CO₂ uptake, but the additional pore structure introduced also adversely affected the structure of concrete waste-biochar aggregate (CWBA), and that a BC dosage of 5% yielded the optimal performance. In addition, different treatment methods, including water washing, ethanol modification, ultrasonic modification, and alkali modification, exert different effects on the properties of BC. Previous research showed that alkali-modified BC exhibited the best overall performance, whereas water-washed BC achieved the best balance between performance and cost. In comparison, ethanol modification and ultrasonic treatment generally require additional solvent-recovery processes or specialized equipment, which may increase operational complexity and cost and restrict their large-scale engineering applications. Therefore, considering modification effectiveness, economic feasibility, and engineering applicability, water washing and alkali modification were selected for comparison in this study. However, the influence mechanism of different modified BCs on the performance

of CWBA remains unclear. Therefore, it is very important to explore the effect of different modified BCs on aggregate performance.

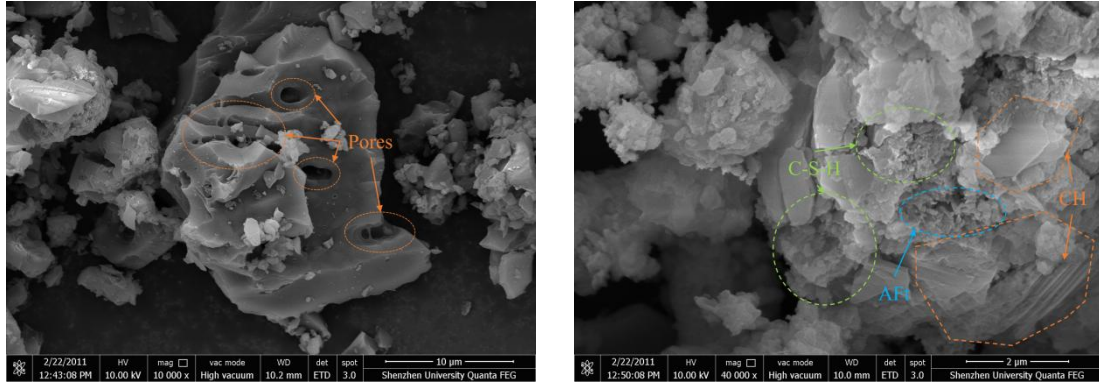
In summary, this study investigates the effect of different modified BCs on the performance of CWBA, and further explores the synergistic mechanism of BC on CWBA. A BC dosage of 5% was adopted, and the types of modified BC included those obtained by water washing and alkali modification, while unmodified BC was used as a control. The macro-performance of CWBA artificial aggregates with different modified BCs was evaluated through physical properties tests, single particle compressive strength test, and CO₂ uptake test. In addition, XRD and SEM tests were conducted to evaluate the micro-performance of different CWBAs, in order to elucidate the influence mechanism of different modified BCs on CWBA.

2. Materials and methods

2.1 Materials

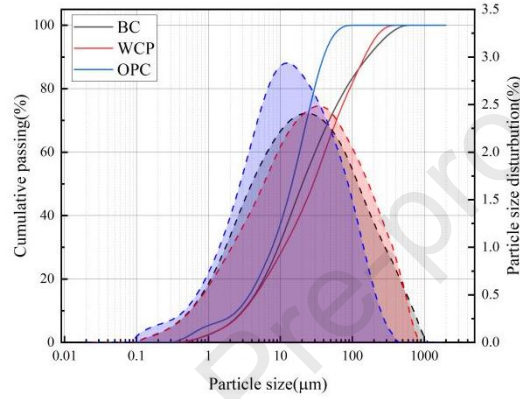
BC was produced using bamboo as the raw material and purchased from an energy saving company in Henan. Before use, BC was ground for 5 min using a laboratory-scale steel grinder and then passed through a 200-mesh sieve. And the measured water absorption capacity of BC was 62.2%. Conch brand P.O 42.5 OPC was adopted, which is in accordance with GB 175-2023. (Ministry of Industry and Information Technology, 2023). CWP was obtained from Conch P.O 42.5 OPC. SEM observations were performed on dried and gold-coated samples using a Quanta 250 FEG. Particle-size distributions were measured using a Mastersizer 3000 after ethanol dispersion, while XRD and XRF analyses were conducted using a D8 Advance on powders finer than 75 µm and an Axios WDXRF spectrometer on pressed pellets, respectively. These analyses followed JY/T 0584-2020, ISO 13320:2020, JY/T 0587-2020, and JY/T 0569-2020, respectively. The microstructural morphology, particle-size distributions, and XRD patterns of the raw materials are shown in **Fig. 1**, and the XRF result of the raw materials is presented in **Table 1**. It should be noted that the results

presented in **Table 1** are reported as relative elemental compositions because the purpose of the XRF analysis was to compare the distribution of inorganic elements among OPC, CWP, and BC. The SEM images show that the BC particles exhibited distinct tubular channels and abundant pores, whereas numerous crystalline structures were observed in CWP. OPC exhibited the finest particle-size distribution, whereas BC and CWP showed similar particle-size distributions because both materials had been passed through a 100-mesh sieve. In addition, the XRD results indicated that quartz was the predominant crystalline phase in BC, which was mainly attributed to the silicon-containing components of the biomass feedstock and was consistent with the elemental composition reported in Table 1. The XRD results of CWP showed that its principal constituents included portlandite, quartz, calcite, and C-S-H, most of which were derived from OPC hydration. In addition, the C, H, N, and O contents of BC were determined using Elementar: Vario UNICUBE. This method is commonly used for BC characterization (Sadrolodabae et al., 2026a, 2026b). The result is presented in Table 2. The C/O ratio of BC was 1.5, indicating a relatively high oxygen content and suggesting the possible presence of oxygen-containing functional groups, such as hydroxyl (-OH) and carboxyl (-COOH) groups. Its C/H ratio was 25, suggesting a relatively high degree of carbonization and aromaticity, together with a low hydrogen content and high chemical stability. The C/N ratio was 50, indicating a relatively low nitrogen content, which may be attributed to the decomposition and volatilization of protein-derived nitrogen compounds during pyrolysis. In addition, the BET result of BC is presented in **Table 3**, and the measurements were conducted in accordance with GB/T 19587-2017.

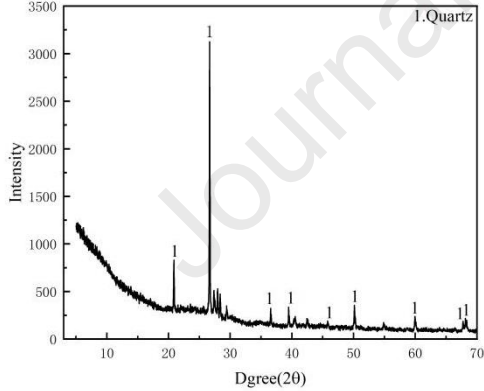


(a) Microstructure of BC

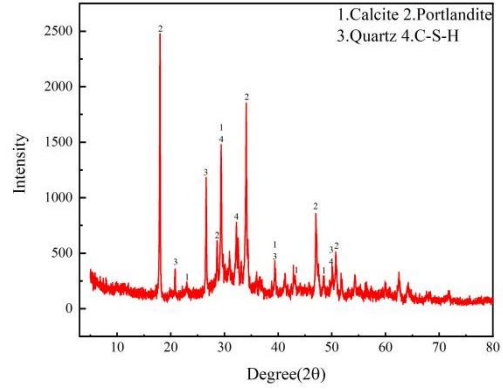
(b) Microstructure of CWP



(c) Particle size distribution of different raw materials



(d) BC-XRD



(e) CWP-XRD

Fig. 1. Morphology, particle-size distributions, and XRD patterns of the raw materials**Table 1. Relative elemental composition of the raw materials determined by XRF (wt.%)**

Type	Na	Mg	Al	Si	P	S	Cl	K	Ca	Others
OPC	0.12	2.64	3.74	15.76	0.04	1.79	0.07	0.98	69.28	5.58
CWP	0.18	2.57	4.14	15.35	0.06	1.49	0.13	0.85	69.14	6.09
BC	2.07	3.72	8.86	34.28	3.96	5.98	4.47	18.25	8.71	9.70

Table 2. CHNS / O test result of BC (wt.%)

Type	N	C	H	O
BC	1.498	74.924	3.046	50.0111

Table 3. BET test result of BC

Type	BET Specific Surface Area (m ² /g)	BJH Total Pore Volume (10 ⁻³ cc/g)	BJH Average Pore Diameter (nm)	D10 (μm)	D50 (μm)	D90 (μm)
BC	3.8	5.0	14.4	5.5	26.2	110.0

2.2 Modification methods of biochar

Fig. 2 shows the modification methods of BC. 200 g of BC was placed in a 2 L beaker, and 2 L of deionized water was added. The mixture was stirred using a mixer at a rotational speed of 550 rpm $\pm$ 10 rpm for 24 h, and then allowed to stand for 24 h until the solution stratified. The supernatant was decanted, and the BC precipitate at the bottom was taken and dried. The BC obtained by this procedure was designated as WW. Part of the WW was taken, placed into a 2 L beaker, and a 1 mol/L NaOH solution (pH = 14.00, 293.15 K) in an amount 10 times the mass of BC was added. After stirring for 24 h, the mixture was allowed to stand for 24 h until the solution stratified. The supernatant was then decanted, and an equal mass of deionized water was added. This dilution step was repeated five times, and the pH value of the solution was recorded each time. Finally, the BC was dried to obtain alkali-modified BC (named as AM).

