

## Research Progress on CO<sub>2</sub> Capture Materials and Mineral Carbonation Technologies from Coal Gasification Slag

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**Abstract.** Coal gasification slag (CGS), an important by-product of the coal chemical industry, is rich in elements such as silicon, carbon, calcium and magnesium with a unique mineral composition, and has become an important raw material for CO<sub>2</sub> capture and mineralization. Utilization of CGS as a precursor for CO<sub>2</sub> capture material synthesis and CO<sub>2</sub> mineralization can significantly reduce the costs of traditional technologies, achieve the high-value utilization of CGS, and promote the synergistic reduction of pollution and CO<sub>2</sub> emissions in the coal chemical industry. Herein, a comprehensive review on the research progress regarding CO<sub>2</sub> capture and mineralization using CGS was provided. The synthetic strategies and mechanisms of silicon-based, carbon-based, and carbon-ash composite adsorbents derived from CGS for CO<sub>2</sub> capture were presented in detail, while the effects of different treatments on the structural properties and CO<sub>2</sub> capture capacity of adsorbents were summarized and compared. In addition, the mechanisms and the research progress on different technologies using CGS for CO<sub>2</sub> mineral carbonation were introduced. Finally, the problems existing in the application research of CGS in CO<sub>2</sub> capture and mineralization were discussed. It was recommended that more in-depth studies should be conducted focusing on performance enhancement, preparation process optimization, coupling technology development for the preparation of CO<sub>2</sub> capture materials and mineralization from CGS, as well as economic feasibility evaluations.

**Keywords:** coal gasification slag; CO<sub>2</sub> capture; silicon-based adsorbents; carbon-ash composite adsorbents; mineralization

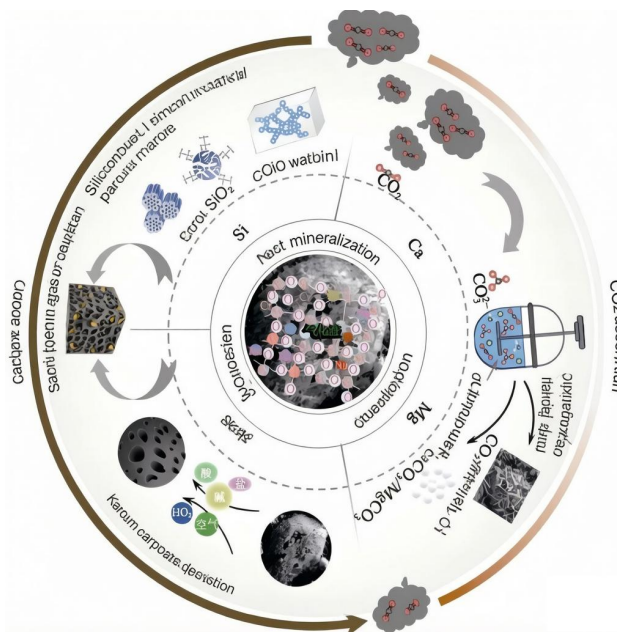
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### 1 Introduction

Coal remains the predominant energy resource and industrial feedstock in China, consistently representing in excess of 55% of national energy consumption. Amid escalating global energy requirements, coal combustion has emerged as the foremost contributor to worldwide CO<sub>2</sub> discharges. Empirical data indicate that carbon dioxide emissions attributable to coal utilization constitute over 75% of China's aggregate carbon footprint. therefore, reducing CO<sub>2</sub> emissions from the coal industry has become the key to China's carbon reduction efforts[1-2]. Although the cost of developing and utilizing renewable energy sources such as solar and wind power has gradually decreased and their economic benefits have steadily improved, they still cannot meet the huge energy demand[3]. Under the circumstance that fossil fuels cannot be completely abandoned in the short term, carbon capture, utilization and storage (CCUS) technology is an important implementation pathway to achieve large-scale carbon emission reduction in the coal industry in the medium and short term, while ensuring energy security and sustainable utilization[4]. According to the International Energy Agency, by the mid-21st century, the application of CCUS technology will cumulatively contribute approximately 15% of global carbon emission reductions and 3%–25% of China's carbon emission reductions[5]. Meanwhile, captured CO<sub>2</sub> can be converted into high-value-added products such as chemicals, biofuels, and fertilizers through physical, chemical, and biological processes, generating additional economic benefits. However, constrained by high costs, technical safety, and reliability concerns, CCUS technology has not yet been deployed on a large scale. As the key component of CCUS technology and the main source of cost and energy consumption, CO<sub>2</sub> capture accounts for 65%–85% of the total project cost[6]. Developing low-cost, high-performance CO<sub>2</sub> capture materials and high-value-added CO<sub>2</sub> utilization technologies has always been a research focus.

Coal gasification technology, as the cornerstone of clean coal conversion and coal-to-chemicals process industries, consumes approximately 250 million tons of raw coal annually[7]. However, incomplete combustion of coal with oxidants during the gasification process results in the co-production of about 30% coal gasification slag per ton of processed coal, with an annual average output exceeding 33 million tons[8-9]. Coal gasification slag obtained via high-temperature quenching processes typically contains abundant inorganic minerals such as SiO<sub>2</sub> (mass fraction 28%–52%), Al<sub>2</sub>O<sub>3</sub> (mass fraction 8%–28%), CaO (mass fraction 4.3%–28.0%), and MgO (mass fraction 0.7%–4.3%), as well as residual carbon (mass fraction 20%–50%), providing necessary conditions for its resource utilization and high-value-added applications[10].



**Figure 1** CO<sub>2</sub> capture and mineral carbonation technologies from coal gasification slag

Nevertheless, the elevated residual carbon fraction in gasification slag precludes compliance with ignition loss specifications mandated for construction and building material applications. Furthermore, the inherent phenomenon of carbon-ash fusion and encapsulation constrains its broad-scale valorization within resource recovery frameworks. Currently, most coal gasification slag is still disposed of via open-air stacking or landfilling, with only approximately 8% by mass being utilized as a resource. This situation not only leads to resource squandering but also consumes extensive land area, elevates ecological contamination risks, and potentially endangers public health [11-12]. In alignment with the "Dual Carbon" objectives and advancing CCUS technology research, leveraging the residual carbon, silicon, calcium, magnesium, and other elemental constituents within gasification slag as feedstock for CO<sub>2</sub> capture material synthesis and mineral carbonation sequestration offers an innovative avenue for slag valorization. The substantial SiO<sub>2</sub> content and residual carbon fraction, characterized by preserved porous architectures, expansive specific surface areas, and elevated pore volumes, can function as economically viable silicon and carbon precursors, respectively. Through physical and chemical treatments and acid leaching for impurity removal (Ca, Mg, etc.) as pre-/post-treatment processes, mesoporous SiO<sub>2</sub>, activated carbon, and other silicon-based, carbon-based, and carbon-ash composite adsorbents can be prepared for CO<sub>2</sub> capture. The captured CO<sub>2</sub> can then react with CaO, MgO, and other alkaline earth metal oxides in gasification slag, or with the Ca- and Mg-rich acid leachate generated during the preparation of gasification slag-based CO<sub>2</sub> capture materials via carbonation reactions, to produce high-value-added carbonate products. This further realizes CO<sub>2</sub> utilization and sequestration and recovers valuable resources such as Ca and Mg from gasification slag. As two major pollutants in the coal chemical sector, CO<sub>2</sub> and gasification slag offer advantages such as large output, wide availability, and low cost compared with commonly used raw materials such as coal, peat, and natural minerals when used as precursors for CO<sub>2</sub> capture material preparation and CO<sub>2</sub> mineralization. Moreover, gasification slag is abundantly produced in industrial parks near coal chemical CO<sub>2</sub> emission sources, enabling on-site utilization, greatly reducing the costs of slag transfer, treatment, disposal, and CO<sub>2</sub> mineralization transportation. This effectively achieves "pollution

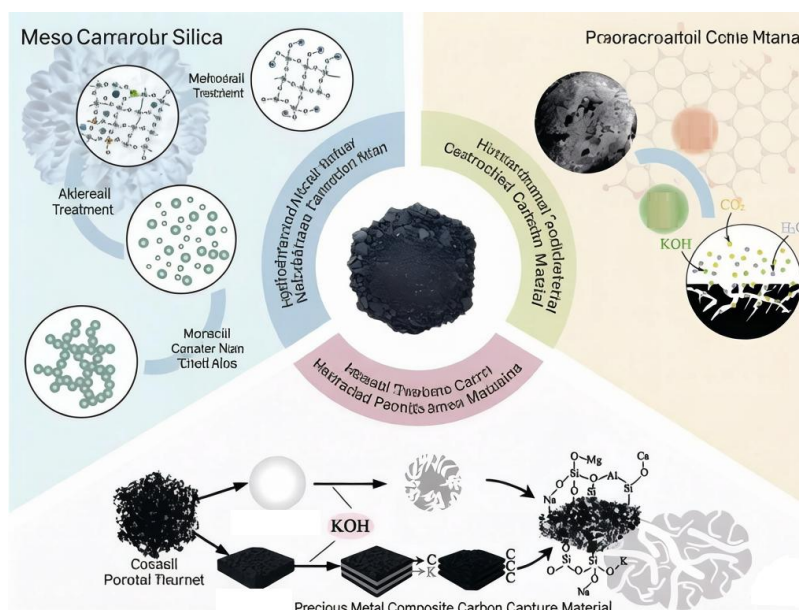
control with pollutants” in the coal chemical field, alleviates environmental pollution caused by improper gasification slag disposal, promotes synergistic pollution reduction and carbon emission mitigation in the coal chemical industry, and delivers dual economic and environmental benefits. In recent years, research has reported on the application of gasification slag in construction and building materials, ecological environment remediation, residual carbon utilization, and preparation of high-value-added materials[10,13-14]. With the proposal of China’s “Dual Carbon” goals, research on utilizing gasification slag for CO<sub>2</sub> capture and mineralization has attracted increasing attention.

This paper systematically summarizes and analyzes the current preparation methods, reaction mechanisms, advantages, disadvantages, and application prospects of silicon-based, carbon-based, and carbon-ash composite CO<sub>2</sub> capture materials derived from gasification slag. It conducts a comparative evaluation of how diverse fabrication methodologies influence the structural attributes and CO<sub>2</sub> sequestration performance of the resulting adsorbents, while also reviewing and critically examining the contemporary research landscape concerning coal gasification slag utilization in post-capture CO<sub>2</sub> mineralization and long-term storage applications. On this basis, the main problems existing in the application of gasification slag for CO<sub>2</sub> capture and mineralization are pointed out, and future research directions are proposed. A novel concept of utilizing the acid leachate by-produced during the preparation of gasification slag-based CO<sub>2</sub> capture materials for mineralization is put forward, aiming to realize the coupling of gasification slag-based CO<sub>2</sub> capture material preparation and mineralization technologies and the synchronous recovery and utilization of multiple valuable resources. This advancement will facilitate the deployment of gasification slag within carbon capture, utilization, and storage frameworks, while furnishing theoretical foundations and empirical evidence to broaden slag resource valorization and advance the environmentally sustainable evolution of the coal sector.

