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Research

PREPARATION AND CHARACTERISATION OF ACTIVATED CARBON FROM *DELONIX REGIA* SEEDS FOR THE REMOVAL OF METHYLENE BLUE DYE

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ABSTRACT

In the present work, activated carbon (AC) have been prepared from *Delonix regia* seeds by chemical activation with H_2SO_4 and has been evaluated for the removal of methylene blue dye by varying pH, contact time, concentration of adsorbate, adsorbent dosage and temperature. The prepared AC was characterised using SEM, FTIR and BET analysis. Scanning electron microscopy images showed a high pore development, with BET surface 259.2714 m^2/g and pore size 26.428 Å. Fourier-transform infrared spectroscopy allowed characterizing the presence of functional groups on the surface of activated carbons. From the optimised conditions, 90% removal of methylene blue dye could be achieved in 300 min with an adsorbent dosage of 0.3 g. The adsorption isotherm, kinetic and thermodynamic parameters was analysed using MATLAB software. The adsorption process followed Freundlich isotherm model with maximum monolayer adsorption capacity of 59.38 mg/g and the nature of adsorption was found to be chemisorption. The kinetic parameter model for the adsorption of methylene blue on to activated carbon (AC) from *Delonix regia* followed pseudo second order kinetics with correlation coefficients of higher than 0.99. The adsorption capacity of this seed can be utilized for the treatment of strongly polluted industrial waste waters containing dye.

INTRODUCTION

Water pollution has been a major challenge to environmental engineers today due to the release of toxic heavy metals from various industries. Synthetic dyes and heavy metals are the main pollutants, which are excessively released into the environment due to rapid industrialization and have created a major global concern. Dye contamination in wastewater causes problems in several ways: the presence of dyes in water, even in very low quantities, is highly visible and undesirable; color interferes with penetration of sunlight into waters; retards photosynthesis; inhibits the growth of aquatic biota and interferes with gas solubility in water bodies (O¨zer and Dursun, 2007). These dyes are purged from multiple sources such as textile, cosmetic, paper, leather, rubber and printing industries (Singh and Arora, 2011). It has been estimated that about 10% to 15 % of these dyes are released as effluents during the dyeing processes (Gómez, *et al*., 2007). These materials are complicated organic compounds which is resistant against light and microbial invasions. Thus, they cannot be decomposed easily. Direct discharge of dyes containing effluents into municipal environment may cause the formation of toxic carcinogenic breakdown products. The highest rates of toxicity were found amongst basic and diazo direct dyes. Therefore, it is highly necessary to reduce dye concentration in the wastewater. The conventional methods for treating dye containing waste waters are electrochemical treatment (Lata, *et al.,* 2007; Wang, *et al*., 2008), coagulation and flocculation (Fan, *et al.,* 2008), chemical oxidation (Gurses, *et al.,* 2002), liquid–liquid extraction (Muthuraman, *et al.,* 2008) and adsorption (Mohan, *et al.* 2002; Wang, *et al*., 2005a; Wang, *et al*., 2005b). Methylene blue (3,7 bis[dimethylamino] phenazothionium chloride) is the most commonly used material for dyeing cotton, wool and silk. Methylene blue specifically causes eye burn, which may be responsible for permanent injury to the eyes of humans and animals. It can give rise to short period of rapid or difficult breathing on inhalation while ingestion through the mouth

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produces a burning sensation and may cause nausea, vomiting, profuse sweating and mental confusion. Methylene blue dye serves as a model compound for adsorption of organic contaminants from aqueous solutions (Avom, *et al.*, 1997; Kavitha and Namasivayam, 2007).

