

Review of the Adsorption of Sulfur-Containing Gaseous Pollutants by Biochar: Progress, Challenges, and Perspectives

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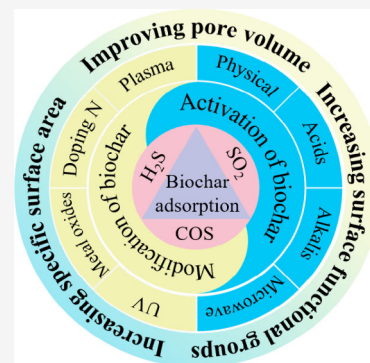
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ABSTRACT: Sulfur-containing gaseous pollutants (e.g., H_2S , SO_2 , and COS) emitted from various industries are important sources of air pollution. Because of the advantages of a simple and clean process, renewability of adsorbent, and cheap raw materials, adsorption removal of sulfur-containing gaseous pollutants by biochar has a good development prospect. However, due to scarce active functional groups and active sites on biochar and an underdeveloped pore structure, the development and application of biochar are still limited in this field. Activation and modification of biochar are one of the most effective ways to raise biochar surface active sites and active functional groups, and improve the pore structure of biochar. This paper reviews the recent research progress of activation and modification methods of biochar for adsorbing sulfur-containing gaseous pollutants (H_2S , SO_2 , and COS). Various activation and modification methods are classified and summarized, including acid/alkali activation, microwave activation, metal oxide modification, nitrogen doping modification, and other emerging modification methods. The main process parameters, advantages, and disadvantages of each modification and activation method are introduced in detail. The activation and modification mechanisms as well as the adsorption mechanisms of pollutants are also discussed and summarized. Among these methods, photochemical modification technology has the advantages of a simple and clean process and low energy consumption, and especially it can simultaneously produce oxygen-containing functional groups on the carbon surface and improve the biochar pore structure, showing superior development prospects. This review will provide needed guidance and inspiration for researchers in related areas to develop new biochar-based desulfurization adsorbents.



1. INTRODUCTION

Sulfur-containing gaseous pollutants, mainly including hydrogen sulfide (H_2S), sulfur dioxide (SO_2), and carbon oxysulfide (COS), are extremely harmful atmospheric pollutants. H_2S mainly exists in coal/biomass gasification, wastewater treatment, and oil/gas production, which poses a serious threat to human health due to its high toxicity.^{1,2} In addition, H_2S will easily corrode equipment (reducing its service life) and poisons the downstream catalysts.³ H_2S is also easily oxidized to form SO_2 , which will become a source of acid rain.⁴ SO_2 is mainly produced by volcanic eruptions and the burning of sulfur-containing fuels.⁵ The SO_2 discharged into the atmosphere will also form acid rain and smog, which will destroy the ecological environment and damage industrial and civil facilities.^{6–8} COS is present in various chemical processes using coal as a feedstock, such as blast furnace gas, yellow phosphorus exhaust gas, and fossil fuel combustion gas, which can corrode equipment and catalysts.^{9,10} The presence of COS can also promote the formation of SO_2 by hydrolysis, which thus leads to the production of acid rain and smog, thereby causing air pollution.^{11–13} Therefore, the development and

research of economically efficient sulfur-containing gaseous pollutant removal technologies is an important topic in this research area.

Most countries in the world have documented laws and regulations to limit sulfur-containing gaseous pollutants emissions. According to the Clean Air Act, the U.S. Environmental Protection Agency (EPA) had a maximum allowable concentration of H_2S of 7 ppm, which is more stringent for special areas such as schools or hospitals. For SO_2 , the Clean Air Act specified no more than 500 μg per cubic meter of the atmosphere, while it also lists COS as a hazardous air pollutant. The Economic Commission for Europe had set a Europe-wide emission limit of 10 mg/m^3 for H_2S and 0.2 $\text{mg}/$

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Table 1. Pore Parameters and Preparation Conditions of Raw Biochars and Their H₂S and SO₂ Adsorption Capacity and Mechanisms

biochar raw material	pollutant	pore parameters				IC ^c (ppm)	AT ^d (°C)	adsorption capacity (mg/g)	main active sites/adsorption mechanism	ref
		pyrolysis temperature (°C)	SSA ^a (m ² /g)	TPV ^b (cm ³ /g)	pH					
Camphor	H ₂ S	400	20.35	/	9.55	50 μL/L	25	109.3	The physisorption is dominant, and the chemical adsorption is promoted by C=O, COO-groups and other functional groups. HS ⁻ + O → S ⁰ + OH ⁻	34
Bamboo										
Rice hull			58.01		10.21			336.7		
Potatoes	H ₂ S	500	115.34	/	10.56	1000	/	382.7		
Reed	H ₂ S	500	63	/	10	1000	/	53	High pH and C–H, C–O, C=O groups promote H ₂ S adsorption.	35
Lignin	H ₂ S	450	78	/	9.8	1000	25	60	Containing-oxygen functional groups (–OH, C–O, C=O) promote H ₂ S oxidation.	36
Sludge	H ₂ S	650	60	/	10	1000	25	70	Physorption is dominant. C–O and C–H groups promote chemisorption.	37
Coconut shell	H ₂ S	/	48	0.281	9.34	1000	25	20.2	H ₂ S reacts with metals or metal oxides to produce SO ₄ ²⁻ .	38
Sargassum	H ₂ S	800	1030	0.52	6.89	3000	25	112	The physisorption is dominant, and alkaline surface promotes the H ₂ S dissociation.	39
Enteromorpha	H ₂ S		26.20	0.034	9.4	200	25	5.8	The physisorption is dominant, and containing-oxygen functional groups, C–O, C=O, COO-, promote H ₂ S oxidation.	40
Coffee granules	H ₂ S	500	21.15	0.466	9.1			0.582		
Almond	H ₂ S		/	/	/	970	25	22	HS ⁻ + O ₂ → S ⁰	41
Perilla	H ₂ S	700	473.4	0.1	10.6	300	50	230	HS ⁻ + H ₂ O + O ₂ → SO ₄ ²⁻	42
Soybean straw								18.3	Larger surface area, higher pH, and more containing-nitrogen functional groups promote H ₂ S removal.	
Korean oak	H ₂ S	700	420.3	0.2	11.3	300		10.5		
Japanese oak	H ₂ S	400	270.8	0.1	10.2	300	50	6	The adsorption process of H ₂ S is governed by both physisorption and the local pH within the pores.	
Cork	H ₂ S	500	475.6	0.2	9.9	300		5.7		
		550	/	/	8.6	750	35	160.44	The physisorption is dominant, and containing-oxygen functional groups (C–O, C=O, COO–) promote the oxidation of H ₂ S; H ₂ S → HS ⁻ + H ⁺ and HS ⁻ + O ₂ → S ⁰ .	43
Hardwood	H ₂ S	800	/	/	9.6			106.32		
		550	/	/	8.2	750	35	143.47		43
		800	/	/	8.7			105.14		
Pig manure	H ₂ S	500	47.4	/	10.5	1000	20	59.6	C=O and –OH groups, high pH, and metal ions promote	44
Sewage sludge			71.6		8.9			43.97	H ₂ S to S ⁰ and SO ₄ ²⁻	
Cow dung	SO ₂	500	4.83	0.014	10.2	5000	25	64.3	Ca ²⁺ , K ⁺ , Fe ³⁺ , and other metal ions, and high pH together cause SO ₂ to produce sulfate.	45
Rice hull			41.5	0.041	9.78			13.3	The excellent pore structure promotes physical adsorption.	
Sludge			10.1	0.022	8.9			34.5	SO ₂ reacts with metal ions to form sulfate.	
Palm oil fiber	SO ₂	450	1.314	0.0267	6.78	2000	26	18.62	The physisorption is dominant, and C–O, C=O, COOH groups promote the oxidation of SO ₂ .	27

^aSSA: Specific surface area. ^bTPV: Total pore volume. ^cIC: Inlet concentration. ^dAT: Adsorption temperature.

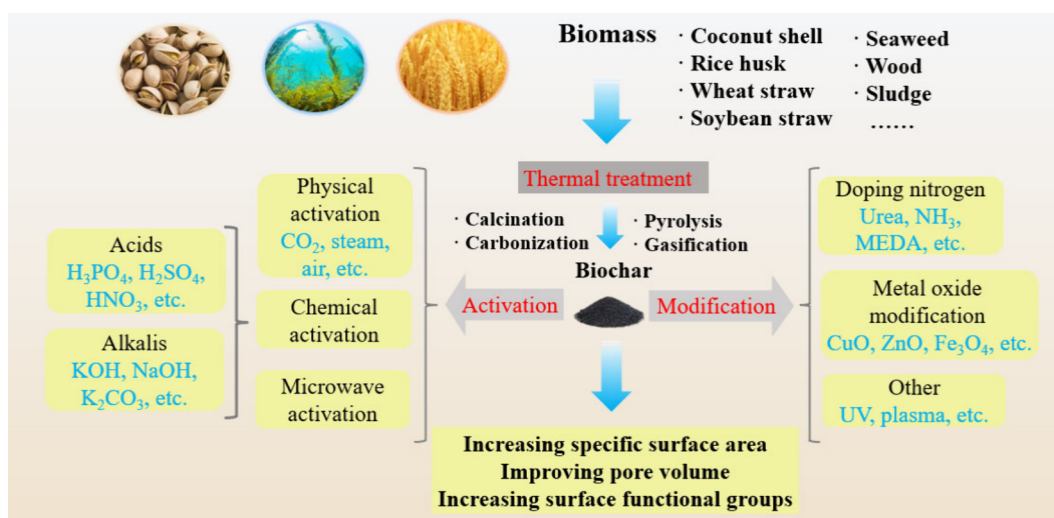


Figure 1. Biochar preparation and activation/modification methods.

m³ for SO₂ in accordance with the Europe-wide Maximum emission limit for H₂S from combustion installations. In 2018, China had revised the “Natural Gas” to make the requirements for the content of H₂S in natural gas. In this regulation, the content of H₂S in Class I natural gas is less than 6 mg/m³, and the content of H₂S in Class II natural gas is less than 20 mg/m³. In 2019, the Ministry of Ecology and Environment of China issued the “Measures to Promote the Implementation of Ultra-Low Emission Standards in Iron and Steel Industry”. This regulation stipulates that the SO₂ emission standard for hot blast furnaces, iron-making and heating furnaces, lime kilns, etc. is 50 mg/Nm³, while the SO₂ emission standard for gas boilers is 35 mg/Nm³.

At present, common desulfurization technology (i.e., sulfur-containing gaseous pollutant removal technologies) can be roughly divided into wet desulfurization and dry desulfurization. Wet desulfurization technology mainly uses amine solution or alkaline solution to absorb sulfur-containing gaseous pollutants through gas–liquid reactions, which usually can meet the requirements of a high desulfurization load.^{9,14} However, there are some problems such as solvent volatilization, high energy consumption, equipment corrosion and scaling, complex product postprocessing and secondary pollution.¹⁵ Therefore, some scholars have begun to pay more attention to dry desulfurization.^{16–18} The representative dry removal methods mainly include plasma removal, ozone oxidation, photocatalytic oxidation, photodecomposition, adsorption removal, etc. Among them, the adsorption method has a good prospect because of its advantages such as a simple process, no wastewater generated, and renewable adsorbent. The development of economical and efficient desulfurization adsorbents is the key point in the current research area.

At present, activated carbon (AC), zeolite, metal oxide, metal organic framework compounds (MOFs), biochar, carbon nanotubes, etc. are the common adsorbents in the sulfur-containing gas adsorption field.^{19–21} Among them, biochar is an adsorbent with good development prospects due to its wide range of raw materials and low cost. It has been widely shown that biochar can effectively adsorb H₂S, SO₂, NO_x, Hg⁰, CO₂, and other gaseous pollutants,^{22–27} which possesses good developing prospect and has received increasing attention. Table 1 summarizes the applications of raw biochar in

adsorbing sulfur-containing pollutants. However, biochar still has shortcomings such as scarce surface active sites/active functional groups and an underdeveloped pore structure, so it is needed to take various activation and modification methods to enhance the pore structure and surface chemical properties of the biochar, thereby overcoming these challenges.²⁸ In the past two decades, researchers have proposed and developed many different activation and modification methods for promoting the adsorption performance of biochar for sulfur-containing gaseous pollutants, which mainly include acid/alkali activation, microwave activation, N-doping modification, metal oxide modification, and some other emerging modification methods. Figure 1 shows the common preparation, activation, and modification methods of biochar.

In the past many years, some review articles have introduced the research progress of biochar in adsorption of H₂S, SO₂, and COS. Gwenzi et al.²⁹ reviewed the development status of air pollutants and greenhouse gases, mainly including SO₂, NO_x, Hg⁰, CO₂ and VOCs by biochar. Bamdad et al.³⁰ reviewed the dynamic and static adsorption isotherms, mechanisms, and process systems of CO₂ and H₂S removal by biochar. Sun et al.³¹ reviewed the research advances of the desulfurization process using carbon-based materials, mainly including the introduction of adsorption mechanism and modification methods of H₂S by biomass activated carbon. Wei et al.³² reviewed the research situation and principle of adsorption of COS from blast furnace gas using porous biochar. Xu et al.³³ reviewed the mechanism and influencing factors of H₂S adsorption by biochar and discussed the methods to enhance the adsorption capacity of H₂S by biochar.

