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A review: preparation of sludge derived carbons and their performance in wastewater treatment

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ABSTRACT

The tremendous pressure of annually increasing amount of sewage sludge producing from wastewater treatment plants and the imposition of more stringent legislation on its disposal and utilization prompt a drive for alternative uses of sewage sludge. Considering abundant energy embedded in sewage sludge, pyrolysis for producing biochar is deemed to be an especially promising method. In this review, preparation methods of sludge derived carbons (SDCs) including pretreatment, pyrolysis temperature and modification, coupled with its application as adsorbents and catalysts in wastewater treatment are discussed. Due to the excellent properties of well-developed porosity and various functional groups on the surface of SDCs, they have high adsorption capacities of heavy metals and organic pollutants. Besides, the metals indigenously existed in sludge are recognized as active components, which makes the SDCs excellent catalysts or catalyst supports in advanced oxidation processes. In order to sufficiently explore the sludge-based biochar, it is recommended that further studies should involve its application in practical wastewater and its environmentally-safe use as well.

Keywords: Sludge derived carbons; Wastewater treatment; Organic pollutants; Heavy metals; Adsorption; Catalytic degradation

1. Introduction

The operation of municipal wastewater treatment plants (MWTPs) is accompanied by the production of large amounts of waste activated sludge which usually contains hazardous organic micro-pollutants, pathogens, herbicides, heavy metals (HMs) and other harmful substances [1]. Thus, without proper disposal, it could cause secondary pollution to the environment and threaten human health. By the end of 2016, the amount of sewage entered MWTPs in China had reached c.a. 150 million m^3/d, inducing that 30 million tons of sludge are generated when considering moisture content of 80%. It is expected that the amount of sludge in China will exceed 60 million tons in 2020, which is ascribed to the increased amount of sewage and the enactment of more strict legislation on sewage treatment. On the basis of current rapid urbanization and continuous growth of economics, it is inferred that the increased rate of prediction on the amount of sludge over a 10 y period steadily retains at a high value of c.a. 10% per year. Thus, this issue is bound to be a disturbing waste problem all around the world [2–4].

The conventional methods for sludge disposal include sanitary landfill, land application, incineration and ocean dumping, which can lead to secondary pollution and a waste of resources. Nevertheless, sludge is recognized as a bio-resource for the reason of being rich in organic substances. Therefore, it is vital to develop and impel a range

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of technologies with economic feasibility and environmental friendship for the purpose of recycling the resource and energy embedded in sludge while eliminating the pollutants. In the 1980s, sludge pyrolysis was proposed as a new process of sludge treatment. The sludge is heated in the absence of oxygen or hypoxia to produce gaseous, liquid and solid residues. Combustible gases such as CO, CH_4, C_2H_6 and C_3H_8 are included in the gas phase, while tar substances and carbonaceous species with high calorific value mainly exist in the liquid and solid phases, respectively. In comparison with traditional disposals, pyrolysis can significantly minimize the volume of sewage sludge, kill parasites and pathogens, fix carbon element and immobilize HMs in the sludge-based biochar [5,6]. Therefore, pyrolysis is considered an eco-friendly treatment of sewage sludge and a solution in the circular economy, greenhouse gas reduction, soil fertilization and improvement of water retention [7].

Sewage sludge has shown great potential as a feedstock, with low costs and yields comparable to those common crops [8]. Unlike the agriculture wastes, sewage sludge is composed of both organic and inorganic matters, including different chemicals, mineral salts, bacteria and viruses [9,10]. Accordingly, the sludge-based biochar possesses a large proportion of ash and inorganic matters. For example, diverse cationic metal ions (e.g., K^+, Na^+, Mg^{2+} and Ca^{2+}) are included in these biochars, which enables them to remove HMs from wastewater via ion exchange besides adsorption effect [11]. Sludge derived carbons (SDCs) are versatile owing to the effects of surface functional groups, the existence of microporous and mesoporous structure, as well as the presence of metal ions. Therefore, subsequent to the studies on SDCs as adsorbents of organic contaminants and HMs, a wealth of research has been conducted on its role as catalysts or catalyst supports. This paper seeks to review the production of SDCs, including the pretreatments, carbonization temperature and modification. In addition, this paper illustrates the application of these biochars in wastewater treatment with an emphasis on their utilization as catalysts which has nearly not been summarized by now. Based on the merits of sludge pyrolysis, such as low cost and excellent properties of the biochar products, there would be more meaningful work to be conducted on this scientific subject.

2. Preparation methods

2.1. Sludge dewatering pretreatments

In MWTPs, sludge dewatering is an essential procedure for reducing the sludge volume and subsequent transport/disposal costs [12]. During the preparation of SDCs, the dehydration and drying of sewage sludge should be carried out first before being pyrolyzed, which can enhance the efficiency of the subsequent carbonization and lower the cost as well. The pretreatment methods, such as chemical conditioning, thermal hydrolysis and advanced oxidation process, allow the degradation of sludge gel structure, partial solubilization of sludge and release of linked water [13].

With regard to traditional methods, inorganic flocculants (e.g., $Al_2(SO_4)_3$, $Fe_2(SO_4)_3$, $AlCl_3$ and $FeCl_3$), inorganic polymer flocculants (e.g., polyaluminum chloride), or organic polymer flocculants (e.g., polyacrylamide) is added into sludge, and then surface adsorption water along with internal hydration water is converted into free-water which can be easily removed by mechanical dewatering. However, by the general method, the moisture content of sludge only gets decreased to the level of approximately 80% [14]. Therefore, the consumption of much energy cannot be avoided during the production of SDCs for the purpose of driving the water out of the solid phase. Wang et al. [15] utilized the dewatered sludge with moisture of c.a. 80% to prepare biochar according to the procedure of drying, grounding, mixing with sesbania powder, and pyrolyzing. This sludge carbon was applied for the degradation of *m*-cresol in catalytic wet peroxide oxidation (CWPO), and its conversion rate reached 96% when the total organic carbon (TOC) removal rate attained 47%. Supposing that the extra energy input is not considered, the catalytic efficiency of SDC is rather high. However, the homogenous catalysis contributing to the degradation was overlooked since the leaching of HMs in SDCs was not discussed. The leaching issue not only affects the stability of catalyst but also is related to an evaluation of its environment-friendly application.

Recently, Wang et al. [4] obtained higher quality biochar from sludge by combining hydrothermal pretreatment with the subsequent pyrolysis (HTP) process (>500°C) [4]. Compared with the traditional method, this new coupling method lowered the toxicity of SDCs and their environmental risk by confining the HMs in the biochar matrix. Hydrothermal pretreatment has been used for enhancing the sludge dewatering or anaerobic fermentation properties [16]. But for the purpose of producing SDCs, the gas inflated into the hydrothermal reaction was inert gas rather than air or oxygen. The dewatered sludge and some deionized water were placed into a reactor at 220°C with an atmosphere of 99.99% pure argon. After HTP, the HMs leaching including Cu, Zn, Cr, Ni and Cd, decreased by 20%–40%, resulting in no risk presented by Cr, Pb and Cu and lower risk of Cd, Zn and Ni in sludge carbon. Some interactions, such as adsorption, precipitation and complexation, between HMs and crystal lattices of the sludge might occur, especially at the temperature higher than 200°C during hydrothermal treatment [17,18]. Therefore, the sludge being pretreated by hydrothermal technology is likely to be a promising precursor for SDCs with high quality and less toxicity. However, it should be noted that hydrothermal pretreatment requires a high-pressure reactor.