Activated carbon (AC) is a well known adsorbent that can be used efficiently for removal of a broad spectrum of pollutants from air, soil and liquids. Examples are activated carbon (adsorbs mainly organics), silica gel and activated alumina (absorb moisture), zeolites and molecular sieves and synthetic resins. Among them, AC is more efficient adsorbent for elimination of many pollutants (organic, inorganic, and biological) of concern in water and wastewater treatment. This is because of their highly developed porosity, large surface area (that can reach 3000 m^2/g), variable characteristics of surface chemistry, and high degree of surface reactivity. These unique characteristics make AC very versatile materials, which have been studied not only as adsorbents, but also as catalysts and catalyst supports used for different purposes such as the removal of pollutants from gaseous or liquid phases and the purification or recovery of chemicals. In order to be commercially attractive, an adsorbent should embody a number of features: (i) the adsorbent should have a large internal surface area, which should be accessible through pores big enough to admit the molecules to be adsorbed; (ii) the adsorbent should have high affinity and capacity for target compounds; (iii) the adsorbent should be capable of being easily regenerated; (iv) the adsorbent should not age rapidly, that is lose its adsorptive capacity through continual recycling; (v) the adsorbent should have tolerance for a wide range of wastewater parameters; (vi) the adsorbent should be mechanically strong enough to withstand the bulk handling and vibration that are feature of any industrial unit. However, due to their high production costs, these materials tend to be more expensive than other adsorbents. Most commercial systems use activated carbon as adsorbent to remove dyes in wastewater because of its significant adsorption capacity. Although, activated carbon is a preferred adsorbent, its widespread use is restricted due to its cost. In order to decrease the cost of treatment, some attempts have been made to find low cost alternative adsorbents. Researchers have studied the production of activated carbon from palm-tree cobs (Avom, *et al.*, 1997). plum kernels (Wu, *et al.*, 1999) cassava peel (Rajeshwarisivaraj, *et al.*, 2001), bagasse (Tsai, *et al.*, 2001) jute fiber (Senthilkumaar, *et al.*, 2005) rice husks (Yalc, *et al.*, 2009), olive stones (El-Sheikh, *et al.*, 2004), date pits (Girgis and El-Hendawy, 2002), fruit stones and nutshells (Ayg¨un, *et al.*, 2003) A. The advantage of using agricultural by-products as raw materials for manufacturing activated carbon is that these raw materials are renewable and potentially less expensive to manufacture. Activated carbons have the advantage of exhibiting a high adsorption capacity for colour pollutants due to their high surface area or porous structure. Besides these physical characteristics, the adsorption capacity of a carbon produced from different sources is strongly influenced by the chemical nature of the surface. Although the determination of the number and the nature of the activated carbon groups began earlier, the precise nature of the functional groups is not entirely established. Due to these functional groups such as carboxyl, hydroxyl, aldehyde, and anhydride, the carbons have an acid–base character. It is now known that the acid or base character of carbon depends on its preparation and treatment conditions at which it was oxidised.

Flamboyant tree plantation is grown around the world for ornamental reasons, preserving the soil and also conserving the environment. Apart from these benefits, the tree species from the Fabaceae family is known for its fern-like leaves and flamboyant display of flowers. It grows in southern Brazil and is also found in other parts of the world, such as the Caribbean, the USA, Hawaii, Puerto Rico, the Virgin Islands, the Canary Islands, India, Hong Kong, Taiwan, southern China, and Australia. The pods gradually fall off the trees after a period of maturation. Due to their abundance, they may be a very interesting precursor for the production of activated carbon. In this research, a new raw material, flamboyant seeds (*Delonix regia*), was used.

In the present work, the activated carbon (AC) was prepared from flamboyant seeds (*D. regia*) using sulphuric acid as activating agent and it was characterised by SEM, FTIR and BET analysis. The prepared AC was evaluated on the performance of AC for the removal of methylene blue dye. The adsorption isotherm and kinetic parameters were determined using MATLAB software.

MATERIALS AND METHODS

Preparation of activated carbon

AC was prepared from *Delonix regia* seeds, collected from trees located at SRM University by carbonisation and chemical activation using sulphuric acid. The raw material was initially washed with distilled water to remove dust, other impurities and then dried in sunlight. The sample was pre-carbonised at a temperature of 300°C for 3 hrs in an air tight crucible and soaked in sulphuric acid in the ratio 1:1 for 24 hrs. It was then washed; pH was adjusted to 7.0 and activated at a temperature of 800°C for 1 hr in order to render it as porous in nature. The AC was then powdered and stored for further analysis.

Preparation of adsorbate

Methylene blue is a basic dye with heterocyclic aromatic chemical moiety. The molecular formula is $C_{16}H_{18}N_3SCl$ (3, 7-bis (dimethylamino)-phenazathionium chloride) with absorption maxima at 665 nm. The dye stock solution 1000 ppm was prepared by dissolving 1 g dye in 1000 ml distilled water. The experimental solutions were obtained by diluting the dye stock solution to the desired initial concentrations.

Characterisation of AC

The specific surface area (m^2/g) was calculated using Brunauer–Emmett–Teller (BET) isotherm equation. The surface morphology of the AC was determined using scanning electron microscopy (SEM). The IR spectra were recorded in KBr on a Fourier transform infrared spectrophotometer.