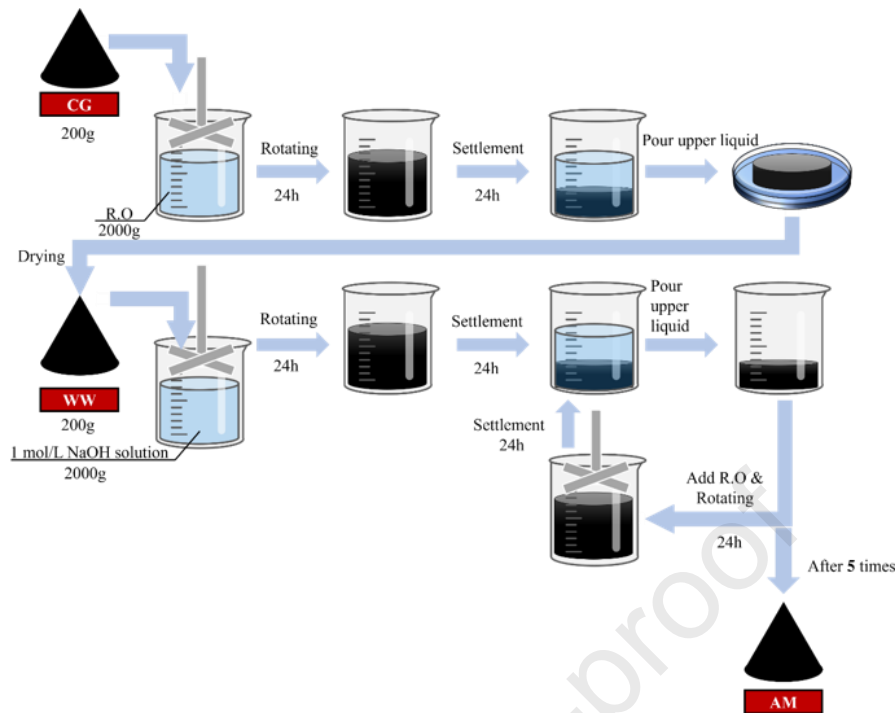


Fig. 2. Modification methods of biochar

2.3 Mix proportion design

In this experiment, four variables and three curing methods were set, resulting in a total of 12 mix proportions, as shown in **Table 4**. Except for the NB group, which contained no BC, the OPC:CWP mass ratio was maintained at 10:90, and BC was externally added at 5% of the combined mass of OPC and CWP. According to Gupta (Gupta and Kashani, 2021), this mix proportion yields the optimal aggregate performance. And preliminary granulation trials showed that stable aggregates were difficult to form at the other OPC:CWP mass ratios examined. The OPC:CWP mass ratio of 10:90 was selected to limit OPC consumption while providing sufficient binding for granulation and maintaining a high CWP utilization rate. In our preliminary study comparing BC dosages of 0%, 5%, 10%, and 15%, the 5% dosage provided the best balance between mechanical performance and CO₂ uptake, whereas higher BC dosages introduced additional pores and weakened the aggregate matrix (Liu et al., 2024). Based on the different pretreatment methods of BC, the groups were designated as conventional BC (abbreviated as CG), water-washed BC (WW), and

alkali-modified BC (AM). For standard curing (S), the aggregates were kept in a curing room at 20 ± 2 °C and RH = $95 \pm 5\%$ until the specified ages of 3, 7, 14, and 28 d. For water-sealed curing (W), the aggregates were completely immersed in tap water in sealed containers and kept in the same curing room. For carbonation curing (C), the aggregates were first water-sealed cured to the specified pre-curing age, dried at 65 °C to constant mass, and then carbonated for 48-72 h in the self-made carbonation box described in Section 2.5.3. The ages of 3, 7, 14, and 28 d refer to the pre-curing ages, and all carbonated groups were exposed to CO₂ for the same duration. Standard curing was conducted in accordance with the Chinese standard GB/T 50081, whereas carbonation curing and water-sealed curing followed the procedures reported by Liu et al (Liu et al., 2024). These curing regimes were selected to separate the effects of moisture supply and CO₂ exposure: standard curing served as the moist-air reference, water-sealed curing provided sufficient water for hydration, and carbonation curing was used to evaluate the additional effect of accelerated CO₂ exposure.

Table 4. Mix proportion of modified BC CWBA artificial aggregate

Group	Type	OPC	CWP	BC	Modification method	Curing method
NB	NB-W	10	90	0	None	Water-sealed curing
	NB-S					Standard curing
	NB-C					Carbonation curing
CG	CG-W	10	90	5	None	Water-sealed curing
	CG-S					Standard curing
	CG-C					Carbonation curing
WW	WW-W	10	90	5	Water-washed	Water-sealed curing
	WW-S					Standard curing
	WW-C					Carbonation curing
AM	AM-W	10	90	5	Alkali-modified	Water-sealed curing
	AM-S					Standard curing
	AM-C					Carbonation curing

2.4 Granulation method

Granulation was carried out using a disc granulator. OPC, CWP, and BC were weighed in a bucket according to the mix proportions and roughly mixed by hand. The mixture was then poured into a disc granulator (Liu et al., 2023a). After the raw materials were uniformly mixed in the disc of the granulator, about 20% of the mixture was taken out and set aside. Subsequently, water was continuously sprayed into the rotating mixture using a spray bottle until the powders agglomerated into balls. The amount of water used was approximately 14 wt.% of the total dry mass of the raw materials. The previously set-aside mixture was then added to shape the aggregates, and the rotation continued for another 10 min. The aggregates were then removed and placed in a standard curing room. For the water-sealed curing group, the aggregates were randomly sampled after 1 day and placed in sealed water bags for curing.

2.5 Experimental methods

2.5.1 Physical properties tests

The physical properties were measured in accordance with GB/T 17431.2-2010 (China Building Materials Federation, 2010). After 28 days of curing, specimens falling in the 10-15 mm size range were taken and oven-dried at 65 °C to constant mass. The dry weight of each specimen was recorded as m_l . They were then submerged in water inside a measuring cylinder. Following 1 hour of immersion, the specimens were removed and gently wiped with a damp towel until their surfaces appeared dry. Each specimen was subsequently transferred to the graduated cylinder containing a certain amount of clean water. By recording the water level rise, the specimen volume V_t was calculated. The density was then determined using **Eq. (1)**:

$$\rho_{ap} = \frac{m_l \times 1000}{V_t - 500} \quad (1)$$

Using the above method, specimens were taken and dried. The specimens were then immersed for 24 h. After immersion, these specimens were brought to a saturated surface-dry condition following the above

method, weighed, and recorded as m_0 . The water absorption rate was then determined using **Eq. (2)**:

$$\omega_a = \frac{m_0 - m_1}{m_1} \times 100 \quad (2)$$

The apparent density and water absorption tests were repeated twice and three times, respectively.

2.5.2 Single particle compressive strength test

The single particle compressive strength of the aggregates was tested using a universal testing machine (model ETM305D) after they had been cured to the specified ages (Liu et al., 2022). Aggregates with a particle size ranging from 10 to 15 mm were selected. The peak load at failure P and the diameter d of each aggregate were recorded. The compressive strength of individual particles was subsequently determined by means of **Eq. (3)** (Liu et al., 2023b). The single-particle compressive strength test was repeated six times for each group and curing age.

$$\sigma = \frac{2.8P}{\pi d^2} \quad (3)$$

2.5.3 CO₂ uptake test

The self-made carbonation box (Liu et al., 2024) was used in the test. The effective gas volume was 25 L, and the test was performed at 293.15 K and approximately 75% RH, with an initial CO₂ concentration of approximately 10.7%. The chamber was operated under static conditions without continuous gas flow, and the calculated CO₂ partial pressure was 11.61 kPa. For the CO₂ uptake test, the aggregates were first placed in an open container, which was then transferred into the chamber. Three beakers were also placed in the chamber, containing (i) a saturated NaCl solution, (ii) deionized water, and (iii) 20 g of solid citric acid and 10 g of solid sodium bicarbonate as the CO₂ source, respectively. The CO₂ concentration in the cabin was monitored by a gas detector, which was recorded every 1 min for 72 h. An empty-chamber test was performed under identical conditions to correct for CO₂ loss caused by chamber leakage, dissolution, and background

variation. And the CO₂ uptake was calculated according to Eqs (4)-(7). To ensure data reliability, each test was repeated twice. Meanwhile, the aggregates were broken after curing, and a 1% phenolphthalein solution was sprayed onto the cross-sections. The pink regions were considered uncarbonated, whereas the colorless regions were considered carbonated.

$$\Delta = \frac{n_{CO_2}}{n_{air}} \quad (4)$$

$$n_{CO_2} = (n_{out} + n_{CO_2}) \times \Delta \quad (5)$$

$$n_{CO_2} = \frac{(n_{out} \times \Delta)}{(1 - \Delta)} \quad (6)$$

$$m_{CO_2} = n_{CO_2} \times 44 \quad (7)$$

where n_{CO_2} is the amount of absorbed CO₂ (mol), n_{OUT} is the amount of substances in the air excluding CO₂ (mol), Δ is the difference in CO₂ concentration between the empty-chamber and experimental groups, and m_{CO_2} is the mass of absorbed CO₂ (g).

