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Removal of dye by adsorption onto activated carbons: review

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Abstract: *The production of dye is increased rapidly due to the development of various industries. The removal of dye could be carried out by using activated carbon through adsorption process. Currently, several types of agricultural wastes and fruits were used to produce activated carbon. In this work, removal of Congo red, crystal violet, turquoise blue, reactive black 5, and malachite green onto activated carbon was studied. The adsorption and equilibrium data were investigated using different models. Experiment results show that adsorption process is endothermic and spontaneous process.*

Key words: *Activated carbon, agricultural wastes, surface area, wastewater treatment, adsorption.*

INTRODUCTION

The increase of dye production and its application in various industries (textile, food industry, pharmaceutical industry, dyeing industry, paper and leather industry) leads to environmental pollutions [1-3]. Colour in the water affects the nature of water and inhibit sunlight penetration. The toxicity of dye causes allergic dermatitis, skin irritation, cancer and mutation in humans [4-6]. The dye wastewater must be filtered before discharging into the environment.

The removal of dye from wastewater could be carried out in various methods [7-10] including biodegradation, ion exchange, oxidation, adsorption and solvent extraction. Among these methods, adsorption is popular method. Adsorption has many advantages [11-15] such as low cost, simple design, low investment, low consumption of reagent. Activated carbon has been used for purification of water since few decades ago. These adsorbents show higher surface area and larger micro porosity (Figure 1). Generally, activated carbon could be produced by using various raw materials such as Borassus bark, rice husk, Borassus aethiopum flower, orange peel, Pandanus leave, grape seed, walnut shell, palm shell, sugarcane bagasse, pinus tree, neem husk, Green peas shell, bean pod, groundnut shell, date palm leaf, Ferula Orientalis L stalk, aloe vera leave shells, neem leaves, barnacle shell, Xanthiyam strumarium and Martynia Annua L.

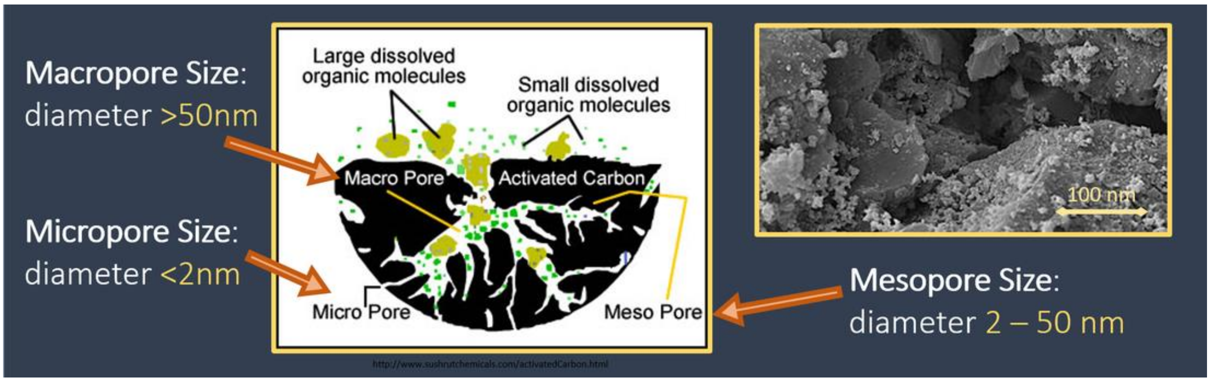

Fig 1: Porosity of activated carbon [16]

In this work, removal of Congo red, crystal violet, turquoise blue, reactive black 5, and malachite green onto activated carbon was studied. The kinetic and equilibrium data were investigated using Langmuir isotherm, Freundlich model, first or second order model.

Literature survey

Paul Bottiger has successfully discovered Congo red in 1883. It is classified as anionic dye and has aromatic structure. Generally, Congo red was used in textiles, printing, dyeing, paper, rubber and plastic industries. The properties of this dye including water soluble, very toxic and stable to biological degradation. Yusef and co-workers [17] have reported the preparation of activated carbon by using aloe vera leave shells. Results obtained revealed that the best contact time of the uptake process is 20 minutes at pH 2. The amount of Congo red was increased from 200 to 929 mg/g with increasing dye content (100 to 500 mg/L). Finally, they concluded that the maximum of dye uptake capacity is 1850 mg/g, fitted second-order-kinetic and Freundlich isotherm models. The removal of dye has been reported by Shiv and co-workers have synthesized activated carbon using neem leaves [18]. They found that adsorption of Congo red increases with an increase in dye concentration (50-150 mg/L) and adsorbent dosage (0.25-0.75 g). At the very beginning, the removal of dye using activated carbon increased with increasing activated carbon dose. Because of increasing surface area of adsorbent and the availability of more adsorption sites. Adsorption kinetics were studied and fitted Langmuir isotherm. Jasim and co-workers [19] have proposed the production of low cost adsorbent by using barnacle shell. The optimum time removal, dye concentration and pH were observed to be 180 minutes, 30 mg/L and pH 4-5.5, respectively. Jayaraj and co-workers [20] have reported the preparation of activated carbon by using marine valoria bryopsis. The samples were treated using sulphuric acid for 8 hours. The removal of Congo red was maximum at pH 5, and adsorbent dosage (200 mg). The presence of H^+ and OH^- ions influence the adsorption process. There are competing between anionic dye (Congo Red) and hydroxide ions exist in the basic medium. The free energy is negative value (from -6.7 to -11.2 kJ/mol), indicating a spontaneous process as shown thermodynamic parameters. Vadivel and co-workers [21] have pointed out the synthesis of activated carbon by Xanthiyam strumarium and Martynia Annua L. These carbons have high surface area (532-1142 m^2/g), iodine number (438-1052 mg/g) and phenol adsorption capacity (4.2-6.6 mg/g). Scanning electron microscopy (SEM) analysis supported that the prepared adsorbents have honey comb shape, well-developed pores with uniform distribution on the surface.

