

Review Article Efficiency of Sawdust as Low-Cost Adsorbent for Dyes Removal

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In recent years, the removal of dyes from wastewater has attracted considerable attention due to their harmful effects to ecosystem and human health. Adsorption as a facile and effective technique has been widely used to eliminate a large variety of dyes from aqueous solutions. Activated carbon is the most preferred adsorbent to treat wastewater but its use is limited because of high cost. Therefore, several low-cost and natural materials and wastes have been used as precursors for the preparation of alternative adsorbents. Among them, sawdust as an abundant and low-cost by-product has been explored as adsorbent for the removal of dyes from wastewater. This review focuses on the various sawdust used as a precursor for the preparation of activated carbons. Extensive literature information about sawdust, its compositions, activation methods, its efficiency for dyes removal, and environmental conditions effects has been reviewed. The applicability of various adsorption kinetic models and adsorption isotherm models for dye removal by sawdust-derived activated carbons has been also reported. Finally, this paper highlights the use of sawdust as base material for various composites and mixture which can be used as granular activated carbon.

1. Introduction

Industrial development has increased the amount of effluent discharged into the environment. Water as an essential constituent of the planet has been affected by various pollutants such as dyes, heavy metals, organic substances, pesticides, etc. [1, 2]. Dyes are being extensively used in many fields such as paper, leather, rubber, textile, plastics, etc. [3, 4]. It is approximated that more than 100,000 commercial dyes are industrially used and more than 7×10^5 tons of dyestuff is produced every year worldwide [4]. The release of large amounts of synthetic dyes to the water streams has posed a serious risk to the environment [5]. The presence of very small amounts of dyes in water (less than 1 ppm for some dyes) is highly visible and undesirable [6]. Most dyes molecules are complex and stable and can be resistant to degradation upon contact with water, detergents, or any other thing [7].

In order to remove the dyes from the wastewater, a wide range of methods are applied such as adsorption, coagulation and flocculation, ion exchange, membrane filtration, advanced oxidation process, electrochemical process, etc.

[8, 9]. Adsorption is one of the most effective widely used wastewater treatment methods to reduce hazardous pollutants present in industrial effluents. Commercial activated carbon is a preferred adsorbent for dye removal, but its use is limited due to its relatively high cost and its regeneration problems [5, 9]. In recent years, attention has focused on various natural, nonconventional, and low-cost adsorbents. Many researchers have developed novel adsorbents using various precursors such as clays, zeolites, biosorbents, agricultural solid waste, industrial by-product, etc. [8, 10-13]. The lignocellulosic materials are the most common precursor materials to obtain low-cost activated carbon [14]. The selection of each precursor should be based on its availability, its low cost, and its abundance. Thus, sawdust as a lignocellulosic material, freely accessible and abundant (up to 24.15 million m³ per year worldwide [15]), can be utilized as a good precursor for alternative activated carbon.

Up to now, numerous reviews have been published in the area of the dyes removal from wastewater by various adsorbents [4–6, 8, 9]. But none of them specifically addressed the efficiency of sawdust as low-cost adsorbent for dyes removal. Therefore, a review focusing on the different

activated carbons produced from sawdust and their adsorption performance of dyes is still needed. In this review, dyes and their removal techniques and the different adsorbents used for dyes removal are presented and described. The aim of this paper is to provide a summary of up-to-date progress of various sawdust used for dyes removal. Thus, the chemical and elemental compositions of this material are identified. Then, the preparation methods and applications of sawdust-derived activated carbon are summarized and compared. Additionally, the effects of environmental conditions such as the adsorbent dosage, the initial dye concentration, the contact time, the temperature, and the solution pH on the dyes removal process are discussed. In the end, we summarized the kinetic and isotherm adsorption studies usually used to evaluate the adsorption capacity of various adsorbents. We draw attention to novel composites and mixture developed using sawdust as a base material to remove different dyes.

2. Dyes

Dyes as colorful substances have been utilized by humans since 3500 BC in various applications using natural extracts of flowers, fruits, certain insects, etc. These natural dyes constitute a very limited range of colors and are produced in low quantities. However, after the discovery of synthetic colors by W. H. Perkins in 1856, a wide range of dyes are used in various fields to color their product such as paper, leather, rubber, textile, plastics, etc. [3, 4]. Synthetic dyes are developed and have replaced natural dyes gradually in different industries because their molecules are stable and can resist degradation upon contact with water, detergents, or any other washing agents [7]. As shown in Table 1, dyes can be classified based on their chemical structure into azo, anthraquinone, indigoid, nitroso, nitro, and triarylmethane dyes [5]. Sometimes, they are classified by their application or by their solubility in water; vat, azoic, sulfur, and disperse dyes are insoluble, while reactive, direct, basic, and acid dyes are highly soluble [16]. Figure 1 presents types of dyes used in different industries.

As we all know, dyes are indispensable to modern life. However, their discharge as industrial waste into water body caused a threat to living things in the environment [18]. Dyes may affect the photosynthesis of aquatic life due to decreased sunlight transmission [19]. Furthermore, most of these dyes are harmful and potentially carcinogenic, mutagenic, or teratogenic for aquatic organisms. They can also cause severe damage to human beings such as dysfunction of the kidney, reproductive system, liver, brain, and central nervous system [8, 20, 21]. Additionally, synthetic dyes are chemically stable and resistant to degradation even when exposed to extreme heat sources, oxidizing agent, or strong light [22]. For these reasons, highlighting efficient treatment methods of dye wastewater is necessary.

2.1. Dyes Removal Methods. Ever since the negative effects of dyes were identified by scientists. A wide range of methods has been developed for the removal of synthetic dyes from

waters and wastewaters to decrease their impact on the environment. Thus, all the biological, physical, and chemical methods are applied such as biodegradation, adsorption, coagulation or flocculation, ion exchange, membrane filtration, reverse osmosis, advanced oxidation process, electrochemical process, etc. Table 2 presents the main advantages and disadvantages of these methods.

The biological treatment is the commonly and extensively utilized dyes removal method to decolorize wastewater. Numerous microorganisms are used to treat effluents loaded with synthetic dyes under aerobic or anaerobic or mixed conditions [24]. However, the biological process can be complex. In fact, most of these dyes compounds are chemically stable and resistant to microbiological degradation [25]. The efficiency of this method is often not satisfying due to production of excess sludge and takes long time.

Chemical dyes removal methods may be ineffective due to the interaction between the contaminants in the wastewater and the nature of chemical used. These methods are efficient at decolorizing water and, at the same time, are costly because they will consume large amounts of chemical reagents and electrical energy. Additionally, the production of sludge may add to the cost of the process [24]. Moreover, secondary pollution problem can arise due to toxic chemical resulting at the end of a chemical dyes removal process [26]. For all these reasons, these methods are commercially unattractive.

