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Removal of Dyes from Wastewater by Adsorption onto Activated Carbon: Mini Review

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Abstract

Nowadays, wastewater from dyeing industries became a challenging issue in the world. Researchers have reported several techniques to treat those effluents based on their projects. Adsorption is the most common method because of cheap, simple and effective method. In this work, activated carbon was used for dye adsorption purpose. This adsorbent has high surface area and high porosity to remove dye. This review highlighted some important results of the last few years regarding the use of activated carbon in wastewater treatment. Research findings supported that adsorption process is spontaneous in nature. Adsorption data confirmed Langmuir model, indicating the chemisorption occurred.

Keywords

Activated Carbon, Wastewater Treatment, Dye, Adsorption, Surface Area

1. Introduction

Dye consisted of complex aromatic molecular structures, therefore very hard to degrade. Dyes have wide applications, causing the production was expected to grow from year to year. Currently, the top and second largest producer and exporter countries are China and India, respectively (Global Dyes Market Outlook, 2019). Researchers have reported that more than 100,000 commercially available dye are known annually (Menaka & Rana, 2017). The obtained dyes are used in carpet, textile industry, food industry, paper, pharmaceutical industry, and leather industry. Wastewater produced from factories causing environmental pollutions (Konsowa, 2003), allergic dermatitis, skin irritation, and mutation in humans. Dyes could be grouped in several types such as direct dyes, reactive dyes, vat dyes, disperse dyes, azo dyes (Gregory, 1990). Reactive dye and disperse dye have accounted for top and the second largest share in dyes market (Global Dyes Market Projections, 2018).

Currently, some techniques have been applied to remove dye from wastewater as reported by many researchers. For example, ion exchange (Tijen & Filiz, 2019), biodegradation (Sathian et al., 2013), oxidation, membrane process, adsorption and solvent extraction (Marcelo, 2011) were discussed. Disadvantage of ion exchange is high cost, while membrane process is low flow rates. Limitation of biodegradation includes slow process and nutrition requirement. Among these techniques, adsorption method is one of the most popular techniques, because it is a rapid, inexpensive, simple, and low investment method. Activated carbon has been selected as absorbent in wastewater treatment (Ho, 2018). This absorbent has high BET surface area (Ho, 2019) and highly developed microporosity which enhanced adsorption process (Ho, 2017). Generally, several raw materials have been used for the production of activated carbon (Ho & Munir, 2020). For example, orange peel (John et al., 2020), grape seed (Irem et al., 2014), walnut shell (Yu et al., 2019), date palm (Tanweer et al., 2012), neem leaves (Qadir & Chhipa, 2017), rice husk (Khu & Thuy, 2019), sugarcane bagasse (Mahanta et al., 2019), aloe vera leave (Karnan et al., 2016), watermelon rind (Jawad et al., 2019), bananas peel (Achmad et al., 2018) lemon peel (Juhaina et al., 2016), pomelo peel (Zhen et al., 2018), mango kernel (Joaquin et al., 2019) have been reported by many researchers.

The objective of this work was to prepare activated carbon by using various raw materials. Activated carbon served as the adsorbent in order to remove dyes from wastewater. Lastly, adsorption kinetic and isotherm were highlighted.

2. Literature Survey

2.1. Removal of Vat Dye Using Activated Carbon

Vat dyes were used for few thousand years ago. Basically, vat dyes are resistant to sunlight and washing. The molecular formula of Alizarin yellow (**Table 1**) is $C_{13}H_8N_3NaO_4$. It is insoluble in water, could be used in nylon and dyeing wool. However, it irritates the eyes, skin, gastrointestinal tract, and respiratory tract. It changes from yellow (less than 5.2), red (pH 6.8 to 10) to blue-violet colour (more than pH 12) at various pH values. Date palm seed was used to produce activated carbon (Nasiru & Magaji, 2016) in the presence of phosphoric acid as activating agent. It is major biomass sources in United Arab Emirates (UAE). Carbonization was carried out in muffle furnace for 90 minutes at 400˚C. Physico-chemical characterization of obtained activated carbon such as moisture content (15.6%), ash content (2.9%), volatile matter (22.2%), surface area (781 m^2/g), bulk density (0.65 g/mL) and iodine number (711.2 mg/g) was reported.

