

Experimental study on the micro-mechanism and compressive strength of concrete with calcined coal gangue coarse aggregate

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ABSTRACT

Coal gangue, a byproduct of coal beneficiation, has low strength and high carbon content, which negatively affects the mechanical properties of concrete when used as a substrate for natural gravel. However, high temperature calcination can activate its pozzolanic properties, improving its suitability for concrete production. This study investigates the microstructural and mechanical property of concrete incorporated with calcined coal gangue coarse aggregate at different temperatures (500°C, 600°C, 700°C, and 800°C). The experimental results indicate that metakaolin in coal gangue begins to decompose at 500°C, increasing the content of reactive SiO₂ and Al₂O₃, which participate in secondary hydration reactions with calcium hydroxide (CH). This process forms additional calcium silicate hydrate (C-S-H) gel, ettringite (AFT) crystals, and afwillite, enhancing the concrete microstructure. Notably, coal gangue concrete calcined at 800°C exhibited a porosity of 1.29 % and a compressive strength of 38 MPa after 28 days of curing, meeting the C30 concrete standard. Furthermore, the correlation between the compressive strength of CCGC and its microstructure exceeded 0.72, indicating that fractal dimension analysis is an effective method for predicting concrete strength.

1. Introduction

Coal gangue, a solid waste generated during coal mining, development, excavation, and washing processes, consists primarily of low-carbon content rock [1,2]. As of 2023, China has accumulated over 7 billion tons of coal gangue, with an additional 150 million tons produced annually [3]. The surface disposal of coal gangue not only occupies vast tracts of land but also causes significant environmental pollution, contaminating nearby soil, water resources, and ecosystems [2]. Given its mineral composition and physical properties, coal gangue has the potential to partially or fully replace natural aggregates in concrete production [4]. This approach could reduce the reliance on natural aggregate extraction while promoting an alternative waste management strategy for coal gangue disposal that benefits both the environment and the economy.

Several studies have demonstrated that coal gangue is a feasible material for concrete preparation, particularly in low-strength applications [5,6]. However, coal gangue aggregates typically exhibit lower strength, a more porous structure, and higher carbon

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content compared to natural gravel [4,7,8]. These properties negatively affect the mechanical performance of concrete, typically restricting its replacement rates to below 40 % [6]. Such limitations hinder large-scale utilization, necessitating strategies to enhance coal gangue's properties to increase its viability as a concrete aggregate.

Raw coal gangue has a stable crystalline structure with limited bonding ability to cement matrices [9]. Enhancing its cementitious activity is key to improving its suitability for concrete [10]. Among activation methods, thermal activation has proven to be the most effective in improving coal gangue's reactivity by disrupting its mineral crystal structure and removing carbon and other organic components [10,11]. High-temperature calcination converts coal gangue's stable crystalline phases into amorphous forms, such as metakaolin, which exhibits high pozzolanic activity [12,13]. Metakaolin plays a critical role in secondary hydration reactions, generating additional calcium silicate hydrate (C-S-H) gel and ettringite (AFT) crystals, thereby enhancing concrete's microstructure and strength [13]. Concrete with higher metakaolin content has demonstrated improved hydration, denser interfacial transition zones, and enhanced mechanical properties [7]. Thus, calcined coal gangue presents a promising solution for large-scale utilization in concrete production.

The improvement in the mechanical properties of concrete due to calcined coal gangue is primarily attributed to changes in the concrete's microstructure. Hydration products formed during the reaction fill voids within the concrete matrix, creating a more homogeneous and durable structure [14]. The generation of additional cementitious materials reduces the proportion of large pores while increasing the volume of smaller gel pores, which contributes to fewer microcracks and inhibits the propagation of existing ones [15]. Non-destructive testing methods, such as nuclear magnetic resonance (NMR), have been widely used to assess the pore size distribution of concrete [16]. NMR relaxation curves are closely associated with pore structure, porosity, and pore connectivity in concrete materials [17]. Previous studies have employed NMR to evaluate the effects of coal gangue fine aggregates on pore distribution, curing age, and water-cement ratio in concrete [18,19]. Additionally, fractal dimension analysis has proven useful in quantifying the complexity of concrete's microstructure and pore size distribution, with larger pore distributions having a greater influence on porosity and compressive strength [20].

Coal gangue can be used as cementing material to make cement or replace natural river sand to make concrete by thermal activation [14]. Despite the growing body of research on calcined coal gangue in concrete, most studies have focused on its use as a fine aggregate, with limited investigation into the effects of high-temperature calcination on coal gangue's mineral composition and its suitability as a coarse aggregate. Furthermore, the mechanisms by which calcined coal gangue influences concrete's mechanical properties and microstructure remain underexplored. Therefore, it is essential to study the impact of calcined coal gangue coarse aggregates on the hydration process and mechanical performance of concrete for high-value utilization of coal gangue.

This study investigates the micro-mechanisms and mechanical property of calcined coal gangue concrete (CCGC) via a series of laboratory tests. Specifically, it aims to analyze the hydration process using X-ray diffraction (XRD) and X-ray fluorescence (XRF); characterize the microstructure using NMR and scanning electron microscopy (SEM); evaluate the mechanical performance of CCGC across different calcination temperatures and curing ages; and establish correlations between compressive strength and microstructure of CCGC. By addressing these knowledge gaps, this study seeks to enhance the large-scale utilization of calcined coal gangue in concrete production.

2. Materials and methods

2.1. Raw materials

The coal gangue used in this study was obtained from the Moxinpo Mine in Beibei District, Chongqing, China, where it has been utilized in concrete construction, filling and support engineering applications in Chongqing. After being crushed and screened, coal

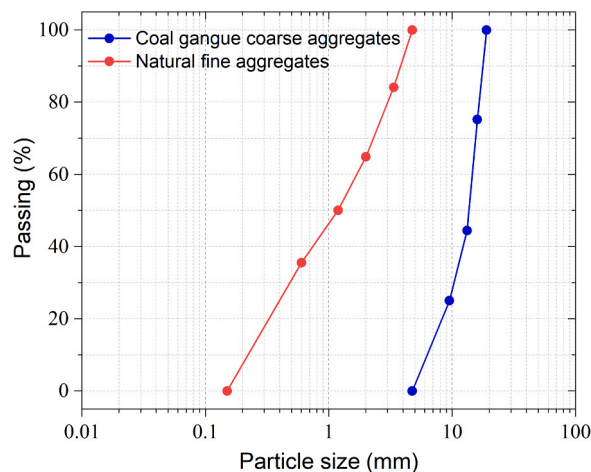


Fig. 1. Particle size distributions of coal gangue and fine aggregates used in this study.

gangue with a particle size of 5–20 mm was selected as coarse aggregates to prepare C30 concrete. The gradation of the coal gangue was determined through standard sieve analysis. The proportion of each particle size was then adjusted following Talbot's equation (Eq. (1)), with a grading shape factor n of 0.5.

$$P_i = 100 \times \left(\frac{d_i}{D_{\max}} \right)^n \quad (1)$$

where P_i is the percentage of coal gangue by weight passing the i th sieve size; d_i is the opening size of this particular i th sieve; $D_{\max}$ is the maximum size of coal gangue particles; and n is the shape factor of the gradation curve.

The fine aggregates, with a particle size ranging from 0.15 mm to 4.75 mm, were sourced from natural river sand from the Qu River in Chongqing. Fig. 1 shows the particle size distribution of coal gangue and natural fine aggregates. The fine aggregates used in the test exhibited a fineness modulus of 3.34, classifying it as coarse sand. XRD tests were conducted to analyze the mineralogical composition of the coal gangue. The primary mineral components of coal gangue were calcite (57.63 %), quartz (14.65 %), and kaolinite (7.6 %), classifying the sample as calcareous coal gangue according to its oxide composition [21]. The cement used in this study was Composite Portland cement (P.C. 42.5) produced by Chongqing Qijiang Red Lion Cement Co., Ltd. Laboratory-neutral tap water was used to mix and stir the coal gangue concrete. Polycarboxylic acid superplasticizer with water-reducing rate of 20 % was selected as the water-reducing agent to coal gangue concrete in the present study.

2.2. Specimen preparation

2.2.1. Preparation of calcined coal gangue

Brindley et al. [22] established that kaolinite is dehydroxylated to metakaolin at 500–600°C, while at 800°C, amorphous SiO_2 and Al_2O_3 are formed. At temperatures above 1000°C, metakaolin recrystallizes into mullite, thereby reducing its pozzolanic activity. Additionally, our experimental results indicated that the strength of the calcined coal gangue aggregate significantly decreases and becomes more prone to breakage when the pre-treatment temperature reaches 900°C. Therefore, the optimal calcination conditions for the current samples range between 500°C and 800°C. The muffle furnace YTH-5-12 (35 cm in length, 20 cm in width, 15 cm in height, and 2 cm in wall thickness) was utilized for the high-temperature calcination of coal gangue (particle size ranging from 4.75 mm to 20 mm) at temperatures 500°C, 600°C, 700°C, and 800°C, respectively, with a controlled heating rate of 5°C per minute. The heating durations were 96, 116, 136, and 156 minutes for 500°C, 600°C, 700°C, and 800°C, respectively. Once the target temperature was reached, samples were maintained for 120 minutes, then cooled to room temperature (24°C) for at least two hours.

Fig. 2 illustrates the color changes in coal gangue during calcination. Raw coal gangue is typically black or gray-black in color but its surface color changes significantly after high-temperature calcination. At 500°C, the coal gangue turns reddish-brown, while at 800°C it predominantly exhibits a bright red color. The observed color change indicated that the carbon, sulfur, and other organic materials attached to the surface of the coal gangue were completely burnt off as the calcination temperature increases. In addition, the calcination process altered the mineral composition of the coal gangue, resulting in the carbonation of silicate mineral and basic oxide.

The weight of coal gangue was measured before and after calcination to quantify the influence of the calcination process on its mass. Five groups of coal gangue were randomly selected and measured to calculate the average weight loss percentage caused by high-temperature calcination. The weight loss percentage increased progressively with rising calcination temperature, indicating greater material decomposition at higher temperature. At 800°C, the mass loss stabilized at an average of 9.5 %.

