Review Article

Recycling of Waste Sludge: Preparation and Application of Sludge-Based Activated Carbon

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With the rapidly increasing industrial and agricultural development, a large amount of sludge has been produced from much water treatment. Sludge treatment has become one of the most important environmental issues. Resource utilization of sludge is one of the important efficient methods for solving this issue. Sludge-based activated carbon (SBAC) materials have high adsorption performance and can effectively remove environmental pollutants including typical organic matter and heavy metals through physical and chemical processes. Therefore, developing efficient SBAC materials is important and valuable. At present, preparation, modification, and application of SBAC materials have gained widespread attention. This paper provides a review of the research on SBAC preparation and modification and its utilization in removing environmental pollutants. It included the following topics present in this review: conventional and new methods for preparation of SBAC were clearly present; the effective methods for improving SBAC performance via physical and chemical modification were reviewed; and the correlation of their physico-chemical properties of SBAC with pollutants' removal efficiencies as well as the removal mechanisms was revealed. SBAC has a better adsorption performance than commercial activated carbon in some aspects. Furthermore, it is a cost-effective technique and has a wide range of raw materials. However, there are still some drawbacks to its research; thus, some suggestions for further research were given in this review.

1. Introduction

With the rapid development of water treatment industry, the amount of activated sludge produced by treating sewage increases [1]. Because activated sludge is characterized by high water content, loose structure, a large amount of organic matter, and poisonous and harmful substances (such as microorganism, heavy metal, and poisonous organism) [2], it has become an important environmental issue and needs to be treated through an effective pathway. At present, sludge utilization has become one of the important methods for sludge disposal [3–5]. For example, sludge-based activated carbon (SBAC) material which is prepared with treated activated sludge can be used to adsorb and treat pollutants [6, 7]. The material is a black amorphous carbon material made by activated sludge that came from water treatment, which is

made by blending, carbonization, activation, and so on. It has the characteristic of a dense pore, complex pore structure, and large specific surface area; so, it has higher absorbability [8]. In the 1970s, using sewage sludge as raw material to prepare SBAC has been reported [9]. Its preparation and application have become one of the pathways to dispose the waste sludge.

SBAC is an environment-friendly adsorbent [10], which has good adsorption performance, wide range of source of raw material, stable chemical properties, and repeated utilization and recovery. In addition, it has good thermal stability and also has been reported to be safe for use at present [11]. Therefore, its focus has been moved to how to enhance the performance of SBAC. The selections of carbonization activation process and carbonization material are therefore important for the preparation. For example, prepared by

phosphoric acid activation and microwave pyrolysis, the SBAC has excellent performance in adsorption of organic matters. As a result, the maximum methylene blue adsorption value by the SBAC is higher than that by the activated carbon for purified water [12]. Good SBAC material is important for economic and social development; preparation and application of SBAC have received extensive attention in the field of environmental protection. Also, more and more researches are reported and push the development of sludge utilization exploration. At present, the direction of the preparation method of SBAC material which has previously obtained the characteristics of a specific physicochemical property is not clear. Therefore, this paper adopts different classification models to summarize the preparation and application process of SBAC and to clarify the method of constructing and applying SBAC with specific physicochemical structure. The short review was aimed to review the development of SBAC in its modification and environmental application using different classification models.

2. The Preparation of SBAC

SBAC can be prepared by direct pyrolysis method [13, 14], physical activation process [15, 16], chemical activation process [17, 18], physical-chemical activation process [19], microwave activation [20], and so on. These methods can be used to produce porous carbon-adsorbing materials. The physical activation often uses traditional Muffle furnace heating method, while microwave activation has received much attention because of its high efficiency and easy control. Menéndez et al. [21] have demonstrated that the microwave preparation of activated carbon is highly feasible. The carbon content of SBAC would be low; adding carbon source material (e.g., corn kernel [22, 23], wood chip [24], peanut shell [25], and hazelnut shell [26]) is the possible increase of carbon source supply, which potentially enhances its efficient use in large scale. For instance, adding 20% of the peanut shells to the municipal sludge as carbon sources, the SBAC developed by Jingjing [27] exhibited abundant pore structures and strong adsorption performance.

Figure 1 shows the modification and preparation pattern of SBAC, including direct pyrolysis, physical activation process, chemical activation, and physical-chemical activation. Direct pyrolysis is carried out under inert gas to obtain SBAC by directly drying and pulverizing activated sludge [28]. It consists of three stages: the dehydration stage, the second stage in which the large amount of volatile components are dissolved out in the pyrolytic zone, and the third stage where the residual material continues to be slowly pyrolyzed [29]. Physical activation (gas activation method) is able to directly pyrolyze and dry the grinding-activated sludge under inert gas protection and then pyrolyze it again thus obtaining SBAC under the other protective gas (such as CO_2 [30], water vapor [31], and flue gas [32]). Chemical activation puts raw sludge materials and chemical reagents together at a ratio [33] or dip-dried sludge in chemical reagent solution according to a certain solid-liquid ratio [34] and then pyrolyzes the hybrid product for SBAC [35], as shown in Figure 2. Physical-chemical activation mixes the sludge with

FIGURE 1: The mode for preparation of SBAC.

the chemical reagent in a proportion ratio; thereafter, they are pyrolyzed to obtain SBAC under the protection of inert gas [19]. Microwave activation pyrolyzes and carbonizes sludge into SBAC through microwave heating [36–38]. Compared with the traditional activation processes, the microwave activation has the advantages of high efficiency, low energy consumption, low cost, and small pollution [39].

