

Article

Towards Carbon-Negative Concrete Using Low-Carbon Binders and Carbonated Recycled Aggregates: MAA-Based Mix Design Optimization, Carbon Emission and Cost Assessment

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Abstract

Developing low-carbon building materials is essential for achieving sustainability in the construction sector. This study proposes a carbon-negative concrete (CNC) system that combines low-carbon binders derived from industrial by-products with carbonated recycled aggregates capable of CO₂ absorption. To enhance particle packing and mechanical performance, the Modified Andreasen–Andersen (MAA) model was adopted for mix design optimization and experimentally validated. The optimized CNC mixture containing 22% coarse aggregate achieved the minimum residual sum of squares between the graded particle distribution and the theoretical MAA curve, as well as the highest strength performance. Compared with a 14% coarse aggregate mixture, the 22% mix exhibited 13.5% and 19.8% increases in compressive strength at 7 and 28 days, confirming the applicability of the MAA model for CNC proportioning. Carbon emission assessment, limited to raw material production, demonstrated significant environmental benefits. CNC incorporating both low-carbon binders and carbonated recycled aggregates reduced total emissions and CO₂ intensity by 87.1% and 86.2%, respectively, compared with ordinary concrete of the same strength grade. Economic evaluation further showed that CNC reduced material cost by 48.1% relative to ordinary concrete. It should be emphasized that the reported CO₂ reduction and negative emission effects are limited to the defined raw material production boundary and do not represent a fully net-negative life cycle. Overall, these results confirm the technical, environmental, and economic feasibility of CNC as a sustainable alternative to traditional concrete.



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Keywords: carbon-negative concrete; low-carbon binders; carbonated recycled aggregates; mix design optimization; carbon emissions

1. Introduction

The global construction sector is under increasing pressure to reduce carbon emissions while sustaining the mechanical performance and durability required for engineering applications. As the most widely used construction material, conventional concrete contributes substantially to the embodied carbon of infrastructure systems, primarily due to

the production of ordinary Portland cement (OPC) and the extensive extraction of natural aggregates. In China, emissions from concrete and its upstream cement industry represent about 25~30% of national totals [1,2]. Driven by carbon neutrality targets and the urgent need for sustainable construction, the development of carbon-negative concrete (CNC), a new class of concrete capable of achieving net CO₂ reduction through material innovation and optimized mixture design, has become a research frontier with significant practical potential [3].

A promising pathway for CNC development lies in the combined use of low-carbon binders [4–9] and carbonated recycled aggregates (CRAs) [10,11]. Low-carbon binders, such as industrial by-product-based binders, can significantly reduce cement-related emissions, while CRAs derived from waste concrete can effectively sequester CO₂ through mineral carbonation, further lowering the carbon footprint of the final material. Compared with natural aggregates, CRAs not only provide carbon storage benefits but also help reduce environmental burdens related to aggregate extraction and waste disposal. Integrating these two strategies offers a holistic approach to developing CNC that meets both engineering performance requirements and environmental sustainability goals [12,13].

In addition to materials innovation, the mixture design methodology plays a crucial role in achieving the desired performance–carbon–cost balance. Classical mixture design models such as Feret’s packing theory [14] and the Fuller gradation curve [15,16] have historically guided aggregate optimization. However, these models were originally developed for relatively simple aggregate systems and are primarily applicable to concrete composed of natural aggregates and single-binder matrices. Their underlying assumptions limit their adaptability when extended to modern multi-material concrete systems.

In CNC, the material system typically consists of multiple solid constituents, including low-carbon binders derived from industrial by-products, recycled aggregates with high porosity and water absorption, and carbonated aggregates with altered surface characteristics. In such systems, significant differences in particle size distribution, morphology, and physical properties coexist, making it difficult for classical models to accurately represent the overall packing behavior. Moreover, traditional models offer limited flexibility in independently adjusting the volumetric contribution of each constituent, which constrains their effectiveness in optimizing complex multi-phase mixtures.

In contrast, the Modified Andreasen–Andersen (MAA) model provides a more versatile framework for mix design optimization by allowing volumetric weighting of individual components and continuous control of the target particle size distribution. As shown in Equation (1) [17]. This feature makes the MAA model particularly suitable for multi-component systems with diverse material characteristics, such as carbon-negative concrete incorporating low-carbon binders and carbonated recycled aggregates. Nevertheless, the application of the MAA model to carbon-negative concrete remains limited, and its effectiveness in simultaneously optimizing mechanical performance, carbon emission reduction, and material efficiency has not been sufficiently explored.

$$P(D) = 100 \left(\frac{D^q - D_{\min}^q}{D_{\max}^q - D_{\min}^q} \right) \quad (1)$$

where $P(D)$ is the cumulative passing percentage of particles smaller than size D ; D is the particle size under consideration; $D_{\min}$ represents the minimum particle size; $D_{\max}$ represents the maximum particle size; q is the distribution modulus, typically ranging from 0.21 to 0.37. A smaller q value indicates a higher proportion of fine particles in the system, whereas a larger q value corresponds to a greater proportion of coarse particles.

Recent experimental studies [17] have shown that concretes designed using the MAA model exhibit significantly higher mechanical performance and allow for an approximately

10% reduction in binder content compared with traditional methods, reflecting both engineering and environmental advantages.

Given these benefits, the present study adopts the MAA model as the primary theoretical basis for CNC mixture design. By integrating low-carbon binders, carbonated recycled aggregates, and an optimized particle packing approach, this research aims to develop CNC mixtures that achieve superior mechanical performance while substantially reducing both CO₂ emissions and material cost.

Building on these considerations, this study develops a comprehensive framework for designing CNC by integrating low-carbon binders, CRAs, and particle packing optimization based on the MAA model. Through this approach, the research aims to enhance packing density, reduce binder demand, and improve mechanical performance, while simultaneously lowering carbon emissions and overall material costs. By systematically examining the mechanical behavior, carbon footprint, and economic implications of the proposed mixtures, this work provides a scientifically grounded pathway for advancing CNC technologies and supports their practical transition from experimental development to real-world engineering applications.