Adsorption kinetics

Adsorption equilibrium and kinetics were determined by the batch method. Adsorption isotherms were performed in a series of 100 ml conical flasks where dye solutions (50 mL) with different initial concentrations (20, 40, 60, 80, 100 ppm). Equal masses of 0.3 g of activated carbon were added to dye solutions. The flasks were placed in a shaker, 100 rpm at room temperature. Residual concentration of MB was determined using UV spectrophotometer at 665 nm. The percentage removal of dye from solution was calculated by the following:

$$\%\,\text{Removal} = \frac{C_0 - C_t}{C_0} \times 100\% \qquad (1)$$

C_0 : Initial concentration (mg/l)

C_t : Final concentration at time *t* (mg/l)

The amount of adsorbate uptake by AC in each flask was calculated using the mass balance equation:

$$q_e = \frac{(C_0 - C_e)\text{V}}{W} \qquad (2)$$

q_e: Amount of adsorbate adsorbed by AC (mg/g)

C_0 : Initial concentration (mg/l)

C_e: Final concentration (mg/l)

V: Volume of solution (L)

W: adsorbent weight (g)

DISCUSSION

Characterisation of activated carbon

SEM analysis

Surface morphology of AC is shown in Fig. 1a, the micrograph clearly shows the presence of highly porous nature and differences in pore sizes due to chemical activation by sulphuric acid. As a result of the creation of pores, there was an increase in both the surface area and the pore-volume in the activated carbon. Fig. 1b shows the surface morphology of AC after adsorption of methylene blue dye. The micrograph shows that the dye molecules, which are well bound to the inner walls of the pores in the carbon matrix. The dye molecules enter into the deep pores of the carbon matrix and after filling the pores they reside at the entrance of the mouth of the pores and thereafter it spreads to the outer pore surface area.

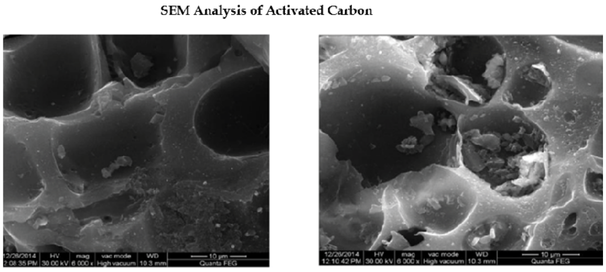

SEM Analysis of Activated Carbon

Fig. 1 Scanning electron micrograph showing surface morphology of (1a) AC before adsorption and AC (1b) after adsorption.

FTIR analysis

Fig. 2 shows the functional groups and surface properties of the adsorbent by FTIR spectra before and after adsorption of MB dye. The spectra of the adsorbent were measured in the range of 4000 cm^{-1} to 400 cm^{-1} wave number. The FT-IR spectrum (Fig. 2a) reveals the complex nature of the adsorbents as evidenced by the presence of a large number of peaks. The peak obtained at 3442.95 cm^{-1} indicates the existence of free and intermolecular bonded hydroxyl groups. Peaks around 1443.74 cm^{-1} corresponds to the C=O stretching that may be attributed to the lignin aromatic groups. The peak at 618.980 cm^{-1} corresponds to C-H bending. The peak at 1115.602 indicates S=O stretching, which confirms the chemical activation using sulphuric acid.

Evaluate the performance of AC for the removal of methylene blue dye

Effect of adsorbent dosage

In order to study the effect of adsorbent mass on the adsorption of methylene blue, a series of adsorption experiments was carried out with different adsorbent dosages with an initial dye concentration of 100 ppm. Fig. 3 show the effect of adsorbent dose on the removal of methylene blue. Along with the increase of adsorbent dosage from 0.1 g to 0.5 g, the percentage of dye adsorbed increased from 10% to 87.38%. This is because an increase in dosage increases the active sites for adsorption of dye. There may also be a decrease in the dye removal with an increase in concentration of the dye, mainly due to the fact that some sites may be saturated during the process. Above 0.3 g of adsorbent dose, the adsorption equilibrium of dye was reached and the removal ratio of the dye was constant.

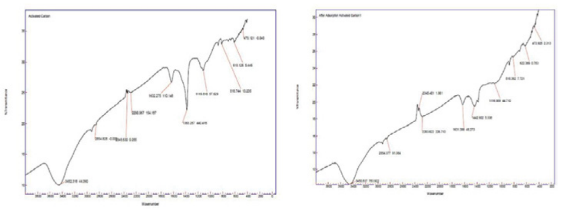

FTIR spectra of Activated Carbon

Fig. 2a Fig. 2b

Fig. 2 FTIR spectra of AC prepared from *Delonix regia* seeds (2a) before adsorption and (2b) after adsorption of Methylene blue dye.