2.5.4 XRD test

After the compressive test, the aggregate samples were submerged in anhydrous ethanol in order to terminate hydration, then ground until they passed through a 75 μ m sieve, and the mineral composition of the samples was analyzed using an X-ray diffractometer (D8 Advance). The samples were scanned at angles from 15° to 55°, with a scanning time of 0.2 s per point (Li et al., 2023).

2.5.5 SEM test

After curing for 28 days, the raw materials or samples were fractured, and fragments with a long axis of 3-4 mm were picked out. These fragments were submerged in anhydrous ethanol for 48 hours. Subsequently, they were oven-dried. Before observation, a gold coating was applied to the samples to enhance their conductivity. Finally, Quanta TM 250 FEG was employed to obtain SEM images (Zhang et al., 2022).

3. Results and discussion

3.1 CO₂ uptake

The CO₂ uptake test results of CWBA are shown in **Fig. 3**. **Fig. 3(a)-(d)** present the CO₂ uptake results of groups NB, CG, WW, and AM over time, respectively. Before 1 h, due to uneven gas mixing, the concentration values detected by the detector fluctuated considerably. Therefore, only the test data from 1-72 h were used.

From **Fig. 3(a)-(d)**, it can be observed that, except for the CG group, the carbonation rate at 7 days in the early stage or throughout the whole test period was higher than that at 3 days for each group. Notably, for the NB group, except at 3 days, the slope of the curve decreased significantly after 32 hours at all other ages. The carbonation rate curve can be divided into surface carbonation and internal carbonation. The surface carbonation rate is related to the content of hydration products (Ca(OH)₂) and largely depends on the surface area, whereas the internal carbonation rate is mainly related to porosity. Surface carbonation is governed mainly by the exposed surface area, whereas carbonation within the aggregate depends on CO₂ transport through the pore network and the volume of accessible carbonatable phases. In terms of total CO₂ uptake, internal carbonation accounts for the majority. Regarding the carbonation rate, since surface carbonation is much less difficult than internal carbonation, surface carbonation plays a dominant role in the early stage of carbonation. At early ages, due to incomplete hydration, the Ca(OH)₂ content generated by hydration is relatively low. Therefore, the surface Ca(OH)₂ content at 7 days is higher than that at 3 days, resulting in a steeper curve slope in the early carbonation stage. The higher the internal porosity, the higher the slope at later stages.

The main component of the aggregates is CWP. After crushing and sieving, the surface of the powder particles is relatively stable, making carbonation difficult. In contrast, the added clinker has a shorter

254 hydration age and higher porosity compared to CWP, and is therefore more easily carbonated. Meanwhile,
255 the amount of clinker added is much lower than that of CWP, so the limit of carbonation depends on the
256 carbonation amount of CWP, while the early carbonation rate depends on the carbonation amount of the
257 clinker hydration products.

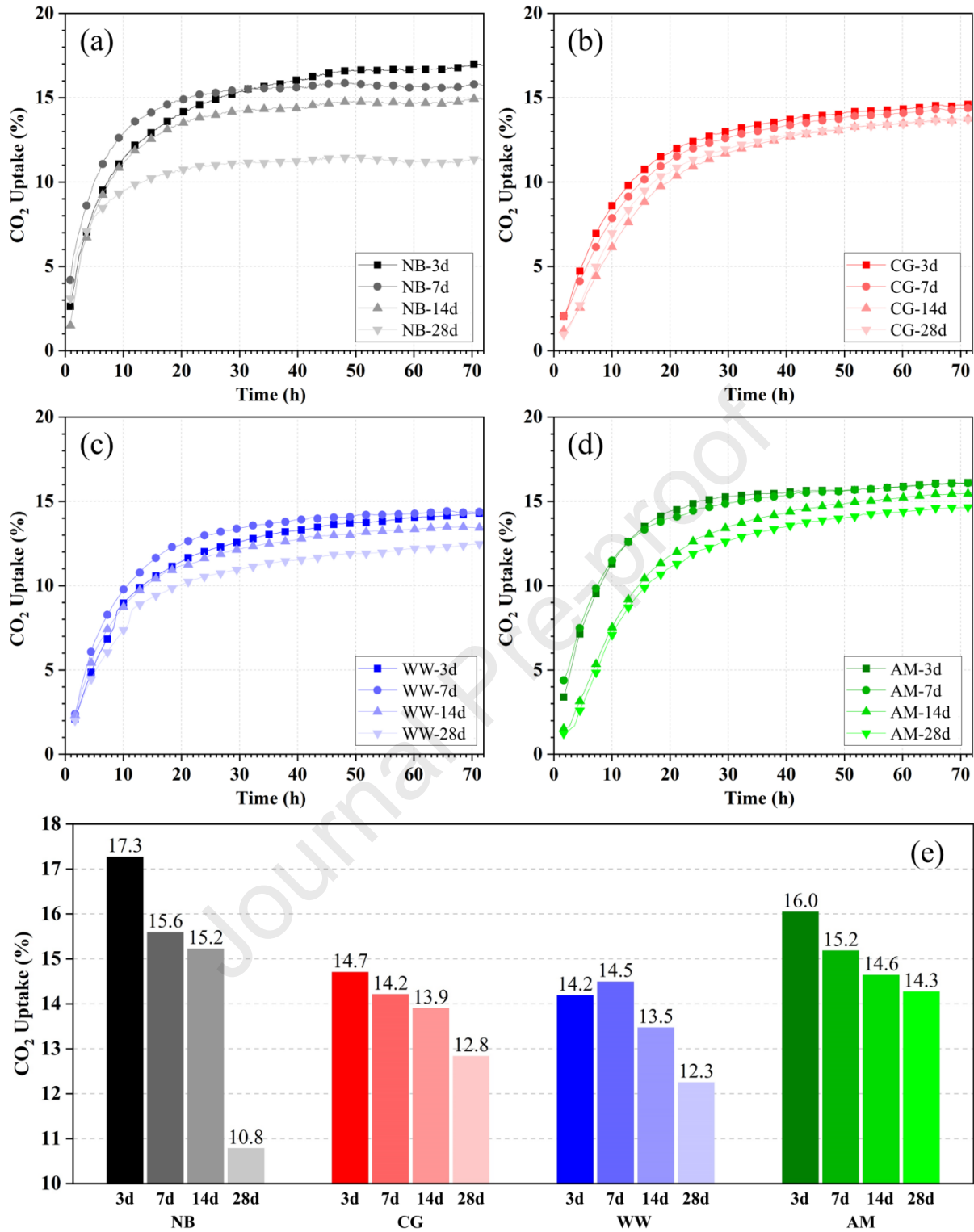
Fig. 3. CO₂ uptake test results

Fig. 3(e) shows the total CO₂ uptake rate of each group. Overall, with increasing age, the CO₂ uptake rate of each group showed a decreasing trend. Compared to the NB group, the groups with BC added showed a smaller decline. At short ages (3 d, 7 d) and long ages (28 d), the change in CO₂ uptake was not significant;

however, the NB group exhibited a substantial drop in CO₂ uptake at 28 d. This is because the CO₂ transport channels provided by BC may offset part of the negative effect. At early ages (≤ 14 d), the groups with BC added had a generally lower CO₂ uptake rate, which is consistent with the result in (Liu et al., 2024). The addition of a small amount of BC promoted the hydration of OPC particles, thereby reducing carbonation efficiency. Previous studies have shown that BC porosity can improve CO₂ adsorption and facilitate its transport in cementitious materials (Chen et al., 2025; Mesnage et al., 2025). The higher CO₂ uptake rates of the WW and AM groups were due to the modified BC being more favorable for CO₂ transport, thus improving carbonation efficiency. Specifically, from 14 d to 28 d, the CO₂ uptake of the NB group decreased by 41.19%, while the CG and WW groups decreased by 8.27% and 9.96%, respectively, and the AM group decreased by only 2.60%. This indicates that BC plays a promoting role in carbonation.