As a matter of fact, the usual dewatering method is difficult to complete deep dehydration which is significant and necessary for the sludge treatment [14]. Despite the high efficiency of the thermal dewatering method, high energy consumption was accompanied, which limits its application. Among the deep dewatering approaches, advanced oxidation is one of the most promising methods due to its low environmental impact and high efficiency [19]. In recent years, zero-valent iron (ZVI) combined with persulfate has been proved to be effective in improving the sludge dewaterability [20,21]. In the ZVI-persulfate system, ZVI can effectively activate persulfate ($S_2O_8^{2-}$) to produce sulfate radical anion ($^{\bullet}SO_4^-$), a species with a strong oxidation capacity [22]. During this process, the structure of sludge is efficiently destroyed and then bound water is converted into free water. However, the sludge residue which is

introduced with ZVI might intensify the threat on the ecosystem and human health, so post-treatment is necessary to separate iron species from dewatered sludge. Chen et al. [23] treated the ZVI-persulfate dewatered sludge through preparing magnetic biochar which was stated to be an excellent biosorbent to remove HMs from wastewater. And an excellent adsorption property of Pb^{2+} was found by this biochar in lead-battery wastewater. Interestingly, the iron content in ZVI-persulfate sludge carbon was 40.97% while only 4.82% iron existed in the sludge carbon by conventional dewatering methods, so this sustainable biochar showed strong magnetic ability which was beneficial for recycling.

After being dewatered, the sludge still presents a low calorific value of approximately 145 kcal/kg. In China, sludge of about 1,100 kcal/kg is free for incineration and sludge of more than 2,000 kcal/kg would be sold at 7.5–22.5 $/t. Comparatively, it generally costs 22.5 $/t for treating sludge with a calorific value of 70 kcal/kg. Liu et al. [24] confirmed that sawdust conditioning for sludge deep dewatering could make a great profit (79.2 $/t sludge with the moisture of 80%) from energy recovery [24]. The water content of sludge should be better decreased to a value below 15% for the disposal of pyrolysis. During thermal treatment, the required drying energy (Q) is closely related to the moisture content of dewatered sludge (Y_{dew}) and dried sludge (Y_{dry}). For instance, when Y_{dew} is 60%, Q is in the range of 1,233–3,273 kJ/kg dry sludge; when Y_{dew} is 80%, the value of Q is between 7,664 and 9,757 kJ/kg dry sludge (the specific data depends on the value of Y_{dry}) [25].

2.2. Pyrolysis

For sludge pyrolysis, most studies are focused on the traditional pyrolysis due to its uncomplex reactors and easy operation. In this pyrolysis, heat is delivered externally to the sludge surface by convection, conduction and radiation in an electric or gas heater, so it takes a long time to complete the transformation of sludge into biochar. In recent years, the microwave irradiation pyrolysis of sludge has attracted much attention owing to its advantages (e.g., enhancing and accelerating chemical reactions) compared to the traditional pyrolysis [26]. The microwave-assisted pyrolysis forms the heat from the inner side of sludge towards the surface, allowing high-efficient heat transfer [27]. Even at a lower temperature than that reported in conventional heating, some feedstock can be pyrolyzed by microwave-assisted pyrolysis [28]. Interestingly, biochar fabricated from the sludge treated by microwave irradiation has an undeveloped porous structure, which makes it a poor adsorbent [29]. However, this less porous biochar is more resistant to the leaching of organic substances and HMs than the sludge derived biochar by conventional pyrolysis [29,30]. In Raček et al. [31], they also found microwave pyrolysis could immobilize most HMs in biochar structure which might allow its use in agriculture. Hence, microwave-assisted pyrolysis is suggested to be a better approach for sludge pyrolysis when considering the safe application of biochar in the environment. Nevertheless, the studies on using microwave irradiation heating for the product of biochar are much less than the liquid and gas products [26,32]. Deep and systematic researches still need to be conducted for the practical use of microwave-induced biochar from sludge.

Apart from the different types of pyrolysis, the pyrolysis temperature is also an important factor in influencing the yield of the sludge-based biochar, the ash content, the component of elements, the surface area and the structure. Higher pyrolysis temperature induces lower biochar yield of SDCs, which is caused by the massive decomposition of organics substances in sludge during the pyrolysis [4]. When the original sources for preparing biochar are different, the yield is various even at the same pyrolysis temperature. The biochar yields greatly depend on the content of inorganic substances (or ash) in the feedstock [33]. Higher pyrolysis temperature induces a higher percentage of ash in sludge carbon owing to the same reason for the trend of its yield. A large number of organics are decomposed, leaving more content of inorganics in the prepared sludge carbon. In Wang et al. [4], the yield of SDCs reduced by approximately 15%, as the temperature increased from 300°C to 700°C [4]. Meanwhile, the value of carbon content was decreased from 17 to 10 wt.%, while the amount of other elements relative with inorganics rose. Mahapatra et al. [34] developed SDCs by using sludge originated from the food processing industry as the feedstock. And they found that the specific surface area Brunauer–Emmett–Teller (S_{BET}) was larger with an increase of pyrolysis temperature at a low level (<300°C) but the micropores were destructed and S_{BET} decreased as the temperature was further improved. Apart from the S_{BET}, the pyrolysis temperature affects the ingredient and acid-base properties of sludge carbon as well. Gu et al. [35] found that among three carbonization temperatures (600°C, 800°C and 1,000°C), the sludge carbon obtained at 600°C had the highest content of Fe_3O_4 which was beneficial for its recycling. Hossain et al. [36] reported that the sludge carbon prepared at the temperature of 300°C–400°C was acidic, while those obtained at higher carbonization temperature was alkaline. Thus, they suggested that the SDCs pyrolyzed at various temperatures could be applied as a soil conditioner to regulate its pH. A low pyrolysis temperature prompts the transform of macromolecules such as proteins into acidic substances [37]. Conversely, during the carbonization at a rather high temperature, the alkali salts in the ash of sludge are deprived of the pyrolytic structure and the collapse of acidic surface functional groups happens when the percentage of oxygen reduces [38,39].

2.3. Surface modification

Normally, the S_{BET} of sludge carbon prepared through the only pyrolysis is much low, the pore structure is underdeveloped and the functional groups are limited [40]. Therefore, efforts are ongoing to substantially improve the potential of sludge carbon surface by suitable treatment methods which enable sludge carbon to enhance its performance in the removal of specific contaminants from the aqueous phase. In general, methods of the pristine SDCs activation/modification can be divided into two categories: (I) physical treatment such as steam and (II) chemical treatment, for example, acid and alkali.

Steam activation utilizes the redox reaction between steam and carbon under high temperature, which is low-cost

and easy-operated. The carbon compounds in edges and defects of graphite microcrystals are much active and easy to be oxidized to volatile gas and escape from the sludge carbon, which facilitates the formation of developed pore structure. The mechanism belongs to water gas reaction: $C + H_2O \rightarrow CO + H_2$, with some presence of water gas shift ($CO + H_2O \rightarrow CO_2 + H_2$) and Boudouard reaction ($C + CO_2 \rightarrow 2CO$) [41].