## 2 Gasification Slag-Based CO<sub>2</sub> Capture Materials and Their Capture Performance

Carbon dioxide capture denotes the separation and enrichment of CO<sub>2</sub> from emission streams through diverse gas separation methodologies. Presently, prevalent CO<sub>2</sub> capture routes primarily encompass pre-combustion separation, oxy-fuel combustion, and post-combustion recovery. Pre-combustion capture technology isolates CO<sub>2</sub> from fuel or shifted syngas prior to the combustion of carbonaceous or hydrogenous fuels. This approach offers elevated CO<sub>2</sub> partial pressure in the processed gas stream, reduced contaminant levels, and diminished energy penalties, though its current deployment remains largely confined to integrated gasification combined cycle power generation facilities [15]. Oxy-fuel combustion technology substitutes pure oxygen for atmospheric air in fossil fuel combustion, yielding flue gas with substantially elevated CO<sub>2</sub> concentration. A portion of the flue gas is recycled and mixed with oxygen to prevent excessive combustion temperatures, while the remaining portion undergoes cooling, compression, and other processes to separate CO<sub>2</sub>. This technology effectively reduces pollutant emissions from traditional fuel combustion processes but increases the energy consumption of the oxygen production system and the total investment[16]. Post-combustion capture technology separates and recovers CO<sub>2</sub> from blast furnace flue gas. Its operational protocol is comparatively straightforward, the underlying technology is well-established, and capital investment requirements are modest, rendering it the most extensively deployed carbon capture methodology currently [17]. Conventional post-combustion capture approaches encompass liquid amine absorption, solid-phase adsorption, membrane separation, and cryogenic fractionation [18]. Among these, the liquid amine scrubbing technique represents the most mature and industrially prevalent technology, distinguished by robust CO<sub>2</sub> absorption capacity and rapid uptake kinetics. Nevertheless, this method is encumbered by substantial regenerative energy demands, pronounced corrosive behavior, and considerable absorbent degradation and volatilization losses. Solid adsorption methods, owing to their high adsorption efficiency, simple operation, low regeneration cost and energy consumption, absence of corrosion, high configuration flexibility, and wide applicability, are the most commercially promising carbon emission reduction strategies[19]. As the core of adsorption-based CO<sub>2</sub> capture technology, porous carbon capture materials are generally required to possess high CO<sub>2</sub> adsorption capacity and selectivity, fast adsorption/desorption kinetics, chemical and thermal stability, and reversible regenerability. In recent years, extensive research has been conducted on the development of various types of solid adsorbents, such as carbon-based adsorbents (activated carbon, carbon nanotubes, etc.), silicon-based adsorbents (zeolites, SiO<sub>2</sub>, etc.), and metal-organic frameworks. The abundant silicon and carbon elements in gasification slag can serve as cost-effective silicon and carbon sources for preparing various silicon-based, carbon-based, and carbon-ash

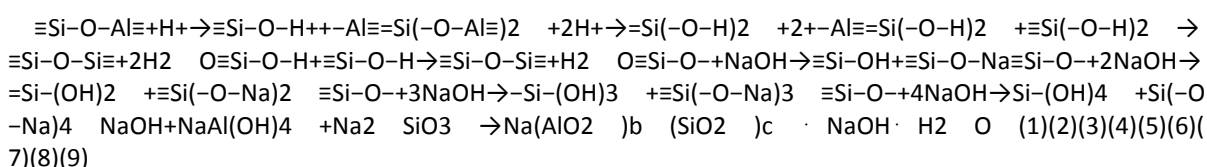
composite CO<sub>2</sub> capture materials. The types and preparation schematic of these materials are shown in Figure 2.



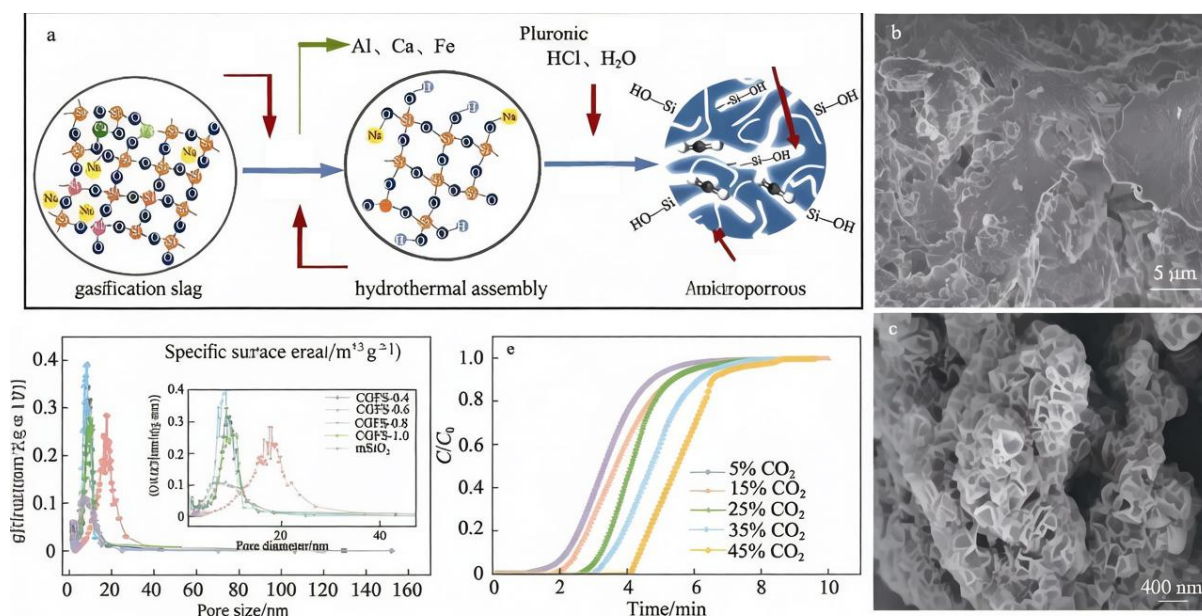
**Figure 2** Schematic diagram of types and preparation methods of carbon capture materials based on gasification slag

## 2.1 Silicon-Based CO<sub>2</sub> Capture Materials

Currently, commonly used silicon-based CO<sub>2</sub> capture materials mainly include mesoporous SiO<sub>2</sub>, zeolites, silica gel, lithium silicate, etc.[20]. Among these, mesoporous silica has garnered extensive application in CO<sub>2</sub> sequestration owing to its adjustable pore architecture, expansive specific surface area, profuse surface silanol functionalities (Si–OH), and favorable thermal and mechanical robustness [21]. Traditional mesoporous SiO<sub>2</sub> is mainly prepared by using organic molecules as templates, leveraging the synergistic interaction between inorganic silicon sources and template interfaces or supramolecular self-assembly to form assemblies, followed by removal of the organic matter via high-temperature calcination or extraction to retain the inorganic framework. This process is usually costly and difficult to scale up for mass production[22]. The SiO<sub>2</sub> rich in coal gasification slag can serve as a silicon source precursor for preparing mesoporous SiO<sub>2</sub> CO<sub>2</sub> capture materials, thereby reducing the production cost of mesoporous SiO<sub>2</sub>. Since most of the silicon in gasification slag exists in the form of amorphous silicate glass phases with low reactivity, pretreatment via acid leaching to dissolve acid-soluble metals and alkali activation is required to obtain soluble sodium silicate as the silicon source precursor. During acidic leaching, cationic species in solution disrupt the chemically inert Si–O–M linkages (M = Al, Ca, Fe) within the gasification slag matrix, inducing bond scission and generating reactive Si–O and Al–O termini. This process triggers depolymerization of amorphous aluminosilicate frameworks and concurrent dissolution of contaminant elements including aluminum, calcium, iron, and magnesium. The resultant activated Si–O–H functionalities subsequently undergo condensation polymerization, yielding amorphous silica precursors characterized by relatively weak Si–O–Si connectivity [Eqs. (1)–(4)]. Under alkaline activation conditions, Si–O–Si bonds undergo hydrolytic cleavage and transform into Si–O–Na linkages, producing soluble sodium silicate intermediates [Eqs. (5)–(9)] [23]. Then, using a certain template agent, mesoporous SiO<sub>2</sub> CO<sub>2</sub> capture materials are prepared via a hydrothermal self-assembly process.



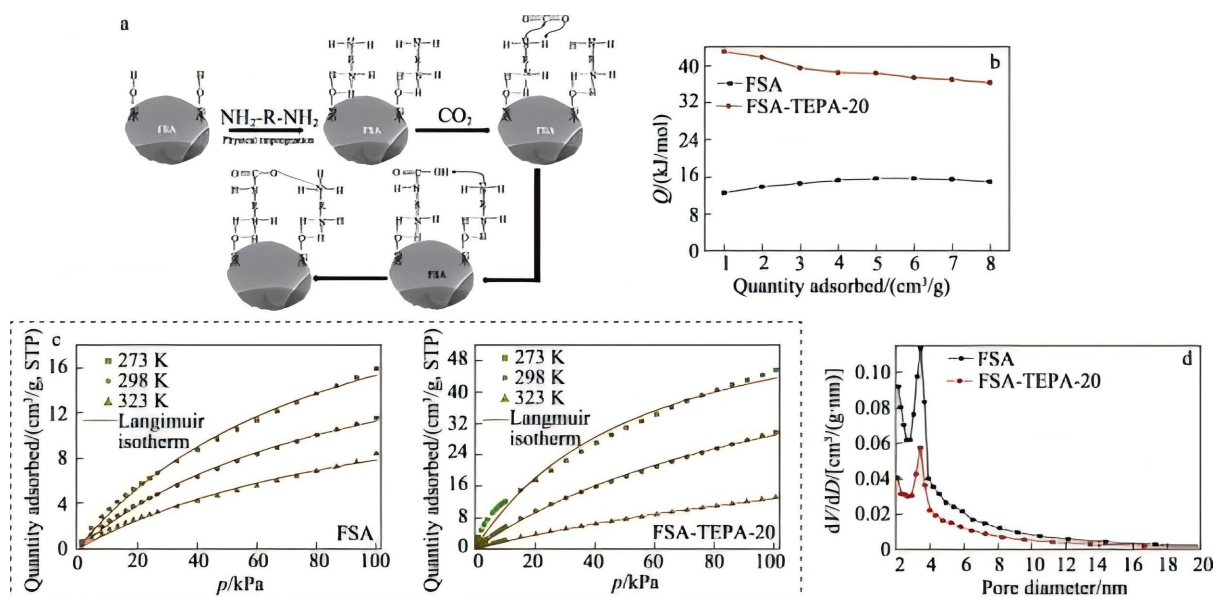
In recent years, to improve the capture capacity of silicon-based CO<sub>2</sub> capture materials, a series of hierarchical porous SiO<sub>2</sub> CO<sub>2</sub> capture materials with micropores, mesopores, and macropores have been developed using gasification slag as a raw material. The hierarchical porous architecture incorporating diverse pore dimensions not only amplifies the specific surface area and pore volume of silicon-based CO<sub>2</sub> adsorbents, thereby furnishing additional active sorption sites, but also attenuates CO<sub>2</sub> diffusion mass transfer resistance through the presence of mesoporous and macroporous channels, consequently enhancing overall CO<sub>2</sub> capture capacity. Wei et al. [23] employed sodium silicate solution derived from sequential acid leaching and alkaline dissolution of gasification fine slag as the silicon precursor, utilizing Pluronic triblock copolymer (PEO<sub>20</sub>-PPO<sub>70</sub>-PEO<sub>20</sub>) as the organic structure-directing agent. Self-assembly was carried out in a ternary hydrothermal system of Pluronic polymer, HCl, and H<sub>2</sub>O, followed by high-temperature calcination to remove the organic template, preparing a hierarchical porous nano-silica adsorbent (CGFS, Figure 3a). When the alkali-to-slag mass ratio was 0.6, the prepared adsorbent (CGFS-0.6) had a specific surface area of 457.17 m<sup>2</sup>/g and a pore volume of 2.34 cm<sup>3</sup>/g. At 20 °C, 0.1 MPa, and a CO<sub>2</sub> volume fraction of 15%, it reached adsorption equilibrium within only 10 min, with a CO<sub>2</sub> adsorption capacity of 2.87 mmol/g. After 20 adsorption-desorption cycles, it retained 89.90%–96.47% of its adsorption capacity, demonstrating high adsorption-desorption cyclic stability (Figures 3b–e). GAO et al.[24] used pre-decarbonized gasification fine slag (ash mass fraction 99.82%) as a raw material. After alkali dissolution to obtain a silicon-containing precursor, acidification with HCl caused Si<sup>4+</sup> in the solution to re-polymerize into nanoscale SiO<sub>2</sub> particles, which gradually bonded and cross-linked via condensation reactions to form a three-dimensional network structured SiO<sub>2</sub> wet gel. Drying at 105 °C under atmospheric pressure yielded hierarchical porous SiO<sub>2</sub> aerogels, avoiding the complexity and risks associated with the traditional supercritical drying method for SiO<sub>2</sub> wet gels. Under optimized process parameters, the synthesized material exhibited a specific surface area of 643.48 m<sup>2</sup>/g and total pore volume of 0.88 cm<sup>3</sup>/g. The CO<sub>2</sub> sorption capacity at 273 K attained 1.53 mmol/g, with merely 5.22% capacity attenuation following five consecutive adsorption-desorption cycles, demonstrating excellent regenerative stability. However, the aforementioned porous SiO<sub>2</sub> CO<sub>2</sub> capture materials mainly capture CO<sub>2</sub> through the physical adsorption mechanism of the microporous structure. Under actual flue gas conditions with higher temperatures and lower CO<sub>2</sub> partial pressures, their CO<sub>2</sub> capture capacity is poor, limiting their practical application.