2. Materials and Test Methods

2.1. Raw Materials

2.1.1. Low-Carbon Binders

The raw materials used in this study were blast furnace slag (BFS) and silica fume (SF), both purchased from Longze Water Purification Materials Co., Ltd., Gongyi, China. The BFS selected was of S105 grade, with a white color and a density of 2.93 g/cm³; its physical properties are listed in Table 1. The SF appeared bluish-gray and contained 97.37% SiO₂, with its physical properties provided in Table 2. The chemical compositions of BFS and SF are shown in Table 3. The XRD results (Figure 1) indicate that both BFS and SF are primarily amorphous, with BFS also containing minor amounts of CaCO₃ and akermanite (Ca₂MgSi₂O₇) [18,19]. The particle size distributions of BFS and SF are shown in Figure 2, with mean particle diameters of 12 μm and 0.2 μm, respectively.

Table 1. Physical properties of BFS.

Test Items (%)	Flowability Ratio	7-Day Activity Index	28-Day Activity Index	Loss on Ignition	Moisture Content
Standard values	≥95	≥95	≥105	≤1.0	≤1.0
Measured value	102	98	109	0.96	0.4

Table 2. Physical properties of SF.

Test Items (%)	Total Alkali Content	7-Day Activity Index	Chlorine Content	Water Demand Ratio	Loss on Ignition
Standard values	≤1.5	≥105	≤0.1	≤125	≤4.0
Measured value	0.6	128	0.01	106	2.6

Table 3. Chemical compositions of SF and BFS.

Oxide (wt. %)	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	MgO	P ₂ O ₅	SO ₃	Na ₂ O	K ₂ O	TiO ₂	Others
SF	97.37	0.20	0.57	0.04	0.51	0.12	0.82	0.06	0.29	-	0.01
BFS	28.12	15.26	42.4	0.46	8.37	0.02	2.50	0.52	0.41	1.23	0.69

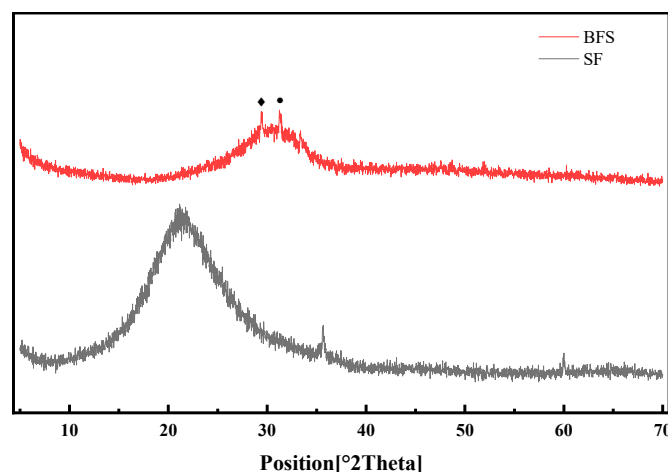


Figure 1. XRD patterns of SF and BFS. (♦, Akermanite; •, CaCO_3).

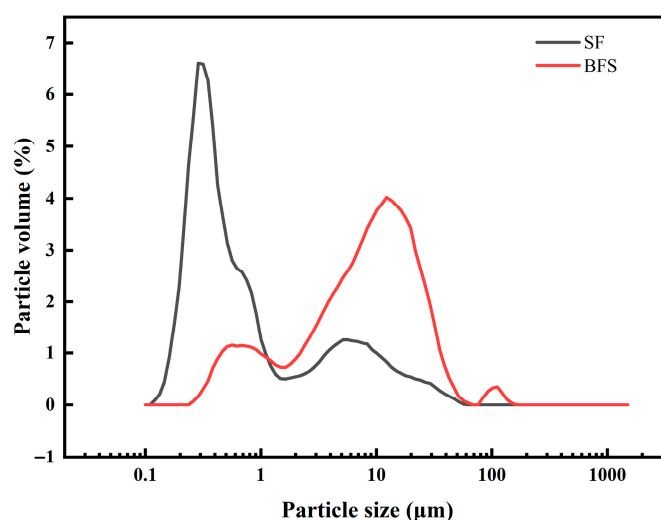


Figure 2. Particle distribution of SF and BFS.

Calcium oxide (CaO) was used as the alkaline activator, obtained from Yingge Mining Co., Ltd., Fenyi County, China. It was in powdered form with a fineness of 150~200 mesh. The chemical activators selected were calcium sulfate (CaSO_4) and calcium chloride (CaCl_2). The CaSO_4 , of superior purity (>99%), contained $\leq 0.1\%$ alkali and metal impurities and was purchased from Damao Chemical Reagent Factory, Tianjin, China. The CaCl_2 , of analytical grade (>96% purity) with a pH range of 8.0~10.0, was obtained from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. In the experiments, CaCl_2 was used in the form of white solid granules.

2.1.2. Fine Aggregates

The fine aggregate used in this study was carbonated recycled fine aggregate (CRFA), as shown in Figure 3a. CRFA produced from demolished concrete structures collected in Shanghai, China, and processed at a local construction waste recycling facility. The carbonation treatment was conducted using a carbonation chamber manufactured by Cangzhou Dongyi Rongke Testing Instrument Co., Ltd., Cangzhou, China. The cumulative sieve residue of the CRFA, measured experimentally, is listed in Table 4. CRFA was produced by subjecting recycled fine aggregate (RFA) to a controlled carbonation treatment. Specifically, the RFA was first dried in an oven at $60 \pm 2^\circ\text{C}$ and then placed in a carbonation chamber under ambient pressure, with a relative humidity of $75 \pm 5\%$ and a CO_2 concentration of $20 \pm 2\%$. The carbonation treatment was maintained for more than 14 days to ensure

sufficient carbonation of the recycled aggregates [20]. In this study, the carbonation degree of recycled aggregates was qualitatively verified using phenolphthalein testing, ensuring complete carbonation of the treated aggregates.