3. Modification of SBAC

The factors that influence SBAC-specific surface area, adsorption yield of activated carbon, surface functional groups of activated carbon, and other characters include activation reagent class, activation temperature, activator concentration, pyrolysis time, impregnation ratio, and excess substance ratio [41]. The pretreatment of sludge also affects the performance of SBAC, such as the sludge treated with Fenton pretreatment, which can effectively improve SBAC characters [42]. The electro-Fenton (EF) process is a new electrochemical process that produces free radicals and iron ions. When using iron as an anode, H_2O_2 is produced by dissolved oxygen electrolysis and Fe^{2+} is produced by electrolytic corrosion of anode. The combination of H_2O_2 and Fe^{2+} can produce strong oxidative free radicals, having a high oxidation potential up to 2.8 eV. In the EF process, the sludge flocculation products are to be broken and destabilized and the decomposed flocculation products are complexed with oxidized Fe^{3+} to enhance metal dispersion and modification [43]. Gu et al. [44] showed a simple method for loading the magnetic carbon to Fe nanoparticle (EF-SBAC) using EF pretreatment sludge, as shown in Figure 3. A new-type magnetic sludge-derived carbon was synthesized by continuous EF activation and carbonization, which had a good physicochemical property, including high Fe insertion rate (74%), small size (4.77 nm) of nanoparticles, and good dispersion. The EF-SBAC revealed good catalytic activity, stability, and availability. For example, EF-SBAC could remove 96.1% methyl orange within 60 min, while its iron leaching rate was only 1.4%.

FIGURE 2: The preparation of SBAC using corn stalks mixing with sludge by chemical activation method [40].

FIGURE 3: The preparation of EF-SBAC [44].

The SBAC modification methods can be divided into surface physical modification and surface chemical modification. The structural characteristics of SBAC can be observed by instrumental analysis such as scanning electron microscope (SEM), Fourier infrared transform spectrum, X-ray diffraction and X-ray photoelectron spectroscopy, and other parameter analyses such as BET surface area analysis denoted as BET analysis in this paper.

3.1. Surface Physical Modification. The physical structure modification of SBAC is mainly to increase the capacity of SBAC adsorption, which is affected by BET size, pore size,

internal volume structure of micropore and volume capacity, micropore distribution on its surface, and so on [45–48]. To prepare an efficient SBAC having high BET value and wide and uniform pore distribution, we usually modify its surface structure by temperature activation, time adjustment, and oxidation corrosion of chemical reagent. The SBAC effectiveness can be improved significantly by the specific modification of SBAC but the selection of modification method is related to removal of pollutant. The SBAC adsorption characteristic was performed when the molecular size of pollutants was less than or equal to aperture of SBAC, and then the pollutant enters inside the aperture and absorbed and

FIGURE 4: (a) The SEM images of SBAC (a) without corn bar; (b) with corn stalk [40].

separated [49]. In the process of carbonization and activation of SBAC, the physical structure of SBAC was significantly affected by external physical conditions and modifier [50]. Li et al. [40] added 25% corn stalk into the sludge using 4 mol/L ZnCl_2 as activator, with the activation temperature at 600°C and pyrolysis time at 60 min, to prepare a SBAC, which had a BET value as high as $769.0 \text{ m}^2/\text{g}$. As shown in Figures 4(a) and 4(b), it showed that the SEM images of SBAC after modification through hybridizing corn stalk changed. The modified SBAC had an obvious longitudinal deep-hole structure evenly distributed on the surface. In the process of sludge pyrolysis, ZnCl_2 can promote the secondary pyrolysis, reducing the formation of tar and promoting the dehydration of activated sludge and aromatization of organic matter [51]. The iron salt can effectively improve the pore structure of SBAC and improve the SBAC adsorption performance [52–54]. Jin et al. [55] showed that iron additive as catalyzer during activation was added into sludge, which could accelerate the reaction between carbon and high thermal water vapor and the pore structure adjustment of SBAC. Su et al. [56] added iron salts to the sludge to prepare cost-effective iron-containing porous carbon (Fe-SBAC). The pyrolysis mixture of sludge and iron salts was favorable for adsorption of lead. With specific surface area, total pore volume, and average pore width of $321 \text{ m}^2/\text{g}$, $0.25 \text{ cm}^3/\text{g}$, and 3.17 nm , respectively, it resulted in the hydroxyl groups and carboxyl groups significantly. Fe-SBAC adsorption capacity can be as high as 128.9 mg/g , while the adsorption capacity of activated carbon is 79.1 mg/g .

The performance of SBAC was significantly affected by carbonization time and carbonization temperature. Because the sources of raw materials are different, the preparation temperature of SBAC required to reach maximum BET would be also different. For example, Jeyaseelan and Lu [57] showed that in the carbonization process, the best temperature is 850°C ; Inguanzo et al. [58] found that the best temperature was 650°C ; Bagreev et al. [59] indicated that 950°C was the suitable temperature; Rio et al. [60] had the best temperature fixed at 1000°C . When Fan et al. [61] added chitosan to modify SBAC, it was found that both carbonization time and carbonization temperature could change BET value and pore volume of SBAC. Compared with the carbonization time, carbonization temperature had a much

more significant influence. Silva et al. [62] heated sludge materials at room temperature till 300°C for 2 hours in the presence of N_2 flow. With temperature elevated to a different level (750°C , 800°C and 800°C) and activation under CO_2 flow for 1 h, different kinds of SBAC were prepared. It showed that the performance of SBAC was different by varying activation temperature.