Fig. 4 shows the phenolphthalein color development of the aggregates. The last column labeled “W” represents the specimens after 28 days of water-sealed curing. All these specimens remained pink, indicating that no obvious carbonation occurred and that the sealing conditions were effective. Large pink regions were also observed in the 3-day carbonation-cured specimens, indicating that most of the cross-section remained uncarbonated. With increasing age, a colorless outer layer and a pink inner region gradually became visible in the carbonation-cured specimens, showing that carbonation mainly developed from the surface toward the interior. Meanwhile, the critical pH for phenolphthalein coloration is 11.00, which requires a Ca(OH)₂ content of 0.037 g/L. Even in a saturated CaCO₃ solution prepared in the laboratory (pH = 10.58), the addition of phenolphthalein also shows a light pink color. Therefore, the phenolphthalein coloration range can indicate the presence of trace amounts of Ca(OH)₂, but the intensity of the color cannot be used as a criterion for differences in Ca(OH)₂ content.



Fig. 4. Phenolphthalein color development results of CWBA

As shown in Fig. 5, from a local microscopic perspective, at early ages, the carbonated particle surfaces form a dense CaCO_3 layer, which makes it more difficult for the internal minerals to contact CO_2 . As CaCO_3 accumulates in the more extensively carbonated regions, local transport resistance increases, causing CO_2 to preferentially migrate through the remaining permeable pathways toward the aggregate interior. It can also be inferred that at this stage, the CaCO_3 formed by carbonation is insufficient to fill the matrix pores. CO_2 molecules can more easily diffuse into the interior, which on one hand increases CO_2 uptake, and on the other hand raises the degree of carbonation. At early ages, the relatively porous matrix allowed CO_2 to penetrate into the aggregates, resulting in an indistinct carbonation boundary. At later ages, continued hydration and CaCO_3 formation densified the matrix and restricted further CO_2 penetration, producing a clearer boundary

between the colorless carbonated outer layer and the pink uncarbonated inner region. At the same age, the carbonation boundary of the BC-containing groups was less distinct than that of the NB group because the pore structure of BC provided additional pathways for CO₂ transport into the aggregate interior. Consequently, carbonation was less concentrated near the surface, thereby delaying the appearance of a distinct surface carbonation layer in the BC-containing groups.

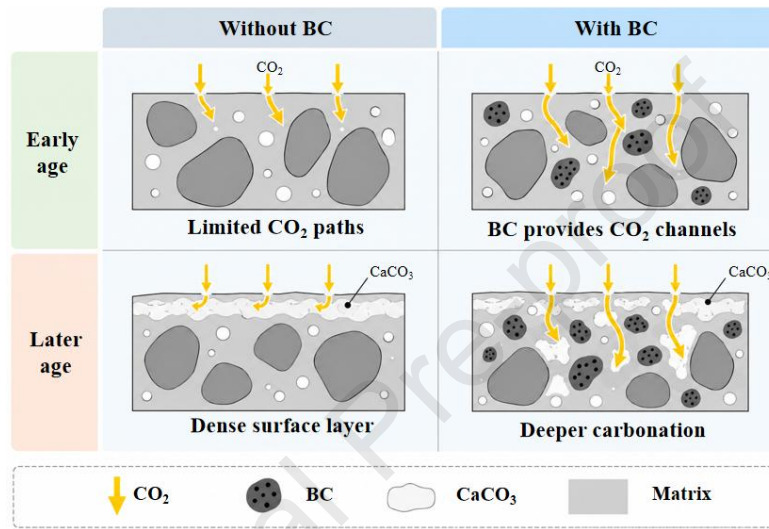


Fig. 5. BC carbonation synergistic mechanism

3.2 Physical properties

As shown in **Fig. 6**, except for the CG-W group, the aggregate densities were primarily distributed within 1400-1650 kg/m³, classifying them as lightweight aggregates, and the water absorption rates ranged from 11.89% to 26.33%. Among the different curing methods, the density increased in the order of water-sealed curing, standard curing, and carbonation curing (W > S > C), while the water absorption rate decreased in the same order, indicating that carbonation increases aggregate density. In terms of the density increase from W to S treatment, the NB group showed the smallest increase (2.20%), while the CG group exhibited the largest increase (17.03%). This is because the NB group, without BC addition, had low carbonation efficiency in the natural environment, whereas the CG group, with BC, effectively improved carbonation efficiency. The remaining two groups showed increases of about 4%. Regarding the density increase from S

to C treatment, the CG group showed the smallest increase (1.46%), while the AM group showed the largest increase (7.75%). The NB group showed an increase of 6.20%. This is because the degree of carbonation of the CG group in the natural environment was close to its limit under accelerated carbonation, making further carbonation difficult. In contrast, the AM group could still undergo deeper carbonation under accelerated conditions, indicating that its carbonation efficiency is higher than that of the CG group, demonstrating that AM has a higher carbonation capacity. The high increase in the NB group is because it is difficult for it to absorb and fix CO_2 from the air under natural carbonation conditions, whereas under accelerated carbonation conditions it can effectively fix CO_2 from the air.

It is worth noting that the AM-C group had the highest density. However, considering that compared to the NB-C group, the AM-C group contained BC with higher porosity and lower density, the higher density of the AM-C group relative to the NB-C group can be attributed to the formation of CaCO_3 beyond that shown in the figure, which filled this density deficit.

Regarding water absorption rate, a decreasing trend was observed from W-S-C. In terms of the reduction in water absorption from W to S treatment, the NB group showed the largest decrease (17.24%), while the WW group showed the smallest decrease (7.15%). This is because during sealed curing, the WW group contained BC, which can serve as hydration sites and improve the degree of hydration, making the internal structure denser. Regarding the reduction in water absorption from S to C curing, the CG group exhibited the largest decrease (33.69%). This may be due to the hydrophobic nature of the tar layer on CG itself (Parthasarathy et al., 2021), which makes it difficult for water to enter the interior of the aggregates. On the other hand, the CaCO_3 formed from the carbonation of minerals in CG blocked the pore channels of BC, further increasing the difficulty of water diffusion.

In contrast, for AM and WW, due to being washed, their tar content was lower, allowing water to enter

the interior of the aggregates through BC. Since the surface of BC can provide nucleation sites for cement hydration products (Jeon et al., 2025), tar-derived residues remaining after bamboo pyrolysis may affect its accessible surface area and interaction with the cement paste (Parthasarathy et al., 2021). This may partly explain the lower density of the CG-W group, although this mechanism was not directly verified.

BC may facilitate local water transport through its pore network, but this contribution becomes limited once hydration or carbonation densifies the surrounding matrix and restricts water movement to and from the BC particles. However, once the internal matrix becomes dense due to hydration or carbonation, BC cannot absorb a large amount of water, and the water is hindered both before contacting BC and after passing through BC. Therefore, the addition of BC does not significantly change the water absorption rate. Furthermore, any increase in water absorption may be attributed to incomplete hydration in some areas, leading to matrix porosity. The pore volume within BC is smaller compared to the pore volume of the matrix, so a small dosage of BC does not significantly affect the water absorption rate. Rather, the main effect of BC is to promote hydration, which densifies the matrix and reduces the water absorption rate.

It was found that using modified BC to prepare CWBA can indirectly improve the performance of CWBA, especially in terms of CO₂ uptake efficiency. Carbonation is crucial for enhancing the performance of CWBA, and the degree of carbonation can be reflected in the various performance improvements of CWBA. Meanwhile, the interaction between BC and CWBA is also manifested in the substances contained within. For example, after unmodified BC is used to prepare CWBA, due to its high tar content and the hydrophobic nature of the tar, BC's internal curing effect may be reduced.

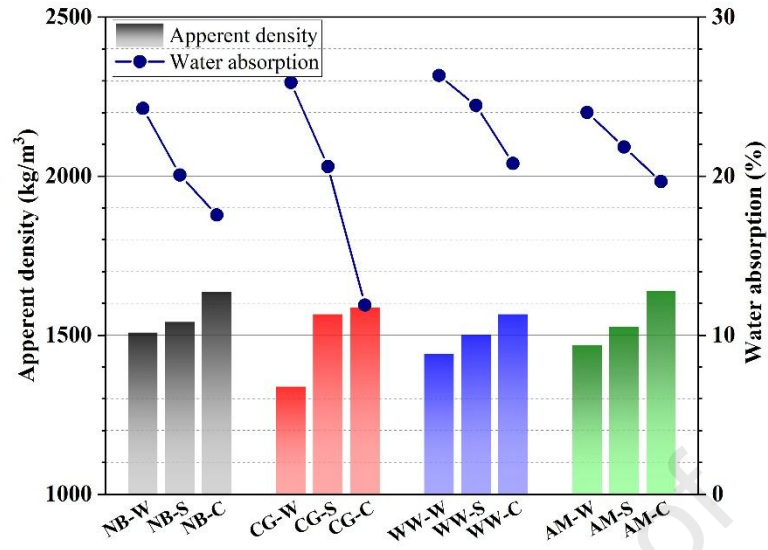


Fig. 6. Physical properties test results

3.3 Mechanical property

Fig. 7 shows the single particle crushing strength of CWBA. Except the C curing group, the strength of all other groups increased with increasing age. In the groups of NB, the standard curing strength at 3 days was higher than that of water-sealed curing. Thereafter, the strength development of the water-sealed curing group exceeded that of the standard curing group, and the strength surpassed that of the standard curing group at 28 days. This is because the main component of the aggregates is CWP. Compared to OPC, CWP is an inert particle with water absorption capacity, which absorbs part of the water during granulation, however, it contributes little to strength development because of its limited pozzolanic activity. The strength of the aggregates is mainly influenced by the hydration products formed from the hydration of clinker, which bind the particles together.