Table 1. Examples of vat dyes.

Molecular formula	Molecular Weight (g/mol)	Dye
$C_{13}H_9N_3O_5$	287.23	Alizarin yellow

Adsorption study of Alizarin yellow revealed that all data fitted into Langmuir model and pseudo-second kinetic. The Langmuir isotherm was used to explain chemisorption process (Luo & Deng, 2019). It happened when ionic bond or covalent bond (Endre, 2019) was produced between adsorbent such as activated carbon and adsorbate such as dye. Basically, adsorbate adsorption is limited to monolayer (Rackley, 2017). Production of mango seed based activated carbon through carbonization and activation process was described (Abdus & Buhari, 2014). The seed is not the consumable part of the mango, usually discarded as waste (Babatope & Funmilayo, 2017). The obtained activated carbon showed surface area of 819.8 m^2/g, percentage of yield (62.3%), contained 52.3% carbon, and 3.38% hydrogen. Based on the experiment results, the percentage removal of Alizarin yellow dye increased as the agitation time and temperature were increased. Thermodynamic parameters highlighted that adsorption process is spontaneous (negative free energy value), endothermic (positive enthalpy value) and increased randomness (positive entropy value).

2.2. Removal of Basic Dye Using Activated Carbon

Basic dye is insoluble in water because of less hydrophilic group. However, soluble in water in the presence of alcohol or acetic acid. Basic dye is also called cationic dye, available in synthetic form, and can react with substances contained negatively charged. Methylene blue and crystal violet (**Table 2**) are examples of basic dyes. These dyes are low cost and were used in paper, veterinary and textile industries. Production of activated carbon by using coal under activating agent (zinc chloride), activation temperature (500˚C) and activation time (1 hour) (Ramazan et al., 2016). Surface area, micropore volume and total volume are 696 m^2/g, 0.44 cm^3/g and 0.59 cm^3/g, respectively. Adsorption studies confirmed that all data fitted well with Langmuir isotherm. Adsorption capacity is found increased at higher temperature, indicating accelerates transportation of crystal violet to activated carbon surface. Almond shell was used to prepare activated carbon (Ahsaine et al., 2018) in the presence of KOH as activating agent. The loading of zirconium oxynitrate onto surface of activated carbon was reported. Researchers point out that percentage removal of dye increases at higher dosages because of binding sites and surface were promoted. Adsorption studies supported the Freundlich isotherm, indicating multilayer adsorption. Removal of methylene blue and crystal violet were observed to be 208 and 204 mg/g, respectively. The date palm waste was used to prepare activated carbon (Mashael et al., 2013). Thermodynamic investigations confirmed spontaneous adsorption, exothermic process and good affinity between adsorbent and dyes. Adsorption data fitted well into Langmuir

Table 2. Examples of basic dyes.

Molecular formula	Molecular Weight (g/mol)	Dye
$C_{16}H_{18}ClN_3S{\cdot}3H_2O$	373.9	Methylene blue
$C_{25}H_{30}ClN_3$	407.99	Crystal violet

model if compared to Freundlich, Temkin and Elovich model. Effect of temperature onto adsorption process was studied. Researchers found that removal of crystal violet dye reduced from 94% to 89% when the temperature was increased from 25˚C to 55˚C. Rice husk was used to synthesis activated carbon under activating agent such as sulfuric acid and zinc chloride (Kaustubha et al., 2006). Sulfuric acid treated activated carbon (64.9 mg/g) showed higher adsorption capacity if compared to zinc chloride (61.6 mg/g). The kinetic data were described by using Lagergren, and pseudo-second order model. Researchers concluded that intraparticle diffusion plays great role in the removal of crystal violet from wastewater.