Using chemical reagent activation treatment can modify BET and pore volume of SBAC and increase its adsorption efficiency. Linru et al. [63] showed that an increase in the size of BET and pore of SBAC occurred after nitric acid modification. The possible reason for the increase was that the oxidation corrosion of HNO_3 destroyed the inner channel structure, which broke through the small pore and increased the surface area. Daoxiong [64] found that the use of nitric acid, ammonia, or hydrogen peroxide could improve the physical surface properties of SBAC, especially the treatment of ammonia, which could improve the pore diameter structure and increased the volume of micropore. Zhai et al. [65] studied the HNO_3 and NH_3 modification influence in the physical properties of the SBAC surface. The results showed that the BET of SBAC increased and the total pore volume and the pore volume also increased. The maximum BET and pore volume of SBAC prepared at 750°C were $258 \text{ m}^2/\text{g}$ and $0.198 \text{ cm}^3/\text{g}$, respectively. Yang et al. [54] used iron as activator and pyrolyzed biological sludge to prepare SBAC, as shown in Figure 5. It shows that pyrolysis temperature and mass ratio (activator/dry sludge) significantly affected the BET value and pore characteristics of SBAC. It was included that iron activation could promote the development of the porous structure of SBAC; the best preparation condition for the pyrolysis temperature was 750°C and the mass ratio (activator/dry sludge) was 0.5.

The washing of different solutions on the sludge affects the surface physical structure of SBAC as shown in Table 1. Alvarez et al. [66] studied the washing influence of hydrochloric acid and sodium carbonate in the SBAC prepared under carbon dioxide activation. During the whole activation process, the carbon content of pretreated sludge with hydrochloric acid and sodium carbonate decreased, while the content of ash increased. The increase of ash content was more obvious after pretreatment of sodium carbonate.

FIGURE 5: The preparation of SBAC using activated iron [54].

TABLE 1: Surface physicochemical characteristics of SBAC generated with pretreatment by different substances.

Substance	SBET ($\text{m}^2 \cdot \text{g}^{-1}$)	Surface property change	Reference
KOH	984	SBAC (KOH-800) is mainly mesoporous. The SBET and total pore volume of SBAC are the closest to those of commercial AC (the BET surface and total pore volume of the commercial AC were $984 \text{ m}^2/\text{g}$ and $0.663 \text{ cm}^3/\text{g}$, resp.).	[18]
NaOH	179	After modification with NaOH, the shape of SBAC is mesoporous and macroporous, showing a stronger adsorption capacity for CO_2 than that of commercial AC.	[67]
HCl	269	After the sludge was acid washed with HCl (5 M), slight pore development was observed and the BET surface area is $269 \text{ m}^2 \cdot \text{g}^{-1}$ when activated under CO_2 at 800°C .	[19]
H_3PO_3	<5	Chemical activation with H_3PO_4 was ineffective for an efficient porosity development in the SBAC preparation process.	[19]
Na_2CO_3	440	The Na_2CO_3 washing step significantly promotes the development of the porous structure in CO_2 activation of the treated char.	[66]

FIGURE 6: The SEM images of sludge: rapid pyrolysis (a); precursors after pickling (b); sodium carbonate treatment (c) [66].

The difference of surface characteristics by varying pretreatment reagents is shown in Figure 6.

3.2. Surface Chemical Modification. SBAC surface chemical modification is to change the chemical properties of the surface of SBAC and control the hydrophilicity and hydrophobicity of the adsorption process and the binding capacity between SBAC and pollutant. The activation methods often contain acid activation, alkali activation, and salt activation, and the activators mainly include ZnCl_2 [68], H_3PO_4 [69], NaOH [70], KOH [71], and H_2SO_4 [72]. There are different types of functional groups formed on the surface of SBAC if using different types of activators, such as alkaline functional groups, acidic functional groups, oxygen-containing functional groups, and nitrogen-containing functional groups. The coupling of various activation methods can improve SBAC adsorption efficiency and overcome the

defect of single activation [73]. At present, the SBAC surface chemical modification has been widely used, but there is still a deficiency of corrosivity in the chemical activation method. Table 2 illustrates the typical methods for preparing SBAC by surface chemical modification.

The acid modification and alkaline modification of SBAC can enhance its adsorption performance. Huijun et al. [86] used zinc chloride activation and nitric acid to modify SBAC. The result showed that the nitric acid modification can greatly improve the performance of SBAC. The organic matter and inorganic matter decomposition of sludge evaporated more completely, providing more functional groups which was a better adsorption to Cd^{2+} . Lu et al. [87] studied the catalytic oxidation of chlorobenzoic acid (p-cba) with two kinds of SBAC which were modified by $(\text{NH}_4)_2\text{S}_2\text{O}_8$ and NaOH. The study showed that the removal effects of two kinds of SBAC were significantly different. The main reason for the

TABLE 2: Typical methods for preparation of SBAC.