2.2.2. Preparation of CCGC specimens

The raw and calcined coal gangue treated at 500°C, 600°C, 700°C, and 800°C) was used as coarse aggregate for concrete, with particle size of 4.75 mm to 19 mm. Due to the high-water absorption, the coal gangue was pre-wetted for 2 hours before use [4]. Each cubic coal gangue concrete specimen (100 mm × 100 mm × 100 mm) was prepared using 375 kg/m³ of cement, 642 kg/m³ of sand, 1193 kg/m³ of coal gangue, and 185 kg/m³ of water. A polycarboxylic acid superplasticizer was added at 1 % of the total mixture as a water-reducing agent for all specimens. The cement, sand and coal gangue were mixed manually, after which water was added and stirred for three minutes until a homogenous and dense mixture was obtained. The concrete was emptied into the mold and vibrated on the shaking table until all air bubbles eliminated. The After 24 hours of casting, the CCGC specimens were removed from the molds and

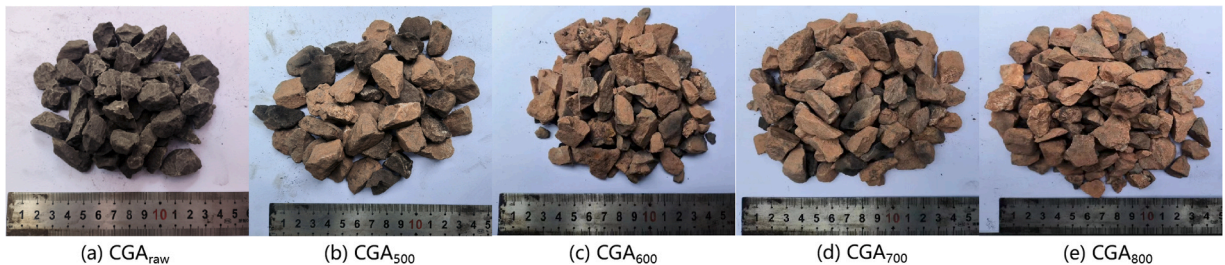


Fig. 2. The color of coal gangue before and after high-temperature calcination: (a) raw coal gangue; (b) coal gangue after calcination at 500°C; (c) coal gangue after calcination at 600°C; (d) coal gangue after calcination at 700°C; (e) coal gangue after calcination at 800°C.

cured under standard conditions ($T = 20 \pm 2^\circ\text{C}$, and $\text{RH} \geq 95\%$). The CCGC specimens were divided into five groups based on calcination temperatures (i.e., CCGC_{raw} , $\text{CCGC}_{500^\circ\text{C}}$, $\text{CCGC}_{600^\circ\text{C}}$, $\text{CCGC}_{700^\circ\text{C}}$, and $\text{CCGC}_{800^\circ\text{C}}$). Each group included specimens at three different curing ages (3 days, 7 days, and 28 days) with three duplicates per condition. In total, 45 specimens (5 calcination temperatures $\times$ 3 curing ages $\times$ 3 duplicates) were prepared for uniaxial compression testing in accordance with [23].

2.3. Concrete testing

2.3.1. XRF and XRD tests

The mineral composition of the raw and calcined coal gangue was investigated using XRD tests (X'Pert PRO MPD produced by PANalytical B.V., Almelo, Overijssel, Netherlands). The instrument operated at a working voltage of 60 kV and a current of 60 mA. The scanning range in 2θ was set from 5° to 80° , with a step size of 0.02° , and a scanning speed of $0.5^\circ/\text{s}$. The chemical composition of the samples was measured by XRF tests (ARL Perform'X produced by Thermo Fisher, United States). To analyze the mineral phase changes in the hydration products of CCGC, cement mortar pieces were extracted from the CCGC specimens. The pieces were immersed in anhydrous ethanol for 24 hours to prevent further hydration, followed by drying in an oven at 60°C for 5 hours. Finally, the dried samples were ground to pass through the No. 200 mesh sieve before being analyzed by XRD.

2.3.2. NMR and SEM tests

The peaks of the NMR T_2 relaxation curves were highly correlated with the pore size, and the area and size of the peaks indicated the proportion of the corresponding pore size. The CCGC specimens for NMR tests (Fig. 3(b)) were prepared as cylinders (50 mm in diameter and 100 mm in height) to investigate the impact of calcination temperature and curing age on porosity of CCGC. To ensure complete saturation, the specimens were placed in a ZYB-II type vacuum pressurized saturation device, where the water pressure was increased to 10 MPa following a thirty minute vacuum period. The specimens were soaked in apparatus for one day (i.e. 24 h). Prior to the test, the surface of the specimen was wiped to achieve a saturated surface dry state. The pore structure of the concrete specimens was analyzed using NMR instrument (MacroMR12-150H-I, Suzhou, China), which calculates the volume of water in the pores based on the NMR signals generated by the hydrogen nuclei in the water [22].

Crushed CCGC mortar blocks were carefully selected and ground into smooth thin sections with a thickness of 1 mm. To prevent further hydration, the specimens were immersed in anhydrous ethanol for 24 hours. The thin sections were then polished using a dedicated polishing machine to achieve a smooth surface, mounted with conductive adhesive, and subsequently coated with a 10 nm gold film to minimize charging effects during SEM observation (Fig. 3(d)).

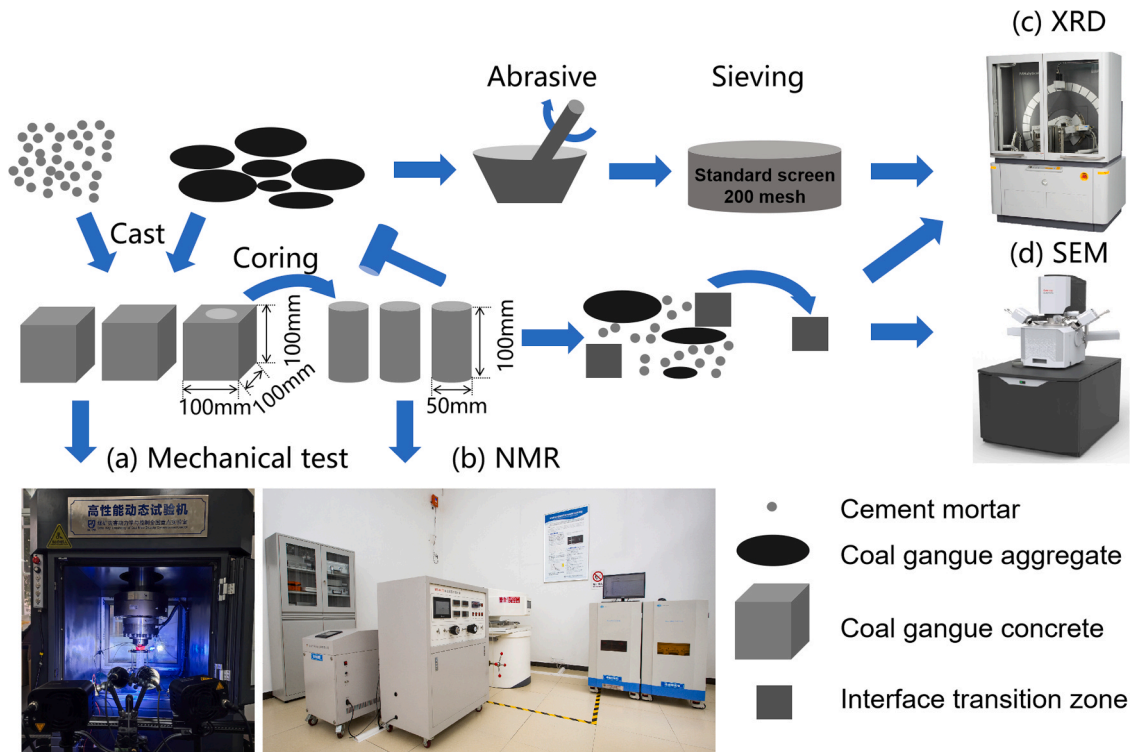


Fig. 3. Testing equipment and scheme: (a) high-performance dynamic testing machine; (b) nuclear magnetic resonance instrument; (c) X-ray diffraction; (d) environmental scanning electron microscope.

2.3.3. Uniaxial compression test

The compressive strength of concrete was tested according to Chinese National Standard GB/T 17671–1999 [23]. Uniaxial compression tests were carried out using a high-performance dynamic testing machine (DDZ-2000, Sichuan dexkcyq instrument Co., Ltd) (Fig. 3(a)). The system recorded failure models and the full stress-strain curve of the CCGC. The apparatus was comprised of a loading frame (2000 kN), a load control system (Loading rate: 0.1–50KN/s, measuring accuracy 0.5 % FS), a displacement controller (displacement loading rate 0.01–10 mm/s, measuring accuracy 0.5 % FS). The uniaxial loading process was controlled by displacement, and the loading rate was 0.01 mm/s. The apparatus automatically recorded the load and displacement every 200 ms until the specimen was destroyed. The compressive strength (σ) of CCGC was calculated using Eq. (2):

$$\sigma = \frac{F}{A} \quad (2)$$

Where σ is the compressive strength (MPa); F is the compressive failure load (N); A is the pressure area of the specimen (mm^2).

2.3.4. Environmental test

Environmental leaching tests were conducted according to HJ/T 781–2016 [24]. After 28 days of curing, the CCGC specimens were subjected to nitric and sulphuric acid treatment to simulate leaching conditions. The concentrations of isobaric metals elements in the leachates were then measured using an inductively coupled plasma mass spectrometer (ICP-MS, PerkinElmer Optima 4300DV).

3. Hydration reaction of CCGC

3.1. Mineral and chemical composition of calcined coal gangue

Fig. 4 displays the XRD patterns of the raw and calcined coal gangue at temperatures of 500°C, 600°C, 700°C, and 800°C. The primary rock minerals in raw coal gangue were calcite (57.63 %), quartz (14.65 %), and kaolinite (7.6 %). The characteristic diffraction peaks of kaolinite at 11.62° and 32.04° in the coal gangue disappeared after calcination at 500°C. This is likely due to the dehydroxylation of kaolinite during high-temperature calcination, which transformed into reactive metakaolin [7]. As the temperature increased to 600°C, the diffraction peak curves of the original kaolinite further diminished, indicating its complete decomposition. At temperatures above 600°C, the most noticeable changes were observed in the diffraction peak intensities of calcined coal gangue. The diffraction peaks of calcite also gradually decreased with increasing temperature, with the peak at 64.74° completely disappearing at 800°C. Additionally, siderite in the raw coal gangue was transformed into hematite (mainly composed of Fe_2O_3) under high-temperature and oxygen-rich conditions [14]. The formation of hematite was the primary reason for the red coloration observed in calcined coal gangue, with higher hematite content at 800°C contributing to the most intense red coloration. The chemical reaction equations involved in the calcination process of coal gangue were as follows [14,25]:

Fe^{2+} in siderite was oxidized to a more stable Fe^{3+} under high temperature and oxygen-rich conditions, as shown in (3) and (4).



At 500°C, the kaolinite was gradually transformed to metakaolin due to dehydroxylation. This process weakened the stability of Si-

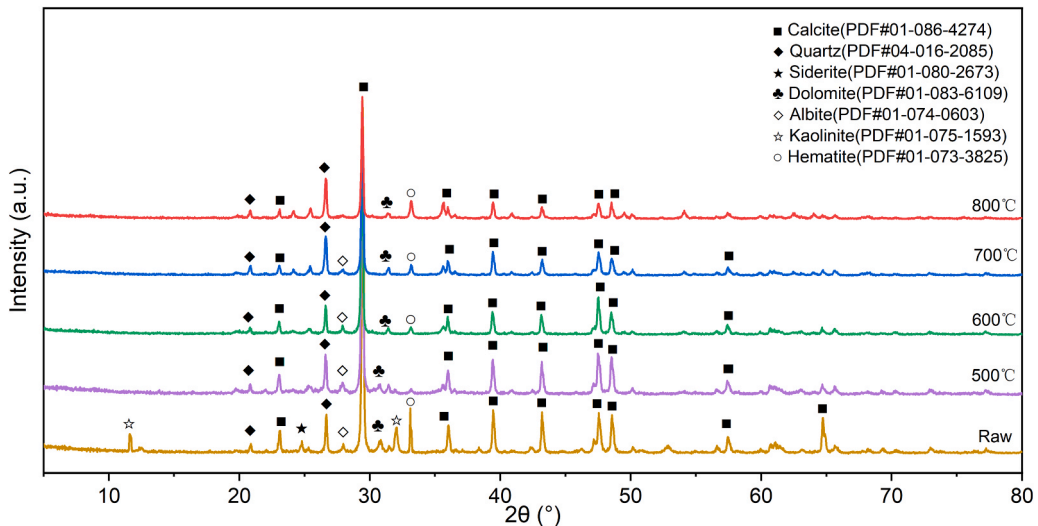


Fig. 4. XRD patterns of raw and calcined coal gangue coal gangue aggregates.

O and Al-O bonds in metakaolin, resulting in the formation of metastable aluminosilicates with high pozzolanic activity [10].