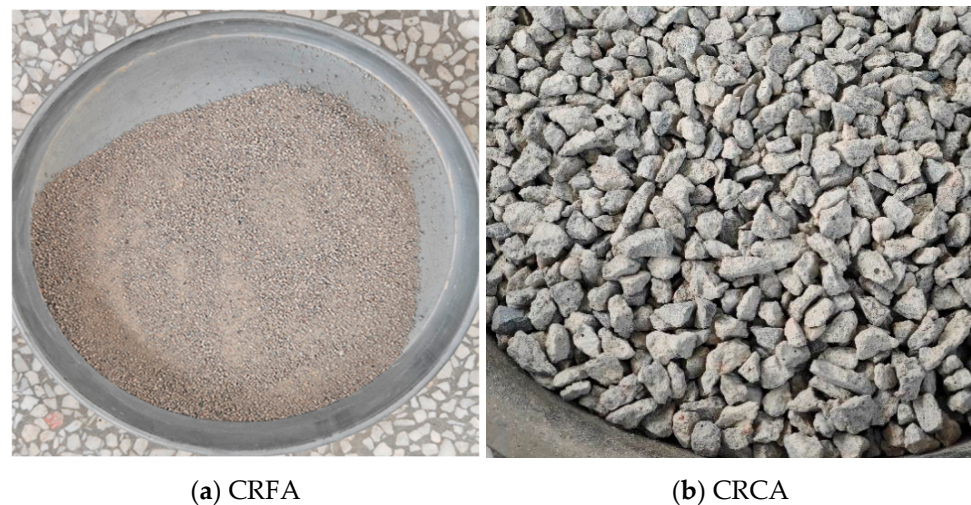


Figure 3. Carbonized recycled aggregate.

Table 4. Cumulative sieve residue of CRFA.

Sieve Size (mm)	0	0.15	0.3	0.6	1.18	2.36	4.75
CRFA	15.7	34.8	52.3	84	99.3	99.79	99.94

The fine aggregate meets the requirements of GB/T 14684-2022 [21], and its basic properties are summarized in Table 5. As reported by Kaliyavaradhan [22], carbonation increases the apparent density of recycled aggregates and reduces their water absorption. This is attributed to the formation of calcium carbonate, which fills part of the pores on the aggregate surface.

Table 5. Physical properties of CRFA.

Aggregate	Fineness Modulus	Crushing Value (%)	Apparent Density (kg/m ³)	Water Absorption (%)	Moisture Content (%)
CRFA	3.4	35.75	2301	10.94	5.37

2.1.3. Coarse Aggregates

The recycled coarse aggregate (RCA) used in this study was crushed from waste concrete obtained from demolished concrete structures in Shanghai. The original concrete was made with ordinary Portland cement and natural aggregates. After demolition, the waste concrete was mechanically crushed, sieved, and subsequently subjected to a controlled carbonation treatment to produce carbonated recycled coarse aggregate (CRCA), as shown in Figure 3b. Both CRCA and CRFA were processed in the same facility and underwent identical treatment procedures. The cumulative sieve residue of CRCA, measured experimentally, is presented in Table 6, and its basic properties are summarized in Table 7.

Table 6. Cumulative sieve residue of the CRCA.

Sieve Size (mm)	2.36	4.75	9.5	16	19	26.5	31.5
CRCA	50	99	100	100	100	100	100

Table 7. Physical properties of CRCA.

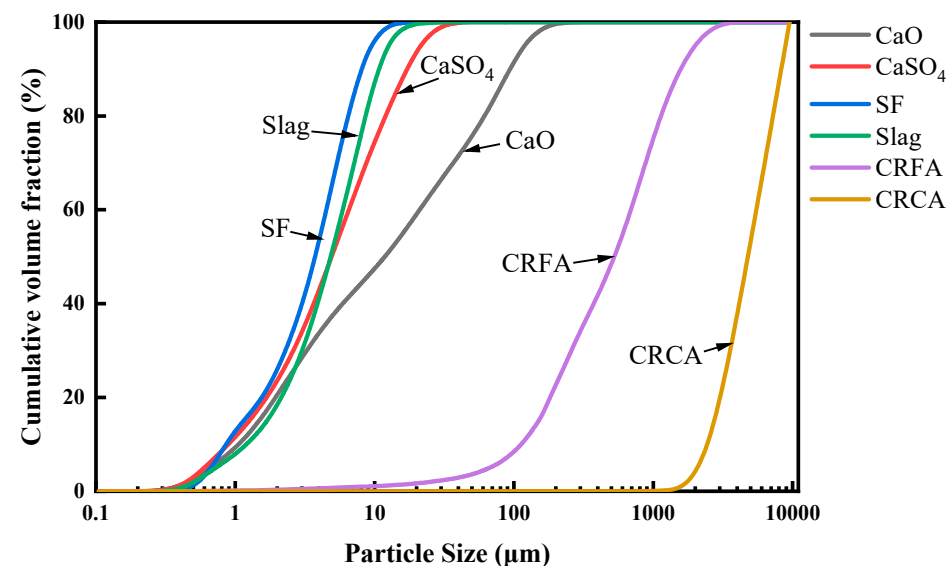
Aggregate	Crushing Value (%)	Apparent Density (kg/m ³)	Water Absorption (%)	Moisture Content (%)
CRCA	12.3	2667	5.09	3.07

2.1.4. Water and Chemical Additives

Tap water was used for mixing, and a polycarboxylate-based high-performance water-reducing admixture, conforming to GB 8076-2008 [23], was employed. The admixture serves to disperse agglomerated binder particles, release entrapped free water, and thereby reduce the total mixing water required.

2.2. Raw Material Particle Size Distribution and Mixing Ratio

Combined with the cumulative sieve residue results of CRFA and CRCA, the particle size distributions of all solid raw materials are presented in Figure 4. Since CaCl_2 was fully dissolved in water prior to mixing, its particle size distribution was not considered in the particle size analysis. Nevertheless, the dissolved Ca^{2+} and Cl^- ions inherently participate in the hydration and reaction processes of the concrete system.

**Figure 4.** Particle size distribution of raw materials.

Previous studies have shown that a lower porosity in the concrete system indicates higher packing density, which in turn improves mechanical performance. To verify whether the mixture optimized based on the MAA model is indeed optimal, mix proportions were first designed for a CNM matrix containing only binder and fine aggregate, following existing research and the JGJ 55-2011 [24] concrete design code. On this basis, a small amount of CRCA was used to replace part of the paste volume, and the mixture was recalculated accordingly. To evaluate the significance of the optimized mix, CRCA replacement levels of 14%, 18%, 22%, and 26% were designed, with 22% representing the optimized proportion obtained from the MAA model. The detailed CNC mix proportions are listed in Table 8. Additionally, the mixing curves of each formulation were compared with the MAA model curve by calculating the residual sum of squares (RSS) values.