Using various acids (e.g., HNO_3, H_2SO_4 and HCl) for the modification of SDCs under appropriate conditions renders an increase in the number of functional groups. Fig. 1 displays some common oxygen-containing groups including carboxyl, hydroxyl/phenol, lactone, carboxylic anhydrides, carbonyl, ether, quinone and pyrone-type structure. Xi et al. [42] found that the sludge carbon modified at room temperature with sulfuric acid could sorb 61.3 mg g^{-1} of Cr(VI) while the adsorption amount was only 27.7 mg g^{-1} by the pristine sludge carbon. The reason might be that the modified SDC contains rich oxygen functional groups such as–COOH, –C=O and –OH. However, varying the concentration of acid, modification temperature, time or other parameters may decrease the number of oxygen-containing functional groups. Wang et al. [43] reported that SDCs modified by nitric acid at low temperatures included less C–O and –C=O than those of the original SDCs [43]. The reason might be that some basic functional groups were eliminated by HNO_3 when the ash was rinsed off. Conversely, high temperature and concentration of acid could help to introduce the oxygen-containing groups during the modification process. For instance, carboxyl appeared at 300°C while lactone and carboxylic anhydride presented at 650°C. Therefore, only under suitable conditions can the number and types of oxygen-containing groups be enhanced in SDCs by acid modification. Compared with non-oxidizing acids, oxidizing acids are more frequently applied in the SDCs modification. It could be explained by that the acids oxidization is beneficial for the generation of more functional groups which positively impact catalytic degradation of pollutants. For example, Yu et al. [44] studied the effects of several acids (sulfuric acid, nitric acid, hydrochloric acid, perchloric acid and glacial acetic acid) on the modification of SDCs and found that SDCs treated by H_2SO_4, HNO_3 and $HClO_4$ could convert 100% *m*-cresol to smaller molecules substances while HCl and glacial acetic acid could achieve only less than 5% *m*-cresol conversion rate.

Alkali metal hydroxides such as potassium hydroxide (KOH) and sodium hydroxide have been successfully used as activating agents to improve the adsorption capacity of SDCs through increasing the S_{BET} and the amount of oxygen-containing functional groups. Among all the reagents, KOH can most effectively activate SDCs, allowing a S_{BET} as high as 1,900 m^2/g [45]. The porosities in SDCs prepared from sewage sludge are higher than those made from other carbonaceous precursors under similar experimental conditions. This observation indicates that sewage sludge is a promising feedstock for producing adsorbents. The existence of inorganic fraction in SDCs is one important issue which is accounted for its high adsorption. Monsalvo et al. [46] activated SDCs with KOH allowing a much higher development of porosity (a mesopore volume higher than 0.35 cm^3/g; a micropore volume of 0.75 cm^3/g) and S_{BET} above 1,800 m^2/g. Hwang et al. [47] discussed the best activating condition of alkaline hydroxides including KOH and NaOH. The optimum conditions for KOH-activation were 1 mol/L, 800°C and 1.5 h, whereas those for NaOH-activation were 1.25 mol/L, 850°C and 1.5 h. As for the treatment of the effluent waster of MWTPs, the modified SDCs had the highest removal efficiencies for biochemical oxygen demand (BOD), chemical oxygen demand (COD) and SS of ca.56%, 58% and 82%, respectively. Nunthaprechachan et al. [48] investigated the effects of different types of activating agents (e.g., $ZnCl_2$, HNO_3 and KOH) on the adsorption performance of SDCs. It was found that the SDC prepared by KOH-activation had the highest adsorption capacity which was up to

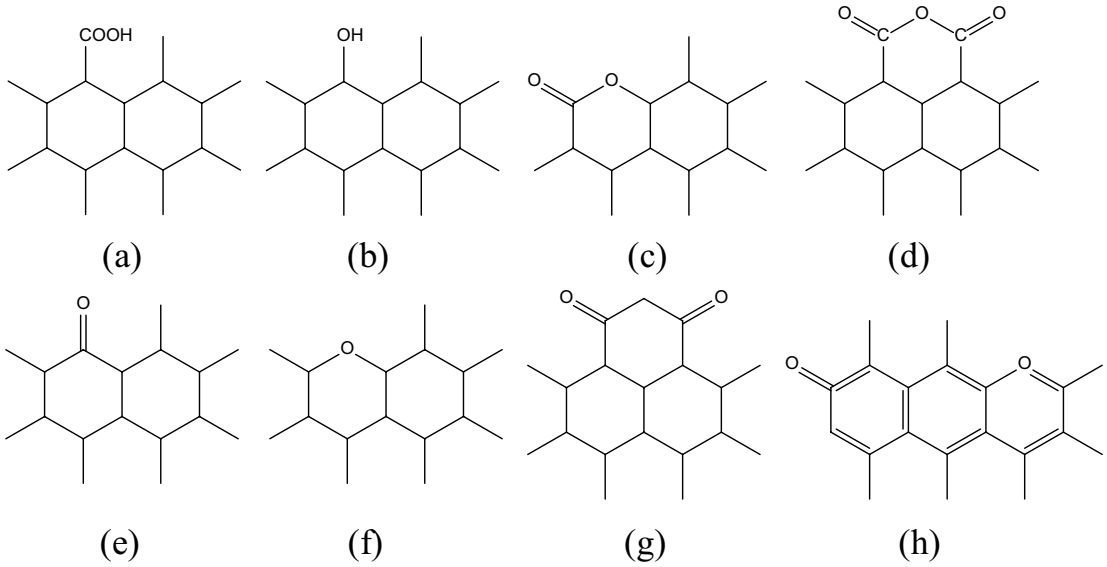

Fig. 1. Surface oxygen groups on carbonaceous materials: (a) carboxyl, (b) hydroxyl/phenol, (c) lactone, (d) carboxylic anhydrides, (e) carbonyl, (f) ether, (g) quinone, and (h) pyrone-type structure.

14.12 mg/g and the corresponding removal of dibenzothiophene (DBT) was around 70.6%. This result was owing to the highest amount of oxygen-containing functional groups, especially the carbonyl group.

3. Application in wastewater treatment

3.1. Adsorption

In the 1990s, it was proposed that sewage sludge might be a potential precursor for the production of activated carbon or adsorbents due to its carbonaceous nature [49]. At the beginning of the 21st century, the researches on SDCs were mainly to obtain the structure with large S_{BET} and high development of porosity which was beneficial for the adsorption of pollutants. Table 1 lists sources, carbonation conditions, characteristics and application of SDCs. Based on the properties of the porous structure, abundant surface functional groups and other unique characteristics, SDCs are confirmed to be an effective adsorbent for organic pollutants and heavy metals [4,52].

