

## A review on adsorbent parameters for removal of dye products from industrial wastewater

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### ABSTRACT

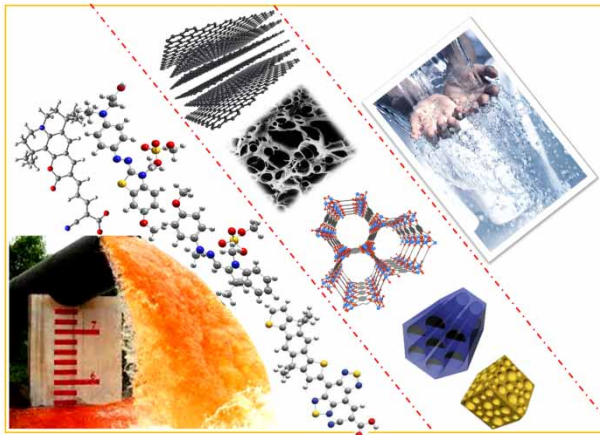
Industrial effluents are usually one of the major industries polluting the environment and surface water. It is estimated that the worldwide production of dyes is about 70 tons/year. To overcome this problem, innovative processes are suggested for the treatment of industrial effluents containing dyes and heavy metals. The goal of the processes is often to reduce the toxicity of these pollutants in order to meet treatment standards. Recently, great attention has been paid to innovative processes for physical and chemical removal techniques such as adsorption on new adsorbents, biomass adsorption, membrane filtration, irradiation, and electrochemical coagulation. In this study, the application of adsorbents in the adsorption process to remove dye pollutants from industrial effluents has been studied. Factors affecting dye adsorption such as pH, temperature, initial dye concentration, and adsorbent amount are also presented. The obtained results revealed that more than 80% of the dye adsorption on the surface of adsorbents are endothermic processes and more than 95% of the processes obey the pseudo-second-order kinetic model.

**Key words:** adsorbent, adsorption, adsorption isotherm, dye removal, industrial effluent

### HIGHLIGHTS

- Investigation of the influence of different adsorbents and different adsorption process parameters.
- Factors affecting dye adsorption.
- Regenerability of adsorbents in the adsorption process to make the adsorption process more economical.
- Application of adsorbents in the adsorption process to remove dye from industrial effluents.

### GRAPHICAL ABSTRACT



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## 1. INTRODUCTION

In recent years, adsorption processes have shown effective results on water and wastewater treatment technology in different industries. In these techniques, a series of natural or synthesized adsorbents have been used to treat contaminants such as metals, dyes, and pharmaceutical products in solutions (Hajipour *et al.* 2021). These adsorbents can be regenerated or consumed after the adsorption process. Synthetic dyes are widely used in many advanced industries such as various textiles, paper, leather, food process, plastics, cosmetics, printing, and industrial dyes (Benkhaya *et al.* 2020). The entry of synthetic dyes into industrial effluents and their discharge into aqueous solutions has caused considerable environmental issues in recent years (Al-Sakkaf *et al.* 2020). The presence of dye pollutants in water will reduce sunlight penetration and negatively affect the photosynthetic activity (Misra *et al.* 2020). To date, more than 100,000 industrial dyes with an annual production of more than  $7 \times 10^5$  tons/year are known (Adegoke & Bello 2015). Textile industries consume more than 100,000 tons/year dyes, and about 100 tons/year of dye enters the effluent water (Mosbah *et al.* 2019). There is no exact information on the amount of dye released from various processes to the environment. However, scientists have identified the release of actual amounts of artificial colors into the environment as an environmental challenge (Lellis *et al.* 2019; Javidparvar *et al.* 2020). Various methods such as adsorption (Konicki *et al.* 2017), coagulation (Kosaka *et al.* 2018), advanced oxidation (Javaid & Qazi 2019) and separation of membranes and nanosorbents (Karim *et al.* 2014; Abdi *et al.* 2018) have been used to remove dyes from wastewater.