The empty fruit bunch was used to prepare activated carbon by using KOH solution in order to enhance surface area and adsorption capacity (Egbosiuba et al., 2020). The highest surface area of 2114 m^2/g was observed at 600˚C, 45 minutes and 1.5 M KOH solution. Adsorption data supported Langmuir model indicating monolayer adsorption. Empty fruit bunch based activated carbon and empty fruit bunch ultra-sonicated activated carbon showed adsorption capacity of methylene blue about 400 and 435 mg/g, respectively. Eucalyptus waste was employed to produce activated carbon via carbonization and activation process (Han et al., 2020). Experimental results showed that surface area (108 to 1545 mg/g), yield (30-9 to 54.3%), total pore volume (0.085 to 1.88 cm^3/g) and adsorption capacity (114.69 to 977 mg/g) were strongly depended on the concentration of phosphoric acid. A well-developed pore structure was created during the impregnation process in the presence of phosphoric acid. The methylene blue adsorption process is spontaneous, exothermic, fitted well into Langmuir model and pseudo-second order kinetics. Teak wood was used to produce activated carbon under activating agent such as sulphuric acid (Gurumoorthy et al., 2019). The physico-chemical behaviors including bulk density (0.53 g/cc), moisture content (6%), total ash (2.3%) and iodine number (1012 mg/g) were investigated. Adsorption data followed Langmuir model, indicating the chemisorption occurred. Removal of methylene blue was found to obey pseudo-second order kinetic.

2.3. Removal of Acid Dye Using Activated Carbon

Characteristics of acid dyes including anionic, were used for wool, food colorants and medical industries at acidic conditions. Several examples of acid dyes (**Table 3**) such as acid blue 83, acid black 1, acid orange 7, acid blue 1 and acid yellow 36. Hazalnut bagasses were used to prepare activated carbon at 600˚C by using zinc chloride as activating agent (Hakan et al., 2008). Absorbent has surface area of 1489 m^2/g. Adsorption capacities increased at lower pH, higher concentration of dye (acid blue 350) and higher temperature. Based on the experiment results, the highest adsorption capacity about 450 mg/g, at 45˚C, pH 2. Adsorption data supported Langmuir model. The wood of Ailanthus altissima was used to produce activated carbon under various temperatures such as 400˚C and 800˚C (Bangash & Alam, 2009). Acid blue 1 is an anionic triphenylmethane dye.

Table 3. Examples of acid dyes.

Molecular formula	Molecular Weight (g/mol)	Dye
$C_{45}H_{44}N_3NaO_7S_2$	825.97	acid blue 83
$C_{22}H_{14}N_6Na_2O_9S_2$	616.49	acid black 1
$C_{16}H_{11}N_2NaO_4S$	350.33	acid orange 7
$C_{27}H_{31}N_2NaO_6S_2$	566.67	acid blue 1
$C_{18}H_{16}N_3NaO_3S$	377.39	acid yellow 36

Molecular weight of this dye is 566.68. Experiment revealed that it has maximum wavelength absorption about 635 nm. Adsorption data confirmed the first order and Bangham model. Free energy, entropy and enthalpy decreased, when the temperature was increased. At higher temperature (800˚C), the carboxyl group disappeared to improve adsorption capacity. The car scrap tire was used to produce activation under KOH solution (Edris et al., 2012). Surface area and total pore volume were reported as 185 m^2/g and 0.58 cm^3/g, respectively. Adsorption data matched well with Langmuir model if compared to Freundlich and Temkin model. Maximum removal of Acid Black 1 dye and Langmuir constant were 14.5 mg/g and 0.044 mg/L, respectively. Kenya tea residue was utilized to synthesis activated carbon by using phosphoric acid under various carbonization temperatures (Behnaz et al., 2017). The obtained carbon has surface area about 832 m^2/g. The highest percentage removal of acid orange 7 about 98.4% at pH 2, and dosage of 10 g/L. Adsorption data supported Freundlich model with correlation coefficient (R) of 0.97 if compared to Langmuir model (R = 0.855). Rice husk and sawdust were used to produce activated carbon (Malik, 2003). Adsorption capacities of acid yellow 36 dyes were 183.8 mg/g and 86.9 mg/g in sawdust and rice husk based activated carbon, respectively. Kinetic data supported pseudo first order model. Researcher found that acidic pH (pH 3) is favorable intra particle diffusion of dye during the experiment. Jatropha husk was used to prepare activated carbon through carbonization and activation process (zinc chloride as activating agent) (Kumaravel et al., 2017). Adsorption kinetic data matched well pseudo-second order model and Freundlich isotherm. Experiment findings confirmed that lower pH (pH 2) and higher temperature are favorable conditions for the removal of acid blue 83 dye.