Physical dye removal methods are very extensive including membrane filtration and adsorption. Membrane filtration is an efficient technique including microfiltration, ultrafiltration, nanofiltration, and reverse osmosis [27]. Membrane filtration processes are associated with few disadvantages; for example, the service life of membrane is short and it is easy to be polluted and costly as well [27]. From the literature, adsorption is one of the most widely used wastewater treatment methods which has been shown to be effective and efficient in the science of wastewater remediation because it can remove contaminants and discolor water resources [28, 29]. Activated carbon adsorption has been cited by the U.S. Environmental Protection Agency as one of the best available control techniques. However, its use is restricted by its high cost [6]. In recent years, attempts have been made to produce low-cost adsorbents from the agricultural by-products and industrial wastes for dye removal [30-32].

2.2. Adsorption. In general terms, adsorption refers to the accumulation of adsorbate at the interface between two phases (liquid-solid interface or gas-solid interface). There are two types of adsorption: chemical adsorption (chemisorption) and physical adsorption (physisorption) [33]. The difference between the two types of adsorption is that chemical adsorption involves the formation of strong chemical associations between adsorbate and adsorbent surface through electron exchange [24]. On the other hand, physical adsorption is characterized by weak Van der Waals or hydrogen bonds between adsorbate and adsorbent [34].

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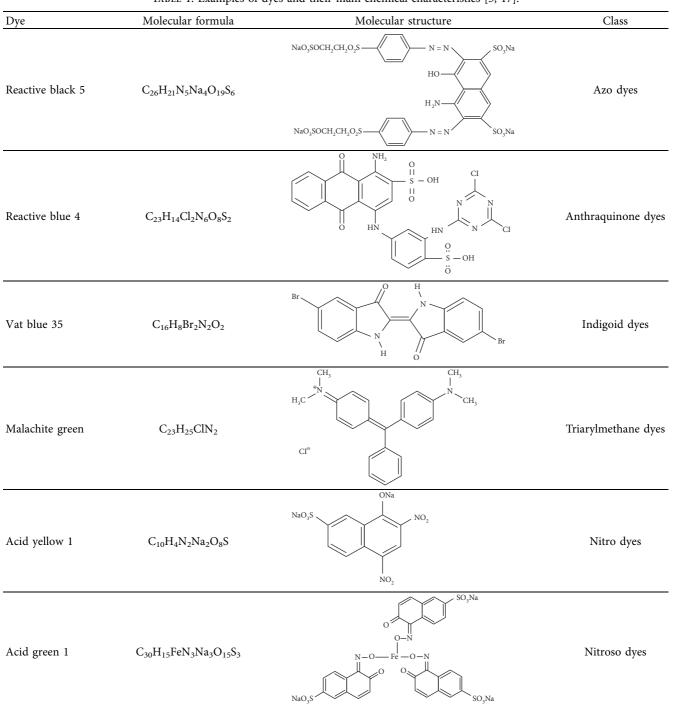


TABLE 1: Examples of dyes and their main chemical characteristics [5, 17].

Often, adsorption on most of the adsorbent is controlled by physical forces with minor exceptions of chemisorption. In adsorption processes, the adsorbent retains dye particles (adsorbate) from dye solution hence accumulating it on its surface [35]. After a certain period of treatment time, the saturated adsorbent will have to be regenerated/replaced depending on the feasibility of regeneration [24].

Katheresan et al. [35] reported that five factors can affect the rate of adsorption which are the adsorbent dosage, contact time, the dye concentration, the solution pH, and the temperature of the solution; they found the following:

- (i) The rate of adsorption increases with increasing of adsorbent dosage and vice versa
- (ii) The rate of adsorption can be high if contact time is long and vice versa
- (iii) High rate of adsorption is found if dye concentration is low and vice versa
- (iv) Low rate of adsorption is obtained when pH is low and vice versa
- (v) Low rate of adsorption is obtained if temperature is too low or too high

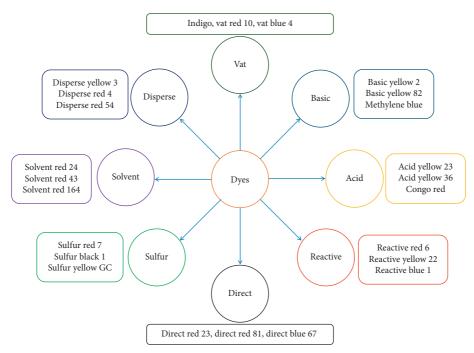


FIGURE 1: Types of dyes used in different industries [17].

TABLE 2: Different dyes removal methods along with their advantages and disadvantages [8, 9, 23	TABLE 2: Different	dyes removal	methods alo	ong with th	heir advantages	and disady	vantages [8, 9, 23]
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Method	Advantages	Disadvantages
Biodegradation	Simple, economically attractive	It requires strict environmental conditions, produces sludge, and takes long time
Adsorption	Excellent removal method for a wide variety of dyes, and simple operation process	Adsorbents can be costly and have regeneration problems
Coagulation/ flocculation	Economically attractive, simple	It generates huge amounts of concentrated sludge
Ion exchange	Can be regenerated, good dye removal method	It is ineffective to certain dyes and costly
Membrane filtration	Effective for water recovery and reusing	It has short lifetime, produces concentrated sludge, and is costly
Reverse osmosis	Effective for decoloring and desalting a variety of dyes	It requires high pressure and is expensive
Advanced oxidation process	Can remove dye in unusual conditions, good dye removal method	It is expensive and produces undesirable by-products
Electrochemical process	Effective, rapid	There is high cost of electricity and it is economically unfeasible

Yagub et al. [5] reported that many factors affect dye adsorption such as solution pH, temperature, amount of adsorbent, and initial dye concentration. They found the following:

- (i) At pH < pH_{pzc} (the pH at which the surface charge of adsorbent is zero), the adsorption of anionic dyes is favored, while the adsorption of cationic dyes is favored at pH > pH_{pzc}, where the surface charge becomes negative.
- (ii) If the adsorption process is endothermic, the amount of adsorption increases with increasing temperature, whereas it decreases with increasing temperature if the adsorption process is exothermic.
- (iii) The percentage of dye removal increases with increasing adsorbent dosage.

(iv) The percentage of dye removal decreases with an increase in the initial dye concentration.