It can be seen that most of the existing reviews mainly focus on the introduction of adsorbent performance and their applications in different fields and mainly use relatively small spaces to introduce the key activation and modification methods of biochar. Besides, most of the existing reviews have been published for over 3–5 years, and some of the results and data in these reviews appear outdated. In recent years, a large number of new research results have been published in the field of sulfur-containing gaseous pollutants by biochar adsorption. It is necessary to publish a new review specifically introducing the recent research advances in activation and modification methods of biochar for adsorbing

Table 2. Summary of Pore Parameters, Adsorption Properties, and Mechanism of the Activated Biochar: Acid/Alkali Activation and Microwave Activation

biochar raw material	pollutant	activation reagents	adsorption performance					main active sites/adsorption mechanism	ref
			pore parameters		adsorption capacity (mg/g)				
			SSA ^a (m ² /g)	TPV ^b (cm ³ /g)	AT ^c (°C)	BD ^d	AD ^e		
Wood	H ₂ S	H ₃ PO ₄	1620	1.27	25	/	17.3	High specific surface area and carboxylic acids promote the adsorption of H ₂ S.	80
Oil palm shell	H ₂ S	H ₂ SO ₄	1014	/	25	46	76	Phenols and carboxylic acids promote the oxidation of H ₂ S.	62
		KOH	1148				68	Surface alkalinity is increased, and pyran promotes the decomposition of H ₂ S.	
Sargassum	H ₂ S	H ₂ SO ₄	556.56	0.436	25	5.8	8.5	It is mainly physical adsorption, and the pore structure of biochar is improved significantly.	63
Enteromorpha		H ₂ O ₂	310.63	0.335		0.58	7.28	Containing-oxygen functional groups such as C–O and COO– promote the oxidation of H ₂ S.	
Olive pomace	SO ₂	H ₃ PO ₄	1456	0.9	30	/	380.17	It is mainly physical adsorption, which is promoted by high specific surface area and big pore volume.	65
Coconut shell	SO ₂	HNO ₃	971	0.2	100	26	44	C=O and transition state (π – π^*) promote the catalytic oxidation of SO ₂ by biochar.	66
Oil palm shell	SO ₂	H ₃ PO ₄	1563	/	25	14.55	16.08	It is mainly physical adsorption, and high specific surface area promotes the SO ₂ adsorption.	64
		KOH	1408	/			15.34	Pyranone and pyranone promote the adsorption of SO ₂	
Walnut shell	H ₂ S COS	KOH	1239.92	0.766	/	2.5	52.67	Chemisorption and hydroxyl group promotes the oxidation of H ₂ S.	70
Corncoobs	H ₂ S	KOH	1618	0.8117	25	/	164.54	High specific surface area and good porosity promote the adsorption of H ₂ S.	81
Chlorella	H ₂ S	KOH	428.95	0.28	25	Almost	59.1	Containing-oxygen functional groups (–OH, C–O, and C=O) promote the adsorption of H ₂ S.	74
Spirulina			350.91	0.22		0	42.3		
Sludge and corn stalks	H ₂ S	KOH	369.27	0.256	50	/	7	The improvement of pore structure and the increase of alkalinity of biochar promote the adsorption of H ₂ S.	72
Coffee	H ₂ S	CO ₂	2000	/	25	97.3	281.5	Basic functional groups promote H ₂ S to form S ⁰ .	82
Coconut shell	H ₂ S	NaOH	815	0.38	25	11.6	105	High pH value and abundant surface metal ions promote the dissociation of H ₂ S.	83
Coconut shell	H ₂ S	NaOH	1234	0.44	30	16.7	25	Chemisorption and H ₂ S reacts with basic compounds to promote the adsorption of H ₂ S.	84
Coconut shell	SO ₂	KOH	719	/	550	14	301	The –OH groups on the biochar surface promote the oxidation of SO ₂ to sulfate and SO ₃ .	77
					230	0.04 mmol/g	0.53 mmol/g		
Waste wood powder	SO ₂	KOH	584	0.33	45	60	90	Adsorption capacity of biochar for SO ₂ is enhanced by higher specific surface area and better porosity.	78
Silver birch	SO ₂	KOH	1700	0.7	25	20.4	35.3	Higher specific surface area promotes the physical adsorption of H ₂ S.	79
Sewage sludge	H ₂ S	Microwave	476.87	0.212	35	/	78.4%	Higher specific surface area and more metal irons promote the adsorption of H ₂ S.	85
Chlorella	H ₂ S	Microwave	747.2	0.44	25	59.1	96.12	Higher specific surface area and more –OH, C–O, C=O groups promote the adsorption of H ₂ S.	86
Spirulina		KOH	568.3	0.4		42.3	69.42		
Rice straw	H ₂ S	Microwave	1999.96	1.191	25	/	57.91	Containing-oxygen functional groups such as C=O, C–O, and COOH promote the oxidation of H ₂ S to S ⁰ and sulfate.	87
Wheat straw		KOH	877.16	0.527			61.47		
Sewage sludge	H ₂ S	Steam	169.9	0.252	30	/	9.149	Physical adsorption is dominant.	88
		Microwave							
		CO ₂	42.1	0.135			3.415	In the chemisorption, C–O and C=O promote the adsorption of H ₂ S to produce sulfate and sulfide.	
Coconut shell	H ₂ S	Microwave	3121	/	/	25.9	77.47	The superior specific surface area and pore structure promote the physical adsorption of H ₂ S.	89
Conifer sawdust	H ₂ S	Microwave	367	0.23	25	/	2.6	The presence of mineral substance is favorable for H ₂ S adsorption.	90
Sludge	H ₂ S	Microwave H ₂ O	50.98	/	30	1.47	22.83	Oxygen-containing functional groups and ferric oxide promote the oxidation of H ₂ S to FeS and SO ₄ ^{2–} .	91
Coconut shell	SO ₂	Microwave	646.3	0.344	60	10.9	16.9	Higher specific surface area promotes SO ₂ adsorption.	92
								Microwave radiation reduces the acid group, which thus promotes the adsorption of SO ₂ .	

Table 2. continued

biochar raw material	pollutant	activation reagents	adsorption performance					main active sites/adsorption mechanism	ref
			pore parameters		adsorption capacity (mg/g)				
			SSA ^a (m ² /g)	TPV ^b (cm ³ /g)	AT ^c (°C)	BD ^d	AD ^e		
Coconut shell	SO ₂	Microwave	983.7	0.39	60	/	16.36	It is mianly physical adsorption, and the microporous structure promotes the adsorption of SO ₂ .	93
		Microwave K ₂ Cr ₂ O ₇	866.6	0.28			40.9	Chemisorption, and alcohol and carboxyl groups promote the SO ₂ oxidation.	
Oil palm fiber	SO ₂	CO ₂	270.6	0.16	100	18.62	33	Higher specific surface areas enhance the adsorption capacity of biochar for SO ₂ .	27
Walnut shell	COS	KOH	484.26	0.272	60	2.5	52.67	Larger specific surface area and pore volume promote the COS adsorption.	70
Wood	H ₂ S CS ₂							The hydroxyl group promotes the oxidation of COS.	
	COS	CO ₂	473.61	0.22	/	/	0.234 cm ³ /g	It is mainly physical adsorption, and a small part of COS is removed by COS + H ₂ O → CO ₂ + H ₂ S.	94
								The surface metals facilitate this removal process.	
Coconut shell	COS	KOH	858	0.47	40	5.81	40.64	COS reacts with —OH to form H ₂ S, which is further oxidized to elemental sulfur and sulfate.	95
Tobacco pole	COS	KOH	1639.8	0.78	60	/	45.25	The excellent pore structure and C—O, -OH functional groups promote the adsorption and hydrolysis of COS.	96
	H ₂ S CS ₂	CO2	699.53	0.32			184.68	The C—O, C=O groups and basic groups promote the hydrolysis of COS and the oxidation of COS to S ⁰ and sulfate.	

^aSSA: Specific surface area. ^bTPV: Total pore volume. ^cAT: Adsorption temperature. ^dBD: Before activation. ^eAD: After activation.

sulfur-containing gaseous pollutants (H₂S, SO₂, COS). Therefore, this review discusses the recent research advances of activation and modification methods of biochar for adsorbing sulfur-containing gaseous pollutants (H₂S, SO₂, and COS). Various activation and modification methods are classified and summarized, mainly including acid and alkali activation, microwave activation, metal oxide modification, nitrogen doping modification and other emerging modification methods. The main process parameters, advantages, and disadvantages of each modification and activation method are introduced in detail. The activation and modification mechanisms, as well as the adsorption mechanisms of sulfur-containing gaseous pollutants, have been discussed and summarized. This review will provide needed guidance and inspiration for researchers in the related research areas to develop new biochar-based desulfurization adsorbents.

2. ACID AND ALKALI ACTIVATION

In recent years, carbon-based materials such as activated carbon have been widely used in the adsorption of gaseous pollutants due to their large specific surface area and abundant surface functional groups.^{46–48} However, the general commercial activated carbon is made from coal, petroleum, and other fossil fuels, which is expensive, and the source is limited. Compared with activated carbon, biochar is a solid byproduct converted from the pyrolysis of agricultural and forestry waste, etc.^{49–51} Due to the wide range of raw material sources and low cost, biochar has become a potential alternative to activated carbon and has received extensive concern in various fields such as water treatment, flue gas purification, and soil remediation.^{52–55}

Although biochar has a wide range of sources and low price, its pore structure is underdeveloped, and the active functional groups and active sites on carbon surface are few. Therefore, activation and modification of the biochar are needed through various methods. Activation can generally be divided into physical and chemical activation. The main physical activators

are CO₂, O₂, steam, and air or a mixture of them.^{56,57} Physical activation is a relatively green way of activation, which can increase the porosity of biochar and also introduce some containing-oxygen functional groups on the surface of biochar.⁵⁸ However, the physical activation process requires more energy because the process takes place at high temperatures for a long time.^{59,60} Chemical activation mainly uses acids (HNO₃, H₃PO₄, and H₂SO₄) and alkali (KOH, NaOH, and K₂CO₃) to activate biochar. In the process of chemical activation, the pore structure of biochar will be more developed due to the production of gaseous byproducts, and the functional groups introduced on the surface of biochar can also enhance the affinity of biochar for gaseous pollutants. Compared with physical activation, the advantages of chemical activation are that the pore structure is more developed, and the surface sites and active functional groups are also more abundant, which usually has better activation effects.^{29,60,61}

Table 2 describes the summary of pore parameters, adsorption properties, and mechanisms of the acid/alkali-activated biochars for adsorbing SO₂ and H₂S.

2.1. Acid Activation. **2.1.1. H₂S.** Sulfuric acid is one of the most common acid activators of biochar. Guo et al.⁶² used a 40% H₂SO₄ solution to activate oil palm shell biochar at room temperature to remove gaseous H₂S. The results showed that the H₂S adsorption capacity of biochar was 76 mg/g at room temperature, which is better than that of CO₂ and H₂O-activated biochars (46 mg/g and 53 mg/g, respectively). This performance improvement is mainly attributed to the significant increase in oxygen content on the H₂SO₄-activated biochar, which is mainly due to the increase of containing-oxygen functional groups such as carboxyl, phenolic, lactone, and carbonyl groups. The presence of these oxygen-containing functional groups enables the activated oil palm shell biochar surface to undergo chemisorption by forming hydrogen bonds, which can oxidize H₂S into S⁰, thus effectively facilitating the removal process of H₂S.

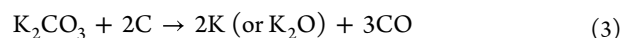
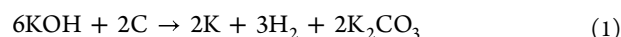
In order to further increase the content of containing-oxygen functional groups, Chen et al.⁶³ used a $\text{H}_2\text{SO}_4/\text{H}_2\text{O}_2$ combination reagent to activate two types of algal biochars (i.e., *Sargassum* (S) and *Enteromorpha morpha* (E)) to remove H_2S . Studies have shown that sulfuric acid can improve the pore structure of biochar, while hydrogen peroxide can provide more abundant containing-oxygen functional groups on the biochar surface. Therefore, after the combined activation of these two activators, the specific surface area of biochar is greatly increased by nearly 20 times (S: 26.2 m^2/g to 556.56 m^2/g , E: 21.15 m^2/g to 310.63 m^2/g), and a large number of containing-oxygen functional groups (such as COO^-) are produced. Compared with one activator alone, the H_2S adsorption capacity of biochar was better after the combining activation via two activators (S: 5.8 to 8.5 mg/g ; E: 0.58 to 7.28 mg/g). It is shown that sulfate is produced after H_2S adsorption on the biochar, which is due to the existence of a large number of containing-oxygen functional groups on the surface of the modified seaweed biochar, so that H_2S molecules are adsorbed on the active sites, and a series of oxidation and/or substitution reactions occur on the modified seaweed biochar surface.

2.1.2. SO_2 . Phosphoric and nitric acids are the other two most common acid activation reagents for biochar. Guo et al.⁶⁴ activated oil palm shell biochar using H_3PO_4 and KOH with different concentrations, and compared the effects of activator concentration on surface morphology and SO_2 adsorption capacity of biochar. Their study showed that biochar activated with 40% H_3PO_4 had the largest specific surface area (1563 m^2/g) and porosity (72.9%). However, the biochar obtained by the activation of 10% KOH was more conducive to the adsorption of SO_2 (15.34 mg/g). This is because after activation of KOH, some functional groups such as $\text{C}=\text{O}$, $\text{C}=\text{C}$, $\text{C}-\text{O}$, $\text{C}-\text{H}$, and so on were produced, which were conducive to the adsorption of SO_2 . Yavuz et al.⁶⁵ used olive factory waste residue as a raw material and H_3PO_4 as an activator to prepare an olive core biochar for adsorption of SO_2 . The results showed that the SO_2 adsorption capacity of biochar was improved after the activation of H_3PO_4 , and the SO_2 adsorption capacity of biochar was 380.17 mg/g at 30 $^\circ\text{C}$. The specific surface area significantly increased from 525.2 to 1456 m^2/g and micropore volume increased from 0.22 to 0.59 cm^3/g of biochar with the increase of H_3PO_4 concentration, but the pore size remained unchanged. It is proven that the large specific surface area and pore volume of biochar promote the adsorption of SO_2 .

