

Article

Synergistic Effects of Alkali Activator Dosage on Carbonation Resistance and Microstructural Evolution of Recycled Concrete: Insights from Fractal Analysis and Optimal Threshold Identification

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Abstract: The synergistic mechanism between alkali activation and carbonation in fly ash recycled aggregate concrete (FRAC) remains a critical challenge for enhancing durability and promoting solid waste utilization. This study systematically investigates the effects of CaO-based alkali activator dosages (0%, 4%, 8%, 12%) on carbonation resistance, compressive strength, and pore structure evolution. The results demonstrated 8% CaO maximized compressive strength (48.6 MPa, 10.76% higher than the control group) and minimized porosity (22.87% vs. 39.33% in untreated samples), with enhanced carbonation resistance (35% depth reduction after 28 days). Fractal dimension (FD) analysis revealed that 8% dosage optimized pore complexity (FD > 1.9), forming a dense C–S–H/AFt network that suppressed CO₂ diffusion. CaO addition introduces embodied carbon (9.84 kg CO₂/m³), and the synergy between fly ash’s cement replacement (120 kg CO₂/m³ reduction) and extended service life (theoretically, 15–20 years) ensures a net carbon benefit. These findings establish 8% as a critical threshold for optimizing alkali activation efficiency and durability in low-carbon concrete design. These findings offer theoretical and technical foundations for low-carbon concrete design and sustainable solid waste recycling in construction.

Keywords: recycled aggregate; alkali activation; carbonation resistance; microstructure evolution; fractal dimension of pore structure; solid waste utilization



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1. Introduction

The construction materials industry is transitioning toward a low-carbon and sustainable direction, where the synergistic use of fly ash (FA) and recycled aggregates (RA) is one of the important paths to achieve the “dual-carbon” goal. According to the International Energy Agency (IEA), global carbon emissions from the cement industry account for 7% of total anthropogenic CO₂ emissions, while each cubic meter of concrete production generates an average of 300–400 kg CO₂ [1]. In China, construction waste is projected to exceed 4 billion tons by 2024, yet the comprehensive utilization rate of FA remains at 70%, and the utilization rate of RA is only 38% [2]. There is significant potential for the environmental and economic benefits of solid waste resource utilization. The synergistic use of FA and RA is regarded as a key technology direction to realize the decarbonization of concrete. FA has a pozzolanic effect that can partially replace cement and improve the durability of concrete [3–6], and the application of RA can significantly reduce the consumption of natural

resources and landfills of construction waste [7–9]. However, the synergistic utilization of the two faces significant challenges: the low activity of FA leads to its hydration reaction relying on $\text{Ca}(\text{OH})_2$ provided by cement, while the old mortar attached to the surface of RA has reduced $\text{Ca}(\text{OH})_2$ content due to long-term carbonation [10–12], forming a “low alkali reserve–inhibited activation cycle”. In addition, the porosity of RA (usually, 5–15%) is significantly higher than that of natural aggregate (NA) (1–3%), and its internal defects and the weak layer of the interfacial transition zone (ITZ) together constitute a CO_2 diffusion channel, which accelerates the process of the carbonation of concrete [13–15] and seriously threatens the service life of the structure.

Existing studies have been conducted to enhance the performance of recycled concrete (RAC) through two strategies: physical reinforcement and chemical excitation. Physical reinforcement is mainly based on nanomaterial modification by adding nanomaterials to fill aggregate pores and optimize the ITZ structure. Chen et al. [16] found that the incorporation of 3% nano- SiO_2 could reduce the ITZ width of RAC from 50 μm to 40 μm and increase the compressive strength by 25.55%. Cheng et al. [17] reported that the combined addition of nano- CaCO_3 and FA significantly enhances the compressive strength of concrete by 7.1 MPa, reduces electrical flux by 23.2%, lowers hydroxide (CH) content to 9.12%, markedly decreases pore structure, and greatly improves density. However, the high cost and agglomeration difficulties of nanomaterials limit their large-scale application. Chemical excitation, on the other hand, is centered on alkali excitation technology, which activates FA activity by introducing exciters such as Na_2SiO_3 and CaO . Alsarhan H et al. [18] have comprehensively analyzed the potential of industrial by-products (e.g., fly ash, slag) in alkali-activated systems, demonstrating their ability to balance mechanical performance with environmental benefits. Liu [19] demonstrated that the incorporation of Na_2SiO_3 endows FRAC with remarkable early-age strength characteristics. Specifically, the 3-day compressive strength reached 65% of the 28-day strength, and the 7-day strength achieved 75–80% of the 28-day value. Jiang et al. [20] investigated the compound addition of Na_2CO_3 , NaOH , and Na_2SiO_3 . They found that increasing the Na_2CO_3 content prolonged the setting time. At an alkali dosage of 6% and Na_2CO_3 substitution ratio of 40%, the geopolymer cementitious material exhibited the maximum slump flow and attained a 28-day compressive strength of 94.4 MPa, representing the optimal performance in their study. However, the existing research focuses on the influence of a single factor, and the systematic research on the mechanism of the coupling of the “aggregate pore space-alkali environment” is still blank.

The carbonation process exerts a dual effect on recycled concrete: (1) CO_2 reacts with $\text{Ca}(\text{OH})_2$ to form CaCO_3 , filling micropores and enhancing surface densification [21]; (2) the internal pH reduction triggers the decalcification of the C–S–H gel, leading to mechanical degradation [22]. Xiao et al. [23] observed that the full replacement of NA with RA tripled the carbonation rate. The old mortar attached to the surface of RA not only reduces the bond with cement paste [24] but also has more connecting pores, which becomes a channel for Mg^{2+} , SO_4^{2-} , and Cl^- to enter into the interior of the concrete, which in turn reduces the chlorine-ion penetration resistance of RAC [25,26]. Alkali activators enhance carbonation resistance by increasing the alkalinity of the pore solution, which stabilizes hydration products (C–S–H and Aft) and delays CO_2 ingress. However, in RAC, pre-carbonated old mortar on aggregate surfaces reduces the available $\text{Ca}(\text{OH})_2$, creating a “low-alkali reserve” that accelerates carbonation. FA mitigates this by providing reactive SiO_2 and Al_2O_3 , which react with CaO to form additional C–S–H gels, densifying the matrix and blocking CO_2 diffusion paths [27]. The excessive addition of alkali activations may trigger the abnormal generation of Aft, which will exacerbate the pore connectivity [28]. This dynamic interaction of carbonation alkali activation–pore evolution has not been fully revealed,

which makes it difficult to break through the bottleneck of durability enhancement in the existing modification techniques.