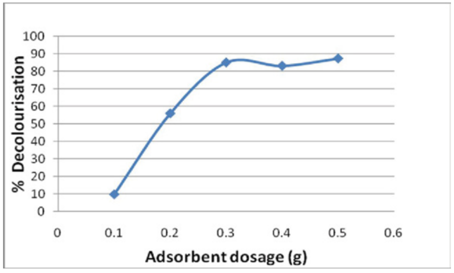

Evaluate the performance of AC for MB removal
Effect of dosage

Fig. 3 Effect of adsorbent dosage on removal of methylene blue.

Effect of contact time

A series of contact time experiments for MB has been carried out at the initial concentration of MB (100 ppm), agitation-100 rpm and at room temperature. The amount of adsorbed MB onto the activated carbon versus time (0 min to 300 min) was studied and shown in Fig. 4. The amount of dye removal increases as time increases and remains constant as equilibrium time was reached. The time taken to reach equilibrium is almost 200 min, after which the amount of decolourisation is constant. The adsorption of methylene blue onto jute fiber carbon was investigated (Senthilkumaar, *et al.,* 2005). It was reported that during adsorption of dyes, initially the dye molecules reach the boundary layer; then they have to diffuse into the adsorbent surface; and finally, they have to diffuse into the porous structure of the adsorbent. Therefore, this phenomenon will take a relatively longer contact time (Senthilkumaar, *et al.,* 2005).

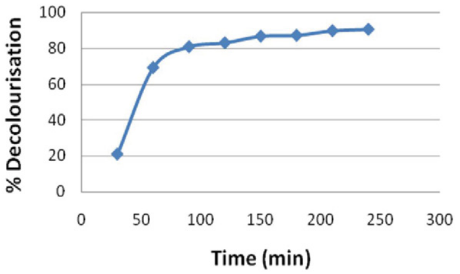


Fig. 4 Effect of contact time on removal of methylene blue dye.

Effect of pH

The pH of the aqueous solution is an important factor to affect the dye adsorption process through changing the surface charge of an adsorbent and the ionization behaviour of adsorbent and dye (Qada, *et al.*, 2006). The surface properties of the carbon change with the change in pH. The effect of the initial pH value of the solution on the adsorption process was investigated in conditions of 100 mg/l of initial dye solution concentration, 0.3 g of adsorbent, 300 min of contact time, and 150 rpm stirring speed. The pH of the dye solution was varied between 2.0 and 10.0, as it can be seen in Fig. 5-12. The amount of dye removal increases as pH increases. At high pH values, the surface of the adsorbent is negatively charged and the adsorption of dye molecules increases due to the electrostatic attraction between the surface and cationic dye molecules. At low pH values, since the surface of the adsorbent is charged positively, there is a decrease in adsorption of dye due to the electrostatic repulsive between cationic dye molecules and the surface of the adsorbent. At pH 2.0 and 10.0 there was an increase in the percentage of dye removal. At high pH the increase in adsorption can be explained as the induction of negative charges on the surface of activated carbon. However, the reason for the increase in dye adsorption at a lower pH can be as the replacement of hydrogen ions on the surface of the adsorbent with the cationic dye molecules in the solution. The negatively charged surfaces can enhance the adsorption properties of the positively charged MB cations by the adsorbents through electrostatic forces of attraction.

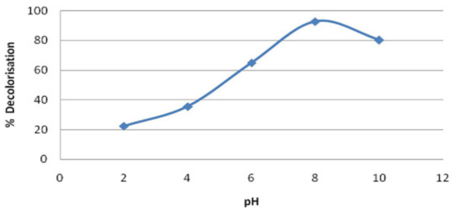
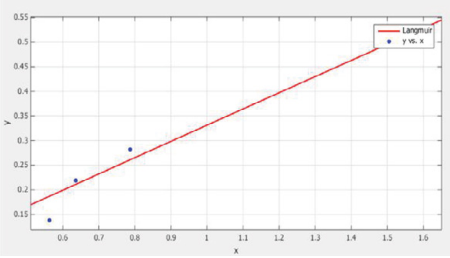
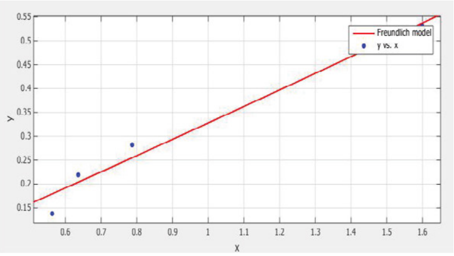

Effect of pH

Fig. 5 Effect of pH on removal of methylene blue dye.



