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Technical Paper

Effect of Carbonation–Treated Recycled Aggregates on the Properties of Permeable Geopolymer Concrete

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Abstract: To address technical challenges such as the high water absorption of recycled aggregates (RA) and the low mechanical performance of permeable concrete, this study systematically investigates the optimization of carbonation modification processes for RA and their coupling mechanisms with geopolymer binder systems. By adjusting CO_{22} concentration and environmental humidity, the densification of RA was enhanced, resulting in a 24.6% reduction in water absorption and a 15.4% decrease in crushing value, significantly improving their applicability in green permeable concrete. Experimental results demonstrated that carbonation treatment induced in situ deposition of calcium carbonate on aggregate surfaces, effectively sealing microcracks and enhancing interfacial properties. The geopolymer concrete exhibited excellent permeability and mechanical strength, with a maximum compressive strength of 52.1 MPa, while maintaining performance consistency under controlled porosity (minor deviation from design values). Further analysis revealed that optimized binder compositions mitigated strength degradation caused by increased porosity and compensated for interfacial bonding deficiencies in RA. Freeze-thaw cycle tests indicated that RA-based specimens exhibited poor durability, with accelerated deterioration at higher porosity, while optimized binder formulations significantly enhanced frost resistance. A regression model was established to quantify the effects of porosity and aggregate type on durability.

Keywords: Recycled aggregates; Geoplymer; Permeable Concrete ; Mechanical Properties

1. Introduction

With the rapid advancement of global infrastructure construction, cement production generates massive carbon emissions (0.8-1 ton of CO_2 per ton of cement[1]), creating a sharp contradiction with the "dual-carbon" strategic goals. Meanwhile, China's annual construction waste has reached 3 billion tons, with waste concrete accounting for over 40%. Insufficient pavement permeability leads to urban waterlogging, intensified heat island effects, and slippery road surface. By crushing and sieving waste concrete into recycled aggregates and combining them with oligomeric gel which emits only 1/5–1/10 of the CO_2 of cement[2], permeable recycled concrete can be produced[3]. This approach simultaneously achieves solid waste recycling, low-carbon building materials, and improved road ecological environments, offering

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significant eco-economic benefits.

However, recycled aggregates exhibit significant performance degradation compared to natural aggregates due to their rough surfaces, high porosity, and microcracks, limiting their widespread engineering applications[4-6]. Enhancing the performance of recycled concrete has thus become a key research focus. Geopolymer-based recycled aggregate permeable concrete demonstrates early-age strength and high mechanical performance[7,8], along with excellent chemical resistance, partially compensating for the mechanical deficiencies of recycled coarse aggregates[9-11].

Carbonation treatment is a method that utilizes the reaction between CO_2 and calcium compounds in aggregates. The mineralization process involves CO_2 diffusion and dissolution, Ca^{2+} leaching from hydration products, and subsequent reactions with CO_3^{2-} in pore solutions to form $CaCO_3$ precipitates[11]. The filling of microcracks by $CaCO_3$ is an effective way to improve the physical properties of recycled aggregates[13]. Studies indicate that humidity is a critical factor in CO_2 mineralization curing-optimal moisture conditions are required for efficient reactions, while excessive humidity hinders CO_2 penetration[14]. Chen Xuyong[15]demonstrated that the quality, dosage, and single/mixed incorporation of recycled aggregates significantly affect concrete's mechanical properties. As the recycled aggregate content increases, compressive strength, splitting tensile strength, and axial compressive strength decline. However, higher-quality recycled aggregates can mitigate these adverse effects. Xuan [12]found that carbonation treatment can absorb 7.9 kg of CO_2 per ton of recycled aggregates (5-20 mm particle size). Carbonation not only enhances the mechanical properties and durability of recycled aggregates but also contributes to CO_2 sequestration, offering a novel solution for low-carbon road engineering[16].

Thus, the incorporation of oligomers and carbonation treatment can not only improve the performance of recycled concrete but also reduce atmospheric CO_2 levels, holding great promise in the context of global carbon reduction. Meanwhile, permeable concrete has gained increasing attention in urban road construction due to its ecological benefits, such as mitigating urban flooding, alleviating heat island effects, and improving groundwater recharge[15]. Nevertheless, the inferior properties of recycled aggregates may negatively impact the strength, permeability, and durability of permeable concrete. Therefore, in-depth research on carbonation treatment technology and its application in permeable concrete is of great significance.