**Figure 3** Schematic diagram of preparation process of hierarchical porous silica nanoparticles from coal gasification slag(a), SEM images of CGFS(b) and CGFS-0.6(c), pore size distribution curves of different samples(d) and breakthrough curves of CGFS-0.6 at different CO<sub>2</sub> volume fractions(e)[23]

At higher adsorption temperatures, the CO<sub>2</sub> capture performance of carbon capture materials relies more on surface chemical properties rather than micropore structure. Studies have shown that amine-functionalized modification of silicon-based CO<sub>2</sub> capture materials via physical impregnation, chemical grafting, and in-situ

polymerization can effectively increase the surface basicity and electron density of the materials. This enhances the Lewis acid-base interaction between CO<sub>2</sub> and the modified basic groups. The surface-loaded amino groups can react reversibly with adsorbed CO<sub>2</sub> to form ammonium carbamate or ammonium bicarbonate, effectively improving the CO<sub>2</sub> capture capacity and selectivity of the carbon capture materials through chemical adsorption[25]. To elucidate the influence of organic amine identity and loading level on the CO<sub>2</sub> capture performance of silicon-based adsorbents derived from gasification slag, Zhang et al. [26] employed acid-leached, impurity-removed gasification slag (FSA) as the precursor material and synthesized a series of amino-functionalized FSA sorbents through physical impregnation using ethylenediamine, diethylenetriamine, triethylenetetramine, and tetraethylenepentamine. The fabrication protocol and CO<sub>2</sub> sorption characteristics are illustrated in Figure 4.



**Figure 4** Schematic diagram of preparation mechanism of amino-modified FSA adsorbent(a); Isothermic heats of CO<sub>2</sub> adsorption of FSA-TEPA-20(b); CO<sub>2</sub> adsorption isotherms(c) and pore size distribution curves(d) of FSA and FSA-TEPA-20[26]

The findings demonstrated that escalating organic amine molecular weight correlated with increased amino group loading density, expanded chemisorption site availability, and enhanced CO<sub>2</sub> capture capacity. Nevertheless, excessive amine incorporation engendered pore blockage within the FSA framework, impeding intraporous CO<sub>2</sub> diffusion and consequently diminishing sorption performance. Therefore, identifying an optimal amine loading to harmonize pore structural attributes with surface basicity represents a critical determinant for maximizing CO<sub>2</sub> capture efficiency. At 20% TEPA mass fraction loading, FSA-TEPA-20 achieved superior performance, exhibiting a specific surface area of 185 m<sup>2</sup>/g and pore volume of 0.543 cm<sup>3</sup>/g. Under conditions of 298 K and 5% CO<sub>2</sub> volumetric concentration, the CO<sub>2</sub> sorption capacity reached 1.32 mmol/g, corresponding to a 156.39% enhancement relative to pristine FSA. Following ten adsorption-desorption cycles, CO<sub>2</sub> capacity declined by 10.61%, attributable to amine volatilization and degradation during thermal regeneration. Adsorption kinetic analysis and isosteric heat calculations revealed that CO<sub>2</sub> uptake on the modified material transitioned from purely physisorption to a hybrid physisorption-chemisorption mechanism, with an increasingly dominant chemisorption contribution. Although the physical impregnation method is simple to operate and allows high amine loading, the organic amines are mainly loaded onto the carrier via van der Waals forces and electrostatic interactions, resulting in poor thermal stability and susceptibility to agglomeration that blocks pores. SUN Jiajun et al.[27] adopted a chemical grafting method to graft 3-aminopropyltriethoxysilane (APTES) onto acid-leached slag (AFS) prepared from hydrochloric acid-treated coal gasification slag, producing an amino-functionalized adsorbent (AFS-A- $x$ , where  $x$  is the addition amount of APTES in mL) with excellent CO<sub>2</sub> capture capacity under low CO<sub>2</sub> pressure. Acidic leaching not only dissolved the majority of metal oxide constituents, thereby enriching the porous architecture of the material, but also induced scission of Si-O-Al linkages within the gasification slag matrix during the process, generating nascent

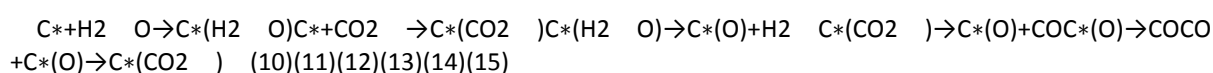
Si–OH functionalities favorable for subsequent amino group grafting. At 60 °C and 0.1 MPa, the CO<sub>2</sub> adsorption capacity of AFS-A-3 reached 0.61 mmol/g, a 19.6% increase compared to AFS, and after 10 adsorption/desorption cycles, the product's adsorption capacity decreased by only 5%, demonstrating good adsorption cyclic stability. Although organic amine modification can effectively improve the CO<sub>2</sub> capture capacity of silicon-based carbon capture materials, problems such as easy blockage of carrier pores, severe amine loss, and low adsorption capacity remain. It is necessary to improve the spatial structure of silicon-based CO<sub>2</sub> capture materials, study methods for stably loading organic amines on the surface of silicon-based CO<sub>2</sub> capture materials, and enhance the CO<sub>2</sub> capture capacity of silicon-based CO<sub>2</sub> capture materials derived from gasification slag.

## 2.2 Carbon-Based CO<sub>2</sub> Capture Materials

Activated carbon, as one of the most commonly used CO<sub>2</sub> capture materials, offers advantages such as wide raw material availability, low cost, well-developed pore structure, and wide application range, with easily modifiable surface groups, and is widely used in industrial adsorption separation processes[28]. Following coal gasification, the residual carbon fraction within gasification slag contains minimal uncombusted organic matter and volatile components, while exhibiting substantial specific surface area and microporous volume. In contrast to conventional activated carbon feedstocks such as coal, biomass, and synthetic polymers, activated carbon derived from gasification slag residual carbon obviates the carbonization step, thereby streamlining the overall process. Nevertheless, ash particulates in gasification slag frequently coat the residual carbon surface or become entrapped within its pore network, causing pore occlusion. Consequently, preliminary separation and enrichment of residual carbon through screening, gravity concentration, froth flotation, or acid leaching is imperative [29]. In addition, the high temperature of the coal gasification process causes carbon structure polycondensation, crystallite growth, and aliphatic side chain scission, resulting in a high graphitization degree of the residual carbon, low surface functional group content, and limited CO<sub>2</sub> adsorption sites[30]. Hence, subsequent physical and/or chemical activation treatments are necessary to modulate the specific surface area, pore architecture, and surface chemical characteristics of the enriched residual carbon fraction, thereby fabricating activated carbon materials with enhanced CO<sub>2</sub> capture performance.