Source of sludge	Modifier	Synthetical method	Treated compounds	Synthetical conditions	Results	Reference
Yanzhuang sewage treatment plant in high-tech zone	Coke powder	Chemical activation method (coupling of acid and salt : sulfuric acid, zinc chloride)	Ammonia nitrogen, chromaticity	Activation temperature: 750°C; activation time: 2 h; mass ratio of sludge to blue charcoal: 5 : 2; V (sludge-blue charcoal) : V (activator) = 1 : 2.5	The SBAC pore structure was dominated by the middle hole. The BET area was small. The preparation of SBAC contained many types of hydrocarbon and oxygen-containing functional groups.	[72]
A sewage treatment plant in Songjiang district	Loading metal Co and Fe	Hydrothermal method	Dye wastewater: Congo red	Hydrothermal time: 3 h; temperature: 180°C; and ultrasonic time: 10 min	The surface and internal hole of SBAC occurring after adding nanocobalt, which increased the BET; SBAC had good magnetic separation performance; the separation of solution and adsorption material was easy to separate, which were conducive to the recovery and utilization of SBAC.	[74]
Ningbo APP Asia pulp paper deinking sludge	Nothing	Chemical activation method (acid activation : phosphoric activation)	Modeling iodine solution and methylene blue as standard of evaluation	Activation time: 90 min; activation temperature: 450°C; mass impregnation ratio of sludge to phosphoric acid solution is 1 : 1.3; phosphate concentration : 70%	The BET of SBAC was large and contained a large number of hydroxyl groups. The middle hole was developed and stable. The microcrystal was irregular and the crystallization degree was high.	[75]
A sewage treatment plant in Zhejiang province	Loading manganese and iron oxide	Chemical activation method (salt activation: ZnCl ₂)	Effluent of biochemical treatment of coal gas wastewater	5 mol/L ZnCl ₂ solution; impregnated time: 24 h; mass of solid : liquid ratio: 1 : 3; activation temperature: 600°C and time: 3 h	Metal manganese and iron oxide were loaded to SBAC. The catalytic process was dominated by the indirect oxidation reaction of hydroxyl radical and the synergistic effect of various oxidation reactions.	[76]
Xi'an fourth sewage treatment plant	Wood chips	Chemical activation method (salt activation: ZnCl ₂)	High-concentration coal wastewater	Activation temperature: 650°C; activation time: 30 min; solid : liquid ratio: 1 : 1.5	The wood chip SBAC had rich tubular pore structure, and the pore arrangement was relatively uniform and orderly, which mainly was the middle pore. The wood chip increased the carbon content of SBAC and greatly increased the area of BET and the total pore volume.	[24]
Sewage sludge in a paper mill in Zhejiang province	Loading iron and copper	Chemical activation method (acid activation: nitric acid, hydrochloric acid)	Dye wastewater: synthetic methylene blue	Pyrolysis temperature: 800°C and time: 2 h; activation conditions: 5 mol/L sulfuric acid (solid : liquid ratio : 0.1)	The SBAC-based catalytic carrier was loaded with iron and copper binary metal as a nonhomogeneous Fenton catalyst. The pore structure was developed and the surface functional groups were rich, which mainly had hydroxyl radical reaction. The pH range of catalytic carrier reaction system was greatly expanded.	[77]

TABLE 2: Continued.

Source of sludge	Modifier	Synthetical method	Treated compounds	Synthetical conditions	Results	Reference
A sewage treatment plant in Yan'an	Peanut shells, wood chips, and walnut shells	Chemical activation method (alkali activation: KOH)	Oil-containing wastewater	Activation temperature: 350°C; activation time: 40 min; 0.5 mol/L KOH infusion, mass ratio of peanut shell quality to sludge: 20%	In the preparation of peanut shell SBAC, the activation of KOH and the high temperature in carbonization process played an important role in the making of holes, so that it had better adsorption capacity. The surface was rough and had a porous structure, which also had the phenolic hydroxyl group, ether bond, lactone bond, and so on. In the activation process, the surface of the carbonized material was formed into holes and scattered short bars. The pores of the cross-section were dense and honeycombed. Most of the holes were micropores; some of which were porous, with a relatively developed pore structure and strong adsorption capacity.	[25]
A factory waste paper deinking sludge in Rizhao	Wood powder	Chemical activation method (alkali activation: KOH)	Bleaching wastewater from alkali extraction in paper mill	Mass ratio of wood powder to sludge: 20%; ratio of carbon to alkali 1 : 1; activation temperature: 675°C; the activation time: 75 min		[78]
Low organic matter dehydration sludge of a sewage treatment plant in Chongqing	Nothing	Chemical activation (acid activation: phosphoric acid); microwave activation method	Methyl-purple dye wastewater	Soaked time: 24 h; microwave power: 464 W; microwave activation time: 4 min	SBAC had a tube-shaped hole and its pore diameter was wider. SBAC had acidic surface functional groups such as the hydroxyl group and carboxyl group, which were beneficial to the adsorption of alkaline dyes.	[79]
A petrochemical refinery dries up the remaining sludge	Datong bituminous coal	Chemical activation method (salt activation: ZnCl_2)	Phenol	Activation temperature: 550°C; activation time: 30 min; mass ratio of ZnCl_2 solution and sludge: 2 : 1	The surface groups were mainly acidic groups, while the alkaline groups were less, and the choice of zinc chloride as activator was beneficial to the formation of acidic groups.	[80]
Shanghai east district sewage treatment plant	Loading manganese	Chemical activation method (acid and salt activation: ZnCl_2 , H_2SO_4)	Oxalic acid wastewater	Volume ratio of 3 mol/L H_2SO_4 to 3 mol/L ZnCl_2 solution: 1 : 2; heating rate: 10°C/min; activation temperature: 550°C; activation time: 60 min	The reaction mechanism of hydroxyl radical was consistent with the SBAC-based catalytic ozone process. Oxalic acid was adsorbed on the surface of the catalyst and reacted with the ozone molecules and hydroxyl radicals produced by the ozone molecules. The free radicals formed on the surface of the catalyst diffused into the solution and reacted with oxalic acid.	[81]
Chengdu municipal sewage treatment plant	Iron additive 铁细	Chemical activation method (salt activation: ZnCl_2)	Modeling iodine solution	Pyrolysis temperature: 400°C; pyrolysis time: 70 min	Iron compounds could promote the decomposition of carbon organic compounds that were difficult to convert, and its own decomposition could leave pores and regulated the structure of the activated carbon pore. When the additive was small, the dispersion was uneven and the local overheat was burnt into a large hole. While more was added, the catalytic activity could be increased to enhance the catalytic action, to form a micropore and enlarge the original microhole, increasing the adsorption iodine value.	[55]

TABLE 2: Continued.