This study focuses on the carbonation treatment of recycled aggregates, systematically investigating its enhancement mechanisms and evaluating the performance of carbonated recycled aggregates in permeable concrete. By analyzing the microstructure, physical properties, and interfacial interactions with cementitious materials, we optimize carbonation process parameters and elucidate their effects on the mechanical strength, permeability, and durability of permeable concrete. The findings provide a scientific basis for the efficient utilization of recycled aggregates and technical support for the large-scale application of permeable concrete in green, low-carbon road engineering.

2. Materials and Methods

2.1 Materials

Using a sealed carbonation reaction chamber to prepare recycled aggregates with varying CO_2 concentrations, humidity levels, and carbonation durations. First, standard sieves are used to screen the recycled aggregates, selecting particles with sizes of 3-5 mm and 5-10 mm as the treatment subjects. These particles are then evenly spread on a perforated shelf inside the reaction chamber. The CO_2 gas supply system is activated, and the intake flow rate is precisely controlled using a mass flow controller to gradually increase the CO_2 volume concentration inside the chamber to the target level, maintaining dynamic equilibrium. The carbonation reaction proceeds for the predetermined duration. After carbonation treatment, the residual CO_2 inside the chamber is rapidly purged to terminate the surface carbonation reaction, preventing any interference from unreacted acidic gases in subsequent aggregate performance tests。The recycled aggregates and the carbonation recycling device are shown in Figure 2.1.

(a)recycled aggregates (b) carbonation recycling device

Figure 2.1 The recycled aggregates and the carbonation recycling device

The fly ash and granulated blast furnace slag used in this study were supplied by a company based in Zhengzhou, China. The sodium silicate solution was purchased from Youreka Refractories Co., Ltd., with a solid content of 43.65% and a silica modulus of 2.25. Solid sodium hydroxide (96% purity, industrial grade) was obtained from Shanghai Aladdin Bio-Chem Technology Co., Ltd., and was added to the sodium silicate solution to adjust its modulus to 1.5.

The aggregates consisted of 4.75–9.5 mm basalt, commonly used in road construction, and were sourced from a concrete mixing plant in Shanghai. The recycled aggregates were obtained by crushing waste concrete from a decommissioned bridge section in Jiangxi Province. Tap water provided by the municipal water supply system in Shanghai was used for mixing.

Based on the results of preliminary experiments, three geopolymer binder formulations with optimized mechanical and functional properties were selected for the preparation of permeable concrete, considering both performance requirements and economic feasibility. The detailed mix proportions are presented in Table 2.1 and Table 2.2

Table 2.1 Oxide Composition of Granulated Blast Slag, and Fly Ash (%)

Type	SiO_2	Al_2O_3	Fe_2O_3	CaO	TiO_2	MgO	Na_2O	SO_3
Fly ash	53.97	31.15	4.16	4.01	2.04	1.13	0.89	0.73
Granulated blast slag	34.50	17.70	1.03	34.00	-	6.01	-	1.64

Table 2.2 Mix Proportions of Geopolymer Binder (Unit: g)

Mix ID	Fly Ash	Slag	Activator	Activator Modulus	Water
A	30	70	68.63	1.5	3.21
B	30	70	61.72	1.3	7.61
C	40	60	71.21	1.3	2.63

The mix proportions were determined using the volumetric method, following the guidelines outlined in the Technical Specification for Permeable Cement Concrete Pavements (CJJ/T 135-2009). Detailed formulations are provided in Table 2.3.

Table 2.3 Mix Proportions of Permeable Geopolymer Concrete (Unit: kg/m³)

Mix ID	Slag	Fly ash	Natural Aggregate	Recycled Aggregate	Activator	Water	Designed Porosity(%)
A1	245.9	101.1	1768.5	—	240.9	10.8	10.0
A2	245.9	101.1	—	1768.5	240.9	10.8	10.0
A3	179.6	73.4	1510.8	—	175.8	7.9	15.0
A4	134.2	54.8	1412.1	—	131.4	5.8	20.0
B1	245.9	101.1	1768.5	—	215.8	26.0	10.0
B2	245.9	101.1	—	1768.5	215.8	26.0	10.0
B3	179.6	73.4	1510.8	—	157.5	18.9	15.0
B4	134.2	54.8	1412.1	—	117.8	14.1	20.0
C1	209.7	137.3	1768.5	—	250.2	8.7	10.0
C2	209.7	137.3	—	1768.5	250.2	8.7	100
C3	153.0	100.0	1510.8	—	182.7	6.3	15.0
C4	114.3	74.7	1412.1	—	136.8	4.7	20.0