From the perspective of microscopic mechanisms, the carbonation process of concrete is regulated by the alkali reserve of the cementitious system and the fractal characteristics of the pore structure. The former determines the buffering capacity of the CO₂ neutralization reactions, and the latter influences the CO₂ diffusion rate. Traditional studies predominantly employ a single index to characterize the durability of concrete, often failing to establish multi-scale correlations. In recent years, fractal theory has provided a new paradigm for the quantitative characterization of pore structures. For instance, Ren et al. [29] utilized the box-counting dimension method to reveal a significant positive correlation between the multifractal spectral width of aggregate distribution and the fractal dimension (FD) of crack distribution ($D = 1.2252\text{--}1.3955$), and Gong et al. [30] developed a FD calculation method based on SEM images. By applying grayscale image binarization and Otsu threshold segmentation, they quantified the microstructural roughness of the interfacial transition zone (ITZ). Their results demonstrated that a higher FD corresponds to a rougher surface, implying greater complexity and densification of the ITZ microstructure. However, there is still a lack of systematic research on the pore fractal evolution law of alkali-activated recycled concrete and its quantitative mapping relationship with macroscopic properties.

Based on this, this present study designed different alkali activator dosages (0%, 4%, 8%, and 12%) to systematically investigate the synergistic mechanism of carbonation alkali activation on the performance of recycled concrete. Through carbonation depth tests, compressive strength analysis, and multi-scale characterization methods (SEM, XRD, and pore fractal analysis), the regulatory effects of alkali activator dosage on the carbonation resistance and microstructure of concrete were revealed. This research provides theoretical foundations and technical support for solid waste resource utilization and low-carbon concrete design.

2. Materials and Methods

2.1. Materials

The materials used in this test are shown in Figure 1. The cement used in this test was Shilin brand P.O42.5 ordinary silicate cement produced in Kunming City, and its physical properties and indexes are shown in Table 1. The FA used was first-grade ash produced by Shijiazhuang Shang'an Power Plant, and its test report is shown in Table 2. The fine aggregate was sand with a fineness modulus of 2.3 and an apparent density of 2250 kg/m³. NA is made of natural granite gravel with continuous grading; its apparent density is 2746 kg/m³, its bulk density is 1491 kg/m³, and its saturated surface dry water absorption rate is 0.6%. RA was sourced from waste roadbed concrete; it was mechanically crushed using a PE-150×250 jaw crusher (manufactured by Nanchang Hengshui Testing Equipment Manufacturing Co., Ltd., Nanchang, China) to a particle size of 5–20 mm and subjected to air classification in the laboratory to remove loose particles and impurities. The physical properties of RA were as follows: apparent density of 2560 kg/m³, bulk density of 1295 kg/m³, water absorption of 5.0%, crushing value of 18.7%, and stone powder content of 4.5%. No chemical pretreatment was applied to preserve the inherent reactivity of the aggregate. The alkali activator employed was calcium oxide (CaO).

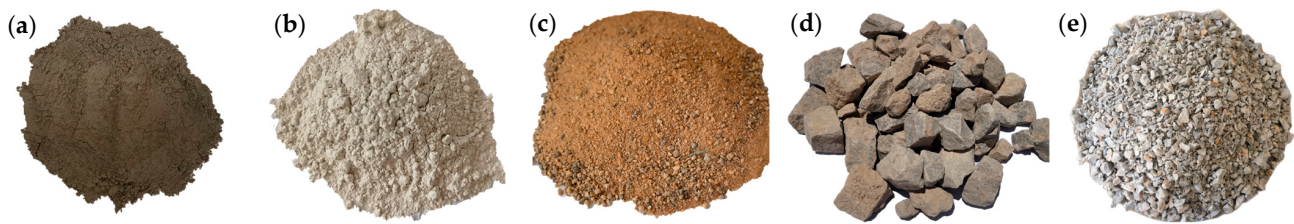


Figure 1. Raw materials ((a) cement; (b) FA; (c) fine aggregate; (d) NA; (e) RA).

Table 1. Basic properties of cement.

	Compressive Strength/MPa		Flexural Strength/MPa		Solidification Time/min		Stabilizing
	3 d	28 d	3 d	28 d	Initial Set	Final Set	
Standardized value	≥ 17.0	≥ 42.5	≥ 4	≥ 6.5	≥ 45	≤ 600	Conformity
Measured value	17.6	45.5	5.1	7.8	1545	371	Conformity

Table 2. Fly ash test report.

Chemical Composition	SO ₃	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO
Quantity contained (%)	0.8	5.6	43	23	2.5	0.95

2.2. Mix Proportion

In this test, the water-to-binder (w/b) ratio was 0.5, RA mixing was 30%, FA mixing was 30%, and alkali activator was externally mixed according to the proportion of FA, according to different alkali activator mixing (0%, 4%, 8%, 12%), to design four experimental groups (J0, J4, J8, J12). The specific mixes are shown in Table 3.

Table 3. Mix proportions of fly ash recycled concrete (kg/m³).

Group	Cement (kg/m ³)	Recycled Aggregate (kg/m ³)	Coarse Aggregate (kg/m ³)	Fine Aggregate (kg/m ³)	Water (kg/m ³)	Fly Ash (kg/m ³)	CaO (kg/m ³)
J0	287	338.33	789.42	607.25	221	123	0
J4	287	338.33	789.42	607.25	221	123	4.92
J8	287	338.33	789.42	607.25	221	123	9.84
J12	287	338.33	789.42	607.25	221	123	14.76

Note: the alkali activator dosage in the table is calculated based on the amount of FA, and the water contains additional water.

2.3. Carbonation Test Method

According to the specification GB/T50081-2019 [31] “Standard Test Methods for Mechanical Properties of Ordinary Concrete”, the specimens were made into cubic specimens of 100 mm × 100 mm × 100 mm and put into the standard curing box to maintain for 28 d. According to the specification GB/T50082-2024 [32] “Standard Test Methods for Long-Term and Durability Properties of Ordinary Concrete”, the specimens were baked at 60 °C for 48 h, and after drying, the specimens were sealed with heated paraffin wax on the rest of the surface except for the two opposite sides, and then were placed in the carbonization box so that the carbon dioxide concentration in the box could be maintained at (20 ± 3)%, the relative humidity could be controlled at (70 ± 5)%, and the temperature could be controlled in the range of (20 ± 2) °C. The specimens were placed in the carbonization box at (20 ± 2) °C. In the carbonization process at 3 d, 7 d, 14 d, and 28 d, respectively, the specimens were taken out, their compressive strength was tested, and then they were sprayed with a 1%

concentration of phenolphthalein alcohol solution on the cleavage surface. After waiting for about 30 s, they should have been in line with the original marking of every 10 mm at a measuring point with a steel plate ruler to measure out the depth of carbonization at each point, and then the average depth of carbonization was calculated.