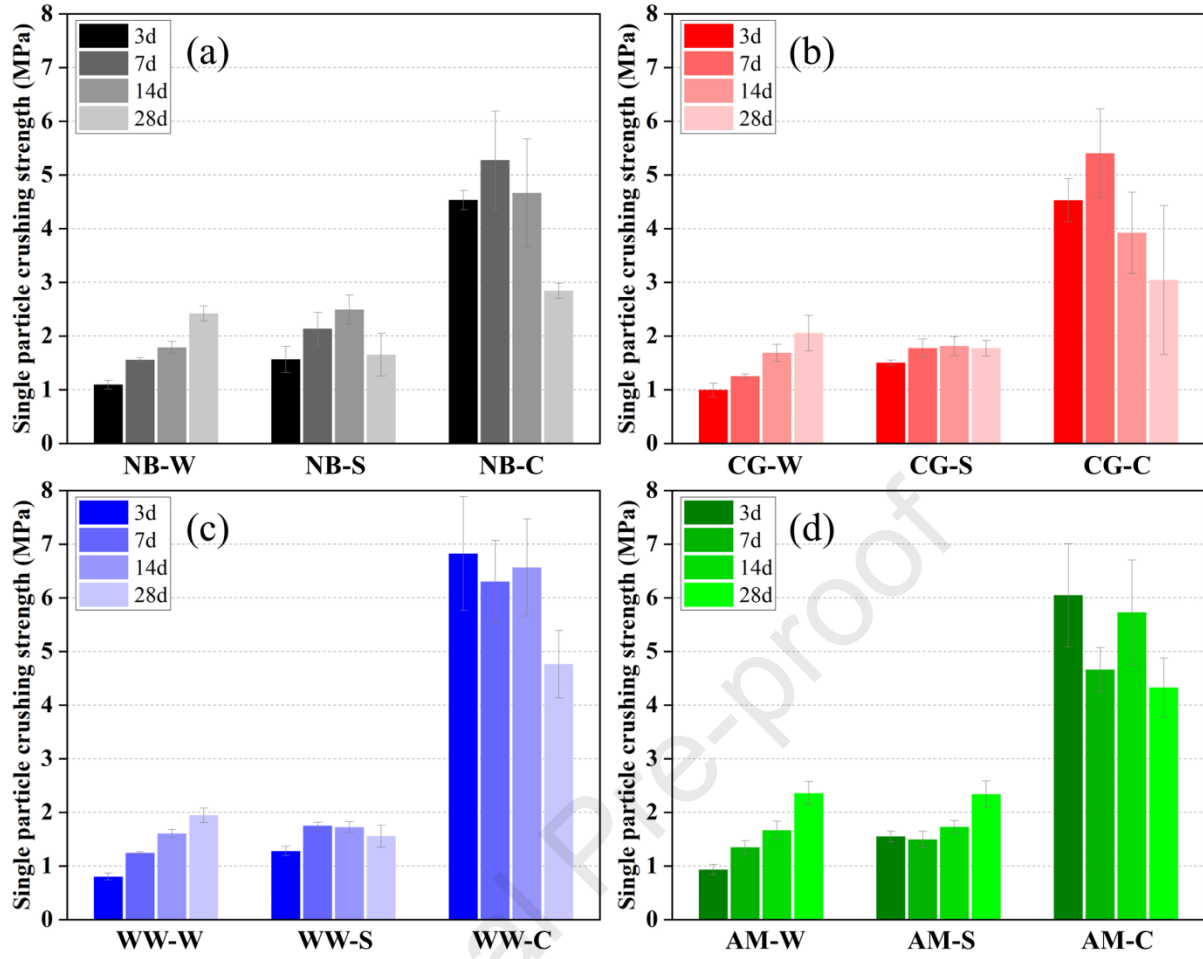


Fig. 7. Single particle compression test results

During granulation, the BC may absorb part of the added water. As hydration proceeds and the surrounding matrix dries, part of the stored water may become available to the adjacent cement particles, providing a possible internal-curing effect (N. Song et al., 2023; Wang et al., 2025). Within the first 24 h after granulation, all curing groups were not yet distinguished and were placed together; only after 24 h were the different curing groups placed in their respective curing environments. During this period, part of the clinker inside the aggregates began to hydrate, binding the components together. Under water-sealed curing conditions, the interior of the aggregates was filled with water, which dissolved part of the CWP particles, forming hydration products such as $\text{Ca}(\text{OH})_2$. The dissolved ions saturated the environment, and according to Le Chatelier's principle, the equilibrium shifted in a direction unfavorable to clinker hydration. At the same time, the dissolution of the CWP particle surfaces made it difficult for crystallization sites to be fixed on the

CWP surfaces, resulting in poor adhesion between particles. These factors together led to poor early-age strength development in the water-sealed curing group. As the age increased, under water-sealed conditions, the clinker became fully hydrated, binding the particles together, and the strength developed significantly. It should be noted that the clinker hydration rate develops slowly under high Ca^{2+} concentration, instead of stalling. In the standard curing group, the hydration rate was faster at early ages, and the CWP particles hardly dissolved, which both reduced the Ca^{2+} concentration in the environment and provided a rough surface more suitable for crystal attachment, thus achieving higher strength. Later, because water vapor in the air could not effectively reach the internal unhydrated particles in time to participate in hydration, some clinker remained unhydrated, leading to slower strength development at later ages.

In the water-sealed curing group, the strengths of NB-W and AM-W were relatively high, reaching 2.42 MPa and 2.36 MPa, respectively, which were slightly higher than those of CG-W (2.05 MPa) and WW-W (1.95 MPa). In the standard curing group, AM-S significantly outperformed NB-S, CG-S, and WW-S, with values of 2.34 MPa versus 1.65 MPa, 1.77 MPa, and 1.55 MPa, respectively. It can be observed that the AM group exhibited superior strength performance among the non-carbonated groups. It can also be considered that non-alkali-modified BC at a dosage of 5% has no significant effect on aggregate strength. The addition of BC to the aggregates may promote the hydration degree of OPC particles in two ways: at early ages, it may provide additional hydration sites; at later ages, it may act through internal curing. At the same time, BC improves carbonation efficiency. However, BC also has higher porosity and lower strength compared to the matrix, forming weak points inside the aggregates. These two effects are antagonistic, with one dominating depending on the dosage. In this study, the AM group at a dosage of 5% showed a more pronounced effect in enhancing hydration and carbonation, indicating that alkali modification can improve the performance of BC in aggregates. The better performance of the AM group may be related to the removal of tar from the BC

surface during alkali treatment. Removing the tar exposed more pores and improved the hydrophilicity of BC, which favored cement hydration. The exposed pores also provided more pathways for CO₂ transport. In contrast, the tar remaining on unmodified BC blocked some pores and hindered the transport of water and CO₂. A lower BC dosage might lead to greater strength improvement but could result in poorer CO₂ uptake performance (Liu et al., 2024). In the analysis of the carbonated groups, it is evident that BC has a marked influence on aggregate performance.

Fig. 8 shows the effect of BC and carbonation on the improvement of compressive strength. The corresponding water-sealed group at the same pre-curing age was used as the uncarbonated reference for calculating the percentage increase. Carbonation treatment can significantly increase the aggregates' strength, with an increase ranging from 117%-851%. Carbonation transforms calcium-containing minerals inside the aggregates into CaCO₃, which is stronger and denser, thereby improving aggregate strength. At early ages (<14 d), the strengthening effect is more pronounced, with an average strength increase of about 500%; at 28 days, the average increase decreases to 173.4%. As the curing age increases, the hydration degree inside the aggregates becomes higher, the aggregates become denser, and CO₂ diffusion into the interior becomes more difficult, leading to a lower degree of carbonation. Notably, the strength increase of the NB group decreased from 414% to 117.5% with increasing age, whereas the increase of the AM group ranged from 648.7%-183.4%, far exceeding that of the NB group. Combined with the CO₂ uptake performance results, it can be seen that at early ages, there was no significant difference in CO₂ uptake between the AM and NB groups before 28 days, with the AM group only showing a significant advantage at 28 days. The difference in strength increase is mainly attributed to the random distribution of BC within the aggregates. During carbonation, CO₂ molecules move randomly. In regions without BC, the carbonation morphology of the aggregates is similar; in regions containing BC, CO₂ molecules move rapidly through the pores inside BC and then diffuse,

forming irregular carbonated zones. A large number of irregular carbonated zones effectively enhance the interlocking effect, allowing the carbonated layer and the interior to bear loads synergistically. In contrast, in aggregates without BC, the carbonation boundary is relatively smooth and continuous, and when subjected to pressure, internal forces are transmitted along the boundary, making them more prone to failure.