2.2 Experimental Method

2.2.1 Porosity

The porosity was determined using the drainage method and calculated according to Equation (1), with results rounded to 0.1%:

$$P=\left(1-\frac{m_1-m_2}{\rho_w V}\right)\times 100\% \tag{1}$$

Where: m_1 is the dry mass of the specimen (g),m_2 is the mass of the specimen submerged in water (g),V is the volume of the specimen (cm^3), ρw is the density of water (g/cm^3), P is the porosity (%).

2.3.2 Permeability Coefficient

In this study, the permeability of the concrete was evaluated using the variable head permeameter method. Tests were conducted with an initial water head of 150 mm on specimens measuring 100 mm × 100 mm × 100 mm. The permeability coefficient (K) was calculated using Equation (2):

$$K = \frac{H}{T} \qquad (2)$$

Where: H is the water head height (mm),T is the test time interval (s) and K is the permeability coefficient (mm/s).

2.3.3 Compressive Strength and Flexural Tensile Strength

Compressive and flexural strength tests were performed in accordance with the Standard for Test Methods of Concrete Physical and Mechanical Properties (GB/T 50081-2019). The flexural tensile strength of the permeable geopolymer concrete was calculated using Equation

$$f_f = \frac{FL}{bh^2} \qquad (3)$$

Where: ff is the flexural tensile strength of permeable geopolymer concrete (MPa),F is the ultimate load (N), L is the distance between the two points of support for the concrete (mm), b is the side length of the square cross-section of the concrete beam (mm) and h is the height of the specimen (mm).

2.3.4 Freeze–Thaw Resistance

In this study, the freeze-thaw resistance of the concrete is evaluated using the rapid freezing method, referring to the "Standard for Test Methods of Long-term Performance and Durability of Ordinary Concrete " (GB/T50082-2009). The mass loss ratio and strength(ΔW) loss ratio(ΔP) of the specimens are calculated using equations (4) and (5):

$$\Delta W = \frac{W_0 - W_N}{W_0} \times 100 \qquad (4)$$

$$\Delta P = \frac{P_0 - P_N}{P_0} \times 100\% \qquad (5)$$

Where:W0 is the saturated mass of the specimen before freezing (g),WN is the mass of the specimen after N cycles of freezing and thawing (g),ΔW is the mass loss rate (%),P0 is the flexural tensile strength of specimens without freeze-thaw cycles (MPa),PN is the flexural tensile strength of specimens after N cycles of freezing and thawing (MPa) and ΔP is the strength loss rate (%).

3. Results and discussion

3.1 Effects of Carbonation Conditions on Recycled Aggregate Performance

3.1.1 Physical Properties

Owing to the presence of adhered old mortar on the surface and microcracks induced during the crushing process, recycled aggregates demonstrate significantly elevated crushing values and water absorption rates compared to natural aggregates. These inherent deficiencies substantially compromise the mechanical performance and durability of concrete, thereby posing a critical challenge to the large-scale engineering application of recycled aggregates. The carbonation modification effects are illustrated in the figure.

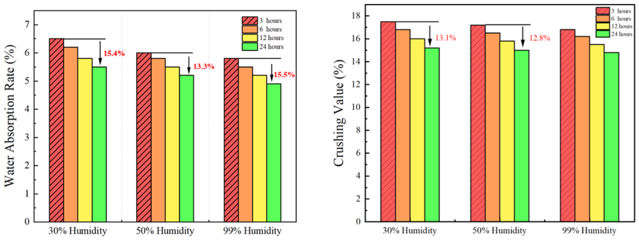


Figure 3.1 Effects of Carbonation on the Physical Properties of Recycled Aggregates