Green peas shell was used to produce activated carbon under different activation agents. Thermodynamic parameters indicated the negative value of enthalpy (exothermic process) and free energy (spontaneous process). Langmuir isotherm describes the adsorption data very well, indicating crystal violet could be adsorbed in monolayer coverage. The removal of dye is reduced [22] (Dandge et al., 2016) because of an excess of H^+ ions in pH 2 (52%) if compared to pH 7 in NaOH treated sample (94.5%), and H_2SO_4 treated sample (97%). Bean pod and groundnut shell were employed to prepare activated carbon [23] (Akinola 2015). The removal of crystal violet reached 80.8 % at pH 6 (groundnut shell) and 84.4 % at pH 5 (bean pod). The adsorption kinetic data fit pseudo second order model and Freundlich isotherm. Date palm leaf was utilized as starting material to synthesis activated carbon [24] (Maliha 2015). The highest percentage removal of crystal violet was observed in nitric acid treated sample if compared to sulfuric acid treated sample. The influence of mesh size on the properties of activated carbon was studied. They conclude that the smallest size (<300 µm) exhibited the highest adsorption if compared to other sizes (300, 425, 710 µm). Adsorption data supported the pseudo second order model and followed Langmuir isotherm. The adsorption of crystal violet from wastewater was investigated using activated carbon prepared from Ferula Orientalis L stalk [25] (Aysu 2015). It has high surface area (1476 m^2/g) based on BET analysis. Equilibrium data fitted Freundlich isotherm and pseudo-second order model. The thermodynamic parameters show that adsorption of dye onto activated carbon is spontaneous (ΔG=-3.6 to -4.1 kJ/mol) and endothermic (ΔH=3.06 kJ/mol) process.

The adsorption of reactive turquoise blue onto activated carbon prepared from sugarcane bagasse increased as the initial concentration was increased in acidic medium. The obtained carbons have high surface area (1843 m^2/g). They highlight that the total pore volume increased with increasing activator (zinc chloride) proportion [26] (Li et al., 2017). Pinus tree was used as starting material to produce activated carbon [27] (Schimmel et al., 2010). Maximum dye removal capacity was observed at pH 2 and 30 °C. Pseudo second order model was used to study the kinetics of dye. FTIR spectrum was used to identify the functional groups on the surface of activated carbon such as hydroxyl (3600 cm^{-1}), aldehydes (1500 cm^{-1}), N-O (1350 cm^{-1}) and ketone (1700 cm^{-1}). Neem husk was used to prepare activated carbon

[28] (Alau et al., 2010). The experimental findings supported high adsorption for turquoise blue. The adsorption data fitted Langmuir isotherm (R^2= 0.904 to 0.997). The influence of activator was investigated. The obtained carbons treated with $ZnCl_2$ gave the highest percentage of removal (99 %), followed by H_3PO_4 (79 %), and KOH (68 %).

Grape seed was used to produce activated carbon [29] (Mojtaba 2017). The highest percentage (83 %) removal of reactive black 5 could be achieved by raising the contact time (2 hours), amount of adsorbent (10g/L carbon) and at lower pH value (pH 2). The kinetic data supported Langmuir adsorption isotherm (R^2=0.999). Walnut shell [30] (Mahboobeh, 2010) was utilized to synthesis activated carbon. The surface area obtained is 249 m^2/g. The maximum dye removal (reactive black 5) was found in acidic pH. The fitting of adsorption data to Dubinin-Radushkevich model and Pseudo-second order kinetic model was confirmed. Palm shell was used to produce activated carbon [31] (Wei, 2015). The adsorption capacity to be favorable (25 mg/g) in acidic medium (pH 2). Pseudo-first-order model and Langmuir model were confirmed through experiment. Thermodynamic studies show that endothermic process with an activation energy (12.6 kJ/mol).