Adsorption is an effective process in advanced wastewater treatment techniques to reduce the effluent risks of high pollutants and minerals (Khulbe & Matsuura 2018). Many textile industries use commercial activated carbon to treat colored effluents (Briton *et al.* 2020). Many researchers have reported the possibility of using cheap adsorbents derived from natural materials, industrial, and agricultural solid waste (Sulyman *et al.* 2017). The use of biosorbents to improve the adsorption capacity has been reported by researchers (Adewuyi 2020). Several groups of dyes are stable molecules resistant to decomposition by light, chemicals, biological, and other factors and are considered mutagens for humans (Badran & Khalaf 2019; Javaid & Qazi 2019). Most dyes are complex organic molecules that require resistance to external agents such as detergents (Julkapli *et al.* 2014). Dye molecules are made up of two components: the dye agent, which is responsible for the color formation, and the enhancer, which is responsible for amplifying the dye agent, its solubility in water, and binding to the surface (Bisht & Lal 2019). Therefore, dye-contaminated wastewater must be treated appropriately before being released into the environment (Basheer 2018). In general, colors can be classified according to their structure, application, the charge of particles, or color in the solution. Table 1 shows the classification of colors based on their different applications (Hou *et al.* 2011; Yu *et al.* 2016).

Base colors have a high color intensity and can be seen in low concentrations (Nelis *et al.* 2019). Complex dyes are generally based on chromium, which is carcinogenic (Chakraborty *et al.* 2020). Azo dyes are toxic due to the presence of toxic amines in the effluents (Sarkar *et al.* 2017). Anthraquinone-based dyes are also the most resistant dyes to decomposition and remain in the effluent for a long time (Ray *et al.* 2020). Reactive dyes are soluble in water, and 5–10% of these dyes enter the dye bath and are present in the colored effluent, which creates serious problems for the environment (Hynes *et al.* 2020). Removal of reactive dyes and their degradability in biological systems using conventional wastewater treatment techniques is low (Dayi *et al.* 2020). For this reason, focusing on specific methods of removing dye from industrial effluents helps to select a treatment system appropriate to the type of pollution. The main purpose and novelty of this review study are to

**Table 1** | Classify different colors based on their application (Hou *et al.* 2011; Yu *et al.* 2016)

Category	Sub-layer
Acidic	Wool, nylon, silk, ink, leather, and paper
Basic	Ink, paper, polyacrylic and nitrile, processed nylon, and polyester
Direct	Nylon, rayon, paper, leather, and linen
Disperse	Polyamide, acrylic polyester, acetate, and plastic
Reactive	Wool, linen, silk, and nylon
Sulfur	Rayon and linen
Vat	Cotton, wool, and linen

investigate the application of new and inexpensive adsorbents for the removal of dye compounds from industrial wastewater. In addition, physical and chemical adsorption processes and parameters affecting adsorption capacity are presented. Adsorption kinetics, isotherm models, thermodynamics, and adsorption capacity are collected under different process conditions. Therefore, an overview of the most comprehensive updated information on the adsorption of different colors from aqueous solution by a wide range of adsorbents has been provided in the current work. Over the past two decades, attempts have also been made to use a wide range of adsorbents to analyze color adsorption information (Ali 2013).

## 2. AVAILABLE TECHNOLOGIES FOR DYE REMOVAL

There are currently not many ways to remove dye from the effluents. Despite the availability of many techniques for eliminating dye contaminants from wastewater, such as flocculation, chemical oxidation of membranes, separation processes, electrochemical and aerobic, and microbial anaerobic (Rajasulochana & Preethy 2016; Zaied *et al.* 2020), inherent limitations have been involved by each of these methods (Ghoreishi & Haghghi 2003).

Utilizing biological processes to remove dye from wastewater has grown recently, gaining several research groups' attention. In this method, a new strain of microbe resistant to the microbe contacted the toxic waste that would convert into a less harmful form. The biodegradation mechanism is based on the biotransformation enzymes, which would lead to the biodegradation of recalcitrant compounds in the microbial system. Several enzymes such as tyrosinase, hexane oxidase, demethylase, laccase, or lignin peroxidase were used as the biodegradation agent. However, the degradation of azo dyes by enzymes or a combination of physical methods has gained attention from several biotechnological approaches (Kumar *et al.* 2012).

Physical methods operate based on the mass transfer mechanism and usually consist of straightforward systems to remove dye from wastewater. Ion exchange, irradiation, adsorption, coagulation, and membrane filtration are the conventional physical processes that were used for the separation process. However, among the mentioned methods (chemical, biological, and physical), physical methods are implemented to remove the dye commonly due to ease of use, simple operation conditions, and more efficient processes. Adsorption method has been considered as the very effective method for utilizing in the dye removal process because of several factors, such as low operation cost, reusability of adsorbent, low material consumption, or excellent properties of adsorbent material (e.g., high surface area and high adsorption capacity) (Ghoreishi & Haghghi 2003). Several adsorbents with different features were utilized for the adsorption of dyes, as tabulated in Supplementary Material, Table S2. The pore diameter is also one of the most important features of adsorbent materials that would affect adsorbent selectivity based on the diameter of adsorbed material (Kumar *et al.* 2012).