In the removal of organics, a typical example is the direct utilization of SDCs to treat the dye wastewater which contains various types of dye compounds refractory to conventional treatments. Methylene blue (MB) is a common type found in the effluents discharged from the printing and dyeing industries [56]. And for exploring the performance of dyes removal from aqueous solution, MB is often selected as a target pollutant [57]. The removal mechanism of MB is summarized and drawn simply in Fig. 2. Fan et al. [50] used SDCs to remove MB from aqueous solution and nearly 100% of the dye was adsorbed within 10 h. The maximum adsorption capacity of SDCs for MB reached 29.85 mg/g, which might be attributed to their coarse and porous texture structure. But the S_{BET} of 25 m^2/g was not higher than other materials shown in Table 1. It could be owing to the binding sites furnished by the abundant functional groups and various ions (e.g.,

Table 1
Preparation, characteristics of sludge derived carbons and their application in the adsorption of organics and heavy metals

Sludge source	Carbonation conditions				S_{BET} (m^2/g)	V_{micro} (cm^3/g)	Adsorption capacities of contaminants (mg/g)	References
	Temperature (°C)	Time (h)	Heating rate (°C/min)	Activation				
Sewage sludge	600	1	10	KOH	629	0.4839	29.0 (MB) 14.12 (DBT)	[48]
Sewage sludge	550	2	10	None	25	0.047	29.85 (MB)	[50]
Sewage sludge	300, 700	2	5	$CaSO_4$	14.27	0.0008	131.9 (MB)	[51]
Sewage sludge	800	1	10	$ZnCl_2$ and lime	631.8	0.5098	287.1 (Allura Red) 640.7 (Crystal Violet)	[52]
Sewage sludge	1,000	1	20	None	96	0.036	182 (Phenol)	[53]
Aerobic granular sludge	750	0.5	10	KOH	1,832	0.75	325 (4-chlorophenol)	[46]
Sewage sludge	650	0.5	40	None	60	0.04	64.9 (Hg^{2+}) 40.3 (Pb^{2+}) 6.7 (Cu^{2+}) 3.0 (Cr^{3+})	[54]
Sewage sludge	650	0.083	5	$ZnCl_2$	472	0.1	175.4 (Hg^{2+}) 64.1 (Pb^{2+}) 30.7 (Cu^{2+}) 15.4 (Cr^{3+})	[54]
Dewatered iron-containing sludge	600	1.5	15	None	No data	No data	49.96 (Pb^{2+}) 36.75 (Cu^{2+}) 33.91 (Cd^{2+}) 30.1 (Zn^{2+}) 29.5 (Ni^{2+})	[23]
Anaerobic sludge	600	1.5	15	None	No data	No data	47.0 (Pb^{2+}) 22.5 (Cd^{2+}) 21.2 (Cu^{2+}) 18.1 (Zn^{2+}) 16.2 (Ni^{2+}) 8.3 (Cr^{6+})	[11]
Limed sludge	800	1	20	None	63	0.023	277 (Cu^{2+})	[55]

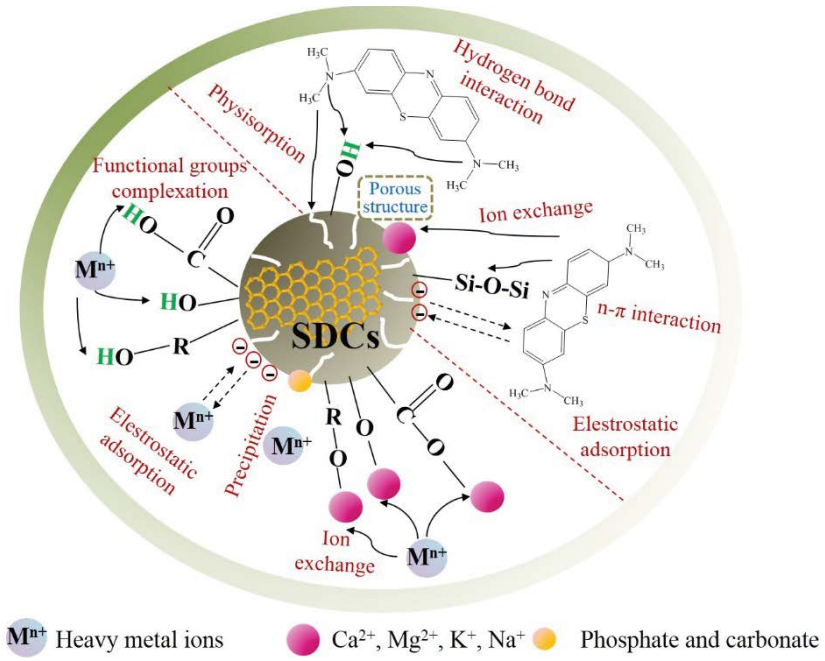

Fig. 2. Adsorption of methylene blue and heavy metals on SDCs with various mechanisms.

Ca^{2+}, Mg^{2+}, Na^{+} and K^{+}) for MB adsorption through the ion exchange, electrostatic interaction, hydrogen bond interaction and n–π interaction which might occur between the aromatic structure of MB and Si–O–Si of SDCs (Fig. 2). However, the mechanisms are varied according to the types of SDCs, contaminants and conditions of aqueous solutions. Liu et al. [51] used calcium as a binder to make granular activated carbon (4 mm in diameter and 9 mm long) from sludge and its adsorption of MB reached 131.9 mg/g which was dominated by physisorption. This adsorption mechanism significantly differed from that in Fan's study [50]. They declared that the MB adsorption process was well described by the pseudo-second-order kinetic model and the MB diffusion in micropores was the potential rete-controlling step. By the analysis of adsorption thermodynamics, the ΔH value of 6.63 kJ/mol indicated MB adsorption onto the SDCs was endothermic and physical sorption (in the range of 2.1–20.9 kJ/mol). Large surface-specific areas of SDCs provide sufficient area for the contact between contaminants and adsorbents. Therefore, researchers incline to prepare SDCs with high S_{BET} by various approaches. For example, Streit et al. [52] developed a high-quality activated carbon by using the feedstock of biological sludge which was produced from a beverage wastewater treatment plant. The novel method was that prior to pyrolysis, the mixture of 250 g sludge, 200 g $ZnCl_2$ and 50 g lime was dried in an oven and then ground to the particles with a diameter lower than 355 μm. This was obviously different from the method of modification/activation after the pyrolysis process, which was mentioned in section "surface modification". Consequently, the acquired SDCs exhibited a much high S_{BET} of 683.1 m^2/g and a well adsorption of Allura Red (287.1 mg/g at pH 2.0) and Crystal Violet (640.7 mg/g at pH 8.0). This novel research suggested that the modification could also proceed before the pyrolysis process. Apart from the dye adsorption, SDCs are also applied as adsorbents for miscellaneous pollutants such as benzoic acid and phenol which is presented in Table 1. For these substances, functional groups rather than the surface-specific areas are responsible for the removal mechanism [53]. Since the phenol molecule (~0.62 nm) is small, phenol adsorbents usually show a high degree of microporosity, a property generally not associated with SDCs. So surface chemical interaction between pollutants like phenol and the functional groups of SDCs can surmount the limitations of an inadequate porosity.