3. Adsorbents for the Removal of Dyes from Aqueous Phase

In recent years, there has been an increasing amount of literature on the study of adsorbents for adsorption dye removal. A good adsorbent should have a high adsorption capacity and high surface area [36]. Additionally, a short adsorption period and adsorbent diversity in removing a wide range of pollutants are also desired factors for the selection of an adsorbent [37]. Thus, a selected adsorbent should be able to function in varying dye concentrations and a wide range of pH as well as temperature. 3.1. Activated Carbon as Adsorbent. Nowadays, activated carbon as a carbonaceous solid is the most commonly adopted adsorbent. It can be produced from any material containing high levels of carbon in its composition. Activated carbon is extensively used for the removal of a large variety of dyes from wastewater [32, 35, 37]. However, activated carbon poses several disadvantages; for example, it is quite expensive (about US\$ 2.0–2.2/kg) [8], has a problem in regeneration, and is nonselective [38].

Due to the mentioned problems previously, more recent attention has focused on the production of alternative adsorbents to replace the costly activated carbon. Because of their low cost and local availability, numerous natural materials and certain waste products from industrial operations are classified as low-cost adsorbents.

3.2. Low-Cost Adsorbents. Over the past decades, a number of researchers have been gathering considerable research papers on low-cost adsorbents and held discussions in reviews to prove the existence of cheap as well as effective adsorbents [4, 6, 8]. In order to develop a low-cost adsorbent from a given precursor, many factors should be considered. The precursor should preferable be freely available, inexpensive, and nonhazardous for nature [5]. The cost is an important parameter for the comparison and selection of adsorbents. An adsorbent can be assumed as "low-cost" if it requires little processing and is abundant in nature [6]. Many low-cost adsorbents have been used for the removal of dyes. Also, some waste products from industrial and agricultural operations, natural materials, and biosorbents represent potentially economical alternative adsorbents. Many of them have been tested and proposed for dye removal. Sawdust as an abundant by-product of the wood industry with negligible price is one of the most widely used adsorbents; it has been tested and proposed for dye removal. Table 3 reports the list of low-cost adsorbents used for dyes removal.

4. Sawdust

Nowadays, sawdust plays an important role in the adsorption of wastewater pollutants, because it contains numerous functional groups such as carboxyl, hydroxyl, phenolic, and amide groups in its structure, which may be favorable for adsorbing a large variety of dyes [45]. Furthermore, sawdust can be modified by acid and alkali to increase its adsorption properties. A considerable amount of literature has been published on the use of wood sawdust: its composition, its preparation methods, and its capacity to remove dyes from effluents [21, 46–48].

4.1. Chemical Composition of Sawdust. Sawdust as a lignocellulosic material can be utilized as a sustainable precursor to produce activated carbon. In fact, the major chemical constituents of sawdust are hemicellulose, cellulose, and lignin [12, 15, 49]. On average, the quantitative percentages of hemicellulose, cellulose, and lignin in the sawdust are observed in the range of 15–35, 35–60, and 15–30%,

TABLE 3: List of low-cost adsorbents used for dyes removal.

Adsorbents	Dye name	References
Activated clay	Methylene blue	[39]
Rice husk	Direct red 23	[5]
Walnut cake	Methylene blue	[40]
Chitosan	Acid orange 10	[41]
Powdered marble	Methylene blue	[11]
Sugar scum using	Methylene blue	[31]
Prickly pear seed cake	Methyl orange	[32]
Argan shells	Methylene blue	[42]
Tectona grandis sawdust	Crystal violet	[43]
Terminalia arjuna sawdust	Crystal violet	[44]

respectively [13, 46, 50]. Tarrés et al. [46] have determined the composition of *Eucalyptus* sawdust as raw material and they found that hemicellulose accounts for 20.65%, cellulose accounts for 52.39%, and lignin accounts for 24.09%. Similarly, Azargohar et al. [51] have studied the composition of sawdust collected in Saskatchewan (Canada) and they observed that cellulose, hemicellulose, and lignin are the major components with 37.7 wt%, 31.4 wt%, and 15.9 wt%, respectively. As shown in Figure 2, Rusanen et al. [52] have determined the composition of pine sawdust and they found that hemicellulose accounts for 26%, cellulose accounts for 44%, and lignin accounts for 26%.

Due to the important role that chemical composition plays in the formation of pores, pore volume, and surface area, more attention has focused on the determination of sawdust chemical composition. The elemental compositions of various sawdust are shown in Table 4.

4.2. Preparation Methods of Sawdust. Generally, there are two different methods for activated carbon preparation: chemical and physical activation methods [47, 48]. Physical activation treatments consist of two steps: in the first, precursors will be carbonized in presence of inert atmosphere such as nitrogen or helium, whereas in the second step, the carbonized substance is placed in air, oxygen (O₂), carbon dioxide (CO₂), steam, or the mixed atmosphere at much elevated temperature. On the other hand, in chemical treatments, precursors are impregnated by an activating reagent, followed by heating process under an inert atmosphere [59–63].

In physical activation, the purpose of the carbonization is to eliminate the volatile matters at relatively low temperature (400–700°C) from the raw material and then to convert the resulting char with higher content of fixed carbon for activation purpose. At high temperature (800–1200°C), activation treatment with oxidizing gas takes place to form carbon oxides, and pores are created inside the carbon material by releasing gas [47, 48, 64].

Chemical activation method refers to the impregnation of the raw material with a dehydrating chemical agent and heating at a high temperature under inert atmosphere. A variety of chemical activating agents such as potassium hydroxide (KOH), zinc chloride (ZnCl₂), phosphoric acid (H₃PO₄), sulphuric acid (H₂SO₄), nitric acid (HNO₃), potassium carbonate (K₂CO₃), etc. can be used [13, 65–68]. The

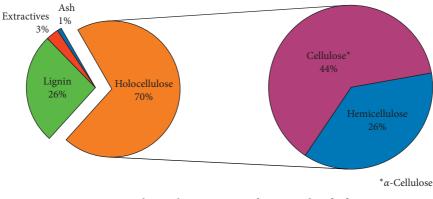


FIGURE 2: Chemical composition of pine sawdust [52].

Sawdust		D.C			
Sawdust	С	Н	0	Ν	References
Pine sawdust	46.41	6.27	47.23	0.06	[53]
Meranti sawdust	42.38	5.27	42.41	0.14	[54]
Chinese fir sawdust	48.95	6.54	53.74	0.11	[55]
Rubber-wood sawdust	44.0	8.04	47.5	0.45	[56]
Sawdust (Canada)	45.2	6.7	48.0	0.1	[51]
Sawdust (Colombia)	48.5	6.6	44.9	_	[57]
Red wood sawdust	27.1	_	72.5	_	[15]
Parkia biglobosa sawdust	57.62	_	35.93	_	[58]

TABLE 4: The elemental compositions of various sawdust.

utilization of chemical agents aims to facilitate the development of the pores in the activated carbon using degradation, dehydration, and complexation with organic carbon molecules of precursor materials [12].