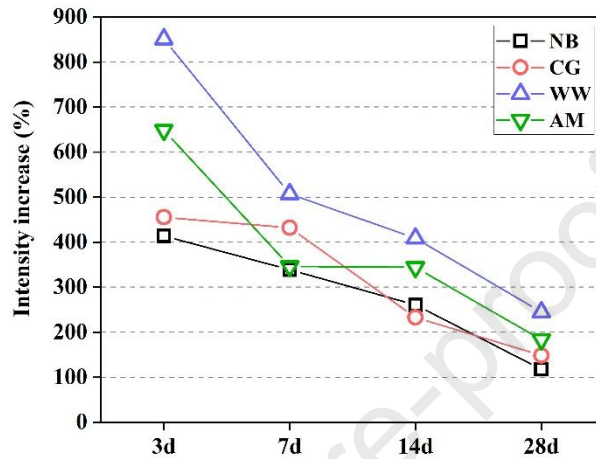


Fig. 8. Improvement effect of compressive strength

3.4 XRD

The XRD test results are shown in Fig. 9, with corundum used as an internal standard at a dosage of 20%. It can be seen that the 2θ angles corresponding to portlandite are 18° , 28.62° , 34.02° , and 47.04° . The 2θ angles corresponding to calcite are 29.36° , 39.44° , 42.82° , and 47.5° . The 2θ angles corresponding to quartz are 20.84° , 26.58° , and 50.14° . The 2θ angles corresponding to corundum are 25.58° , 35.15° , 43.35° , and 52.55° . The 2θ angle corresponding to aragonite is 26.18° .

From the qualitative analysis, the addition of BC has no effect on the types of minerals, indicating that BC is an inert material and does not participate in the OPC hydration reaction. In the carbonation curing group, the peak intensity of portlandite almost disappeared before 14 days compared to the water-sealed and standard curing groups, indicating that carbonation curing can consume a large amount of portlandite.

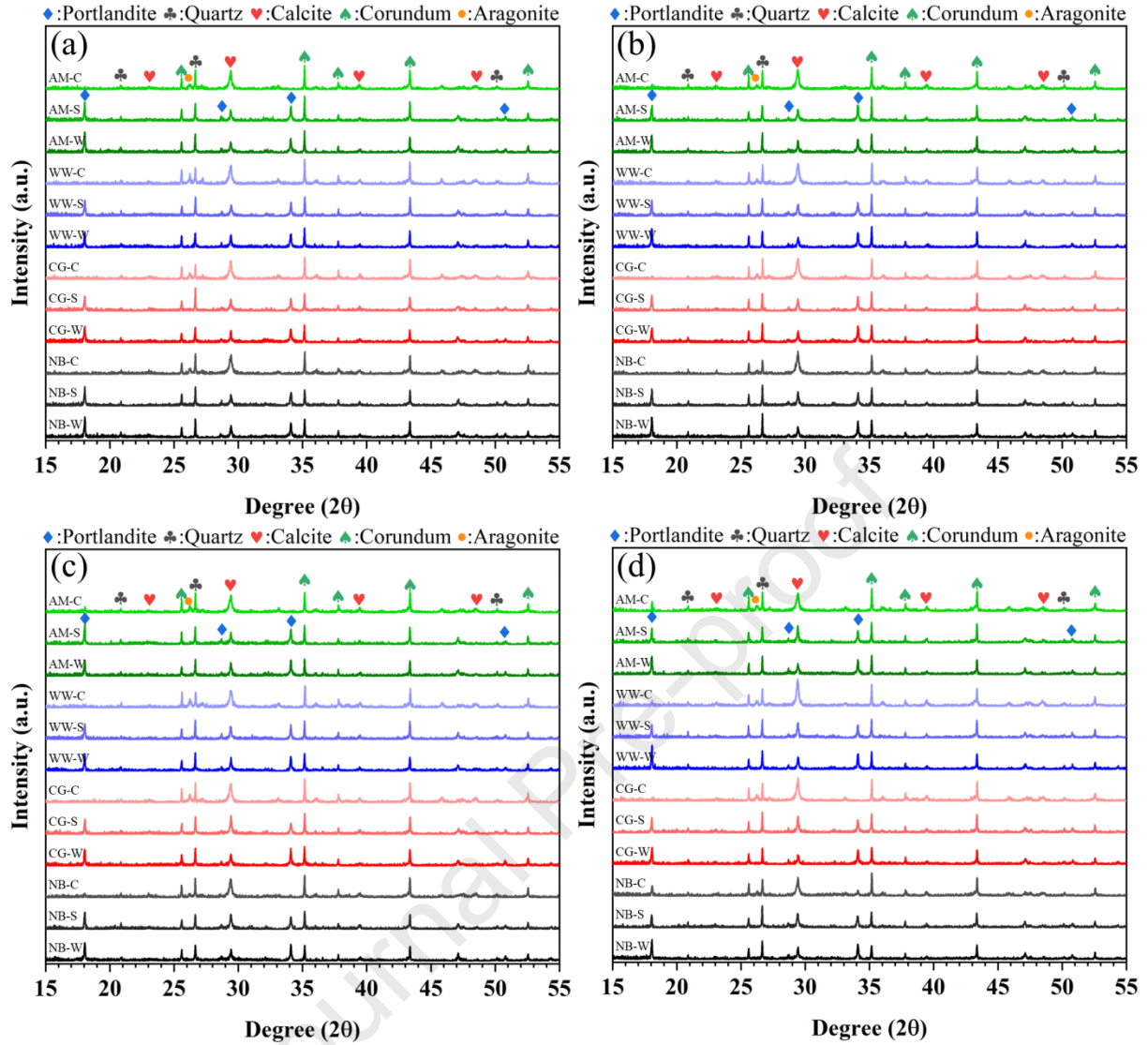
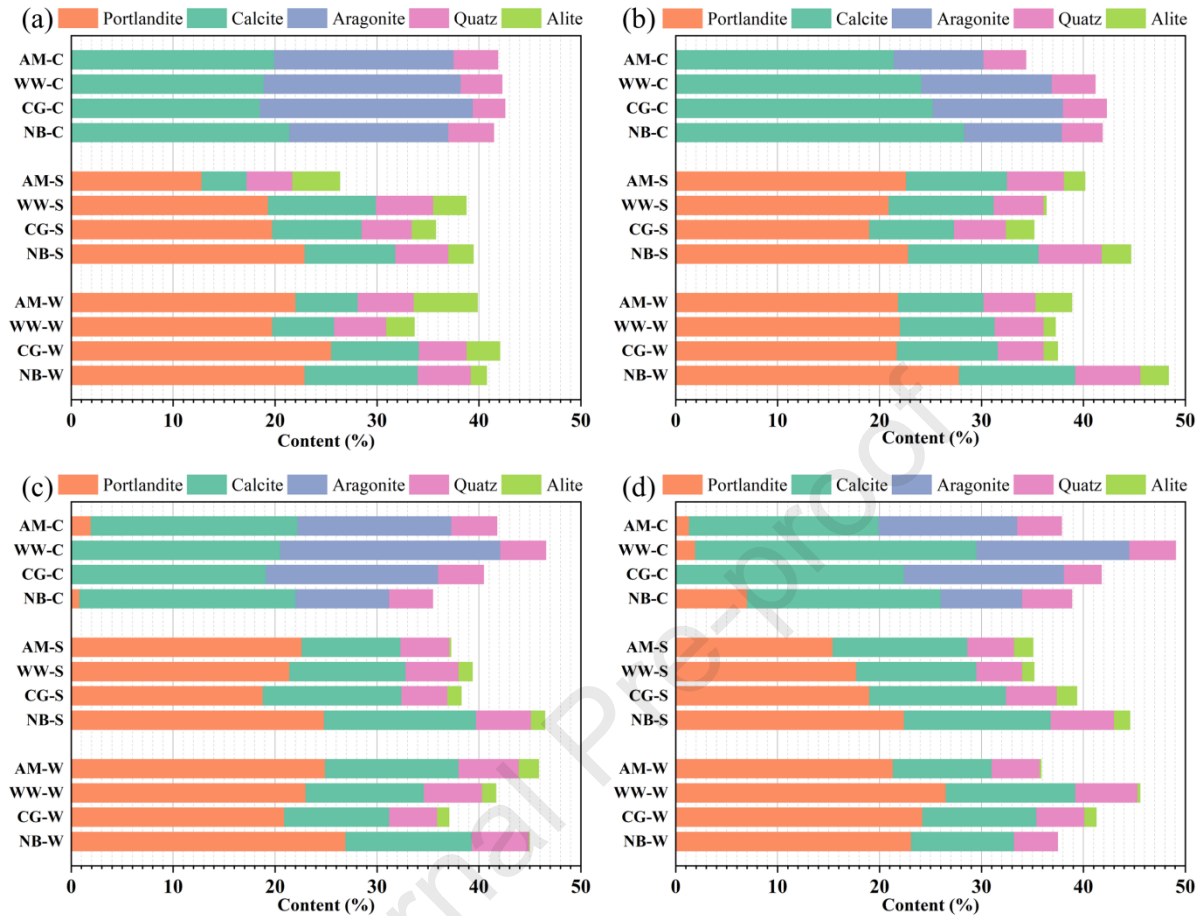


Fig. 9. XRD results of (a) 3 d; (b) 7 d; (c) 14 d; and (d) 28 d.