As illustrated in Figure 3.1, under a CO_2 concentration of 99% and relative humidity of 30%, the water absorption of non-carbonated RA measured 6.5%, which decreased to 5.5% following 24 hours of carbonation treatment. This confirms that carbonation effectively densifies RA by precipitating calcium carbonate within pore structures. Concurrently, the crushing value of RA exhibited a gradual decline with extended carbonation duration; under identical CO_2 and humidity conditions, it reduced from 17.5%

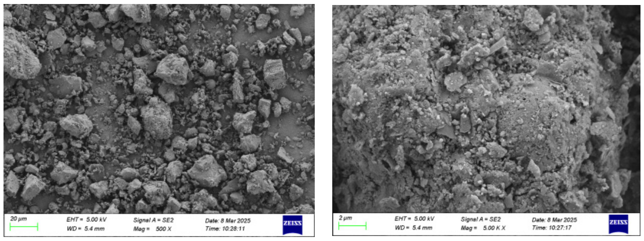
in the non-carbonated state to 15.2% after 24 hours of carbonation. These observations suggest that carbonation not only enhances aggregate densification but also improves compressive strength, likely due to pore refinement and healing of internal microcracks.

Furthermore, experimental data reveal that the water absorption and crushing valueof RA are significantly influenced by ambient humidity during carbonation. At lower humidity levels , carbonation efficiency is limited, evidenced by relatively higher water absorption (5.5% after 24 hours) and crushing values (15.2%). However, increasing humidity to 50% markedly accelerates the carbonation reaction, further reducing water absorption and crushing values. Specifically, at 99% CO_2 concentration, water absorption decreased from 5.5% (30% RH) to 4.9% (70% RH) after 24 hours, while crushing values correspondingly declined from 15.2% to 14.8%. These results underscore the pivotal role of humidity in enhancing carbonation kinetics, thereby facilitating pore structure refinement, aggregate densification, and improved mechanical performance.

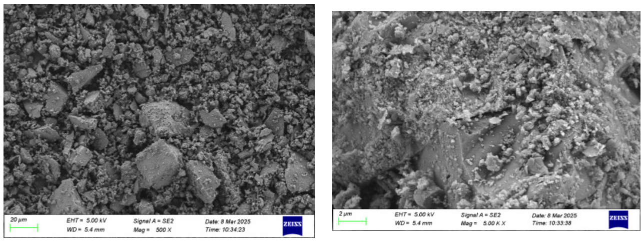
3.1.2 Microstructural Analysis

To further clarify the effects of carbonation on recycled aggregates, the surface morphology of RA before and after carbonation treatment was examined via scanning electron microscopy (SEM), as shown in Figure 3.2.

Comparative analysis of the surface microstructure of recycled aggregates before and after carbonation revealed significant modifications induced by the treatment. Non-carbonated RA exhibited loose adhered mortar debris with intersecting microcrack networks and irregular pores , forming a porous and defective structure that facilitated water penetration and stress concentration, critically contributing to their inferior physical properties. In contrast, after 24-hour carbonation, the RA surface underwent notable densification, where nanometer- to micrometer-sized calcium carbonate ($CaCO_3$) crystals nucleated in situ at the old mortar interfaces. These crystals grew in clustered or layered configurations, tightly filling pre-existing pores and microcracks to form a continuous, dense shell structure.This $CaCO_3$-derived micro-barrier not only sealed intrinsic defects but also enhanced the bonding strength of the interfacial transition zone between old and new mortar via crystal cementation. Under external loading, the densified shell uniformly redistributed stress, suppressing microcrack initiation and propagation. These microstructural improvements provide a mechanistic foundation for the enhanced mechanical performance of carbonated RA-based concrete.



（a）Before carbonation (×500)　（b）After 24-hour carbonation（×5.00k）



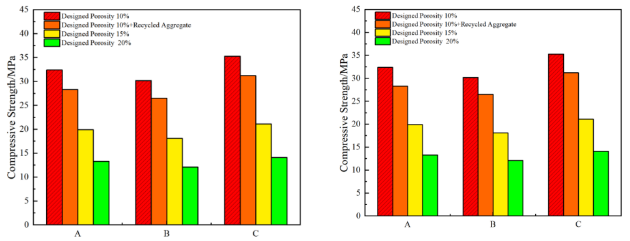
（c）After carbonation（×500）　（d）After 24-hour carbonation（×5.00k）

Figure 3.2 SEM Morphology of Recycled Aggregates Before and After Carbonation

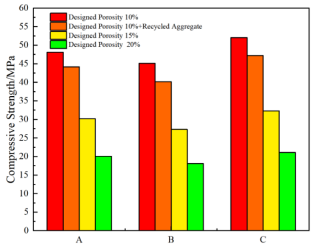
3.2 MecMechanical

3.2.1 Compressive Strength

Compressive strength is a critical indicator for evaluating the load-bearing capacity of concrete, directly affecting its structural safety in practical engineering applications. In this study, the compressive strength of pervious geopolymer concrete at various curing ages was determined through experimental testing. The results are presented in Figure 3.3.