Calcite began to decompose at 700°C–800°C, converting to quicklime and releasing carbon dioxide [15].



To further analyze the chemical composition of ram and calcined coal gangue, XRF tests were conducted (Table 1). Raw coal gangue primarily consisted of oxides such as CaO, SiO₂, Al₂O₃, and Fe₂O₃. After high-temperature calcination, the carbon and sulfur in the coal gangue were completely eliminated, resulting in modifications to the mineral phases. The contents of SiO₂ and Al₂O₃ in calcined coal gangue were higher compared to the raw coal gangue with Al₂O₃ primarily existing as metakaolin (Al₂O₃·2SiO₂). The larger mass ratio of SiO₂ to Al₂O₃ means higher content of quartz crystal, while a smaller ratio indicated a higher content of reactive Si and Al in coal gangue [7]. The content of reactive SiO₂ and Al₂O₃ in the coal gangue gradually increased with calcination temperature. The mass ratio of SiO₂ to Al₂O₃ in raw coal gangue was 3.23, while it decreased to 1.92 at calcination temperature of 800°C. Additionally, the Fe₂O₃ content in coal gangue increased significantly, reaching 22.56 % at 800°C. The increase in Fe₂O₃ content suggested that the variation in hematite content was the primary reason for the different shades of red observed in calcined coal gangue.

3.2. Hydration products of CCGC

XRD tests were also carried out on CCGC (including CCGC_{raw}, CCGC_{500°C}, CCGC_{600°C}, CCGC_{700°C}, and CCGC_{800°C}) to determine the hydration products. The concrete fragments were soaked in anhydrous ethanol for 7 days before testing to terminate the hydration process [26].

The XRD diffraction peaks for the various CCGC samples exhibited a broadly similar profile (Fig. 5). It suggested that the hydration products formed in the cement mortar containing raw and calcined coal gangue were indeed the same. The hydration products of CCGC_{raw} primarily include CH crystals, C-S-H gel, and AFT crystals. Notably, there was an intense calcium hydroxide diffraction peak at 31°, which gradually disappeared with increasing calcination temperature. The calcium hydroxide content in CCGC_{raw} was higher than in the calcined samples, likely due to the low reactivity of raw coal gangue, which did not participate in secondary hydration reactions. In contrast, the metakaolin on the surface of the calcined coal gangue participated in secondary hydration reactions, consuming the excess calcium hydroxide. The XRD analysis also revealed intense abazite diffraction peaks at 27.9° in CCGC_{600°C}, CCGC_{700°C}, and CCGC_{800°C} while CCGC_{raw} exhibited almost no abazite. This indicated that the secondary hydration reaction not only produced more C-S-H gel and AFT crystals but also generated a new hydration product, abazite (CaAl₂Si₂O₈·4 H₂O).

The hydration products of studied CCGC were mainly composed of Quartz (23.5 %–59 %), Calcite (10 %–25 %), Calcium hydroxide (0 %–4 %), Hydrate Calcium Silicate (3 %–15 %), Ettringite (4 %–8 %), Abazite (0 %–32.7 %) (Table 2). The amount of hydration products in CCGC_{raw} after 28 days of curing was significantly lower than that for CCGC_{500°C}, CCGC_{600°C}, CCGC_{700°C}, and CCGC_{800°C}. As the calcination temperature increased, the calcium hydroxide content progressively declined, and was completely consumed at 800°C. Proportion of Quartz and Calcite in CCGC_{800°C} decreased to 23.5 % and 10 %, respectively, indicating that coal gangue exhibited the highest activity at this temperature. Potassium Aluminum Iron Silicate Hydroxide (K(Al, Fe)₂AlSi₃O₁₀(OH)₂) was identified in CCGC_{800°C}, potentially contributing to the enhanced strength of the concrete [14].

3.3. Hydration reaction of CCGC

In this section, the hydration reaction process of CCGC was analyzed based on the mineral and chemical composition of calcined coal gangue and concrete. The main components of Composite Portland cement include tricalcium silicate (C₃S), dicalcium silicate (C₂S), tricalcium aluminate (C₃A), and tetracalcium aluminoferrite (C₄AF) [27]. Upon mixing cement with water, hydration products including C-S-H gel and CH crystals were formed [28]. After high-temperature thermal activation, coal gangue coarse aggregates exhibited a notable degree of pozzolanic activity. The active SiO₂ and Al₂O₃ abundant in kaolin participate in secondary hydration reactions with Ca(OH)₂ from cement hydration products, producing AFT crystals and C-S-H gel, along with other hydration products (Fig. 6). The calcite present in the calcined coal gangue provided additional Ca(OH)₂ for secondary hydration reactions. Calcined coal

Table 1

The main chemical composition of raw and calcined coal gangue.

Composition (%)	Raw	500°C	600°C	700°C	800°C
CaO	63.71	46.98	45.27	42.93	34.29
SiO ₂	17.22	22.71	21.21	24.47	24.03
Al ₂ O ₃	5.34	9.47	8.31	11.07	12.51
Fe ₂ O ₃	5.58	10.21	13.63	13.33	22.56
MgO	1.98	1.63	1.03	1.40	2.27
TiO ₂	1.69	2.20	2.11	1.42	1.28
S	0.67	0	0	0	0
SO ₃	0	3.11	2.59	4.61	5.25
Na ₂ O	0.46	0.59	0.40	0.42	0.34
K ₂ O	0.45	0.59	0.39	0.60	0.50

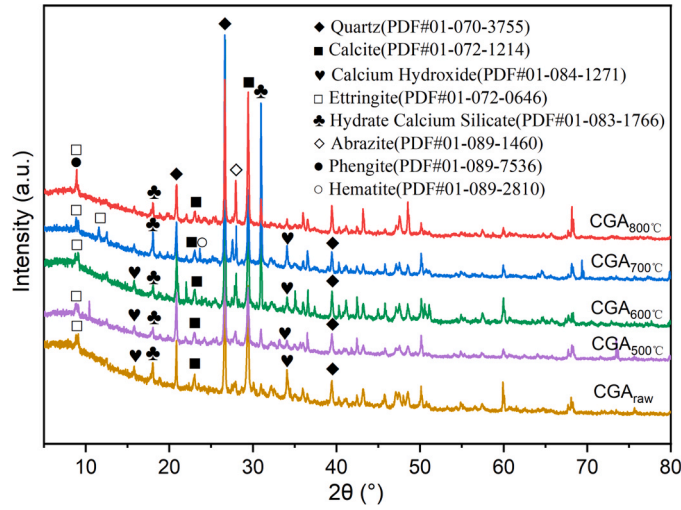


Fig. 5. XRD patterns of hydration products for different CCGC.

Table 2

The proportion of hydration products of CCGC.

Hydration products	CCGC _{raw}	CCGC _{500°C}	CCGC _{600°C}	CCGC _{700°C}	CCGC _{800°C}
Quartz (%)	59	53.6	42	37.1	23.5
Calcite (%)	25	10.1	11	19.9	10
Calcium hydroxide (%)	4	3	1	0.5	0
Hydrate Calcium Silicate (%)	4	3	15	14.8	9.1
Ettringite (%)	8	6.1	6	4	7.3
Abrazite (%)	0	21.2	22	19.8	32.7
Phengite (%)	0	0	0	0	17.4
Hematite (%)	0	0	0	4	0

gangue provided free SiO_4^{4-} and AlO_2^- ions for secondary hydration reactions, while cement hydration provided free Ca^{2+} ions. In alkaline environments (OH^-), these free ions interact with each other on the surface of coal gangue to swiftly form C-S-H gel and AFT crystals. The reaction equations were as follows:



Calcined coal gangue promoted cement hydration, especially during the early stages of hydration. The rate of cement hydration reaction was primarily controlled by the dissolution of $\text{Ca}(\text{OH})_2$, while the CaO produced from calcite decomposition provided a stronger alkaline environment for the reaction. Calcite decomposition was most pronounced above 700°C , resulting in more complete hydration process in CCGC_{700°C} and CCGC_{800°C} after 28 days of curing. In the later stages of hydration, active SiO_2 and Al_2O_3 in coal gangue react with $\text{Ca}(\text{OH})_2$ to form hydration products such as C-S-H gel and AFT crystals. The XRD results confirmed that calcined coal gangue also promoted the formation of abrazite. The higher the concentration of active SiO_2 and Al_2O_3 precipitating from calcined coal gangue, the more intense the secondary hydration reactions, leading to a denser and stronger hydration layer, which contributed to the substantial increase in the mechanical performance of CCGC.

4. Microstructure of CCGC

4.1. Microstructure of the interfacial transition zone (ITZ)

The ITZ is considered the weakest location of the concrete material and plays a crucial role in determining its compressive strength,

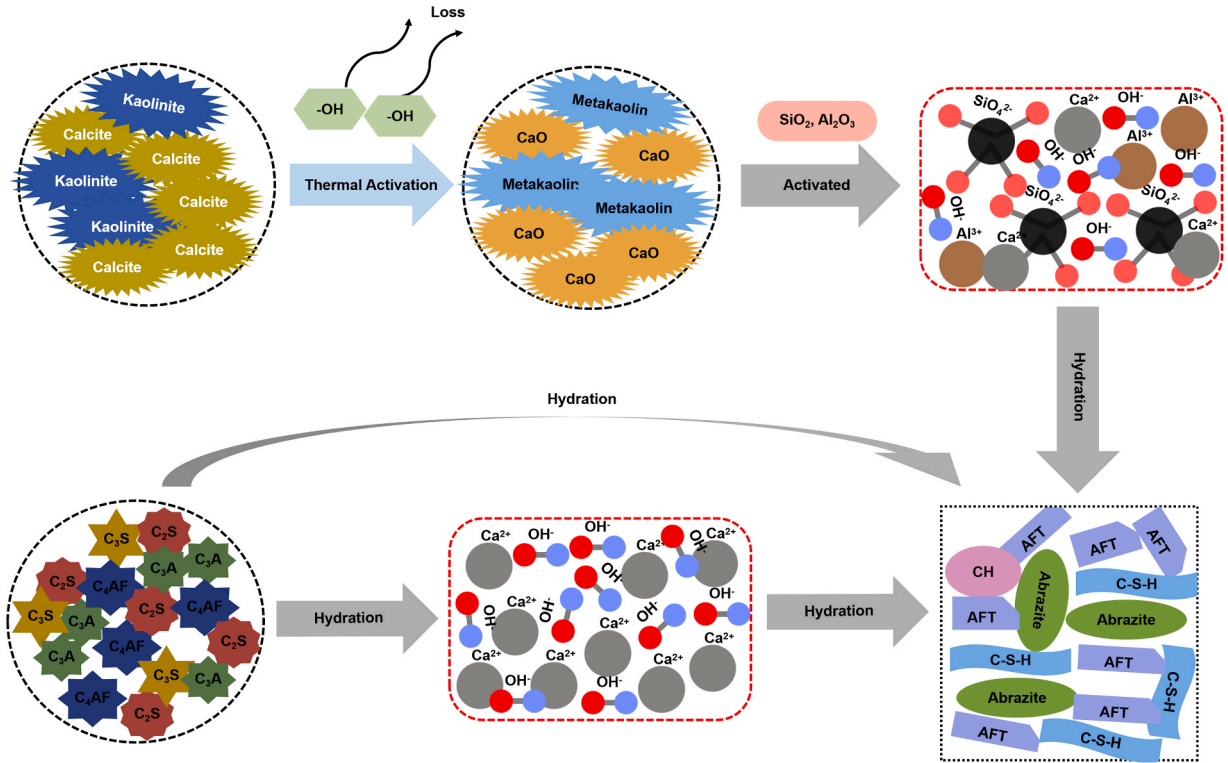


Fig. 6. Schematic of hydration reaction of CCGC.

therefore the observing the ITZ is essential in analyzing the failure model of concrete. SEM is an effective method to distinguish between coal gangue coarse aggregates and the cement matrix. This method was used to capture the ITZ of CCGC after 28 days of curing under uniaxial compression loading (Fig. 7). The microstructure of the ITZ varied significantly from that of coarse aggregates of CCGC. Some noticeable cracks were observed in the ITZ of CCGC_{raw}, indicating that the bonding between raw coal gangue and cement matrix was relatively weak (Fig. 7(a)). In contrast, calcined coal gangue demonstrated tight bonding with the cement matrix, leading to a gradual reduction in both the width and length of cracks within the ITZ. Compared with CCGC_{500°C} (Fig. 7(b)), the tighter ITZ existed in CCGC_{800°C} (Fig. 7(c)). The aggregates closely adhered to the cement matrix, rendering it difficult to distinguish the ITZ of the concrete. The microstructure of ITZ in CCGC was optimized by high temperature calcination of coal gangue coarse aggregates. It was also apparent that microcracks mainly appeared in ITZ and the cement matrix was relatively densely packed (Fig. 7(b)). With the increased of calcination temperature, the cracks extended inside the aggregates (Fig. 7(c)). The dense Microstructure of the ITZ strengthened the bond between the coal coarse aggregates and cement matrix. Thus, the ITZ was no longer the weakest point of strength in concrete, shifting the failure mode of CCGC from ITZ damage to aggregates damage.