Liu et al.⁶⁶ used HNO_3 to activate coconut shell biochar loaded with MnO_2 to study the effect of HNO_3 concentration on the adsorption of SO_2 by biochar. Their results showed that when the adsorption temperature was 100 $^\circ\text{C}$ and SO_2 was 500 ppm, the maximum sulfur capacity was 44 mg/g , while the biochar without HNO_3 activation was only 26 mg/g . They found that the specific surface area was decreased from 1012 to 971 m^2/g , and the pore volume was slightly increased from 0.17 to 0.20 cm^3/g after HNO_3 activation. In addition, the number of containing-oxygen functional groups ($\text{C}-\text{O}$, $\text{C}=\text{O}$) was increased after the HNO_3 activation. When the concentration of HNO_3 was 10 mol/L , the content of the oxygen group $\text{C}=\text{O}$ was the highest (16.55%). The increase in carbonyl carbon ($\text{C}=\text{O}$) and transition ($\pi-\pi^*$) groups on the surface of biochar after the HNO_3 activation effectively promoted the removal of SO_2 .

2.2. Alkali Activation. Alkali (KOH, NaOH, K_2CO_3 , etc.) is also extensively used to activate biochar because this technology has a simple process and can simultaneously improve the pore structure and surface chemical properties of carbon-based materials.^{67–69} The alkali activation can increase the specific surface area, micropore volume basic groups (hydroxyl), and surface alkalinity of biochar, which is conducive to acid gas adsorption. Other studies^{70–72} have shown that the alkaline environment is conducive to the formation of elemental sulfur and the improvement of sulfur adsorption capacity.

Activation of biochar by KOH is beneficial to the improvement of its surface morphology, the increase of alkaline groups, and the enhancement of H_2S and SO_2 adsorption capacity. In the process of high temperature activation, CO_2 , H_2 , and CO are released, which will promote the formation of pores and the improvement of the pore structure. The activation mechanism is described as follows:⁷³



2.2.1. H_2S . Among alkaline activators, KOH is often used as an activator for activating biochar due to its effectiveness and low cost. Li et al.⁷⁰ used KOH to activate walnut shell biochar, and studied its ability to simultaneously remove H_2S , COS, and CS_2 . Results showed that when the calcination temperature was 600 $^\circ\text{C}$, the KOH/biochar mass ratio was 0.5, and the activation temperature was 700 $^\circ\text{C}$, and the sulfur capacity of the activated walnut shell biochar was 52.67 mg/g . The biochar activated with KOH exhibited the highest specific surface area (increased from 1.28 m^2/g to 1239.92 m^2/g) and pore volume (increased from 0.005 cm^3/g to 0.673 cm^3/g) compared to several other activators (H_3PO_4 , ZnCl_2 , K_2CO_3). In addition, increasing the mass ratio of KOH to biochar resulted in the formation of more micropores and the generation of more alkaline hydroxyl groups, which is favorable for the adsorption of H_2S . XPS analysis showed that most of the products chemisorbed on the biochar surface were elemental sulfur ions (S^0) and sulfate ions (SO_4^{2-}).

Zeng et al.⁷² used KOH-activated sludge and corn straw biochar for adsorption of H_2S . Research indicated that when the mass ratio of sludge, corn straw, and potassium hydroxide was 3:7:2, the desulfurization performance was the best, and the H_2S adsorption capacity was 7.00 mg/g , which was 5.74 times that of the unactivated sample. They suggested that KOH activation led to the increase of the specific surface area from 93.215 to 369.27 m^2/g and the increase of the pore volume from 0.14 to 0.256 cm^3/g . While the decrease of lactone groups and the increase of alkaline groups (2.666 mmol/g) created a more alkaline environment, which is conducive for the H_2S removal. During the adsorption, H_2S reacted with the basic functional groups to form elemental sulfur and sulfate.

Ma et al.⁷⁴ used KOH solution to activate two kinds of algal biochar (*Chlorella* and *Spirulina*) at room temperature for adsorption of H_2S . Their results showed that the H_2S adsorption capacity of KOH-activated *Chlorella* biochar

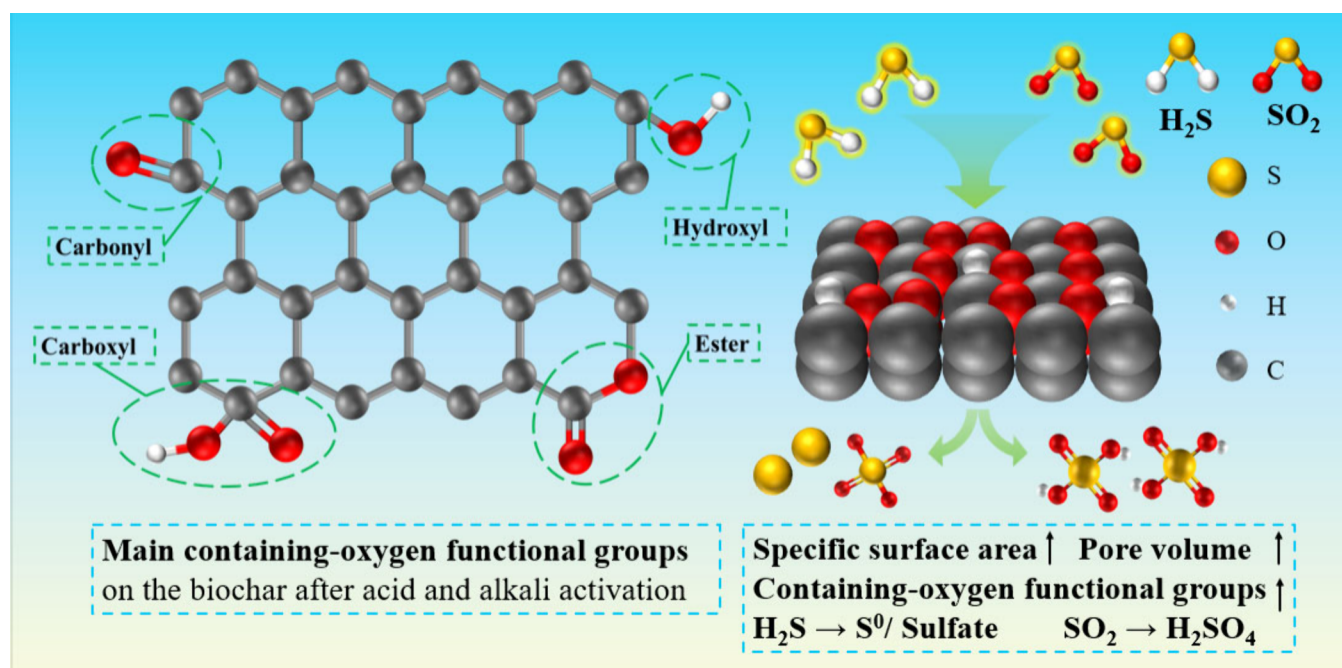


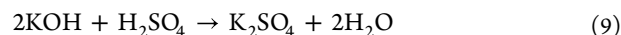
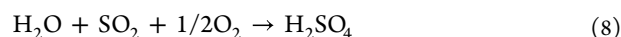
Figure 2. Adsorption mechanisms of H_2S and SO_2 on the biochar after acid/alkali activation.

reached 59.1 mg/g, and that of spirulina biochar was 42.3 mg/g, while the unmodified sample had almost no adsorption capacity under the same studied conditions. After KOH activation, the specific surface area and pore structure of both biochars were significantly improved. The specific surface area of chlorella biochar was increased from 5 to 428.957 m^2/g , while that of spirulina biochar was also increased from 5.653 to 350.91 m^2/g . By measuring the surface pH value of biochar, it was found that the surface pH value was increased from 8.7 to 9.7 after the activation of KOH. In addition, the activation of KOH increased the number of functional groups such as $-\text{OH}$, $\text{C}=\text{O}$, and $\text{C}-\text{O}$, which promoted the oxidation reaction of H_2S , and the reaction process generated elemental sulfur and sulfate.

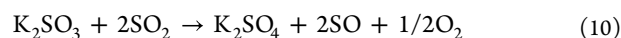
In addition to KOH activator, NaOH is also a relatively common alkaline activator due to its cheaper price. Bagreev et al.⁷⁵ used NaOH to activate wood biochar to improve its adsorption capacity for H_2S . Results showed that the adsorption breakthrough time of H_2S was increased significantly from 112 to 320 min after NaOH activation. This significant improvement is attributed to the promotion of the H_2S dissociation process by the alkaline environment. Under alkaline conditions, H_2S is more easily dissociated into HS^- , and the addition of sodium hydroxide further increases the pH value of biochar, thus increasing the concentration of HS^- . Subsequently, the HS^- produced is further oxidized and converted into sulfate or carbonate, thus achieving the effective adsorption removal of H_2S on the activated wood biochar.

2.2.2. SO_2 . Activation by alkali (KOH, NaOH, etc.) can contribute to the increase of containing-oxygen functional groups on the biochar surface, and the containing-oxygen functional groups are extensively thought to be the key catalytic centers for the oxidation of SO_2 to SO_3 .⁷⁶ Lee et al.⁷⁷ used KOH solution as an activator and prepared a coconut biochar for adsorption of SO_2 . Results showed that the desulfurization ability of biochar was greatly increased from 0.04 to 0.5298 mmol/g after KOH activation. The character-

ization results of the biochar surface physical and chemical properties showed that the surface containing-oxygen functional groups ($\text{C}=\text{O}$, $\text{C}-\text{O}$, $-\text{OH}$) of biochar were greatly increased after KOH activation, and these functional groups played an important role in providing adsorption sites for the selective adsorption of SO_2 . They found that in the chemisorption process, the containing-oxygen functional groups could react with SO_2 to form $-\text{SO}_4$ groups and $-\text{SO}_3$ groups, which greatly promotes the adsorption of SO_2 . Due to the action of potassium, the adsorbed SO_2 existed in the form of stable oxide crystals on biochar, such as K_2SO_3 and K_2SO_4 . The relevant reactions are as follows:⁷⁷



At high temperatures, due to evaporation of H_2O , the following reaction process can further occur:⁷⁷



Atanes et al.⁷⁸ activated waste wood meal biochar by KOH to remove SO_2 from flue gas. Studies had shown that the SO_2 adsorption capacity of the activated biochar was increased from 20 to 90 mg/g at 45 °C. After KOH activation, the pore structure was optimized, and the specific surface area of biochar was greatly increased from 7 m^2/g to 584 m^2/g . Besides, more containing-oxygen functional groups (such as alcohols, phenols, and carboxyl groups) were introduced onto the activated biochar surface. Their results showed that the adsorption process of SO_2 was mainly physical adsorption. The higher specific surface area and better porosity enhanced the adsorption capacity of biochar for SO_2 , and the lower surface acidity was more conducive to the adsorption of SO_2 .

Table 3. Summary of Pore Parameters, Adsorption Properties, and Mechanisms of the Modified Biochar: Metal Oxide Modification, Nitrogen Doping, and Other Modification Methods