Unlike the physical activation, the chemical activation can be carried out at lower temperature and does not need the participation of oxidizing atmosphere, but it requires an additional step of flushing activated carbon with hot and cold water to remove the unreacted chemicals and chemical by-products [69]. The elimination of chemicals from the carbon structure by washing will develop the porosity in resulting activated carbon [70]. The diagram is shown in Figure 3. Generally, both activation methods have been used by researchers to obtain the best activated carbon from sawdust as low-cost precursor. Table 5 presents various activation conditions of sawdust.

4.2.1. Activation Process. Among the various dehydrating agents used in chemical activation, $ZnCl_2$ and H_3PO_4 are the most important and commonly used for activation purpose for lignocellulosic materials [83]. Chemical activation by $ZnCl_2$ enhances the pore development in the carbon structure and the yields of carbon may be usually high because of the effect of chemicals [70]. Mohanty et al. [73] reported large surface area of an activated carbon prepared from *Tectona grandis* sawdust by zinc chloride activation. Further, in order to know the effect of chemical impregnation ratio on the surface area and total pore volume, the experiments were carried out for different chemical ratio (100–300%) at carbonization temperature 500°C and

carbonization time 60 min. The BET surface area as well as the pore volume increased with the increase in chemical ratio. The weight loss of the carbon products decreased with the addition of chemical agents to the precursor, resulting in an increase of the carbon yield. They concluded that the best conditions for the production of high surface area activated carbon from *Tectona grandis* sawdust by chemical activation are chemical ratio (activating agent/precursor) of 200%, carbonization time of 1 h, and carbonization temperature of 500°C. At these optimal conditions, the BET surface area and micropore volume obtained were 585 m²/g and 0.442 cm³/g, respectively.

Zhang et al. [84] have prepared activated carbon from sawdust by chemical activation using zinc chloride as an activating agent. In order to optimize operation conditions, they studied the effects of operation parameters such as impregnation ratio, activation temperature, and time on the adsorption properties of activated carbons. The authors reported that the impregnation ratio ZnCl₂/sawdust was the most important operation parameter and the activation temperature was the second variable which had significant effect on the yield of carbon and the adsorption capacity of prepared activated carbon for iodine and methylene blue. The yield of activated carbon increases with increasing the impregnation ratio from 50-100% at 500°C activation temperature. However, a further rise in impregnation ratio (100-300%) did not have any noticeable effect on activated carbon yield. The effect of activation time and temperature on yield showed that the increase in these parameters leads to a decrease of yield. The similar adsorption behaviors for iodine and methylene blue of activated carbon prepared in

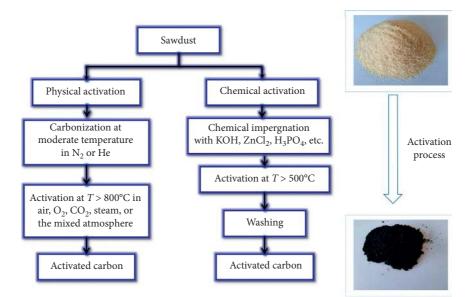


FIGURE 3: Diagram for activation process of sawdust-derived activated carbon.

Raw material	Carbonization condition	Activation condition	Chemical treatment	Impregnation ratio	Activation method	Ref.
Pine sawdust	450°C	_	H_3PO_4	_	Chemical activation	[71]
Mahogany sawdust	500°C/1 h	800°C/1 h	Steam	_	Physical activation	[72]
<i>Tectona grandis</i> sawdust	500°C/1 h	_	$ZnCl_2$	200%	Chemical activation	[73]
Pine sawdust	_	500°C	$ZnCl_2$	1:1	Chemical activation	[74]
Teak sawdust	600°C/1 h	750–900°C/h	Steam	_	Physical activation	[75]
Rattan sawdust	700°C	_	KOH	1:1	Chemical activation	[76]
Chinese fir sawdust	400–600°C/1 h	_	$ZnCl_2$	_	Chemical activation	[77]
Pine sawdust	_	800°C/2 h	CO_2	_	Physical activation	[78]
Sawdust of Algarroba wood	_	800°C	CO_2	_	Physical activation	[14]
Pine sawdust	400°C	_	$ZnCl_2$	0.5-2	Chemical activation	[53]
Rubber-wood sawdust	_	500°C/1 h	$ZnCl_2$	1:1-2:1	Chemical activation	[79]
Rubber-wood sawdust	740°C/1 h	740°C/1 h	CO ₂	_	Physical activation	[80]
<i>Eucalyptus grandis</i> sawdust	_	_	$ZnCl_2$	2:1	Chemical activation (microwave-assisted)	[81]
Pine sawdust	450°C	—	H_3PO_4	_	Chemical activation	[71]
Sawdust (Colombia)	600°C	900°C	CO ₂	_	Physical activation	[57]
Sawdust	425°C/2 h	—	H ₃ PO ₄	15 wt%	Chemical activation (ultrasound-assisted)	[82]

TABLE 5: Carbonization and activation condition of various precursors.

different operation variables had been observed. The adsorption of iodine and methylene blue by activated carbon can reach a maximum value in each case of different impregnation ratio, different activation time, and/or different activation temperature. Finally, they concluded that the better adsorption properties of activated carbon can be obtained in the zinc chloride activation process under the optimum operation conditions with the impregnation ratio of 100% ZnCl₂/sawdust at the activation temperature of 500°C for activation time of 60–90 min.

As described above, chemical activation with $ZnCl_2$ is efficient in producing activated carbon with microporous

structure, higher surface area, and higher yield. However, when compared to phosphoric acid, zinc chloride could be unfriendly for environment. Moreover, the activated carbon obtained using $ZnCl_2$ cannot be used in food and pharmaceutical industries [85]. H₃PO₄ as an activating agent helps in the formation of micropores and mesopores in activated carbon. It also gives activated carbon with higher yield and has nontoxic properties [40, 86].

Zhang et al. [87] reported that H_3PO_4 is widely used as an activating agent in the preparation of activated carbon from lignocellulosic products because of its nonpolluting character compared to $ZnCl_2$ and its easy elimination by washing with water. Therefore, the authors have prepared activated carbons from sawdust by chemical activation with H₃PO₄. Then, they studied the effects of various operational parameters such as the impregnation ratio, activation temperature, and time on the nitrogen adsorption isotherms, specific surface areas, and pore volumes of the activated carbons. It was observed that the H₃PO₄/sawdust impregnation ratio (R) was the most important operational parameter in chemical activation with phosphoric acid. At low values of the impregnation ratio (R < 1.0), phosphoric acid essentially promotes the development of micropores. When *R* increased (R > 1.0), some of the micropores were converted into mesopores and some of the mesopores were also converted into macropores, resulting in decrease of BET surface area. The high value of BET surface area was $1642 \text{ m}^2/\text{g}$ for R = 1 at activation temperature 500°C for 90 min. Additionally, better nitrogen adsorption properties were found for activated carbons prepared with R = 1.0-1.5heated at an activation temperature of 500°C for 60-90 min.