Table 8. Mix proportions of CNC.

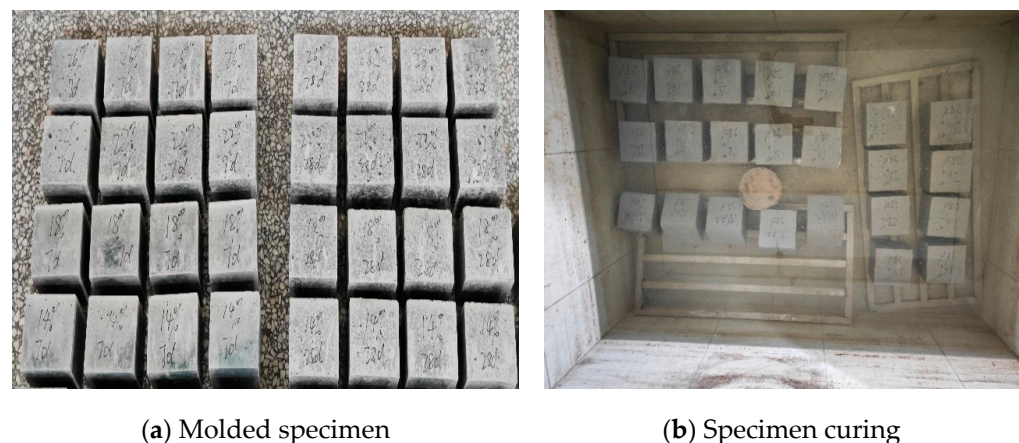
CRCA Dosage	Water/Binder Ratio	Sand/Binder Ratio	CRFA	CRCA	Water	SP	RSS
CA14%	0.4	1.5	918	373	244.8	1%	2734
CA18%	0.4	1.5	875	480	233.6	1%	1658
CA22%	0.4	1.5	833	587	222	1%	1027
CA26%	0.4	1.5	790	693	210.8	1%	3226

2.3. Specimen Preparation and Molding

Before mixing, both CRFA and CRCA were conditioned to a surface-saturated dry (SSD) state, ensuring that their water absorption did not adversely affect the effective water-to-binder ratio. This procedure helped maintain stable workability across all mixtures without requiring additional mixing water. To ensure high-quality sample preparation, a two-stage mixing approach (TSMA) was employed for concrete batching. The TSMA was selected instead of conventional one-step mixing to improve mixture homogeneity and particle dispersion in CNC. This method is particularly beneficial for systems incorporating CRFA and CRCA, as the initial addition of part of the mixing water allows aggregates to be pre-wetted, thereby mitigating excessive water absorption and improving the subsequent coating of binders on aggregate surfaces. As a result, a more uniform microstructure and stable fresh mixture can be achieved, which is advantageous for the mechanical performance of CNC.

A key feature of the TSMA method is that the mixing water is divided and added in two separate stages. The procedure was as follows: initially, all aggregates were placed into the mixer and stirred for 60 s; next, approximately half of the total mixing water was added and the mixture was stirred for an additional 60 s; then, the binder were introduced and mixed for 30 s; finally, the remaining mixing water was added, and the mixture was stirred for 120 s. It should be noted that a polycarboxylate-based high-performance water-reducing admixture was used in all mixtures, and the water-to-binder ratio was kept constant. As a result, all mixtures exhibited acceptable workability for laboratory casting without segregation or bleeding.

Prior to casting, a release agent was applied to the inner surfaces of the molds to facilitate demolding. The prepared mixture was then poured into molds measuring $100 \times 100 \times 100$ mm, and the surfaces were leveled using a trowel. The specimens were initially cured in a standard curing chamber maintained at 20°C and relative humidity above 95% for 24 h, after which they were demolded. Following demolding, the specimens were immersed in water (maintained at $20 \pm 1^\circ\text{C}$) for continued curing, as shown in Figure 5.

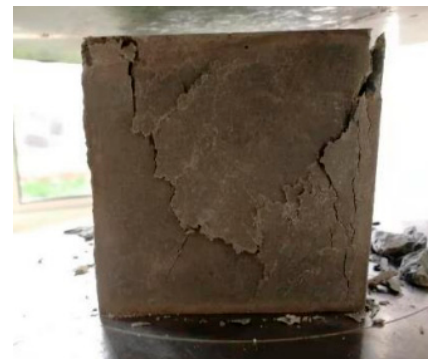
**Figure 5.** Molded specimen and specimen curing.

2.4. Compressive Strength Test

The compressive strength tests of CNC were conducted strictly in accordance with GB/T 50081-2019 [25]. A WAW-2000 universal testing machine (Figure 6a), manufactured by Jinan Xinguang Testing Machine Manufacturing Co., Ltd., Jinan, China, was used for the experiments. The procedure was as follows: specimens were first removed from the curing chamber, and surface water was wiped off with a dry towel. Each specimen was then carefully positioned at the center of the lower platen of the testing machine. The hydraulic pump was activated, the loading rate was set to 0.5 MPa/s, and the machine's reading was reset to zero. The test was initiated by pressing the start button, and loading continued until the specimen developed large cracks indicative of imminent failure, as illustrated in Figure 6b. At this point, loading was stopped, and the failure data were recorded in detail. It should be noted that the specimens used were cubic with a side length of 100 mm, which are non-standard sizes; therefore, the measured strength values were multiplied by a size conversion factor of 0.95 to obtain the corrected compressive strength [25].



(a) Hydraulic pressure testing machine



(b) Specimen failure diagram

Figure 6. Concrete testing instruments and failure diagrams.

3. Experimental Results and Discussion

3.1. Mix Design Based on the MAA Model

Based on the particle size distribution curves of the binder and aggregates, the CNC mix design method can be summarized as follows: starting from several known distribution functions, these functions are used to optimize and fit a target function. During the fitting process, the proportions of the individual distribution functions are continuously adjusted until the fitted curve closely matches, or coincides with, the target function. Once the optimal proportions are determined, they define the final CNC mix.