（a）7d Compressive Strength （b）14d Compressive Strength



（c）28d Compressive Strength

Figure 3.3 Experimental Results of Compressive Strength for Permeable Geopolymer Concrete

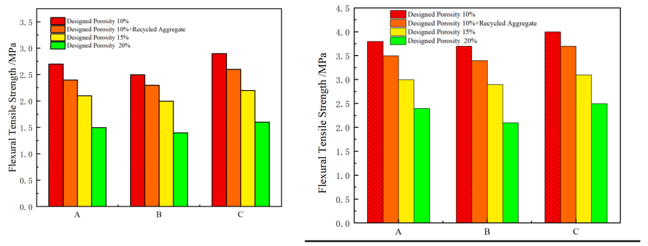
The experimental results demonstrated that Group C specimens exhibited the optimal mechanical performance across all curing ages, with 7-day, 14-day, and 28-day compressive strengths ranging from 35.3 to 52.1 MPa, representing an 8.9%–8.3% increase compared to Group A and a 16.9%–15.5% improvement over Group B. Notably, the optimal mix formulation in Group C (C1) achieved a 28-day strength of 52.1 MPa, surpassing Group A1 and B1 by 8.3% and 15.5%, respectively. Even at a porosity of 15.1%, Group C maintained a 28-day strength of 32.3 MPa, significantly outperforming Groups A and B at equivalent porosity levels, indicating that binder system optimization effectively mitigates strength loss caused by increased porosity.

Natural aggregate specimens consistently exhibited a 2.5–4.3 MPa strength advantageacross all groups. For instance, at 28 days, Group A NA specimens achieved 48.1 MPa, while their recycled aggregate counterparts dropped to 44.2 MPa. This performance gap widened in Group C, with NA specimens reaching 52.1 MPa and RA specimens attaining 47.2 MPa. This disparity may stem from enhanced aggregate-matrix interfacial bonding enabled by the optimized binder system, partially offsetting the detrimental effects of RA surface defects.

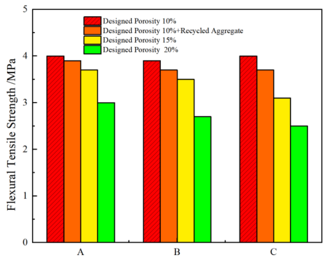
Porosity exerted a significant influence on compressive strength degradation. When porosity increased from 10.2% to 20.7%, the 28-day strength of Group A specimens declined from 48.1 MPa to 18.1 MPa , corresponding to a decay rate of 1.5 MPa per 1% porosity increase. In contrast, Group C exhibited a lower decay rate of 1.1 MPa per 1% porosity increase, reducing the strength loss by 26.7% compared to Group A. Notably, when porosity exceeded 15%, all groups experienced a 30%–50% acceleration in strength decay rates. For example, Group B specimens at 15.3% porosity showed strengths below 27.3 MPa, while Group C specimens at the same porosity retained significantly higher performance.

3.2.2 Flexural Tensile Strength

Flexural tensile strength, as the core mechanical index for measuring the ability of concrete to resist bending loads, directly determines its applicability in pavement and bridge deck..The test results are shown in Figure 3.4.。



（a）7d flexural tensile strength （b）14d flexural tensile strength



（c）28d flexural tensile strength

Figure 3.4 Results of Flexural Tensile Strength of Permeable Geopolymer Concrete

The flexural tensile strength of pervious geopolymer concrete exhibited a developmental trend similar to that of compressive strength, as illustrated in the figure. At 28 days, Group C specimens achieved the highest flexural strength (up to 4.3 MPa), slightly outperforming Groups A and B. Specifically, the optimal mix in Group C (C1) reached a 28-day flexural strength of 4.3 MPa, which was higher than Group A and B reference specimens, demonstrating the advantage of the optimized binder. Even at 15.1% porosity, Group C maintained a strength of 3.3 MPa, confirming that the binder optimization mitigates porosity-related losses.