### 2.2.1 Physical Activation

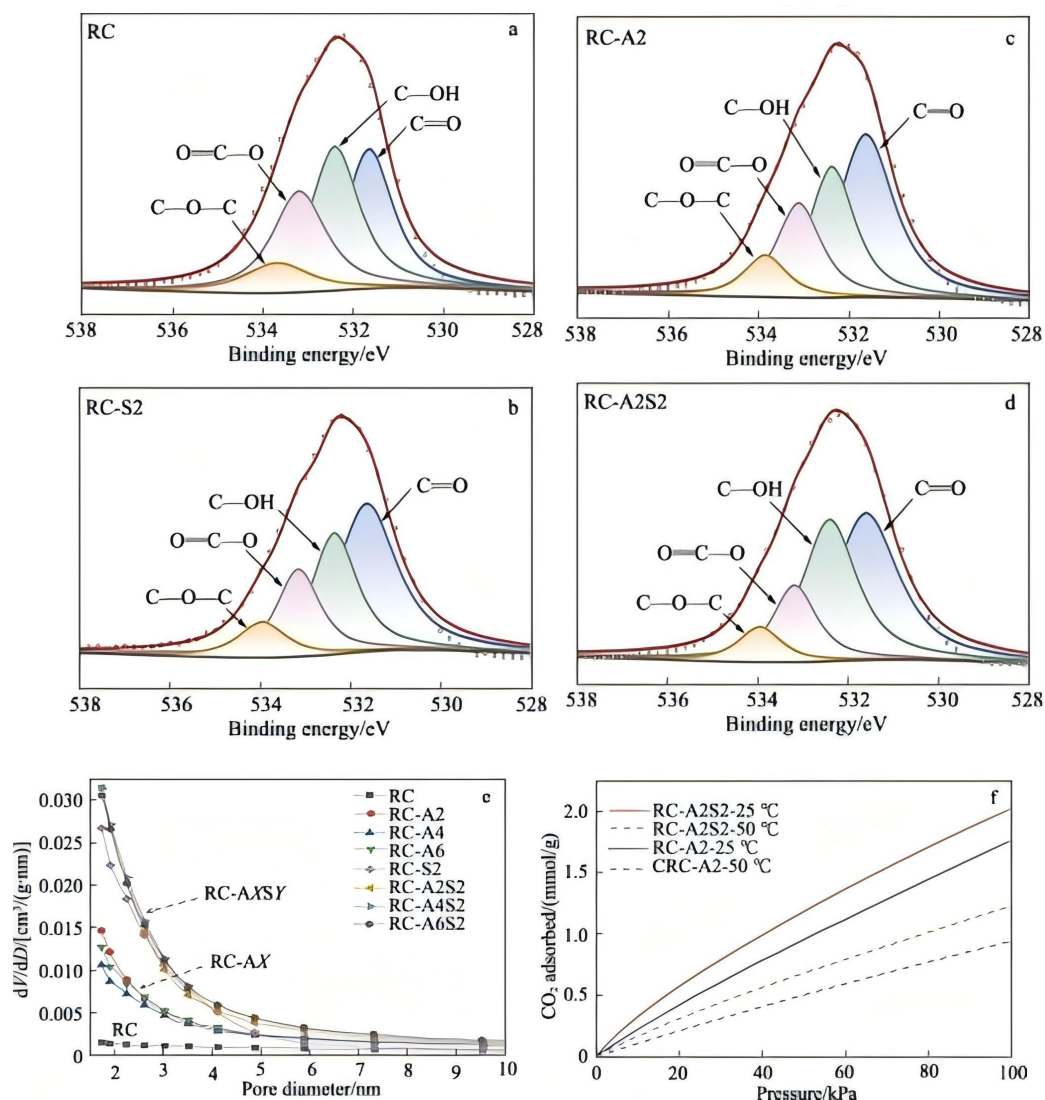
Physical activation refers to the use of oxidative gases such as water vapor, CO<sub>2</sub>, air, or their mixtures as activating agents. At temperatures of 300–800 °C, redox reactions occur between the activating agents and carbon atoms in the residual carbon lattice of gasification slag, causing carbon atoms to escape in gaseous form. This creates “cavities” at the original active sites, forming a large number of pores, thereby achieving pore creation and expansion of the raw material. Simultaneously, physical activation can also remove residual inorganic minerals and tar produced during the gasification process, and introduce active oxygen-containing functional groups by oxidizing the carbon surface via the oxidizing agent, thereby improving the structural characteristics and surface properties of the residual carbon to enhance the CO<sub>2</sub> capture capacity of carbon-based CO<sub>2</sub> capture materials. During physical activation, gaseous activating agents such as water vapor and carbon dioxide diffuse toward and adsorb onto surface active sites of the residual carbon fraction at relatively moderate temperatures. With progressive elevation of activation temperature, carbon atom reactivity intensifies, promoting reaction with adsorbed water vapor or CO<sub>2</sub> to yield transient oxygenated intermediates of limited stability. These intermediates further decompose to produce CO, or react with CO to generate CO<sub>2</sub>, which then desorbs and diffuses into the gas stream, forming a large number of new micropores within the residual carbon and creating more active sites for continued reaction. The specific reaction mechanisms are shown in Eqs. (10)–(15)[31].



Where: C\* represents the active site on the residual carbon lattice of gasification slag; C\*(H<sub>2</sub>O) and C\*(CO<sub>2</sub>) represent H<sub>2</sub>O and CO<sub>2</sub> attached to the active sites of gasification slag residual carbon, respectively; C\*(O) represents the unstable oxygen-containing intermediate.

The effectiveness of physical activation is usually influenced by factors such as the type of activating agent,

activation temperature, reaction time, and activating agent flow rate.



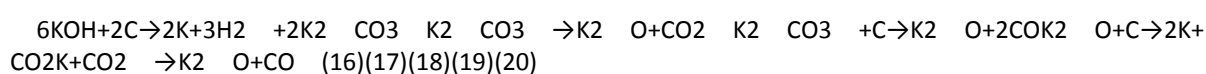
**Figure 5** Effects of RC, RC-A2, RC-S2, RC-A2S2 on surface functional groups(a~d), pore volume(e) and CO<sub>2</sub> adsorption capacity(f) of residue carbon from coal gasification slag[32]

REN et al.[32] used residual carbon (RC, ignition loss 91.29%) separated and enriched from gasification fine slag via ultrasound-assisted flotation as a raw material. They activated the residual carbon using air (300 °C), water vapor (600 °C), and air-water vapor mixtures, respectively, to prepare a series of gasification slag-based activated carbon CO<sub>2</sub> capture materials, and investigated the effects of different process parameters on the CO<sub>2</sub> capture capacity of the products (Figure 5). The results showed that compared with air activation, water vapor has smaller molecular dimensions, faster diffusion rates in residual carbon pores, lower reaction activation energy with carbon, and stronger reactivity. Therefore, more intense oxidation reactions occurred in RC under water vapor activation, leading to more significant improvements in specific surface area and total pore volume. Compared with unactivated RC, after 2 h of activation with air, water vapor, and air-water vapor mixtures, the specific surface areas of the residual carbon (denoted as RC-A2, RC-S2, and RC-A2S2, respectively) increased from 12.00 m<sup>2</sup>/g to 108.00, 289.24, and 327.03 m<sup>2</sup>/g, respectively, and the total pore volumes increased from 0.04 cm<sup>3</sup>/g to 0.12, 0.20, and 0.24 cm<sup>3</sup>/g, respectively. At 25 °C and 0.1 MPa, RC-A2S2 exhibited the maximum CO<sub>2</sub> adsorption capacity, reaching 2.00 mmol/g. GAO et al.[33] used dried gasification fine slag (CGFS) as a raw material and prepared activated carbon CO<sub>2</sub> capture materials via a one-step low-temperature air activation method. During the low-temperature air activation at 485 °C, slight oxidation of the carbon matrix, combustion of ash and tar blocking the pores, and decomposition of some inorganic mineral carbonates opened up blocked

pore channels. Compared with CGFS, after 20 min of low-temperature air activation, the specific surface area and pore volume of the sample (CGFS-20) increased from 247.53 m<sup>2</sup>/g and 0.2895 cm<sup>3</sup>/g to 300.84 m<sup>2</sup>/g and 0.3093 cm<sup>3</sup>/g, representing improvements of 21.5% and 6.8%, respectively. At 25 °C, the maximum theoretical CO<sub>2</sub> adsorption capacity of CGFS-20 reached 0.94 mmol/g. The physical activation method features a simple process flow, low cost, minimal environmental pollution, no equipment corrosion, and the produced activated carbon can be used directly without washing, facilitating large-scale promotion and application. However, gaseous activating agents such as water vapor and CO<sub>2</sub> have relatively weak reactivity; the activation process usually takes a long time, consumes high energy, and cannot form a rich pore structure. Therefore, the specific surface area of the products is often lower than that from chemical activation methods, resulting in relatively lower CO<sub>2</sub> capture capacity.