To further observe the ITZ microstructure characteristics, the ITZ of CCGC_{raw}, CCGC_{500°C}, and CCGC_{800°C} were magnified by a factor of 10 and 20. The ITZ of CCGC_{raw} exhibited noticeable microcracks (Fig. 7(a)), while the ITZ of CCGC_{500°C} and CCGC_{800°C} was primarily characterized by pores (Fig. 7(b), (c)). The improvement of the microstructure of concrete was mainly attributed to the production of a large amount of active SiO₂ and Al₂O₃ by calcining the coal gangue coarse aggregates. The calcined coal gangue would promote generating more cementitious materials in concrete, which appeared as needle-like and columnar structures. A large amount of needle-like and clustered cementitious materials filled the microcracks and pores, forming a dense ITZ and pore structure (see Fig. 7(c)).

The primary components of ITZ of CCGC_{raw} were flaky CH crystals and C-S-H gel, with a relatively low presence of flocculent AFT crystals. As the calcination temperature increased, the number of CH crystals decreased, while the amounts of AFT crystals and C-S-H gel increased. The denser microstructure was observed in the ITZ of CCGC_{800°C}, which was completely covered by AFT crystals and C-S-H gel. It demonstrated that the secondary hydration reaction promoted the formation of cementitious products and consumes more CH crystals. There was a small amount of fly ash adhered to the surface of ITZ of CCGC_{raw} (Fig. 7(a)). The presence of carbon led to a relative reduction of substances involved in hydration reactions in the ITZ, and the pores near the fly ash were not filled with cementitious materials. It indicated that carbon was mainly present in the form of defects in the ITZ of the concrete.

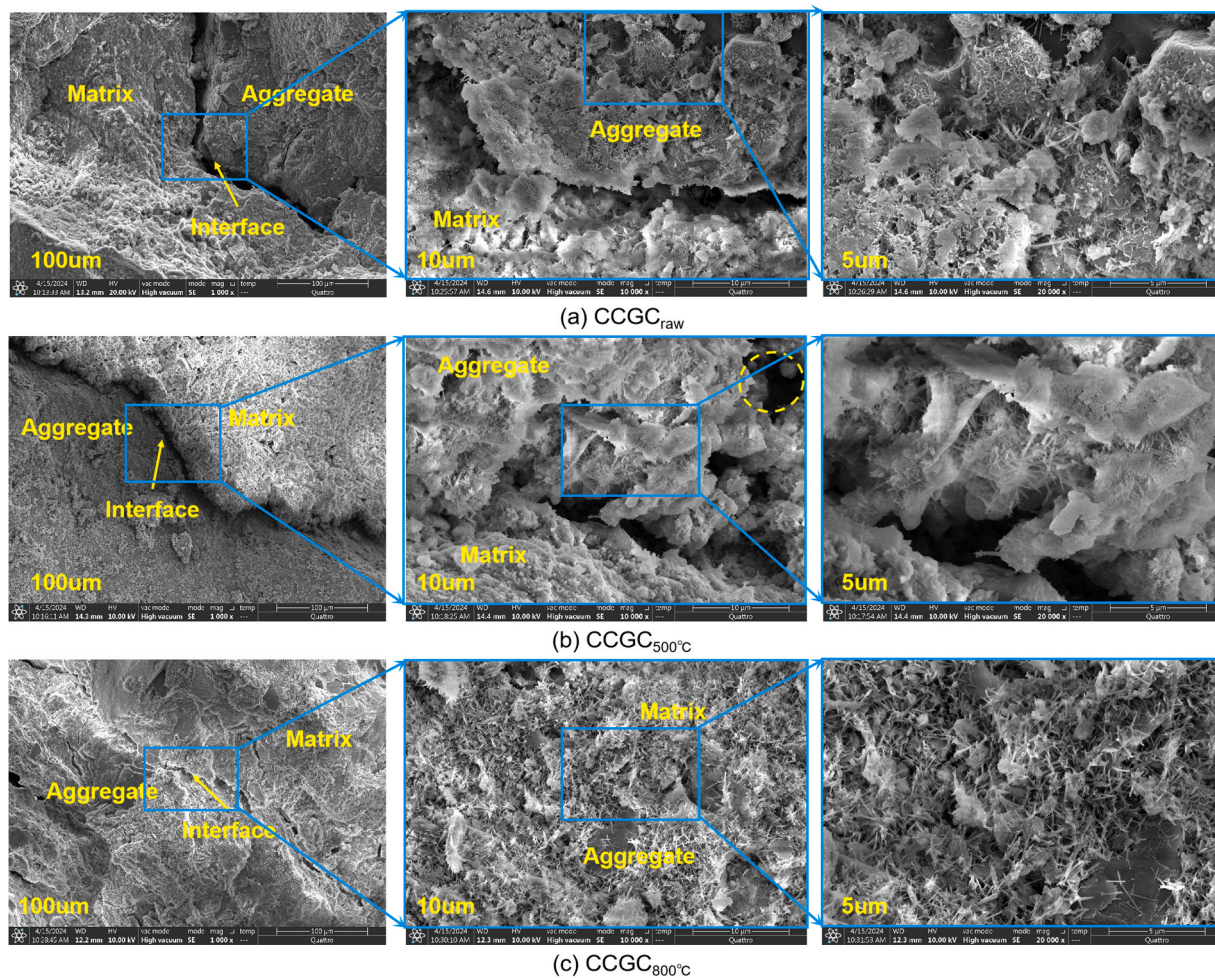


Fig. 7. SEM images of ITZ of CCGC: (a) CCGC_{raw}, (b) CCGC_{500°C}, and (c) CCGC_{800°C}.

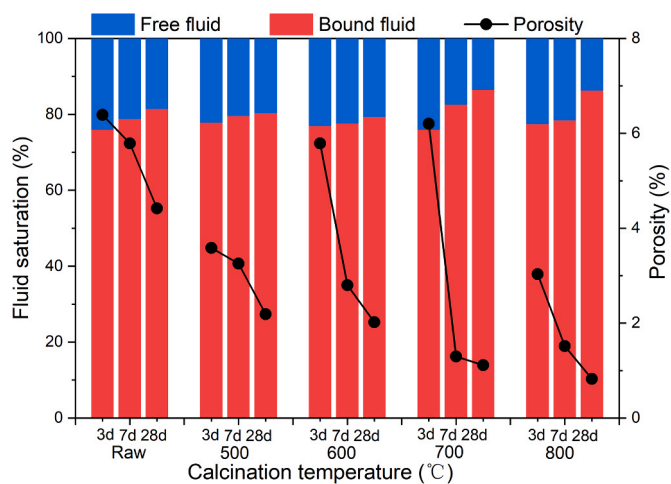


Fig. 8. The fluid Saturation and porosity of CCGC with various curing ages.

4.2. Pore structure of CCGC

4.2.1. Porosity and fluid saturation

The variation in fluid saturation and porosity of CCGC across different temperatures (raw, 500°C, 600°C, 700°C, and 800°C) and curing ages (3d, 7d, and 28d), as shown in Fig. 8. The proportions of bound fluid saturation were all greater than 75 %, indicating that

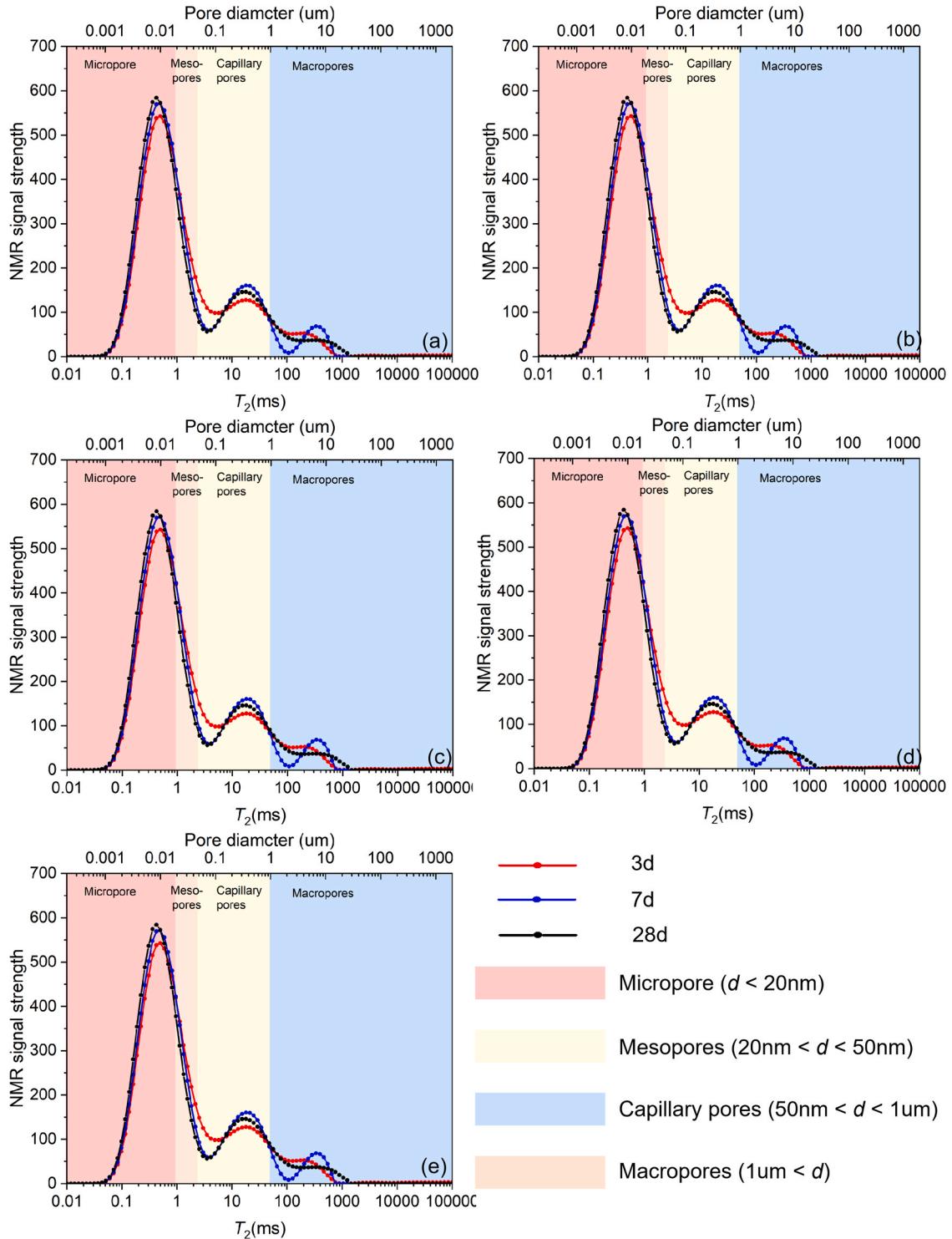


Fig. 9. The NMR T_2 spectra of CCGC with various curing ages: (a) CCGC_{raw}; (b) CCGC_{500°C}; (c) CCGC_{600°C}; (d) CCGC_{700°C}; (e) CCGC_{800°C}.