Source of sludge	Modifier	Synthetical method	Treated compounds	Synthetical conditions	Results	Reference
Sewage treatment plant of Shanxi University of Science and Technology	Nothing	Chemical activation method (salt activation: ZnCl_2)	Acid red G solution	ZnCl_2 concentration of 30%; mass ratio of sludge to ZnCl_2 solution: 1 : 2; carbonization temperature: 500°C; carbonization time: 1.5 h	The SBAC surface was rough, with more porous structures and irregular porous structures. Acid red G was adsorbed on SBAC as single-layer adsorption; the regenerated SBAC had a good adsorption capacity for acid red G, and the regeneration ability was good. The heat treatment method was better than alkali treatment.	[82]
A sewage treatment plant in Lu'an city	Nothing	Chemical activation method (alkali activation: KOH)	Electroplating wastewater	Mass ratio of sludge to KOH solution: 1 : 3; activator concentration: 40%; activation temperature: 500°C; activation time: 60 min	The control of activation temperature and time could make SBAC compact microporous. KOH had a hole-forming effect on the sludge, and when the hole was saturated, too many activators could cause the channel to collapse or clog up and destroy the formed channel structure, resulting in the decrease of adsorption performance.	[83]
Sludge from sewage and of a refinery in Shijiazhuang 炼油厂	Nothing	Chemical activation method (salt activation: ZnCl_2)	Modeling iodine solution	Mass ratio of sludge to ZnCl_2 solution: 1 : 3; carbonization time: 1 h	SBAC presented an irregular porous structure, with more transition holes extending into the interior, and it had the properties of easy adsorption of macromolecular organic matter. The process of chemical activation and pyrolysis had a good effect on the hole making, which made the sludge have better adsorption capacity.	[84]
Beijing Xiaohongmen sewage treatment plant	Corn kernel	Direct pyrolysis and modification with $(\text{NH}_4)_2\text{S}_2\text{O}_8$ acid and NaOH alkali	Para-chlorobenzoic acid (p-CBA) solution	30% ZnCl_2 solution; activation time: 1 h; activation temperature: 600°C activation	NaOH-modified surface had a rich alkaline functional group, and the removal p-cba by SBAC-based catalytic ozone oxidation was enhanced. $(\text{NH}_4)_2\text{S}_2\text{O}_8$ destroyed the pore wall structure of activated carbon, so that the micropore and middle hole were expanded into large holes, resulting in the decrease in the level of BET.	[85]

difference was that NaOH could enrich the alkaline functional groups of SBAC; thus, the efficiency of the catalytic ozone oxidation on removal of p-cba could be improved.

The adsorption capacity of SBAC is related to not only the molecular size of SBAC but also the hydrophilicity and hydrophobicity between SBAC and pollutant. Karin and Li [88] studied the SBAC adsorption effect on eight hydrophobic organic compounds (HOCs) that were often detected in rainwater. Kinetic studies showed that most of the HOCs were absorbed within 10 minutes. In the batch test, the adsorption capacity was negatively correlated with the hydrophobicity of the compounds and positively associated with decreasing molecule size, suggesting that molecular sieving limited adsorption. However, in the repeated adsorption test, the competition between HOCs was more likely to occur, and the pollutant adsorption loading was positively correlated with the hydrophobicity of the compounds. Din et al. [89] used sludge and waste oil sludge to prepare bulk adsorbents for dealing with eleven antibiotics in a batch experiment. The variety of antibiotics was adsorbed on the surface of the sludge owing to its high degree of heterogeneity linked to the pores of specific sizes of hydrophobic nature and existence of the reactive/polar phase that dispersed in the pore walls. This ensured not only the accessibility of the high energy adsorption centers but also the possibility of precipitation of surface reaction products within the pore system.

The chemical activation of new activators can enhance the mesoporous structure of SBAC effectively, which also promotes the interaction of SBAC with pollutants. Yang et al. [54] showed that in a batch experiment, the iron-activated SBAC had higher adsorption capacity than the unactivated SBAC. The iron activation enhanced the mesoporous structure to facilitate the diffusion of tetracycline into the pore, and the iron oxide and oxygen-containing functional groups of SBAC could be complexed on the surface, resulting in a good effect on adsorption of pollutants. The functional groups generated by surface chemical modification can be removed by electrostatic force resulting from the adsorbed ions. Al-Malack and Dauda [90] put activated sludge under the activation temperature of 700°C for 60 mins, using 5 M ZnCl_2 as activator, and the prepared SBAC had a BET of 319.5 m^2/g . At the same time, the mechanism for adsorption of Cd^{2+} was also studied, showing that the Cd^{2+} was mainly removed by the electrostatic adsorption between the functional group with negative charge on the surface of SBAC and the positively charged metal ions. The negatively charged hydroxy or carboxyl group on the surface of SBAC played a major role in the removal of Cd^{2+} .

In the process of chemical modification of SBAC, there would appear specific metal complexes and oxygen-containing functional groups can form strong bonds with adsorbent materials. Hanyu et al. [91] used reed straw and activated sludge as raw materials to prepare SBAC under the condition of 500°C and to study its adsorption effect on the removal of norfloxacin. The results showed that there were lots of oxygen-containing functional groups in SBAC, which provided adsorption sites for the adsorption of norfloxacin. The presence of oxygen-containing functional

groups was conducive to the formation of strong hydrogen bonds between norfloxacin and SBAC, where the hydrogen bond played a dominant role in SBAC adsorption of norfloxacin. Therefore, the chemical functional groups can enhance the adsorption capacity of SBAC. Xin et al. [92] adopted the ZnCl_2 activation method to prepare SBAC, which showed that the content of Al and Fe in the oxidation form in the SBAC was higher than that of commercial active carbon, which can enhance the adsorption of SBAC to organic matter in water. Fan et al. [61] prepared SBAC with chitosan, which contained a large amount of nitrogen and oxygen-containing functional groups. The types and quantities of the desulfurization and denitration active groups in the surface of SBAC were enriched, which could help the adsorption and the oxidation of SO_2 and NO. Also, adding chitosan could significantly increase SBAC properties such as the BET value and the middle-hole volume. As a result, the physical adsorption of SO_2 and NO gas was then increased, thus improving its desulfurization and denitration performance under the appropriate condition control. Zou et al. [93] prepared a clean sludge (P-SBAC) which did not contain inorganic impurities (e.g., Si and Al) in activated sludge. The morphological characteristics, surface elements, and functional groups of SBAC were different from P-SBAC: P-SBAC had a BET and pore volume ratio of about 3 times more than that of SBAC; the oxygen-containing groups of hydroxyl and epoxide in P-SBAC were rich, which were conducive to the removal of organic pollutants from the wastewater. Furthermore, the P-SBAC had better adsorption effects in the removal of rhodamine B and phenolic compounds.