2.4. Removal of Direct Dye Using Activated Carbon

Direct dye has some properties such as cheap, available in all colours, easily applied, water soluble. It also has an affinity to cellulosic fiber, and low photosensitivity. Hazelnut was used to prepare activated carbon by using zinc chloride as activating agent (Yavuz & Aydin, 2006). Direct yellow 50, direct red 80 (**Table 4**) and direct blue 71 exhibited maximum absorbance value in 398, 527 and 587 nm, respectively. The removal of these dyes were observed to be 11 mg/g (direct yellow 50), 14 mg/g (direct red 80) and 26 mg/g (direct blue 71), respectively.

Table 4. Examples of direct dyes.

Molecular Formula	Molecular Weight (g/mol)	Dye
$C_{45}H_{26}N_{10}Na_6O_{21}S_6$	1373.08	direct red 80
$C_{40}H_{23}N_7Na_4O_{13}S_4$	1029.87	direct blue 71
$C_{35}H_{24}N_6Na_4O_{13}S_4$	956.82	Direct yellow 50
$C_{32}H_{14}CuN_8Na_2O_6S_2$	780.16	direct blue 86
$C_{29}H_{19}N_5Na_2O_8S_2$	675.6	Direct red 81
$C_{30}H_{16}Cl_2N_4Na_2O_8S_2$	741.49	direct blue 106

Orange peel was used as raw material to produce activated carbon (Ahmed at al., 2009). The maximum removal of dye (92%) was observed in pH 2, 6 g/L (adsorbent), 100 mg/L (dye concentration). Adsorption of direct blue 86 supported Langmuir model and pseudo-second kinetic. Bamboo sawdust was utilized to synthesis activated carbon (Sarita et al., 2012). The percentage of removal of direct red 81 dye about 89%. Adsorption data matched well with Freundlich and Halsey model. Kinetic data fitted into pseudo-second order. Adsorption process is endothermic, spontaneous and controlled by intra particle diffusion. Pomegranate peel was used to produce activated carbon (Amin, 2009). Adsorption process is exothermic and spontaneous in nature. The adsorption of direct blue 106 dye increased, when the adsorbent increased, low temperature and reduce in initial dye concentration. The adsorption findings confirmed pseudo-second order and intra particle diffusion. Palm ash was used to prepare low cost activated carbon (Ahmad et al., 2007). Adsorption of blue 71 dye confirmed the Freundlich model in the range of 50 - 600 mg/L. The kinetic data fitted well with pseudo-second order model. Almond shell has been employed to prepare activated carbon (Ardejani et al., 2008). The percentage of removal of direct red 80 dye about 97 % after 1 hour. Adsorption process supported pseudo-second order kinetic and Langmuir model. Orange peel was used to synthesis activated carbon (Azza et al., 2009). Adsorption data revealed that removal of direct navy blue 106 dye is physical process, fitted well to Freundlich model. This work confirmed pseudo-second order kinetic with intraparticle diffusion process.

2.5. Removal of Reactive Dye Using Activated Carbon

Reactive dyes were used in cotton, viscose, dyeing protein and cellulose because of cheap, brightness, simple application and have wide color range. They can produce covalent bond between the fibre polymer and reactive group. Bagasse pith was used to produce activated carbon in the presence of activating agent such as phosphoric acid and zinc chloride (Nevine, 2008) under physical and chemical activation. Removal of reactive orange dye fitted well with pseudo-second-order kinetic and intraparticle diffusion process. Jatropha curcas was employed to synthesis activated carbon (Palanivel et al., 2012). About 95% removal for Remazol Brilliant Blue R (**Table 5**) was observed at the specific conditions such as 0.2 g

Table 5. Examples of reactive dyes.