Heavy metals (e.g., lead, copper, cadmium nickel and chromium), toxic and non-biodegradation, can threaten human and ecological health if being discharged to the environment without disposal. Among the methods of removing HMs from wastewater, SDCs have been found to be high-efficient sorbents, whose removing mechanism is displayed in Fig. 2 [4,11,54]. In 2008, Rozada et al. [54] investigated the feasibility of using sludge-based adsorbents for separating mercury, lead, copper and chromium from aqueous solutions. At optimum pH, SDCs activated by $ZnCl_2$ exhibited the adsorption capacities of 175.4, 64.1, 30.7 and 15.4 mg/g for Hg^{2+}, Pb^{2+}, Cu^{2+} and Cr^{3+}, respectively (Table 1). Comparatively, those corresponding to the original SDCs were 64.9, 40.3, 6.7 and 3.0 mg/g, respectively. The higher adsorption capacity by the modified SDCs was attributed to its high S_{BET} (472 m^2/g) and micropore volume (0.1 cm^3/g), whereas the SDCs had rather low S_{BET} of 60 m^2/g and V_{micro} of 0.04 cm^3/g (Table 1). It seems that a higher adsorption capacity was obtained when using the SDCs with larger specific surface areas. But in Rio et al. [55], the SDC carbonized at 800°C could uptake 277 mg/g Cu^{2+} although it had a low BET surface area of 63 m^2/g. Its prominent adsorption capacity was attributed to the high content of calcium in SDCs by the pretreatment with

lime. Therefore, the mechanism for the HMs removal was attributed to ion exchange between Cu^{2+} in wastewater and Ca^{2+} ions in SDCs. The ion exchange mechanism was also confirmed in the research by Seredych and Bandosz [58]. As the pyrolysis temperature improved, the high degree of mineralization was achieved, thus hindering the cation exchange. Seredych and Bandosz prepared SDCs at the temperature of 950°C and found that despite the inhibition of cation exchange capacity, the SDCs still attained higher Cu^{2+} adsorption through the complexation of Cu and its subsequent surface precipitation as hydroxides of hydroxyl carbonate entities [59]. From the perspective of resource and energy utilization, anaerobic digestion of sludge prior to pyrolysis seems to be more appropriate for producing SDCs since the sludge could be used to produce a considerable amount of methane or hydrogen. In 2017, Gu et al. [11] made a research on the efficiency of biochar produced from anaerobic sludge on the removal of HMs and the sorption mechanism. This study indicated that this SDC had better adsorption of HMs especially Pb^{2+} than that of the biochar made directly from sewage sludge. As shown in Table 1, the biochar produced from anaerobic sludge has a competitive adsorption capacity of HMs in comparison with other SDCs.

3.2. Catalytic degradation

In the 2000s, using sludge as a precursor for the production of biochars or activated carbon was studied primarily on the adsorption of organic pollutants and heavy metals [35]. However, with the development of advanced oxidation processes, researches on SDCs as efficient catalysts or catalyst supports in liquid phase reactions appeared and attracted more attention during the last decade [1,15,60]. As is well known, biochars initially prepared from agricultural and forest residues comprise a high proportion of carbon. However, when considering industrial by-products and municipal wastes as the feedstock, the obtained biochars usually have a larger amount of ash and inorganic components [61]. That is a crucial reason for the catalytic performance of these biochars prepared from sewage sludge. It is reported that waste activated sludge is abundant in iron species, about 100 times as that found in commercial activated carbons, resulting in its catalytic characteristics [62]. The relative content of metals in SDCs is improved obviously after carbonization. Therefore, the metals immobilized in SDCs can act as the active constituent of catalysts. In light of the unsatisfactory catalytic performance, some researchers attempted to add extra metals to improve the catalytic characteristic of SDCs. It should be noticed that whether the contaminant removal is caused by adsorption or the catalytic process. Prior to the catalytic study, the adsorption experiment is conducted by adding SDCs materials without oxidizing agents. The adsorption contribution is less than 5% due to the rather low S_{BET} and volume of pores in comparison to the SDCs adsorbents obtained via different methods [15,63]. In previous studies, it was reported that the SDCs have been successfully applied as catalysts or catalyst supports in CWPO, catalytic wet air oxidation (CWAO), catalytic ozone oxidation (CWOO), Fenton, persulfate and other advanced oxidation processes (AOPs) for the disposal of wastewater containing refractory organics [44,60,64,65]. Table 2 presents some SDCs as catalysts or catalyst supports for organic pollutants in various AOPs.

In Fenton-like degradation, the SDCs showed higher catalytic activity in 1-diazo-2-naphthol-4-sulfonic acid (1,2,4-Acid) oxidation than commercial Fe_3O_4 MNPs [35]. Marques et al. [66] investigated for the first time the potential of applying SDCs as catalytic materials in the degradation of phenolic compounds in a CWAO reactor. Since CWAO was conducted at a high temperature of 160°C and under a partial oxygen pressure of 4.2 bar, the conversion of phenol (X_{con}) and TOC removal (X_{TOC}) reached above 93% and 78% with the SDCs catalyst activated by physical and chemical ways when using a rather high concentration of phenol (5 g/L). The active sites, whether present as surface functional groups or active metals (e.g., Fe), are necessary to the decomposition of organic pollutants. Unfortunately, this study didn't give a clear correlation between the phenol degradation process and the active sites. Furthermore, the removal rate of phenol with SDCs didn't surpass that with a granular commercial activated carbon (X_{con} = 97%, X_{TOC} = 88.1%). Considering the low cost and environmental friendship, pyrolysis for producing catalysts or supports is still a desirable alternative for the sludge disposal. In recent years, the persulfate-based advanced oxidation process has developed rapidly and gained much attention. Unlike CWAO and Fenton-like systems, this process is under milder temperature and pressure at a wide pH range. It is important to develop efficient and cost-effective activators for the production of sufficient oxidative radicals which play a key role in pollutant degradation [75]. Interestingly, the SDCs are proved to be an efficient activator. Wang et al. [67] used this approach for the degradation of triclosan, and 99.2% of triclosan was converted and 32.5% of TOC was removed within 240 min. Huang et al. [68] synthesized biochar via facile one-pot pyrolysis of sewage sludge and applied it to treat bisphenol A in persulfate decomposition, resulting in TOC removal of ca. 80%. As mentioned above, the SDCs have been tested with desirable catalytic activity in the decomposition of organic pollutants during various AOPs. However, prior to the practical application, the problems need to be demonstrated detailed and solved appropriately, including the degradation mechanism, catalytic efficiency, HMs leaching, and the attrition resistance of these SDCs.

The identification of intermediates generated from the catalytic process by gas chromatography-mass spectrometer (GC-MS) can help simply inferring the reaction pathway for pollutant degradation. And the possible mechanism can be concluded via integrating the degradation pathway with the detection of radicals and the characteristics of SDCs. Yuan and Dai [69] detected that $^{\bullet}OH$ rather than $^{\bullet}OOH/^{\bullet}O_2^-$ was the main active species arising from the decomposition of H_2O_2 with SDCs supported iron via the catalytic process [69]. Thus, they suggested that rhodamine B (RhB), the target pollutant, could transfer an electron to Fe(III) on the catalyst, assist H_2O_2 decomposition and $^{\bullet}OH$ generation, and initiate the degradation and mineralization subsequently. Wang et al. [15] prepared an efficient sludge carbon catalyst by the modification of HNO_3. In order to probe into the mechanism, determining the intermediates during the reaction was carried out by GC-MS and subsequently,

four degradation pathways for *m*-cresol were proposed. Interestingly, the means of the density functional theory method was successfully utilized to confirm the possibility and feasibility of these four decomposition pathways. Fig. 3 shows the degradation mechanisms of *m*-cresol in CWPO and CWOO. The reason for the fast degradation of organic pollutants could be attributed to functional group interaction, π–π interaction and the catalytic disintegration of an oxidizing agent by iron on the surface of SDCs.