This optimization process involves three key components: the target values, the adjustable variables, and the constraints. The target values define the desired outcome of the optimization; the adjustable variables allow the proportions of each distribution function to be modified to achieve better fitting; and the constraints ensure that the resulting CNC mix is practical and feasible for engineering applications.

Target values: The primary objective of CNC mix optimization is to achieve an ideal particle packing of the multicomponent system, which requires the particle size distribution of the mixture to approximate the theoretical maximum packing model, as expressed in Equation (1). Thus, the mix design problem becomes a curve-fitting problem between the particle size distribution of the solid mixture, $P_i(D_i)$, and the target MAA model curve, $P_m(D_i)$. The key in this optimization is to minimize RSS between the observed values $P_i(D_i)$ and the theoretical model $P_m(D_i)$. Based on the least squares principle, the objective function quantifies the deviation between the two datasets. Specifically, the constraints

are formulated as in Equation (2), and optimal fitting is achieved by minimizing the RSS between the observed and predicted values:

$$RSS = \sum_{i=1}^i [P_i(D_i) - P_m(D_i)]^2 \quad (2)$$

where $P_i(D_i)$ is the cumulative passing percentage of the mixture at particle size D_i , and $P_m(D_i)$ is the cumulative passing percentage of the MAA target curve at D_i .

Adjustable variables: To solve the mix design problem, the particle size distributions of all solid raw materials were measured, combined with information on the volumetric proportions of each material in 1 m³ of concrete. Suppose the concrete contains n solid materials, including CaO, CaSO₄, SF, BFS, CRFA, and CRCA. Let m denote the number of solid components considered in the MAA calculation, with a total volume V_s (m³). The volume of each material k is V_k (m³, $k = 1, 2, \dots, m$), and its cumulative passing percentage at the particle size D_i is $P_k(D_i)$. The volume fraction of each material k in the solid mixture is expressed by Equation (3), and the cumulative passing percentage of the total mixture at D_i , $P_m(D_i)$, is calculated as the weighted sum of each material's passing percentage according to its volume fraction (Equation (4)).

$$V_k = \frac{V_k}{\sum_{m=1}^m V_k} = \frac{V_k}{V_s} \quad (3)$$

$$P_m(D_i) = \frac{\sum_{m=1}^m P_k(D_i) \bullet V_k}{\sum_{m=1}^m P_k(D_i)} = \sum_{m=1}^m V_k \bullet P_k(D_i) \quad (4)$$

Constraints: According to Brouwer's particle packing theory [26], the distribution modulus n in the maximum packing criterion is adjustable. Previous studies indicate that a higher modulus ($n \geq 0.5$) is suitable for systems with a wide aggregate size range, whereas systems with a higher fine aggregate content favor a lower modulus ($n \leq 0.25$). Experimental and model results show that a distribution modulus n in the range of 0.22~0.28 yields a packing state close to the maximum density [27]. Hunger [28] further suggested that, for self-compacting concrete, a packing parameter q between 0.22 and 0.25 provides optimal performance. Considering the relatively small maximum coarse aggregate size and the high overall density of the concrete in this study, a packing parameter of $q = 0.23$ was selected based on previous research [17,29].

Based on preliminary experiments, the binders were blended according to the designed proportions to obtain the particle size distribution of the powder fraction P . In practice, when the coarse aggregate volume fraction is below 0.4, concrete may exhibit insufficient stability. Therefore, in this study, the volumetric proportions of powder P , CRFA, and CRCA were adjusted to simultaneously satisfy the principle of maximum packing density and ensure adequate workability and practical applicability. Considering all these factors, the constraints for the system parameters can be summarized as follows:

$$0.151 < V_p < 0.226 \quad (5)$$

$$0.278 < V_{\text{CRFA}} < 0.417 \quad (6)$$

$$0.1 < V_{\text{CRCA}} < 0.4 \quad (7)$$

During the solution process, parameter optimization was performed using MATLAB (Version R2024a) and Solver tools in Excel. Following the procedure described above, the

optimized fitting curve was obtained, as shown in Figure 7, and the corresponding CNC mix proportions calculated based on the MAA model are presented in Table 9. Analysis of the results indicates that, compared with the theoretical optimal particle size distribution curve of the MAA model, the fitted curve exhibits only minor fluctuations around the target, while the overall trend aligns closely with the theoretical curve. This demonstrates a high degree of agreement and correlation between the experimental fitting and the theoretical model.

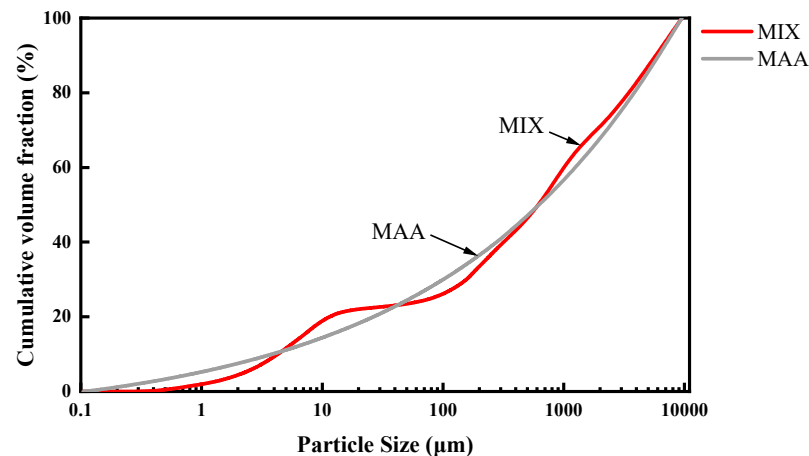


Figure 7. Fitting curves of the optimization solution.

Table 9. Mix proportions optimized based on the MAA model.