4.3. Applications of Sawdust-Derived Activated Carbons in Dyes Removal. Over the past few decades, sawdust has been used as precursor to produce low-cost activated carbon with high porosity, large surface area, and various functional groups for different applications. In the field of dyes removal, activated carbons derived from sawdust have been studied by several researchers and research groups for the elimination of various types of water-soluble dyes such as methylene blue, congo red, methyl violet, malachite green, etc. Table 6 summarizes applications of sawdust-derived activated carbons in dyes removal. It was observed that the adsorption capacity of sawdust varied between 3.42 and 526.3 mg/g. This difference is due to the operating conditions, the adsorbent nature, and the dye molecules. Ferrero [88] reported that the amount of methylene blue adsorbed per unit gram of adsorbent increases from 27.78 to 59.17 mg/g by varying the adsorbent under the same operating conditions. Similarly, the adsorption capacity of walnut sawdust varied from 36.98 to 59.17 mg/g for acid blue 25 and methylene blue, respectively, under the same operating conditions.

4.4. Factors Affecting Adsorption of Dyes. The adsorption process of dyes by sawdust-derived activated carbon can be affected by many factors such as the adsorbent dosage, the initial dye concentration, the contact time between the adsorbent and the adsorbate, the temperature of the solution, and the solution pH. Several researchers have studied the effects of these parameters on adsorption capacity. To ensure achievement of the better capacity of adsorption, optimization of such conditions should be considered when conducting lab-scaled experiments.

4.4.1. Effect of Adsorbent Dosage. Adsorbent dosage is an important process parameter to determine the capacity of an adsorbent for a given amount of the adsorbent at the operating conditions. Generally, the percentage of dye removal

increased with an increase in adsorbent mass. However, the amount of molecules adsorbed per unit mass of adsorbent decreased [111]. Increase in the adsorption with adsorbent dosage can be attributed to the increased adsorbent surface area and the availability of more adsorption sites [107].

Gupta and Lataye [110] studied the adsorption of crystal violet dye by *Acacia nilotica* sawdust activated carbon and they found that the percentage of dye removal increases when the adsorbent dosage increases up to 8 g/L. Further, increasing the adsorbent dosage from 8 to 20 g/L results in a marginal increment in removal percentage and, beyond that, the removal of dye is almost constant and unaffected by an increase of adsorbent dose. Aljeboree et al. [107] have obtained similar results from adsorption of Maxilon blue dye into Fugas sawdust. They found that the percentage of dye removal increased from 45 to 85% with an increase in adsorbent dosage from 0.3 to 1 g/L at temperature 303 K, pH 6.7, contact time 1 h, and initial dye concentration 10 mg/L, while the adsorption capacity decreased from 17.5 to 8 mg/g with same increase in adsorbent dosage.

4.4.2. Effect of Initial Dye Concentration. The effect of initial dye concentration C_0 on the removal of dyes depends on the immediate relation between the concentration of the dye and the available sites on an adsorbent surface. Generally, the percentage of dye removal decreases with the increase in the initial dye concentration, although the amount of dye adsorbed (mg/g) increased with increased dye concentration. This may be due to the high driving force created by the high initial concentration to overcome all mass transfer resistance of the dye between the aqueous and solid phases [100].

Agarwal et al. [112] studied the effect of initial dye concentration on the adsorption of methylene blue (MB) by *Ephedra strobilacea* sawdust (ESC) and modified using phosphoric acid (ESP) and zinc chloride (ESZ) and they noticed that, in concentration range from 30 to 100 mg/L, the removal efficiency of MB decreases from 97.19, 99.17, and 97.96% to 74.42, 76.2, and 85.55% for ESC, ESP, and ESZ adsorbents, respectively. The authors explain that by the saturation of the adsorbent surface or possible repulsive force between adsorbed layers and remaining bulk molecules.

4.4.3. Effect of Contact Time. The equilibrium time is a very important parameter in study of adsorption process. Traditionally, there are three periods of time for adsorption process. In the first, the adsorption process is very rapid and the rate of removal is higher because of the large amount of adsorbate attached to the adsorbent; this can be attributed to the presence of a larger surface area presented by the adsorbent and the availability of free active sites at the beginning time for the adsorption of adsorbate. In the second time, the adsorption rate gradually becomes relatively slower with the increase of contact time and, at some point in time, reached a constant value where no more adsorbate was removed from the solution. Finally, the process attains the