biochar raw material	pollutant	modification reagents	pore parameters		adsorption performance			main active sites/adsorption mechanism	ref
			SSA ^a (m ² /g)	TPV ^b (cm ³ /g)	AT ^c (°C)	BD ^d	AD ^e		
Grapefruit skin	H ₂ S	Cu(NO ₃) ₂ ·3H ₂ O	966	0.417	25	12.1	358.3	CuO + H ₂ S → CuS + H ₂ O	116
Rice straw	H ₂ S	Cu(NO ₃) ₂ ·3H ₂ O	60.78	0.053	125	/	1191.1	2CuS → Cu ₂ S + S	111
	H ₂ S	Cu(NO ₃) ₂ ·3H ₂ O	36.05	0.034	120	/	1000.6	CuO + H ₂ S → CuS + H ₂ O	118
Coconut shell	H ₂ S	Cu(NO ₃) ₂ ·3H ₂ O	/	/	25	148 min	287 min	2CuS → Cu ₂ S + S	119
Coffee base	H ₂ S	CuCl ₂ ·2H ₂ O	1422	0.655	20	/	132.22	C–O, O–H, C–H groups	117
Peanut shell	H ₂ S	CuCl ₂ ·2H ₂ O	/	/	20	/	97.63	Cu ₂ O + H ₂ S → Cu ₂ S + H ₂ O	67
Coconut shell	H ₂ S	Zn(NO ₃) ₂	622.6	0.33			289.3	C–O, C–H, C≡O groups	
	H ₂ S	KMnO ₄	191.1	0.15	/	37.7	229	ZnO + H ₂ S → ZnS + H ₂ O	123
Maize straw	H ₂ S	Zn(NO ₃) ₂ + KMnO ₄	450.3	0.38			334.7	MnO ₂ + 2H ₂ S → MnS ₂ + 2H ₂ O	124
Maple	H ₂ S	FeCl ₃ ·H ₂ O	34.9	/	/	3.3	8.2	3Mn ₃ O ₄ + H ₂ S → SO ₂ + 9MnO + H ₂ O	
Coconut shell	H ₂ S	Cu(CH ₃ COO) ₂ ·H ₂ O K ₂ CO ₃	830	0.49	120	242.1	373.4	Fe ₃ O ₄ + 4H ₂ S → 3FeS + 4H ₂ O + S 3S + H ₂ O + 4O ₂ + Fe ₃ O ₄ → 3FeSO ₄ + H ₂ O	158
Coconut shell	H ₂ S	KMnO ₄	390.9	0.131	40	13.8	110.7	The synergistic effect of potassium carbonate and copper oxide promote the production of ·O ₂ [−] and then further improve the catalytic activity.	
Walnut shell	H ₂ S	Mn(NO ₃) ₂	921.7	/	350	/	72.4	MnO ₂ + 2H ₂ S → MnS ₂ + 2H ₂ O	159
Palm kernel shell	H ₂ S	MgNO ₃ ·7H ₂ O	274	0.15	/	30%	98.6%	3Mn ₃ O ₄ + H ₂ S → SO ₂ + 9MnO + H ₂ O	125
	H ₂ S	Ce(NO ₃) ₃ ·6H ₂ O NaOH	660	0.33	30	1.8	83.1	MnO + H ₂ S → MnS + H ₂ O	160
Wood	H ₂ S	Fe(NO ₃) ₃ ·9H ₂ O Zn(NO ₃) ₂ ·6H ₂ O	1403	0.8	25	5.6	122.5	MgO + H ₂ S → MgS + H ₂ O	
Waste rice	H ₂ S	ZnCl ₂ FeCl ₃	1065	0.56	25	12.11	228.29	C≡O and COOH groups promote the oxidation of H ₂ S.	126
Waste rice	H ₂ S	Mg(NO ₃) ₂ FeCl ₃ Melamine	222.91	0.31	25	15.49	1052.83	Ce ₂ O ₃ + 3H ₂ S → 2CeS + 3H ₂ O + S	108
Rice	H ₂ S	ZnCl ₂ CaCl ₂	523.6	0.47	25	29.31	834.48	ZnFe ₂ O ₄ + 4H ₂ S → ZnS + Fe ₂ S ₃ + 4H ₂ O	129
Wood	H ₂ S	Cu(NO ₃) ₂ Fe(NO ₃) ₃ ·9H ₂ O	1176	0.752	25	214	667	ZnS + 2FeOOH + 3H ₂ O + 3S	130
Coconut shell	SO ₂	Cu(NO ₃) ₂ ·3H ₂ O	843	0.416	250	/	43	MgFe ₂ O ₄ + 4H ₂ S + 3/2O ₂ →	131
Walnut shell	SO ₂	TiO ₂	661.8	0.377	90	70	228.62	MgS + 2FeOOH + 3H ₂ O + 3S	
								Pyridinic-N promotes the dissociation of H ₂ S.	133
								H ₂ S + CaCO ₃ → CaCO ₃ + H ⁺ + HS [−] ZnO + 4H ₂ S + 3/2O ₂ → ZnS + 4H ₂ O + 3S	134
								CuFe ₂ O ₄ + 4H ₂ S → CuS + Fe ₂ S ₃ + 4H ₂ O	161
								SO ₂ + 1/2O ₂ → SO ₃ SO ₃ + CuO → CuSO ₄	
								CuO + 1/2O ₂ + SO ₂ → CuSO ₄	
								Metal oxide produces ·O ₂ [−] through electron transfer, which promotes the catalytic oxidation of SO ₂ .	

Table 3. continued

biochar raw material	pollutant	modification reagents	pore parameters		adsorption performance			ref
			SSA ^a (m ² /g)	TPV ^b (cm ³ /g)	AT ^c (°C)	BD ^d	AD ^e	
Palm shell	SO ₂	Fe ₂ O ₃	791.0	0.687			125	162
		Ce(NO ₃) ₃ ·6H ₂ O	807	0.33	150	18.62	121.7	
Straw straw	SO ₂	Ethylendiamine	138.1	0.36	25	11.1	194.6	113
		Tetraethylenepentamine	139.2	0.42			260	
Soybean stalks	SO ₂	MgCl ₂ ·6H ₂ O	355.6	/	75	96	146	163
		Microwave Fe(NO ₃) ₃					146	
Coconut shell	H ₂ S	Urea	808	0.432	30	27.2	54.1	140
		Melamine	732	0.386			71.9	
Palm shell	H ₂ S	Urea	1000.62	0.51	30	61.1	356.9	144
		Urea phosphate	1189	0.63	30	0.24	54.8	
Rice hull	H ₂ S	NH ₃ ·H ₂ O	170	0.1	25	2.54	23	143
		NH ₃	354	0.18			9.58	
Starch	H ₂ S	NH ₃ ·H ₂ O	2084.1	1.004	25	27.13	50.2	145
		Alanine	1613	0.796			110.7	
Poplar sawdust	H ₂ S	Melamine cyanuric acid ester	1914.3	0.82	30	1.0	1827	149
		Melamine	1839.0	1.12	25	12.5	426.2	
Cypress wood	H ₂ S	Melamine cyanurate (MCA)	805	0.5	25	48.76	390.1	165
		Urea	1065	0.78	25	72.9	356.5	
Sludge Pine sawdust	SO ₂	NH ₃	371.1	/	30	92.8	175.9	155
		NH ₃ ·H ₂ O	108	0.09	70	46.3%	115.4	
Corn cobs	SO ₂	Diethanolamine (MDEA) -methanol	654.0	0.32	120	58.0	216.9	156
		MDEA	Near 0		120	57.8	156.2	
Soybean straws	SO ₂	NH ₃ +CO ₂	500.2	/	30	98.6	201.9	142
					120	58.6	143.68	

Table 3. continued

biochar raw material	pollutant	modification reagents	pore parameters		adsorption performance			main active sites/adsorption mechanism	ref
			SSA ^a (m ² /g)	TPV ^b (cm ³ /g)	AT ^c (°C)	BD ^d	AD ^e		
Walnut shell	H ₂ S	NH ₃ -plasma	640.9	0.2954	60	/	390 min	The pore structure is improved, and the functional groups such as C≡O/O-O-O-, O-C≡O-, amino groups promote the oxidation of H ₂ S to S ⁰ and SO ₄ ²⁻ .	167
Corn stalks	H ₂ S	UV H ₂ O ₂	535.44	0.31	25	2 min	24 min 90.9	UV/H ₂ O ₂ modification increases the surface containing-oxygen groups, such as -OH, C-O, COO- groups, which promotes the removal of H ₂ S.	168
Soybean straws	SO ₂	UV CO ₂	347.28	0.37	70	/	100.181	CO ₂ activation promotes the formation of micropores.	169
								UV modification promotes the formation of containing-oxygen functional groups (carboxyl).	
Tobacco stem	COS	Cu(NO ₃) ₂ Fe(NO ₃) ₃	554	0.29	60	/	231.28	Metal oxides facilitate the hydrolysis of COS and promotes the following process:	170
								COS + CuO + H ₂ O → CuO·H ₂ S + CO ₂	
Coconut shell	COS	Cu(NO ₃) ₂	768.4	0.3	20	7.5	14.8	Containing-oxygen functional groups (such as hydroxyl, carboxyl, etc.) and metal oxides participate in the adsorption of COS, and the adsorption product is sulfate.	171
Tobacco pole	H ₂ S COS CS ₂	Cu(NO ₃) ₂	514.77	0.26	60	/	161.93	Metal oxides and -OH groups promote the hydrolysis of COS, and the H ₂ S produced by hydrolysis is oxidized to elemental sulfur and sulfate.	96

^aSSA: Specific surface area. ^bTPV: Total pore volume. ^cAT: Adsorption temperature. ^dBD: Before modification. ^eAD: After modification.

Braghiroli et al.⁷⁹ compared the SO₂ adsorption capacity of birch biochar activated by CO₂, steam and KOH, respectively. Research indicated that the biochar activated by KOH had the largest specific surface area and porosity (1700 m²/g and 0.7 cm³/g), and the SO₂ adsorption capacity was increased from 20 to 35.3 mg/g. Functional groups such as phenols, lactones, and carboxyl groups were abundant on the surface of biochar activated by KOH. In contrast, the steam-activated biochar had a relatively lower specific surface area (590 m²/g), but possessed a higher SO₂ adsorption capacity (76.9 mg/g). This is because the steam-activated biochar has fewer acidic groups and therefore resulted in a higher SO₂ adsorption capacity. Figure 2 shows the types of the main oxygen-containing functional groups on the surface of biochar after the acid/alkali activation and the adsorption mechanisms of H₂S and SO₂.

3. METAL OXIDES MODIFICATION

Metals oxides (e.g., Fe, Mn, V, Ti, Ce, Cu, Co, Ni, etc.) are often used as desulfurization adsorbents due to their strong affinity with sulfur-containing gases.^{97–102} However, pure metals and metal oxides used as adsorbents usually suffered from evaporation, sintering, and mechanical disintegration, which will result in the loss of specific surface area and porosity, thereby adversely affecting their performance and lifetime.¹⁰³ To overcome these problems and improve the removal performance, the metals can be loaded onto the support materials with a high specific surface area.^{103–107} The large surface area and high porosity of the support materials can contribute to the high dispersion of metal oxides, and the pore channels of the support materials can limit the growth of metal oxide particles.^{108–110} Related research^{41,60,111–114} had shown that the combination of metals and biochar materials could effectively increase the surface active sites of biochar, enhance the electron transfer on carbon surface, and improve the reactivity between biochar and gas pollutant molecules, thereby strengthening the adsorption capacity of adsorbent for H₂S and SO₂. Table 3 shows the summary of pore parameters, adsorption properties, and adsorption mechanisms of the metal oxide-loaded biochar for adsorption of H₂S and SO₂. Figure 3

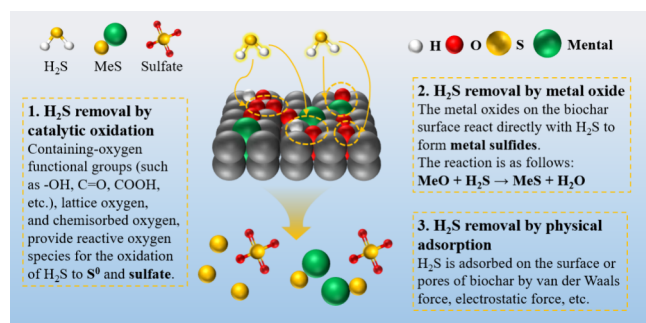
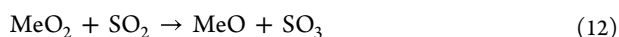
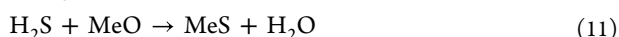


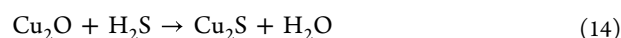
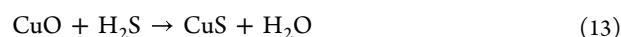
Figure 3. Mechanism of H₂S adsorption by metal oxide-supported biochar.

and Figure 4 show the adsorption processes of H₂S and SO₂ on biochar loaded with metal oxides, respectively. The corresponding chemical reactions are as follows:¹¹⁵



3.1. H₂S. **3.1.1. Single Metal Modification.** Copper-based oxides are one of the most common biochar modification reagents for enhancing the H₂S removal because of its high sulfur balance constant, high desulfurization accuracy, and good dispersion.^{111,116–119} Cui et al.¹¹¹ used microwave-activated rice straw biochar as the carrier, and prepared a CuO-loaded rice straw biochar (CuWRS) to study its adsorption capacity of H₂S. Studies showed that the breakthrough time of H₂S increased from 11 to 78 min after loading CuO. When the calcination temperature was 300 °C and the Cu/WRS mass ratio was 2.56, the adsorption effect was the best, and the sulfur capacity could reach up to 1191.1 mg/g, which is more than many similar Cu-based adsorbents. It was found that although copper loading reduced the specific surface area of biochar (280 m²/g to 60.78 m²/g), a large number of CuO adsorption active sites with strong oxidation ability were formed on biochar, which greatly enhanced H₂S removal. In the adsorption process, lattice oxygen participated in the chemisorption, and Cu²⁺ in CuO was reduced. The main products were CuS and small amounts of CuSO₄ and Cu₂S.