Materials	Slag	CaSO ₄	CaCl ₂	CaO	SF	W	CRFA	CRCA
Volume (m ³)	0.142	0.016	0.013	0.013	0.013	0.222	0.362	0.22
Mass (kg)	416.3	41.6	27.8	41.6	27.8	222	832.6	586.7

3.2. Design Mix Proportion Strength Verification

The compressive strength results of CNC with varying CRCA replacement levels at 7 and 28 days are shown in Figure 8. As observed, the compressive strength initially increases and then decreases with increasing CRCA content. At CRCA volume fractions of 14% and 18%, the strength differences are minimal. When the CRCA content is increased to 22%, a significant enhancement in compressive strength is observed, with 7-day strength increasing by 13.5% and 28-day strength by 19.8% compared to the 14% CRCA mixture. This improvement is mainly attributed to the formation of a stable, rigid skeleton structure by an appropriate amount of CRCA, which enhances the interfacial bonding and mechanical interlock, effectively impeding crack propagation and improving overall mechanical performance [30]. However, when CRCA content exceeds the optimal level, the continuous granular skeleton becomes disrupted, leading to packing inefficiency, increased voids, and stress concentration. This packing failure is reflected by the increased RSS value and the corresponding reduction in compressive strength observed for CA26% [31]. Considering compressive strength comprehensively, the optimal CRCA volume fraction is approximately 22%.

In addition, RSS was used to quantify the closeness between the particle size distribution curve of the mixture and the theoretical MAA target curve. A minimum RSS indicates the most optimal packing density of the system. As shown in Figure 8, under different mix proportions, the RSS value exhibits a clear inverse relationship with compressive strength. With increasing coarse aggregate content, the RSS initially decreases and then increases, a trend that is more pronounced in Figure 8b. The RSS data listed in Table 8 show the same pattern, further validating this relationship.

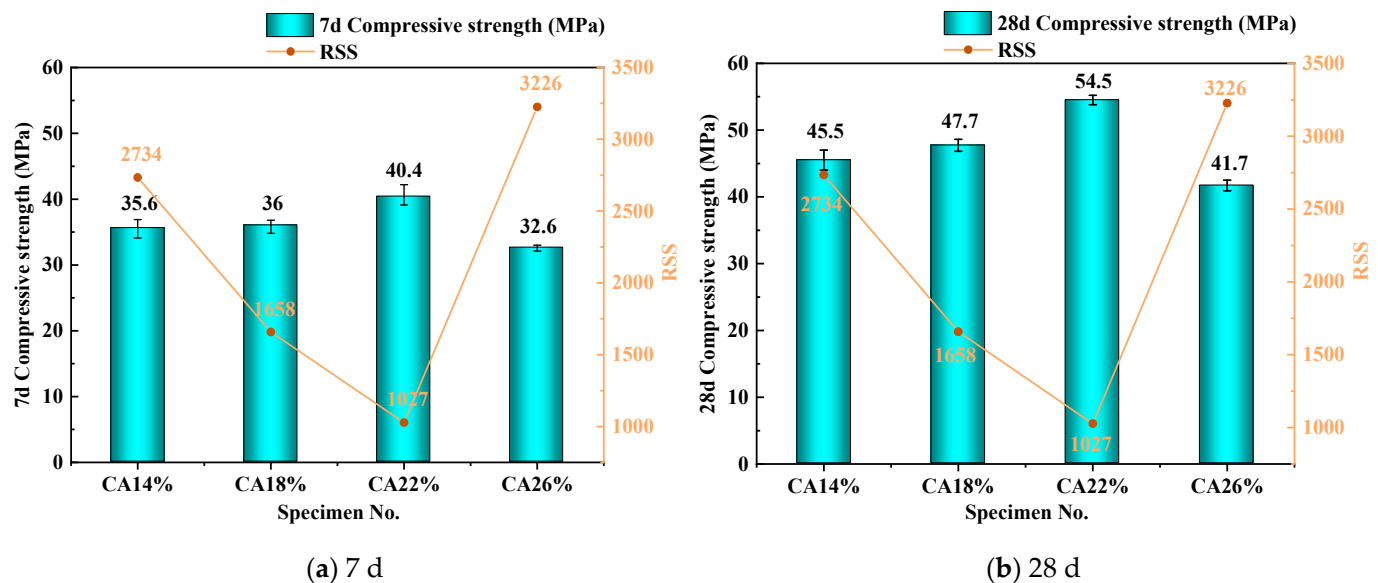


Figure 8. CNC compressive strength and RSS.

3.3. Carbon Emission Analysis

When calculating carbon emissions, it is essential to define the system boundary appropriately based on the research objectives. Life cycle carbon assessment (LCA) typically adopts a “cradle-to-grave” approach, covering all stages from raw material extraction, processing, and transportation, to concrete mixing, placement, service life, and eventual demolition. However, due to the limited experimental timeframe and practical constraints on raw material procurement in this study, a full life cycle assessment is not applicable.

Therefore, to facilitate comparison and ensure computational feasibility, the system boundary for carbon emission accounting in this study is limited to the raw material production stage. Carbon emission factors for different materials were obtained from a comprehensive review of relevant literature and standards, and are summarized in Table 10. For carbonated recycled aggregates, the calculated emissions include the processes of crushing and sorting the recycled aggregates, CO₂ collection, and CO₂ absorption. Representative experimental mixtures were then compared with ordinary concrete of the same strength grade to evaluate the carbon emission benefits of the CNC at the material production stage. It should be noted that the term “carbon-negative” refers to the CO₂ absorption capacity of CRFA and CRCA. During the carbonation treatment of recycled aggregates, a portion of CO₂ is sequestered and stored within the aggregate structure. This captured CO₂ is accounted for as negative emissions in the raw material production stage. In addition, the negative CO₂ emission factors adopted for CRFA and CRCA in this study were derived from literature sources [32] rather than experimentally quantified for the specific aggregates used. Consequently, the reported negative emission values represent literature-based estimates and may vary depending on aggregate source characteristics, carbonation efficiency, and treatment conditions.

A process-based carbon accounting method was employed to calculate the carbon emissions of the various concrete types [40]. Specifically, according to the emission calculation formula established for different concrete raw material production routes (Equation (8)) [32], the carbon emissions per unit volume of concrete were obtained by multiplying the material consumption M by the corresponding carbon emission factor C_{ef} . This method is simple to apply, widely applicable, and reliable, and has been extensively used for quantifying carbon emissions at the material production stage.

$$\text{Carbon emission}_i = \sum_{k=1}^n M_{ik} Cef_k \quad (8)$$

where Carbon emission_i is the total carbon emission of the concrete production process ($\text{kg CO}_2 \text{ eq/m}^3$), M_{ik} is the consumption of material k (kg), and Cef_k is the carbon emission factor of material k ($\text{kg CO}_2 \text{ eq/t}$).