The quantitative results are shown in Fig. 10. The chemical composition of both calcite and aragonite is CaCO_3 . In the water-sealed and standard curing groups, the aragonite mineral did not appear. Unlike the common trigonal crystal form of calcite, aragonite has an orthorhombic crystal form. Therefore, it can be considered to be formed under carbonation curing. With increasing age, the proportion of portlandite increases and the content of alite decreases. Fig. 10(a) shows the quantitative analysis results of the NB group. It can be found that, except for the C group, the content of portlandite varied within a small range (less than 3%), and the calcite content in the S group did not change significantly over time. It can be inferred that the NB groups' hydration had largely concluded during the early period, and carbonation development was not

447 obvious. Notably, for the NB-C group, portlandite could still be detected at 28 days.



448
449 **Fig. 10. Crystal content of (a) 3 d; (b) 7 d; (c) 14 d; and (d) 28 d.**

450 3.5 SEM

451 **Fig. 11** presents the SEM test results. It can be observed that the crystallinity of the groups with added
452 BC is better than that of the NB group. On one hand, this is because BC can promote the hydration of clinker.
453 On the other hand, it may be because the CWP particles are hydration products of OPC, the main products
454 of which include Ca(OH)_2 , C-S-H gel, AFt, AFm, and so on. From the SEM images of the raw materials, it
455 can be seen that there are no obvious single crystals on the surface. Among these products, Ca(OH)_2 has a
456 relatively high content and is relatively soluble. During granulation, after CWP dissolves, Ca(OH)_2 crystals
457 can reform, but the dissolved amount is very low, and recrystallization occurs during subsequent curing. At
458 the same time, a large number of crystal seeds remain on the surface after dissolution, which serve as good

nucleation sites. Due to the low clinker content, the ion concentration in the environment is relatively low, which is clearly insufficient to form large crystals at every location. From the principle of minimum energy (entropy increase theorem), ions are more inclined to combine with crystal seeds rather than concentrate at a single location to form distinct crystals. As a result, a large amount of Ca^{2+} combines with numerous crystal seeds, forming poorly crystalline $\text{Ca}(\text{OH})_2$ crystals in various places.

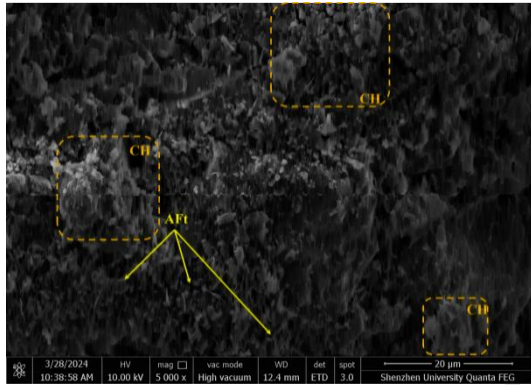
The crystallinity of the standard curing group is better than that of the water-sealed curing group because crystallization requires a high-concentration environment, whereas the environment provided by the water-sealed group reduces the ion concentration inside the aggregates. Therefore, the standard curing group exhibits better crystal morphology. On the other hand, as mentioned earlier, due to Le Chatelier's principle, clinker hydration is hindered by CWP particles, and this effect is more pronounced in the water-sealed group. Inside the aggregates, crystals are mainly contributed by clinker hydration, and the degree of clinker hydration is reflected in the extent of crystal growth. Crystal growth requires sufficient space and generates crystallization pressure during formation. The aggregates have relatively high porosity, providing relatively free space for crystal growth. However, between CWP particles, the high stiffness of the CWP particles makes it difficult to release crystallization stress, thereby inhibiting crystal growth. In contrast, BC has lower stiffness, allowing better release of crystallization stress during crystal growth, so crystals grow more freely. Consequently, more crystals appear near BC. Meanwhile, during granulation, the oxygen-containing functional groups on the BC surface may undergo deprotonation, resulting in a negatively charged surface (Chambers et al., 2025). Positively charged cations may therefore preferentially adsorb onto the BC surface, promoting the formation of initial nucleation sites for hydration products (Berti et al., 2021) and resulting in abundant crystals around BC, although the detailed process remains complex. The surface of CWP particles also has numerous nucleation sites. However, on the BC surface, due to charge balance and electrostatic

repulsion, the number of nucleation sites within a given area is limited. Moreover, the BC surface is smoother than that of CWP, so the number of nucleation sites on BC is much lower than on CWP, but the crystallinity is better than that on CWP. During hydration, crystals grow and gradually envelop BC, embedding BC into the matrix. During curing, the internal curing effect allows water to move freely and be released at various pores, enabling nearby clinker to continue hydrating and crystallizing. The boundary of BC is observed to be relatively clear, indicating that BC is chemically stable and does not react with OPC particles. It can also be observed that hydration products are inversely distributed with distance from BC, meaning that the farther away from BC, the fewer hydration products are present.

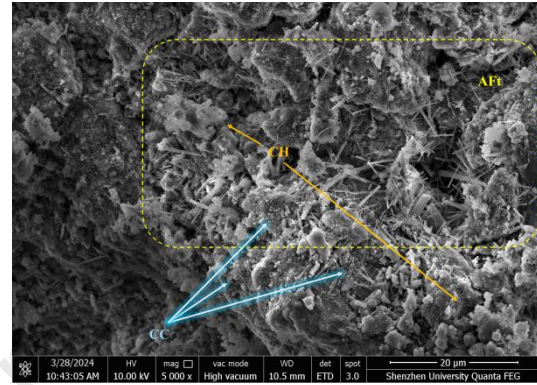
Modifying BC causes the minerals on its surface to be washed away and dissolved, making the surface appear more “smooth” in the raw material SEM images. For OPC particles, this may make it more difficult to attach to the BC surface. However, at the same time, modification removes the oil film on BC, increases its hydrophilicity, enhances nucleation sites and internal curing, and allows a spatial network structure to form around BC. This explains why the crystallinity of the AM group is better than that of the CG and WW groups. Finally, the WW group shows the poorest crystallinity, likely because washing reduces the number of surface minerals while some oil film remains, resulting in poor hydrophilicity. The combined negative effects lead to inferior crystallinity.

In the carbonation treatment groups, it can be observed that the NB-C group has a large amount of CaCO_3 on the surface, indicating that carbonation transformed the other minerals into CaCO_3 . The surface particles are relatively small and show no obvious crystal structure. This is because the sample was carbonated from NB-W, and the crystal structure inherited that of NB-W. In the groups with added BC, distinct hydration products can be seen, closely surrounding BC. This is also because they were obtained from water-sealed curing. The difference is that a large amount of C-S-H gel structure can be observed,

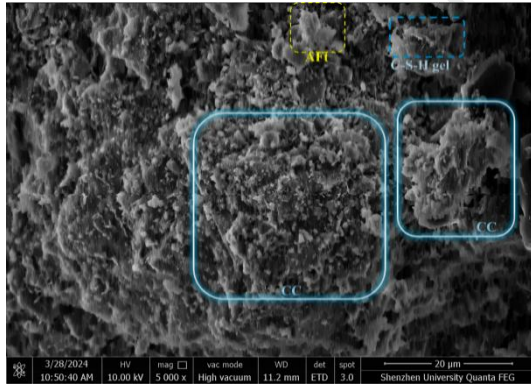
503 indicating thorough hydration. Since C-S-H gel is generated only by clinker hydration, and CWP contains a
 504 large amount of carbonatable components, the carbonation priority of $\text{Ca}(\text{OH})_2$ is higher than that of other
 505 minerals, so the C-S-H gel is preserved. The distribution of CaCO_3 in NB-C is relatively uniform, whereas
 506 in the BC-added groups, the distribution of CaCO_3 is more concentrated. This also indicates that BC may
 507 facilitate CO_2 transport.