2.4. Microstructure Characterization Methods

SEM and EDX were used to observe the microstructural characteristics, elemental species, and content levels of the recycled concrete specimens. The composition of the samples was analyzed with the help of XRD. To characterize the pore structure of alkali-activated fly ash concrete (JFRAC), the BSE-IA method was employed under the following assumptions:

1. The pore structure was assumed to exhibit statistical self-similarity within the analyzed scale range (50 nm to 50 μ m).
2. Pores smaller than the SEM resolution threshold (50 nm) were not captured in the imaging process.
3. Fractal dimension (FD) values derived from 2D images may underestimate the true 3D pore complexity; however, averaging across multiple imaging planes was applied to mitigate this effect.

The distribution of the physical phase of JFRAC was further analyzed using FD calculations based on the box dimension method.

2.5. Flowchart

This paper focuses on the study of the effect of alkali admixture on the carbonation resistance of concrete, and the specific technical route is shown in Figure 2.

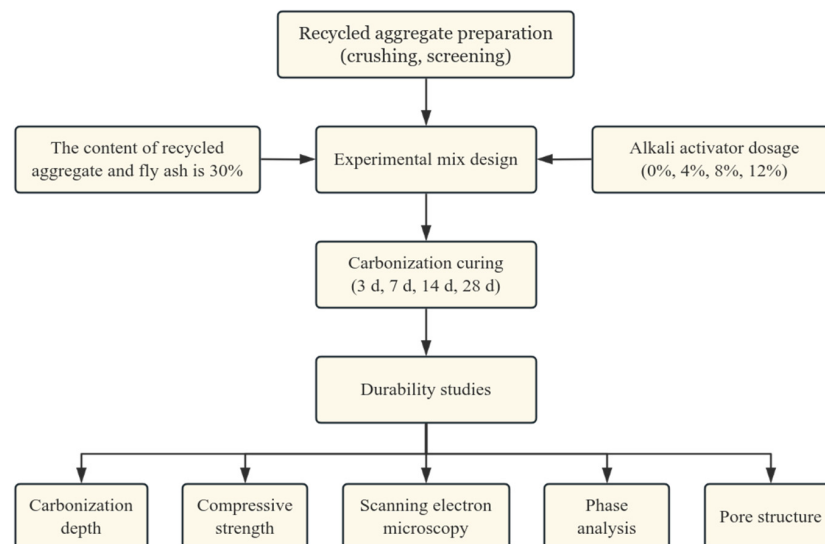


Figure 2. Flowchart.

3. Results and Discussion

3.1. Carbonation Depth and Compressive Strength

Carbonation depth is a critical indicator of concrete durability; the carbonation depth of different specimens is shown in Figure 3. The carbonation depth of specimens increases with the growth of carbonation time, and at the ages of 3 d and 7 d, the carbonation depth of JFRAC decreases with the increase of alkali stimulant dosage. The carbonation depth of the J0, J4, and J8 groups exhibited a faster increase in the early stage, followed by a gradual slowdown in the later stage. The J12 group's carbonation depth development and the J0, J4,

and J8 groups are more consistent, but the lengthening of the carbonation age carbonation depth development rate increased significantly. In summary, the carbonation depth of the concrete and alkali activator admixture is a non-linear relationship, and there is an optimal admixture of alkali activator. The admixture of the alkali activator can significantly inhibit the increase of the carbonation depth in the process of concrete carbonation, and the best inhibition effect is achieved when the admixture of the alkali activator is 8%; however, the long-term inhibition effect is weakened when the admixture is too large, and it was analyzed that cement is the main source of $\text{Ca}(\text{OH})_2$. Additionally, the use of FA to replace cement reduces the content of $\text{Ca}(\text{OH})_2$ in the concrete, and the hydration process of FA consumes $\text{Ca}(\text{OH})_2$ [33], further reducing the alkaline reserve. In addition, the $\text{Ca}(\text{OH})_2$ in the old mortar attached to the recycled aggregate has been partially converted to CaCO_3 , lowering the initial alkalinity decreases and increasing the risk of carbonation. While the high alkaline environment inhibits CO_2 penetration [34], the alkalinity of recycled concrete can be increased by adding alkali activators to delay the rate of carbonation reaction and enhance the carbonation resistance of JFRAC, but the alkalinity of the pore fluid in concrete will increase when the admixture of alkali activators is too large, so that the pore structure is not dense enough. This structural defect will provide more channels for the diffusion of CO_2 , thus accelerating the process of carbonation depth.

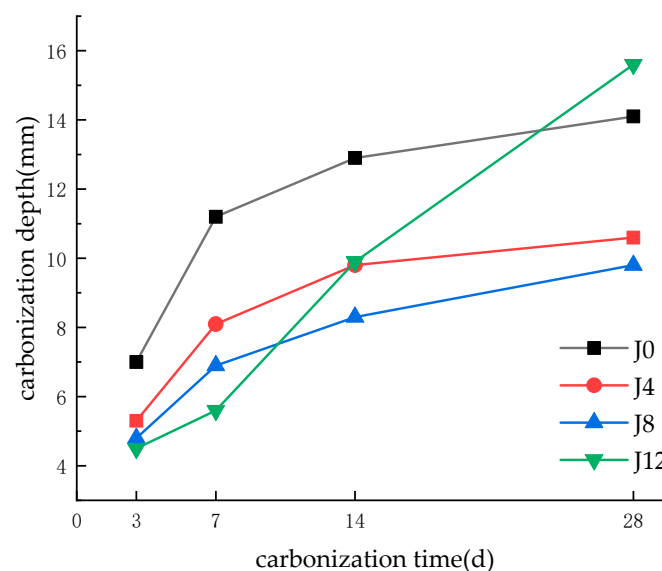


Figure 3. Variation of carbonization depth of JFRAC.

The carbonation divergence among groups stems from the dynamic balance between RA porosity and alkali activation–carbonation synergy. J0 and J4 exhibit continuous carbonation due to insufficient alkalinity or pore filling, while J8 achieves surface stabilization through optimal dosage, and J12's microcracks trigger accelerated carbonation.

The evolution of compressive strength of JFRAC with carbonation time is shown in Figure 4; with the increase of the alkali-exciter dosage, the compressive strength of specimens first increased and then decreased, and with the prolongation of the age of carbonation of specimens in each group, the compressive strength showed an increasing trend. After 28 d of carbonation, the compressive strength of the J4 and J8 groups increased by 3.96% and 10.76%, respectively, compared with that of the J0 group, and the compressive strength of the J12 group decreased by 7.61%, which is consistent with the trend of the carbonation depth. The compressive strength of JFRAC after carbonation exhibits a significant peak at the optimal alkali activator dosage of 8%, while excessive dosage results in reduced strength owing to pore structure inhomogeneity and microcrack propagation. The

increase of alkali activator dosage has a significant promotion effect on the early hydration process of the specimen, which is because alkali activators can induce the dissolution of Ca^{2+} , Al^{3+} , Si^{4+} [35], and other reactive substances in FA and polymerize to produce C–S–H and C–A–S–H gels, promoting the generation of early hydration products, making the internal structure more dense and increasing the compressive strength accordingly. The neutralization reaction between CO_2 and alkalis in the process of carbonation leads to the reduction of the pH value of concrete, which affects the stability of its hydration products and causes a decalcification reaction [36], but the proper supplementation of alkali can effectively avoid this situation. After 28 days of carbonation, the compressive strength of the J12 group exhibited a decreasing trend. Consistent with the findings of Provis [37], highly alkaline environments accelerate the reaction kinetics but may result in the formation of brittle reaction products. CaO is a kind of expanding agent; in addition to the expansion of destruction in the process of mixing, it may also occur in the late stage of concrete hardening. So, when the alkali activator is mixed too much, it leads to the uneven distribution of hydration products and can even produce large holes, instead of reducing the compressive strength of concrete.