Table 10. Carbon emission factors in the production of each raw material.

Raw Materials	Carbon Emission Factor	Unit	Reference
CaO	0.540	kg CO ₂ eq/kg	A. Ababneh et al. [33]
CaCl ₂	0.420	kg CO ₂ eq/kg	I.D.A. [34]
CaSO ₄	0.350	kg CO ₂ eq/kg	I.D.A. [34]
SF	0.028	kg CO ₂ eq/kg	M.S. Meddah et al. [35]
BFS	0.022	kg CO ₂ eq/kg	G. Habert et al. [36]
OPC	0.735	kg CO ₂ eq/kg	GB/T 51336-2018 [37]
Water reducer	1.064	kg CO ₂ eq/kg	Moghadam et al. [38]
CRFA	−0.016	kg CO ₂ eq/kg	
CRCA	−0.007	kg CO ₂ eq/kg	
RFA	3.678	kg CO ₂ eq/t	Xiao et al. [32]
RCA	2.328	kg CO ₂ eq/t	
NFA	2.510	kg CO ₂ eq/t	
NCA	2.180	kg CO ₂ eq/t	
Mix	1.667	kg CO ₂ eq/m ³	Wang et al. [39]

To further evaluate the carbon emission efficiency of different concrete systems, CO₂ intensity was introduced as an indicator, calculated as shown in Equation (9). Here, CO₂ intensity represents the carbon emission per unit volume per unit compressive strength of concrete ($\text{kg CO}_2 \text{ eq/m}^3/\text{MPa}$). Carbon emission is the total carbon emissions of the concrete, and $f_{cu,k}$ is the compressive strength (MPa).

$$\text{CO}_2 \text{ intensity} = \frac{\text{Carbon emission}}{f_{cu,k}} \quad (9)$$

Four representative concrete systems were selected for comparative analysis of total carbon emissions and CO₂ intensity: ordinary concrete (OC) composed of OPC, natural fine aggregate (NFA), and natural coarse aggregate (NCA); low-carbon concrete (LC) prepared with low-carbon binders, NFA and NCA; recycled concrete (RC) incorporating low-carbon binders, RFA, and RCA; and carbon-negative concrete (CNC) consisting of low-carbon binders, CRFA, and CRCA. The calculated total carbon emissions and CO₂ intensities for these concrete systems are presented in Figure 9. Additionally, Figure 10 illustrates the composition of carbon emissions for each system, where positive values represent carbon emissions and negative values indicate carbon uptake.

Figure 9 presents the total carbon emissions and CO₂ intensity of different concrete types. It can be observed that CNC, RC, and LC exhibit significantly lower carbon emissions during the production stage compared to OC, with reductions of 87.1%, 81.0%, and 83.0%, respectively. Among them, CNC, which incorporates CRFA and CRCA, is capable of absorbing a certain amount of CO₂ during production, further reducing its overall carbon emissions. Compared with RC and LC, the production-stage emissions of CNC are decreased by 31.9% and 24.2%, respectively.

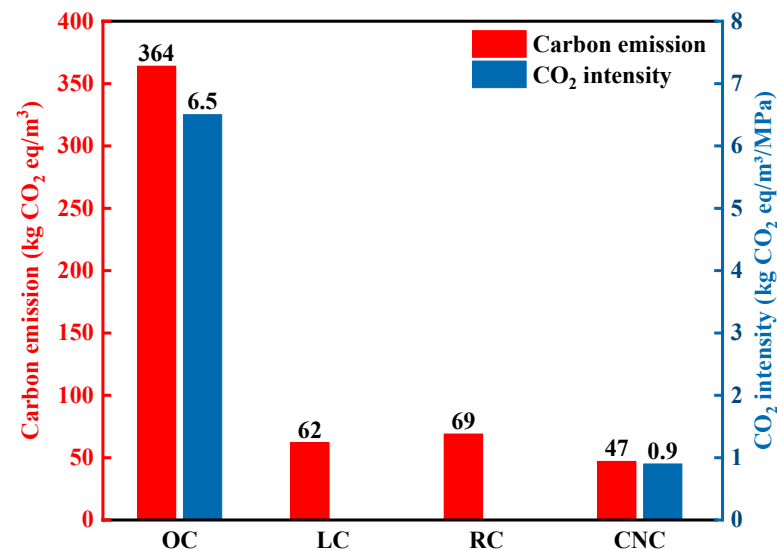


Figure 9. Total carbon emissions and CO₂ intensity.

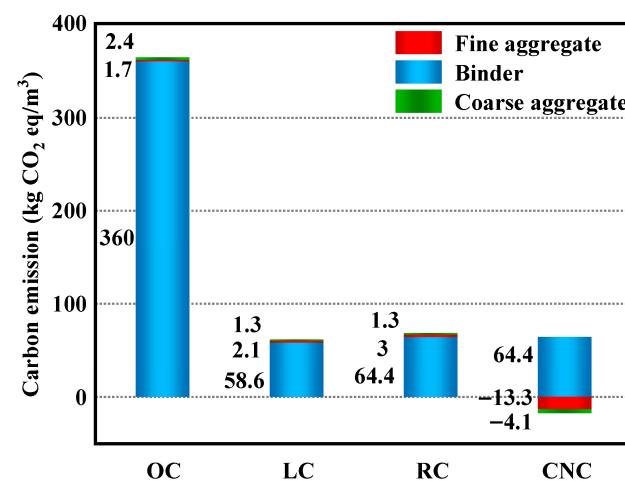


Figure 10. The contribution to carbon emissions.

For CO₂ intensity, the representative CNC was compared with the OC of the same strength grade [41]. The results indicate that at equivalent compressive strength, the CO₂ intensity of CNC is 86.2% lower than that of OC, demonstrating its significant advantage in reducing carbon emissions. It should be noted that the CO₂ intensity values are reported only for concrete systems whose compressive strengths were experimentally determined in this study. Since the compressive strengths of LC and RC were not measured, their CO₂ intensity values are not presented in order to avoid unreliable or misleading comparisons.