the pores of CCGC were mainly fine pores. Water in the fine pores was constrained by capillary and adsorption forces, making it difficult to flow freely. The curing ages had a significant impact on fluid saturation of CCGC. The free fluid saturation tended to decrease significantly (-15 % to -34 %) as curing ages increased from 3d to 28d. The porosity of CCGC also gradually decreased with the curing age (3d, 7d, and 28d). For example, for the specimen of CCGC_{raw}, the porosity decreased from 6.39 % to 4.41 % and the bound fluid saturation increased from 76 % to 81.5 %. This trend suggest that prolongation curing facilitated secondary hydration reactions, promoting microstructural stability of CCGC and further restricting water mobility [29]. The 28 day curing age seemed to be optimal for the pore structure adjustment of CCGC.

The porosity of CCGC_{700°C} and CCGC_{800°C} after 28 days of curing decreased more significantly than that of CCGC_{raw} with Porosity decreases of 82 % and 72.8 %, respectively. The relatively lower porosity and free fluid saturation of CCGC_{700°C} and CCGC_{800°C} (porosity: 1.11 % and 0.82 %, free fluid saturation: 13.51 % and 13.68 %, respectively) compared with other concrete (CCGC_{raw}, CCGC_{500°C} and CCGC_{600°C}) indicated the hydration reaction in CCGC_{700°C} and CCGC_{800°C} consumed more water, contributing to enhanced compactness. When the calcination temperature exceeded 500°C, the reduction in porosity caused by curing age (from 3d to 7d) was pronounced. For example, the porosity of CCGC_{600°C} and CCGC_{700°C} decreased significantly from 5.79 % to 2.8 % (-51.6 %) and 6.2–1.29 % (-79.2 %), respectively, as curing age increased from 3d to 7d. These findings indicated that the incorporation of calcined coal gangue accelerated the early hydration process of concrete. The primary reason for these differences was the increased pozzolanic activity of calcined coal gangue at higher calcination temperatures. A greater amount of active SiO₂ and Al₂O₃ attached to the surface of coal gangue coarse aggregates facilitated secondary hydration reactions with cement hydration products (Fig. 6), generating additional cementitious materials and accelerating the hardening and setting of the concrete [12]. In addition, the decomposition of calcite into CaO at 700°C provided a more alkaline environment for CCGC, where calcium hydroxide particles occupied some of the pores, further enhancing the density and uniformity of the concrete [30]. The porosity trends of CCGC_{800°C} was similar to those in CCGC_{700°C}. However, due to the already low porosity of CCGC_{800°C} after 3 days of curing (3.03 %), the rate of decline was relatively slower.

4.2.2. Pore size percentage

The T_2 relaxation curves of CCGC obtained from NMR tests were employed to quantify the effect of calcination temperatures on the pore structure of concrete. The peak of the T_2 relaxation curve represents the intensity of the NMR signal, with a stronger signal indicating a greater number of pores. The position and area of the peak are directly related to the pore size distribution [31].

The T_2 spectral relaxation curves of CCGC_{raw}, CCGC_{500°C}, CCGC_{600°C}, CCGC_{700°C} and CCGC_{800°C} concrete samples were obtained after curing for 3, 7 and 28 days (Fig. 9). The pores of CCGC can be categorized into four types according to pore diameter, i.e., micropores ($d < 20$ nm), mesopores ($20 \text{ nm} < d < 50$ nm), capillary pores ($50 \text{ nm} < d < 1 \mu\text{m}$), and macropores ($d > 1 \mu\text{m}$) (Zhou et al., 2012, Dongjin and Progress 2014). The T_2 relaxation curves of CCGC typically consisted of three peaks, including one main and two minor peaks. The main peak corresponded to the distribution of micropores, while the two secondary peaks represented capillary pores and macropores, respectively. The magnitude of the peaks was proportional to the frequency of occurrence of the corresponding pores. The main peak of the T_2 relaxation curves of CCGC mainly appeared at 0.5 ms, indicating that micropores were the main component of CCGC pores. As curing progressed, the signal of main peak had the tendency to increase, which indicated that the proportion of micropores were increased. The shape of T_2 spectral relaxation curves showed significant changes with the addition of calcined coal gangue. The T_2 spectral relaxation curves of CCGC_{raw} gradually shifted to the left with an increase in curing. In addition, when the

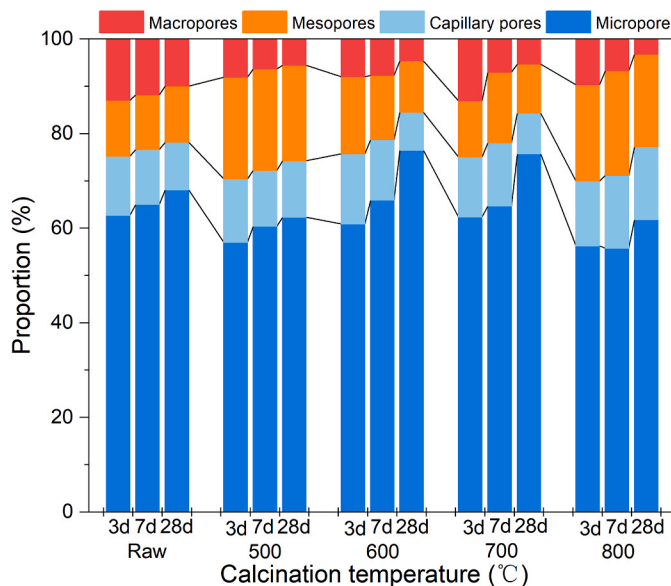


Fig. 10. The distribution of pore sizes in CCGC with various curing ages.

curing age progressed from 7 days to 28 days, the main peak increased by 18 % and the peak area ratio of minor peaks decreased gradually. Compared to CCGC_{raw}, the main peak of concrete incorporated with calcined coal gangue showed a significant increase in curing at 7 days. Changes in minor peaks of concrete with calcined coal gangue presented disordered character. This could be attributed to the higher water desorption rate of calcined coal gangue, which improved the relative humidity inside the concrete and thus promoted the hydration of the cement [7]. The progress of the secondary hydration reaction accelerated the consumption of water, which resulted in increased capillary tensile stresses increasing concrete shrinkage and increased disorder in the minor peaks [32].

To quantitatively analyze the pore size distribution of CCGC over the curing period, the proportion of each pore size was calculated, as shown in Fig. 10. The pores in CCGC were mainly micropores, accounting for over 55 % of the total pores in the specimens. As the curing process progressed, the proportion of macropores in the pore structure continuously decreased, while the proportion of micropores gradually increased. For CCGC_{raw}, the proportion of macropores decreased from 13 % to 10 %, while the proportion of micropores increased from 63 % to 68 %. The transformation was driven by the formation of hydration products, which progressively filled the large pores, reducing their size. In concrete incorporated with calcined coal gangue, the proportion of macropores ranged from approximately 3–6 %, while the proportion of micropores ranged from approximately 62–76 %, demonstrating a denser pore structure compared to CCGC_{raw}. The phenomenon arose from the secondary hydration reactions between active SiO₂ and Al₂O₃ on the surface of coal gangue coarse aggregates and Ca(OH)₂. The reactions promoted the formation of gels including C-S-H gel and AFT crystals, effectively reducing the proportion of deleterious pores in CCGC. Minimal changes were observed in mesopores and capillary pores. A possible explanation is that hydration products filled some mesopores and capillary pores, transforming them into smaller micropores, while the filling of macropores compensated for the reduction in transition pores and capillaries.

5. Mechanical behavior of CCGC under uniaxial compression

5.1. Effect of calcination on the compressive strength of CCGC

Uniaxial compression tests were conducted on 45 CCGC specimens (CCGC_{raw}, CCGC_{500°C}, CCGC_{600°C}, CCGC_{700°C}, and CCGC_{800°C}) to investigate the mechanical behavior. The typical stress-strain curve of CCGC (specimen CCGC_{raw}) under uniaxial compression is shown in Fig. 11. The stress at the maximum compressive strength is defined as the peak stress σ_c , and the corresponding strain is defined as the peak strain ϵ_c . The failure process of CCGC under uniaxial compression can be divided into the following five stages:

(1) Pore and crack compaction stage (OA segment). In the preparation of concrete, the presence of pores is inevitable, leading to a nonlinear curve during the compaction stage of the stress-strain curve for CCGC. (2) Elastic Stage (AB Segment). During this stage, coal gangue concrete experiences elastic deformation, which is entirely recoverable upon unloading. (3) Unstable crack propagation stage (BC segment). Point B is the inflection point that coal gangue concrete transitions from elastic deformation to plastic deformation, also known as the yield point. The stress at point B represents the yield stress, which is approximately 85 % of the peak strength. (4) Failure stage (CE segment). After reaching peak strength, the stable structure of coal gangue concrete was compromised, but it remained largely intact. In the CD segment, cracks rapidly propagate and penetrate through, causing surface spalling of the specimen. Point D represents the limit of crack development. In the DE segment, the specimen relied on the frictional force between the coal gangue aggregate and the cement paste to support the load. (5) Residual Stage (after Point E). After Point E, the coal gangue concrete lost its

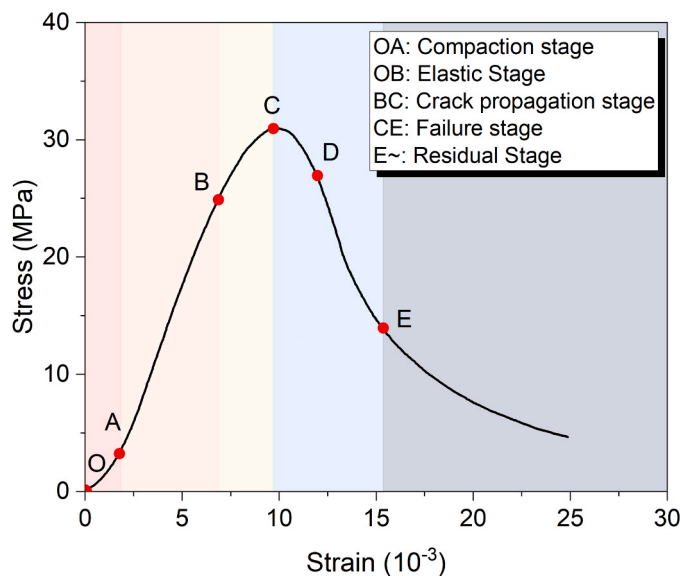


Fig. 11. The typical stress-strain curve of CCGC under uniaxial compression (specimen CCGC_{raw}).

load-bearing capacity.