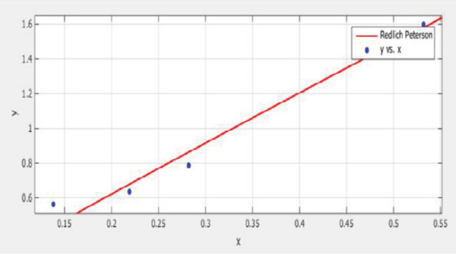
General model:
f(x)=(q*k*x)/(1+(k*x))
Coefficients (with 95% confidence bounds):
k=0.005511(-0.5776, 0.5888)
q=59.38(-6069, 6187)
Goodness of fit:
SSE: 0.002916
R-square: 0.9664
Adjusted R-square: 0.9496
RMSE: 0.03818

Fig. 6 Langmuir isotherm plot for MB adsorption.



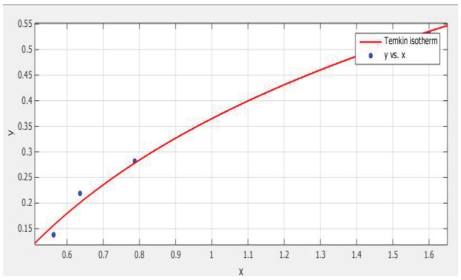
General model:
f(x)=k*x^(1/n)
Coefficients (with 95% confidence bounds):
8k=0.3279 (0.2401, 0.4156) n=0.9521 (0.4128, 1.491)
Goodness of fit:
SSE: 0.002693
R-square: 0.9689
Adjusted R-square: 0.9534
RMSE: 0.0367

Fig. 7 Freundlich isotherm plot for MB adsorption.



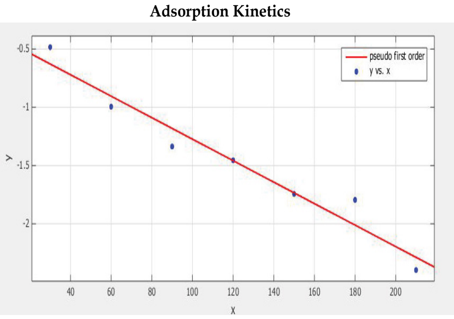
General model:
f(x)=(k*x)/(1+A*x^B)
Coefficients (with 95% confidence bounds):
A=52.44(-1.577e+007, 1.577e+007)
B=0.04826(-286.8, 286.9)
K=154.1(-4.548+007, 4.548+007)
Goodness of fit:
SSE: 0.02397
R-square: 0.965
Adjusted R-square: 0.8949
RMSE: 0.1548

Fig. 8 Redlich-Peterson plot for MB adsorption.



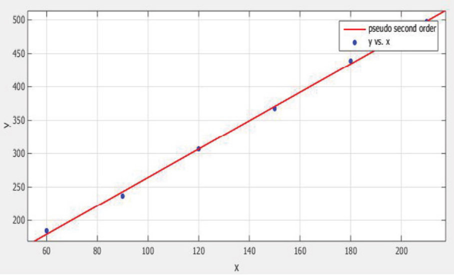
General model: f(x)=b*log(a*x)
Coefficients (with 95% confidence bounds): a=2.735 (2.057, 3.414)
b=0.3628(0.2631, 0.4625)
Goodness of fit: SSE: 0.000702
R-square: 0.9919
Adjusted R-square: 0.9879
RMSE: 0.01874

Fig. 9 Temkin isotherm plot for MB adsorption.



General model:
f(x) =log(g)-((k/2.303)*x)
Coefficients (with 95% confidence bounds) k=0.02124 (0.01566, 0.02682) q=0.7036(0.4749, 0.9323)
Goodness of fit:
SSE: 0.1119
R-square: 0.9504
Adjusted R-square: 0.9404
RMSE: 0.1496

Fig. 10 Pseudo first order kinetics.



General model:
f(x) =(1/(k*(q^2)))+((1/q)*x)
Coefficients (with 95% confidence bounds)
k=0.08734 (0.05217, 0.1225)
q=0.4704(0.4462, 0.4945)
Goodness of fit: 12
SSE: 97.62
R-square: 0.9986
Adjusted R-square: 0.9983
RMSE: 4.94

Fig. 11 Pseudo second order kinetics.