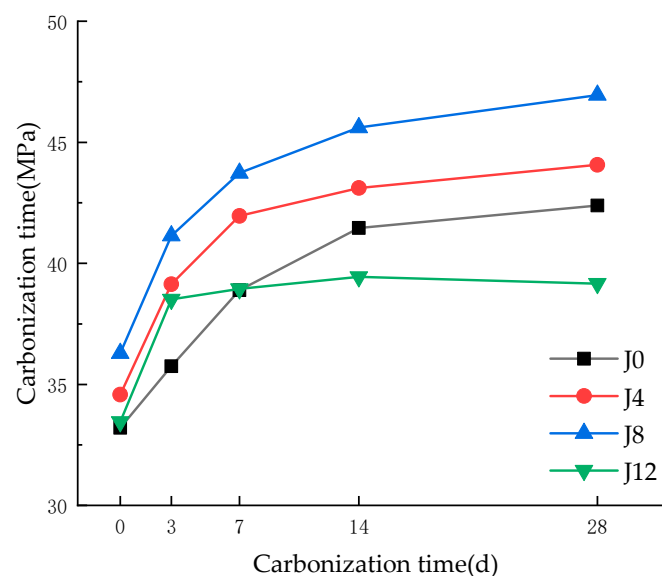


Figure 4. Variation of compressive strength of JFRAC.

3.2. Comparative SEM Analysis of Microstructural Evolution Pre- and Post-Carbonization

In order to investigate the connection between the mechanical properties and microstructure of JFRAC under carbonation conditions, scanning electron microscopy and energy spectroscopy tests were carried out on concrete samples. Figure 5 shows the microstructure of alkali-activated agent mixing of 0%, 4%, 8%, and 12%, respectively, at the curing age of 28 d. The structure of the concrete samples of the J0 group is relatively loose (Figure 5a), with more pores in the interior and the presence of a large number of unhydrated FA microbeads. Only a small number of microbeads attached to the surface of scattered hydration products due to the existence of the natural aggregate side wall effect, while the recycled aggregate surface attached to more hardened cementite; due to the existence of more large pores, its own strength is low, and it has strong water absorption. Therefore, the bonding ability between the recycled aggregate and the cement matrix is weak, resulting in an obvious weak area in the interface transition zone. With the admixture of the alkali activator (Figure 5b), the surface of FA in J4 group samples began to be covered with thin film hydration products, which improved the interfacial bond between FA and cement matrix. However, due to the incomplete hydration of FA with insufficient alkali

activator, the amount and distribution of the hydration products may not be uniform, and the generation of AFt was inhibited, presenting fine needles. The generation of J8 group FA was more complete, and the presence of fly ash microbeads in the diagram was almost not observed, which led to the presence of obvious weak areas in the transition zone. The presence of FA beads can be seen in Figure 5c, and the C–S–H gel with a high degree of polymerization is interwoven and accompanied by needle-like AFt and square crystal calcium carbonate, which together form a reticulated structure. A large number of dense gels are filled with pores to optimize the interfacial transition period. In the specimens in group J12 (Figure 5d), this is mainly due to the high dosage of the alkali stimulant to produce a large number of hydration products covering in the surface of the FA, which can hinder the unhydrated particles from continuing to participate in the reaction of FA. Reacted FA particles continue to participate in the reaction, resulting in an incomplete reaction, and the total amount of gel generated is reduced [38]. When the age reaches 28 d, FA continues to undergo hydration during the later stages of concrete curing, generating the N–A–S–H gel with poor strength and leading to the further development of microcracks. There was also a large number of coarse needle-rod crystals in the specimen, and the EDS spectral analysis showed that the main constituent elements of the crystals were O, Al, S, and Ca; it was presumed that the substance was AFt [39,40]. This is because under high alkaline conditions (e.g., higher pH), the formation rate of AFt is accelerated, and the direction of crystal growth is more inclined to the formation of a coarser rod structure [41]. When the alkali activator admixture exceeds the optimal admixture, the high saturation environment forms in the concrete, which will not only promote the generation of hydration products as well as AFt but also make the morphology of AFt change significantly compared with the lower dosage of alkali excitors [42]. Additionally, the coarser rod-shaped AFt crystals can form and fill the pores in the cement paste rapidly, which helps to improve the early strength of concrete.