4. Applications

4.1. Organic Matter Removal. The chemical sludge containing a large amount of iron and aluminum oxide was modified to prepare SBAC, of which the effect on the removal of UV_{254} and dissolved organic carbon is similar to the activated carbon. SBAC's removal rate of UV_{254} and dissolved organic carbon could be up to 85.8% and 59.7% [94], respectively. The possible organics that can be treated with SBAC also contain toluene [95], phenol [96], nitrobenzene [97], trinitrotoluene [98], rhodamine B [99], and ibuprofen (IBP) [100]. Table 3 shows the mechanism of removing these organic matters, mainly through physic-chemical adsorption and hydroxyl radical oxidation.

The SBAC that was prepared with activated sludge mixing with other substances in a certain proportion is of higher organic adsorption performance. Xin et al. [101] prepared the SBAC using dehydrated sludge as raw materials and an appropriate amount of sawdust and coconut shell, which was activated by ZnCl_2 . The dynamic adsorption experiment of toluene showed that in the initial concentration of the same toluene, the equilibrium adsorption efficiencies were listed in a descending order: shell SBAC, coal-activated carbon, and sawdust-activated carbon. The prepared SBAC showed better adsorption performance. During the analysis of SBAC physical and chemical properties, the mesoporic and chemical adsorption was helpful to the increase in the level of adsorption. Lijun and Wenju [98] used the

TABLE 3: Organic compound removal by SBAC: preparation methods and removal mechanisms.

Organic compounds	Methods	Mechanisms	Reference
Toluene	ZnCl ₂ dipping, 700°C activation	Middle-pore adsorption in physics and chemical adsorption	[101]
Phenol, nitrobenzene	Mix of sludge and corn	Chemical acid group adsorption mainly	[22]
Trinitrotoluene red water	Phosphoric acid-microwave method, chemical modification	The surface of large hole, internal extension of more transition hole, physical adsorption mainly	[98]
Rhodamine B	Biological sludge and chemical sludge compound	The reaction of the catalytic ozone oxidation of rhodamine B following the hydroxyl radical mechanism	[102]
Ibuprofen	Zinc chloride prepared SBAC as an activator	Hydroxyl radical mechanism and physical adsorption	[103]
Oxalic acid	MnOx load modification	Surface reaction, reaction between organic pollutants and hydroxyl radical	[104]

phosphoric acid microwave method to prepare a kind of SBAC which had a large surface and abundant extension holes and studied the adsorption of SBAC to three nitrotoluene red water. The results showed that when the adsorption equilibrium time was 60 min and the total volume was 8%, the removal rate of dichromate oxidizability (COD_{Cr}) was 85.7%, of which the purification effect was better than activated carbon. Daojing et al. [22] obtained a SBAC with different proportions of corn cores and studied its adsorption to phenol and nitrobenzene. The study showed that the higher doping proportion of the corn cob, the larger the activated carbon micropore volume and the larger BET and the SBAC surface was dominated by acidic groups.

The SBAC can also be used as a catalyst or catalyst carrier to provide conditions for the preparation of new composite photocatalytic materials, and the synergetic effect of adsorption and catalysis in removal of pollutants was exhibited significantly [105, 106]. The MnOx/SBAC prepared by Huang et al. [104] has good catalytic activity. In the heterogeneous catalytic ozonation process of oxalic acid mineralization, its catalytic activity was better than single SBAC and the reaction mechanisms include surface reaction and hydroxyl radical reactions, but the surface reaction played a dominant role. Haifeng et al. [107] adopted the impregnation method to load the transition metal manganese and iron oxide onto the SBAC surface, which significantly increased the efficiency of ozone oxidation wastewater pollutants. Under optimal ozonation, the catalyst increased the ozone utilization rate by 40% and operation cost significantly.

The principles of removing organic matter may be different due to different modification methods. Hongjuan et al. [103] prepared SBAC with zinc chloride as an activator and dehydrating sludge and corn cob as raw materials. The reaction mechanisms were different in different stages of catalytic ozone oxidation of IBP. In the instantaneous oxygen-phase reaction, the mechanisms were based on the reaction of OH and IBP; in the slow reaction stage, it is mainly based on SBAC adsorption. Yangyang et al. [102] mixed biological sludge and chemical sludge together to obtain a SBAC, which follows the hydroxyl radical oxidation mechanism in the catalytic ozone oxidation of rhodamine B. Gu et al. [108] studied the properties of Fe₃O₄-separable magnetic porous carbon (F-SBAC) that were synthesized by a new type of activation

and carbonation process without additional iron ions. The properties of F-SBAC synthesized at 600°C, 800°C, and 1000°C were studied by N₂ adsorption and desorption isotherm. F-SBAC has a high surface area (407.7 m²/g) and a porous structure at 600°C. F-SBAC600, F-SBA800, and F-SBA1000 achieved 96.6%, 67.5%, and 38.9% removal rates of 1,2,4-acid. Compared with the degradation efficiency of commercially available nanomagnetic Fe₃O₄ (57.5%), F-SBAC600 showed an excellent performance to catalyze hydrogen peroxide to produce hydroxyl radicals. Sun et al. [109] studied metal-free catalyst activation sulfate peroxide, with urea as additional nitrogen source; the nitrogen-functionalized carbon sludge (N-SBAC) was prepared. The nitrogen functionalization of the N-SBAC significantly affects the chemical microenvironment as well as the microstructure (morphology and porosity), which can be effectively oxidized to remove organic contaminant.