Zhao et al.¹¹⁸ used a waste Cu-based Fenton-like reagent as a modifier reagent to prepare CuO-loaded straw biochar to remove H₂S. Results showed that the maximum H₂S adsorption capacity of the modified biochar was 1000.6 mg/g at a calcination temperature of 300 °C and reaction temperature of 120 °C. The modification of the waste Cu-based Fenton-like reagent greatly promoted the increase of biochar surface metal active sites, but led to the decrease of the specific surface area and the pore volume of the adsorbent from 280.07 m²/g to 36.06 m²/g, and from 0.1 cm³/g to 0.034 cm³/g, respectively. CuO was found to be the key active substance to promote the chemisorption of H₂S on the adsorbent. The main products of the reaction between H₂S and CuO on the adsorbent were determined to be copper sulfide, cuprous sulfide, and elemental sulfur. The reaction processes between copper based-modified biochar and H₂S can be described by the following chemical reactions:^{117,118}



Zinc-based oxides have good potential to adsorb H₂S at room temperature due to good thermodynamics at room temperature, which have been widely studied as a desulfurization agent.^{120,121} Hernandez et al.¹²² prepared plant biochar loaded with ZnO by a precipitation method, and then studied the adsorption capacity of H₂S at room temperature. Studies had shown that the loading of metal oxides provided more active sites for H₂S removal, which made the H₂S adsorption capacity of biochar significantly increase, and the H₂S adsorption capacity was 11.8 times that of the original biochar. After modification, there were well dispersed ZnO particles on the surface of biochar. The specific surface area was increased from 851 to 867 m²/g, and the pore volume was increased from 0.44 to 0.45 cm³/g. The reaction process between H₂S and ZnO on the active sites can be depicted as follows:



Zhang et al.¹²³ used Zn(NO₃)₂ solution to impregnate coconut shell biochar and prepared the coconut shell biochar modified by ZnO to adsorb H₂S. Results showed that the pore

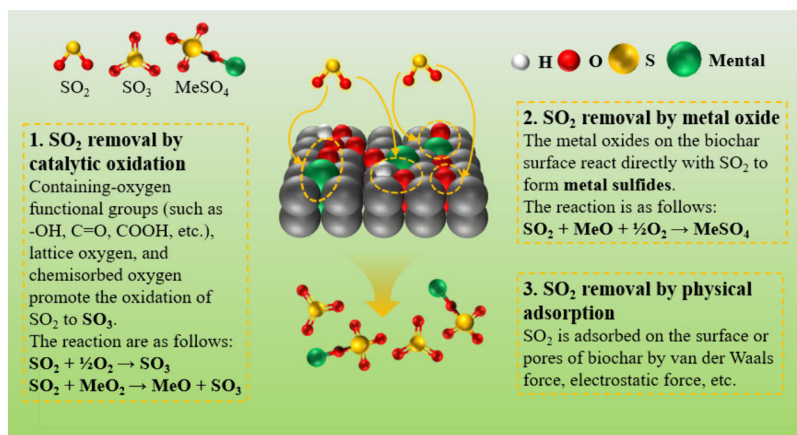
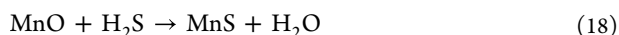
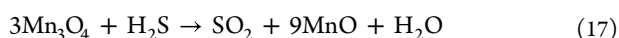


Figure 4. Mechanism of SO₂ adsorption by metal oxide-supported biochar.

plugging (0.36 cm³/g to 0.33 cm³/g) caused by ZnO loading reduced the specific surface area (from 685.7 m²/g to 622.6 m²/g). However, the increase of active sites on biochar greatly increased the H₂S adsorption capacity of biochar (from 37.7 to 289.3 mg/g), and the adsorption effect was better than that of MnO₂-loaded biochar (229 mg/g). On the biochar, H₂S was chemically adsorbed with ZnO and other active sites, and the catalytic oxidation process of H₂S was promoted by the functional groups (C–O, COO[−], and -OH) on the biochar surface.

In addition to copper-based oxides and zinc-based oxides, other metal oxides such as iron-based oxides and manganese-based oxides are also used to modify the biochar to improve the adsorption performance of H₂S. Choudhury et al.^{112,124} used as a modifier to load Fe₃O₄ on corn stover (CSB) and maple (MB) biochar to remove H₂S from biogas. Results indicated that the H₂S adsorption capacity of Fe₃O₄-loaded biochar was increased 2.5–3.9 times. The H₂S adsorption capacity of CSB was increased from 3.3 to 8.2 mg/g, and the H₂S adsorption capacity of MB was increased from 6.1 to 23.9 mg/g. A large amount of ferric oxide on the surface of biochar provided the active sites for the chemisorption of H₂S, and during the H₂S adsorption process, reactive oxygen species and metal oxides on the biochar facilitated the conversion of sulfides to elemental sulfur and sulfate.

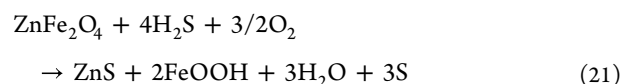
Wang et al.¹²⁵ impregnated walnut shell biochar with supercritical water and Mn(NO₃)₂ solution, and prepared biochar loaded with Mn_xO_y of different valence states to adsorb H₂S. Results showed that when the activation temperature was 350 °C, the biochar had the highest specific surface area (921.7 m²/g), and the H₂S adsorption capacity increased from 42.9 mg/g to 72.4 mg/g. They found that different carriers and different impregnation temperatures had different effects on the valence state of Mn, and the catalytic oxidation activity of Mn_xO_y to H₂S varied with different valence states. XRD results showed that when the impregnation temperature was 350 °C, the active sites on the surface of biochar mainly existed in the form of Mn₃O₄ and a small amount of MnO, and the reactions between these sites and H₂S can be expressed as follows:¹²⁵



Lau et al.¹²⁶ used NaOH and Ce(NO₃)₃·6H₂O to impregnate palm shell biochar and prepared an alkali-activated

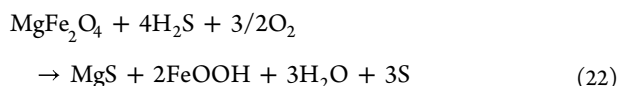
biochar loaded with CeO₂ to remove H₂S. Results demonstrated that when the Ce content was 5%, the H₂S adsorption capacity of biochar was 83.1 mg/g. They found that Ce_xO_y could provide an oxygen component for the oxidation of H₂S through the conversion between CeO₂ and Ce₂O₃. NaOH could improve the surface alkalinity of biochar and increase the contents of C–O, C=O, and COOH and other functional groups, thereby enhancing the adsorption of H₂S on the surface of biochar. During the adsorption process, Ce⁴⁺ was reduced, and S^{2−} was oxidized into S⁰, sulfite, and sulfate (S⁰ was the main product).

3.1.2. Multimetallic Modification. In addition to the use of single metal for modifying biochar, the use of multimetals for modifying biochar has also been widely studied due to the synergistic catalytic effects generated between different metals.^{123,127–129} Yuan et al.¹²⁹ used rice as raw material and ZnCl₂ and FeCl₃ as activators, and then prepared a waste rice biochar modified by ZnFe₂O₄ to achieve the adsorption of H₂S. Results showed that the sulfur capacity of the biochar prepared at 500 °C was greatly increased from 12.1 mg/g to 228.29 mg/g. Compared with the specific surface area of the original biochar of 2.76 m²/g, the modified biochar had a high surface area of 1065 m²/g. It was found that there were hydroxyl groups and nitrogen–oxygen functional groups on the surface of the modified biochar, which contributed to the adsorption and dissociation of H₂S. Besides, the activators ZnCl₂ and FeCl₃ had better pore formation effects for biochar, and the ZnFe₂O₄ was well dispersed on the biochar, without obvious metal oxide agglomeration. The ZnFe₂O₄ loaded on the surface of biochar provided abundant active sites for the H₂S adsorption, which played an important role in the removal of H₂S. The reaction mechanism is as follows:¹⁰⁸

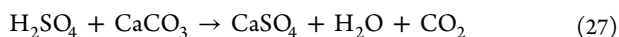
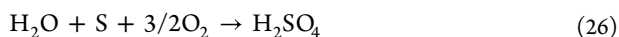
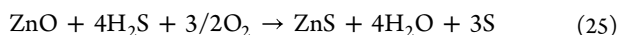
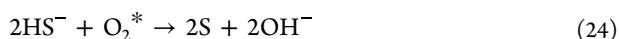


Yuan et al.¹³⁰ used melamine as the nitrogen source, and Mg(NO₃)₂ and FeCl₃ as metal activators to prepare a nitrogen-doped waste rice biochar loaded with MgFe₂O₄ for adsorption of H₂S. Results showed that the sulfur capacity of biochar was greatly increased from 15.49 mg/g to 1052.83 mg/g. After MgFe₂O₄ modification, the desulfurization process was a

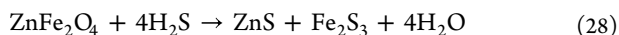
combination of catalytic oxidation and reactive adsorption, in which the presence of pyridine-N and C–O, O–C=O and other containing-oxygen functional groups on the biochar surface greatly enhanced the dissociation and oxidation of H₂S, while the MgFe₂O₄ promoted the direct reactive adsorption of H₂S. The reaction mechanism is as follows:¹³⁰



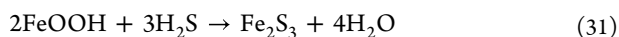
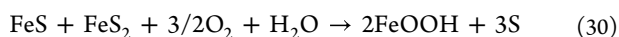
Yuan et al.¹³¹ prepared a CaCO₃–ZnO loaded rice biochar using ZnCl₂ and CaCl₂ solution as the mixed modifier to remove H₂S. Research indicated that the prepared biochar after modification had a high sulfur capacity. At 25 °C, the sulfur capacity was increased from 29.31 to 834.48 mg/g, and the specific surface area was increased from 102 to 523 m²/g after modification. They found that there was a certain amount of nitrogen in the modified biochar, mainly existing in the form of pyridine and pyrrole. The containing-nitrogen groups produced effectively contributed to the formation of alkaline water films to promote the catalytic oxidation of H₂S. The adsorption process of H₂S on the modified biochar was mainly summarized as follows: (i) H₂S is oxidized into S⁰ after dissociation, and part of S⁰ is oxidized into H₂SO₄, which then further reacts with CaCO₃ to produce CaSO₄; (ii) H₂S directly reacts with ZnO to produce ZnS. The reaction mechanism is as follows:¹³¹



Yang et al.¹⁰⁸ studied the ability of ZnFe₂O₄ loaded biochar to remove H₂S at room temperature. Research showed that when the ZnFe₂O₄ loading was 10 wt%, the H₂S adsorption capacity of the adsorbent was the largest, reaching a maximum value of 122.5 mg/g, but the H₂S adsorption capacity of the unmodified wood activated carbon was only 5.6 mg/g. They found that ZnFe₂O₄ was highly dispersed on the biochar matrix and provided a large number of active sites for the H₂S adsorption. The reaction process between ZnFe₂O₄ and H₂S is as follows:¹³²



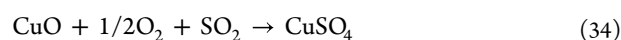
Moreover, due to the instability of the generated Fe₂S₃, the following reactions will occur, which further greatly improves the removal ability of H₂S:¹³²



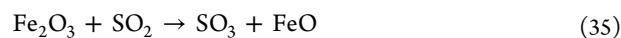
Chen et al.¹³³ used Cu(NO₃)₂ and Fe(NO₃)₃·9H₂O as activators to synthesize the wood biochar loaded with CuFe₂O₄ to adsorb H₂S. Results showed that the H₂S adsorption capacity of the biochar was increased from 214 mg/g to 667 mg/g at room temperature when the metal load was 20 wt%. The removal process of H₂S included two

pathways: active adsorption and catalytic oxidation. In the desulfurization process, the formed FeOOH acted as a catalyst intermediate, which promoted the production of products (including sulfides, elemental sulfur, and sulfates). Micropores could enhance the dispersion of catalysts, provide the adsorption centers, and store the products. The S⁰, as the main product, was mainly produced in the micropores.

3.2. SO₂. Loading metal oxides on biochar can also improve the adsorption capacity of SO₂ through an increase in surface adsorption sites and catalytic centers. Tseng et al.¹³⁴ used Cu(NO₃)₂·3H₂O solution to prepare a coconut shell biochar loaded with CuO to adsorb SO₂. Results showed that when the copper load was 3 wt%, the SO₂ adsorption capacity of biochar was 25 mg/g. In the adsorption process, when O₂ was present, SO₂ was catalyzed on the copper phase to produce SO₃, and then it reacted with CuO to produce CuSO₄. In addition, SO₂ could also react directly with CuO to produce CuSO₄. The reaction mechanism is as follows:¹³⁴



Fan et al.¹³⁵ loaded Fe₂O₃, MnO₂ and pyrolusite onto the walnut shell biochar, respectively, and prepared Fe₂O₃, MnO₂, and Fe₂O₃–MnO₂ loaded biochars to adsorb SO₂. Results showed that the sulfur capacity of the biochar loaded by metal oxides was significantly increased compared to that of the original biochar (123.8 mg/g). When the adsorption temperature was 80 °C, the SO₂ adsorption capacities of the three biochars were 140.6 mg/g, 157.8 mg/g, and 227.8 mg/g, respectively. They observed that Fe₂O₃ loaded biochar had a larger specific surface area and pore volume (515.5 m²/g to 641.1 m²/g, 0.37 cm³/g to 0.43 cm³/g), while MnO₂ loaded biochar was more conducive to the formation of functional groups (such as C–OH, C–O, C=O); thus the MnO₂ loaded biochar had a stronger catalytic effect on SO₂. Compared with single metal oxide loading, Mn and Fe in pyrolusite were more dispersed, and the pore structure development of biochar was more favorable (687 m²/g and 0.45 cm³/g) due to the synergistic effect of the metal mixture in pyrolusite, which resulted in higher catalytic activity for SO₂. In the adsorption process, SO₂ was first oxidized into SO₃, and then it was further converted into H₂SO₄. The related reactions are as follows:¹³⁵



Chen et al.¹¹³ used MgCl₂·6H₂O as a modifying agent to prepare straw biochar loaded with MgO by a coprecipitation method to remove SO₂. Research indicated that, compared with the sulfur capacity of the original biochar (11.1 mg/g), the SO₂ adsorption capacity of the MgO-loaded biochar was up to 194.6 mg/g. It was found that after modification by MgCl₂·6H₂O, the high sulfur capacity of the biochar was attributed to the increases of active sites (abundant Mg–O active sites were formed), and larger specific surface area and pore volume (from 10.6 m²/g to 138.1 m²/g and 0.03 cm³/g to 0.4 cm³/g, respectively). Sumathi et al.¹³⁶ compared the SO₂ adsorption capacities of palm shell biochar loaded with

different metal oxides (NiO , Fe_2O_3 , CeO_2 , V_2O_5). Results showed that the specific surface area and pore volume of biochar were all reduced after the modification of different metal oxides, but the SO_2 adsorption capacities were all increased. The adsorption capacity of SO_2 is based on the following order: $\text{CeO}_2 > \text{V}_2\text{O}_5 > \text{NiO} > \text{Fe}_2\text{O}_3$. The high SO_2 adsorption capacity of palm shell biochar loaded with CeO_2 was attributed to the stronger oxidation ability and higher oxygen storage of CeO_2 .