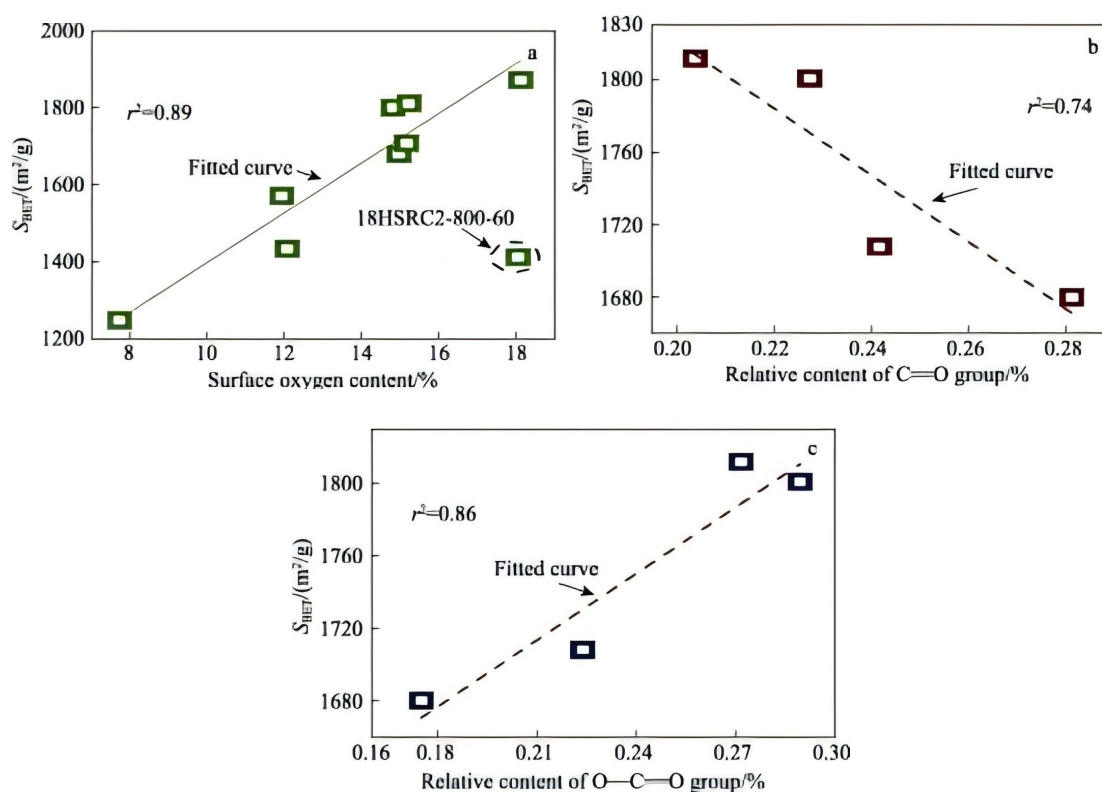
### 2.2.2 Chemical Activation

Chemical activation entails blending or impregnating gasification slag residual carbon with a chemical activating agent at a predetermined proportion, enabling the activator to permeate the internal structure of the carbonaceous material. Subsequent high-temperature activation under inert atmosphere is followed by exhaustive washing to eliminate residual activator and contaminants, yielding the final activated carbon product. Chemical activation not only facilitates the development of the residual carbon pore structure but also introduces basic groups, increasing the surface basicity of the carbon and enhancing the material's CO<sub>2</sub> capture capacity. Commonly used chemical activating agents mainly include strong alkalis (KOH, NaOH), strong acids (HCl, HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, H<sub>3</sub>PO<sub>4</sub>), and salts (ZnCl<sub>2</sub>, K<sub>2</sub>CO<sub>3</sub>, CaCl<sub>2</sub>), among which KOH is the most frequently used [34]. Because of the small radius and low electronegativity of K, the specific surface area of activated carbon after KOH activation increases by approximately 4 times compared to other activation methods [35]. In the KOH activation protocol, the residual carbon fraction is initially immersed in potassium hydroxide solution for a defined duration to ensure complete diffusion of the activator onto the carbon surface and throughout its porous network. Then, under molten conditions induced by heating, a series of redox reactions between KOH and C consume carbon atoms, continuously etching the carbon material and forming new microporous structures. The specific activation mechanisms are shown in Eqs. (16)–(20) [36]. During the activation process, elemental K generated can intercalate into the carbon lattice in vapor form, widening the interlayer spacing of carbon and forming pore structures [37–38]. Additionally, H<sub>2</sub>O and CO<sub>2</sub> evolved from the KOH-carbon reaction can further activate the carbon framework through physical activation pathways as described in Eqs. (10)–(15), generating a synergistic enhancement effect. Miao Zekai et al. [39] employed residual carbon from gasification fine slag (ash content <1.5%) obtained through froth flotation and acid washing for deashing as the precursor material, utilizing KOH as the chemical activator to fabricate a series of activated carbon CO<sub>2</sub> capture sorbents via high-temperature chemical activation. They systematically examined the influence of activation temperature, activator-to-carbon ratio, and activation duration on the porous architecture and CO<sub>2</sub> capture performance of the resulting activated carbons. The findings demonstrated that under optimal conditions of 800 °C, KOH-to-residual carbon mass ratio of 2:1, and 90 min activation time, the synthesized activated carbon exhibited abundant macroporous surface features with micropores and mesopores distributed along macropore walls, collectively constituting an interconnected hierarchical pore network spanning macro-, meso-, and microporous regimes. The specific surface area and total pore volume attained 1596 m<sup>2</sup>/g and 1.297 cm<sup>3</sup>/g, respectively. The optimized product demonstrated exceptionally rapid adsorption kinetics, approaching saturation within approximately 1.5 min. At 0, 25, and 50 °C, the CO<sub>2</sub> sorption capacities reached 4.90, 2.75, and 0.86 mmol/g, respectively. Following ten consecutive adsorption-desorption cycles, no appreciable capacity degradation was observed. Furthermore, the CO<sub>2</sub> capture performance across these three temperatures exhibited strong correlation with narrow micropore (<1.5 nm) volume. The aperture dimensions of these ultrafine micropores approximate the kinetic diameter of CO<sub>2</sub> molecules (0.33 nm), which amplifies van der Waals interactions and electrostatic forces between pore walls and CO<sub>2</sub> molecules, thereby governing the adsorption capacity of the activated carbon sorbent.

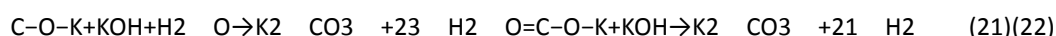


Pre-oxidation treatment of carbonaceous precursors represents an effective strategy for generating abundant porous architectures and enhancing CO<sub>2</sub> capture performance in activated carbons. Through the introduction

of oxygenated surface functionalities onto carbon materials, these groups subsequently react with KOH to generate novel active centers (C–O–K). C–O–K reacts with KOH to generate K<sub>2</sub>CO<sub>3</sub> [Eqs. (21)–(22)], promoting the penetration of KOH into deeper layers of the carbon structure[40–41]. Based on this, MIAO et al.[42] used HNO<sub>3</sub>, NaClO, H<sub>2</sub>O<sub>2</sub>, and air as pre-oxidizing agents to pre-oxidize gasification slag residual carbon, and investigated the effects of different types of pre-oxidation treatments on the pore structure characteristics and CO<sub>2</sub> capture capacity of activated carbon. In comparison with unmodified residual carbon, the surface abundance of C=O and C(=O)–OH moieties on pre-oxidized residual carbon increased markedly, whereas C–OH, C–O–C, and O–C=O functionalities diminished. Correlation analysis between C=O and O–C=O surface concentrations across diverse carbon precursors and the resultant activated carbon specific surface area (Figure 6) demonstrated that O–C=O content exhibited positive association with specific surface area development, while C=O presence constrained pore structural evolution. This may be because O–C=O is acidic and can better react with KOH to form C–O–K active sites, strengthening the chemical activation process via Eqs. (21)–(22); conversely, basic C=O limits the KOH activation process. Compared with samples without pre-oxidation treatment, the activated carbon pre-oxidized with HNO<sub>3</sub> and with an oxygen mass fraction of 15% (15 NARC) had the largest specific surface area and total pore volume, increasing from 1246.00 m<sup>2</sup>/g and 0.88 cm<sup>3</sup>/g for the untreated sample (RC) to 1812.00 m<sup>2</sup>/g and 1.37 cm<sup>3</sup>/g, respectively. At 25 °C and 0.1 MPa, its CO<sub>2</sub> adsorption capacity was maximized, reaching 3.36 mmol/g, and it exhibited a high adsorption rate, achieving 86% adsorption efficiency within 1 min. Although pre-oxidation treatment can effectively improve the pore structure and CO<sub>2</sub> capture capacity of activated carbon, it easily causes carbon loss, leading to a low activated carbon yield of only 42%–56%.



**Figure 6** Relationship between surface oxygen content(a), C=O group content(b) and O–C=O group content(c) and surface area of activated carbon after different pre-oxidation treatment[42]



The formation of the pore structure of gasification slag-based CO<sub>2</sub> capture materials not only stems from the activation process but also from the acid washing process after activation. Acid washing can further improve the pore structure of the product by removing potassium-containing compounds formed during activation and dissolving inorganic mineral particles. MIAO et al.[43] investigated the effects of different acid washing

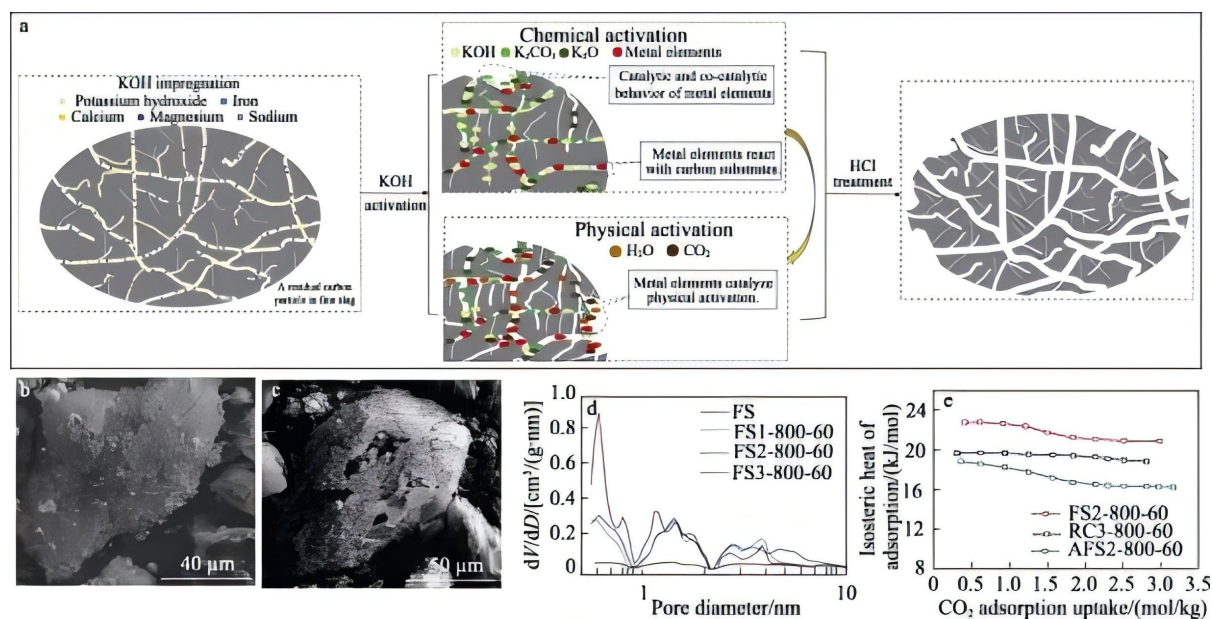
conditions (acid type, acid washing temperature, acid washing time, acid concentration) on the pore structure of porous carbon materials after alkali melting treatment via orthogonal experiments. The results showed that acid treatment to remove inorganic minerals in the micropores of residual carbon is a diffusion-controlled process. Low-temperature, low-concentration acid treatment removes most of the inorganic minerals in the mesopores. With escalating treatment temperature and acid concentration, the acidic medium progressively permeates into microporous channels to dissolve entrapped mineral constituents, thereby exposing previously occluded pores. Post-activation, the carbon capture materials exhibited near-complete elimination of inorganic mineral particulates from both surface and intraporous regions, developing a more refined microporous architecture while preserving the porous carbon framework established during activation. Under optimal conditions of 15% H<sub>2</sub>SO<sub>4</sub> at 25 °C for 4 h, the product attained a maximum specific surface area of 1295 m<sup>2</sup>/g. At 25 and 50 °C, the material achieved CO<sub>2</sub> sorption capacities of 2.64 and 1.02 mmol/g within 2 min, respectively. The adsorption selectivity for N<sub>2</sub>/CO<sub>2</sub>-dominated flue gas reached 10.8, indicating preferential CO<sub>2</sub> affinity. Following ten consecutive adsorption-desorption cycles, the sorption capacity remained at 97% of its initial value, demonstrating excellent regenerative stability.

Relative to physical activation methodologies, chemical activation presents superior reaction kinetics, more extensively developed porous architectures in the resulting activated carbon, greater specific surface areas, and enhanced suitability for CO<sub>2</sub> capture applications. However, existing research mainly uses KOH as the activating agent, and the cleaning process consumes large amounts of acid, which is difficult to recover, easily causes equipment corrosion and secondary pollution, and increases production costs. Low-cost, non-corrosive, and environmentally friendly chemical reagents should be further developed to improve the economic and environmental benefits of the chemical activation method for preparing activated carbon.