Various factors affect the way and capacity of the dye adsorption. The most effective of these factors are solution pH, temperature, initial concentration, and amount of adsorbent (Rápó *et al.* 2020). Optimizing these conditions will greatly affect the development of the dye removal process on an industrial scale. Some of the factors affecting color adsorption that have been investigated in most studies are reviewed in the following sections.

### 2.1. Effect of pH

Acidity of the solution is one of the most important factors affecting the adsorbent capacity in wastewater treatment. Adsorption efficiency depends on the pH of the solution because changes in pH lead to changes in the degree of melting properties (Kumar *et al.* 2019). Therefore, pH is mentioned as an important parameter in color adsorption. The adsorption ability and active centers on the surface depend on the zero electric charge point called the surface  $P_{zc}$  (Skwarek *et al.* 2016). This point is directly affected by the used pH solution,  $pH_{pzc}$ , in which surface charge is zero, typically used to quantify the electromagnetic properties of the surface (Jouniaux & Ishido 2012).

The pH value to describe  $P_{zc}$  is only for systems in which  $H^+/OH^-$  ions are predominant. Many researchers have studied the  $P_{zc}$  point of many different adsorbents prepared from agricultural effluents to understand the mechanism of adsorption. The adsorption of cationic dyes at  $pH > pH_{pzc}$  is desirable due to the functional group,  $OH^-$  group, while the adsorption of anionic dyes at  $pH < pH_{pzc}$  is desirable, where the surface is positively charged (Elwakeel *et al.* 2020).

### 2.2. Effect of dye initial concentration

The effect of initial dye concentration is closely related to the dye concentration and the sites present on the adsorbent surface. In general, the percentage of dye removal decreases with increasing initial dye concentration, which leads to a saturation

of adsorption sites on the adsorbent surface (Son *et al.* 2016). On the other hand, increasing the initial concentration of the dye increases the adsorption capacity due to the high driving force of mass transfer in the initial high concentration of dye.

### 2.3. Effect of temperature

Temperature is another important parameter in the physico-chemical adsorption process due to the fact that the amount of adsorbent capacity can be related to the process temperature (Jiang *et al.* 2018). If the amount of adsorption sites increases with increasing temperature, this indicates that the adsorption process is endothermic. This may be due to the increment of the mobility of dye molecules, as the number of active sites for adsorption increases with increasing the process temperature (Wong *et al.* 2020). On the other hand, there are several adsorption processes in which increases in temperature adsorption capacity have declined, indicating the exothermic adsorption process (Belhamdi *et al.* 2016; Peng *et al.* 2020; Romdhane *et al.* 2020). In this kind of adsorption, the adsorption forces between the dye species and the active sites on the adsorbent surface decreased, resulting in a decrease in the adsorption process.

### 2.4. Amount of adsorbent

The amount of adsorbent is an important parameter for determining the adsorbent capacity for a given amount of adsorbent under operating conditions. In general, the percentage of dye removal increases with increasing the amount of adsorbent with increasing the number of adsorption sites on the adsorbent surface. The effect of the amount of adsorbent material presents an idea for the adsorption process, which is economically viable (Salleh *et al.* 2011).

### 2.5. Kinetic study of dye adsorption

Several kinetic models are used in different laboratory conditions to investigate adsorption mechanisms, such as chemical reaction, diffusion control, and mass transfer. The information obtained from adsorption kinetics can be examined to understand the dynamics of adsorption reactions. The study of adsorption kinetics is useful for designing and modeling the process to predict the adsorption rate. The adsorption capacity at all stages of optimization is calculated from the  $q_e$  relation (Equation (1)).

$$q_e = \frac{(C_0 - C_e)V}{M} \quad (1)$$

where in this equation,  $C_0$ ,  $C_e$ ,  $V$ , and  $M$  are initial dye concentration (mg/L), equilibrium dye concentration (mg/L), solution volume (L), and sorbent weight (g), respectively.