As shown in Figure 10, the production of binder contributes the largest share of CO₂ emissions in concrete. When low-carbon binders are used to replace OPC, the carbon emissions of LC, RC, and CNC are all significantly reduced. In particular, CNC, which incorporates carbonated recycled aggregates that have absorbed CO₂, exhibits further reductions in overall emissions. These results clearly demonstrate that the use of industrial by-products to produce low-carbon binders, combined with carbonated recycled aggregates capable of CO₂ uptake, can effectively reduce carbon emissions during the material production stage of concrete, yielding substantial carbon mitigation benefits.

3.4. Cost Analysis

To further evaluate the feasibility of replacing OC with CNC, it is important not only to achieve significant carbon emission reductions but also to maintain production costs

within a reasonable range. Excessively high costs would limit the practical applicability of CNC in engineering projects. Therefore, this section investigates the market prices of common concrete raw materials (data obtained from wholesale price ranges on the Alibaba platform), with the results summarized in Table 11.

Table 11. Market prices of each raw material (RMB refers to Renminbi (Chinese Yuan); t denotes the SI unit tonne).

Raw Material	Unit Price (RMB/t)	Raw Material	Unit Price (RMB/t)
CaO	450~650	RFA (CRFA)	20~40
CaCl ₂	900~1200	RCA (CRCA)	40~70
CaSO ₄	200~500	NFA	40~160
SF	500~900	NCA	50~100
BFS	240~300	Water reducer	2000~3400
OPC	400~450		

It should be noted that the market prices of raw materials are subject to variability depending on supplier, region, and time. In this study, the median values of the reported price ranges in Table 11 were adopted to provide a representative basis for comparison. Based on the quantities of raw materials required to produce 1 m³ of concrete, the unit costs of different concrete types were calculated, as shown in Figure 11. A simple sensitivity consideration indicates that adopting the lower or upper bounds of the reported material price ranges would change the absolute cost differences by approximately 48%. However, despite this variability in absolute values, the relative cost advantage of the proposed low-carbon concrete system compared with reference concretes remains unchanged.

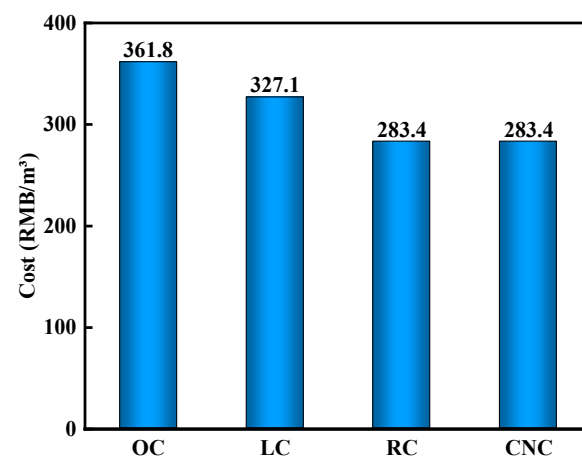


Figure 11. Preparation cost of 1 m³ of different types of concrete.

As illustrated in Figure 11, the cost of producing 1 m³ of OC is higher than that of other concrete types due to the use of OPC and natural aggregates. In comparison, LC employs low-carbon binders but still uses natural aggregates, whereas recycled aggregates generally have lower market prices than natural aggregates. Consequently, RC and CNC have lower costs than both LC and OC. Specifically, RC and CNC reduce costs by 48.1% compared with OC and by 13.4% compared with LC.

4. Conclusions

This study systematically investigated the mix design optimization, carbon emission assessment, and cost analysis of CNC. The main conclusions are as follows:

- (1) MAA-Based Mix Optimization: The optimization results based on the MAA model indicate that a 22%CRCA minimizes the RSS value of the mixture and yields the best

compressive strength. Compared with concrete containing 14%CRCA, the 22%CRCA exhibited increases in 7-day and 28-day compressive strength of 13.5% and 19.8%, respectively, demonstrating the effectiveness and applicability of the MAA model in CNC mix proportioning.

- (2) Carbon Emission Reduction: CNC, prepared using low-carbon binders derived from industrial by-products and carbonated recycled aggregates capable of CO₂ absorption, exhibited significantly reduced carbon emissions during production. Compared with OC, CNC achieved an 87.1% reduction in carbon emissions and an 86.2% reduction in CO₂ intensity, highlighting its substantial advantage in carbon mitigation.
- (3) Economic Performance: Since CNC incorporates low-carbon binders and recycled aggregates, both of which are less expensive than OPC and natural aggregates, the production cost is considerably reduced. The unit cost of producing 1 m³ of RC and CNC decreased by 48.1% and 13.4% compared with OC and LC, respectively, indicating strong economic viability.

It should be noted that the present study has several limitations. The carbon emission assessment was restricted to the raw material production stage and did not include transportation, mixing, construction, or service-life processes; therefore, the results do not represent a fully net-negative life cycle. In addition, the CO₂ uptake of carbonated recycled aggregates was not directly measured but estimated from literature values, which may vary depending on aggregate source and carbonation efficiency. Future research should focus on direct quantification of CO₂ uptake, long-term mechanical and durability performance, microstructural characterization of the interfacial transition zone, and full life cycle assessment under practical engineering conditions.

In summary, the integration of industrial by-products for low-carbon binders, the use of recycled aggregates, and the application of the MAA model for mix design optimization allow CNC to achieve enhanced mechanical performance while substantially reducing both production-stage carbon emissions and cost. These findings demonstrate the feasibility of replacing OC with CNC and provide an economically and environmentally sustainable pathway for the construction industry.

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Abbreviations

CNC	Carbon-Negative Concrete
OC	Ordinary Concrete
RC	Recycled Concrete
CRA	Carbonated Recycled Aggregate
LC	Low-Carbon Concrete
OPC	Ordinary Portland Cement
BFS	Blast Furnace Slag
SF	Silica Fume
NCA	Natural Coarse Aggregate
NFA	Natural Fine Aggregate
RCA	Recycled Concrete Aggregate
RFA	Recycled Fine Aggregate
CRCA	Carbonated Recycled Coarse Aggregate
CRFA	Carbonated Recycled Fine Aggregate
MAA	Modified Andreasen–Andersen
RSS	Residual Sum of Squares
TSMA	Two-Stage Mixing Approach
LCA	Life Cycle Carbon Assessment
RMB	Renminbi (Chinese Yuan)

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