### 2.3 Carbon-Ash Composite CO<sub>2</sub> Capture Materials

Gasification slag contains a large amount of ash, which typically exists in forms such as complete separation, carbon-ash melting, attachment to the residual carbon surface, or embedding within its pores. Using gasification slag to prepare silicon-based or carbon-based CO<sub>2</sub> capture materials usually requires consuming large amounts of flotation reagents and acids for carbon-ash separation and impurity element removal, resulting in high processing costs and low economic benefits. Simultaneously, large amounts of difficult-to-treat wastewater are generated, easily causing secondary pollution, and leading to the waste of abundant resources such as Al, Ca, and Mg, failing to realize the full utilization of valuable resources in gasification slag. Consequently, investigations into the development of composite CO<sub>2</sub> capture materials leveraging the inherent carbon-ash coexistence within gasification slag have garnered escalating research interest, aiming to achieve concurrent valorization of both mineral and carbonaceous fractions. A systematic series of studies by Miao et al. [36,44-46] on mineral-mediated pore generation mechanisms during gasification slag chemical activation revealed that the ash component in gasification fine slag functions as a structural scaffold preventing pore collapse while concurrently facilitating the chemical activation process during fabrication of carbon-ash composite CO<sub>2</sub> sorbents. This mineral fraction thus constitutes a critical determinant governing porous material architecture and ultimate CO<sub>2</sub> capture performance. During the preparation of carbon-ash composite CO<sub>2</sub> capture materials from gasification fine slag via the KOH chemical activation method, KOH diffuses into the pores of residual carbon via impregnation, dissolving part of the Fe, Ca, Mg, Na, K, and other metal elements in the ash, allowing them to disperse evenly on the carbon surface. During the heating process, part of the leached metal elements can bind to the carbon matrix via oxygen atoms to form C–O–M bonds (M = metal element), and convert into low eutectic mixtures such as KFeO<sub>2</sub> and Mg<sub>3</sub>Ca(CO<sub>3</sub>)<sub>4</sub> during the pyrolysis process. These low eutectic mixtures exhibit good fluidity at high temperatures, helping to improve the fluidity of potassium-containing compounds generated from the reaction between KOH and C, allowing them to disperse better on the residual carbon surface and inside pores, promoting the activation reaction and the development of pore structure. Meanwhile, the metal elements in the low eutectic mixtures can catalyze the physical activation process of water vapor and CO<sub>2</sub> generated during the chemical activation process, further enhancing the synergistic effect of physical-chemical activation[48]. MIAO et al.[36] used gasification fine slag (FS) and foam-floated, acid-leached, enriched residual carbon (RC) as raw materials, respectively, to prepare a series of CO<sub>2</sub> capture materials with interconnected macroporous-meso/microporous structures via the KOH activation method. The findings demonstrated that ash incorporation effectively facilitated pore

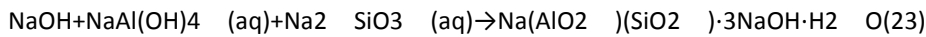
structural development in gasification slag-derived CO<sub>2</sub> capture materials. Under comparable ash content, with an FS-to-KOH mass ratio of 2:1, activation temperature of 800 °C, and activation duration of 60 min, the synthesized carbon capture material (FS2-800-60) attained a specific surface area of 1187 m<sup>2</sup>/g, exceeding the 1114 m<sup>2</sup>/g achieved using residual carbon alone, while reducing activation time by approximately 30 min. Nevertheless, product yield remained comparatively low at 36%–42%. Furthermore, the alkali and alkaline earth metal content within FS furnished supplementary active sites for CO<sub>2</sub> adsorption. At 0 and 25 °C, the FS-based activated carbon achieved CO<sub>2</sub> capture capacities of 4.01 and 2.53 mmol/g within 2 min, respectively. Following five adsorption-desorption cycles, no appreciable capacity degradation was observed, attesting to favorable regenerative stability. The underlying reaction mechanisms and their influence on material architecture and CO<sub>2</sub> capture performance are illustrated in Figure 7.



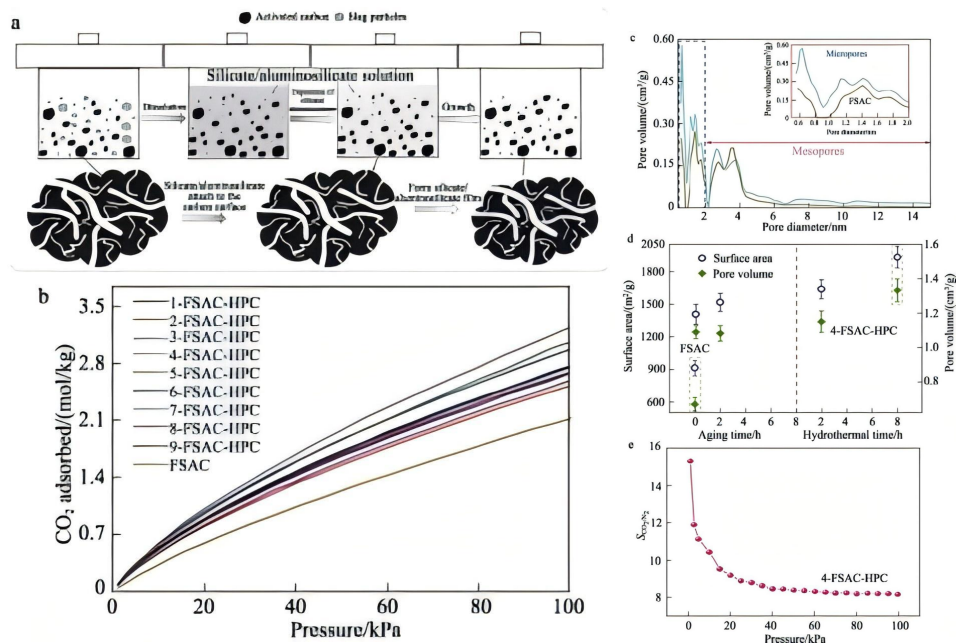
**Figure 7** Schematic diagram of preparation of carbon-ash composite carbon capture materials based on coal gasification slag(a); SEM images of FS(b) and FS2-800-60(c); Pore size distribution curves(d) and isothermic heats of CO<sub>2</sub> adsorption(e) of diverse adsorbents[36]

Mineral particles in gasification slag not only help promote the chemical activation process during the preparation of carbon-ash composite CO<sub>2</sub> capture materials, but the contained SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> can also serve as raw materials for preparing zeolites, synergistically synthesizing porous carbon-zeolite composite adsorbents with residual carbon[49-50]. MIAO et al.[45] mixed KOH-activated samples (FSAC) with NaOH solution, aged for a period, and then subjected them to hydrothermal crystallization. Through 9 sets of orthogonal experiments, they developed a method for preparing gasification slag-based porous carbon-zeolite composite CO<sub>2</sub> capture materials [(x-FSAC-HPC, where x is the orthogonal experiment number)] via chemical activation-hydrothermal treatment (Figure 8). After KOH activation and acid washing, the mineral particles transformed from smooth spherical particles into a layered porous structure with a loose texture. During the NaOH solution aging process, OH<sup>-</sup> ions could enter the interior of mineral particles via pores and react with amorphous aluminosilicates, causing the cleavage of Si–O–Al and Si–O–Si bonds to form new Si–O, Si–OH, Al–O, or Al–OH bonds. These then re-formed silicates or aluminosilicates via interactions of these chemical bonds [Eq. (23)]. Subsequently, the silicates or aluminosilicates continuously grew using nucleation points provided by the porous carbon surface, forming silicate or aluminosilicate films on the surface of the porous carbon substrate. This transformation converted mesoporous and macroporous channels within the porous carbon into microporous and mesoporous regimes, substantially amplifying the specific surface area and pore volume of the resulting materials while furnishing additional active sorption sites for CO<sub>2</sub> molecules. Relative to the FSHPC precursor, the optimized FSHPC-NM specimen exhibited a specific surface area of 2194 m<sup>2</sup>/g and total pore volume of 2.095 cm<sup>3</sup>/g. At 25 °C and 101.325 kPa, the CO<sub>2</sub> adsorption capacity of FSHPC-NM reached a maximum of 3.72 mmol/g, an increase of more than 40% compared to before hydrothermal treatment, and

exhibited a high adsorption rate and adsorption selectivity, with a CO<sub>2</sub> /N<sub>2</sub> adsorption selectivity of 9.5–10.8[44-46]. Adsorption equilibrium could be reached within 3 min, and after 10 adsorption-desorption cycles, 98% of the adsorption capacity was retained. In the process of preparing porous carbon capture materials from gasification slag, large amounts of KOH are often consumed, which is not only costly but also easily causes secondary pollution. To solve this problem, ZHU et al.[51] used deionized water leachate from two types of biomass power plant fly ash (wheat straw and rice husk incineration fly ash) to replace KOH as the alkali and potassium source (potassium carbonate, potassium sulfate, and potassium chloride). They prepared a series of gasification slag-based carbon-ash composite CO<sub>2</sub> capture materials via chemical activation and hydrothermal methods. The results showed that wheat straw incineration fly ash had the highest alkali metal leaching amount and pH. Immersing 10 g of wheat straw power generation fly ash in 50 mL of water yielded a leachate; the introduced alkali and potassium sources along with other metal ions facilitated micropore formation within the material and opened some previously occluded pore channels, generating a well-developed, loofah-like porous architecture that enhanced the material's specific surface area. Relative to the product (CAC) obtained through KOH activation alone, the specific surface area of the fly ash-activated product (CAC-10) increased from 1270 m<sup>2</sup>/g to 1406 m<sup>2</sup>/g. At 25 °C and 101.325 kPa, CAC-10 achieved CO<sub>2</sub> adsorption equilibrium within 3 min, with a maximum sorption capacity of 2.00 mmol/g.



Compared with silicon-based and carbon-based CO<sub>2</sub> capture materials, the preparation of carbon-ash composite CO<sub>2</sub> capture materials can achieve full utilization of gasification slag and reduce by-product generation. However, existing methods still face challenges such as complex process flows, high energy consumption, and difficulty in large-scale utilization.



**Figure 8** Schematic diagram of preparation carbon-ash composite CO<sub>2</sub> capture materials treated by hydrothermal method(a); CO<sub>2</sub> adsorption isotherms(b) and pore distribution curves(c) of diverse samples; Change of specific surface area and pore volume of sample before and after hydrothermal treatment of samples(d); and CO<sub>2</sub> /N<sub>2</sub> selectivity(SCO<sub>2</sub>/N<sub>2</sub>) of 4-FSAC-HPC(e)[45]

## 2.4 Analysis of CO<sub>2</sub> Capture Capacity of Different Carbon Capture Materials

Based on the interaction mechanisms between CO<sub>2</sub> molecules and carbonaceous sorbents, CO<sub>2</sub> capture can be categorized into physisorption and chemisorption pathways. Physisorption predominantly proceeds through