Dye removal percentage (color adsorption efficiency) is also obtained from the following equation:

$$R = 100 \times \frac{(C_0 - C_e)}{C_0} \quad (2)$$

Kinetic and thermodynamic adsorption models are widely discussed in the Supplementary Material.

#### 2.5.1. Application of various isotherm models in dye removal

Equilibrium adsorption isotherm models are essential requirements for the design of adsorption systems and the interaction between adsorbent and adsorbent. Models used to analyze equilibrium adsorption data include the Langmuir and Freundlich models (Khayyun & Mseer 2019).

Langmuir is one of the most prominent isotherm models that describe the nonlinear equilibrium between the amount of adsorbed analyte and its free amount in solution at a constant temperature. This model is simple and provides a good description of the experimental behavior in a wide range of working conditions. This isotherm is based on the assumption of monolayer adsorption on an adsorbent with a homogeneous structure that all adsorption sites are the same and equal in energy. The Langmuir relationship is expressed as follows (Mustapha *et al.* 2019):

$$q_e = \frac{q_{\max} k_L C_e}{(1 + k_L C_e)} \quad (3)$$

or in linear form (Equation (4));

$$\frac{C_e}{q_e} = \left( \frac{1}{q_{\max} K_L} \right) + \left( \frac{C_e}{q_{\max}} \right) \quad (4)$$

In Equation (4),  $q_e$  is the amount of adsorbed analyte per unit mass of the adsorbent and  $q_{\max}$  is the maximum amount of adsorption in the adsorbent monolayer in mg/g,  $C_e$  is the equilibrium concentration of the analyte in solution in mg/L, and  $K_L$  is the Langmuir equation constant.

The Freundlich adsorption model (Equation (5)) is an empirical relation which indicates that adsorption takes place in multiple layers (Na 2020).

$$q_e = K_F C_e^{1/n} \quad (5)$$

or in the linear form as follows;

$$\ln q_e = \ln K_F + \left( \frac{1}{n} \right) \ln C_e \quad (6)$$

where  $n$  is a constant related to the heterogeneity of the surface, and its value usually varies in the range of 0–1. The closer value of  $n$  to 1 means the more homogeneous surface.  $K_F$  is the Freundlich constant that is related to the adsorption capacity. The Freundlich model shows that a higher concentration of initial analyte would lead to a higher adsorption rate of nanoparticles.

Hall *et al.* (1966) introduced the dimensionless component  $R_L$ , called the separation coefficient. This component is used to describe the type and shape of the adsorption isotherm. It is expressed as Equation (7), and the relationship of  $R_L$  with the type of Langmuir isotherm is shown in Table 2.

$$R_L = 1 + K_L C_0 \quad (7)$$

where  $C_0$  is the initial adsorbed concentration in solution (mg/L), and  $K_L$  is the Langmuir equation constant. The Langmuir model is obtained by assuming that the adsorption energy is constant for the active positions of the adsorbent surface and does not depend on its coverage.

The Freundlich model assumes that the number of positions in the adsorption action with free energy can be potentially reduced by increasing free energy (Shikuku *et al.* 2018). According to this assumption, with an increasing solute concentration in the solution, the surface concentration never reaches saturation due to high free energy surface sites for adsorption. The value of  $n$  in Equation (5) describes the type of the Freundlich isotherm. The type of relationship is given in Table 3 (Santhy & Selvapathy 2006).

### 3. DISCUSSION

As was mentioned previously, there are many techniques for eliminating dye contaminants from wastewater. All of these different methods of color removal have the advantages and disadvantages as shown in Table 4.

Fenton reaction, electrochemical method, oxidation, photochemical, UV degradation method, ozonation, or advanced oxidation process are typical processes used for dye removal based on the chemical method. Chemical methods are not

**Table 2** | Correlation of  $R_L$  with the Langmuir isotherm

Isotherm situation	$R_L$
Undesirable	$R_L > 1$
Linear	$R_L = 1$
Desired	$0 < R_L < 1$
Reversible	$R_L = 0$

**Table 3** | Correlation  $n$  with the Freundlich isotherm

Isotherm situation	$n$
Undesirable	$n > 1$
Linear	$n = 1$
Desired	$n < 1$

**Table 4** | An overview of the advantages and disadvantages of dye removal methods (Robinson *et al.* 2001; Salleh *et al.* 2011)