The reaction condition is one of the important factor which influences the degradation efficiency of organic pollutants. In order to find the optimal conditions, various parameters such as the reaction temperature, time, initial pH and the catalyst dosage are measured respectively or interactively. Khataee et al. [70] synthesized a nanocomposite catalyst (ZrO_2-BC) by loading ZrO_2 on biochar produced from wheat husks and sludge. The single-factor method was used to study the impacts of four main parameters including catalyst dosage, solution pH, initial Reactive Yellow 39 (RY 39) concentration and ultrasonic power on the degradation efficiency of the contaminant. Consequently, degradation efficiency as high as 96.8% was obtained under the most suitable condition. However, the single-factor experiments are carried out without concerning interaction and combined effect of factors. Conversely, response surface methodology (RSM) is frequently used to seek the optimal condition and identify the interaction and the combined effects of factors. RSM is an effective tool applied in the design, development and formulation of new products, as well as in the improvement of existing product designs. Wang et al. [43] firstly obtained the optimal condition for *m*-cresol degradation in CWPO with SDCs modified by HNO_3 with the help of RSM. As shown in Table 2, 96% of *m*-cresol was converted and 47% of TOC was decomposed after 2 h under the optimized conditions where the amount of H_2O_2 much lower than the stoichiometric value.

The catalytic degradation of pollutants could be accelerated with some active metals loaded on the sludge carbon supports. In many studies, it was reported that the sludge carbon-supported iron oxide catalyst had a better performance than the pristine sludge carbon [1,69,72]. For example, Tu et al. [73] firstly used SDCs as the support of heterogeneous Fenton-like catalyst and iron was loaded as an active component of the catalyst. In contrast with SDCs, the SDC-supported iron oxide catalyst (Fe-SDC) exhibited a better performance in the degradation of Acid Orange II (AOII). Similar results were found in other studies. A novel Fe-SDC was developed to treat 2-chlorophenol in CWAO [1]. After 5 h of reaction with Fe-SDC catalyst, the conversion and TOC removals were 100% and 76%, respectively. Comparatively, both X_{con} and X_{TOC} were almost as low as 20% when using SDCs. However, homogeneous catalysis was mainly responsible for the removal of organics since 27 mg/L of iron was detected in the solution. Furthermore, the iron leaching was ca. 7 wt.% of the total amount of iron initially in the Fe-SDC. Leaching is a major issue when utilizing catalysts loaded with metal oxides under acidic solutions. The leaching impairs the cyclic and long-term use of catalysts. Efforts are ongoing to completely or partially solve the leaching issue. Minimization of the metal leaching could be effectively attained by the use of acetate buffer (pH = 4.5) while only a slight part of the catalytic activity is prevented. This indicates that controlling the pH value according to appropriate ways (e.g., adjusting the solution

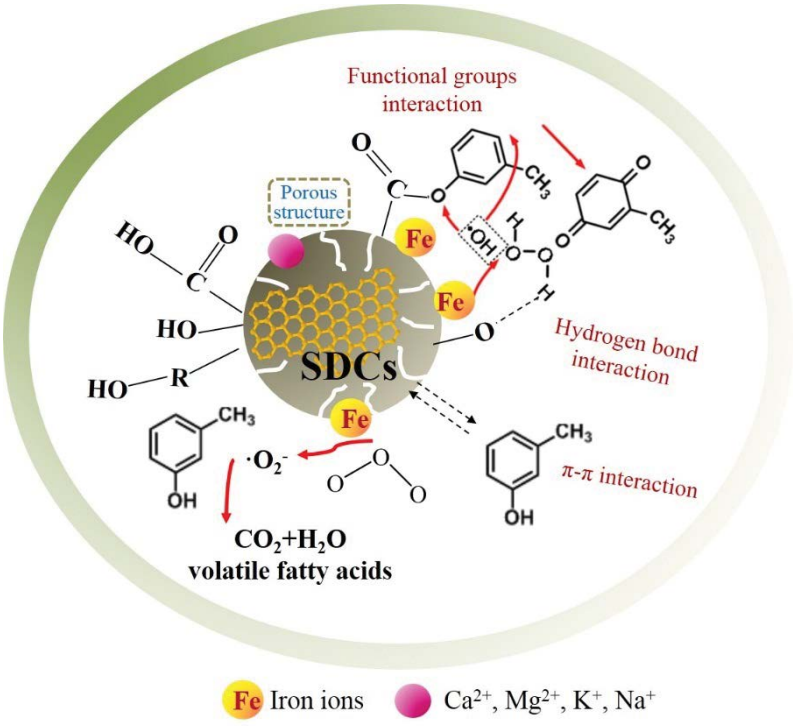

Fig. 3. Degradation of *m*-cresol with SDCs in CWPO and CWOO by various mechanisms.

Table 2
Preparation, characteristics of sludge derived carbons and their application in the catalytic degradation of organics

Catalysts	Carbonation temperature (°C)	Modification	Catalytic system	Dosage (g/L)	Reaction time (h)	Contaminants	Degradation	References
Fe-SDC	800	None	CWAO	3.3	5	2CP (2 g/L)	X_{con} = 100% X_{TOC} = 76%	[1]
SDC	600	HNO_3	CWAO	0.5	3	*m*-cresol (0.1 g/L)	X_{con} = 100% X_{TOC} = 67.1%	[65]
SDC	600	None	Fenton	No data	2	1,2,4-acid	X_{TOC} = 87.2%	[35]
SDC	600	HNO_3	CWAO	5	1.5	*m*-cresol (5 g/L)	X_{con} = 99% X_{TOC} = 67.9%	[63]
SDC	No data	K_2CO_3	CWAO	20	4	Phenol (5 g/L)	X_{con} = 93.2% X_{TOC} = 82.1%	[66]
SDC	No data	Steam and HCl	CWAO	20	4	Phenol (5 g/L)	X_{con} = 93.0% X_{TOC} = 78.0%	[66]
SDC	450	NaOH	Persulfate	1	0.5	Triclosan (10 mg/L)	X_{con} = 99.2% X_{TOC} = 32.5%	[67]
SDC	600	HCl	Persulfate	0.2	0.5	Bisphenol A (10 mg/L)	X_{TOC} = 80%	[68]
Fe-SDC	350	None	UV/Fenton	0.05	0.5	RhB (55.5 mg/L)	X_{TOC} = 81.0%	[69]
Fe-SDC	350	None	Vis/Fenton	0.05	2	RhB (55.5 mg/L)	X_{TOC} = 59.0%	[69]
ZrO_2-BC	600	Ethanol and HCl	Sonocatalyst	1.5	1.12	RY 39 (20 mg/L)	X_{con} = 96.8%	[70]
SDC	600	HNO_3	CWPO	1.2	2	*m*-cresol (100 mg/L)	X_{con} = 96% X_{TOC} = 47%	[43]
SDC	700	$ZnCl_2$	CWOO	0.2	0.67	Oxalic acid (9 mg/L)	X_{con} = 80% X_{TOC} = 57%	[64]
SDC	700	None	CWOO	1.0	0.5	Phenol (0.2 g/L)	X_{con} = 95.4%	[71]
Fe-SDC	600	None	Fenton	1.0	2.5	Black-T (1 mmol/L)	X_{TOC} = 71%	[72]
Fe-SDC	800	None	Fenton	2.0	2	AOII (100 mg/L)	X_{con} = 94.6%	[73]
Fe-SDC	700	HCl	Electro-Fenton	No data	2	Real biologically pretreated CGW	X_{TOC} = 65.5%	[74]