Formula	Molecular weight (g/mol)	Dye
$C_{22}H_{16}N_2Na_2O_{11}S_3$	626.54	Remazol Brilliant Blue R
$C_{27}H_{18}ClN_7Na_4O_{16}S_5$	984.21	reactive red 194

of adsorbent, and 50 mg/L of dye. The maximum adsorption of dye was observed at pH 3, indicating strong interaction between dye and the activated carbon under this condition. Adsorption data showed that fitted well with Langmuir model, pseudo-first-order kinetic and intraparticle diffusion process. Industrial laundry sewage sludge was utilized to produce activated carbon (Tais et al., 2016). Experimental results indicated that surface area (159 to 65 m^2/g) of mesoporous activated carbon reduced when activation temperature was increased from 750˚C to 850˚C. Adsorption kinetic supported Freundlich model and pseudo-second order. Thermodynamic parameters highlighted endothermic reaction is favored at higher temperature. Adsorption capacity of Remazol Brilliant Blue R about 33.5 mg/g at activation temperature of 750˚C. The Brazilian pine fruit shell was used to produce activated carbon (Eder et al., 2008) via chemical treatment in order to enhance surface area and average porous volume. Removal of hydrolyzed reactive red 194 supported Sips isotherm and Redlich-Peterson isotherm for carbon treated with chromium, followed by acid, and chromium without acid, respectively. Researchers point out that the best conditions to reach the equilibrium in pH 2, 24 hours and at 25˚C.

2.6. Removal of Disperse Dye Using Activated Carbon

Properties of disperse dye such as water insoluble, free of ionizing group, the smallest dye if compared to other groups. It is synthetic dye, used for dyeing of polyester and and textile materials. Azo and anthraquinones could be found in disperse dye. Cherry stone was used to produce activated carbon via micro wave assisted phosphoric acid treatment process (Taner & Fatma, 2016). The obtained activated carbon has higher surface area (1998 m^2/g) and micropore volume (0.937 cm^3/g). Removal of disperse yellow 211 (**Table 6**) favored in pH 11, and kinetic data supported pseudo-second order model. Sawdust has been used to produce activated carbon (Rai et al., 2007). The best pH value about pH 2-3 for the removal of disperse blue 56 dye. The experimental data matched well with pseudo-first-order model. Euphorbia rigida was used to prepare activated carbon (Gercel et al., 2008) via chemical activation (sulphuric acid). The removal of disperse orange 25 about 118.9 mg/g at 20˚C and confirmed the Langmuir isotherm. Kinetic data supported pseudo-second-order kinetic. Bamboo was utilized to synthesis activated carbon (Wang, 2013) in the present of phosphoric acid as activating agent. Analysis of data indicating that the highest percentage of removal of disperse red 167 was 90.2% at 15.4 hours, 50˚C, dye concentration of 50 mg/L and 12 g of activated carbon. Adsorption data supported pseudo-first-order kinetic and Freundlich model. Thermodynamic studies revealed that adsorption is spontaneous, endothermic process.

Table 6. Examples of disperse dyes.

Molecular formula	Molecular weight (g/mol)	Name
$C_{15}H_{13}BrN_2O_4$	365.18	disperse blue 56
$C_{24}H_{26}IN_5O_7$	519.93	disperse red 167
$C_{15}H_{12}ClN_5O_4$	361.73	disperse yellow 211
$C_{17}H_{17}N_5O_2$	323.35	disperse orange 25

3. Conclusion

Activated carbon has been prepared by using different raw materials. Experimental results showed that activated carbon could be used as adsorbent to remove dye from wastewater on a laboratory scale. Various types of dyes such as vat dye, basic dye, acid dye, direct dye, reactive dye and disperse dye were successfully removed from effluents. The percentage removal of dye depended on experimental conditions. In future, more and more researchers focus on the design and operational experiences with pilot plant testing of activated carbon adsorption system.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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