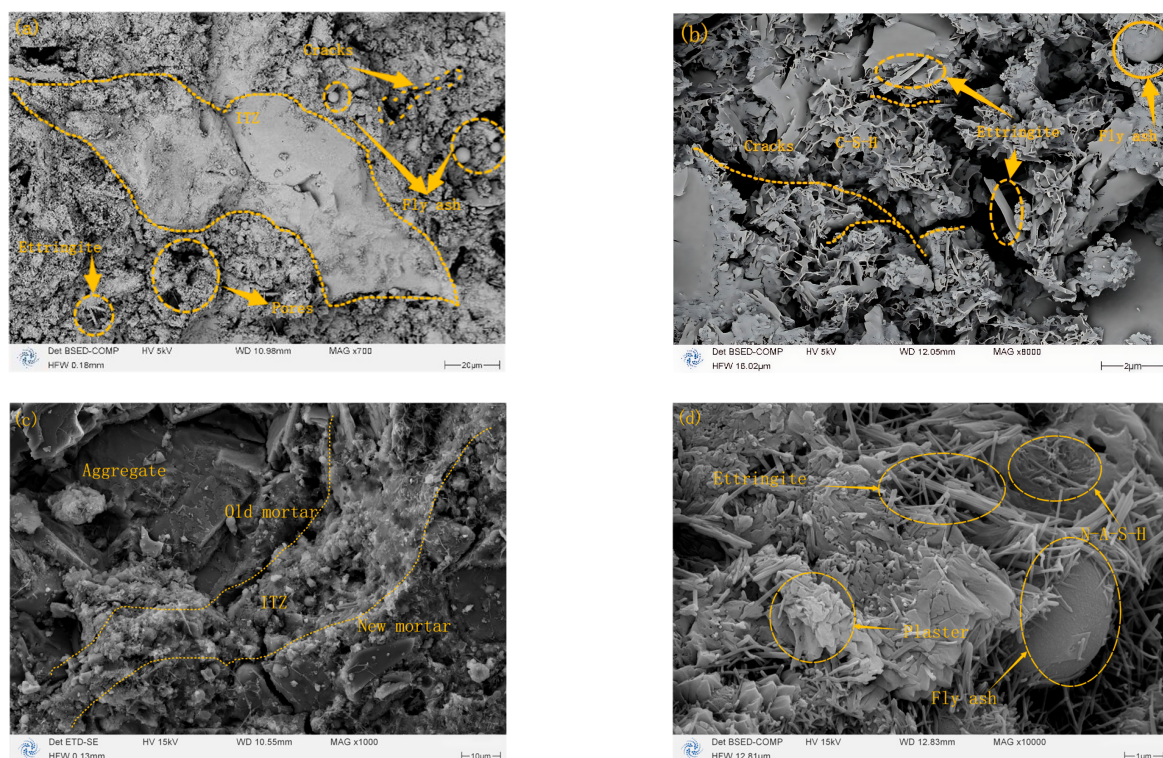


Figure 5. Microstructure of concrete before carbonization: (a) J0; (b) J4; (c) J8; (d) J12.

CaCO_3 generated during carbonation fills surface micropores, slightly enhancing surface hardness, resulting in a slight increase in the surface hardness of the concrete. This increase in surface hardness can improve the abrasion resistance of concrete to a certain extent, but carbonation will also change the acid–base environment inside the concrete and reduce the alkalinity. Figure 6 shows the microstructure of each group of specimens after the carbonation test. The surface layer of the J0 group after carbonation appears as a layer of thin and discontinuous CaCO_3 (Figure 6a), and this is due to the lack of the addition of alkali activators. The overall hydration reaction of the concrete is not sufficient because the amount of calcium carbonate generated is relatively small and its own hydrated calcium silicate generated by the cement is limited. Additionally, the structure was not dense enough, and it was difficult to form a continuous network structure, which also led to the rapid intrusion of CO_2 into the interior, where the pH value decreased rapidly and the unreacted silicate particles were further decalcified. Some of the excess $\text{Ca}(\text{OH})_2$ in the specimens of group J4 reacted with CO_2 to generate CaCO_3 (Figure 6b), filling some pores and blocking some of the diffusion of CO_2 . The densities of the cement paste and the interfacial transition zone were improved, and the width of the interfacial transition zone was reduced, which indicated that the carbonation reaction made the interfacial structure of the recycled concrete denser. However, due to the late secondary hydration of FA in the early stage, the strength of the generated C–S–H gel is low, and its network structure is not good enough, resulting in poorer carbonation resistance of the matrix. Further, the alkali activator is insufficient, and the internal pores of the concrete are even more so. Also, the distribution of CaCO_3 is not uniform enough, and the concrete compactness is poor, so the interior is still carbonated gradually, resulting in the decalcification of the C–S–H gel to generate silica gel. Therefore, the strength enhancement is limited when the dosage of the alkali stimulant is 4%. When the dosage of the alkali stimulant is 8%, the appropriate alkali environment is favorable for the generation of an appropriate amount of AFt (Figure 6c), which fills in the pores of cement stone to form a dense skeleton structure, providing good early strength for the concrete [43,44]. During the carbonation process, the content and development of CaCO_3 inside the concrete increase, which enhances the adhesion cement of the recycled aggregate and effectively improves the defects of the recycled aggregate itself. With the growth of carbonation age, a large amount of CaCO_3 is uniformly distributed in the surface layer of the concrete specimen, which contributes to the increase of the degree of carbonation consolidation, and there is no obvious weak zone at the interface of the sand and the cement paste. FA and the matrix filler are closely combined to inhibit the further invasion of CO_2 , and from the micrographs, it can be seen that in the pore space under the surface of the concrete layer, there is an appropriate amount of AFt as a supporting skeleton. The hydration product is a denser reticulation; the surface of the ordered attachment has an appropriate amount of CaCO_3 , further reducing the porosity of the interfacial transition zone; the concrete is still maintained in the internal high pH environment; the C–S–H and AFt are stabilized; the carbonation resistance is significant; and there is a significant effect on the macroscopic performance of the recycled concrete. The J12 group produces a large number of hydration products in the pre-conservation period (Figure 6d), covering the unreacted FA particles and resulting in a large number of inhomogeneous pores inside the concrete, while AFt as an expansive substance leads to the internal structure of the concrete being loose and porous, and the bonding between the aggregate and the paste is weakened, which makes CO_2 invade the inside of the concrete very quickly. Additionally, the high alkali content leads to an increase in the pH value in the pore solution, which accelerates the rapid development of carbonation and the macroscopic manifestation of this rapid increase in the depth of carbonation. Distinct cracks and abundant CaCO_3 crystals are observed in ITZ at the periphery of the J12 specimen group. The volumetric expansion of

CaCO_3 induces internal stress concentration, exacerbating microcrack propagation caused by AFt expansion [45,46]. This results in CO_2 more easily entering the deeper layers of concrete to react, and due to the more heterogeneous internal structure of concrete, the generated CaCO_3 crystals cannot uniformly fill the space of the pores and cracks, which makes the connecting pores increase; the structure of the interfacial transition zone then becomes loose, and unreacted Ca(OH)_2 combined with microcracks form structurally weak regions. This “carbonated shell–fragile core” configuration ultimately limits the overall mechanical enhancement [47].