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TABLE 6: Adsorption capacities of ac	ivated carbons prepared from	various sawdust for various dyes.
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0 1 /	D		Opera	ating condit	ions		()	D.C
Sawdust	Dye	<i>m</i> (g/L)	$C_0 \text{ (mg/L)}$	t (min)	T (°C)	pН	$q_m (mg/g)$	Ref.
Wood sawdust	Basic blue 69	2	200	_	80		74.4	[89]
Wood sawdust	Acid blue 25	2	200	240	_	_	7.53	[89]
Neem sawdust	Crystal violet	5	12	_	30	7.2	3.789	[90]
Neem sawdust	Malachite green	5	12	_	30	7.2	3.42	[90]
Bamboo Sawdust	Methylene blue	10	100-400	35	30	7.4	143.2	[91]
Coconut tree sawdust	Congo red	2.5	25	3600	31	5.76-5.91	70%	[21]
Coconut tree sawdust	Methyl violet	2.5	25	60	31	4.68 - 4.78	93%	[21]
Coconut tree sawdust	Malachite green	2.5	25	180	31	4.70-4.86	94%	[21]
Treated sawdust	Malachite green	4	250	60	26	7	74.5	[92]
Mahogany sawdust	Acid yellow 36	4	1000	60	30	3	183.8	[93]
Mahogany sawdust	Direct blue 2B	0.6	_	150	30	3	518	[72]
Mahogany sawdust	Direct green B	0.6	_	150	30	3	327.9	[72]
Beech sawdust	Methylene blue	_	1.4-14.0	_	_	8-13	16.05	[94]
Beech sawdust	Red basic 22		2.1-21.0	_	_	8-13	29.1	[94]
Raw beech sawdust	Methylene blue	_	1.4–14.0	_	_	8-13	9.78	[94]
Indian Rosewood sawdust	Methylene blue	4	250	30	26	7	46.1	[95]
Pine sawdust	Malachite green	4	50-2000	120	26	, 	370.37	[74]
Pine sawdust	Acid yellow 132	10	500-2000	120	25	3.5	398.8	[96]
Pine sawdust	Acid blue 256	10	500-2000	120	25	3.5	280.3	[96]
Cedar sawdust	Methylene blue	0.1-1.5	40	300	20	7	142.36	[97]
Rattan sawdust	Methylene blue	1	500	240	30	, 	294.12	[76]
Walnut sawdust	Methylene blue	10	50-500	60	20	2.5-4.2	59.17	[88]
Walnut sawdust	Acid blue 25	10	50-500	60	20	2.5-4.2	36.98	[88]
Cherry sawdust	Methylene blue	10	50-500	300	20	2.5-4.2	39.84	[88]
Cherry sawdust	Acid blue 25	10	50-500	300	20	2.5-4.2	31.98	[88]
Oak sawdust	Methylene blue	10	50-500	60	20	2.5-4.2	29.94	[88]
Oak sawdust	Acid blue 25	10	50-500	60	20	2.5-4.2	27.85	[88]
Pitch-pine sawdust	Methylene blue	10	50-500	60	20	2.5-4.2	27.78	[88]
Pitch-pine sawdust	Acid blue 25	10	50-500	60	20	2.5-4.2	26.19	[88]
Beech wood sawdust	Direct orange 26	2	20-80	240			27.07	[98]
Beech wood sawdust	Acid green 20	2	20-80	240	_	_	18.84	[98]
Beech wood sawdust	Acid orange 7	2	20-80	240		_	24.38	[98]
Crude sawdust	Methylene blue	2	20-80	240	_	6	87.7	[98]
Functionalized sawdust	Methylene blue	2	250	360	_	5	188.7	[99]
Meranti sawdust	Methylene blue	5	50-200	180	60	9	158.73	[100]
Mansonia wood sawdust	Methylene blue		120		26	9	33.44	
Beech wood sawdust	Basic blue 86	2	120	—	20 25	4.43-7.07	136.9	[101] [102]
Beech wood sawdust	Direct brown	2	330-900	_	25 25	4.45-7.07	526.3	
Beech wood sawdust	Direct brown 2	2	320-600	_	25 25	3	416.7	[102] [102]
Treated sawdust	Brilliant green	4	50-300	180	30 22	2.9	58.479	[103]
Raw pine sawdust	Methylene blue	1	1.6-156	240	23	8	41.60	[104]
Treated pine sawdust	Methylene blue	1	1.6-156	240	23	8	65.72	[104]
Eucalyptus wood sawdust	Congo red	4	10-30	240	35 25	7	66.67	[105]
Modified pine sawdust	Methylene blue	2	25-500	480	25	6.5	111.46	[106]
Fugas sawdust	Maxilon blue dye	1	10	60	30	6.7	57.876	[107]
Rattan sawdust	Methylene blue	1	25	480	30 25	7	359	[108]
Bamboo sawdust	Congo red	3.33	100		25		90.51	[109]
Acacia nilotica sawdust	Crystal violet	8	50-500	60	30	6.51	25.253	[110]

m: adsorbent dosage, C_0 : initial dye concentration rang, t: contact time, T: temperature, and q_m : adsorption capacity.

saturation phase. At that time, the lack of free active sites on the adsorbent was the reason behind this behavior [111]. increases of time. This is because the vacant surface sites are difficult to be occupied due to repulsive forces between the solute molecules on the solid and bulk phases.

Ahmad et al. [100] studied the effect of contact time on the removal of methylene blue by meranti sawdust at initial concentrations 50–200 mg/L at 30°C and they found that, with an increase of contact time, the removal of adsorbate was rapid but it gradually slowed down until it reached the equilibrium. When equilibrium was attained, the percentage sorption of methylene blue did not change with further

4.4.4. Effect of Temperature. Temperature plays a very important role in adsorption process because it will change the adsorption capacity of the adsorbent. If the adsorption process is endothermic, the amount of adsorption increases

with increasing temperature. This may be due to increasing mobility of the dye molecules and an increase in the number of active sites for the adsorption with increasing temperature. On the contrary, the amount of adsorption decreases with increasing temperature if the adsorption process is exothermic. This may be due to increasing temperature decreasing the adsorptive forces between the dye species and the active sites on the adsorbent surface as a result of decreasing the amount of adsorption [9].

Gupta and Lataye [110] studied the effect of temperature on the removal of crystal violet (CV) and methylene blue (MB) dyes using *Acacia nilotica* sawdust activated carbon at 283, 293, 303, 313, and 323 K. They noticed that the amount of dye adsorbed per unit gram of adsorbent increases for both CV and MB with the increase in temperature which indicates the endothermic nature of the process.

Mane and Babu [103] reported that the adsorption capacity of Brilliant Green dye by NaOH treated sawdust decreases with an increase of temperature from 288 to 318 K and they concluded the exothermic nature of the adsorption process.

4.4.5. Effect of Solution pH. The solution pH is one of the most important factors which influence the adsorption behavior. The surface charge and the degree of ionization of an adsorbent can be influenced by the variation in pH. Generally, at low pH solution, the positive charge on the solution interface will increase and the adsorbent surface appears positively charged. As a result, the removal efficiency for cationic dye will decrease, while the removal percentage for anionic dyes will increase. In contrast, at high pH solution, the positive charge at the solution interface decreases and the adsorbent surface appears negatively charged, which results in an increase in cationic dye adsorption [9].

In order to understand the adsorption mechanism, it is important to determine an essential parameter called the point of zero charge (pH_{pzc}) at which the surface charge of the adsorbent is zero. The pH_{pzc} value indicates the type of active sites and the adsorption ability of adsorbents. Cationic dye adsorption is favored at $pH > pH_{pzc}$, due to presence of functional groups such as OH^- and COO^- groups. Anionic dye adsorption is favored at $pH < pH_{pzc}$ where the surface becomes positively charged [8].

Zou et al. [106] studied the adsorption of methylene blue (MB) onto citric acid modified sawdust and they found that the value of point zero charge (pH_{pzc}) for the activated carbon was 3.32, while the maximum adsorption capacity of MB was 122.87 mg/g at pH 11; in other words, pH > pH_{pzc}. Islam et al. [108] studied the adsorption of methylene blue by Rattan sawdust, and they reported that the pH_{pzc} value was 4.7 for activated hydrochar and the adsorption performance increases from 92 to 96% removal and MB uptake from 75 to 100 mg/g when the pH is increased from 3 to 7. Özacar and Şengil [96] studied the influence of pH on the adsorption capacity of pine sawdust for metal complex blue (MCB) and metal complex yellow (MCY) as anionic dyes. They noticed maximum removal for MCB and MCY with pine sawdust at

pH 3.5, when the negative charge on the surface is very much reduced due to the excess of protons in solution. They observed also that the amount of adsorbed dye increased from 15.6 to 56.3 mg/g and from 46 to 72 mg/g for MCB and MCY, respectively, with decrease in pH from 11.5 to 3.5.