Method	Advantages	Disadvantages
Chemical purification		
Oxidation	Ease of use	Requires activation agent
H <sub>2</sub> O <sub>2</sub> + Fe (II) salts	Suitable for removal	Sludge production
Ozonation	It can be used in the form of ozone and does not increase the volume of sludge and effluent	Short half-life (20 min)
Biological treatment		
Decolorization by white-rot fungus	Enzymatic removal of dyes is performed	Enzyme production is unreliable
Microbial mixture	Removal in 24–30 h	Azo dyes are not easily metabolized under aerobic conditions
Adsorption by living or dead biomass	Specific colors have a special tendency to bind to microbial species	No effect for all colors
Anaerobic bioremediation of textile dyes	Makes it possible to remove azo and water-soluble dyes	Anaerobic decomposition produces methane and hydrogen sulfide
Physical purification		
Adsorption with carbon	Perfect removal of a wide range of colors	High cost
Membrane filtration	All of dyes removed	Production of concentrated sludge
Ion exchange	Rehabilitation: no loss of adsorbent	No effect for all colors
Irradiation	Effective oxidation on a laboratory scale	Requires high levels of soluble O <sub>2</sub>
Electrochemical coagulation	Economically flexible	High volume of sludge production

commercially favorable due to specific equipment and high electrical energy compared to physical and biological methods. Besides, utilizing the chemical techniques can lead to additional environmental problems due to the consumption of huge amounts of chemicals and releasing of probable toxic material due to chemical reactions (Katheresan *et al.* 2018).

As typical examples, several works studied dye removal, such as low-cost adsorbents for the removal of organic pollutants from industrial wastewater by Ali *et al.* (2012), adsorption of cationic and anionic dyes by agricultural solid wastes by Salleh *et al.* (2011), unconventional low-cost adsorbents for dye removal by Crini (2006), decolorization of effluent dye by biosorbents by Srinivasan & Viraraghavan (2010), biodegradation of industrial dyes by Ali (2010), and adsorption of aqueous methylene on low-cost adsorbents by Rafatullah *et al.* (2010). However, all are shared only with a specific system and have nothing to do with updated information.

The effects of pH on the dye adsorption from different solutions are given in Table 5.

Chowdhury *et al.* (2011) studied the effect of pH of the solution on the adsorption of four green colors by *Ananas comosus* leaf powder. They found that at a pH range of 2–10, the maximum dye removal rate was at pH = 10. Dawood & Sen (2012) studied the effect of pH on the adsorption of red Congo dye by pine cones and found that the maximum adsorption occurs at pH = 3.5. Ibrahim *et al.* (2010) studied the adsorption of RB4 dye by modified barley straw. They found that the complete removal of RB4 occurred at pH = 3, and with increasing pH, the amount of adsorption decreased to less than 50%. Yagub *et al.* (2012) reported that cationic MB's adsorption on pine leaves increases with pH increasing.



**Table 5** | Results of various studies to determine the effect of pH on the adsorption process

Adsorbent	Dye	pH range	Removal range (%)	References
Modified alumina	Crystal violet	2.6–10.8	20–80	Adak <i>et al.</i> (2005)
Activated carbon	Methylene blue	2–11	Additive	Kannan & Sundaram (2001)
Kaolinite	Crystal violet	2–7	65–95	Nandi <i>et al.</i> (2008)
Bentonite	Blue acid 193	1.5–11	Decrease	Sari <i>et al.</i> (2019)
Fly ash	Methylene blue	2–8	36–45	Kumar <i>et al.</i> (2005)
Fe <sub>2</sub> O <sub>3</sub>	Red acid 27	1.5–10.5	27–98	Nassar (2010)
Tobacco	Methylene blue	2–7.93	60–81	Ghosh & Reddy (2013)
Modified sawdust	Methylene blue	2–11	Additive	Zou <i>et al.</i> (2013)

According to Table 5, almost all of the adsorbent performance in dye removal has been enhanced by increasing pH. Typically, crystal violet removal utilizing modified alumina has 60% increased by increasing pH from 2.6 to 10.8, while this manner is almost repeated for other adsorbents. The increase of H<sup>+</sup> ion concentration may lead to a higher protonation degree of the adsorbent surface, which leads to an increase in the electrostatic interaction between the adsorbent and the dye with a negative charge. A higher interaction between the dye molecule and the adsorbent surface leads to a higher adsorption rate. However, increasing OH<sup>-</sup> group concentration on the adsorbent surface (deprotonated surface) in a high pH would lead to a lower adsorption rate (Wong *et al.* 2020), leading to a more inferior adsorption rate dye in high pH conditions. The pH variation may affect the transport of dye molecules to the surface of the adsorbent, which can be considered as the rate-limiting stage in the dye adsorption process (Saratale *et al.* 2011).