The compressive strengths of CCGC were significantly lower than that of natural concrete (Fig. 12). For the natural concrete, the compressive strength after 7 days and 28 days were 33.51 MPa and 45.23 MPa respectively. The compressive strength of CCGC_{raw} after 3 days, 7 days and 28 days of curing were 16.66 MPa, 18.59 MPa and 30.99 MPa respectively. The compressive strengths of CCGC increased gradually with the curing age. The addition of calcined coal gangue influenced the mechanical properties of coal gangue concrete, particularly its compressive strength. After 28 days of curing, the compressive strength ranged from 29.22 MPa to 38.04 MPa, reaching 35.4 %-15.9 % of the strength of natural concrete at the same curing age. The compressive strengths of CCGC_{500°C} was slightly lower than that of CCGC_{raw}, whereas CCGC_{600°C}, CCGC_{700°C} and CCGC_{800°C} exhibited significant strength improvements of 3.13 %, 15.55 % and 22.75 %, respectively. For 7 days of cured specimens, compressive strengths for coal gangue concrete were similar when the calcination temperature was below 600°C. However, as the calcination temperature increased, strength values also increased, reaching 26.39 MPa and 28.29 MPa for CCGC_{700°C} and CCGC_{800°C}, respectively. In contrast, high-temperature calcination exerted a limited effect on the compressive strength of CCGC after only 3 days of curing, with variations in compressive strength ranging from 2.69 % to 15.61 % compared with CCGC_{raw}.

The compressive strength growth rates of CCGC_{raw}, CCGC_{500°C}, and CCGC_{600°C} after 7 days of curing were consistent, at approximately 10 %. However, CCGC_{700°C} and CCGC_{800°C} exhibited significantly higher growth rates of 43.09 % and 36.29 %, respectively. When curing age increased to 28 days, the compressive strength of CCGC_{raw}, CCGC_{500°C}, and CCGC_{600°C} improved significantly, increasing by 66.7 %, 42.3 %, 45.87 %, respectively. In contrast, the compressive strength growth rates for CCGC_{700°C} and CCGC_{800°C} were relatively lower at 35.8 % and 34.46 %, respectively.

These findings indicated that the incorporation of calcined coal gangue can accelerate the curing efficiency of CCGC in the early stages, promoting rapid hardening of the concrete. The carbon content significantly affected the physical and chemical properties of the coal gangue coarse aggregates. High temperatures calcination removed harmful substances such as carbon and sulfur attached to the surface of the coal gangue, which were detrimental to the compressive strength of concrete. Additionally, at elevated temperatures kaolinite decomposition in coal gangue generated more reactive SiO₂ and Al₂O₃, consuming more Ca(OH)₂, which promoted secondary hydration reactions. This process led to the formation of increased amounts of gelling substances contributing to strength development. The presence of clusters of C-S-H gel and flocculent Aft crystals further enhanced the mechanical strength of CCGC. Furthermore, the decomposition of calcite provided an additional source of alkaline material for concrete, contributing to the early hydration reactions. The significant presence of iron-rich phases (hematite) in calcined coal gangue may have also contributed to the observed increase in compressive strength [7].

The water-cement ratio and aggregate gradation also influence the compressive strength of concrete. The effects of water-cement ratio and gradation of coal gangue on compressive strength were therefore evaluated and analyzed by varying water-cement ratios and grading shape factors *n* of CCGC (see Appendix B).

5.2. Failure characteristics

Concrete specimens made with raw coal gangue and calcined coal gangue exhibited similar failure modes during uniaxial compression tests. The damage surface showed two relatively connected quadrilateral cones. The failure of CCGC mainly occurred at the side of the specimens, with minimal overall structural damage (Fig. 13). There were two different types of fracture surfaces in

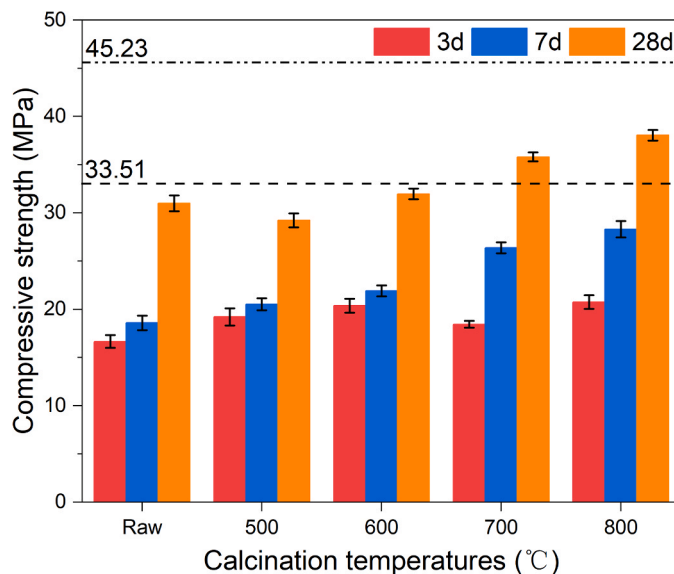


Fig. 12. The compressive strengths of CCGC with various curing ages.

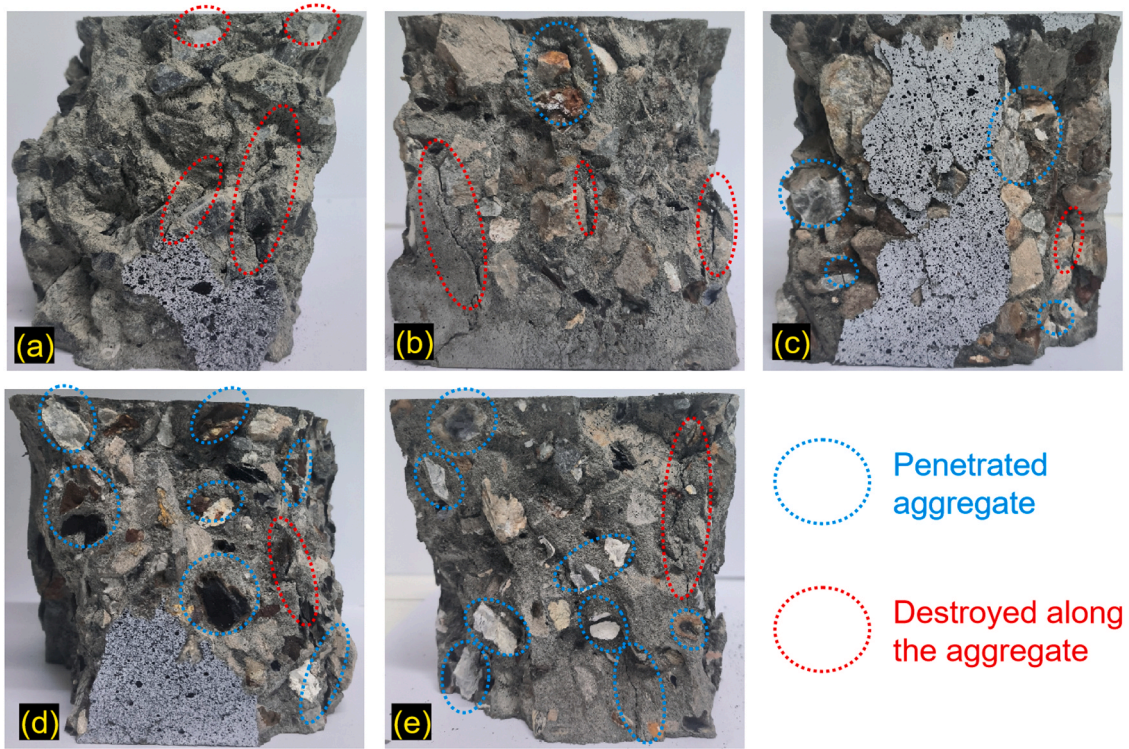


Fig. 13. Failure modes of CCGC during uniaxial compression tests: (a) CCGC_{raw}; (b) CCGC_{500°C}; (c) CCGC_{600°C}; (d) CCGC_{700°C}; (e) CCGC_{800°C}.

CCGC including ITZ fracture and coal gangue aggregate fracture. Fracture surfaces in CCGC_{raw} primarily occurred at the interface between the aggregate and the cement matrix, as well as within the cement matrix (Fig. 13(a)). In contrast, calcined coal gangue coarse aggregates in CCGC_{500°C}, CCGC_{600°C}, CCGC_{700°C} and CCGC_{800°C} were more prone to fracture. For CCGC_{500°C}, crack propagation mainly occurred in the ITZ or within smaller coarse aggregates (Fig. 13(b)). As the calcination temperature increased, larger aggregates were penetrated, and the fracture surface gradually shifted from the ITZ to aggregate fracture. For CCGC_{700°C} and CCGC_{800°C}, most of the coarse aggregates were fractured, with minimal damage observed in the ITZ, resulting in relatively smooth fracture surfaces (Fig. 13(d) and (e)).

Typically, cracks tend to propagate towards the weaker regions of the concrete. The presence of a certain amount of carbon on the surface of raw coal gangue and calcined coal gangue at temperatures of 500°C weakened the bond between the aggregate and the cement. Significant pores and cracks were present in the ITZ, causing concrete to be mainly destroyed along ITZ. The mineral composition of calcined coal gangue under high-temperature calcination changed, with a high content of metakaolin. The pores in CCGC were filled with gellable material produced by the hydration reaction. Consequently, CCGC_{700°C} and CCGC_{800°C} exhibited denser pore structures and ITZ. Therefore, the calcined coal gangue coarse aggregates were penetrated.

5.3. Correlation between compressive strength and microstructure

The pore structure of concrete can be characterized quantitatively using fractal dimensions based on NMR T_2 spectra [33]. According to the relationship between capillary radius and capillary pressure, the cumulative volume fraction of capillaries with a radius less than r can be calculated as V [34]:

$$V = \left[\frac{T_{2\max}}{T_2} \right]^{D-3} \quad (12)$$

where V is the cumulative pore volume fraction for the transverse relaxation time less than T_2 of the total pore volume; $T_{2\max}$ is the maximum transverse relaxation time; T_2 is the transverse relaxation time; D is the fractal dimension.

Taking the logarithm on both sides of the equation:

$$\lg(V) = (3 - D)\lg(T_2) + (D - 3)\lg(T_{2\max}) \quad (13)$$

If the pores in concrete exhibited fractal characteristics, the plot of $\lg(V)$ versus $\lg(T_2)$ will display a linear relationship. The slope of the line is $3 - D$, where the fractal dimension D is calculated as 3 minus the slope of the line [35].