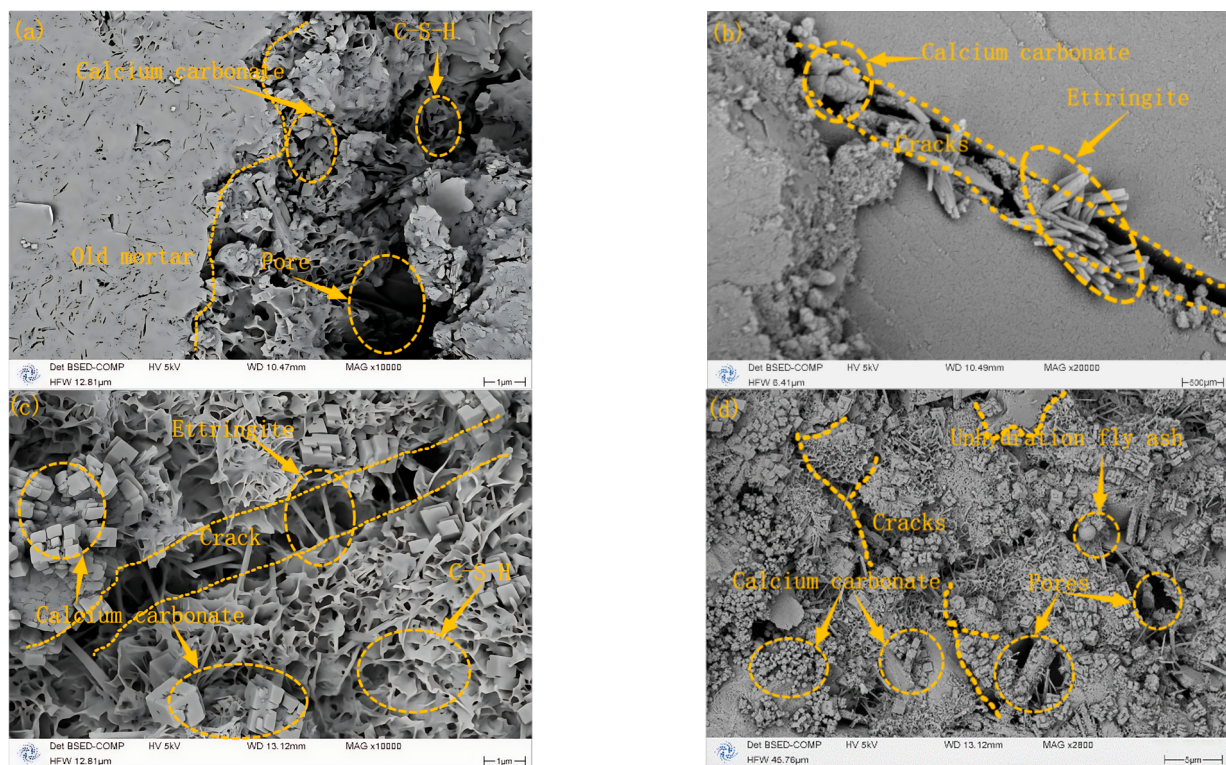


Figure 6. Microstructure diagram of carbonized concrete: (a) J0; (b) J4; (c) J8; (d) J12.

3.3. XRD Analysis of JFRAC

Figure 7 shows the XRD diffraction patterns of the surface materials of the test blocks of groups J0, J4, J8, and J12 after 28 days of carbonization. The diffraction peaks mainly include SiO_2 , AFt, CaCO_3 , Ca(OH)_2 , $\text{CaSO}_2\text{--H}_2\text{O}$, and rhombohedral calcium zeolite, and the diffraction peaks of Ca(OH)_2 in the J0, J4, and J8 groups have completely disappeared. It can be clearly seen that there is a large amount of CaCO_3 , which indicates that the surface layer of the material has been completely carbonized. With the increase of alkali activator doping, the diffraction peak of CaCO_3 is improved, and the diffraction peak of SiO_2 is reduced, which also proves that the hydration of FA in the J8 group is more complete. In the J12 group, compared with that of the J8 group, the diffraction peak of CaCO_3 is reduced, the diffraction peak of SiO_2 is strengthened, and the diffraction peak of Ca(OH)_2 still exists, which is due to the existence of excessive alkali activators that accelerated the hydration rate of the pre-hydrocolloid materials, a large number of hydration products, and the AFt, which quickly wrapped up the non-hydrated colloid materials and alkali activators, making the Ca(OH)_2 peaks more and more visible. The C(–A)–S–H gels have small grains, low crystallinity, and no obvious diffraction peaks, but it can be seen that there are obvious broad peaks in the range of $20\text{--}45^\circ$ for each sample.

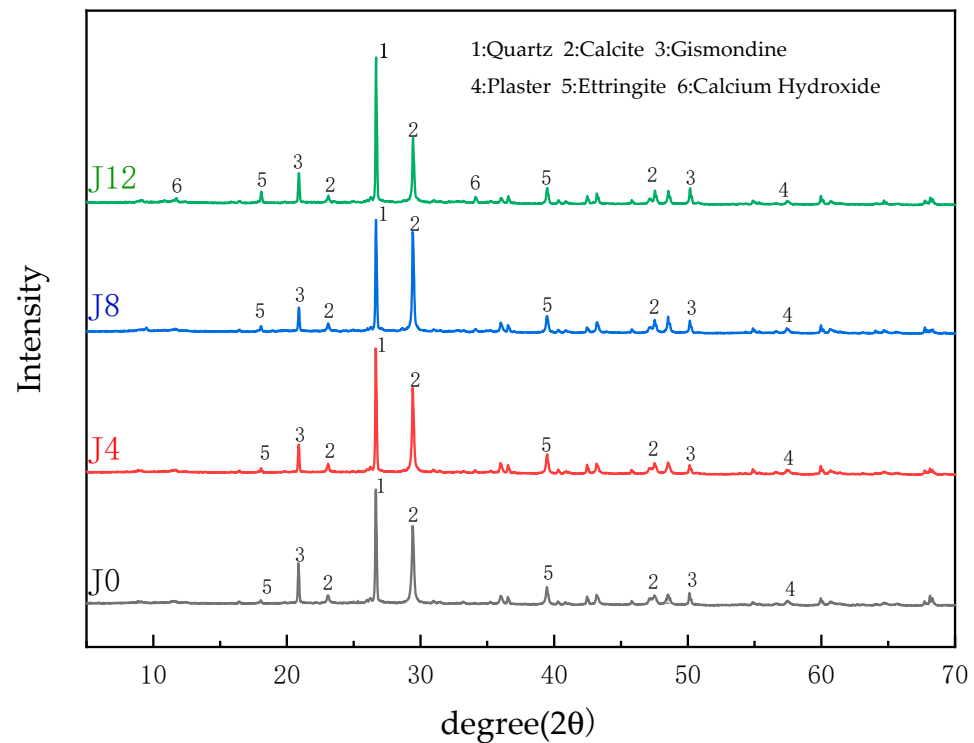


Figure 7. XRD diffraction pattern of carbonization at 28 days with different base activator dosages.

3.4. Effect of Carbonization on the Physical Phase Distribution of Pores

The pore diagrams of hydration and erosion products using MATLAB R2019b in Figure 8, where the red markers are pores, show the pore physical analysis before carbonation. According to the MATLAB analysis statistics, the J0 porosity is 39.33%, and the maximum pore diameter is 1.72 μm ; the J4 porosity is 31.92%, and the maximum pore diameter is 2.06 μm ; the J8 porosity is 22.87%, with a maximum pore diameter of 0.93 μm ; and the J12 porosity is 38.47%, with a maximum pore diameter of 2.27 μm . The porosity of the concrete decreases with the increase of alkali activators, and when the alkali activator dosage is 12%, the pore space of the concrete increases, becoming visible to the naked eye, which corroborates the fact that when the alkali activator dosage is 8%, the FA hydration is the most complete, and the specimen's structure is the densest.