Adsorption isotherm

Langmuir isotherm

The Langmuir adsorption isotherm assumes that adsorption takes place at specific homogeneous sites within the adsorbent and has found successful application in many sorption processes of monolayer adsorption. The Langmuir isotherm can be written in the form:

$$qe = \frac{q_m K_L C_e}{1 + K_L C_e} \tag{3}$$

Where; q_m and K_L are Langmuir constants related to the adsorption capacity and energy of adsorption, respectively. For the Langmuir equation the favourable nature of adsorption can be expressed in terms of dimensionless equilibrium parameter, which is defined by:

$$R_L = \frac{1}{1 + K_L C_e} \tag{4}$$

Where, K_L is the Langmuir constant and C_o is the initial dye concentration (mg/ L). The value of R_L indicates the type of the isotherm to be either unfavorable (R_L>1), linear (R_L=favorable (0<R_L<1), or irreversible (R_L=0). The adsorption isotherm indicates how the adsorption molecules are distributed between the liquid phase and the solid phase when the adsorption process reaches an equilibrium state. The applicability of the isotherm equation is compared by judging the correlation coefficients, R^2 Table 1-3 shows the values of the parameters of the isotherms and related correlation coefficient. The correlation coefficient (R^2) of the linearized form of the equation indicates that the Langmuir model yields a fit for the experimental equilibrium adsorption data. Essential characteristics of the Langmuir isotherms can be described by a separation factor, R_L (Weber, *et al.*, 1974). The value of separation factor (R_L) indicates the nature of the absorption process (Kannan, *et al.*, 2001). In the present study, the values of R_L were in the range of 0 to 1. This indicates that the Langmuir isotherm was favourable for adsorption of MB on the activated carbon prepared from *Delonix regia* seeds. The maximum monolayer adsorption capacity q_m of the adsorbent was found to be 59.38 mg/g.

Freundlich adsorption isotherm

Freundlich model is an empirical equation based on sorption on a heterogeneous surfaces or surfaces supporting sites of varied affinities. It is assumed that the stronger binding sites are occupied first and that the binding strength decreases with the increasing degree of site occupation. The isotherm is expressed as:

Table 1. Proximate and ultimate analysis

Parameter	Value (%)
Moisture	5.21
Ash	12.87
Fixed carbon	5.95
Volatile matter	75.97
Carbon	66.27
Hydrogen	1.92
Nitrogen	2.89

Table 2. Adsorption isotherms' calculated parameters for the adsorption of MB onto AC from *Delonix regia* seeds

Model	Parameters	Values
Langmuir	K_L (l/mg)	0.0055
	q_m (mg/g)	59.38
	R^2	0.966
	R_L	0.946
Freundlich	K_f (mg/g) (L/mg)^(1/?)	0.3279
	N	0.9521
	R^2	0.9689
Redlich-Peterson	K_R (l/g)	154.1
	α_R (l/mg)	52.44
	B	0.04826
	R^2	0.965
Temkin	A (J/mol)	2.735
	B (l/g)	0.3628
	R^2	0.9919

Table 3. Pseudo first order and second order kinetic models' constants for the adsorption of MB onto AC from *Delonix regia* seeds

Order of reaction	Constant	Values
Pseudo first order	q_e (mg/g)	0.7036
	k_1 (min^{-1})	0.2124
	R^2	0.9504
Pseudo second order	q_e (mg/g)	0.4704
	k_1(min^{-1})	0.08734
	R^2	0.9986

$$q_e = K_f C_e^{1/n} \tag{5}$$

Where K_f (mg/g) (L/mg)^(1/n) and 1/n Freundlich constants related to adsorption capacity and adsorption intensity of the adsorbent, respectively. K_f can be defined as the adsorption or distribution coefficient, and it represents the quantity of the dye adsorbed for unit equilibrium concentrations. 1/n is the heterogeneity factor and n is the gauge of linearity from adsorption. Its value indicates the degree of non-linearity between the solution concentration and adsorption as follows: if n=1, the adsorption is linear; if n<1, it implies that the adsorption process is favoured by chemisorption and if n>1, the adsorption is favoured by physisorption. Further the data obtained were analyzed by Freundlich isotherm using the heterogeneity factor. The slope 1/n ranging between 0 and 1 is a measure of adsorption intensity or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero (Haghseresht, *et al.*, 1998). A value of $1/n<1$ indicates a normal Langmuir isotherm while $1/n$ above one is indicative of cooperative adsorption (Fytianos, *et al.*, 2000). From the value of n (0.9521), it is evident that the adsorption process is favoured by chemisorption.