van der Waals interactions, electrostatic forces, and pore volume filling between CO<sub>2</sub> molecules and the internal pore surfaces of carbon capture materials. The sorption capacity is primarily governed by the structural attributes of the sorbent, including pore dimensions, pore volume, and specific surface area [52]. The physisorption process is exothermic in nature; with increasing adsorption temperature, the kinetic energy of CO<sub>2</sub> molecules intensifies, attenuating the intermolecular forces between CO<sub>2</sub> and the carbonaceous substrate, thereby promoting desorption and diminishing physical CO<sub>2</sub> uptake at elevated temperatures [53]. Chemisorption exploits Lewis acid-base interactions and hydrogen bonding between acidic CO<sub>2</sub> molecules and basic surface functionalities on the carbon capture material, undergoing an irreversible exothermic chemical transformation for CO<sub>2</sub> sequestration. The carbon capture performance is mainly affected by the composition of surface functional groups and surface basicity[54]. The CO<sub>2</sub> capture capacity of carbon capture materials is mainly evaluated through indicators such as adsorption capacity, adsorption rate, adsorption cyclic stability, and adsorption selectivity[55]. Table 1 lists the structural characteristics and CO<sub>2</sub> capture performance indicators of different gasification slag-based CO<sub>2</sub> capture materials. As can be seen from Table 1, existing research generally focuses on the adsorption capacity, cyclic stability, and adsorption rate of gasification slag-based CO<sub>2</sub> capture materials, lacking research on their adsorption selectivity. Under certain adsorption conditions, the CO<sub>2</sub> adsorption capacity of gasification slag-based carbon capture materials can reach 0.61–3.72 mmol/g, and adsorption saturation can be achieved within 1.5–10.0 min. After 5–20 adsorption-desorption cycles, they can still maintain a high adsorption capacity. MIAO et al.[56], through correlation analysis between the pore characteristics of gasification slag-based CO<sub>2</sub> capture materials and their carbon capture capacity, found that at 101.325 kPa, the CO<sub>2</sub> adsorption capacity of carbon capture materials had a significant correlation with the pore volume of narrow micropores (<1.5 nm), but had little correlation with total pore volume and specific surface area. Compared with silicon-based CO<sub>2</sub> capture materials, carbon-based and carbon-ash composite CO<sub>2</sub> capture materials have relatively higher adsorption capacities. This may be because carbon-based and carbon-ash composite CO<sub>2</sub> capture materials have relatively higher micropore volumes, and the micropore size better matches the kinetic diameter (0.33 nm) of CO<sub>2</sub> molecules, favoring CO<sub>2</sub> capture. Currently, there are large differences in the test conditions for CO<sub>2</sub> capture performance in the literature, and a unified standard has not yet been formed. Adsorption results cannot be mutually converted, making it impossible to accurately evaluate the CO<sub>2</sub> capture potential of gasification slag. In addition, the CO<sub>2</sub> capture of existing gasification slag-based carbon capture materials is mainly controlled by the physical adsorption mechanism. By selecting appropriate preparation methods and adjusting reaction parameters to regulate the structural characteristics of the materials, their CO<sub>2</sub> capture performance can be improved. As can also be seen from Table 1, under the same adsorption conditions, carbon capture materials after chemical activation have relatively higher specific surface areas, micropore volumes, and CO<sub>2</sub> capture capacities. Among them, the porous carbon-zeolite composite CO<sub>2</sub> capture material (FS-25-HA-HT) prepared from gasification slag via KOH activation and hydrothermal treatment exhibited the maximum CO<sub>2</sub> capture capacity, reaching 3.72 mmol/g. However, owing to variability in feedstock coal properties, gasification reactor configurations, operational parameters, and slag discharge methodologies, the compositional and physicochemical attributes of coal gasification slag exhibit substantial heterogeneity, consequently engendering pronounced disparities in CO<sub>2</sub> capture performance even under identical fabrication conditions. Targeted research needs to be conducted based on different characteristics of gasification slag to determine the corresponding optimal preparation methods for CO<sub>2</sub> capture materials.

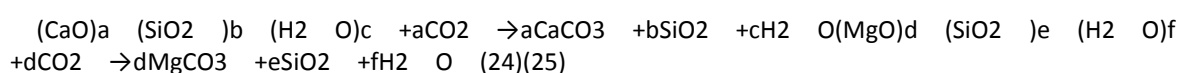
### 3 CO<sub>2</sub> Mineralization Utilization Technologies Based on Gasification Slag

Captured and separated CO<sub>2</sub> needs to be transferred to suitable storage or utilization sites for safe storage via geological sequestration or deep-sea sequestration technologies, or converted into high-value-added chemicals via chemical or biological technologies for utilization. CO<sub>2</sub> mineralization utilization technology refers to the process of reacting captured CO<sub>2</sub> with ores rich in alkaline oxides and hydroxides such as CaO and MgO in nature, or alkaline solid wastes, to convert it into carbonates, facilitating CO<sub>2</sub> sequestration and utilization. Compared with other sequestration technologies, the products of CO<sub>2</sub> mineralization utilization have stable chemical properties, are environmentally friendly, and can achieve permanent storage of CO<sub>2</sub>, effectively avoiding risks associated with traditional geological and ocean sequestration such as CO<sub>2</sub> leakage, groundwater contamination, and ecological damage[57-58]. Meanwhile, purified carbonates obtained from mineralization can be directly sold or converted into high-value-added products, offering higher utilization value. Gasification

slag contains 5%–32% by mass of CaO and MgO with certain reactivity, making it an ideal raw material for CO<sub>2</sub> mineralization utilization. At present, CO<sub>2</sub> mineralization utilization technologies employing gasification slag as feedstock are generally classified into direct mineralization and indirect mineralization pathways based on process routing distinctions.

### 3.1 Direct Mineralization of CO<sub>2</sub> by Gasification Slag

Direct mineralization methods include dry and wet processes. Dry (gas-solid) direct mineralization refers to the direct reaction of CO<sub>2</sub> gas with mineralization raw materials to generate carbonates, requiring no additional chemical reagents. The relative humidity during the gas-solid carbonation process is usually <20%, with low reaction rates and conversion rates. Pretreatment such as grinding and thermal activation of raw materials, or increasing temperature and pressure, is required to accelerate the reaction rate[59]. Since Ca and Mg in gasification slag usually exist in an embedded or encapsulated form with aluminosilicates[60], their reactivity for dry direct mineralization with CO<sub>2</sub> is low, resulting in low carbon conversion rates and CO<sub>2</sub> sequestration capacity. Aqueous-phase direct mineralization can enhance carbonation kinetics and elevate mineralization conversion efficiency through a gas-solid-liquid three-phase reaction framework, constituting a more promising approach among direct mineralization strategies. The wet direct mineralization process occurs in an aqueous environment, where CO<sub>2</sub> first dissolves in water to generate H<sub>2</sub>CO<sub>3</sub>/HCO<sub>3</sub><sup>-</sup>, which then reacts with alkali metals dissociated from the mineralization raw materials in water to form carbonates[61]. ZHANG Yi[62] used ground, dried, and 80-mesh sieved gasification slag (GS) and modified gasification slag (nGS) after ethanol wet ball milling as raw materials to evaluate the wet direct mineralization capacity of gasification slag before and after modification based on the wet direct carbonation reaction. The results showed that after ball milling modification, the particle size of gasification slag decreased, the specific surface area increased, and the original surface structure was destroyed, exposing more calcium-based active sites and increasing the probability of the mineralization reaction between nGS and CO<sub>2</sub>. Compared with GS, the CO<sub>2</sub> mineralization and sequestration capacity of nGS increased from 70.44 g/kg (calculated based on the mass of gasification slag, the same below) to 112.27 g/kg. HUO et al.[63] used Gibbs free energy minimization software to evaluate the CO<sub>2</sub> mineralization capacity of gasification slag slurry with a water-to-cementitious material mass ratio of 0.8 prepared via NaOH activation when used as goaf backfill material. Under alkali excitation, Si–O and Al–O bonds in the gasification slag broke, forming a series of low-polymeric silicon (aluminum) tetrahedron units. These oligomers further dehydrated and condensed, undergoing hydration reactions with Ca<sup>2+</sup> and Mg<sup>2+</sup> to form C–S–H and M–S–H gels, which could further react with CO<sub>2</sub> to generate CaCO<sub>3</sub> and MgCO<sub>3</sub> [Eqs. (24)–(25)]. Under the premise of ensuring the strength of the gasification slag slurry used as goaf backfill paste, when the CO<sub>2</sub> volume fraction was 20%, the theoretical CO<sub>2</sub> storage capacity of coal gasification slag could reach 215.77 g/kg.



In addition, the National Energy Group Guoyuan Electric Power Co., Ltd.[64] is conducting an industrial demonstration project for a 10,000-ton-level CO<sub>2</sub> mineralization absorption coupled with mine fire prevention (extinguishing) materials. Based on coal mine backfill mining technology, coal-based solid wastes (fly ash, coal gangue, bottom slag, desulfurization gypsum, gasification slag, etc.) are used to react with CO<sub>2</sub> for mineralization to excite the activity of the materials and prepare carbon-sequestering backfill cementitious materials. It is expected to sequester 10,000 tons of CO<sub>2</sub> annually.

Furthermore, gasification slag is rich in amorphous SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, and Fe<sub>2</sub>O<sub>3</sub>, resembling the composition of Portland cement, and has good hydration activity and pozzolanic activity. Under certain conditions, its latent cementing ability can be excited, allowing it to be used as an admixture or aggregate. Via a CO<sub>2</sub> mineralization curing process, it can be used to prepare cement-based materials or geopolymers, realizing CO<sub>2</sub> mineralization sequestration. The calcium carbonate crystals precipitated during mineralization can adhere to or become embedded within the calcium-silicate-hydrate gel hydration products, occupying voids and micro-fissures in the interfacial transition zone. This enhances aggregate-matrix interfacial adhesion, mitigates transition zone imperfections, establishes robust bonding, and consequently improves mechanical performance and long-term durability of the composite materials [65]. Wang et al. [66] employed gasification

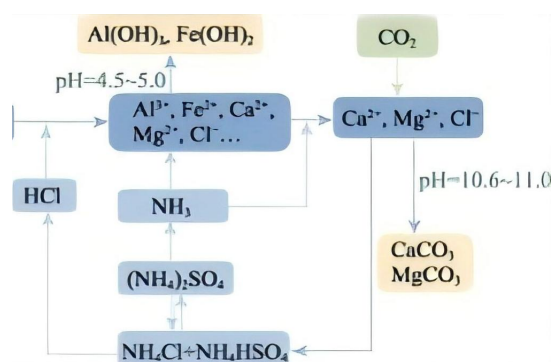
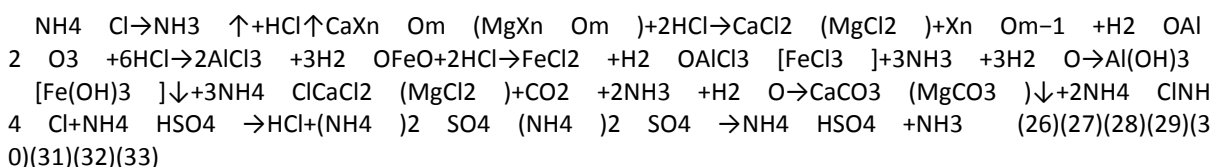
slag and magnesium slag as feedstocks to fabricate a low-carbon composite concrete through CO<sub>2</sub> mineralization curing. Their investigation revealed that silicate mineralization and hydration processes in magnesium slag created favorable conditions for aluminosilicate depolymerization within gasification slag. The aluminosilicate constituents in gasification slag facilitated composite matrix reconstruction and carbonation reactions through generation of C–A–S–H and C–(A–F)–S–H gels, thereby enhancing the mechanical properties of the composite material. At an optimal magnesium slag-to-gasification slag mass ratio of 0.4, the composite concrete achieved maximum compressive and flexural strengths of 38.9 MPa and 9.1 MPa, respectively, following 28 days of CO<sub>2</sub> mineralization curing, with a CaCO<sub>3</sub> yield of approximately 9.21%. Utilizing gasification slag via CO<sub>2</sub> mineralization curing concrete technology for CO<sub>2</sub> capture and utilization is an effective pathway to achieve gasification slag resource utilization and carbon emission reduction. During the CO<sub>2</sub> carbonation process of gasification slag, heavy metals can be co-precipitated and physically adsorbed, thereby preventing heavy metal leaching. However, relevant research remains limited. Future studies should further fully investigate the feasibility and mechanism of gasification slag CO<sub>2</sub> mineralization curing concrete technology, and deeply discuss the effects of mineralization temperature, pressure, alkali concentration, liquid-solid ratio, and other factors on the CO<sub>2</sub> mineralization efficiency of gasification slag.