A large number of studies have shown that the appropriate amount of alkali activators will promote the hydration of concrete and CaO as an alkali activator in the carbonation process for the internal reaction of the concrete to provide Ca^{2+} , so as to improve the amount of carbon sequestration of concrete [48]. Figure 9 shows the pore distribution of each group after 28 d of carbonation, and it can be seen that the porosity of each group after carbonation is significantly reduced. According to the statistical data, its porosity was reduced by 51.46%, 70.11%, 79.72%, and 50.09%. The pores of the J8 group are small, few, and uniformly distributed; the pores of the J4 group and J8 group are significantly smaller than those of the J0 and J12 groups; and the irregularities of porosity of the J8 group are significantly lower than those of J4. The maximum diameter of the pores is reduced compared with that before carbonation, and the structure of the concrete after carbonation with 8% alkali stimulated admixture is the most dense, which is consistent with its macroscopic performance.

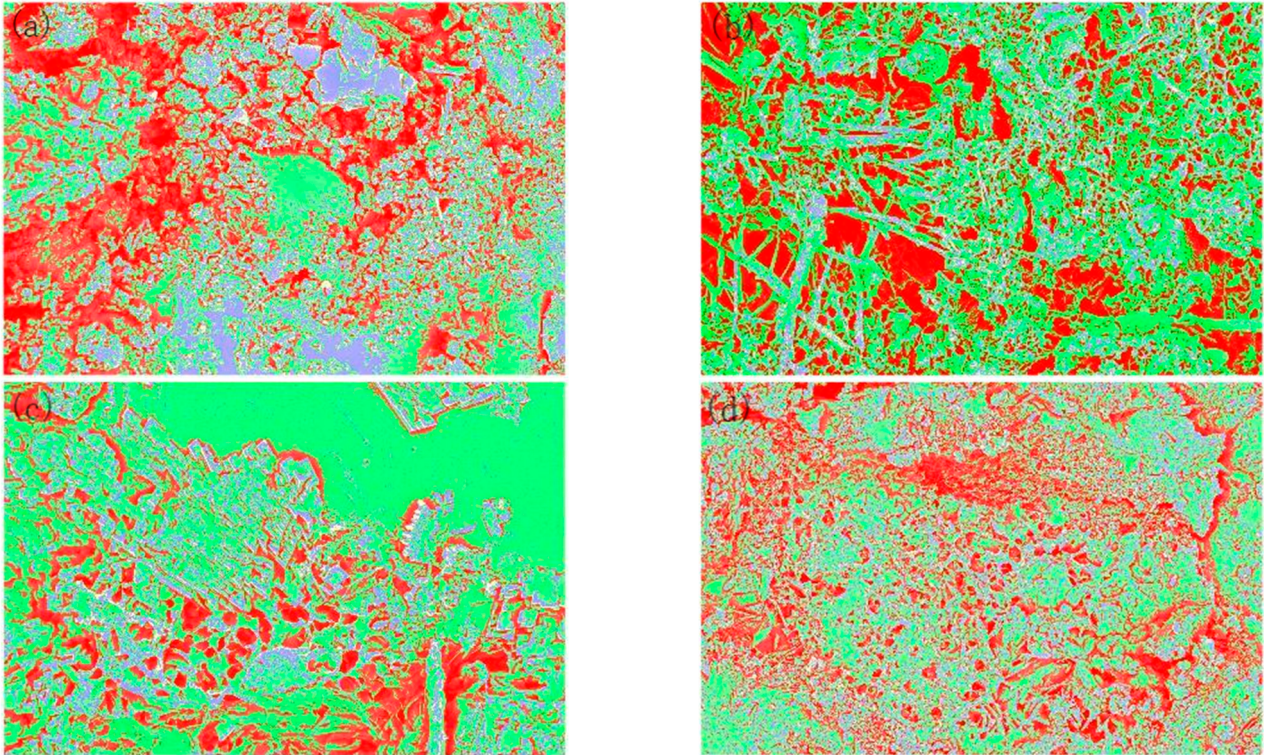


Figure 8. Pore distribution before carbonization: (a) J0; (b) J4; (c) J8; (d) J12.

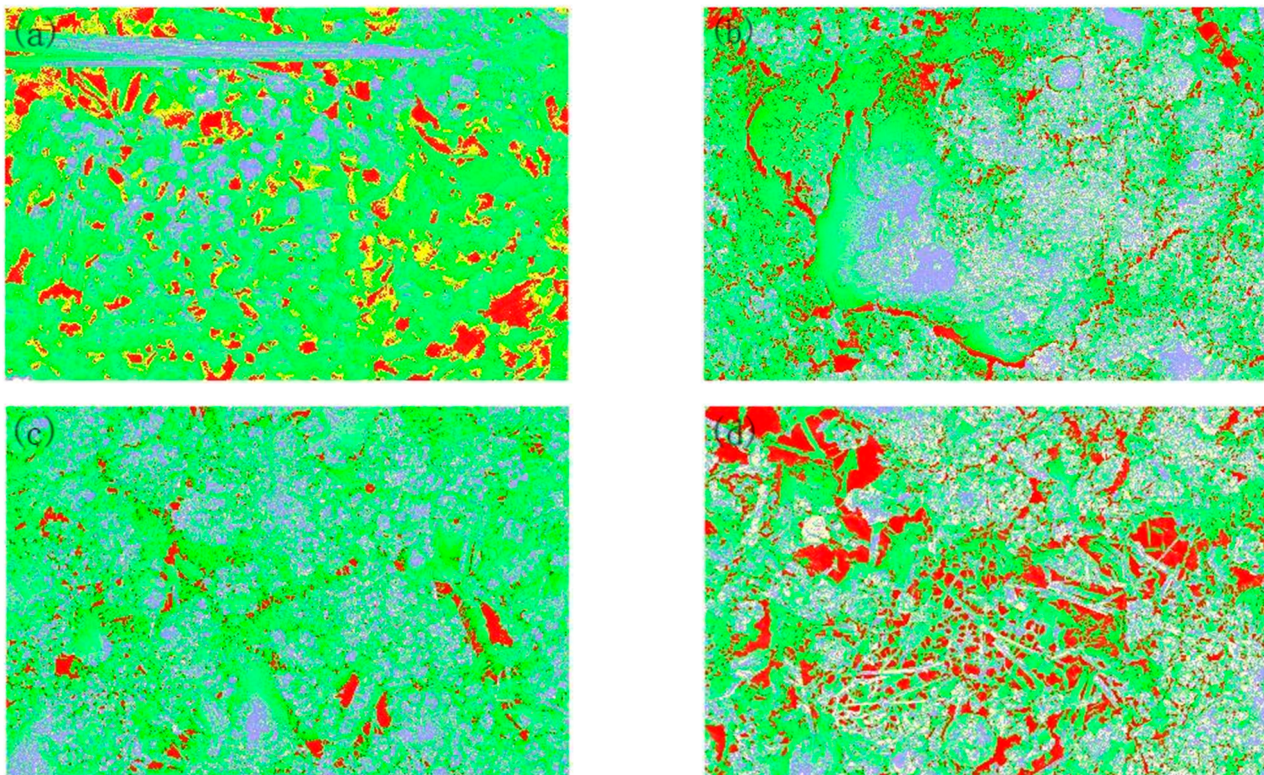


Figure 9. Pore distribution after carbonization: (a) J0; (b) J4; (c) J8; (d) J12.