As can be seen in Table 6, the increment of the initial dye concentration leads to an increase in dye removal by implementing all presented adsorbents, while as a typical example, methylene blue removal over kaolinite, pine leaves, or mango kernel powder has increased from 62, 41, and 92.5% to 90, 96, and 99%, respectively. The presence of more dye initial concentrations leads to the increase of dye molecules in constant adsorbent amounts, resulting in higher dye removal and adsorption capacity. Moreover, due to the saturation of adsorbent sites, the remained dye molecules in the solution do not adsorb on the adsorbent. Zhang *et al.* (2012) studied the adsorption of methyl orange by chitosan/alumina. The results of their study showed that when the concentration of methyl orange increased from 20 to 400 mg/L, the percentage of dye removal decreased from 99.53 to 83.55%. Yagub *et al.* (2012) studied the effect of initial dye concentration on the adsorption of methylene (MB) blue from pine trees. They found that the color decreased from 96.5 to 40.9% in 240 min.

However, based on the data as shown in Table 7, adsorbents such as kaolinite or Na-bentonite exhibit exothermic adsorption, while other adsorbents in Table 7 show an endothermic process. Presented adsorption results for kaolinite and Na-bentonite indicate a slight adsorption decline of Red Congo by increasing temperature from 279 to 333 K, so must a low-temperature condition be considered for Red Congo adsorption on the kaolinite and Na-bentonite. Additionally,

**Table 6** | Results of various studies in determining the effect of initial dye concentration on the adsorption process

Adsorbent	Dye	Concentration range (mg·L <sup>-1</sup> )	Removal range (%)	References
Kaolinite	Methylene blue	10–40	62–90	Motamedi <i>et al.</i> (2011)
Fly ash	Red Congo	5–30	84–99	Mall <i>et al.</i> (2005)
Red clay	Purple acid	10–40	12.52–26.2	Namasivayam <i>et al.</i> (2001)
Activated carbon	Ariochrome block T	30–150	10–45	Luna <i>et al.</i> (2013)
Mango kernel powder	Methylene blue	50–250	92.5–99	Senthil Kumar <i>et al.</i> (2013); Anitha <i>et al.</i> (2016)
Pine leaves	Methylene blue	10–90	41–96	Ramaraju <i>et al.</i> (2013)
Rice husk	Green Malachite	10–30	71–82.5	Kahraman <i>et al.</i> (2011)
Apricot kernels	Black Astrazone	50–500	62–91	Argun <i>et al.</i> (2008)

**Table 7** | Results of various studies to determine the effect of temperature on dye adsorption

Adsorbent	Dye	Temperature range (K)	Process	References
Kaolinite	Red Congo	279–333	Exothermic	Vimonses <i>et al.</i> (2009)
Na-Bentonite	Red Congo	279–333	Exothermic	Vimonses <i>et al.</i> (2009)
Pine leaves	Methylene blue	313–333	Endothermic	Yagub <i>et al.</i> (2012)
Activated bamboo waste	Reactive black 5	303–323	Endothermic	Ahmad <i>et al.</i> (2013)
Residual sludge	Green Naphthol	303–323	Endothermic	Attallah <i>et al.</i> (2012)

exothermic adsorption of Red Congo over kaolinites and Na-bentonite was approved by the exhibited negative enthalpy ( $\Delta H$ ) values from  $-15.25 \text{ KJ}\cdot\text{mol}^{-1}$  at 279 K to  $-15.532 \text{ KJ}\cdot\text{mol}^{-1}$  at 333 K. On the other hand, other adsorbents exhibit an endothermic adsorption process, in which adsorption capacities have increased by an increase in temperature for pine leaves, activated bamboo water, and residual sludge adsorbents. Typically, methylene blue adsorption over pine leaves has grown at a higher temperature, indicating endothermic adsorption of methylene blue over pine leaves. Besides, exhibited positive enthalpy ( $15.29 \text{ KJ}\cdot\text{mol}^{-1}$ ) confirms the endothermic process of methylene blue.