The fractal characteristics of CCGC with different calcination temperatures and curing ages were analyzed, as shown in Fig. 14. The

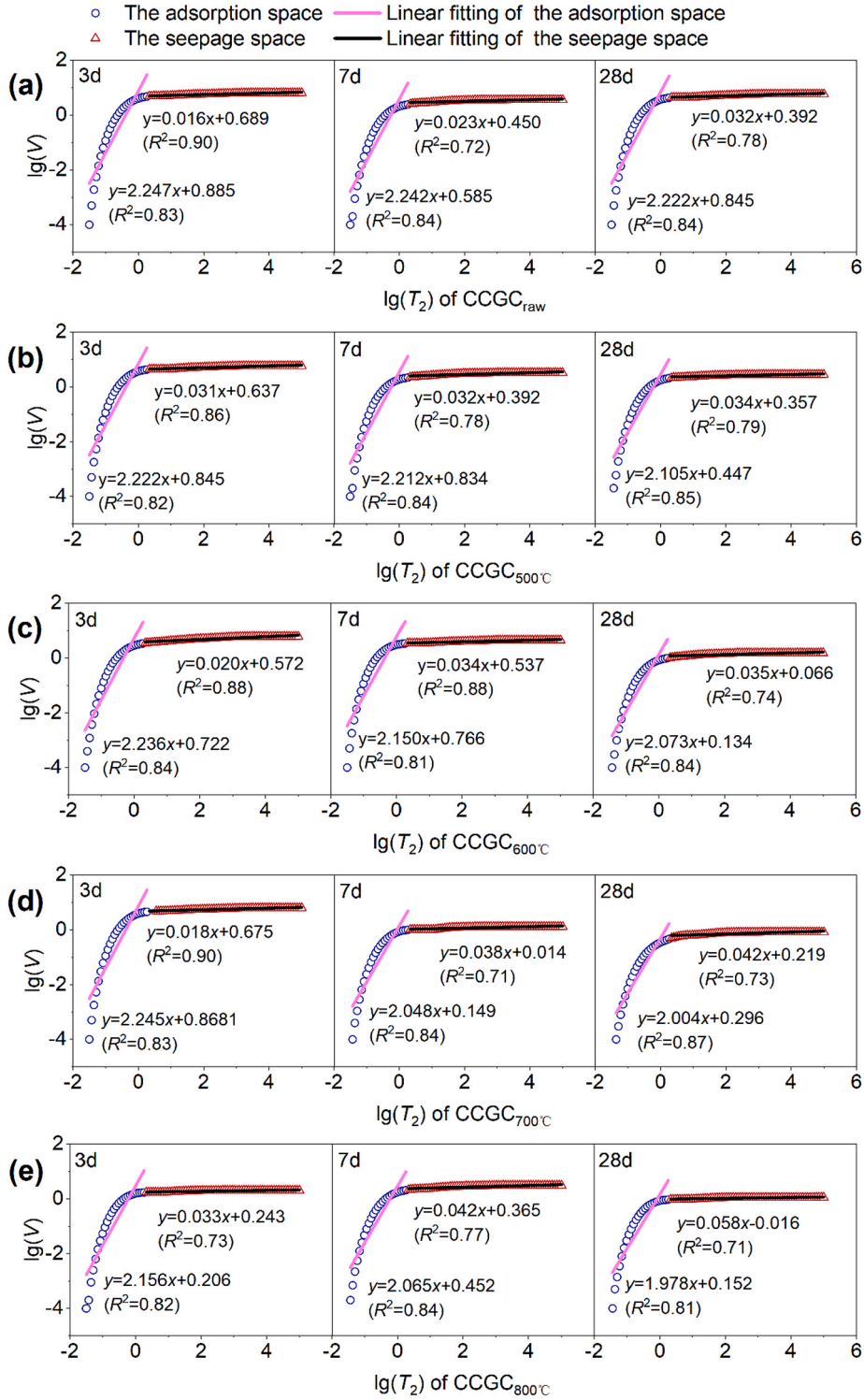


Fig. 14. Fractal dimensions of pore structures in CCGC: (a) CCGC_{raw}; (b) CCGC_{500°C}; (c) CCGC_{600°C}; (d) CCGC_{700°C}; (e) CCGC_{800°C}.

$\lg(V)$ and $\lg(T_2)$ curves of different CCGC have similar trends (Fig. 14). The curves exhibited a distinct inflection point, indicating that the pores in the CCGC exhibited dual fractal characteristics at different relaxation times. The pore structure of CCGC was relatively complex, making it difficult to describe using a single fractal dimension. Therefore, two different stages were fitted separately, resulting in two fractal dimensions $D_{\min}$ and $D_{\max}$, for the CCGC. $D_{\min}$ represented the fractal dimension corresponding to the specific

surface area of the adsorption space ($T_2 < 10$ ms), with a range from 0.753 to 1.022, and an average value of 0.853. $D_{\max}$ represented the fractal dimension corresponding to the seepage space volumes ($T_2 > 10$ ms), with a range from 2.942 to 2.984, and an average value of 2.971. This is generally consistent with the results of previous research [35]. $D_{\min}$ was selected to represent the fractal dimension of fine pores based on the relaxation time. The smaller the pore size and the more micropores present in the specimen, the larger the fractal dimension $D_{\min}$, indicating a more complex pore structure [29]. The larger the fractal dimension $D_{\max}$, the larger pores were present in concrete, indicating a higher quantity of large pores.

The correlation coefficient of the fitting curve for $D_{\min}$ was higher than 0.82, while that for $D_{\max}$ was higher than 0.7. It indicated that the pore structure of CCGC exhibited good fractal characteristics. The fractal dimension $D_{\min}$ values of CCGC increased with curing age. It indicated that the number of fine pores in CCGC increased with curing time. It also aligned with the trends observed in the T_2 relaxation curves. Although the fractal dimension $D_{\max}$ exhibited a decreasing trend with increasing curing age, the extent of change was relatively minimal. The fractal dimensions $D_{\min}$ of CCGC_{raw} and CCGC_{500°C} after 7 days of curing increased by 0.67 % and 1.23 %, respectively. The fractal dimensions $D_{\max}$ of CCGC_{raw} and CCGC_{500°C} decreased by 0.25 % and 0.04 %, respectively. On the contrary, the fractal dimensions $D_{\min}$ of CCGC_{700°C} and CCGC_{800°C} showed higher growth rates after 7 days of curing, increasing by 23.15 % and 10.89 %, respectively. The fractal dimensions $D_{\max}$ of CCGC_{700°C} and CCGC_{800°C} decreased by 0.69 % and 0.31 %, respectively. The results indicate that the number of fine pores in concrete continued to increase as the curing process progressed, while the number of large pores decreased. The calcined coal gangue promoted the early hydration of cement and improved the pore structure of concrete. The fractal dimension $D_{\min}$ of CCGC was greater than that of CCGC_{raw} after 7 and 28 days of curing, while $D_{\max}$ is generally lower. The fractal dimensions $D_{\min}$ and $D_{\max}$ of CCGC_{800°C} after 28 days of curing increased by 31.24 % and decreased by 0.86 %, respectively, compared to CCGC_{raw}. This demonstrated that high-temperature calcination can improve the pozzolanic activity of coal gangue, enabling it to play a more effective role in hydration reactions [36]. As the pores of CCGC were filled with secondary hydration products, a more complex and denser pore structure was formed [33].

Concrete porosity was influenced by its internal pore structure, and fractal dimension can effectively describe the internal spatial structure of these pores [18]. The relationship between fractal dimensions (i.e., $D_{\min}$ and $D_{\max}$) and porosity of CCGC is shown in Fig. 15. The quadratic polynomial model was able to fit the porosity and fractal dimension with a coefficient of determination (R^2) greater than 0.91 (Fig. 15). The analysis results indicated that fractal dimension $D_{\min}$ was negatively correlated with porosity ($R^2=0.91$), while fractal dimension $D_{\max}$ exhibited a strong positive correlation with porosity ($R^2=0.94$). The correlation coefficient of fractal dimension $D_{\min}$ was lower than that of fractal dimension $D_{\max}$, indicating that changes in porosity in CCGC were primarily influenced by larger pores. The increase in fractal dimension $D_{\min}$ revealed an increasing complexity of fine pores in CCGC, suggesting that the formation of hydration products reshaped its internal pore structure. The decrease in fractal dimension $D_{\max}$ illustrated that as the curing process progressed, the large pores in the concrete were gradually filled with hydration products. As the pore size decreased, the complexity of pore shape and connectivity also decreased accordingly [18,33].

The compressive strength of CCGC was closely related to the microscopic pore structure of concrete, which can be described by the fractal dimensions $D_{\min}$ and $D_{\max}$ [18]. A quadratic polynomial function was used to perform regression analysis on the relationship between fractal dimensions and compressive strength of CCGC, with a coefficient of determination (R^2) greater than 0.72 (Fig. 16). The results indicated that fractal dimension $D_{\min}$ was positively correlated with the compressive strength of CCGC (Fig. 16(a)), while fractal dimension $D_{\max}$ was negatively correlated with compressive strength (Fig. 16(b)). As the micro and macro pores in concrete were continually filled, the proportion of harmless or less harmful pores contributing to compressive strength increased, while the proportion of harmful pores decreased. Comparing with Fig. 15 and Fig. 16, it can be found that the correlation between the compressive strength and fractal dimensions $D_{\min}$ and $D_{\max}$ were not strong. In particular, the compressive strength of concrete after 28 days of curing was more discrete. The most probable reason for this result is that the hydration process in CCGC after 28 days of curing was basically over, the ITZ was no longer the weakest part of the concrete strength. In the initial stage of curing (3d and 7d), the change of compressive strength was mainly resulted from the variation in pore structure of CCGC. The compressive strength was strongly influenced by the aggregate strength during the final stage of curing (28d).

6. Discussion

In this study, we investigated the microstructure and mechanical properties of concrete prepared with calcined coal gangue as coarse aggregate. The thermal activation of coal gangue significantly altered its mineral composition, improving its pozzolanic activity. The environmental impact of concrete produced with calcined coal gangue was also evaluated by testing the leaching concentrations of heavy metals of CCGC (see Appendix A). The concentrations of heavy metals leached from CCGC were within the

Table 3
Fractal dimension of five groups of CCGC.

Specimens	Curing age 3d		Curing age 7d		Curing age 28d	
	$D_{\min}$	$D_{\max}$	$D_{\min}$	$D_{\max}$	$D_{\min}$	$D_{\max}$
CCGC _{raw}	0.753	2.984	0.758	2.977	0.778	2.968
CCGC _{500°C}	0.778	2.969	0.788	2.968	0.895	2.966
CCGC _{600°C}	0.764	2.980	0.850	2.966	0.927	2.965
CCGC _{700°C}	0.755	2.982	0.952	2.962	0.996	2.958
CCGC _{800°C}	0.844	2.967	0.935	2.958	1.022	2.942

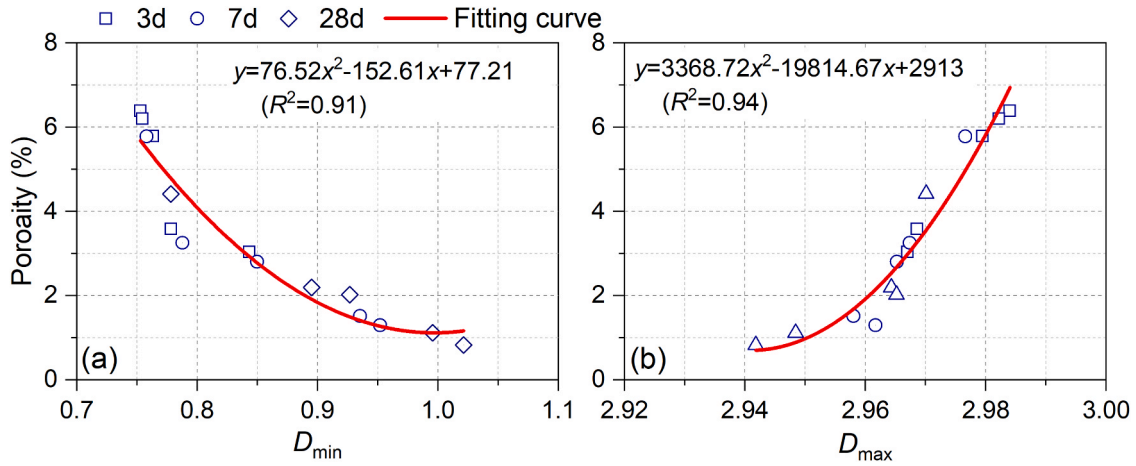


Fig. 15. Correlation between fractal dimensions ((a) D_{min} and (b) D_{max}) and porosity of CCGC.