Redlich-Peterson isotherm

The Redlich-Peterson isotherm is a combination of Langmuir-Freundlich model. The equation is given as:

$$q_e = \frac{K_R C_e}{1+\alpha_R \mathrm{C}^{\beta}{}_{\mathrm{e}}} \qquad (6)$$

where K_R is the Redlich-Peterson isotherm constant (l/g), α_R is the Redlich-Peterson isotherm constant (l/mg), and β is the exponent which lies between 0 and 1. The constant β can characterize the isotherm, if β=1, Langmuir isotherm will be preferable, while if β=0, Freundlich isotherm will be preferable. The Redlich-Peterson isotherm model predicted the value of β as 0.0482 almost 0, which is an indication that it follows Freundlich isotherm.

Temkin isotherm

Temkin isotherm contains a factor that explicitly takes into account the adsorbent–adsorbate interactions. The heat of adsorption of all the molecules in the layer would decrease linearly with coverage due to adsorbent–adsorbate interactions. The adsorption is characterized by a uniform distribution of binding energies, up to some maximum binding energy. The Temkin isotherm is expressed as:

$$q_e = B \ln(AC_e) \qquad (7)$$

B (J/mol), which is the Temkin constant related to heat of sorption, whereas *A* (l/g) is the equilibrium binding constant corresponding to the maximum binding energy. Moreover, the results from Temkin isotherm also confirm that the adsorption of methylene blue dye on to the activated carbon from *Delonix regia* seeds follow chemisorption, since the value of binding energy is less than 1 (0.3628).

Adsorption kinetics

The kinetic adsorption data were processed to understand the dynamics of adsorption process in terms of the rate constant order using MATLAB software. Thus, the kinetic of MB adsorption onto activated carbon prepared from *Delonix regia* was analyzed using pseudo-first-order and pseudo-second order kinetic models. The rate constant of adsorption is determined from the pseudo-first-order equation given by Langergren and Svenska:

$$\log(q_e - q_t)\ \log q_e - \frac{k1}{2.303}t \qquad (8)$$

where, *qe* and *qt* are the amounts of MB adsorbed (mg g^{-1}) at equilibrium and at time t (min), respectively, and k_1 (min^{-1}) is the adsorption rate constant. A plot of log (q_e-q_t) versus *t* should give a linear relationship, from which k_1 and q_e can be determined from the slope and intercept of the plot, respectively.

The pseudo-second-order kinetic model is expressed as:

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

Where, h can be regarded as the initial sorption rate (mg g^{-1} min^{-1}) as q_t/t_0, and hence, $h=k_{qe}{}^2$ and k^2 (g mg^{-1} min^{-1}) is rate constant of second-order adsorption. If the second-order kinetics is applicable, then the plot of t/q_t versus t should show a linear relationship. Values of k_2 and equilibrium adsorption capacity q_e were calculated from the intercept and slope of the plots t/q_t versus t. Values of the correlation coefficients of the pseudo-first-order and second-order kinetic model are compared, the R^2 values for pseudo-second-order kinetic model are higher, and also the experimental and theoretical values of qm are same for pseudo second order kinetics (q_{exp}=0.426 mg/g, q_{theo}=0.470 mg/g). Hence it can be concluded that the process of MB dye adsorption onto AC from *Delonix regia* seeds follows pseudo second order kinetics.

CONCLUSION

The adsorption of MB from aqueous solution onto activated carbon prepared from *Delonix regia* seeds has been studied. 90% decolonization could be achieved in 300 min with 0.3 g dosage of 2N H_2SO_4 AC and at a pH 8. Equilibrium data was best fitted to Freundlich isotherm model. The kinetic modelling of the MB adsorption onto the AC from *Delonix regia* followed the pseudo second-order rate model with the correlation coefficients of higher than 0.99. As a result of this study, AC from the *Delonix regia* can be used as an effective adsorbent for the removal of methylene blue from wastewater.

REFERENCES

Avom, J., Mbadcam, J.K., Noubactep, C. and Germain, P. 1997. Adsorption of methylene blue from an aqueous solution onto activated carbon from palm-tree cobs. *Carbon.* 35 : 365-369.

Ayg¨un, A., Yenisoy, S. and Duman, I. 2003. Production of granular activated carbon from fruit stones and nutshells and evaluation of their physical, chemical and adsorption properties. *Micropor. Mesopor. Mater.* 66 : 189-195.