pH initially or using a buffer reagent during the reaction) could reduce the leaching of metals [1]. Apart from pH control, the preparation methods of SDCs are also important for controlling metals leaching. In Yuan et al. [69], they advised an approach of preparing sewage sludge-derived mesoporous materials via a facile synthesis method. The experimental results showed that the original iron in sludge and additional iron from chemical agents were both identified as the active sites of the SDCs catalyst in photo-Fenton reaction with UV or visible light. Notably, this mesoporous catalyst loaded with Fe exhibited excellent stability of catalytic activity and low Fe-ion leaching (<0.7 mg/L). Wen et al. [72] pretreated the sludge by radical oxidation combined with iron impregnation and then the pretreated sludge was carbonized to prepare the carbon encapsulated Fe nanoparticles. This catalyst was highly stable and active as being used in a heterogeneous Fenton-like system to degrade Black-T with a high TOC removal of 71%. Compared to conventional impregnation, the catalyst exhibited well-dispersed iron nanoparticles encapsulated in a carbon matrix, which permitted a high catalytic activity for degradation of organics and superior stability against iron leaching. The bulk and surface iron concentrations were 27% and 17.2%, respectively. Therefore, most irons were dispersed on the surface of the catalyst, which was one reason for the high catalytic efficiency. In addition, the pore walls which were composed of ash including iron species could help to promote the significant reduction of TOC by polar interaction. Therefore, optimizing the methods of preparing SDC catalysts is a potential way to prevent the metals leaching.

The sludge-based carbon catalysts cannot only prompt the decomposition of pollutants directly but also improve the biodegradability of wastewater by converting refractory organics into less toxic substances. Accordingly, the AOPs with SDCs as catalysts can be combined with post-biodegradation technology for discharged industrial wastewater to satisfy the discharge standards. Zhuang et al. [76] strengthened the biodegradability of the Lurgi coal gasification wastewater (CGW) by the technology of catalytic ozonation. The SDCs activated by $ZnCl_2$ were supported with manganese and ferric oxides for exhibiting better catalytic performance. The BOD_5/COD value increased from 0.06 to

only 0.15 by ozonation alone, while this value was improved to 0.55 with catalyst. The effluent can be considered totally biodegradable as BOD_5/COD is higher than 0.4 [77]. As illustrated in Table 2, the TOC removal rate was in the range of 62.7%–66.0%. Following the catalytic ozonation, the biological treatment process could be selected to be a feasible method for safe and cost-effective treatment. Similar results were also found in other researches. In three-dimensional electro-Fenton for the treatment of biologically pretreated coal gasification wastewater, the Fe-SDC as both catalytic particle electrode and catalyst could enhance total phenols removal efficiency to 93.5%, therefore the biodegradability was dramatically improved (BOD_5/COD from 0.09 to 0.54) [74]. In Wang et al. [15], the BOD_5/COD of wastewater mainly comprised of *m*-cresol was enhanced from the original 0.28 to 0.88 at the end of the reaction. These researches show that SDCs are catalytic materials of high efficiency in decreasing the acute toxicity of wastewater during the AOPs prior to the biological treatment.

3.3. Risk analysis in application

The excellent performances of SDCs in adsorption or catalytic degradation for removing pollutants have been illuminated, but the potential risk of the biochar in wastewater treatment is advised to be explored [78]. In comparison with precursors rich in lignin and cellulose, sludge generated from municipal or industrial wastewater treatment is more diverse, especially in metal types and content. As aforementioned, the existence of various cationic metal ions is important to the catalytic and adsorption performance of SDCs. However, it also poses a threat to the environment and organisms if the HMs are released from the solid phase. Since some heavy metals originally exist in sludge, the feedstock of SDCs, decreasing the leaching potential of HMs is also an important subject in the safe application of SDCs. Many researches have been carried out on the risk analysis of biochar made from agriculture wastes or soils for bioavailability and eco-toxicity of the HMs [61,79]. However, seldom studies are on a similar field of biochar made from sewage sludge. It is found that HMs are accumulated in the biochar matrix after the pyrolysis of sludge. The environmental risk of HMs highly depends on their chemical fractions or binding forms in SDCs.

As mentioned in the previous section "pyrolysis", microwave-assisted pyrolysis is an effective method for reducing the HMs leaching from SDCs [26,29,31]. Besides, the pyrolysis temperature can importantly influence the chemical behavior of HMs in SDCs. Devi and Saroha [5] prepared biochar from the sludge which was produced in pulp and paper mill wastewater treatment plant. The impact of pyrolysis temperature in the range of 200°C–700°C was examined. The leaching potential of HMs declined with an increase in the pyrolysis temperature because mobile and bioavailable fractions of HMs were inclined to be transformed to the relatively stable fractions at a higher temperature. Therefore, it is indicated that higher pyrolysis is beneficial for the immobilization of HMs in SDCs. However, the properties of SDCs such as porosity, chemical components and functional groups may be significantly altered once extreme high temperature is employed. In addition, the sludge structure may be destroyed and converted into aromatic compounds [80]. Thus, there is a vital need for a balance between safe application and efficient utilization.

In addition to the pyrolysis temperature, the pretreatment of sewage sludge prior to the carbonization can also influence the amount of HMs released from SDCs. Noteworthily, Wang et al. [4] prepared SDCs by a novel approach, coupling hydrothermal pretreatment under the atmosphere of argon rather than air or oxygen, with pyrolysis. Compared with direct sludge pyrolysis, the SDCs hydrothermally treated at 220°C and pyrolyzed at a higher temperature (≥500°C) had a better quality along with low toxicity. The result can be explained by some interactions such as complexation, precipitation and adsorption between the crystal lattices of sludge and HMs at a higher temperature of pretreatment [17,18].

Recently, the co-pyrolysis of sludge with other biomass has been proposed for more valuable biochar with multiple possible applications [81]. In Yang et al. [82], co-pyrolysis of sludge and sawdust was proved to be an excellent way of reducing the leachable metal concentration. This phenomenon could be ascribed to: (1) more percentages of aromatic carbon in biochar through co-pyrolysis, which supplying π-electron to bond HMs; (2) the reduction of H and N polar sites in biochar by adding sawdust; (3) the charge neutralization between heavy metal ions and inorganic structures with a negative charge like [SiO_4] and [AlO_4] tetrahedrons in biochar. It should be noted that not all the immobilization mechanisms of HMs in biochar are the same via co-pyrolysis. In Dong et al. [83], they mixed sludge with rice straw and obtained biochar that could be used for soil amendments. They insisted that the decrease in HMs leaching was a result of the transformation of HMs from the acid-extractable fraction to the oxidizable fraction under the promoted reductive atmosphere, and the enhanced specific surface area which could adsorb and restabilize some released HMs during co-pyrolysis.