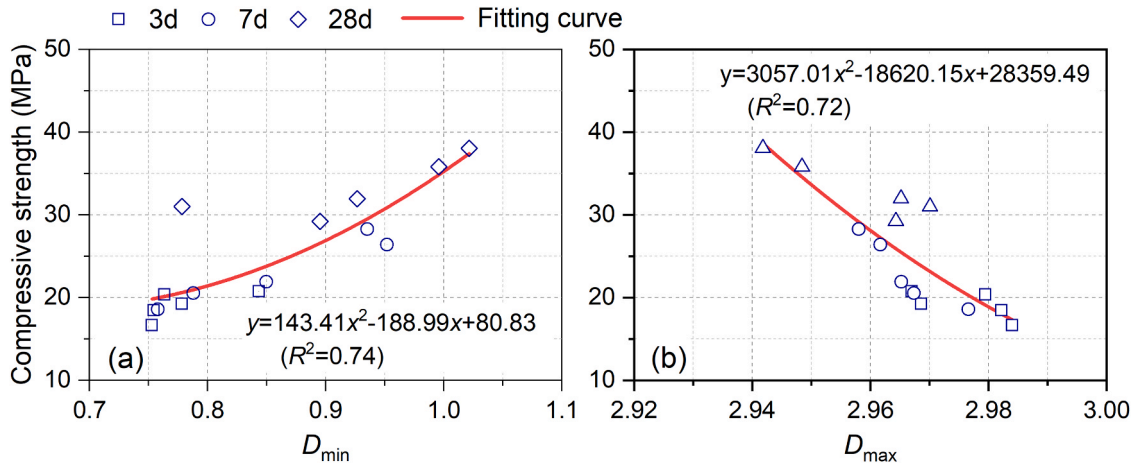


Fig. 16. Correlation between fractal dimensions ((a) D_{min} and (b) D_{max}) and compressive strength of CCGC.

permissible limits set by the GB 5085.3–2007 standard, and the leached heavy metal concentrations from CCGC_{800°C} were the lowest, indicating that thermal activation can mitigate the environmental impact of CCGC.

The integrity of the interfacial transition zone (ITZ) is crucial in determining the failure mechanisms of concrete [37]. The pozzolanic reaction induced by metakaolin generates additional gel products, reducing ITZ porosity and enhancing its compactness [8]. Nanoindentation tests have shown significant improvements in the thickness, hardness, and elastic modulus of the ITZ in metakaolin-containing concrete [38,39]. While this study qualitatively analyzed ITZ failure modes using SEM, future research should incorporate nanoindentation techniques for a quantitative assessment of ITZ strength in concrete incorporating calcined coal gangue at various temperatures. Even with the complete replacement of natural aggregates by calcined coal gangue, the concrete's compressive strength meets the C30 standard. However, this study focuses solely on compressive strength, without evaluating flexural strength or durability. For calcined coal gangue to be widely adopted in construction materials, it must fulfill both mechanical performance and durability requirements, particularly for applications such as pavement engineering [40]. Future research should include freeze-thaw cycle testing and carbonation resistance assessments to ensure that CCGC meets the durability criteria necessary for infrastructure applications [8,41,42].

Fractal analysis of the pore structure in CCGC revealed two distinct regions based on the specific surface area of adsorption space and seepage space volumes. However, the fractal characteristics of different pore types remain unclear. Future studies should categorize pore regions based on pore size classification and investigate the evolution of pore fractal characteristics. Fractal dimension was able to characterize the pore structure variation of concrete. However, microstructure is not the sole determinant of strength; factors such as aggregate strength, water-cement ratio, and grading must be fully considered in future work [19].

In high-temperature environments, the accelerated evaporation of water within concrete increases porosity and weakens the structure, leading to a reduction in strength [43]. Elevated temperatures also cause the expansion of water vapor and entrapped air, which can induce surface cracking, compromising the concrete's permeability and durability and making it more susceptible to

environmental erosion and degradation [44]. Furthermore, high temperatures accelerate cement hydration, shortening the setting time. This can disrupt construction scheduling, affect molding quality, and create uneven internal stress distributions, thereby increasing the risk of cracking. Therefore, the thermal stability of calcined coal gangue concrete must be carefully evaluated to ensure its long-term performance and structural integrity.

Sulfate erosion is a major cause of concrete deterioration, particularly in sulfate-rich soils and aquatic environments [45]. Elevated sulfate concentrations accelerate concrete degradation, highlighting the need to assess the sulfate resistance of heat-activated gangue concrete to ensure its long-term stability [46]. The denser pore structure observed in CCGC, compared to conventional raw coal gangue concrete, can effectively mitigate acid penetration, thereby enhancing both strength and sulfate resistance. However, given the complex factors influencing sulfuric acid resistance, further experimental investigations are necessary to confirm its reliability for engineering applications.

7. Conclusions

In this study, the microstructure and mechanical properties of CCGC was investigated by using SEM NMR, and high-performance dynamic testing machine. The effect of calcined coal gangue incorporation on the microstructure and mechanical properties of concrete were studied. Based on fractal theory, the correlation between fractal dimension $D_{\min}$ and $D_{\max}$ and porosity and compressive strength of concrete were analyzed. The main conclusions were as follows:

- (1) The chemical composition of coal gangue underwent significant changes after calcination. Kaolinite was completely transformed into metakaolin at 600°C, and the content of reactive SiO_2 increased with rising calcination temperature, indicating enhanced pozzolanic activity. Siderite was oxidized to hematite under high-temperature, oxygen-rich conditions, causing the coal gangue to turn red.
- (2) Concrete incorporating calcined coal gangue exhibited a denser pore structure and ITZ. The microstructural enhancements were attributed to secondary hydration reactions that consumed CH crystals and generated beneficial products such as C-S-H gel, ettringite, and afwillite crystals. These hydration products effectively filled large pores and mesopores, transforming them into capillary pores and micropores. The porosity of CCGC decreased with curing time (from 3 to 28 days), as the calcined coal gangue promotes early hydration and accelerates cement hardening.
- (3) The incorporation of high-temperature calcined coal gangue significantly improved the mechanical properties of concrete. Compressive strength increased with calcination temperature, with CCGC_{800°C} achieving a 28-day compressive strength of 38.04 MPa, which was 22.75 % higher than CCGC_{raw}. This result meets the requirements for C30 concrete and suggests that 800°C is the optimal temperature for maximizing the pozzolanic activity of calcareous coal gangue.
- (4) The $\lg(V)$ and $\lg(T_2)$ curves of the 15 CCGC samples exhibited clear inflection points, indicating dual fractal behavior. The complexity of the small pore structure, represented by $D_{\min}$ (0.753–1.022), was lower than that of the large pore structure, represented by $D_{\max}$ (2.942–2.984). A strong correlation ($R^2 > 0.91$) was observed between the porosity of CCGC and the fractal dimensions ($D_{\min}$ and $D_{\max}$), suggesting that the internal pore characteristics were effectively described by fractal theory. However, the correlation between compressive strength and fractal dimensions was weaker ($R^2 < 0.74$), indicating that factors beyond pore structure, such as the strength of coarse aggregates, also influenced compressive strength.

CRedit authorship contribution statement

Zhang Yuyu: Writing – review & editing. **Zhang Zhenyu:** Writing – review & editing, Supervision. **Hao Shengpeng:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **May Bambesiwe M.:** Writing – review & editing, Validation. **Xu Wenyu:** Writing – review & editing, Writing – original draft, Visualization, Validation, Formal analysis, Data curation.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Shengpeng Hao reports financial support was provided by National Natural Science Foundation Project of China. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A

To evaluate the environmental impact of concrete produced with calcined coal gangue as a substitute for natural aggregates, the leaching concentrations of heavy metals were tested using nitric acid and sulfuric acid leaching methods (Table A-1).

The results (Table A-1) show that the concentrations of heavy metals leached from CCGC were within the permissible limits set by the GB 5085.3–2007 standard. Notably, the leached heavy metal concentrations from CCGC at 800°C were the lowest, indicating that thermal activation can mitigate the environmental impact of CCGC.

Table A-1

Leaching concentrations of heavy metals of CCGC

Heavy metals (mg/L)	CCGC _{raw}	CCGC _{500°C}	CCGC _{800°C}	Permissible limits
Cr	0.08896	0.07227	0.04571	1
Cu	0.05230	0.00555	0.00805	1
As	0.00106	0.00117	0.00123	5
Mn	0.00149	0.00280	0.00112	1
Cd	0.00010	0.00043	0.00005	1
Zn	0.03031	0.01911	0.01355	1
Fe	0.11816	0.15580	0.18504	10
Pb	0.010.33	0.00126	0.00429	5

Appendix B

The material proportions of CCGC specimens varied with the water-cement ratio. For a water-cement ratio of 0.4, the mix contained 462 kg/m³ of cement, 612 kg/m³ of sand, 1138 kg/m³ of coal gangue, and 185 kg/m³ of water. For a water-cement ratio of 0.6, the mix included 308 kg/m³ of cement, 666 kg/m³ of sand, 1238 kg/m³ of coal gangue, and 185 kg/m³ of water. The grading shape factors considered in this study were 0.4, 0.5, and 0.6.

Fig. B-1 presents the compressive strength of CCGC under different calcination temperatures, water-cement ratios, and gradations. The labeling follows the format: calcination temperature (raw and 800°C) – water to cement ratio (0.4, 0.5, and 0.6) – grading shape factor n (0.4, 0.5, and 0.6). Regardless of whether tested at raw or 800°C, the compressive strength increased when the water-cement ratio was raised from 0.4 to 0.5 but decreased when further increased to 0.6, suggesting an optimal water-cement ratio of approximately 0.5 (Fig. B-1). A high water-cement ratio reduces the proportion of cementitious material generated during hydration, making it difficult to fill the pores between particles, which in turn weakens the concrete structure [47]. Additionally, water evaporation increases porosity, further compromising the mechanical properties of concrete [48]. Conversely, an excessively low water-cement ratio can hinder cement hydration, negatively affecting the setting and curing process [49]. Within a certain range, improved gradation uniformity positively influences compressive strength; however, more specimens with different coal gangue gradations should be tested to evaluate the effect of CCGC composites.

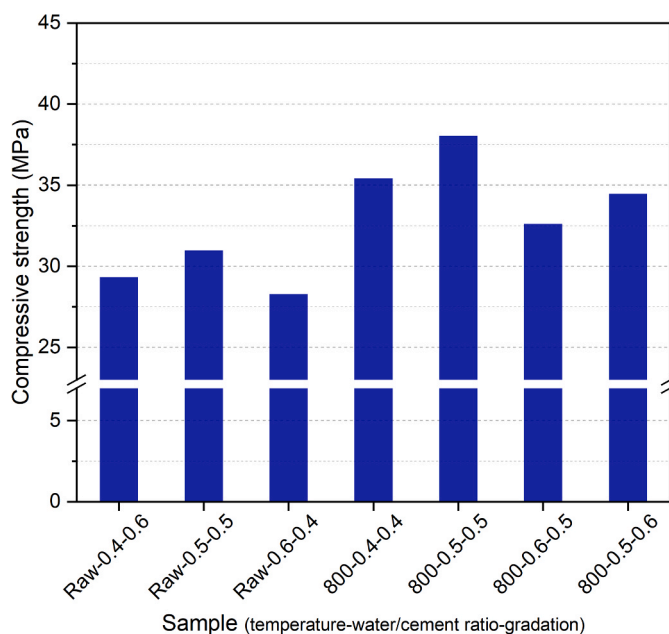


Fig. B-1. Compressive strength of CCGC with different water-cement ratios and the grading shape factors n of coal gangue

Data availability

Data will be made available on request.

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