Most researchers studied the effect of each factor independently of variations in the others, although there may be potential interactions between effects. Thus, there has been an increasing interest in the study of interaction between factors affecting the adsorption process to obtain the optimum adsorption conditions [113, 114].

4.5. *Kinetic Study*. The study of adsorption kinetics still arouses considerable interest because of its importance in the evaluation and application of adsorbents. In order to predict the controlling mechanism of the adsorption process such as chemical reaction, mass transfer, or diffusion control, several mathematical models have been proposed to describe adsorption data. The common kinetic models are the pseudo-first-order [115], pseudo-second-order [116], and intraparticle diffusion [117].

4.5.1. The Pseudo-First-Order Model. The linearized form of the pseudo-first-order model is generally expressed as

$$\ln\left(q_e - q_t\right) = \ln q_e - k_1 t,\tag{1}$$

where q_e and q_t (mg/g) are the adsorption amounts at equilibrium and at time t (min), respectively, and k_1 (min⁻¹) is the adsorption rate constant of pseudo-first-order model. Kinetic parameters, q_e and k_1 , can be determined experimentally from the slope and intercept of the plot ln $(q_e - q_t)$ versus t.

4.5.2. The Pseudo-Second-Order Model. The linearized form of the pseudo-second-order equation is given as follows:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e},$$
 (2)

where q_e and q_t (mg/g) are the adsorption amounts at equilibrium and at time t (min), respectively, and k_2 (g/mg min) is the adsorption rate constant of pseudo-second-order model. Kinetic parameters, q_e and k_2 , can be obtained experimentally by plotting ln ($q_e - q_t$) as a function of t.

4.5.3. The Intraparticle Diffusion Model. The adsorption of soluble species from a solution involves their transfer in several steps from the liquid phase to the surface of an adsorbent [117]. The intraparticle diffusion model is expressed as follows:

$$q_t = k_{\rm id} t^{1/2} + c,$$
 (3)

where $q_t (mg/g)$ is the adsorption amount at time t (min), $k_{id} (mg/g \cdot min^{1/2})$ is the intraparticle diffusion adsorption rate constant, and c is a constant related to the thickness of the boundary layer.

4.5.4. Application of Various Kinetic Models on Dye Adsorption. Generally, the selection of the best-fit model can be chosen based on the values of the linear regression correlation coefficient R^2 and by the normalized standard deviation q (%), defined by following expression:

$$\Delta q(\%) = 100 \sqrt{\frac{\sum \left[(q_{\exp} - q_{cal})/q_{\exp} \right]^2}{N - 1}},$$
(4)

where q_{exp} and q_{cal} are the experimental and calculated equilibrium adsorption capacity value, respectively, and N is the number of data points.

The higher value of R^2 and the lower value of q (%) will be considered to best fit. Table 6 summarizes the various kinetic models of dye adsorption onto various sawdustderived activated carbons.

4.6. Adsorption Isotherm Models. The adsorption isotherm models are important tools for describing the interaction mechanisms between the adsorbate molecules and the adsorbent at constant temperature [8, 118]. Therefore, they play an important role to understand the adsorption process. Several isotherm models are presented in the literature [118]. Langmuir and Freundlich models are the most commonly used to describe the adsorption isotherm.

4.6.1. Langmuir Isotherm Model. The Langmuir isotherm model is a monolayer adsorbed model which assumes that all sites should have equal affinity towards the adsorbate. Each molecule adsorbed on the surface has the same adsorption activation energy, forming a monolayer. The Langmuir equation is expressed by the following relation [119]:

$$q_e = \frac{q_m \cdot K_L \cdot C_e}{1 + K_L \cdot C_e},\tag{5}$$

where q_e is the amount of dye adsorbed at equilibrium time (mg/g), C_e is the equilibrium concentration of dye in solution (mg/L), q_m is the maximum adsorption capacity (mg/g), and K_L is the isotherm constant for Langmuir (L/mg).

Webber and Chakkravorti [120] have defined a dimensionless constant, commonly known as separation factor (R_L), which can be determined from the following relation:

$$R_L = \frac{1}{1 + K_L \cdot C_0},\tag{6}$$

where K_L (L/mg) is the Langmuir constant and C_0 is the adsorbate initial concentration (mg/L). In this context, R_L value indicates the type of adsorption which may be irreversible ($R_L = 0$), favorable ($0 < R_L < 1$), linear ($R_L = 1$), or unfavorable ($R_L > 1$).

4.6.2. Freundlich Isotherm Model. Unlike the Langmuir isotherm model, Freundlich isotherm model is not restricted to the monolayer formation. It considers that the adsorption

surface is heterogeneous and has unequal available sites with different energies of adsorption. The Freundlich adsorption isotherm model is represented as follows [118]:

$$q_e = K_F C_e^{1/n},\tag{7}$$

where q_e is the amount of the adsorbate at equilibrium time (mg/g), C_e is the equilibrium concentration of dye in solution (mg/L), K_F is the capacity of the adsorbent, and n is the intensity of adsorption constant for Freundlich. Several researchers have used Langmuir and Freundlich models to study the adsorption performance of sawdust-derived activated carbon to different dyes. Table 7 shows dyes adsorption isotherms onto various sawdust.

4.7. Sawdust Composites and Mixture. The efficiency of sawdust as a lignocellulosic material in different dyes removal has been approved as raw material [94, 99] or treated by various activating agents [53, 71, 73, 76]. However, adsorbents obtained from a single source have a limitation of their specific nature and therefore have limited applications related to their discharge and regeneration [121, 122]. To overcome this problem, many researchers have used sawdust as a base material with a variety of precursors for making some composites with specific properties [123–125] or as mixture with other materials [126] for the removal of various dyes. Table 8 presents some composites and mixture used for dyes removal.

4.8. Desorption Study. In order to ensure the long-term reusability of used sawdust or its composites and recovery of the adsorbed compounds, desorption studies were planned by using relatively inexpensive salts, acids, organic acids, and bases. An effective eluent should desorb the dye completely without damaging the structure and functional groups associated with the biomass [125]. Desorption process is usually done by mixing a suitable solvent with the dye-saturated substrate and shaking them together for fixed time, until the dye is extracted on the solvent, and then filtration is used to separate the adsorbent. The dye-solvent mixture is dried at high temperature to evaporate the solvent. The desorbed dye is then determined by spectrophotometer [9, 44].