3.5. Fractal Analysis of Pore Structure Evolution in Concrete During Carbonation

Fractal dimension (FD) quantifies the complexity [49–51] and irregularity of pore structures, providing critical microscopic insights into the evolution of concrete durability. Based on the box-counting method (Table 4, Figure 10), the FD values of carbonated concrete

increased significantly ($R^2 > 0.998$), indicating that carbonation refines large pores ($>1 \mu\text{m}$) into smaller micropores ($<0.5 \mu\text{m}$) through CaCO_3 precipitation, thereby enhancing the spatial filling capability of the pore network. For the J0 group, the FD increased from 1.891 to 1.930 after carbonation, reflecting a 2.03% improvement in pore complexity. The optimal dosage group, J8, exhibited the highest FD increase from 1.915 to 1.947, with uniform pore distribution (Figure 9c) and minimized maximum pore size. The FD showed a strong negative correlation with carbonation depth and a positive correlation with compressive strength, confirming that high FD values (>1.9) correspond to low pore connectivity, effectively suppressing CO_2 diffusion pathways. Excessive alkali activators reduced the FD to 1.926 due to the accelerated formation of coarse AFt crystals (Figure 5d) and microcrack propagation (Figure 6d), which disrupted pore self-similarity. These heterogeneous pores (maximum size $2.27 \mu\text{m}$) formed rapid CO_2 diffusion channels, increasing the carbonation depth by 35.2% (Figure 3).

Table 4. Fractal dimension.

	Pre-Carbonation		Post-Carbonization	
	Fractal Dimension Estimation	R^2	Fractal Dimension Estimation	R^2
J0	$y = 0.16961 - 1.8912x$	0.998	$y = 0.11657 - 1.9296x$	0.999
J4	$y = 0.14308 - 1.9041x$	0.999	$y = 0.11349 - 1.9306x$	0.999
J8	$y = 0.14077 - 1.9147x$	0.998	$y = 0.09003 - 1.9474x$	0.999
J12	$y = 0.16993 - 1.8915x$	0.998	$y = 0.12384 - 1.9257x$	0.998

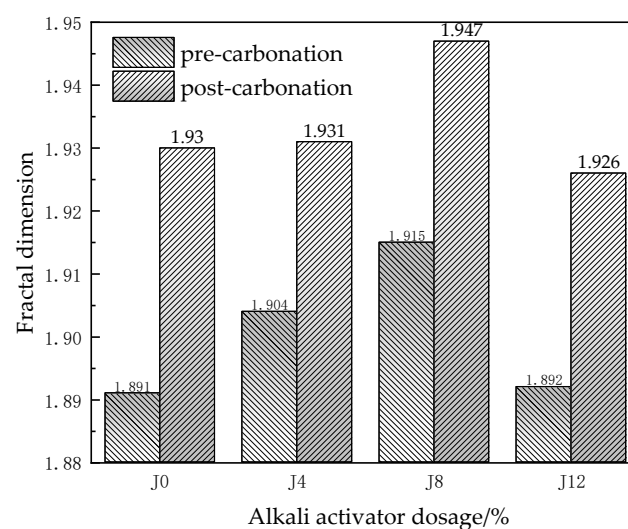


Figure 10. Fractal dimension.

3.6. Embodied Carbon Analysis

The embodied carbon of CaO was calculated based on its dosage in each mix. For J8 (8% CaO by FA mass, FA = 123 kg/m^3), the CaO content was 9.84 kg/m^3 , resulting in upstream emissions of $\sim 9.84 \text{ kg CO}_2/\text{m}^3$ ($1.0 \text{ kg CO}_2/\text{kg CaO}$ [52]). While this increases the initial carbon footprint, the enhanced carbonation resistance (35% depth reduction) may extend the service life by 15–20 years in aggressive environments, potentially offsetting emissions by deferring reconstruction ($\sim 300 \text{ kg CO}_2/\text{m}^3$ per cycle [53]). A full lifecycle assessment is warranted to validate this balance.

4. Conclusions

This study systematically investigates the synergistic effects of alkali activator dosage (0%, 4%, 8%, 12%) on the carbonation resistance and microstructural evolution of FRAC. The results showed that:

- (1) **Optimal Alkali Dosage Enhances Carbonation Resistance and Microstructural Integrity:** The incorporation of 8% CaO as an alkali activator significantly improves the carbonation resistance of fly ash recycled aggregate concrete (FRAC), achieving a 35% reduction in carbonation depth and a 10.76% increase in compressive strength compared to the control group. This optimal dosage promotes the formation of dense C–S–H/AfT networks, reduces porosity to 22.87%, and optimizes pore fractal complexity ($FD > 1.9$), effectively suppressing CO_2 diffusion. However, excessive alkali activation (12% CaO) induces microcracks and pore interconnectivity, leading to a 7.6% strength reduction and accelerated carbonation.
- (2) **Fractal–Pore Relationships Reveal Durability Mechanisms:** Fractal dimension (FD) analysis establishes a quantitative link between pore complexity and macro-performance. A threshold of $FD > 1.9$ correlates with low pore connectivity and high durability, validating its role as a critical indicator for carbonation resistance. However, FD sensitivity to image resolution and binarization methods necessitates standardized protocols for future studies.
- (3) **Environmental and Practical Implications:** Despite the embodied carbon of CaO ($9.84 \text{ kg } CO_2/m^3$), its synergy with fly ash ($\approx 120 \text{ kg } CO_2/m^3$ reduction) and extended service life (15–20 years) ensure a net carbon benefit. This highlights the potential of alkali-activated FRAC in sustainable construction.

5. Study Limitations and Future Directions

5.1. Limitations

- (1) The current scope focuses on compressive strength and carbonation resistance. Other durability metrics (e.g., chloride ingress, sulfate attack) remain unexplored.
- (2) Thermogravimetric analysis (TGA) was not performed to quantify carbonation products.
- (3) The volumetric strain induced by CaO expansion was not directly measured.

5.2. Future Directions

- (1) **Multidisciplinary Modeling:** integrate DeepLabv3+ and EfficientNet-B7 for automated microstructural analysis, enabling the pixel-level quantification of pores, cracks, and ettringite morphology.
- (2) **Extended Testing:** evaluate carbonation behavior beyond 28 days to capture long-term degradation patterns.
- (3) **Durability Expansion:** validate the 8% threshold against chloride penetration, freeze–thaw cycles, and acid exposure.

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