4. DOPING NITROGEN MODIFICATION

Nitrogen doping modification for biochar has attracted extensive attention because of the several advantages such as rich containing-nitrogen functional groups and developed porous structure.^{22,137,138} The doped nitrogen atoms can replace the adjacent carbon atoms in porous carbon to improve the electronegativity, which can raise the electron transfer in carbon skeleton and enhance the electrostatic dipole–dipole effect, thereby resulting in better adsorption and catalytic activity.^{22,139,140} Besides, nitrogen doping modification can also improve the alkalinity and hydrophobic degree of the biochar surface,^{137,139,141,142} thereby resulting in better adsorption effects for SO_2 and H_2S . Common nitrogen doping reagents mainly include urea, ammonia, melamine, and so on. The common nitrogen doping methods mainly include endogenous nitrogen doping and exogenous nitrogen doping. Related studies^{143–145} had showed that using different nitrogen sources, doping methods, and process parameters to modify biochar would lead to different pore structures, nitrogen contents, and containing-nitrogen functional group types on the carbon surface, which will result in different adsorption performances of biochar for sulfur-containing gaseous pollutants. This section mainly introduces the recent research progress on the adsorption of sulfur-containing gaseous pollutants by nitrogen-doped biochar. Figure 5 shows the

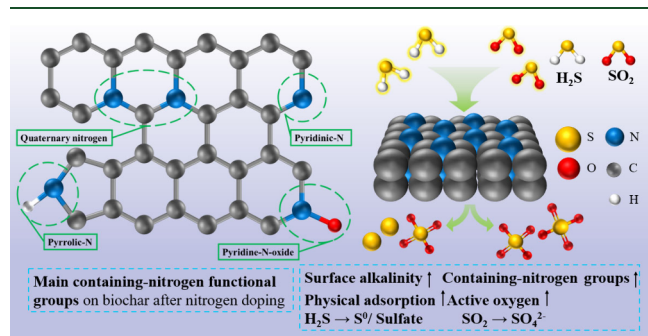


Figure 5. Adsorption mechanism of H_2S and SO_2 on N-doped biochar.

adsorption mechanisms of H_2S and SO_2 by biochar after nitrogen loading. Table 3 shows a summary of pore parameters, adsorption properties, and mechanisms of the nitrogen-doped biochar for adsorption of H_2S and SO_2 .

4.1. H_2S . Nitrogen-doped biochar has been shown to be an efficient metal-free catalyst for H_2S removal at room temperature.^{146,147} Nor et al.¹⁴⁴ used urea to modify palm shell biochar with a microwave-assisted activation to remove H_2S . Results showed that the H_2S adsorption capacity of the modified biochar was greatly increased from 61.14 mg/g to 356.94 mg/g, which were mainly attributed to the following aspects: (i) the specific surface area of the modified biochar

was increased, and some new pores were formed in the existing pores; (ii) new containing-nitrogen functional groups were produced on the biochar, which effectively increased the surface alkalinity and Lewis basic sites of the biochar.¹⁴⁸ Hu et al.¹⁴⁵ used $\text{NH}_3\cdot\text{H}_2\text{O}$ and alanine as nitrogen sources to prepare nitrogen-doped starch porous biochar by a hydrothermal carbonization method and then used it to study the adsorption capacity of H_2S . Results indicated that the H_2S adsorption capacity of the alanine-modified biochar was 110.7 mg/g, which was better than that of the $\text{NH}_3\cdot\text{H}_2\text{O}$ -modified biochar (50.2 mg/g), while the original biochar was only 27.13 mg/g. They discovered that $\text{NH}_3\cdot\text{H}_2\text{O}$ could promote the formation of micropores and mesoporous pores, expand the storage space of reaction products, and increase the specific surface area and pore volume of biochar ($1491.5 \text{ m}^2/\text{g}$ to $2084.1 \text{ m}^2/\text{g}$, and $0.72 \text{ cm}^3/\text{g}$ to $1.0 \text{ cm}^3/\text{g}$). However, alanine was more conducive to the formation of containing-nitrogen functional groups, forming pyridine-N and quinone groups on the biochar, which had strong adsorption, dissolution, and oxidation capacity for H_2S . In the desulfurization process, pyridine-N and quinone groups promoted the removal of H_2S . The main product of the desulfurization process was S^0 , and the byproducts were sulfuric acid and sulfonic acid. After nitrogen doping, the contents of S^0 and sulfuric acid were increased by 5.65 and 1.73 times, respectively.

Setiawan et al.¹⁴³ used biogas fermented digestion solution as a sustainable ammonia source to prepare a nitrogen-doped biochar (N2) and compared the H_2S removal ability of the biochar modified with pure NH_3 (N1). Results showed that the adsorption of H_2S on the biochar was significantly improved after doping with nitrogen. The adsorption capacities of N1 and N2 for H_2S were 9.58 and 23.0 mg/g, respectively, while the H_2S adsorption capacity of the original biochar was only 2.54 mg/g. They indicated that the surface of N1 had more containing-nitrogen functional groups, including pyridine-N, pyrrole-N, and graphite-N, among which pyridine-N dominated. The presence of pyridine-N promoted the formation of the O^* species on the biochar, which effectively enhanced the redox reaction, thus promoting the chemical adsorption of H_2S . The improvement of N_2 adsorption capacity was due to the higher utilization rate of micropores so that the diffusion of H_2S in micropores was not hindered by the formation of sulfate. Wu et al.¹⁴⁹ prepared nitrogen-doped poplar sawdust biochar (NBAC) with a high nitrogen content and microchannel structure for H_2S adsorption using melamine cyanuric acid (MCA) as a nitrogen source. Results showed that the adsorption capacity of NBAC for H_2S reached 1827 mg/g, while the biochar without MCA was only 1 mg/g. They found that the prepared biochar had a microchannel structure with layered pores, which was conducive to mass transfer and storage of elemental sulfur, and H_2S could continue to be catalyzed and oxidized on NBAC after multiple layers of sulfur covered the nitrogen sites. They indicated that during the H_2S adsorption process, the ultrahigh nitrogen content (17.2 atom %) on the surface of biochar provided a large number of active sites (such as pyridine N, pyrrole N, etc.) for the catalytic oxidation of H_2S and the formation of sulfur radicals. These active sites effectively enhanced the dissociation of H_2S on the carbon surface, which enabled the rapid oxidation of HS^- into S^0 , thus promoting the removal of H_2S .

Through using urea as a nitrogen source, and sludge and pine sawdust as raw materials, Li et al.¹⁵⁰ prepared a nitrogen-

doped biochar for adsorption of H_2S . Their results showed that the H_2S adsorption capacity of biochar was significantly increased from 72.9 mg/g to 365.5 mg/g after urea modification. The specific surface area of biochar was increased from 155 to 1065 m^2/g , and the pore volume was increased from 0.07 to 0.78 cm^3/g . Moreover, the modified biochar surface is rich in nitrogen-containing functional groups such as pyrrole nitrogen and pyridine nitrogen, which enhanced the conductivity and molecular polarity of biochar and promoted the adsorption of H_2S . It was found that during the adsorption, H_2S was mainly oxidized into S^0 and sulfate.

4.2. SO_2 . As a Lewis acid, SO_2 tends to interact with basic surfaces; that is, the sulfur atom in SO_2 as the electron acceptor can effectively form stable complexes with the bases as the electron donor.¹⁵¹ Relevant studies^{60,152,153} had shown that the alkaline groups on the surface of biochar, such as alkenes, quinones, benzene, etc., were the groups with high affinity for SO_2 , which was conducive to the adsorption of SO_2 . To investigate the effects of containing-nitrogen functional groups on the adsorption of SO_2 , Qu et al.¹⁵⁴ investigated the possible interaction between SO_2 and N species on carbon surface by DFT. Their results showed that nitrogen doping could improve the physical adsorption of SO_2 on carbon surface by increasing the van der Waals force between carbon surface and SO_2 molecule, and the presence of various containing-nitrogen functional groups effectively contributed to electrostatic interactions for adsorption of SO_2 (such as hydrogen bonding and dipole interactions).

Zhang et al.¹⁴² explored the SO_2 adsorption properties of soybean straw biochar modified by high temperature treatment of $\text{CO}_2\text{-NH}_3$. Results showed that after the modification, the specific surface area of biochar was greatly increased from 0.04 m^2/g to 500.21 m^2/g , and the micropore volume was also significantly increased from 0.1 cm^3/g to 0.22 cm^3/g . Besides, the contents of the containing-nitrogen groups such as pyridine-*n*-oxide, quaternary N, and pyridine on the biochar surface were clearly increased. Due to the significant improvement in the physicochemical properties of biochar, when the adsorption temperature was 120 °C, the sulfur capacity was increased from 57 to 145.68 mg/g. It was found that trimers and bridging hydrogen bonds were the main adsorption sites for SO_2 adsorption. With the increase of adsorption temperature, the containing-nitrogen groups as the main adsorption sites could combine with SO_2 to form sulfamide (60 °C), which eventually existed in a more stable form of SO_4^{2-} (120 °C). Zhang et al.¹⁵⁵ used NH_3 to modify soybean straw biochar to remove SO_2 . Results showed that the SO_2 adsorption capacity of the NH_3 -modified biochar was increased from 100 to 175.9 mg/g at 30 °C and that the sulfur capacity was positively correlated with the nitrogen content of biochar. They indicated that during the modification process, due to the decomposition of NH_3 into H, N, NH_3 , amino, and other free radicals, thermal corrosion occurred on the surface of biochar, which formed new narrow micropores (specific surface area and pore volume increased from 0.04 to 371.11 m^2/g and 0.1 to 0.2 cm^3/g , respectively) and introduced containing-nitrogen functional groups on the surface of biochar. The adsorption process was mainly a physical adsorption, and after NH_3 modification, a large number of pyridine and pyrrole appeared on the surface of biochar to promote the removal of SO_2 .

Zhang et al.¹⁵⁶ used methyl diethanolamine (MDEA)-methanol solution to prepare nitrogen-doped corn cob biochar

for SO_2 removal, and they studied the effect of NO on SO_2 removal. Results showed that the nitrogen content of biochar was greatly increased from 0.44% to 9.5% after MDEA impregnation. At 120 °C, the sulfur content of the corn cob biochar was increased from 58 to 216.9 mg/g. They found that at a low concentration of NO, NO and SO_2 combined to form $\text{NO}_2\text{-SO}_3$ intermediates, which promoted the adsorption of SO_2 . However, at high concentrations of NO, NO, and SO_2 competed for the same adsorption sites, thereby hindering the adsorption of SO_2 . The main adsorption sites on the biochar were found to be the N-Q group of quaternary nitrogen, which was transformed to N oxide after the adsorption of SO_2 , while SO_2 was converted to SO_4^{2-} in the presence of H_2O and oxygen. Wang et al.¹⁵⁷ modified sewage sludge biochar with ammonia–water to adsorb SO_2 . Results showed that after ammonia–water modification, the surface containing-nitrogen functional groups of the biochar was increased significantly (0.11% to 3.57%). The doping of nitrogen atoms changed the uniform electrostatic potential distribution and the surface polarity of the biochar. H_2O , pyridine functional groups, and pyrrole functional groups could promote the adsorption of SO_2 on biochar. Pyridine enabled the formation of dipole–dipole interactions and hydrogen bonds between SO_2 and H_2O , while pyrrole enabled the formation of hydrogen bonds between SO_2 and H_2O .

Nitrogen-doped biochar can effectively improve the adsorption of SO_2 by biochar. It is found that the containing-nitrogen functional groups can not only improve the physical adsorption capacity of biochar for SO_2 but also promote the chemical adsorption of biochar for SO_2 . Among them, the enhancement of SO_2 physical adsorption caused by nitrogen incorporation is always related to the increase of basicity, the change of local electron density, the polarity of carbon atoms, and the charge distribution on the carbon surface. At the same time, the excess π -electrons in the containing-oxygen functional group can be transferred to adsorbed oxygen to form superoxide ions, which can oxidize SO_2 into SO_3 and then further produce sulfuric acid.^{142,152,157} In general, nitrogen modification and doping are beneficial for the development of pores, the enhancement of electronic transfer capability, and the increase of the specific surface area and pore volume of the biochar. Besides, through increasing containing-nitrogen functional groups, the surface alkalinity of biochar is also improved, which is conducive to the adsorption and oxidation of SO_2 ($\text{SO}_2 \rightarrow \text{SO}_4^{2-}$). Figure 5 shows the adsorption mechanism of H_2S and SO_2 by biochar after nitrogen modification and doping.

5. MICROWAVE ACTIVATION

Traditional heating can not provide a uniform temperature for samples of different shapes and sizes, and the temperature gradient in the sample will lead to high energy consumption and a long heating time.¹⁷² As a kind of electromagnetic energy, the microwave can overcome the disadvantage of traditional heating from outside to inside, which can achieve uniform and rapid heating of objects without a temperature gradient.^{173–175} Microwave radiation may produce materials with new microstructure and induce some unique chemical reactions that can not be induced by conventional heating.¹⁷⁶ In addition, microwave heating also has the advantages of a shorter heating time, lower energy consumption, selective heating, greener process, etc. Based on the above several advantages, microwave heating has attracted more and more

attention in the field of materials synthesis and catalysis field.^{173–175} Related studies^{85,90,93,174–176} have shown that the use of microwave heating to activate biochar can effectively improve the pore structure of biochar and introduce some new functional groups onto biochar, thus strengthening the adsorption of gaseous pollutants. This section introduces the adsorption performance and mechanisms of sulfur-containing gas pollutants by microwave-activated biochar.