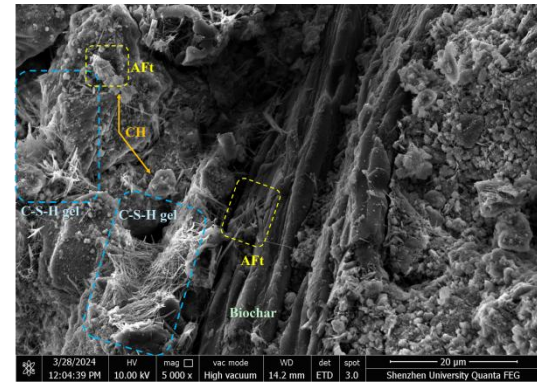
(a)NB-W



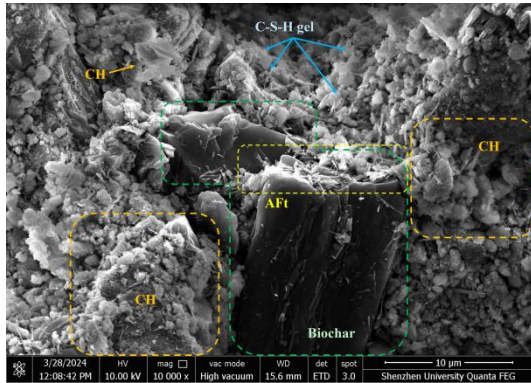
(b)NB-S



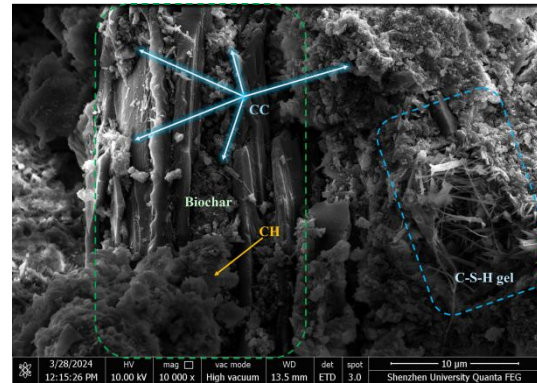
(c)NB-C



(d)CG-W



(e)CG-S



(f)CG-C

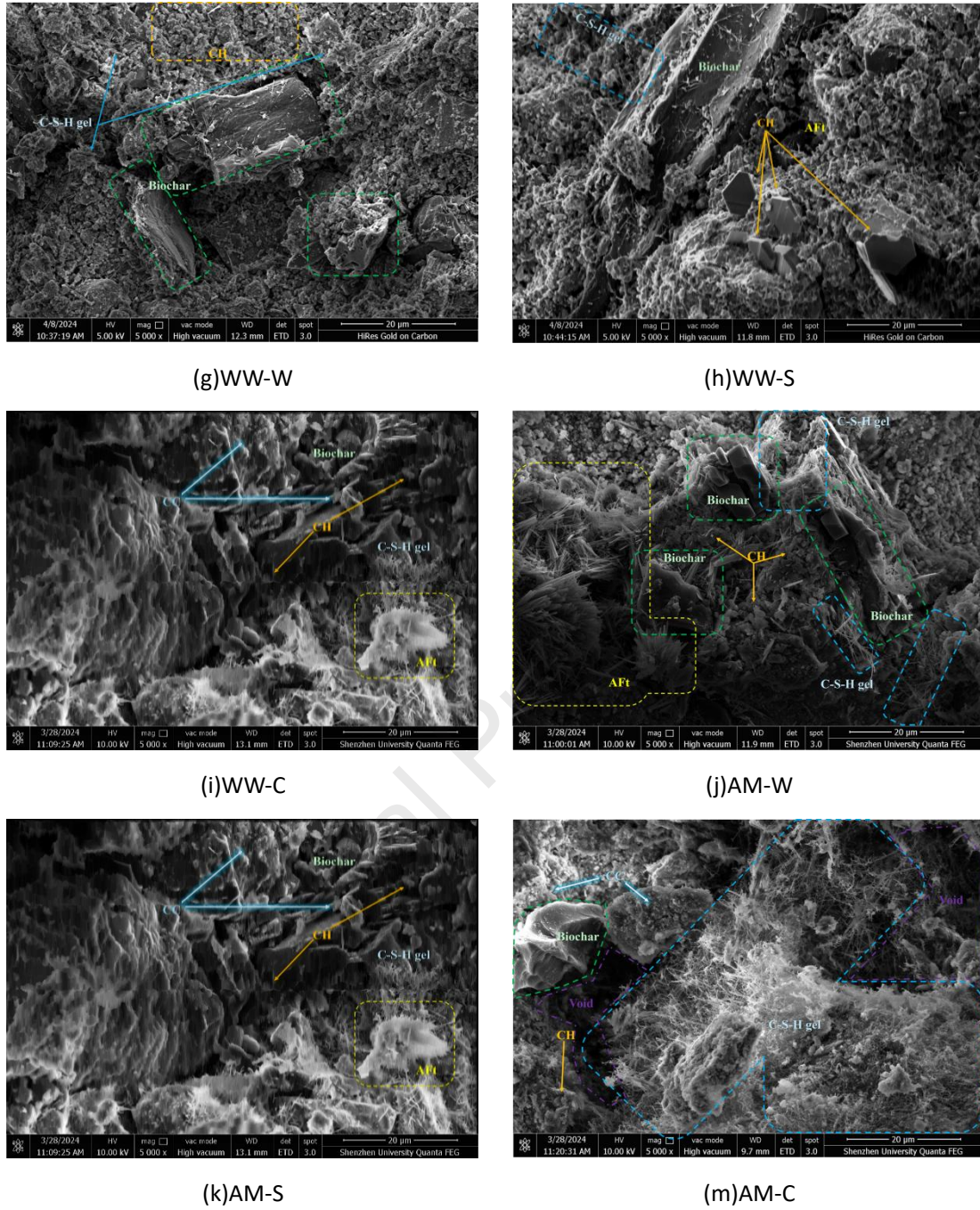


Fig. 11. Microscopic morphology of CWBA

4. Conclusions

In this study, BC modified by different methods was used to prepare CWBA, and the mechanical properties and CO₂ uptake performance were investigated. Based on the excellent pore structure of BC and its CO₂ adsorption capacity, the synergistic mechanism between BC and CWBA was further studied, and the carbonation mechanism and the effects after carbonation were discussed. The conclusions are as follows:

(1) The CO₂ uptake of the artificial aggregates ranged from 8% to 19%, and decreased with increasing age. The NB group showed the most significant decreasing trend, while the AM group exhibited the most gradual decline. This is because BC attracts CO₂ and assists the aggregates in carbon sequestration, offsetting the effect of matrix densification after curing that would otherwise reduce CO₂ uptake.

(2) The density of the artificial aggregates ranged from 1400 - 1650 kg/m³, and the water absorption rate ranged from 11.89% to 26.33%. Among all the tested groups, AM-C and NB-C exhibited the highest aggregate densities. As the amount of CO₂ exposure increased with the curing method, the density increased.

(3) The compressive strength of the artificial aggregates ranged from 0.7-6.8 MPa, and the strength could be increased by up to 851% after carbonation. At 3 days, the WW group showed the greatest carbonation gain, while at 28 days, the AM group exhibited the greatest carbonation gain, and the NB group showed the smallest carbonation gain.

(4) The NB group, without BC addition, exhibited significantly lower CO₂ uptake performance at all ages compared to the groups with BC. Due to the lack of pozzolanically active materials in the mix proportion of the artificial aggregates in this study, the hydration products generated by hydration alone were limited. The main gain in macro-performance came from carbonation. Consequently, after carbonation treatment, the NB group showed the poorest performance.

(5) The performance of the aggregates in this study is highly dependent on the degree of carbonation. The strength gain from surface carbonation is much smaller than that from full-section carbonation.

(6) The tar layer on the surface and in the pores of BC blocked part of its pore structure and reduced its hydrophilicity. Water washing and alkali modification might partially remove this layer and expose more pores, thereby facilitating water and CO₂ transport and promoting cement hydration and carbonation. Alkali modification was more effective in removing the tar layer, resulting in better overall performance of the AM

group than the CG group.

In summary, at an OPC:CWP mass ratio of 10:90 and a BC content of 5%, the use of an appropriate BC modification method improved the performance of CWBA. This approach can promote the utilization of CWP and contribute to China's goal of achieving carbon neutrality. Future research should focus on expanding biochar modification methods to seek more effective and cost-efficient modification approaches for BC.

Conflict of interest

None.

Acknowledgements

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Highlights

- Alkali-modified biochar significantly stabilizes carbon sequestration.
- Alkali-modified biochar shows the largest long-term strength gain.
- Biochar pores serve as efficient CO₂ diffusion pathways.
- Alkali treatment creates abundant stable hydration nucleation sites.
- Modified biochar effectively raises the comprehensive performance.

Declaration of interests

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

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: