

## Review article

Biomass-based carbon materials for CO<sub>2</sub> capture: A reviewCui Quan <sup>a</sup>, Yingying Zhou <sup>a</sup>, Jiawei Wang <sup>c</sup>, Chunfei Wu <sup>b</sup>, Ningbo Gao <sup>a,\*</sup><sup>a</sup> Xi'an International Joint Research Center for Solid Waste Recycling and Utilization, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an 710049, China<sup>b</sup> School of Chemistry and Chemical Engineering, Queen's University Belfast, Belfast BT7 1NN, UK<sup>c</sup> Department of Chemical Engineering and Applied Chemistry, Aston University, Aston Triangle, Birmingham B4 7ET, UK

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## ABSTRACT

Carbon capture and sequestration technologies are essential to reduce CO<sub>2</sub> emissions which are responsible for global warming. Carbon-based materials can play an important role in the reduction of CO<sub>2</sub> emissions. These materials are normally produced from biomass through technologies such as pyrolysis and hydrothermal carbonization. The type of biomass feedstock and biomass conversion conditions can significantly affect the textural properties and surface chemistry of the carbon materials. Various modification methods such as material activation or N-doping can improve the properties of carbon materials to obtain better CO<sub>2</sub> capture effects. This review summarizes recently reported research in the areas of using biomass-based materials for CO<sub>2</sub> capture. The technologies of biomass conversion to carbon materials and modification of the carbon materials are critically analyzed. Meanwhile, the mechanisms of the CO<sub>2</sub> capture process and research of different modification carbon materials for CO<sub>2</sub> capture are also discussed. Finally, potential future research directions are suggested to promote carbon capture using biomass-based materials.

## 1. Introduction

Global warming is always an urgent environmental problem. With the development of industrial activities, carbon dioxide, the main greenhouse gas, has continually been released into the atmosphere. Fig. 1(a) shows global carbon dioxide emissions from 2014 to 2020. It is evident that global CO<sub>2</sub> emissions are rising yearly, except the CO<sub>2</sub> emission in 2020 was reduced due to the covid-19 pandemic. The contribution of CO<sub>2</sub> emissions is from industrial manufacturing [1]. As shown in Fig. 1(b), fossil fuel combustion is the main source of CO<sub>2</sub> emissions.

Therefore, it is urgent to curb the continuing increase in atmospheric CO<sub>2</sub> concentrations. Various carbon capture and sequestration (CCS) technologies have emerged to reduce the emission of CO<sub>2</sub> [2]. Adsorption is a cost-effective and efficient carbon capture and storage technology. Carbons materials, zeolites and MOF, etc. are the popular adsorbents for carbon capture. Compared with carbon materials, the CO<sub>2</sub> uptake process of zeolites is influenced by the moisture, adsorption temperature, and pressure significantly. Although the selectivity may be enhanced by the specific metals in MOF, the preparation method of MOF is more complicated and costly, resulting in difficulty in

industrialization [3]. The structure of MOF may fail due to its moisture sensitive during CO<sub>2</sub> uptake process. Therefore, carbon materials have obvious superiority for CO<sub>2</sub> capture as they have a wide range of inexpensive sources. The preparation method of carbon materials is simple and easy to control. Its high specific surface area and developed pore structures contribute to excellent CO<sub>2</sub> capture. Additional, carbon materials are not moisture sensitive.

Biomass is an excellent resource for preparing carbon materials. It is abundant, renewable, and low-pollution. Biomass generally refers to any form of organics, including all animals, plants, and microorganisms, such as agricultural waste, animal manure, municipal organic solid waste, etc. Up to now, extensive research has been done on converting biomass into carbon materials. Pyrolysis is the most commonly used biomass thermochemical technology. Biomass is decomposed into biochar, tar, and gas through pyrolysis. Different pyrolysis conditions affect the yield, specific surface area, or surface chemistry of biochar, etc. Yang et al. prepared biochar at 250, 350, 450, 550, and 650 °C, respectively. BET data showed that the specific surface area of biochar increased with the increasing temperatures, while the char yield decreased [4].

The adsorption properties of carbon materials are closely related to textural structures and surface chemistry. Numerous studies have shown

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that pristine biochar is less effective in adsorbing pollutants. Therefore, many modification methods have emerged. Steam activation, acid/alkaline activation, and heteroatom doping et al. are all effective methods [5]. For CO<sub>2</sub> capture, enriching pores and introducing basic sites are keys to enhancing the adsorption capacity of carbon sorbents. N-doping is a typical modification method. The use of polyetherimide (PEI), triethylenephosphoramidate (TEPA), and other N-containing chemicals doped carbon materials allow the introduction of new N groups such as pyridinic N or pyrrolic N [6,7]. Hernández et al. [8] reported a kind of carbon material with high nitrogen content. They thought that pyridinic N and pyrrolic N played important roles in CO<sub>2</sub> capture. These nitrogen-containing groups can attract acidic CO<sub>2</sub> molecules. He et al. doped the chitosan into the carbonized material, and the doped activated carbon had a better CO<sub>2</sub> adsorption capacity of 5.83 mmol g<sup>-1</sup> at 273 K [9]. Metal salts or oxides also are used to modify carbon materials, which can introduce alkaline metal oxides or hydroxides. Therefore, the alkalinity of carbon materials is increased, which facilitates the adsorption of acidic CO<sub>2</sub>. Lahijani et al. [10] impregnated walnut shell biochar with several metal nitrates. The authors reported that Mg-biochar composites showed superior CO<sub>2</sub> adsorption capacity to other metal-biochar composites (Al, Fe, Ni, Ca, Na). In the area of using biomass-based carbon materials for CO<sub>2</sub> capture, a few articles have reviewed the research progress of biochar or activated carbon for CO<sub>2</sub> capture [1,11,12]. However, few reviews comprehensively summarize the preparation and modification methods of biomass-based carbon materials and their application in CO<sub>2</sub> capture.

Hence, this work critically reviews the preparation technologies of biomass-based carbon materials and the conversion conditions influencing the properties of carbon materials. Then, emerging carbon material modification techniques are discussed. Moreover, the application of carbon materials for CO<sub>2</sub> capture and its capture mechanisms are discussed. Finally, future research directions and challenges are discussed.

## 2. Pristine carbon materials from biomass

Biomass sources include plant sources (i.e., straw, rapeseed, sawdust, peanut shell), animal sources (i.e., pig manure, pigskin, wool, fishbone), and municipal waste (i.e., sludge). The main elements of biomass are C, H, and O, which also contain a small amount of N and S [13]. After the proper treatment, biomass can be converted into carbon materials with good adsorption performances. But there are also some biomasses such as rice husk with high Si content, so they are often used to prepare SiC [14]. Therefore, selecting a suitable biomass is the first step in preparing a good carbon-based adsorbent. This chapter mainly reviews biomass thermochemical conversion technologies (pyrolysis, gasification, and hydrothermal carbonization) and the factors affecting the properties of carbon materials (feedstock, temperature, residence time, and heating

rate).

### 2.1. Biomass thermochemical conversion technologies

#### 2.1.1. Pyrolysis

Pyrolysis is the most mature thermochemical conversion technology. Pyrolysis refers to a chemical process in that biomass is decomposed into solid (char), liquid (oil), and gas (syngas) at relatively high temperatures [15]. Lignin, cellulose, and hemicellulose are the typical components of biomass. Fig. 2 shows the temperature range of these components during thermal decomposition. During biomass pyrolysis, the first stage is the evaporation of water. The second stage is the decomposition of hemicellulose at the temperature of 200–350 °C. Then, the cellulose is broken down with the increase in reaction temperature. Lastly, lignin is decomposed at over 400 °C. It should be noted that the whole biomass pyrolysis process is not a simple superposition of single-component pyrolysis.

According to different temperature, heating rate, and residence time, pyrolysis can be divided into slow, fast, and flash pyrolysis. Slow pyrolysis occurs at temperature range of 300–600 °C and a low heating rate of 0.1–1 °C s<sup>-1</sup>, which has a long residence time and the main product is biochar. The biochar yield of slow pyrolysis generally varies from 30%–50% [17]. Fast pyrolysis and flash pyrolysis are effective technologies to obtain bio-oil. The most obvious difference between them is that flash pyrolysis has a very high heating rate (1000 °C s<sup>-1</sup>) and a very short residence time (<1 s) [18]. Therefore, the particle size of feedstock in flash pyrolysis is suggested to be less than 250 μm for better heat transfer [11].

Fig. 3(a) exhibited that fast and flash pyrolysis gave much higher bio-oil yields than slow pyrolysis. This is due to the short residence time of fast and flash pyrolysis, which greatly reduces secondary cracking reactions, thus increasing bio-oil yield. To obtain maximum bio-oil yield, proper pyrolysis temperature is essential. In general, the increase in pyrolysis temperature promotes the production of bio-oil. But too high

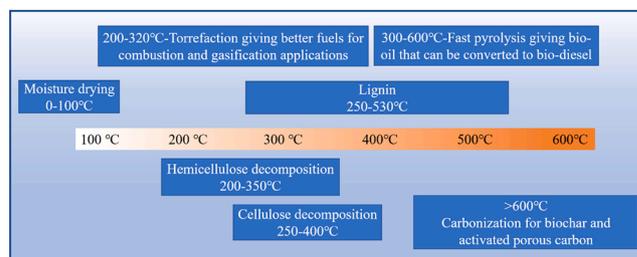


Fig. 2. Thermal decomposition of lignin, cellulose and hemicellulose in biomass [16].

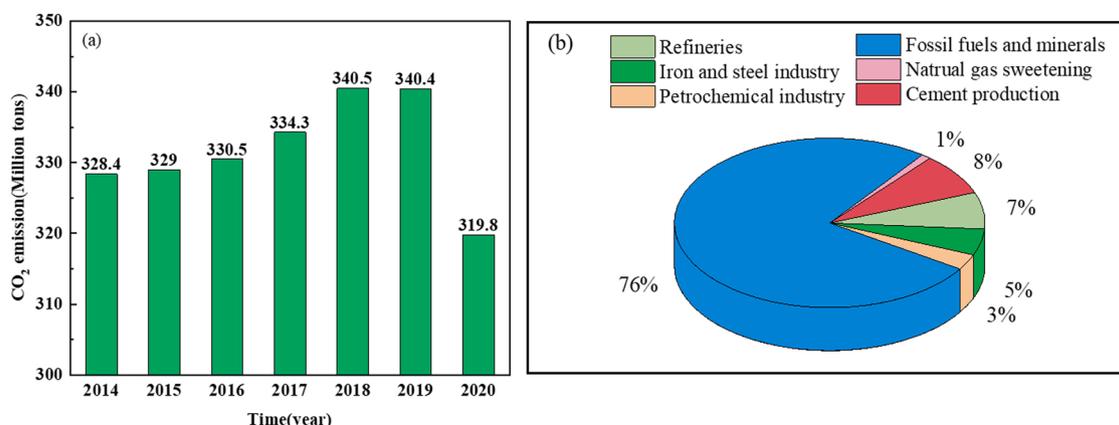


Fig. 1. Status of CO<sub>2</sub> emissions: (a) Global CO<sub>2</sub> emissions from 2014 to 2020; (b) Contribution of each CO<sub>2</sub> emission source [1]

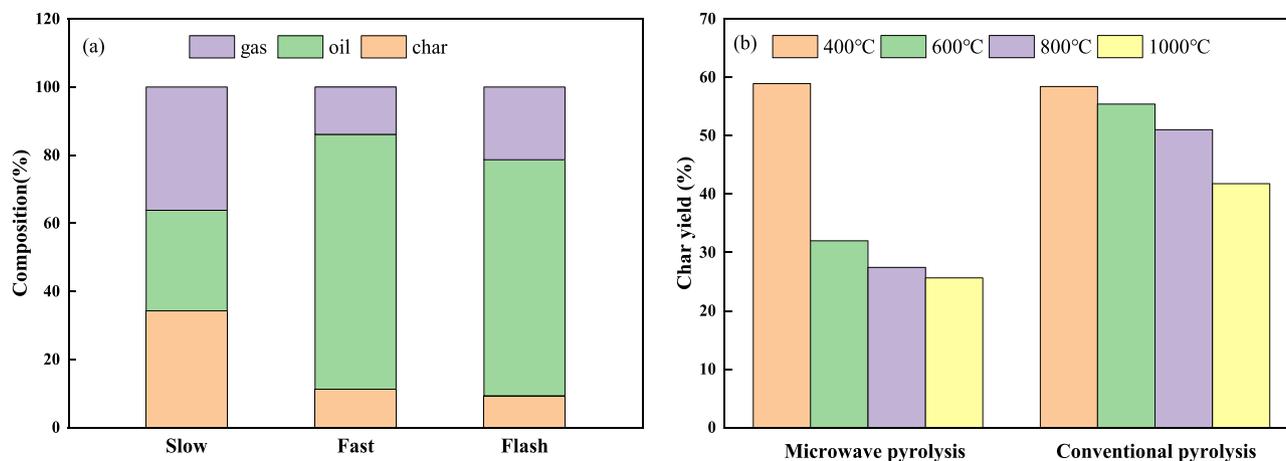


Fig. 3. Comparison of different pyrolysis technologies: (a) slow, fast, and flash pyrolysis; (b) microwave and conventional pyrolysis [20,21].

temperatures are counter-productive. This is because secondary cracking of volatiles can occur at very high temperatures, resulting in increased gas yield and decreased liquid yield. It has been reported that 450–550 °C is a suitable temperature range to obtain maximum bio-oil yield [19].

However, the conventional pyrolysis technologies mentioned above have some defects. The conventional pyrolysis process mainly uses electricity to heat the walls of the pyrolysis furnace and then transfers the heat to the material surface, which requires a long heating time to reach the target temperature. This heating method has low heat transfer efficiency, high heat loss, and requires a long reaction time, resulting in high cost.

Different from conventional pyrolysis, microwave pyrolysis can directly penetrate the material and heat the materials uniformly through the interaction between molecules and an electromagnetic field [22]. It has a fast heating rate and short reaction time, thus increasing production efficiency and reducing production costs. In addition, water is a strong absorbing medium for microwaves, so biomass with high moisture content can be directly microwave-pyrolyzed without pretreatment. Mašek et al. [23] compared the difference between microwave and slow pyrolysis. They found that at the same temperature condition microwave pyrolysis obtain biochar with higher specific surface area and porosity. Nevertheless, the char yield of conventional pyrolysis was higher than that of microwave pyrolysis, as is shown in Fig. 3(b). This is because microwave pyrolysis promotes secondary cracking of biomass and increases gas production, resulting in lower biochar and bio-oil yield.

### 2.1.2. Hydrothermal carbonization

Hydrothermal carbonization (HTC), a low-cost and sustainable thermochemical conversion technology, is known as hydrous pyrolysis. It includes exothermic reactions, which mainly work through dehydration and decarboxylation reactions to reduce the content of O and H in biomass. Specifically, in a closed reactor, with water as the reaction medium, the biomass undergoes chemical reaction processes such as hydrolysis, dehydration, decarboxylation, concentration polymerization, and aromatization.

Hydrothermal carbonization includes high-temperature hydrothermal carbonization (>220 °C, HHTC) and low-temperature hydrothermal carbonization (≤220 °C, LHTC). HHTC mimics the natural formation process of coal to produce 'coal-like' materials [24], while LHTC is prepared under mild conditions and is more widely used in laboratories. The current LHTC technology has been able to produce functional carbon materials with different morphologies, such as honeycomb and fibrous [25]. HTC is often used to treat biomass with high water content, such as municipal sludge, which saves energy consumption to a large extent and makes hydrothermal carbonization technology a green

thermochemical conversion technology.

The main products of biomass from HTC treatment are hydrochar, bio-oil and gas. It has been proved that the obtained hydrochar includes matrix and coke microparticles. The matrix is formed by condensation, dehydration, and decarboxylation of unhydrolyzed biomass, which still maintains the original structure. For the hydrolyzed biomass, the organic molecules first polymerized to form aromatic molecules and then continuously aggregated to form primary micro-nuclei. The surface of the nuclei was rich in -OH, C=O, and -COOH, which can react with organic molecules such as furans in the liquid phase, thus forming the coke microparticles [26].

### 2.2. Factors influencing the properties of carbon materials

Since pyrolysis is the most commonly used biomass conversion technology, this section mainly focuses on the factors that influence the production and properties of carbon materials during pyrolysis. In addition to the effect of biomass itself, the reaction parameters are also critical. Pyrolysis temperature, residence time, heating rate, and the atmospheres all have some effects on carbon materials. These parameters not only influence the char yield but also determine the properties of char, such as surface area, pH et al. Due to the strong association between adsorption capacity and properties of sorbents, it is of great importance to discuss factors affecting carbon materials.

#### 2.2.1. Feedstock

Different biomass has different element compositions, which determines the carbon materials' properties and thus its adsorption characteristics. Feedstocks have a significant effect on carbon materials, such as pH, ash content, and char yield. Biomass can be roughly classified as animal biomass and plant biomass. Animal biomass is abundant in mineral elements (Na, K, Ca, Mg), which can explain animal biochar's high pH and ash content. These mineral elements exist in the forms of  $\text{CO}_3^{2-}$ ,  $\text{PO}_4^{3-}$ ,  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ , which can combine with heavy metals to produce precipitation. Plant biomass is mainly composed of lignin, cellulose, and hemicellulose, which are more likely to form porous carbon structures. Therefore, animal-source biochar is more suitable for removing heavy metals, while carbon materials derived from plant biomass are more suitable for the disposal of organic contaminants and  $\text{CO}_2$  capture [27]. Some studies have shown that lignin plays a dominant role in the formation of carbon materials with excellent properties. Eid Gul et al. [28] reported that pure lignin was a good substrate for producing activated carbon and the quality of biochar produced from lignin-rich biomass was better than that of others. Another study also showed that the lignin-rich biomass resulted in high biochar yields [29]. This is because lignin is heat resistant and does not decompose easily.

In addition, the particle size of feedstocks also should be considered. Biomass with a larger particle size can slow down the heat transfer during pyrolysis, which is beneficial to the formation of carbon materials. Wang et al. [30] investigated the effect of biomass particle size on the structure of biochar. Results showed the specific surface area of biochar increased from 240 to 566 m<sup>2</sup> g<sup>-1</sup> with biomass particle size changing from < 100 μm to 450–2000 μm, and then decreased to 428 m<sup>2</sup> g<sup>-1</sup> at a particle size of 0.5 cm. Therefore, it should be noted that when the particle size is too large, the volatile substances need a longer diffusion path, which may result in pore blockage. Biomass with different particle sizes also affects biochar yields, but most studies suggested that the char yield showed slight change or unclear variable trends. Qureshi et al. [31] prepared biochar from 10 different particle sizes of biomass (0.25–10 mm) and studied the effect of particle size variation on biochar production. The product distribution showed little change and had no clear variation tendency, with char yields of approximately 20%. Therefore, there was no clear mechanism to explain the relationship between particle size and char yield.

### 2.2.2. Temperature

Besides feedstock, the effect of operating conditions also cannot be ignored. Compared with other parameters, the impact of synthesizing temperature on carbon materials is relatively more significant. Generally, as the pyrolysis temperature increases, the specific surface area and pH increase, while the char yield decreases. Yang et al. [4] reported the pyrolysis of rice husk at five different temperatures from 250 to 650 °C. The results showed that the biochar yield decreased from 65.44% to 8.76%, while the pH and surface area increased from 7.64 to 11.15 and from 1 to 14 m<sup>2</sup> g<sup>-1</sup>, respectively. However, the surface and pore

structure of biochar may be destroyed by melting and collapsing at an excessive temperature to decrease the specific surface area. As shown in Fig. 4(a), the char yield decreases with the temperature rising. This may be attributed to the breakdown and rearrangement of C<sub>x</sub>H<sub>y</sub> in biomass that occurs at high temperatures. Fig. 4(b) shows the relationship between pyrolysis temperature and the pH of char. PH is positively correlated with temperature. This is due to the enrichment of alkaline minerals (Na, K) under high-temperature pyrolysis. High pyrolysis temperature also can facilitate the removal of volatile substances, which contribute to forming more developed pore structures, as shown in Fig. 4c and Fig. 4(d). Pooya Lahijani et al. [10] studied the effect of pyrolysis temperature on CO<sub>2</sub> capture. They found that biochar prepared at high pyrolysis temperature has a better CO<sub>2</sub> capture effect. This is because the carbonization temperature increases, the carbon particles are crushed, the average pore size decreases, and micropores are formed. The microporous structure makes a significant contribution to CO<sub>2</sub> adsorption.

### 2.2.3. Residence time

Residence time can be interpreted as the reaction time of biomass carbonization. Generally, the biochar yield may decrease with the prolongation of residence time. This is because the prolonged residence time results in continuous decomposition and release of volatile substances in biomass. However, Zhao et al. [43] pointed out that there was no significant correlation between char yield and residence time. Therefore, the residence time is not an effective factor in determining the char yield. It can be determined that longer residence time promotes the aromatization of biochar highly. Jeong et al. showed that the H/C and O/C of biochar decreased gradually with the prolonged residence

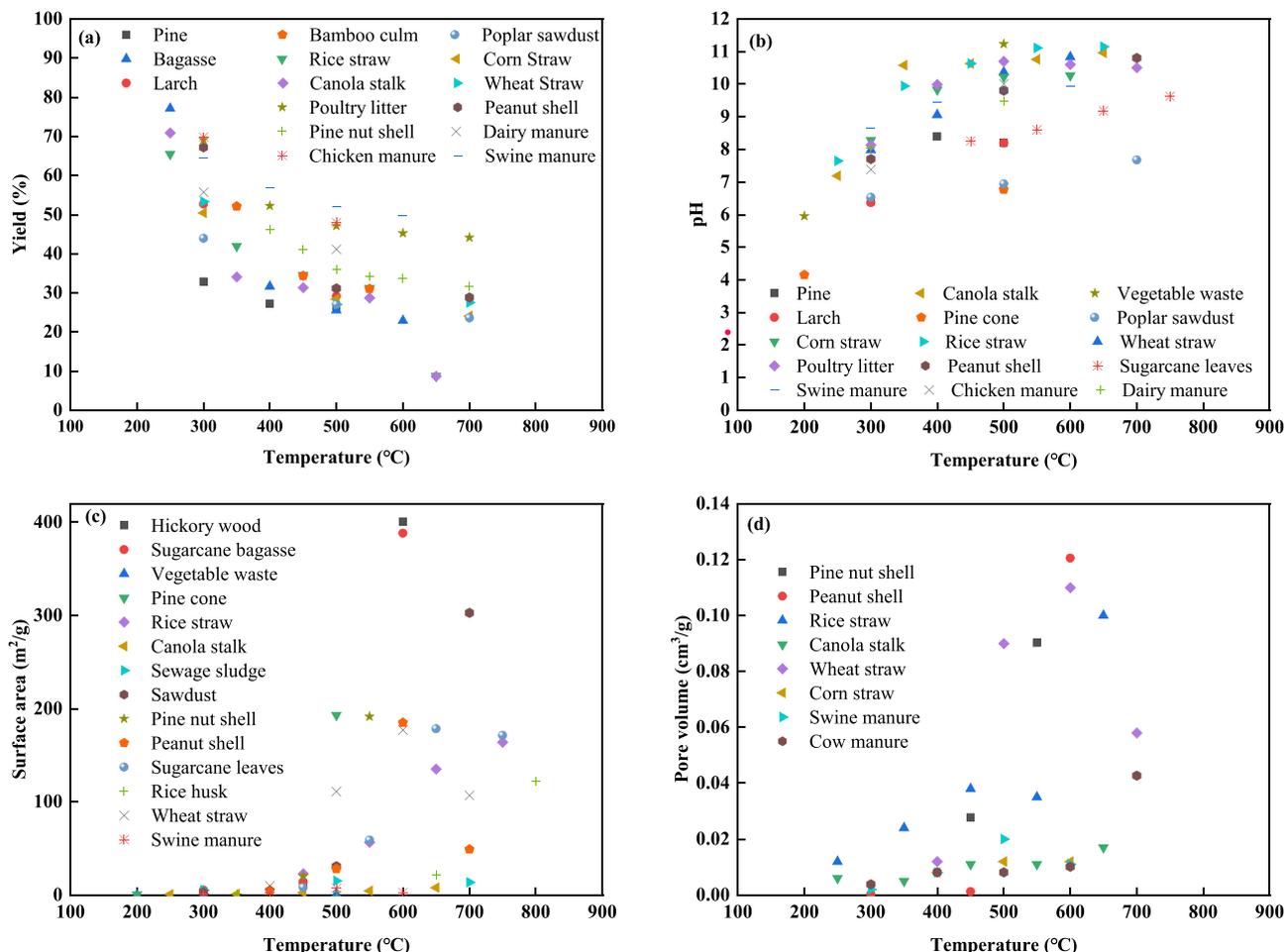


Fig. 4. Variation of biochar produced from different feedstocks under different temperatures: (a) yield, (b) pH, (c) surface area, (d) pore volume [4,17,32–42].

time. This means that biochar is highly carbonized and highly aromatic [44]. During the hydrothermal carbonization process, a longer reaction time is favorable for generating high-heating-value hydrochar. Gao et al. reported that the heating value of water hyacinth hydrochar gradually increased from 13.78 to 20.63 MJ kg<sup>-1</sup> as time increased [45]. Appropriate extension of residence time can increase the char's specific surface area. Wang et al. [46] prepared five biochar with the residence time of 30, 60, 90, 120, and 150 min, respectively, producing biochar with specific surface areas of 9, 26, 31, 24, 21 m<sup>2</sup> g<sup>-1</sup>, respectively. This was because the longer the residence time, the organic components in the biomass were fully decomposed and volatilized, which contributes to the development of biochar pore structure. However, if the pyrolysis time is too long, the pore structure of biochar would collapse and the specific surface area would decrease.

#### 2.2.4. Heating rate

Heating rate has an important influence on the mechanism of the pyrolysis carbonization process and the properties of the resulting biochar. The increase in the heating rate could result in the improvement of the weight loss of biomass. For example, Li et al. studied the pyrolysis process of lignin under different heating rates. It was reported that with the increase of heating rate from 5 to 20 °C min<sup>-1</sup>, the yield of biochar decreased from 55.5% to 50.1%, and the yield of gas increased from 18.2% to 22.2% [47]. The increase in heating rate also promotes the cracking rate of organic components and volatile in biochar to accelerate the gas generation. In addition, some people have a one-sided view that an increase in heating rate must lead to an enhancement in the specific surface area of biochar. However, another different result was given by Hu et al. [48]. They found that the specific surface area of biochar decreased from 362 to 146 m<sup>2</sup> g<sup>-1</sup> by the rise of the heating rate from 2 to 15 K min<sup>-1</sup>, then it increased to 327 m<sup>2</sup> g<sup>-1</sup> at 25 K min<sup>-1</sup>. They attribute this to the intense decomposition and diffusion of volatiles at a steep heating rate.

### 3. Modification methods for carbon materials

The pristine carbon materials from biomass have certain limits in chemical composition and pore structure, thus limiting the CO<sub>2</sub> adsorption capacity of carbon materials. Therefore, the modification of carbon materials is of great importance for CO<sub>2</sub> capture.

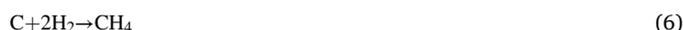
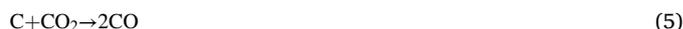
There are two operating ways for carbon material modification, as shown in Fig. 5. Pre-treatment is a modification process in which the raw biomass is directly mixed with acids or metal salts and then carbonized. Post-treatment generally refers to the process of obtaining modified carbon materials by activating carbonized biomass with modifiers such

as acids, bases, metal salts, or gases. But it needs to be noted that before modification, the raw biomass should be treated with diluted acid to remove the ash in biomass, avoiding blocking pores during carbonization process. In this chapter, we discuss the modification methods into three kinds of physical treatment (steam/air/CO<sub>2</sub> activation), chemical treatment (acid/alkaline activation, metal or metal oxide impregnation, heteroatom doping), and additional methods (ball milling, microwave, ultrasound, plasma).

#### 3.1. Physical treatment

##### 3.1.1. Steam activation

Steam activation can improve the textual structure of carbon materials. During the steam activation process, the steam could diffuse into the carbon materials and facilitate the formation of more pores [49]. The mechanisms of carbon materials steam activation are as follows [50]:



It was observed that specific surface area and pore volume could be significantly improved through steam activation of biochar. Rajapaksha et al. [51] utilized steam activation to modify tea waste biochar. They found that the specific surface area of biochar increased from 342 to 576 m<sup>2</sup> g<sup>-1</sup> and the pore volume was enhanced from 0.022 to 0.109 cm<sup>3</sup> g<sup>-1</sup>. The improvement in pore structure by steam activation can be divided into three ways: (1) the removal of pore-clogging tar, (2) the formation of new micropores, and (3) the expansion of original pores. Different steam activation parameters such as temperature and reaction time have different activation effects. With the increase of activation time, the inner surface and the total pore volume of the activated carbon become larger, the pore width distribution becomes wider, and the proportion of mesopores increases [52]. Young-Jung Heo et al. [53] used cellulose fibers as feedstock and activated them at different temperatures by steam activation. They found that steam activation could promote the formation of new pores and affect the expansion of pore sizes. The activated material had an excellent CO<sub>2</sub> adsorption capacity of

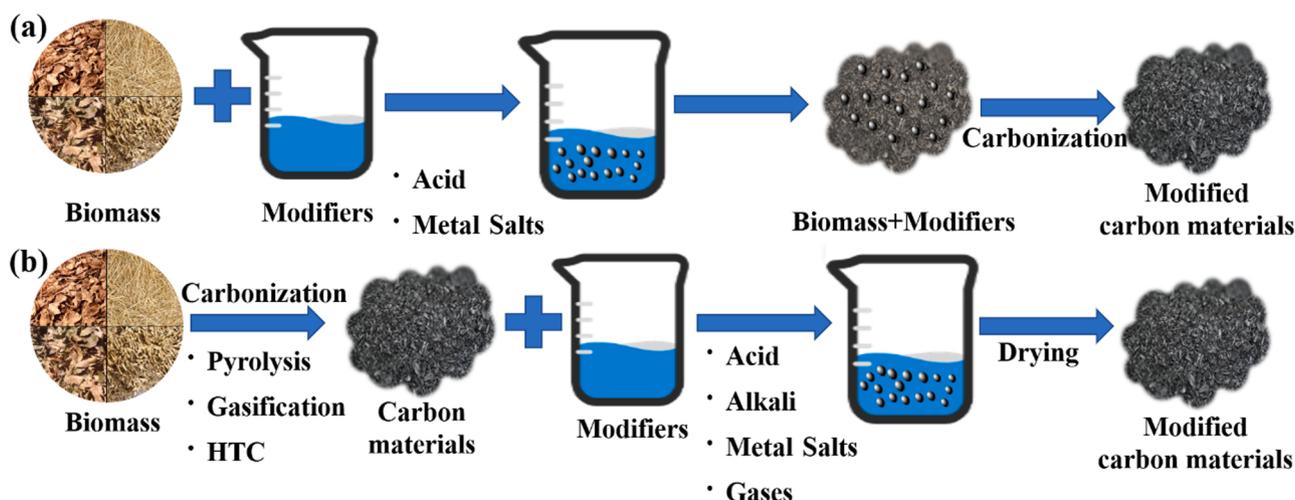


Fig. 5. Operating methods for carbon material modification: (a) pre-treatment, (b) post-treatment.

3.78 mmol g<sup>-1</sup>, which was ascribed to the microporous structure. Igalavithana et al. [54] produced biochar from pine sawdust and performed steam activation. The authors observed that not only the textual properties of biochar were improved, but also the surface functional groups were enriched. The specific surface area of biochar was enhanced from 316 to 582 m<sup>2</sup> g<sup>-1</sup> and the pore volume increased from 0.17 to 0.25 cm<sup>3</sup> g<sup>-1</sup>. The XPS analysis showed that the oxygen content of biochar increased from 9.16% to 11.05% by steam activation, due to an increase in C-O, C-OH, and O-C=O.

### 3.1.2. Air activation

Oxygen is also an effective oxidizing reagent. Due to the high cost of pure oxygen, the air is generally used for activation instead of oxygen. The mechanism for air activation can be described by the following reactions [55]:



Air oxidation can introduce new functional groups into biochar. However, the oxidation process may result in a slight reduction of surface area, which could be attributed to the new functional groups blocking the pores. Suliman et al. [56] found that after air oxidation, the micropores of biochar were enriched, and O-groups were also introduced, while the specific surface area decreased. Meanwhile, carbon materials produced at low temperatures are more easily oxidized to introduce more -COOH and C=O. Compared with steam activation, air oxidation is more likely to introduce O-containing groups. Bardestani et al. [57] reported that after steam activation, the surface area of biochar increased from 50 to 1025 m<sup>2</sup> g<sup>-1</sup>, while the concentration of functional groups decreased. However, after air oxidation, the concentration of functional groups in biochar increased from 44 to 104.6 μmol m<sup>-2</sup>. Xiao et al. [58] performed air activation on biochar at 400 °C and 500 °C, respectively. The FTIR patterns indicated that carbon granules after air activation showed considerable enhancement in the surface groups of -OH, C=O and C-O. This can be attributed to the reactions between O<sub>2</sub> and lignocellulosic components or aromatic substances. They also focused on the effect of activation time on the activation effect. Results showed that the increase of the micropores occurred in the first 15 min and then the mesopores of biochar began to increase. The extension of activation time may lead to the destruction of the micropores; hence selecting an appropriate activation time is very important. For carbon materials used for CO<sub>2</sub> capture, an optimal oxygen activation treatment time should be selected.

### 3.1.3. CO<sub>2</sub> activation

CO<sub>2</sub> activation can occur either during or after biomass pyrolysis. Studies showed that pyrolysis using CO<sub>2</sub> atmosphere was more beneficial to retaining C and N in biomass. The mechanism of CO<sub>2</sub> activation involves the following reactions [50]:



CO<sub>2</sub> activation can significantly improve the porosity of carbon materials and enhance the specific surface area. Lee et al. [59] showed that the porous carbon fiber after CO<sub>2</sub> activation was mainly composed of micropores, and the specific surface area was up to 1012 m<sup>2</sup> g<sup>-1</sup>. However, there are also other carbon materials that obtain a large amount of mesoporous structure after CO<sub>2</sub> capture. This may be related to the type of pristine materials or activation conditions. Kozyatnyk et al. [60] reported that when the three kinds of biochar, wheat straw, softwood, and peach stones, were activated by CO<sub>2</sub>, the mesoporous structure of wheat straw and softwood biochar was more developed,

while the peach stones biochar had some improvement in microporosity. CO<sub>2</sub> activation involves the Boudouard reaction, which refers to an endothermic reaction. Hence increasing temperature is beneficial to carbon dioxide activation. Sun et al. [61] prepared CO<sub>2</sub>-activated biochar. Results showed that the improvement effect in pores increases with the increase of activation temperature. The authors performed CO<sub>2</sub> activation at 800 °C, 850 °C, 900 °C, and 950 °C. The specific surface area of biochar increased from 392 m<sup>2</sup> g<sup>-1</sup> to 506, 854, 1059, and 2186 m<sup>2</sup> g<sup>-1</sup>, respectively. Furthermore, the pores in all the samples are dominated by micropores less than 2 nm.

## 3.2. Chemical treatment

### 3.2.1. Acid activation

Acid activation can occur either before or after carbonization. It can not only facilitate the decomposition of the organic compounds during pyrolysis but also inhibit the generation of tar and other by-products, leading to an increase in char yield. Phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) is the most common acid activator used in the preparation of activated carbon for carbon capture. According to Olivares-Marín et al. [62], phosphoric acid polyphosphoric acids to polyphosphate in the activation process. The activation mechanisms of phosphoric acid are as follows:



Phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) activation not only promotes the formation of pores but also provides new functional groups for carbon materials. Chen et al. [63] used 85 wt% H<sub>3</sub>PO<sub>4</sub> immerse Lantana camara biomass to obtain activated biochar. Results showed that compared with un-activated biochar, the specific surface area of biochar increased from 117 to 1177 m<sup>2</sup> g<sup>-1</sup>, and the pore volume was increased by about 20 times. In addition, the O content of biochar was increased, and XPS confirmed the presence of C-PO<sub>3</sub>, C-P<sub>2</sub>O<sub>6</sub>, and C-P<sub>2</sub>O<sub>5</sub> groups on the surface of the modified biochar. Cao et al. also found that the biochar produced by phosphoric acid activation at 400–600 °C with amounts of surface groups C-O-PO<sub>3</sub> and C-PO<sub>3</sub> [64]. But carbon dioxide is an acidic molecule, it has been proved that the acidic P<sub>x</sub>O<sub>y</sub> group generated from H<sub>3</sub>PO<sub>4</sub> activation is detrimental to CO<sub>2</sub> chemisorption in carbon materials. Sánchez et al. [65] found that the surface chemistry and porous structures can be easily adjusted by changing the concentration and treatment temperature of H<sub>3</sub>PO<sub>4</sub>. Porous structure plays a key role in CO<sub>2</sub> capture at 0 °C, while surface chemistry such as functional groups is more important in high-temperature adsorption. Therefore, more care should be taken in the preparation and application of H<sub>3</sub>PO<sub>4</sub>-activated carbon. Additionally, some researchers also use H<sub>2</sub>SO<sub>4</sub> or HNO<sub>3</sub> to prepare activated carbon. Li et al. [66] synthesized three kinds of porous carbon for gas adsorption by H<sub>3</sub>PO<sub>4</sub>, H<sub>2</sub>SO<sub>4</sub>, and HNO<sub>3</sub> treatment, respectively. For CO<sub>2</sub> adsorption, the H<sub>2</sub>SO<sub>4</sub> treated porous carbon had a good CO<sub>2</sub> adsorption capacity (3.60 mmol g<sup>-1</sup>), which is 39.5% higher than the untreated sample.

### 3.2.2. Alkaline activation

Alkaline activation operates similarly to acid activation. Alkaline activation benefits the formation of pores and the introduction of alkaline groups to facilitate the adsorption of carbon dioxide. Potassium hydroxide (KOH), sodium hydroxide (NaOH), and amino (-NH<sub>2</sub>) are the most commonly used alkaline activators.

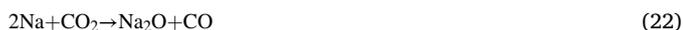
Potassium hydroxide (KOH), as a strong basic metal hydroxide, is often used as an activator of carbon materials. During the KOH activation process, KOH is first mixed thoroughly with the carbon precursor. The redox reaction of KOH and carbon precursors approximately starts at 400 °C, forming K<sub>2</sub>CO<sub>3</sub>. Then, KOH is completely converted into K<sub>2</sub>CO<sub>3</sub> at around 600 °C. When the temperature is higher than 700 °C, K<sub>2</sub>CO<sub>3</sub> begins to decompose into CO<sub>2</sub> and K<sub>2</sub>O. K<sub>2</sub>CO<sub>3</sub> and K<sub>2</sub>O can also be reduced to K at high temperatures. K can become vapor, which is easy

to intercalate into the carbon lattices. This is beneficial for the formation of micropores. The specific reaction equations are shown below [67]:



KOH activation enriches the surface groups on carbon materials, such as -OH, C=O, C-C, and =C-H. However, with the synthesis temperature increase, the O-containing groups on the surface of the carbon materials break down when heated to produce CO and CO<sub>2</sub> [68]. Due to the acidity of carbon dioxide, it is beneficial to introduce the Lewis base on the surface of carbon material for CO<sub>2</sub> capture. A study showed that the biochar prepared from a mixture of food and wood waste after activation via KOH and KOH-CO<sub>2</sub>, respectively, the biochar through KOH activation showed a better microporous structure and enhanced biochar CO<sub>2</sub> adsorption capacity. In contrast, KOH-CO<sub>2</sub> activation could not further improve them [69]. Qiu et al. [67] investigated the contribution of KOH activation to the preparation of porous carbon from biomass as supercapacitors. They found that KOH affected the microstructure of the produced char. The specific surface area of optimized carbon materials was up to 2790 m<sup>2</sup> g<sup>-1</sup>. Graded pore structure significantly improves the rate performance of carbon materials and the maximum specific capacitance is 327 F g<sup>-1</sup>.

Compared with KOH, sodium hydroxide (NaOH) activation has better oxidizing properties and is more friendly to the environment. According to the research, the redox reaction of NaOH with carbon contributed positively to forming micropores and mesopores of carbon materials. During the activation process, NaOH also reacts with carbon materials in the following ways [70]:



The reaction process releases active intermediates CO, CO<sub>2</sub> and H<sub>2</sub>. Meanwhile, Na and Na<sub>2</sub>CO<sub>3</sub> also enter the carbon layer, resulting in the carbon atomic structure layer being larger. Therefore, the specific surface area and pore diameter of carbon materials also increase. However, if the concentration of NaOH is continuously increased, excessive NaOH will promote the vigorous gasification reaction of carbon and destroy the carbon structure, thus reducing the effective area. Moreover, a high concentration of NaOH can induce a series of reactions (e.g. C-NaOH, C-Na<sub>2</sub>CO<sub>3</sub>, C-Na<sub>2</sub>O, C-Na, C-CO<sub>2</sub> and C-CO), promoting the rupture of C-C and C-O-C, thus reducing the yield of carbon materials [71]. Wei et al. synthesized carbon materials from macroalgae by NaOH activation, which had a high surface area of 1238 m<sup>2</sup> g<sup>-1</sup>. Another research showed that NaOH activation helped to enhance the aromatic properties of biochar, thus facilitating the π-π interaction when it was adsorbing pollutants [72].

Introducing amino groups into carbon materials also contributes greatly to CO<sub>2</sub> capture. Since the charcoal graphite surface is not highly reactive to NH<sub>3</sub>, NH<sub>3</sub> treatment is generally carried out under high-temperature conditions. This method can introduce not only basic functional groups but also has a certain contribution to the removal of acidic groups. During the NH<sub>3</sub> treatment process, ammonia reacts with carboxylic acids to form amides, lactam, and imides. Some functional groups can also form heterocyclic compounds pyrrole and pyridine by dehydration or decarboxylation [73]. In addition to introducing various new groups, ammonia activation can also improve the pore size and specific surface area of carbon materials to a certain extent. Wu et al.

[74] prepared biochar from coconut fiber in a muffle furnace (300, 500, 700 °C) and modified the pristine biochar by NH<sub>3</sub>·H<sub>2</sub>O at 50 °C for 9 h. Results showed that the specific surface area of biochar was increased from 4, 7, and 541 m<sup>2</sup> g<sup>-1</sup> to 9, 24, and 553 m<sup>2</sup> g<sup>-1</sup>, respectively. Geng et al. [75] synthesized microporous N-doped carbon materials in one step, which used NH<sub>3</sub> as the activator. It was indicated that the synergistic effect of pores and N-groups (phenylamine and pyridine-nitrogen) determines the high CO<sub>2</sub> capture and CO<sub>2</sub>/N<sub>2</sub> selectivity of carbon materials.

### 3.2.3. Salts activation

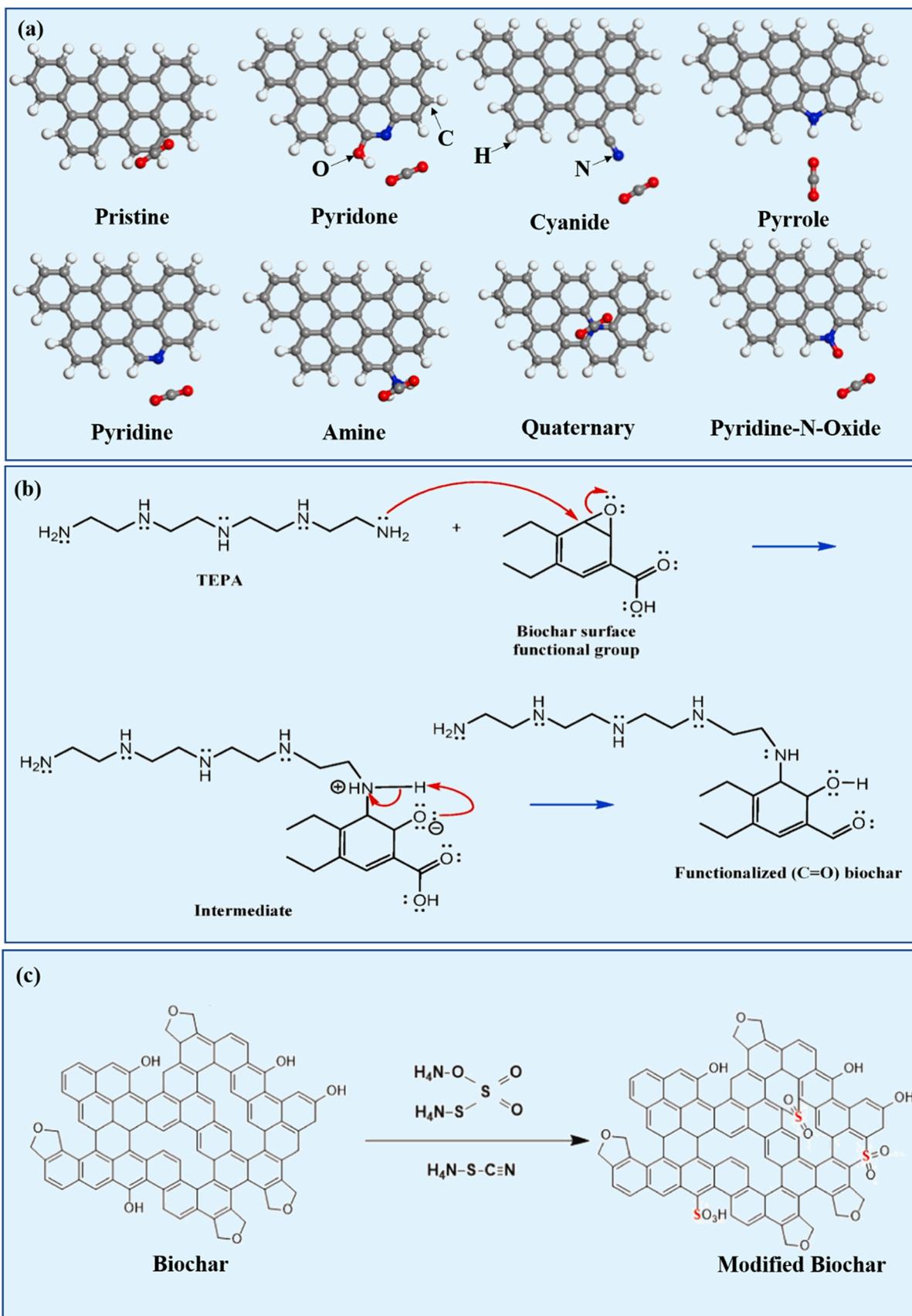
Salts also play an important role in carbon materials activation. Zinc chloride (ZnCl<sub>2</sub>) is the most commonly used salt activation agent, which has a strong dewaterability. Zinc chloride is usually mixed directly with biomass for activation. The whole activation process can be divided into three stages during pyrolysis. Biomass is firstly decomposed into small particles and volatiles with increasing temperature. Then, ZnCl<sub>2</sub> reacts with small particles generating char, tar, and volatiles. Finally, the generated char reacts with ZnCl<sub>2</sub>, thus forming the pores [76,77]. Carbon materials through ZnCl<sub>2</sub> activation showed more mesopores and macropores since the molten ZnCl<sub>2</sub> can penetrate evenly into the carbon lattice, resulting in the expansion of pores [2]. Balou et al. [78] co-activated the hydrochar through ZnCl<sub>2</sub> and CO<sub>2</sub>. They found that the physical-chemical activation significantly increased the specific surface area and total pore volume and even formed ultra-fine pores. Different from ZnCl<sub>2</sub>, ferric chloride (FeCl<sub>3</sub>) treatment is more favorable for forming micropores. Dan et al. [79] prepared activated carbon by FeCl<sub>3</sub> activation, exhibiting an admirable micropore volume of 0.468 cm<sup>3</sup> g<sup>-1</sup>.

Herein, we cannot ignore the problem of Zn pollution in the last century. The EU has proposed in 2017 to ban the use of high concentrations of ZnO. China also began limiting the use of ZnO in the year of 2018. Hong et al. reported that [80] during the activation process, some of the ZnCl<sub>2</sub> was converted to ZnO and adsorbed on the activated carbon. Therefore, waste ZnCl<sub>2</sub>-activated carbon materials should be disposed of properly, otherwise it can contaminate soil or groundwater.

### 3.2.4. Heteroatom doping

Although micropores in carbon materials make a major contribution to carbon dioxide capture. Nevertheless, Ma et al. [81] indicated that during the CO<sub>2</sub> adsorption process, the contribution of surface functional groups was approximately 38%; thus, the chemical adsorption of CO<sub>2</sub> cannot be neglected. Surface alkalinity is an important factor affecting CO<sub>2</sub> adsorption. It has been pointed out that the introduction of heteroatoms improved the CO<sub>2</sub> uptake in carbon materials, which was due to the enhancement of surface alkalinity [2]. N doping is the most widely studied doping method, which can not only regulate pore structure but also enrich the N-containing groups in carbon materials, thus promoting CO<sub>2</sub> adsorption [82]. Generally, the forms of N-containing groups include pyridone, cyanide, pyrrole, pyridine, amine, quaternary, and pyridine-N-oxide, the structure of which is shown in Fig. 6(a). In addition, polyetherimide (PEI), triethylenephosphoramidate (TEPA), diethylenetriamine (DETA) et al. have been proven to be effective N dopants for CO<sub>2</sub> capture [83]. Fig. 6(b) exhibits the mechanism of biochar amination. The addition of TEPA to biochar induces the lone pair of N to attack the epoxy group of biochar, which eventually forms amino groups on the biochar structure [1]. However, additional N-doping treatment incurs many costs. It is essential to find suitable nitrogen-rich biomass synthesizing carbon materials. As is shown in Table 1, biomass with high N content would lead to nitrogenous biochar. The nitrogen content of microalgae was much higher than that of others. However, not all kinds of nitrogen in biomass can be converted to N in carbon materials, as some nitrogen may be converted into NH<sub>3</sub> or other gases [84].

Except for single N doping, other atoms doping, such as S or co-doping, is also efficient. Seema et al. [89] synthesized an S-doped porous carbon, the CO<sub>2</sub> adsorption capacity of which was up to



**Fig. 6.** Heteroatom doping mechanisms: (a) structures of various N-containing groups; (b) amination of biochar using TEPA; (c) N, S, O co-doped porous carbon [90–92].

**Table 1**  
A summary of nitrogen-rich biomass and its produced biochar.

Biomass	Ref.	Biomass	Carbon materials	
Species		N content (wt%)	N content (wt%)	N forms
Microalgae	[85]	13.94	6.57–12.93	C-N-C, pyrrolic-N, pyridinic-N, quaternary-N
Chitosan	[86]	9.4	8.9–9.1	–
Crab shells	[87]	–	5.1–8.5	pyrrolic-N, pyridinic-N, pyridinic-N-oxide and quaternary nitrogen
Glucosamine	[86]	6.5	6.6–6.7	Pyridinic and aromatic graphene structure
Sludge	[88]	5.9	2.1–3.1	Pyridinic-N
Walnut shell	[10]	4.50	0.34–1.54	–

4.5 mmol g<sup>-1</sup>, much higher than similar materials by N doping. This may be because the binding energy of CO<sub>2</sub>-S is higher than that of CO<sub>2</sub>-N. The larger the binding energy, the more stable the binding between adsorbent and adsorbate, and thus the higher the adsorption capacity. Guo et al. [90] used poplar sawdust as raw material and sulfur-containing wastewater as a modifier, N, S, and O were co-loaded onto the activated carbon, obtaining porous carbon with CO<sub>2</sub> adsorption capacity of 5.61 mmol g<sup>-1</sup> at 273 K. The introduced S existed in the forms of O=S=O and SO<sub>3</sub>H (Fig. 6c).

### 3.2.5. Metal impregnation

Metal impregnation refers to the use of metal salts or metal oxides to impregnate biomass or carbon materials. After the impregnation, the metal ions may be attached to the surface or inside of carbon materials, which could provide more adsorption sites.

From a previous study, the introduction of several metals into carbon materials, such as Na, Ca, Mg, Al, Ni, and Fe, showed good potential for CO<sub>2</sub> capture [1]. Especially, Mg is one of the most proper metals for the synthesis of metal-carbon composites for CO<sub>2</sub> capture. The basic MgO is favorable for CO<sub>2</sub> capture at low temperatures. The reaction is as the following equation:



Pooya Lahijani et al. [10] compared the CO<sub>2</sub> capture capacity of different metal-impregnated biochar. They found the biochar modified by Mg has the highest CO<sub>2</sub> capture capacity of 82.0 mg g<sup>-1</sup>, followed by Al, Fe, Ni, and Ca. In addition, Mg-biochar composites also had superior selectivity of CO<sub>2</sub> over N<sub>2</sub>, O<sub>2</sub>, and CH<sub>4</sub>. However, it is worth noting that the CO<sub>2</sub> capture capacities of metal-loaded carbon materials do not endlessly increase with the metal loading amount. Although the loading of metal in carbon materials can provide basic sites for CO<sub>2</sub> capture, excess metals block up the pores and reduce specific surface area and pore volume, thus increasing CO<sub>2</sub> diffusion resistance and reducing CO<sub>2</sub> capture. Therefore, it is important to determine proper metal loading amounts to balance the change between pore structure and surface basicity for high CO<sub>2</sub> capture capacity.

Different impregnation methods may reduce the clogging of metal to carbon materials. Somy et al. [93] found that the activated carbon metal loaded by the slurry method had better performance on surface area and CO<sub>2</sub> adsorption rather than the wet impregnation method. This is because the solution method resulted in the pores of activated carbon clogged, while the slurry method could avoid clogging. Furthermore, the authors proved that the adsorption performance of the activated carbon co-loaded with two metals was much higher than single metal.

As mentioned before, whether biomass or carbon materials were mixed with metal salts, some thermal treatment is still required after impregnation to convert metal salts into metal oxides. It should be noted that for the same metal ions, different metal salts require different calcination temperatures. Zubbri et al. [94] utilized kinds of magnesium salt (MgNO<sub>3</sub>, MgCl<sub>2</sub>, MgSO<sub>4</sub>, Mg(CH<sub>3</sub>COO)<sub>2</sub>) to modify biochar.

MgNO<sub>3</sub>-biochar showed the best CO<sub>2</sub> capture capacity (76.80 mg g<sup>-1</sup>), while there was no improvement of Mg(CH<sub>3</sub>COO)<sub>2</sub>-biochar. The authors suggested that the calcination temperature (500 °C) was insufficient to decompose Mg(CH<sub>3</sub>COO)<sub>2</sub> into MgO (650 °C), while MgNO<sub>3</sub> could be decomposed into MgO at 500 °C. Therefore, metallic oxides are normally used to impregnate biochar or biomass, avoiding the incomplete decomposition of metal salts.

### 3.3. Additional modification methods

In addition to the major modification methods described above, other methods, such as ball milling, microwave radiation, ultrasound treatment, and plasma modification, are expected to combine with the above methods to improve carbon materials further.

Ball milling modification refers to the kinetic energy generated during the ball movement in the machine applied to the material, resulting in the reduction of the particle size of the material or the destruction of chemical bonds. Ball milling modification can change the pore structure of materials. Zhang et al. [95] compared the crayfish shell biochar before and after ball milling, which showed that the specific surface area and micropore volume increased from 128 to 290 m<sup>2</sup> g<sup>-1</sup> and from 0.028 to 0.061 cm<sup>3</sup> g<sup>-1</sup>, respectively. Xu et al. [96] compared the difference between pristine biochar, ball-milled biochar, and ball-milled-N-doped biochar. The results showed that two modified biochar exhibited more excellent pore structures, which also had higher CO<sub>2</sub> adsorption capacities.

Microwave radiation, as an emerging heating method, has the advantages of fast heating speed, convenient control, and uniform heating. Carbon materials have a high absorption capacity for microwaves, a property that allows them to be transformed by microwave heating to produce new carbon with specific properties [22]. Rapid heating may promote the formation of new pores and change the pore structure and surface functional groups. A study reported that the number of functional groups on the molecular bonds of graphene materials increased after microwave treatment, providing more adsorption sites for the adsorption process [97].

Ultrasonic treatment is an emerging method for carbon material modification. Due to the effects of cavitation and microjet, the lumps generated during biomass pyrolysis can be removed, thus reducing pore clogging and introducing more microporous structures [1]. Ultrasonic treatment is usually combined with chemical modification methods. Chatterjee et al. [92] synthesized biochar by ultrasonic treatment and amination at room temperature. Ultrasonic treatment of biochar prior to amination may cause the graphite layer of biochar to peel off and create new pores. The exfoliated biochar is rich in -COOH, -OH, and -CH(O)CH-, which facilitates the grafting of amines onto it. The results showed that the CO<sub>2</sub> adsorption capacity of the biochar treated with ultrasound and amine was 9 times higher than that of the pristine biochar.

Plasma is a neutral substance composed of cations, particles, and free electrons. Plasma used for surface modification is generally generated by corona, glow, and microwave discharge methods. During the modification process, reactions occurring between plasma and the material surface may facilitate the formation of new groups. Shen et al. [98] modified activated carbon by atmosphere pressure plasma. The authors found that the surface area and pore volume of activated carbon were enhanced. Meanwhile, surface oxygen-containing functional groups were also enriched.

## 4. Carbon materials for CO<sub>2</sub> capture

Carbon materials have various applications, such as pollutants adsorption, catalysts, energy storage et al. Recently, carbon materials for CO<sub>2</sub> capture have always been a research hotspot. Due to the rapid development of industry, a mass of carbon dioxide has been released into the atmosphere continually, leading to an increasing problem of global warming. Biomass is a kind of solid waste with high carbon

content, which can be used to prepare biochar or hydrochar. However, pristine carbon materials have some limits in textual structure and chemical composition. Before CO<sub>2</sub> capture, some modifications are needed for carbon materials to improve their properties, thus increasing CO<sub>2</sub> capture capacities. This chapter mainly reviews previous research on biomass-based carbon materials for CO<sub>2</sub> capture and its adsorption mechanism.

#### 4.1. Carbon materials for CO<sub>2</sub> capture

Table 2 summarizes carbon materials prepared by different modification methods and compared their CO<sub>2</sub> capture capacities. The pore structure and surface chemistry of pristine carbon materials are not well developed, which is not conducive to CO<sub>2</sub> capture. As shown in Table 2, the pristine biochar produced from coffee grounds only had a CO<sub>2</sub> capture capacity of 0.14 mmol g<sup>-1</sup>, while it was significantly improved to 4.76 mmol g<sup>-1</sup> after KOH activation (500 °C, 1 h). Therefore, for high CO<sub>2</sub> capture capacities, modifications such as acid or alkaline activation, heteroatom doping, and metal impregnation are necessary. It is very important to choose the right activation conditions. The selection of modifier, treatment temperature, and time all can affect the modification effect. In Table 2, Ma et al. [99] synthesized different porous carbon from tobacco stems by KOH activation at different temperatures. The surface area and micropores volume of porous carbons are increasing with the increase of activation temperatures, while the CO<sub>2</sub> capture capacity rises first and then falls. They found that this is because the higher the activation temperature, the lower the oxygen content of porous carbon. Oxygen groups in the carbon framework improve CO<sub>2</sub> adsorption. According to Peredo-Mancilla et al. [100], the activated carbon was derived from corn stover and activated by H<sub>3</sub>PO<sub>4</sub>, CO<sub>2</sub>, and H<sub>2</sub>O, respectively. Carbon activated by H<sub>3</sub>PO<sub>4</sub> showed the best CO<sub>2</sub> capture capacity of 10.9 mmol g<sup>-1</sup>. Activated carbon obtained by chemical activation has the highest specific surface area and microporous volume, leading to good CO<sub>2</sub> adsorption performance. Rouzitalab et al. [101] synthesized a kind of urea-doped nanoporous carbon from walnut shell. Results showed that the CO<sub>2</sub> capture capacity of carbon material was enhanced with the increase in urea doping amount. Nowrouzi [102] modified activated carbon by NiO and CuO, respectively. Its CO<sub>2</sub> uptake showed a significant enhancement from 3.02 to 6.48 and 6.78 mmol g<sup>-1</sup>, respectively. The following part in this section will further discuss its application of CO<sub>2</sub> capture according to different types of carbon materials (pristine carbon, activated carbon, heteroatom-doped carbon, metal-loading carbon, and carbon-based nanomaterials).

**Table 2**

A summary of CO<sub>2</sub> capture performance of different carbon materials.

Biomass	Ref.	Modification	Sample name	S <sub>BET</sub> (m <sup>2</sup> g <sup>-1</sup> )	V <sub>micro</sub> (cm <sup>3</sup> g <sup>-1</sup> )	Adsorption Condition	CO <sub>2</sub> capture (mmol g <sup>-1</sup> )
Coffee grounds	[103]	/	HC	34	< 0.01	35 °C, 1 bar	0.14
		Melamine	MHC	402	0.09		0.85
		Melamine,KOH	KMHC	990	0.45		2.67
Tobacco stem	[99]	KOH (500 °C 1 h)	OC500	786	0.321	0 °C, 1 bar	4.76
		KOH (600 °C 1 h)	OC600	1086	0.445		6.32
		KOH (700 °C 1 h)	OC700	1922	0.788		7.98
		KOH (800 °C 1 h)	OC800	2399	0.958		6.60
Olive stones	[100]	H <sub>3</sub> PO <sub>4</sub>	AC-H <sub>3</sub> PO <sub>4</sub>	1178	0.45	30 °C, 1 bar	10.9
		CO <sub>2</sub>	AC-CO <sub>2</sub>	757	0.30		5.89
		H <sub>2</sub> O(g)	AC-H <sub>2</sub> O	754	0.28		7.97
Walnut shells	[101]	Urea/carbon 1:3	KNWS-13	1047	0.27	25 °C, 10 bar	5.72
		Urea/carbon 2:3	KNWS-23	2707	0.17		10.06
		Urea/carbon 3:3	KNWS-33	2461	0.13		11.02
Ersian ironwood	[102]	H <sub>3</sub> PO <sub>4</sub>	HP5	1802	0.83	30 °C, 1 bar	3.02
		NiO	HP5/Ni3-1	1945	1.59		6.48
		CuO	HP5/Cu3-1	1954	1.60		6.78

##### 4.1.1. Pristine carbon materials

Pristine carbon materials are obtained directly through the thermochemical conversion of biomass, including biochar or hydrochar. Biochar is recognized as an environmentally friendly adsorbent because of its cheap and wide raw biomass source. Several researchers have shown that biochar has a good CO<sub>2</sub> adsorption effect, which attributes to its polarity, high specific surface area, and functional groups [12]. Huang et al. [104] synthesized composite biochar from sewage sludge and leucaena wood in different mix ratios, and the pure leucaena wood biochar had a good CO<sub>2</sub> uptake of 53 mg g<sup>-1</sup> better than that of pure sewage sludge biochar. The authors observed that the carbon content of biochar increased by the mix ratio of leucaena wood biochar, and the CO<sub>2</sub> adsorption capacity would enhance by the increase of carbon content. Salem et al. [105] synthesized date palm leaf biochar at 300, 400, 500, and 600 °C, respectively, and the CO<sub>2</sub> adsorption capacity of biochar was enhanced from 0.09 to 0.25 kg CO<sub>2</sub>/kg biochar with the increase in temperature. It was also indicated that the increased preparation temperature could enhance the biochar carbon content, which can promote biochar CO<sub>2</sub> capture.

##### 4.1.2. Activated carbon

Activated carbons generally have a higher CO<sub>2</sub> adsorption capacity than biochar. This is because activation treatment improves its textual properties and surface chemistry. This will facilitate the CO<sub>2</sub> adsorption of carbon materials. The mechanism of CO<sub>2</sub> capture by activated carbon includes physical and chemical processes. Van der Waal's force between molecules is the reason for physical adsorption, and surface area and pore size are the decisive factors in the CO<sub>2</sub> adsorption process. Chemical adsorption is mainly attributed to the alkali functional groups on the surface of activated carbon.

Activation can significantly improve the textual properties of carbon materials, thus leading to a higher CO<sub>2</sub> uptake performance. Fiuza et al. [106] successfully synthesized a kind of granular activated carbon from yellow mombin fruit stones and explored the material's CO<sub>2</sub> adsorption capacity. They indicated that the good CO<sub>2</sub> adsorption capacity could be attributed to the micropores between 0.65 and 1.5 nm and surface basic sites. Furthermore, 10 adsorption-desorption cycles proved the stability of the activated carbon for CO<sub>2</sub> capture. Singh et al. [107] prepared nano-porous activated bio-carbons from alligator weed through carbonization and KOH activation. Moreover, the prepared four types of nanocarbons with different KOH dosages have a high surface area ranging from 1779 to 3106 m<sup>2</sup> g<sup>-1</sup> and CO<sub>2</sub> adsorption ranging from 5.1 to 6.4 mmol g<sup>-1</sup>.

Some studies also refer to activated carbon as porous carbon. Pramanik et al. [108] used one-step methods by KOH and alum co-activation to obtain porous carbon from the cotton stalk. The surface

area of porous carbon was up to  $2695 \text{ m}^2 \text{ g}^{-1}$ , much higher than more carbon materials. Its CO<sub>2</sub> adsorption capacity was 6.9 and  $4.24 \text{ mmol g}^{-1}$  at  $0^\circ\text{C}/0.1 \text{ MPa}$  and  $25^\circ\text{C}/0.1 \text{ MPa}$ , respectively. Some researchers studied the activation of N-doped porous carbon by H<sub>3</sub>PO<sub>4</sub>, KOH, and ZnCl<sub>2</sub>, respectively [109]. The result showed that the CO<sub>2</sub> adsorption capacities of these three treatment carbon materials are ZnCl<sub>2</sub> ( $3.54 \text{ mmol g}^{-1}$ ) > H<sub>3</sub>PO<sub>4</sub> ( $2.59 \text{ mmol g}^{-1}$ ) > KOH ( $2.27 \text{ mmol g}^{-1}$ ) under the condition of 298 K and 1 bar respectively. This is because a kind of narrow micropores formed in the N-doped carbon after ZnCl<sub>2</sub> activation, which is more suitable for CO<sub>2</sub> adsorption, and the rich N-groups is another reason for its better adsorption effect than H<sub>3</sub>PO<sub>4</sub> activation. The reason why the adsorption capacity of H<sub>3</sub>PO<sub>4</sub> activation is higher than the KOH activation can be attributed to the difference in surface area ( $1648 > 567 \text{ m}^2 \text{ g}^{-1}$ ). But activation does not necessarily improve the CO<sub>2</sub> adsorption capacity by improving the specific surface area of the carbon materials. Bae et al. [110] activated commercial activated carbon using KOH, Ba(OH)<sub>2</sub>, and a combination of KOH and Ba(OH)<sub>2</sub>, respectively. After combined activation treatment, the specific surface area of activated carbon decreased the most. But its CO<sub>2</sub> adsorption capacity was 1.42 times higher than that of the original activated carbon. Through XPS analysis, they concluded the alkalinity of KOH and Ba(OH)<sub>2</sub> played an essential role in CO<sub>2</sub> adsorption.

#### 4.1.3. Heteroatom-doped carbon materials

As mentioned before, the micropores of carbon materials made a major contribution to CO<sub>2</sub> capture. But micropores also strengthen the adsorption of other gases (such as CH<sub>4</sub>, N<sub>2</sub>). Therefore, the CO<sub>2</sub> adsorption capacity of carbon materials decreases due to the competitive adsorption between various gases. Heteroatom doping can help

improve this defect. Heteroatoms like S, N, and O can give rise to many functional groups such as hydroxyl, amine amid, pyridine, and sulphoxides. These functional groups can exclusively interact with CO<sub>2</sub> leading to a higher CO<sub>2</sub> selectivity. He et al. [9] used husk as raw materials and prepared the activated carbon through carbonization and KOH activation. During the activation step, they doped the chitosan into the carbonized material, and the doped activated carbon had a better CO<sub>2</sub> adsorption capacity of  $5.83 \text{ mmol g}^{-1}$  at 273 K. XPS analysis showed that pyridine-N and pyrrole-N appeared on the surface of the doped activated carbon. These N-species can effectively increase the surface alkalinity of activated carbon, thus promoting CO<sub>2</sub> uptake. According to Yadavalli et al. [111], they modified the activated carbon with ammonium sulfate leading to the improvement of the performance of CO<sub>2</sub> capture. This is due to the presence of the sulfur functional group, which can interact with CO<sub>2</sub> molecules.

N doping is important in heteroatom doping for the improvement of CO<sub>2</sub> capture. Wang et al. [112] used five different amines (TEPA, MEA, DEA, PEI, and DETA) to modify the waste wood ashes for CO<sub>2</sub> capture. As shown in Fig. 7, the TEPA-sorbent showed the best CO<sub>2</sub> capture performance among all the sorbents, which had a capacity of  $1.76 \text{ mmol g}^{-1}$  and a high regeneration rate of 92.6%. In addition, though DEA-sorbents had the highest actual N loading amount, their adsorption capacity was lower than TEPA. This may be because the CO<sub>2</sub> adsorption depends not only on N content. The contribution of micropores also plays a key role. Moreover, it was pointed out that the increase in TEPA content could affect the dispersion of the amine, which could further influence the available active sites. Therefore, an optimum loading amount (45 wt%) was investigated with a better CO<sub>2</sub> capture performance ( $2.02 \text{ mmol g}^{-1}$ ). In addition to porosity and N content

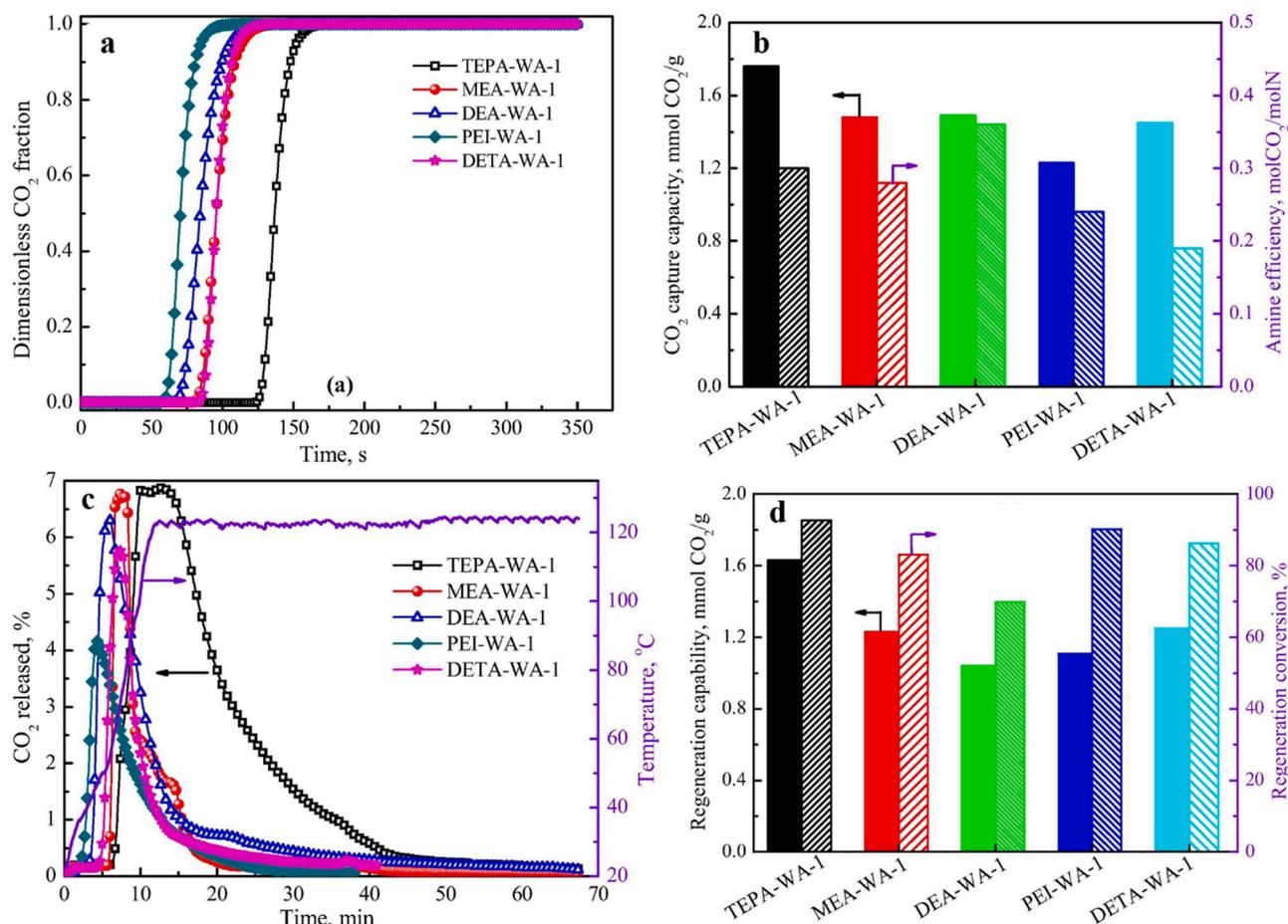


Fig. 7. CO<sub>2</sub> capture and regeneration performances of five wood ash (WA-1) sorbents doped with different amines [112].

affecting CO<sub>2</sub> capture, the type of N-containing functional groups is also pivotal. Wang et al. [113] studied the contribution of different N functional groups in the CO<sub>2</sub> adsorption process. They found that the CO<sub>2</sub> adsorption energies of pyridine-N, pyrrole-N, amine-N, and quaternary-N modified carbons were  $-21.4$ ,  $-18.8$ ,  $-16.4$ , and  $-14.9$  kJ mol<sup>-1</sup>, respectively. For pyridine-N, the high CO<sub>2</sub> adsorption energy can be attributed to the formation of strong dipole-dipole interaction between the N group and CO<sub>2</sub> such as the interaction of H<sup>+</sup> and O<sup>2-</sup> [2].

#### 4.1.4. Metal-loading carbon materials

The introduction of alkaline metals on the surface of carbon materials is very effective in the adsorption of acidic carbon dioxide. In the adsorption process, CO<sub>2</sub> as an electron acceptor can accept electrons from a metal oxide (electron donor) with basic characteristics.

Pu et al. [114] used walnut shell as feedstock and then prepared the carbon composited with Mg and Mg-Al, respectively. As shown in Fig. 8, it was found that after metal loading, the specific surface area decreased due to the clogging of pores. However, the CO<sub>2</sub> capture experiments indicated that the CO<sub>2</sub> capture capacities of different metal-carbon adsorbents had been improved. Especially, the carbon-10%Mg-5%Al composites had an excellent CO<sub>2</sub> uptake of 4.5 mmol g<sup>-1</sup>. The SEM and EDX showed that lots of Mg and Al were distributed on the surface of the carbon material. Meanwhile, the FTIR and XRD patterns showed that after CO<sub>2</sub> adsorption, some metal oxide characteristic peaks of the sorbents were weakened. This indicated that metal-loading enhanced the contribution of sorbent chemisorption in the CO<sub>2</sub> capture process. Therefore, they concluded that the high CO<sub>2</sub> uptake performance of carbon/Mg-Al composites could be attributed to the result of a combination of physical and chemical adsorption. Creamer et al. [115] reported that the CO<sub>2</sub> adsorption capacity of metal-biochar composites was more excellent than the pristine biochar. The biochar modified by AlCl<sub>3</sub> with a ratio of 1:4 showed the best CO<sub>2</sub> uptake performance among all the prepared biochar. They illustrated that the CO<sub>2</sub> capture of

metal-biochar composites mainly resulted from the surface adsorption mechanism, and the metallic oxide should not be ignored.

#### 4.1.5. Carbon-based nanomaterials

Carbon-based nanomaterials are also considered promising CO<sub>2</sub> adsorbents. Typical representatives include carbon nanotubes, carbon nanofibers, and graphene et al. [116]. They are promising CO<sub>2</sub> adsorbents because of their high surface area, developed pores, possible surface functionalization, and ease of regeneration. Due to the complex synthesis process, there is little research into biomass-derived carbon nanomaterials. Ekhlesi et al. [117] utilized populus wood to synthesize graphene for CO<sub>2</sub> capture. Compared with 1 bar adsorption, the graphene showed better adsorption capacities at 10 bars. With the adsorption temperature increasing from 273 to 313 K, the adsorption capacities decreased from 9.4 to 5 mmol g<sup>-1</sup> and from 13.2 to 8.35 mmol g<sup>-1</sup> at 1 and 10 bar, respectively. Zhang et al. [118] synthesized carbon nanotubes-foam composites with developed micropores structure, which exhibited CO<sub>2</sub> adsorption capacities of about 4 mmol g<sup>-1</sup>. The linear relationship analysis showed that pores less than 0.8 nm dominated composites' CO<sub>2</sub> adsorption.

#### 4.2. Mechanisms of CO<sub>2</sub> capture

Fig. 9 shows the possible mechanisms of CO<sub>2</sub> capture by carbon materials. Depending on the surface forces between CO<sub>2</sub> molecules and carbon materials, the mechanisms of CO<sub>2</sub> capture can be divided into physical adsorption and chemical adsorption. The physical adsorption process is mainly determined by van der Waals forces and pore filling, which are heavily influenced by the adsorption temperature. While for chemical adsorption, chemical bonds are formed between the CO<sub>2</sub> and the adsorbents. Lewis acid-Lewis base interactions and hydrogen bonding make major contributions. As shown in Fig. 9(b), micropores, oxygen functional groups, N functional groups, and aromaticity are the main factors affecting CO<sub>2</sub> capture.

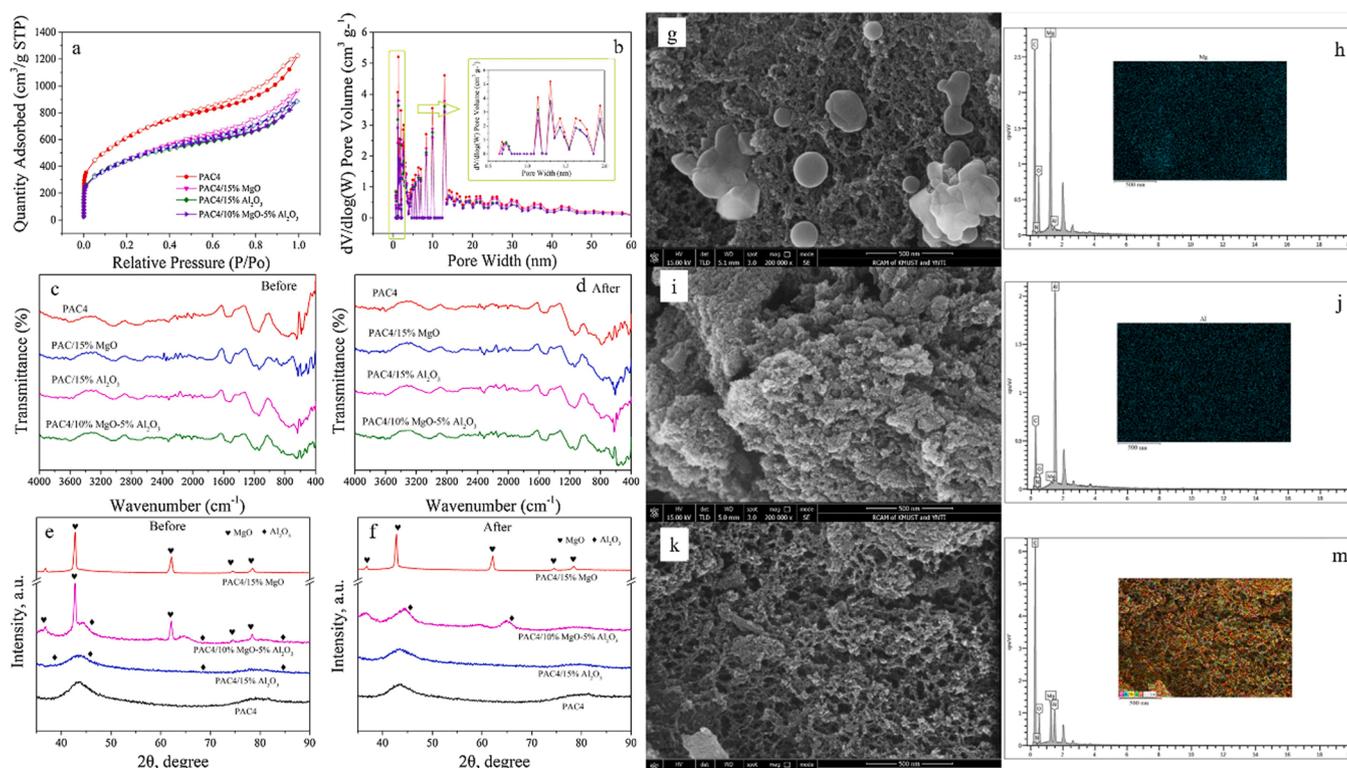


Fig. 8. Characterization of Mg/Al-carbon adsorbents: (a) N<sub>2</sub> adsorption-desorption isotherms; (b) pore size distribution; (c, d) FTIR; (e, f) XRD; and (g, h, i, j, k, m) SEM and EDX [114].

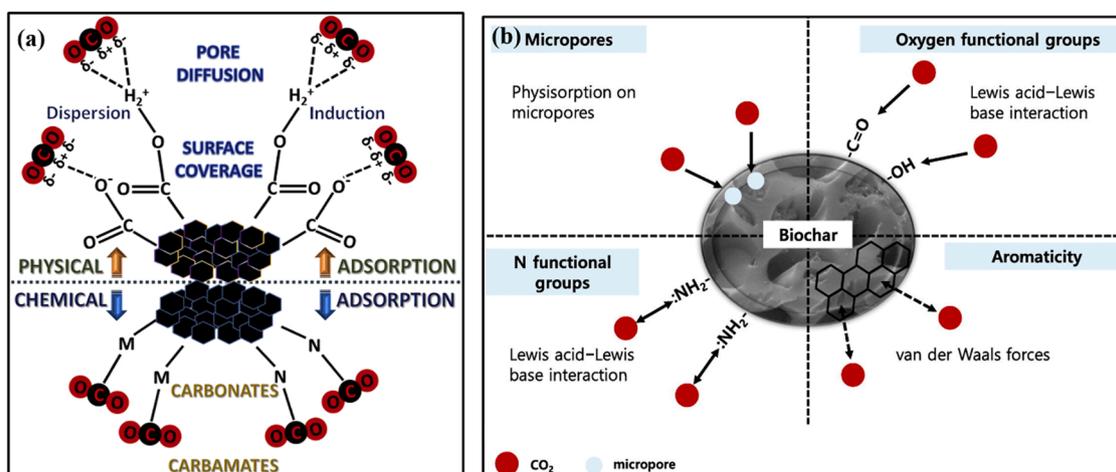


Fig. 9. Mechanisms of CO<sub>2</sub> capture by the carbon materials [16,54].

#### 4.2.1. Physical adsorption

Physisorption is defined as a reversible process that the adsorbates are adsorbed to the adsorbents due to the combined effect of van der Waals forces, electrostatic forces, etc. The physisorption is mainly controlled by the textual properties of carbon materials, including the pore structure and surface area. Generally, large specific surface area and porosity of carbon materials are responsible for the high capacity of CO<sub>2</sub> capture. However, for CO<sub>2</sub> adsorption, the size of the pore is more important than the specific surface area and total pore volume. The pores can be divided into macropores (>50 nm), mesopores (2–50 nm), and micropores (<2 nm) [55]. Under low adsorption pressure, the volume-filling mechanism dominates the whole adsorption process. The micropores had more contribution to CO<sub>2</sub> adsorption. A study conducted by Seul-Yi Lee aimed to determine the most suitable pore size for CO<sub>2</sub> adsorption, and they observed that the enhancement of CO<sub>2</sub> adsorption capacity at 298 K and 1 bar critically depended on the narrow micropores with sizes between 0.5 and 0.7 nm instead of the specific surface area and pore volume [119]. Wahby et al. [120] reported that the pores around 0.4 nm have high selectivity for CO<sub>2</sub>.

Under higher adsorption pressure, adsorption can occur through surface coverage. The specific surface area was more related to the whole CO<sub>2</sub> adsorption process. Li et al. [121] synthesized kinds of porous carbons with different pore sizes ranging from 0.524 to 4.351 nm. The authors found that the porous carbons with a high surface area were more suitable for high-pressure adsorption. When the adsorption pressure increased from 1 to 40 bars, the porous carbon exhibited a great improvement in CO<sub>2</sub> adsorption capacity from 3.65 to 21.97 mmol g<sup>-1</sup>. Adsorption temperature is an important influencing factor for physical adsorption. Generally, the increase in temperature can promote the diffusion of CO<sub>2</sub> and the interaction between CO<sub>2</sub> and carbon materials is weakened and easy to desorption. Ismail et al. [122] carried out a study related to the effect of adsorption temperature and pressure on the adsorption process of CO<sub>2</sub> and investigated CO<sub>2</sub> adsorption under the temperature of 0, 10, 25 °C and the pressure of 1 and 30 bar respectively. The results showed that the CO<sub>2</sub> adsorption volume of 30 bar at different temperatures was always much higher than that of 1 bar. Moreover, under the same pressure conditions, with the increase of temperature from 0 to 10–25 °C, the CO<sub>2</sub> adsorption capacities of biocarbon in 1 and 30 bar decreased from 6.2 to 4.7–3.2 mmol g<sup>-1</sup> and from 26.8 to 25.1–19.9 mmol g<sup>-1</sup> respectively. Another interesting study used CO<sub>2</sub> and KOH for the activation of shoot biochar, respectively. After CO<sub>2</sub> capture experiments, they concluded that KOH-activated carbon was more suitable for low-temperature adsorption at 25 °C, while the CO<sub>2</sub>-activated carbon performed an excellent adsorption capacity at the temperature of 75 °C [123].

#### 4.2.2. Chemical adsorption

In addition to physisorption, chemisorption is also critical. Chemisorption is an irreversible exothermic chemical reaction process in the new chemical bonds (hydrogen, covalent bonding et al.) formed between adsorbents and the adsorbates. Surface functional groups and alkali ions play a key role in carbon materials' chemisorption. Hydrogen bonds are the combination of hydrogen atoms and other atoms with large electronegativity in covalent bonds, but they are not covalent bonds. Hong et al. [124] synthesized a porous carbon with a high CO<sub>2</sub> adsorption capacity of 8.44 mmol g<sup>-1</sup>. Results indicated that the high CO<sub>2</sub> capture performance of adsorbents was not only determined by the porosity of adsorbents but also depended on the hydroxyl groups. CO<sub>2</sub> molecules can form hydrogen bonds with -OH. Further analysis shows that hydrogen bonding reduces the binding energy of adsorbents and CO<sub>2</sub>, and significantly promotes the adsorption of CO<sub>2</sub>. Wu et al. [125] also reported that the porous carbon derived from corn kernels had a good CO<sub>2</sub> capture capacity of 3.63 mmol g<sup>-1</sup> due to the high specific surface area and abundant O-group, especially -OH.

Moreover, due to the effect of Lewis acid-base interaction, covalent coordination bonds can be formed between CO<sub>2</sub> and the adsorbent surface. Many researchers used nitrogen-containing polymers to modify the carbon materials, which can enhance their alkalinity and electron density, thus strengthening Lewis acid-base interaction. Wang et al. [126] prepared a kind of novel N, P co-doped porous carbon, which had a high specific surface area of 1332 m<sup>2</sup> g<sup>-1</sup> and rich mesopores, micropores, and N, P. This resulted in the CO<sub>2</sub> uptake capacity of 5.68 mmol g<sup>-1</sup> at 5 bars with no decrease after 8 cycles. Another study reported that N-doping porous carbon derived from lotus stalks had an excellent CO<sub>2</sub> adsorption capacity of 4.25 mmol g<sup>-1</sup>, which could be attributed to the narrow pore structure and the N groups [127]. Zhou et al. attributed the excellent CO<sub>2</sub> adsorption capacity to the microporosity and the content of N groups. They found that the contribution of microporosity and N groups at 0 °C and 1 bar were almost equivalent, while the N groups played a dominant role in CO<sub>2</sub> adsorption at higher temperatures and lower pressure [128]. This is because microporous adsorption is physical adsorption, which is seriously affected by temperature. However, some researchers have indicated that excessive loading could clog the pores resulting in the reduction of the total adsorption effect. Quan et al. [129] used melamine to modify the tea seed shell activated carbon. They mixed the pristine activated carbon with melamine at different ratios of 1:1, 1:2, 1:3, and 1:4, respectively. After modification, the specific surface area of activated carbon decreased from 1503 to 1065, 1188, 1055, and 706 m<sup>2</sup> g<sup>-1</sup>, respectively. This is because excessive N loading clogs the pores, resulting in a reduced specific surface area. A similar research effort was conducted in another study [130]. S.O. Adio et al. used sugarcane bagasse to prepare

porous carbon and modified it with melamine. They observed that the specific surface area of porous carbon with a carbon-melamine ratio of 1:1 and 1:2 increased from 684 to 1000 and 1111 m<sup>2</sup> g<sup>-1</sup>, respectively, while the carbon-melamine ratio of 1:3 decreased to 727 m<sup>2</sup> g<sup>-1</sup>. The enhancement of surface area can be attributed to the heat-treated at 900 °C for 60 min under N<sub>2</sub> atmosphere after melamine impregnation, but melamine would clog the pores when it was too much. Therefore, there must have a balance between loading amounts and the change of porosity for optimal CO<sub>2</sub> capture performance.

## 5. Summary and prospect

This paper reviews the research progress of synthesis and modification methods of biomass-based carbon materials and discusses CO<sub>2</sub> capture mechanisms and some recent key findings. Firstly, this review includes basic technologies of biomass converted to carbon materials and the factors influencing the properties of carbon materials. Different feedstocks and preparation conditions affect the structure and chemical composition of carbon materials. Therefore, it is important to select the appropriate biomass and preparation parameters. Secondly, some modification methods are summarized. Whether the method is physical or chemical, the essence of modification is to change the pore structures or introduce new functional groups or metals. For CO<sub>2</sub> capture, it is crucial to enrich the micropores and basic sites for carbon materials. Even though progress has been made in this field, some current challenges and future research directions are proposed as below.

(1) Renewable biomass can be converted into carbon material for carbon dioxide capture, which has a low cost. However, pristine carbon materials have some limits in pores or surface chemistry, thus limiting their adsorption capacities. Some modification methods emerged to improve the properties of carbon materials. Nevertheless, some modification processes are too complex and add additional costs, making it difficult to achieve scale applications. Template carbonization is a low-cost method. Although it has more complex preparation steps than pyrolysis and hydrothermal carbonization, it does not need further modification treatment to improve the CO<sub>2</sub> adsorption capacity of carbon materials. Meanwhile, it can effectively control the pore diameter and structure of materials, which is very beneficial for CO<sub>2</sub> capture.

(2) Most of the research on CO<sub>2</sub> adsorption by carbon materials is only in the laboratory stage. However, the actual application environment is generally harsh such as moisture and corrosion, so it should focus on researching and improving the mechanical properties of carbon materials. In addition, the application environment of most research uses a pure CO<sub>2</sub> atmosphere or a mixture of several gases (CH<sub>4</sub>, CO<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O). However, the composition of the gas in the actual application site is often much more complex than the simulated one. Therefore, it becomes essential to implement it in actual industrial installations to assess the feasibility of CO<sub>2</sub> capture by carbon materials. More researchers are also needed to explore the mechanisms of competitive adsorption between different gases. Several simulation programs can be used to predict the adsorption performance of carbon materials in a specific environment.

(3) Most studies focus only on CO<sub>2</sub> capture and barely on conversion. Biomass-based carbon materials have not only adsorption ability but also catalytic effect. In-situ conversion of adsorbed CO<sub>2</sub> will be an interesting research topic in the future. A total solution involving the capture and conversion of adsorbed CO<sub>2</sub> will provide a sustainable process for recovering emitted CO<sub>2</sub>, thereby mitigating the harmful effects of CO<sub>2</sub> emissions. Meanwhile, another problem that needs to be addressed is the regeneration of CO<sub>2</sub>-saturated carbon materials. One suggested solution is to use waste materials for supercapacitors or batteries, etc.

(4) Carbon materials may be contaminated during the adsorption process. Contaminants such as heavy metals and polycyclic aromatic hydrocarbons may affect the age of carbon materials. However, few studies have focused on these, and future research should focus on

studying the effects of various contaminants on carbon materials and how to improve the function of the materials to resist the effects of contaminants.

## CRediT authorship contribution statement

**Cui Quan:** Conceptualization, Validation, Supervision, Writing – review & editing. **Yingying Zhou:** Writing – original draft, review & editing. **Chunfei Wu & Jiawei Wang:** Writing – review & editing. **Ningbo Gao:** Resources, Formal analysis, Supervision.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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