Borassus bark was used to prepare activated carbon. The obtained samples were treated with sulphuric acid [32] (Arivoli, 2007). The removal of malachite green was investigated and the adsorption data supported first order model. Endothermic and spontaneous were observed according to thermodynamic data. The enhancement of adsorption capacity (observed from Langmuir model) at higher temperature (20.7 mg/g at 60 °C) was because of the enlargement of pore size. Rice husk was employed to synthesis activated carbon [33] (Sharma 2009). The removal of malachite green reduced from 94.9 to 93.75 % by increasing initial concentration (60 to 100 mg/l). Monolayer adsorption capacity (63.85 mg/g) onto activated carbon could be seen at room temperature. Borassus aethiopum flower [34] (Nethaji, 2010) was used to produce activated carbon. The adsorption of dye onto adsorbent was fitted with pseudo second-order model and Langmuir model. Orange peel was used to produced activated carbon [35] (Abdussalam et al, 2018). This adsorbent contains ash (5.6%), carbon (4.7%) and moisture content (13.3%). There are several peaks could be observed in FTIR spectra including ethene (1400 cm^{-1}), chloro compound (860 cm^{-1}), (1864 cm^{-1}) ester, (3056-3700 cm^{-1}) alcohol. The ΔG (-553 to -4096 kJ/mol), ΔS (47.2 J/mol) and ΔH (13526 kJ/mol) values were measured. Henry isotherm and pseudo second order were found fit better for the removal of dye by using orange peel. Pandanus leave was used to prepare activated carbon. The samples were treated by using sulphuric acid during the carbonization process [36] (Hema and Arivoli, 2008). The percentage removal of dye was observed reduced with increase in initial dye concentration from 5, 10, 15, 20, 25 to 30 mg/L and increased with increase in temperature from 30 to 60 °C (indicating the physical adsorption). The obtained enthalpy, entropy and free energy values are about 11.2, 44.4 and -3.5 kJ/mol, respectively. Up-to-dare, there are various raw materials could be used to produce activated carbon as indicated in Table 1. Researchers have successfully reported adsorption data and kinetic data through different models such as Langmuir model, Freundlich isotherm, first and pseudo-second order model.

Table 1: De removal by adsorption using activated carbon

Precursor	Dye	Highlighted results	References
Mahogany sawdust	Direct dye	Adsorption data supported Langmuir isotherm and pseudo-second order kinetic. The best conditions for removal of dye: pH 3, 300 mg dye/g adsorbent	Malik [37]
Coconut husk	Maxilon blue GRL and direct yellow DY 12	Adsorption is endothermic process based on thermodynamic parameter. The adsorption data supported Fritz-Schlunder model.	Aseel and co-workers [38]

		Adsorption of dye was favorable when the pH less than 7, with increasing initial dye concentration and time.	
Ficus carica bast	Methylene blue	Thermodynamic parameters such as enthalpy (21.5 kJ/mol), entropy (76.2 J/mol.K) and free energy (-1.55 kJ/mol) were studied Adsorption equilibrium followed Langmuir and Tempkin models.	Deepak and co-workers [39]
Crab shell	Acid brilliant scarlet	Activated carbon treated with KOH showed higher surface area (2197 m^2/g) and large pore volume (1.19 cm^3/g) Langmuir model and pseudo-second order kinetic were found to fit data.	Gao and co-workers [40]
Pomegranate peel	Direct blue -106 dye	Removal of dye was observed increased at pH 2, with increasing amount of activated carbon. Adsorption process was spontaneous process and followed pseudo-second order model.	Amin [41]
Coconut shell	Rhodamine-B	The obtained activated carbon showed higher surface area (1200 m^2/g) and iodine number (600 mg/g). The removal of dye was observed to be fitted to Freundlich (R^2=0.96) if compared to Langmuir (R^2=0.9) model. The best condition for removal of dye at pH 7, initial dye concentration (150 mg/L), and amount of carbon (1 g).	Balasubramani and Sivarajasekar [42]

Cordia myxa	Disperse Blue 56	Removal of dye reached 80 % in 30 minutes, pH 7, 25 °C. Kinetic data were fitted to the Freundlich isotherm and pseudo-second order.	Janabi and Kihc [43]
Mussell shell	Basic Fuchsin dye	Adsorption on dye onto activated carbon was spontaneous and endothermic process. The adsorption data supported Langmuir model and pseudo-second order model.	Haddad [44]
Coffee husk	Fast green	The presence of OH, CO and C=O groups as indicated in FTIR spectra. Equilibrium data was supported by Langmuir (R^2=0.97) and Freundlich (R^2=0.99) model.	Ahalya and co-workers [45]

CONCLUSIONS

Activated carbon can be used to remove dye in wastewater in order to protect environment. In this paper, removal of Congo red, crystal violet, turquoise blue, reactive black 5, and malachite green onto activated carbon through adsorption process. The obtained adsorption data supported either Langmuir or Freundlich model. Researchers point out that adsorption process is endothermic and spontaneous process based on thermodynamic parrameter.

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