The surface functional groups of SBAC were removed by electron donor-receptor reaction. Gupta and Garg [110] reported that the mechanism of SBAC adsorption to phenol is usually based on the electron donor receptor reaction between the aromatic phenolic rings and the functional groups on the surface of the SBAC. Al-Malack and Dauda [90] prepared a SBAC which was characterized by FTIR, indicating the existence of hydroxyl and carboxyl groups on the surface of SBAC. The reason for the decrease of adsorption capacity of SBAC to Cd²⁺ in the presence of phenol may be the competition of adsorption sites of two components on the surface of SBAC. It may also be attributed to the space resistance of the adsorbent phenol on the surface of SBAC. The removal efficiency of phenol was enhanced in the presence of Cd²⁺. That may be related to the stable effect of the SBAC surface adsorption of Cd²⁺ ions, which made the surface functional groups interact freely with the phenol ring molecules. Kong et al. [111] showed that the citric acid-ZnCl₂-mixed fabricating pore agent was a potential technique in green production of sludge-derived char with hierarchical porous for reuse of sludge. The sludge-derived char was considered as a hybrid material containing-elemental carbon, highly aromatic organic species and inorganic ash. Many kinds of benzene derivatives in aqueous solution could be treated. Multiple sorption including pore filling, hydrophobic interaction, and the stronger specific

sorption bindings between carboxyl and SiO_2 can be presented in the sorption of the porous sludge char towards carboxyl-containing adsorbates. Nunthaprechachan et al. [112] studied the effect of dibenzothiophene (DBT) removal from n-octane by a SBAC prepared with the sewage sludge of doping chemical reagents (ZnCl_2 , HNO_3 , and KOH). With the increase of oxygen-containing functional groups, especially carbonyl groups, DBT adsorption increased. SBAC prepared by KOH activation showed the highest adsorption capacity, up to 14.12 mg/g or about 70.6% DBT removal, which was about 1.22–1.28 times more than that of the commercial activated carbon.

4.2. Heavy Metal Removal. The removals of heavy metal ions with SBAC were mainly achieved through ion exchange reaction, chemical adsorption, and physical adsorption. Heavy metal ions can generate an exchange reaction on the surface of activated carbon. The surface of SBAC can be introduced to the special groups to strengthen the absorption of heavy metal ions after adding a reagent or remodification of SBAC. These surface groups form ligands with heavy metal ions and their type and stability can determine the adsorption quantity and adsorption capacity of SBAC. The SBAC adsorption of heavy metals is mainly chemical adsorption. Heavy metal ions are combined with surface functional groups thus forming adsorption products.

The adsorption mechanisms for heavy metal removal by SBAC consist of surface precipitation and ion exchange [113]. Tan et al. [113] prepared SBAC under 900°C by anaerobic pyrolysis. This SBAC has good adsorption to Pb, Zn, Cu, and Cd, and its adsorption capacity is higher than that of commercial activated carbon. When pH was buffered to a fairly high level, the heavy metal ions were converted into hydroxide and precipitated on the surface of SBAC. When pH was low, in addition to producing a very small amount of precipitation, a large number of heavy metal ions were exchanged with Ca^{2+} and subsequently adsorbed by SBAC.

The SBAC surface chemical functional groups could react with heavy metal ions as chemical adsorption, in which the acid groups of the SBAC surface can form stable ligands with heavy metal ions. Hanfeng. [114] studied the process of adsorbing heavy metals by SBAC containing loaded functional groups. The BET and micropore volume ratio of SBAC are smaller than those of coir and coal, and in addition, SBAC has a low adsorption rate. However, the equilibrium adsorption efficiencies of Cu (II), Pb (II), Cr (VI), and Cd (II) are much higher than that of commercial activated carbon, which is possibly attributed to the fact that the content of the acid group in SBAC is too high that it affects the adsorption of heavy metal ions. Weiwei et al. [115] studied the adsorption effect on the removal of Cu (II) and Pb (II) with SBAC prepared by zinc chloride activation and commercial coal carbon. It showed that the BET and pore volume of SBAC were accounted for by the 36.7% and 23.6% of commercial coal carbon, respectively. But on the surface, there was a high content of the acid functional group, its equilibrium adsorbate uptake on Cu (II) and Pb (II) was much higher than that of commercial coal carbon.

Heavy metal ions that are sedimented on the surface of activated carbon are physically adsorbed. The physical adsorption of heavy metal ions in SBAC mainly occurred on the surface. With the elapse of the adsorption process, the adsorption mass gradually got through the bigger hole and transition pores and finally arrived at the micropore. The mass transfer velocity of heavy metal ions gradually became slow among the inner bore, which eventually reached adsorption equilibrium with adsorption capacity increased gradually over time [116, 117]. Tao et al. [118] prepared a SBAC with sludge and bagasse as raw materials through pyrolysis under 800°C for 0.5 hours. After treatment with 60% HNO_3 , BET could be up to 806.57 m^2/g . The changes to pore diameter indicated that HNO_3 oxidation could enhance the adsorption capacity of metal ions.

The SBAC in the removal of heavy metals can also be coupled with the other processes. Qing et al. [119] used activated sludge as raw materials to obtain SBAC through carbonization preparation. They used the impregnation-sintering method to load nano- TiO_2 on SBAC, obtained TiO_2/AC photocatalyst, and studied its removal effect on heavy metal ion Hg^{2+} . The results showed that the adsorption performance and catalytic efficiency of TiO_2/AC photocatalyst were high and the Hg^{2+} removal rate of 20 mg/L aqueous solution was 88.5%.