The type of aggregate also had a clear influence. Natural aggregate specimens consistently outperformed recycled aggregate ones by 0.1–0.5 MPa. For example, the flexural strength of the natural aggregate specimen in Group A was 4.1 MPa, compared with 3.7 MPa when recycled aggregates were used. Although this gap was smaller in Group C, natural aggregates still showed about 10% higher strength, mainly due to adhered mortar on recycled aggregates that promotes microcrack propagation. The optimized binder in Group C, with a denser [Si–O–Al] gel network, partially offset these deficiencies.

Flexural strength was also sensitive to porosity. As porosity increased from 10% to 20%, Group A strength fell nearly 50%, while Group C showed a slower decline. Notably, once porosity exceeded 15%, all groups exhibited accelerated strength loss. Group B dropped to 3 MPa at 15.3% porosity, whereas Group C maintained 81.8% of its baseline strength, further confirming the role of binder optimization in improving failure resistance.

3.3 Frost Resistance Durability of Geopolymer Concrete

3.3.1 Analysis of Mass Loss

Under the action of freeze-thaw cycles, the mass loss of geopolymer concrete shows a significant phased evolution law. The specific test results are shown in Figure 3.5.

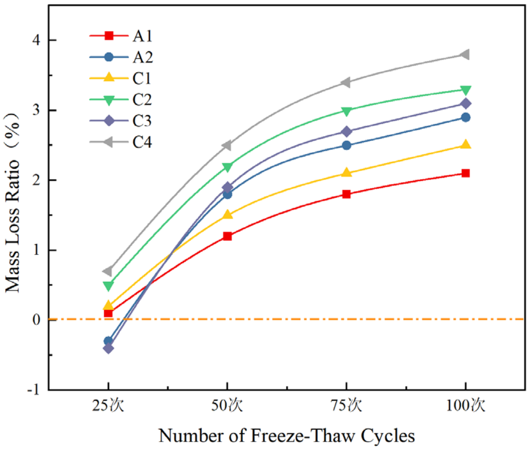


Figure 3.5 Mass Loss Rate of Geopolymer Concrete After 100 Freeze-Thaw Cycles Under Different Porosity Conditions

The data shows that the damage development of specimens with different mix proportions is synergistically influenced by pore structure, aggregate type, and the cementitious system. After 100 freeze-thaw cycles, the mass loss rate of Group C1 is 2.6%, while that of Group C4 increases to 4.9%. The deterioration of frost resistance in the recycled aggregate system is more significant: the mass loss rate of Group A2 (recycled aggregate) after 100 cycles reaches 3.0%, which is 30% higher than that of Group A1 (natural aggregate) with the same porosity.

In the early stage of freeze-thaw (within 25 cycles), some specimens show positive mass growth due to pore water absorption. The closed-pore structure gradually forms a connected network under repeated phase-change stress. At this stage, the expansion of water migration channels enhances capillary adsorption, and the resulting mass gain can even partially offset the mass loss caused by surface micro-cracks, macroscopically presenting as almost unchanged total specimen mass. The water absorption compensation of recycled aggregate Group A2 at 25 cycles is 0.5%, 150% higher than that of natural aggregate Group A1, which is closely related to the capillary adsorption of micro-cracks inside recycled aggregate.

When the number of freeze-thaw cycles exceeds 50, damage-induced mass loss gradually dominates the mass change. The loss rate of recycled aggregate Group A2 reaches 1.2% at 50 cycles, 50% higher than that of natural aggregate Group A1, indicating that the weakness of the interfacial transition zone exacerbates early-stage damage. Before 75 cycles, the loss rate of Group C1 is 0.7% per 25 cycles, and it decreases to 0.4% per 25 cycles in the later stage. The established regression model shows that every 1% increase in porosity leads to a 0.21% increase in the mass loss rate after 100 cycles, and the introduction of recycled aggregate increases the loss rate by 0.15%. Their synergistic effect causes the mass loss rate of Group C2 after 100 cycles to reach 3.5%. The damage evolution of high-porosity specimens exhibits non-linear characteristics: the loss rate of Group C4 surges to 1.6% per 25 cycles after 50 cycles. In contrast, the loss rate of Group A1 stabilizes at 0.4% per 25 cycles after 75 cycles.