In summary, when the physicochemical properties of SDCs satisfy the requirements for pollutant removal, it is recommended to employ microwave-assisted pyrolysis, higher pyrolysis temperature, hydrothermal pretreatment (>200°C) or co-pyrolysis with other precursors. An appropriate method for preparing SDCs cannot only satisfy the biochar quality but also reduce the HMs leaching. Based on the above studies, it is suggested that future researches on SDCs should involve the risk analysis of their application and approaches for reducing these environmental risks.

3.4. Recycling

Although SDCs have been successfully used for the adsorption and catalytic degradation of pollutants in aqueous solution, most researches are performed with the SDCs powder without magnetic property, causing the difficulty in recycling and secondary pollution. For instance, the particle of sludge carbon used in the batch reactor was less than 200 mesh and no magnetism was detected in Wang et al. [43]. Considering the difficulty in recycling the powdered SDCs, it is desirable to find suitable ways to separate the sludge-based biochar from aqueous solution. There are two main ways to complete the target: one is preparing SDCs

with a shape such as granular activated carbon; the other is making magnetic SDCs which is recognized as a more promising strategy [84].

At first, the method of preparing SDCs with large size was tried. Liu et al. [51] used calcium as a binder to make granular activated carbon (4 mm in diameter and 9 mm long) from sludge and its adsorption of MB reached 131.9 mg/g which was also a desirable capacity. However, it was subsequently noted that the granular activated carbons made from sewage sludge performed a low attrition resistance, blocking their application in adsorption processes or adsorption beds with pressure swing [66]. Meanwhile, Smith et al. [85] investigated the pelletization for enhancing the attrition resistance of SDCs and found the most effective method of pellet production which consisted of lignosulphonate binder and the post-pelletization activation. Since the cut-off size between powdered and granular SDCs is 80 mesh (ca. 0.18 mm), it is also hard to recycle the granular SDCs with millimeter-sized diameter. Moreover, the mass transfer resistance between liquid and solid, the attrition resistance, and intensity should be all considered once the size of granular SDCs is too large. Therefore, developing magnetic SDCs by introducing magnetic media, including Fe, Co, Ni and their oxides, into the sludge carbon matrix is an efficient approach to separate these biochars quickly from aqueous solution via magnetic separation technique. Additionally, loading magnetic substances into SDCs could offer a potential to improve the adsorption performance due to the strong sorption affinity of iron oxides with phosphate, selenite and organic arsenic [86]. Dewatering is one important procedure during the disposal of sewage sludge, which is significantly different from other biochars. Persulfate activated by ZVI or Fe^{2+} is proved to be a sustainable technique for enhancing the dewatering of sludge [21,87]. The iron is embedded into the sludge matrix during the dehydration process and the SDCs which contained magnetic iron oxides are obtained after the subsequent pyrolysis. Consequently, there is no need of extra magnetic substances for the preparation of magnetic SDCs. Chen et al. [23] invented a novel method to treat the persulfate-ZVI dewatered sludge by producing magnetic biochar as an environmentally friendly biosorbent to remove HMs from wastewater. This SDC was conformed to the typical model of soft magnetic material and its saturation magnetization reached a high value of 54 emu/g. This indicated that the magnetic SDC could be easily separated from the aqueous solution by using an external magnet.

After recycling, it is worth studying that, whether the used SDCs still have the potential of adsorbing organics and HMs or catalyzing the oxidation of refractory organics. In this aspect, many researches have been carried out. Wang et al. [15] examined the reusability of SDCs catalysts by triple-time batch experiments in CWPO. After the first run, only 0.97% of iron was released into the solution, resulting in high *m*-cresol conversion of 97.6%, 94.1% and 92.3% for the first, second and third runs, respectively. In contrast, Zhang et al. [71] used SDCs in the catalytic ozonation and the phenol removal dropped from 95.4% (first trial) to 59.3% (fourth trial), indicating poor stability. They believed that the poor reuse stability of SDCs catalysts would be attributed to biochar mass loss and change in surface chemistry. Wen et al. [72] compared the reusability of the catalyst prepared by radical assisted impregnation (SC-F-0.2) and catalyst prepared from direct impregnation (SC-I-0.2). The dye removal still reached 91% with SC-F-0.2 after three repeated experiments, while the degradation dropped to 61% with SC-I-0.2. Because the Fe leaching in SC-F-0.2 was only 2.77% in comparison with 17.3% in SC-I-0.2. In order to maintain the catalytic activity of SDCs, the release of active components should be inhibited by the improved preparation method like radical assisted impregnation and nitric acid modification at a low temperature which is suggested in the above literature [15,72].

4. Conclusions and prospects

This literature review suggests that sewage sludge is a highly promising precursor for the generation of adsorbents, catalysts or catalyst supports. Unlike biomass such as agricultural and forest residues, sewage sludge is an available starting material without seasonality. Moreover, this conversion of sludge into useful substances for pollutant removal is deemed to be an attractive alternative to the existing sludge disposal and reuse routes. Considering the poor performance of pristine SDCs in removing pollutants, physical or chemical modification methods are employed to promote the activity of these sludge-based biochars. The huge specific surface area, high porosity and abundant oxygen-containing functional groups on the modified SDCs provide a large number of active sites for binding organic pollutants via hydrogen bond interaction, n–π conjugate action, electrostatic adsorption, ion exchange process or other interactions. Furthermore, the porous structure and abundant functional groups of SDCs facilitate its combination with other functional materials or catalytic active components. These distinguished properties of SDCs make them one of the most promising materials in the application of wastewater treatment. It can be concluded that SDCs are not only excellent adsorbents for heavy metals and organic pollutants especially dye but also efficient catalysts or catalyst supports. Previous studies prove that these synthesized sludge-based catalysts can be effectively utilized in various AOPs during which high toxic pollutants are decomposed to less toxic or even non-toxic substances. As for the pollutants with high toxicity and insignificance of recycling, catalytic degradation is a more suitable alternative than adsorption.

Although many achievements for the sludge-based biochar in wastewater treatments have been made, there still remain some unsolved issues for the preparation of these biochars and their application in pollution control. For example, it is necessary to make a good balance between the removal rate of pollutants and the economy. Meanwhile, the properties of these materials, such as the chemical composition and structure, heavily depend on the sludge types and synthesized methods. Consequently, the precise control of SDCs with desired chemical composition and structures is still difficult to realize. Most literature mainly concerns with the modification methods and there is little information on synthesizing the SDCs at nanoscale for better performance. Furthermore, most researches are on the use of SDCs in the model wastewater rather than the practical effluents which may have one or more characteristics including salinity, extreme acid/alkaline, and high temperature. Since there

are generally more than two kinds of contaminants in practical wastewater, it is necessary to analyze the competition between diverse organic substances during the process of adsorption or catalytic degradation. As stated in section "risk analysis in application", the potential inferior effects of using SDCs on the organisms and environment should be discussed in detail in future work. Finally, prior to the industrial application, the strength, stability and recycling of these sludge-based biochars are inevitable issues that need illuminating.

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