4.3. Gas Pollutant Removal. SBAC has the characteristics of uniform small particles, even pore distribution, high BET, and so on, whose surface loads a certain amount of active component. Its functional groups are rich, crystal surface content increases, and the crystal structure becomes more orderly [120]. Those are beneficial to contact with the reaction gas. Qingbo et al. [121] studied the formaldehyde adsorption properties of SBAC and commercial activated carbon. The results showed that SBAC contained a large number of ultramicropore, micropores, and various nitrogen-containing groups, which form a chemical adsorption center, which were conducive to the adsorption of low-concentration formaldehyde. SBAC has a good adsorption effect on formaldehyde in the air, and the maximum removal rate can reach up to 83%, which is the same with that of commercial activated carbon. SBAC can be used as an important carrier, loading catalyst for removing organic matter. Tao [122] studied the effect of SBAC loading nitric acid iron sludge-based catalyst on NO_x gas, and the maximum conversion rate of NO_x was 98.3%. Yanjing [123] took SBAC as the carrier of the TiO_2 photocatalyst, and the TiO_2/SBAC photocatalytic degradation of acetone gas achieved good results.

Wei et al. [124] adopted ZnCl_2 to prepare SBAC as an active agent, which was modified with cerium. It was utilized in H_2S removal. Their study showed that the SBAC deodorization performance was greatly improved, no matter modified by cerium doping or loading. After modification, the SBAC surface was still the middle hole, but its aperture was smaller and the surface functional group increased, making it more favorable for H_2S material adsorption and catalytic reaction, which was beneficial to H_2S gas removal. Boualem et al. [17] used sludge to chemically react with phosphoric

acid to prepare SBAC. The surface area of the activated carbons was at $300\text{ m}^2/\text{g}$. The activated carbons were mainly mesoporous. SO_2 adsorption capacity was associated with average micropore size, which could be controlled by the impregnation ratio used to prepare the activated carbons.

4.4. Others. The SBAC not only can be used alone but also can be coupled with other water treatment technologies to increase its application prospect thus reducing the operational cost. The BET of SBAC prepared by Dezhi et al. [125] was $397.9\text{ m}^2/\text{g}$. Combining it with membrane bioreactor to treat the waste leachate, SBAC could improve the properties and structure of the cake layer on the membrane surface, so that the membrane had higher filtration performance and water permeability. It has the advantages of reducing membrane fouling and protein as well as humic acid, lengthening membrane operation cycle, and lowering operation costs. Zhai et al. [126] studied the effect of SBAC on sludge liquefaction. It showed that SBAC was conducive to the increase of the yield and energy density of bio-oil at 350°C (denoted as 350-SBAC at this temperature). 350-SBAC was beneficial to lowering the risk of Cu, Zn, and Pb, while 400-SBAC was effective in lowering the risk of Cd, Cu, and Zn. 400°C was preferable for lowering the risk of SR, while special attention needs to be paid to Cd. Considering the bio-oil yield, the use of liquefaction at 350°C with SSAC-550 was preferable.

4.5. Regeneration of SBAC. Recycling of activated carbon is of great significance for environmental protection and resource conservation. Regeneration of SBAC is the use of physical and chemical means to restore the adsorption capacity of SBAC saturated with various pollutants without destroying the original activated carbon structure. At present, the regenerative technologies of activated carbon mainly include the thermal regeneration method [127], wet oxidation regeneration method [128], microwave regeneration method [129], and high-frequency ultrasonic regeneration method [130]. For a lower regeneration cost and an available regeneration cycle, the selection of the methods for adsorption, degradation, and regeneration is bound to be affected by the types and properties of pollutants. Recycling of sludge activity should take into account the cost of recycling and reparation of sludge-activated carbon, the loss of activated carbon during regeneration, regenerative adsorption capacity, secondary pollution, and other issues. For sludge-activated carbon adsorption material recovery, based on the nature of the material being adsorbed, to determine the recovery method, the regeneration mechanism of sludge-based activated carbon needs to be further explored. The safety issues during regeneration are worth considering. Parts of the regeneration methods are still in the experimental stage and can not be widely applied to production practice.

5. Summary

Sewage treatment produce a lot activated sludge. If the sludge is prepared into SBAC, it can reduce environmental pollution and bring considerable economic value. Compared with the

traditional activated carbon, the preparation cost of SBAC is lower and there is a wide range of source. Therefore, the research and application of SBAC have potential value. At present, the research and application of SBAC have obtained a certain achievement. However, some problems still remain to be solved in the preparation and application of SBAC, which needs further process. First, potential release of some toxic and hazardous substances during the preparation and preparation of SBAC were produced. For example, the heavy metals are possible to be released from SBAC. The transformation and its mechanisms for soluble heavy metal into insoluble metal compound are still not clear in preparation of SBAC. Second, the environmental effects of SBAC need to be further studied such as the disposal of waste SBAC, the leakage of its adsorbed substance in the transfer, the effective recycle and reuse of SBAC adsorption materials, the regeneration technology, and regeneration performance comparison between SBAC and commercial activated carbon. Third, the reaction mechanisms for SBAC preparation need deeper study. Because of the complexity of sludge composition and the influence including pyrolysis conditions and pyrolysis equipment and other factors, in the preparation process of activation, the organic matters in activated sludge can produce chemical reaction due to the activation by temperature. Meanwhile, the additives and chemical activators make chemical reaction more complicated. Therefore, studying the variations of the activation process and activation mechanism can give us guidance in the preparation, modification, and application of SBAC at a deeper level.

Conflicts of Interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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