5.1. H₂S. Lin et al.⁸⁵ activated sludge biochar for 10 min at 650 °C by microwave heating and studied the adsorption capacity of the microwave-activated biochar for H₂S. Their results showed that the desulfurization efficiency of the activated biochar was 78.4%. The microwave-activated biochar had a more developed pore structure and higher surface alkalinity, which promoted the adsorption of H₂S. Besides, due to the presence of metal elements on the surface of sludge biochar, these metal species could catalyze H₂S to form metal sulfide and sulfate, thereby increasing the H₂S adsorption capacity of biochar. Cui et al.⁸⁷ studied the effect of microwave-assisted KOH activation on H₂S removal by rice straw biochar (WKRS) and wheat straw biochar (WKWS). Results showed that the sulfur capacity of the microwave-assisted KOH activated biochar was significantly higher than that of original biochar, which was 57.911 and 61.446 mg/g, respectively. The pH value of biochar was increased (WKRS: 9.5 to 9.6; WKWS: 9.9 to 10.3), and the pore structure of biochar was also greatly improved (WKRS: 108 to 1991.96 m²/g, 0.186 to 1.191 cm³/g; WKWS: 154.92 to 877.15 m²/g, 0.217 to 0.527 cm³/g). In addition, the numbers of containing-oxygen functional groups (such as C=O, C–OH, and/or C–O) on the biochar surface were significantly increased, which were involved in the chemisorption process of H₂S, and both S⁰ and sulfate were produced on the biochar surface.

Dou et al.⁸⁶ activated chlorella (CKW) and spirulina (SKW) biochar with microwave-assisted KOH for H₂S adsorption. Results showed that compared with thermal KOH activated biochar, the H₂S adsorption capacity of the two biochars after microwave-assisted KOH activation was enhanced (CKW: 59.1 to 96.12 mg/g; SKW: 42.3 to 69.42 mg/g), and the specific surface area and pore volume of the two biochars significantly increased (CKW: 5 m²/g to 747.22 m²/g, 0.01 cm³/g to 0.44 cm³/g; SKW: 5.65 m²/g to 568.3 m²/g, 0.1 cm³/g to 0.4 cm³/g). Moreover, the contents of surface functional groups (–NH and C–O) were significantly increased, and the contents of C=O and C–H were also slightly increased. They found that the H₂S adsorption process was mainly physical adsorption supplemented by chemical adsorption. When chemical adsorption occurred on the carbon surface, –NH and C–O were involved in the adsorption process of H₂S, and the products were S⁰ and sulfate. Xu et al.⁸⁸ activated the sludge biochar by microwave pyrolysis and CO₂/steam activation, and then studied the H₂S adsorption capacity of the activated biochar. Results showed that the sulfur capacity of the steam-activated sample was higher than that of the CO₂-activated sample, which was 9.149 and 3.451 mg/g, respectively. The specific surface area of the original biochar was only 34.2 m²/g, which was increased to 169 m²/g after microwave steam activation and 42.1 m²/g after microwave CO₂ activation, respectively. They found that there were acidic functional groups (methylene) on the surface of the unactivated samples that were eliminated after microwave activation. The contents of C=O, C–O, and other groups were increased after microwave activation, which

promoted the adsorption of H₂S. In the adsorption process of H₂S, physical adsorption was dominant, while chemical adsorption produced sulfate, sulfone, and sulfide.

Zhu et al.⁹¹ prepared a kind of sludge biochar by a microwave-assisted steam activation method, and then modified it using red mud. They further studied the adsorption capacity of the prepared biochar for H₂S. The results showed that the specific surface area of the biochar after microwave steam activation was 50.98 m²/g compared with 28.5 m²/g of the original biochar, and the H₂S adsorption capacity of biochar was significantly increased from 1.47 to 22.83 mg/g. They found that the number of oxygen-containing functional groups (such as C–O, C=O, and O–C=O, etc.) on the activated biochar surface was increased, which facilitated the adsorption and catalytic oxidation of H₂S. In addition, the iron oxide on the biochar surface reacted with H₂S to form FeS, which was subsequently further oxidized to produce SO₄^{2–}.

5.2. SO₂. Zhang et al.⁹² activated the coir organisms biochar by microwave heating for adsorption of SO₂. Results showed that the SO₂ adsorption capacity of biochar was increased from 10.9 to 16.9 mg/g after microwave activation. They found that after microwave activation, the acidic functional groups on the surface of biochar were gradually decreased from 0.238 to 0.063 mmol/g, and the surface alkalinity of biochar was increased owing to the generation of alkaline functional groups on the surface of biochar, which effectively promoted the adsorption of SO₂ through acid–base interaction. In addition, alkaline functional groups had high electronic conductivity, which thus further enhanced the adsorption of SO₂ on the biochar surface.

Fengrui et al.⁹³ used microwave-assisted oxidant (K₂Cr₂O₇) to activate coir biochar, and then studied the ability of the activated biochar to adsorb SO₂. Research indicated that compared with the biochar activated by microwave-oxidizer (K₂Cr₂O₇–CAC), the biochar activated by direct microwave (CAC) had larger specific surface area and pore volume (983.7 m²/g > 866.6 m²/g, 0.39 cm³/g > 0.28 cm³/g), and a large number of microporous scale structures were found on the surface of biochar. The optimal SO₂ adsorption capacity of K₂Cr₂O₇–CAC was 40.09 mg/g, which was 2.45 times that of CAC. They indicated that the adsorption process of SO₂ was mainly chemical adsorption, and K₂Cr₂O₇ introduced a large number of alcohol and carboxyl groups to the surface of biochar, which greatly promoted the adsorption of SO₂. Table 2 describes the summary of pore parameters, adsorption properties, and mechanisms of the microwave-activated biochars for adsorbing SO₂ and H₂S.

6. MODIFICATION BY OTHER METHODS

In recent years, nonthermal plasma has been widely applied for material surface modification due to its simple process, high modification efficiency, and low secondary pollution.^{23,177,178} Ning et al.¹⁶⁷ used nonthermal plasma to modify walnut shell biochar loaded with Fe₂O₃ for adsorption of H₂S. Research demonstrated that nonthermal plasma treatment significantly improved the H₂S adsorption capacity of biochar. When the treatment time was 10 min and the output voltage was 6.8 kV, the sample maintained 100% H₂S removal efficiency within 390 min, which was much higher than 120 min of the original biochar. The reasons for this enhancement effect are summarized as follows: (i) the specific surface area, pore volume, and micropores of biochar are all increased after nonthermal plasma modification, which is beneficial for the physical adsorption and molecular transfer of H₂S; (ii) the contents of containing-oxygen and containing-nitrogen functional groups such as C=O/O–C–O,

O=C=O, amino groups, etc. on the biochar are all increased after the nonthermal plasma modification, thus enhancing the adsorption of H₂S; (iii) the conversion of chemisorbed oxygen into lattice oxygen after the nonthermal plasma modification effectively promotes the catalytic oxidation removal of H₂S.

Hydroxyl radical ($\cdot\text{OH}$) is recognized as a type of clean and efficient strong oxidant, which has been widely applied in many fields such as wastewater treatment, soil remediation, flue gas treatment and so on.^{179–183} Zhao et al.¹⁶⁸ used UV/H₂O₂ advanced oxidation process to produce $\cdot\text{OH}$, and then used it as a modifier to modify the corn straw biochar to remove H₂S. Results showed that after the modification by UV/H₂O₂ advanced oxidation process, the desulfurization breakthrough time of the modified corn stalk biochar was greatly increased from 2 to 24 min (reaching 90 mg/g), and its H₂S adsorption capacity was better than that of commercial activated carbon and most biochar adsorbents. It was found that the modification by UV/H₂O₂ advanced oxidation process effectively improved the pore structure of biochar, and significantly increased the numbers of containing-oxygen functional groups ($-\text{OH}$, $\text{C}-\text{O}$, $\text{COO}-$) on the surface of biochar, thus greatly promoting the removal of H₂S on the biochar. Elemental sulfur and sulfate are found to be the main adsorption products.

Chen et al.¹⁶⁹ prepared a modified soybean straw biochar using CO₂ as an activator and UV radiation modification for adsorption of SO₂. Results showed that when the UV radiation modification temperature was 100 °C and the modification time was 12 h, the sulfur capacity of the modified biochar was 100.181 mg/g, which was 3.41 times that of the original biochar. They found that UV radiation modification effectively promoted the formation of micropores in the biochar. The specific surface area was increased from 114.22 m²/g to 347.28 m²/g, and the pore volume was increased from 0.12 cm³/g to 0.37 cm³/g. After the UV radiation modification, some lactones and hydroxyl groups on the surface of biochar were oxidized to the carboxyl group. The content of carboxyl group on the surface of biochar was increased from 0.473 mmol/g to 0.539 mmol/g, and the surface alkalinity and nitrogenous basic functional groups (such as pyridine-N and pyrrole-N) were also both increased, which enhanced the adsorption and oxidation capacity of the sample for SO₂. Table 3 describes the summary of pore parameters, adsorption properties, and mechanisms of the biochars modified by other methods for adsorption of SO₂ and H₂S.

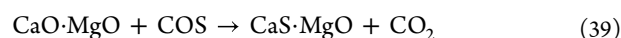
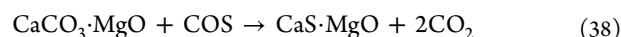
7. COS ADSORPTION BY BIOCHAR

At present, catalytic hydrolysis, as the mainstream COS removal technology, has the advantages of mild reaction, high conversion rate and mature technology.¹⁸⁴ However, the catalytic hydrolysis process will lead to a large amount of sulfur or sulfate deposition on the catalyst surface, resulting in the blockage of pores and the poisoning and deactivation of the catalyst. Besides, the reaction process is long and is easy to corrode the equipment.^{32,185,186} Recently, the adsorption removal technology of COS by biochar has received gradually increasing attention due to its simple and cleaning process. Sakanishi et al.⁹⁴ studied the adsorption and decomposition of H₂S and COS by wood biochar in the temperature range of 300 to 450 °C. Study showed that although the specific surface area of biochar was smaller than the commercial activated carbon (473.61 m²/g > 1059.77 m²/g), the adsorption capacity was higher (0.234 cm³/g > 0.039 cm³/g). This was because the residual metal in biochar could effectively promote the removal of COS, and the COS will form H₂S and CO₂ in the pores. Compared with the chemisorption of H₂S on biochar, the adsorption of COS on biochar was mainly controlled by physical adsorption, most of COS was adsorbed by biochar in the pores, and only a small part of COS was decomposed into CO₂. Sattler et al.¹⁸⁷ compared the adsorption capacity of three carbon-based materials for COS, in which the biochar prepared

by coconut husk and coal was named VPR. They studied the effects of H₂O and H₂S on COS adsorption. Research indicated that the sulfur capacity of VPR was 1.8 mg/g at a 17% relative humidity. With the increase of H₂S concentration and relative humidity, the adsorption capacity of COS by the biochar was decreased because H₂O/H₂S and COS could compete the adsorption sites on the carbon surface.

The comparative study of different activators has a certain reference value for the rational selection of activators. Li et al.⁷⁰ activated walnut shell biochar with different activators (H₃PO₄, KOH, ZnCl₂, K₂CO₃) for simultaneous removal of H₂S, COS, and CS₂ from yellow phosphorus tail gas and closed calcium carbide furnace tail gas. Results showed that KOH-activated biochar had the best desulfurization effect. When the mass ratio of KOH to biochar was 0.5, the sulfur capacity of biochar was increased from 2.5 to 52.67 mg/g. This was because KOH could react with carbon materials to form a large number of micropores at high temperature, which greatly raised the specific surface area and pore volume of biochar (from 1.28 m²/g to 484.26 m²/g and 0.005 cm³/g to 0.272 cm³/g, respectively). Moreover, KOH activation provided a large number of hydroxyl functional groups and increased the alkaline adsorption sites on the biochar. The adsorption products were mainly S⁰ and SO₄²⁻. It was also found that increasing the activation temperature and the amount of KOH increased the specific surface area (1239.92 m²/g) but also led to the transformation from micropores to mesoporous pores, thereby resulting in a decrease in COS adsorption capacity (52.67 to 45.25 mg/g). The micropores with 0.3–1.8 nm was found to play an important role in the COS adsorption process.