According to Table 8, in the presence of higher concentrations of adsorbents the dye removal was increased. By increasing the pine cone adsorbent amount from 0.01 to 0.03 g, Red Congo dye removal has grown from 13.45 to 18.96, while this manner was observed for all mentioned adsorbents. As discussed above, the higher dosage of adsorbent would lead to higher vacant sites availability, so more dye molecules can contact adsorbent active sites that lead to higher dye removal. Additionally, by an increase in the amount of adsorbent, a higher surface area (micropore or mesopore structures) may be available. This higher surface area enables the adsorbent to operate with a higher capacity in dye adsorption (Mall *et al.* 2005).

It is important to regenerate the adsorbent and reuse it in the adsorption process to make the adsorption process more economical. Almost all of the adsorbents have been regenerated by chemical agents such as NaOH or HCl. In most cases, the regenerated adsorbent was suitable for the color adsorption process for at least three cycles; however, the reusability of the adsorbents may decline after several regeneration cycles. According to Table 9, all mentioned adsorbents show

**Table 8** | Results of studies performed to determine the effect of adsorbent on removal percentage

Adsorbent	Dye	Amount of adsorbent	Removal percentage (%)	References
Pine cone	Red Congo	0.01–0.03 mg	13.45–18.96	Dawood & Sen (2012)
Fly ash	Methylene blue	8,000–20,000 mg	45–96	Kumar <i>et al.</i> (2005)
Tea residual	Basic yellow	$2.20 \text{ g}\cdot\text{L}^{-1}$	19–60	Khosla <i>et al.</i> (2013)
Orange peel	Purple acid	50–600 mg/50 mL	15–98	Sivaraj <i>et al.</i> (2001)
Rice husk	Orange reactive	20–80 mg	21.7–56.2	Ong <i>et al.</i> (2007)
Refined sawdust	Green Malachite	200–1,000 mg/100 mL	18.60–86.90	Garg <i>et al.</i> (2003)

**Table 9** | Regeneration of adsorbents implemented for dye removal

Adsorbent	Dye name	Regeneration agent	Reuse cycle	References
Activated pinecone	Alizarin Red S	NaOH	4	Bhomick <i>et al.</i> (2018)
Magnetic rice husk	Methylene blue	NaOH	10	Lawagon & Amon (2019)
Chitosan	Reactive black 5	NaOH	5	Vakili <i>et al.</i> (2019)
Chitosan/bentonite/CTAB	Weak acid scarlet	NaOH	3	Vakili <i>et al.</i> (2019)
Activated bentonite/alginate	Methylene blue	HCl	6	Aichour & Zaghouane-Boudiaf (2020)
Fe <sub>3</sub> O <sub>4</sub> /activated charcoal/-cyclodextrin/alginate	Methylene blue	HCl	5	Yadav <i>et al.</i> (2020)
Modified activated carbon	Methylene blue	HCl	4	Naushad <i>et al.</i> (2019)



proper reusability after that regenerated by a chemical treatment (e.g., HCl or NaOH). However, adsorbent activity may decline after several cycles of regeneration due to part structure collapse or remaining adsorbed molecule in adsorbent structure (Fan *et al.* 2018).

#### 4. CONCLUSION AND FUTURE PERSPECTIVES

In this study, a series of adsorbents such as industrial solids, agricultural by-products, activated carbon, activated carbon-based biomasses, nano-adsorbents, mineral oxides, and mineral soil are reviewed to remove dye or contaminants from aqueous solutions. Adsorbents should be available, cost-effective, porous, recyclable, and have many active sites on their structure. In this way, future studies could focus on the adsorbents with good adsorption/desorption performance, recyclability, and cost-effectiveness. Furthermore, finding cost-effective strategies in the fields of storage and reusability of used adsorbents are other important issues for the future studies. In this way, the safe and economical disposal/reuse of used adsorbents should be considered in future investigations. It is possible to use raw materials and refining materials instead of expensive commercial activated carbon to remove dye from aqueous solutions. In this review study, a large number of articles have been reviewed, and as can be seen, the mechanism and methods of adsorption for the removal of contaminants by the adsorbent depend on the conditions of experimental physical and chemical data such as pH, initial contaminant concentration, adsorbent concentration, and temperature. In this review study, it is found that the Langmuir and Freundlich adsorption isotherm models for measuring the adsorption capacity of adsorbents are different, and the kinetic data for the adsorption of color pollutants usually follow the pseudo-second-order kinetic model.

#### CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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