Mashkoor and Nasar [122] studied the desorption of crystal violet from polyaniline/*Tectona grandis* sawdust using different desorbing agents, namely, 0.1 M each of hydrochloric acid (HCl), acetic acid (CH₃COOH), sodium chloride (NaCl), sodium hydroxide (NaOH), and with water. They found that HCl (0.1 M) is the best desorbing agent among the chosen ones for the recovery of polyaniline/*Tectona grandis* sawdust dye with 60.7% desorption.

Shakoor and Nasar [44] studied the desorption of crystal violet from *Terminalia arjuna* sawdust waste using 0.1 M HCl, 0.1 M CH₃COOH, 0.1 M NaCl, 0.1 M NaOH, and the water obtained by double distillation of deionized water DDDW. They found that the percentage desorption was highest in the case of 0.1 M NaOH (2.86%).

TABLE 7: Various kinetic and isotherm studies for dye adsorption by various sawdust.

Sawdust	Dye	Kinetic model	Isotherm model	Reference
Pine sawdust	Malachite green	Pseudo-second-order	Langmuir	[74]
Rattan sawdust	Methylene blue	Pseudo-second order	Langmuir	[76]
Wood sawdust	Basic blue 69	Pseudo-second-order	Langmuir	[89]
Wood sawdust	Acid blue 25	Pseudo-second-order	Langmuir	[89]
Neem sawdust	Crystal violet	Pseudo-first-order	Langmuir	[90]
Neem sawdust	Malachite green	Pseudo-first-order	Langmuir	[90]
Bamboo sawdust	Methylene blue	Pseudo-first-order	Langmuir	[91]
Mahogany sawdust	Acid yellow 36	Pseudo-first-order	Freundlich and Langmuir	[93]
Mahogany sawdust	Direct blue 2B	Pseudo-second-order	Langmuir	[72]
Mahogany sawdust	Direct green B	Pseudo-second-order	Langmuir	[72]
Raw beech sawdust	Methylene blue	Pseudo-first-order	Freundlich	[94]
Beech sawdust	Methylene blue	Pseudo-first-order	Freundlich	[94]
Beech sawdust	Red basic 22	Pseudo-first-order	Freundlich	[94]
Cedar sawdust	Methylene blue	Pseudo-second-order	Langmuir	[97]
Walnut sawdust	Methylene blue	Pseudo-second-order	Langmuir	[88]
Walnut sawdust	Acid blue 25	Pseudo-second-order	Langmuir	[88]
Cherry sawdust	Methylene blue	Pseudo-second-order	Langmuir	[88]
Cherry sawdust	Acid blue 25	Pseudo-second-order	Langmuir	[88]
Oak sawdust	Methylene blue	Pseudo-second-order	Langmuir	[88]
Oak sawdust	Acid blue 25	Pseudo-second-order	Langmuir	[88]
Pitch-pine sawdust	Methylene blue	Pseudo-second-order	Langmuir	[88]
Pitch-pine sawdust	Acid blue 25	Pseudo-second-order	Langmuir	[88]
Beech wood sawdust	Direct orange 26	Pseudo-first-order	Langmuir	[98]
Beech wood sawdust	Acid green 20	Pseudo-first-order	Langmuir	[98]
Beech wood sawdust	Acid orange 7	Pseudo-first-order	Langmuir	[98]
Crude sawdust	Methylene blue	Pseudo-first-order	Langmuir	[99]
Functionalized sawdust	Methylene blue	Pseudo-first-order	Langmuir	[99]
Meranti sawdust	Methylene blue	Pseudo-second-order	Langmuir	[100]
Beech wood sawdust	Basic blue 86	Pseudo-second-order	Langmuir	[102]
Beech wood sawdust	Direct brown	Pseudo-second-order	Langmuir	[102]
Beech wood sawdust	Direct brown 2	Pseudo-second-order	Langmuir	[102]
Eucalyptus wood sawdust	Congo red	Pseudo-second-order	Redlich and Peterson (R-P)	[105]
Modified pine sawdust	Methylene blue	Elovich model	Langmuir	[106]
Fugas sawdust	Maxilon blue dye	—	Langmuir	[107]
Rattan sawdust	Methylene blue	Pseudo-second-order	Langmuir	[108]
Bamboo sawdust	Congo red	Pseudo-second-order	Freundlich	[109]
Acacia nilotica sawdust	Crystal violet	Pseudo-second-order	Redlich and Peterson (R-P)	[110]

TABLE 8: Composites and mixture used for dyes removal.

Composites and mixture	Dye name	Adsorption capacity	References
Fe ₂ O ₃ /chitosan-bamboo sawdust	Bromothymol blue	217.39 mg/g	[123]
Hydrolyzed oak sawdust-alginate biopolymer	Methylene blue	38.46 mg/g	[124]
Magnetized Tectona grandis sawdust	Methylene blue	172.41 mg/g	[125]
Polyaniline/Tectona grandis sawdust	Crystal violet	263.2 mg/g	[122]
Clay-wood sawdust	Basic red dye 46	85.5%	[126]

5. Conclusion

In this review, sawdust as low-cost precursor for activated carbons preparation has been reviewed based on a significant number of papers published so far. Sawdust-derived activated carbons have been proven to be efficient adsorbents for the removal of different dyes from aqueous solution. The characterization of the prepared activated carbon shows a large surface area and high microporosity. As the raw material is relatively inexpensive, freely accessible, and abundant, so the produced activated carbon can be used as a good alternative low-cost adsorbent to commercially activated carbon in the removal of dyes from its aqueous solution. From the large number of published literatures reviewed here, it is observed that the environmental parameters such as adsorbent dosage, initial dye concentration, contact time, temperature, and pH are critical factors affecting the dyes adsorption. This review article also reveals that the kinetic data of adsorption of dyes by sawdust-derived activated carbons were fitted by both the pseudo-firstorder and pseudo-second-order kinetic models, and the Langmuir and Freundlich adsorption isotherm models are usually used to evaluate the adsorption capacity of various adsorbents.

Several researches are limited to lab-scale or batch adsorption studies. However, for real application in industry scale, effluents treatment needs to be carried out in continuous mode. In the future, the research should therefore concentrate on column study. More work should be carried out using real industrial effluent. Regeneration of adsorbents study should be done in detail. Much attention should be given to the preparation of sawdust composites and mixtures. Also, the preparation of granular activated carbon should be taken into account because of its advantages in regeneration process and in mechanical properties.

Data Availability

All the data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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