In addition to terrestrial biochar, the adsorption performance of seaweed biochar for COS was also been tested. Hanaoka et al.¹⁸⁸ prepared a kind of seaweed biochar for COS adsorption by gasification under 900 °C and CO₂/O₂ atmosphere. Their results showed that the prepared seaweed biochar had a specific surface area of 71 m²/g and a pore volume of 16.3 cm³/g. At 450 °C, the biochar had a good simultaneous removal capacity of H₂S and COS, and nearly 99.5% of the COS was removed by the seaweed biochar. This was because the chemisorbed H₂S promoted the decomposition of COS. At the same time, the metal oxides on the surface of biochar could promote the chemisorption of COS, and COS could react with the metal oxides in biochar to form the (Ca and Mg) crystalline phase. The reaction processes are as follows:¹⁸⁸



Sun et al.¹⁷⁰ prepared tobacco biochar loaded with CuO and Fe₂O₃ by a solution–gel method to simultaneously remove H₂S, COS, and CS₂. Research showed that when the temperature was 60 °C, the humidity was 49%, the oxygen content was 0.5%, and the sulfur capacity reached 231.28 mg/g. It was found that low humidity was conducive to the adsorption of COS on biochar, while high humidity would form a water film on the surface of the biochar to hinder the diffusion of COS in the pores. In the adsorption process, the existence of appropriate oxygen and C=O, $-\text{OH}$ groups on the surface of biochar could enhance the hydrolysis and oxidation process of COS, and metal oxides loaded on the biochar were also conducive to active adsorption of COS (COS + CuO +

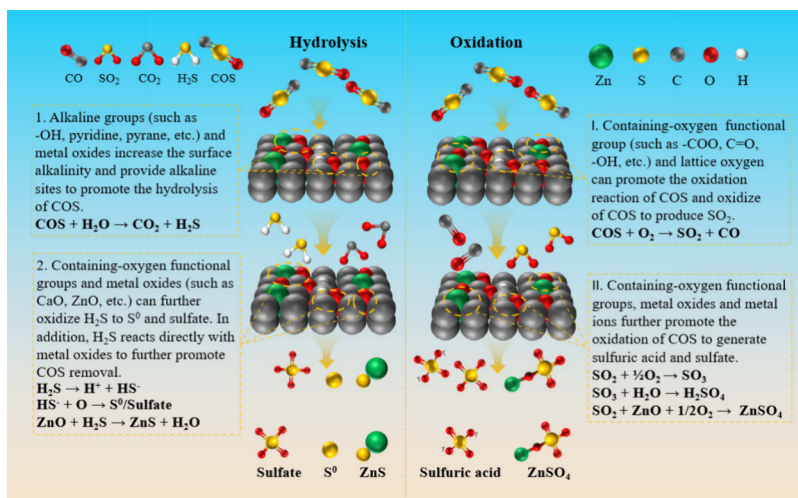


Figure 6. Chemical mechanism of COS removal on biochar.

$\text{H}_2\text{O} \rightarrow \text{CuO} \cdot \text{H}_2\text{S} + \text{CO}_2$). The main adsorption products were the thiol/thioether, S^0 , and sulfate.

Wang et al.⁹⁵ used KOH impregnation to activate coconut shell biochar to study the adsorption capacity of the modified biochar for COS. Their results showed that when the KOH mass fraction was 10%, the COS adsorption capacity of biochar was increased from 5.81 mg/g to 40.64 mg/g. This is due to the increase of the -OH functional group on the surface of biochar after KOH impregnation, which increases the surface alkalinity of biochar and promotes the hydrolysis and catalytic oxidation process of COS. In the adsorption process, COS can react with the -OH functional group to form H_2S (the produced H_2S can be further oxidized into elemental sulfur and sulfate), and it can also be directly oxidized into sulfate. Li et al.¹⁷¹ used an equal-volume impregnation method to prepare two kinds of modified biochars from coconut shell biochar through using AgNO_3 and $\text{Cu}(\text{NO}_3)_2$ as two types of modification reagents, and then further studied their COS adsorption capacities on the prepared two kinds of modified biochars (i.e., Ag/AC and Cu/AC). Their results showed that the maximum COS adsorption capacity of Cu/AC was 14.8 mg/g when the Cu load was 5%, which was better than that of Ag/AC (nearly 10.0 mg/g) and raw biochar (7.5 mg/g). It was found that after modification, the surface of biochar was loaded with metals and metal oxides, among which the surface of biochar after AgNO_3 treatment was loaded with Ag, while the surface of biochar after $\text{Cu}(\text{NO}_3)_2$ treatment was loaded with Cu_2O . Therefore, Cu/AC achieved a stronger COS adsorption activity than Ag/AC. The containing-oxygen functional groups (such as hydroxyl, carboxyl, etc.) and metal oxides all participated in the adsorption process of COS, and the adsorption product was mainly sulfate.

Different modification methods usually have different effects on the physical and chemical properties of biochar.⁹⁶ Ruan et al.⁹⁶ compared the effects of chemical activation and physical activation on the simultaneous removal of H_2S , COS, and CS_2 on tobacco stem biochar. They used KOH, H_3PO_4 , ZnCl_2 , and K_2CO_3 to chemically activate the biochar, respectively. Results showed that the adsorption capacity of the biochar activated by KOH was the highest (45.25 mg/g), while the adsorption capacities of the biochar activated by the other three activators were 10.03, 12.09, and 20.35 mg/g, respectively. This was because the specific surface area and pore volume of the

biochar after activation by KOH were both the largest, reaching 1639.8 m^2/g and 0.78 cm^3/g , respectively. The excellent pore structure effectively promoted COS removal. After the KOH activation, the numbers of C-O, -OH, and other functional groups on the biochar surface were increased, which was conducive to the simultaneous removal of H_2S , COS, and CS_2 . In addition, they used H_2O and CO_2 to physically activate the biochar, respectively. Results showed that the adsorption capacity of the biochar activated by CO_2 was 184.68 mg/g, which was far higher than that of the biochar activated by H_2O (98.32 mg/g). Compared with the H_2O -activated biochar, the CO_2 -activated biochar had a larger specific surface area, more C-O, C=O groups, and alkaline groups, thus promoting the hydrolysis of COS. During the adsorption process, in addition to the physical adsorption of COS, COS was also hydrolyzed to produce H_2S , and then H_2S was further oxidized into S^0 and sulfate. Ruan et al.⁹⁶ studied the simultaneous removal of H_2S , COS, and CS_2 by tobacco stalk biochar modified with different metal nitrates ($\text{Fe}(\text{NO}_3)_3$, $\text{Cu}(\text{NO}_3)_2$, $\text{Co}(\text{NO}_3)_2$, $\text{Ni}(\text{NO}_3)_2$, $\text{Zn}(\text{NO}_3)_2$). Research showed that the adsorption capacity of the biochar modified by $\text{Cu}(\text{NO}_3)_2$ was the strongest, and the adsorption capacity reached 161.93 mg/g when the CuO load was 10%. Then, based on the biochar loaded with CuO, the authors further studied the adsorption capacity of the biochar loaded with two types of metal oxides. Their results showed that the desulfurization capacity of the biochar modified with Cu and Fe was the strongest, up to 231.28 mg/g. They found that the metal oxides promoted the hydrolysis and catalytic oxidation of COS. During the adsorption process, CuO was reduced into Cu_2O , and then COS was finally oxidized into S^0 and metal sulfate. Table 2 and Table 3 further describe the summary of pore parameters, adsorption properties, and mechanisms of the biochars for adsorption of COS.

Based on the above review and analysis, it can be found that the removal mechanism of COS on biochar mainly involves hydrolysis and catalytic oxidation.^{95,186,189} In the hydrolysis process, alkaline groups (such as -OH, pyridine, pyranes, etc.) and metal oxides increase the surface alkalinity and promote the hydrolysis of COS to produce H_2S and CO_2 .^{32,185,190} The generated H_2S is further oxidized into elemental sulfur and sulfate by catalytic oxidation of containing-oxygen functional groups and lattice oxygen on biochar.^{96,170} In addition, H_2S

can react with metal oxides to form metal sulfide.^{188,189} In the catalytic oxidation process, various containing-oxygen functional groups (such as $-\text{OH}$, $\text{C}-\text{O}$, $\text{C}=\text{O}$, etc.), lattice oxygen, and chemisorbed oxygen can promote the oxidation reactions of COS to produce SO_2 and CO .^{95,96,191} After this, the resulting SO_2 can be further oxidized into sulfuric acid and sulfate.^{95,96,171} The above mechanism of COS can also be more intuitively described in Figure 6.

8. CHALLENGES AND PERSPECTIVES

Based on the above analysis and comments, the main challenges and perspectives of this review are summarized as follows:

- (1) Acid and alkali activation can simultaneously improve the pore structure and increase the surface active sites of biochar, making it one of the most widely used methods for biochar activation. However, high temperature and high energy consumption, reagent leakage, and strong corrosion are unavoidable challenges for this technology. Thus, the development of the new modification methods that possess milder operating conditions, lower energy consumption, and a cleaner process is the key direction of development in this field. Advanced oxidation technologies have received more and more attention because they can generate hydroxyl radicals, sulfate radicals, and superoxide radicals with extremely strong oxidizing properties. These free radicals are environmentally friendly and may be applied for modifying biochar to raise the functional groups and active sites and improve the pore structure of biochar. This is likely to become a promising development direction because a large number of advanced oxidation processes have been developed in the fields of water treatment and flue gas treatment. Microwave activation have received increasing attention due to their uniform and rapid heating method, as well as the ability to generate unique physical and chemical changes on the surface and inside of biochar. However, microwave activation can lead to the removal of active functional groups and active sites on the surface of biochar due to extreme conditions. Therefore, after microwave activation, the combined use of other chemical modification methods, especially clean advanced oxidation processes, may be a worthwhile approach to explore.
- (2) Transition metal oxides (e.g., Cu, Zn, Mn, Fe, Ce, etc.) have been proven to be a promising class of sulfur-containing gaseous adsorption and catalytic materials owing to good affinity for sulfur. Single metal modification leads to limited improvement in adsorption capacity, while the combined modification of multiple metals often can produce modification effects beyond expectations due to the synergistic effect between multiple metals. However, current research in this field is still insufficient, especially in the synergistic modification between transition metal oxides and various rare earth elements (utilizing their superior oxygen storage capacity of rare-earth metal oxides), which deserves more research. Besides, excessive metal loading onto biochar can lead to deterioration of the pore structure of adsorbent and a decrease in its activity, as well as waste of metal active components. Single atom catalysts have gained great attention in the field of

catalysis and adsorption due to their relatively low cost, high catalytic activity, and good selectivity, which can realize high catalytic activity, and simultaneously alleviate deterioration of adsorbent and reduce reagent consumption. Therefore, the development of various single metal and multimetal single atom catalysts and the hybrids with biochar is worth exploring.

- (3) Doping nitrogen into biochar can raise the adsorption capacities of H_2S and SO_2 and improve the adsorption selectivity of biochar for these gas molecules through increasing the containing-nitrogen functional groups on biochar. However, most of the nitrogen doping methods currently studied in this field are traditional high-temperature heat treatment methods, which have similar drawbacks to acid and alkali activation processes. Other some modification methods such as plasma discharge, ultraviolet radiation, microwave radiation, and so on may be potential alternatives because they possess relatively lower energy consumption or cleaner process. Besides, codoping of oxygen, phosphorus, sulfur, and/or nitrogen may have good developing potential since they have been shown to achieve faster electron transfer, more alkaline carbon surfaces, and stronger affinity for acidic substances in many other fields, which is worth exploration.
- (4) Existing research on sulfur-containing gaseous pollutants adsorption by biochar mainly focuses on the study of simple experiments and characterization analysis. There is not enough research on the adsorption mechanism, especially using various in situ analysis methods and quantum chemical calculation methods, which leads to insufficient in-depth mechanism revelation. Research on the use of biochar to adsorb COS is relatively scarce. At present, a large amount of biochar-derived porous carbons and modified porous carbons have been developed in many other gas adsorption fields, and thus it is necessary to conduct research and evaluation in the field of COS removal. In addition, it is necessary to study the simultaneous removal of several sulfur-containing gaseous pollutants due to their coexistence in many industrial gas streams, which may greatly reduce the costs of removal system. The activation and regeneration of biochar after adsorption are crucial for reducing adsorption costs and the post-treatment cost of waste adsorbent. However, current research in this field is still not systematic and in-depth enough, and more research should be invested in.
- (5) Adsorption of H_2S on biochar can be divided into physical and chemical adsorption. The large surface area and pore volume of biochar can promote the physical adsorption of H_2S by van der Waals forces. The oxygen-containing functional groups on the biochar surface can promote the catalytic oxidation of H_2S to produce S^0 and sulfate, and the nitrogen-containing functional groups on biochar surface can promote the dissociation of H_2S to produce HS^- and H^+ , which are then further oxidized to S^0 and sulfate. The metal oxides supported on biochar surface can react with H_2S to produce metal sulfide. Adsorption of SO_2 on biochar can be also divided into physical and chemical adsorption. During chemisorption of SO_2 , the oxygen-containing functional groups and chemisorbed oxygen on biochar surface can provide oxygen composition for oxidation of SO_2 .

Addition of nitrogen-containing functional groups (such as pyridine, pyrrole, etc.) on the biochar surface can raise surface alkalinity of biochar, change surface charge distribution and polarity, which can make SO_2 more easily oxidized to SO_4^{2-} , promoting adsorption of SO_2 . The metal oxides on the biochar surface can react with SO_2 to form metal sulfate. In addition to physical adsorption, COS adsorption on biochar surface can be divided into two processes: hydrolysis and catalytic oxidation. In the hydrolysis process, basic groups (such as $-\text{OH}$, pyridine, pyranes, etc.) and various metal oxides can increase surface alkalinity of biochar and promote hydrolysis of COS to H_2S and CO_2 . The producing H_2S is further oxidized to S^0 and sulfate by catalytic oxidation. In the catalytic oxidation process, various oxygen-containing functional groups, lattice oxygen, and chemisorbed oxygen can promote COS oxidation to produce SO_2 and CO . Subsequently, the producing SO_2 is further oxidized to sulfuric acid and sulfate.

9. CONCLUSION

Compared with other removal technologies, adsorption removal of sulfur-containing gaseous pollutants by biochar possesses a good development prospect owing to several advantages such as a simple and clean process, renewability of adsorbent, and cheap raw materials. However, scarce active functional groups and active sites on biochar and underdeveloped pore structure limit the development and application of biochar adsorption removal technology in this field. Activation and modification of biochar are one of the most effective ways to raise biochar surface active sites and active functional groups, and improve the pore structure of biochar. Common activation and modification methods mainly include acid/alkali activation, microwave activation, metal oxide modification, nitrogen doping modification, and some emerging modification methods. Among these activation and modification methods, photochemical modification technology has the advantages of a simple and clean process and low energy consumption, and especially it can simultaneously introduce oxygen-containing functional groups on the surface of carbon materials and improve the pore structure of carbon materials, showing superior development prospects.

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