### 3.2 Indirect Mineralization of CO<sub>2</sub> by Gasification Slag

Indirect CO<sub>2</sub> mineralization technology mainly includes two steps: first, using leaching agents such as HCl, H<sub>2</sub>SO<sub>4</sub>, NaOH, and ammonium salts to extract active components such as Ca<sup>2+</sup> and Mg<sup>2+</sup> from the mineralization raw materials; second, under alkaline conditions, introducing CO<sub>2</sub> into the solution rich in Ca<sup>2+</sup> and Mg<sup>2+</sup> to undergo a carbonation reaction to generate carbonates, realizing CO<sub>2</sub> mineralization utilization [67]. Compared with direct mineralization technology, the indirect mineralization process can remove impurity elements from gasification slag, obtain high-purity, high-value-added carbonate products, and operate under mild reaction conditions. However, using HCl, H<sub>2</sub>SO<sub>4</sub>, NaOH, etc., as leaching agents results in high leaching efficiency but suffers from high acid and alkali consumption, difficulty in extracting agent recovery, and high regeneration energy consumption.

To solve the problem of extracting agent regeneration, the indirect mineralization process based on NH<sub>4</sub>Cl cycling has attracted widespread attention in recent years because it requires no acid/base consumption and allows chemical reagent recycling [68–69]. During the NH<sub>4</sub>Cl cyclic indirect mineralization process, HCl and NH<sub>3</sub> generated from the thermal decomposition of ammonium chloride can be recycled for leaching Ca<sup>2+</sup>, Mg<sup>2+</sup>, etc., from gasification slag and adjusting the pH of the leachate before the carbonation reaction, respectively [Eqs. (26)–(31)]. Li Xiangyu et al. [70] conducted in-depth research on the process of preparing CaCO<sub>3</sub> via a three-step decarbonized gasification slag indirect mineralization cycle comprising Ca<sup>2+</sup>-leaching–CO<sub>2</sub> carbonation–NH<sub>4</sub>Cl recycling. They systematically examined the influence of leaching agent identity, concentration, liquid-to-solid ratio, contact duration, and temperature on calcium extraction efficiency from decarbonized gasification slag, alongside the effects of CO<sub>2</sub> flow rate, reaction temperature, and carbonation duration on both carbonation conversion efficiency and calcium carbonate crystallographic characteristics. Under optimized conditions of 2 mol/L HCl, 20 mL/g liquid-solid ratio, 50 °C reaction temperature, and 30 min contact time, the calcium ion leaching efficiency from decarbonized ash slag attained 96.18%. When CO<sub>2</sub> flow rate was 300 mL/min, reaction temperature 60 °C, and carbonation duration 120 min, the carbonation conversion reached 99.59%. By adjusting the CO<sub>2</sub> flow rate, carbonation reaction temperature, and reaction time, the mutual transformation between different CaCO<sub>3</sub> crystal forms such as vaterite, calcite, and aragonite could be effectively regulated to obtain CaCO<sub>3</sub> products with different crystal structure compositions. However, the aforementioned method still requires the consumption of certain amounts of acid and alkali in the early stage, and does not utilize the large amounts of leached aluminum and iron ions, resulting in resource waste. Yu et al. [71] proposed a new process for CO<sub>2</sub> mineralization utilization by reacting NH<sub>4</sub>HSO<sub>4</sub> with NH<sub>4</sub>Cl to generate HCl and NH<sub>3</sub> under atmospheric pressure and without acid/base consumption, which are used for Ca<sup>2+</sup>-leaching from gasification slag and the carbonation reaction [Figure 9, Eqs. (32)–(33)]. A pilot-scale experiment with a capacity of 1 kg/h (calculated based on CO<sub>2</sub> mass) was conducted using an airlift loop reactor. In this process, based on the difference in solubility product constants (K<sub>sp</sub>) of Al(OH)<sub>3</sub> and Fe(OH)<sub>3</sub>, selective precipitation of Al, Fe, and Ca was achieved by adjusting the pH of the acid leachate to 4.5–5.0 and 10.6–11.0, realizing the synchronous recovery of multiple value-added products including Al(OH)<sub>3</sub>, Fe(OH)<sub>3</sub>, and CaCO<sub>3</sub>. When the CO<sub>2</sub> flow velocity was 1.4

cm/s, pH = 10.6–11.0, and the CO<sub>2</sub> volume fraction was 85%, the carbonation rate reached 91.5%, and the calcium utilization rate reached 75%. Under pilot experimental conditions, per kilogram of gasification slag, this process could generate 344.5 g of crude carbonate (CaCO<sub>3</sub> mass fraction 72.6%) and 328.2 g of aluminum-rich precipitate (Al<sub>2</sub>O<sub>3</sub> mass fraction 47.8%), with a CO<sub>2</sub> mineralization capacity (same basis as above) reaching 121 g/kg. This method requires no acid or alkali throughout the process and realizes the full utilization of the main components of coal gasification slag, greatly improving the economic viability of CO<sub>2</sub> mineralization utilization and possessing great potential for industrial application. In addition, gasification slag is rich in amorphous SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, and Fe<sub>2</sub>O<sub>3</sub>, resembling the composition of Portland cement, and has good hydration activity and pozzolanic activity. Under certain conditions, its latent cementing ability can be excited, allowing it to be used as an admixture or aggregate. Via a CO<sub>2</sub> mineralization curing process, it can be used to prepare cement-based materials or geopolymers, realizing CO<sub>2</sub> mineralization.



**Figure 9** Process flow diagram for indirect mineral carbonation of coal gasification slag based on chemical looping[71]

Although the gasification slag ammonium salt cyclic indirect mineralization process can effectively realize CO<sub>2</sub> mineralization utilization, research on indirect CO<sub>2</sub> mineralization using gasification slag remains scarce. Ammonia escape is inevitable during the ammonium salt cycling process, necessitating further research on the evaluation of mineralization capacity after multiple ammonium salt cycles, mineralization utilization kinetics, and the CO<sub>2</sub> mineralization capacity of different gasification slags. In addition, the overall energy consumption of this process is high, and production costs are elevated, requiring further research on renewable leaching agents and optimization of the process flow to reduce process energy consumption and costs.

## 4 Conclusions and Prospects

The extensive disposal and valorization of gasification slag, coupled with carbon dioxide emission mitigation, constitute critical challenges confronting the sustainable advancement of the coal chemical sector. The abundant Si and C resources in coal gasification slag serve as cost-effective precursors for CO<sub>2</sub> capture materials. After impurity removal, silicon-based, carbon-based, and carbon-ash composite CO<sub>2</sub> capture materials with large specific surface areas, pore volumes, and excellent CO<sub>2</sub> capture capacities can be prepared via hydrothermal templating, physical activation, chemical activation, and organic amine modification. Currently, the CO<sub>2</sub> capture capacity of gasification slag-based carbon capture materials can reach 0.61–3.72 mmol/g (Table 1), and adsorption saturation can be achieved within 1.5–10 min. After 5–20 adsorption-desorption cycles, they still retain a high adsorption capacity. Compared with silicon-based gasification slag CO<sub>2</sub> capture materials, carbon-based and carbon-ash composite CO<sub>2</sub> capture materials exhibit relatively higher CO<sub>2</sub> capture capacities. Meanwhile, CaO, MgO, and other alkaline earth metal elements in gasification slag serve as important raw materials for CO<sub>2</sub> mineralization utilization, enabling CO<sub>2</sub> utilization and sequestration via wet

direct and indirect carbonation technologies. Currently, the CO<sub>2</sub> mineralization capacity can reach 112.27–215.77 g/kg[62-63]. Notably, the acid leachate generated during the preparation of gasification slag-based CO<sub>2</sub> capture materials is rich in Ca, Mg, and other elements, which can be further used as active components for indirect CO<sub>2</sub> mineralization after pH adjustment and recovery of Al, Fe, and other elements. This not only realizes the synchronous recovery and utilization of Si, Ca, C, Mg, Al, Fe, and other resources in gasification slag, but also effectively reduces acid usage and waste liquid generation during the CO<sub>2</sub> capture and mineralization processes, delivering significant economic, environmental, and social benefits. However, research on the coupling technology of gasification slag-based CO<sub>2</sub> capture and mineralization utilization is currently lacking. Although gasification slag shows great potential in the fields of CO<sub>2</sub> capture and utilization, relevant research is still in its infancy, and some shortcomings remain in existing research and large-scale promotion and utilization. In future research on using gasification slag for CO<sub>2</sub> capture and mineralization utilization, efforts should focus on the following aspects:

**(1) Slag-specific strategies for developing gasification slag-based CO<sub>2</sub> capture material preparation and mineralization utilization.** Although different gasification slags all contain elements such as Si, Ca, Mg, and C, influenced by factors such as the type and origin of raw coal, furnace type, gasification process, and slagging method, the composition of different gasification slags varies greatly. Appropriate CO<sub>2</sub> capture material preparation methods should be explored based on the differences in physicochemical characteristics of different gasification slags. The correlations between the physicochemical characteristics of gasification slag and its CO<sub>2</sub> capture and mineralization utilization capacities should be established to quantitatively evaluate its CO<sub>2</sub> capture and mineralization potentials, fully exploiting the CO<sub>2</sub> capture and mineralization utilization potential of gasification slag.

**(2) Developing new strategies for preparing gasification slag-based CO<sub>2</sub> capture materials with excellent yield, adsorption capacity, selectivity, and impurity gas tolerance.** Existing preparation methods for gasification slag-based CO<sub>2</sub> capture materials usually consume large amounts of strong acids and strong alkalis, and generally suffer from problems such as complex processes, high energy consumption, low product yield, susceptibility to equipment corrosion, and secondary pollution. Moreover, there is a lack of evaluation on the CO<sub>2</sub> capture capacity and selectivity from flue gas containing impurities such as water vapor, dust, and sulfides. Based on a deep understanding of the mechanisms of gasification slag for CO<sub>2</sub> capture material preparation and carbonation, feasibility studies should be conducted on using weakly acidic and saline substances with low regeneration energy consumption and environmental friendliness to replace strong acids and strong alkalis for gasification slag-based CO<sub>2</sub> capture and mineralization utilization. Combined with CO<sub>2</sub> adsorption kinetics research, the structure and preparation methods of CO<sub>2</sub> capture materials should be designed and regulated.

**(3) Acid leaching treatment serves as both the impurity removal process in the preparation of silicon-based and carbon-based CO<sub>2</sub> capture materials from gasification slag and the first step of indirect CO<sub>2</sub> mineralization.** The acid leachate generated during the preparation of gasification slag-based CO<sub>2</sub> capture materials can be used for CO<sub>2</sub> carbonation after pH adjustment and recovery of Al, Fe, and other elements. This not only realizes the synchronous recovery of Si, Ca, C, Mg, Al, Fe, and other resources in gasification slag, but also effectively reduces acid usage and waste liquid generation during the CO<sub>2</sub> capture and mineralization processes. Further research is needed on the feasibility of coupling gasification slag CO<sub>2</sub> capture and mineralization utilization technologies, and optimization of related technical processes.

**(4) Most existing studies are still limited to the laboratory research stage, and lack quantitative economic feasibility assessments for their application in the CCUS field.** Efforts should be continuously made to translate related technologies from laboratory research to industrial applications, and conduct economic feasibility studies for large-scale application of related technologies, thereby promoting the large-scale promotion and application of gasification slag in the CCUS field.

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