3.3.2 Analysis of Compressive Strength Loss

Under the action of freeze-thaw cycles, the evolution of the internal pore structure of the material and the stability of the cementitious phase directly determine the service life of the project. The test results are shown in Figure 3.6.

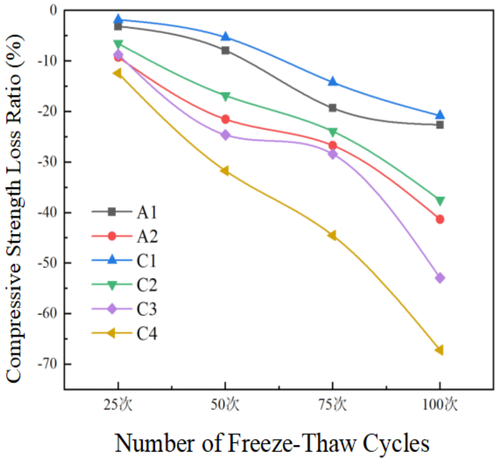


Figure 3.6 Compressive Strength Loss Rate of Geopolymer Concrete at 28 Days After 100 Freeze-Thaw Cycles Under Different Porosity Conditions

As shown in the figure, under the same porosity condition, the compressive strength loss rate of recycled aggregate specimens is significantly higher than that of natural aggregate specimens, and the gap widens with increasing freeze–thaw cycles. In the first 25 cycles, the loss rate was small, but after 50 cycles, recycled aggregate specimens deteriorated more rapidly, reflecting their larger porosity and weaker interfacial transition zones, which promote microcrack generation and propagation.

Higher porosity further aggravated frost damage. When porosity increased from 10% to 20%, both Groups A and C showed significantly higher loss rates. Low-porosity groups (A1, C1) exhibited slow degradation, while high-porosity C4 suffered sharp deterioration after 50 cycles, reaching 67.2% loss by 100 cycles. This confirms that increased porosity accelerates freeze–thaw damage by providing more channels for water penetration and expansion.

After adjusting the slag-fly ash ratio, group C specimens exhibited better frost resistance compared to group A. Particularly under low-porosity conditions, the compressive strength loss rate of group C1 was significantly lower than that of group A1, and the loss rate was relatively slow during freeze-thaw cycles, indicating that optimized mix proportions can improve matrix density, reduce internal defects, and effectively delay the accumulation of freeze-thaw damage. Under high-porosity conditions, although the loss rate of group C4 was higher than other groups, it still showed certain frost resistance advantages compared to group A4, suggesting that reasonable geopolymer mix proportions play a positive role in improving the frost resistance of specimens. In summary, reasonable control of porosity, selection of appropriate aggregate types, and optimization of slag-fly ash ratios are all helpful for improving the frost resistance of specimens and effectively delaying the compressive strength loss caused by freeze-thaw cycles.

Conclusions

This study systematically explored the carbonation modification of recycled aggregates and its synergistic effect with geopolymer binders in the development of high-performance permeable concrete. The main findings are as follows:

1.Carbonation treatment effectively enhances the quality of recycled aggregates, significantly reducing water absorption and improving mechanical and physical properties through the formation of dense CaCO□ phases, as confirmed by SEM analysis.

2.The mechanical performance of geopolymer permeable concrete is governed by binder composition, porosity, and aggregate type. The optimized binder system demonstrated superior strength development and better resistance to strength loss at higher porosities, highlighting the importance of tailored binder design.

3.Freeze–thaw resistance is significantly influenced by aggregate type and porosity, with recycled aggregate concrete showing higher deterioration. The use of optimized slag–fly ash binder systems markedly improved durability, with Group C1 exhibiting the best performance under cyclic freezing conditions.

These findings provide a theoretical basis and experimental support for the design of durable, low-carbon, and high-strength permeable geopolymer concrete materials. However, regarding the engineering application and industrialization of carbonation modification technology, while laboratory-scale treatment demonstrates significant efficacy, practical implementation still faces challenges in CO_2 source availability, energy consumption costs, and process control. Further research is needed to optimize large-scale carbonation processes for economic viability and industrial adoption.

Acknowledgments

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