El-Sheikh, A.H. and Newman, A.P. 2004. Characterization of activated carbon prepared from a single cultivar of Jordanian olive stones by chemical and physiochemical techniques. *J. Anal. Appl. Pyrol.* 71 : 151-164.

Fan, L., Zhou, Y., Yang, W., Chen, G. and Yang, F. 2008. Electrochemical degradation of aqueous solution of Amaranth azo dye on ACF under potentiostatic model. *Dyes Pigments.* 76 : 440-446.

Fytianos, K., Voudrias, E. and Kokkalis, E. 2000. Sorption-desorption behaviour of 2, 4-dichlorophenol by marine sediments. *Chemosphere.* 40 : 3-6.

G¨urses, A., Yalcın, M. and Dogar, C. 2002. Electrocoagulation of some reactive dyes: a statistical investigation of some electrochemical variables. *Waste Manage.* 22 : 491-499.

Girgis, B.S. and El-Hendawy. E. 2002. Porosity development in activated carbons obtained from date pits under chemical activation with phosphoric acid. *Micropor. Mesopor. Mater.* 52 : 105-117.

Gómez, V., Larrechi., M.L. and Callao, M.P. 2007. Callao, Kinetic and adsorption study of acid dye removal using activated carbon. *Chemosphere.* 69 : 1151-1158.

Haghseresht, F. and Lu, G. 1998. Adsorption characteristics of phenolic compounds onto coal-reject-derived adsorbents. *Energy and Fuels.* 6 : 1100-1107.

Hameed, B.H. and Ahmad, A.L. 2007. Adsorption of methylene blue onto bamboo-based activated carbon: Kinetics and equilibrium studies. *J. Hazard. Mater.* 141 : 819-825.

Kannan, N. and Sundaram, M.M. 2001. Kinetics and mechanism of removal of methylene blue by adsorption on various carbons: A comparative study. *Dyes Pigments.* 51 : 25-40.

Kavitha, D. and Namasivayam, C. 2007. Experimental and kinetic studies on methylene blue adsorption by coir pith carbon. *Bioresour. Technol.* 98 : 14-21.

Lata, H., Garg, V.K. and Gupta, R.K. 2007. Removal of a basic dye from aqueous solution by adsorption using *Parthenium hysterophorus*: An agricultural waste. *Dyes Pigments.* 74 : 653-658.

Mohan, D., Singh, K.P., Singh, G. and Kumar, K. 2002. Removal of dyes from wastewater using fly ash a low-cost adsorbent. *Ind. Eng. Chem. Res.* 41 : 3688-3695.

Muthuraman, G., Tow, T.T., Peng, L. and Ismail, N. 2008. Recovery and reuse of methylene blue from industrial wastewater using benzoic acid as a carrier. *Int. Conf. Environ. Res. Technol.* 278-282.

O¨zer, G. and Dursun, G. 2007. Removal of methylene blue from aqueous solution by dehydrated wheat bran carbon. *J. Hazard. Mater.* 146 : 262-269.

Qada, E.N., Allen, S.J. and Walker, G.M. Adsorption of methylene blue onto activated carbon produced from steam activated bituminous coal: a study of equilibrium adsorption isotherm. *Chem. Eng. J.* 124 : 103-110.

Rajeshwari sivaraj, S., Sivakumar, P., Senthilkumar, V. and Subburam, V. 2001. Carbon from cassava peel, an agricultural waste, as an adsorbent in the removal of dyes and metal ions from aqueous solution. *Bioresour. Technol.* 80 : 233-235.

Senthilkumaar, S., Varadarajan, P.R., Porkodi, K. and Subbhuraam, C.V. 2005. Adsorption of methylene blue onto jute fiber carbon: kinetics and equilibrium studies. *J. Colloid Interf. Sci.* 284 : 78-82.

Senthilkumaar, S., Varadarajan, P.R., Porkodi, K.V. and Subbhuraam, C.V. 2005 Adsorption of methylene blue onto jute fiber carbon: kinetics and equilibrium studies. *J. Colloid Interf. Sci.* 284 : 78-82.

Singh, K. and Arora, S. 2011. Removal of synthetic textile dyes from wastewaters: a critical review on present treatment technologies. *Crit. Rev.Environ. Sci. Technol.* 41 : 807-878.

Tsai, W.T., Chang, C.Y., Lin, M.C., Chien, S.F., Sun, H.F. and Hsieh, M.F. 2001. Adsorption of acid dye onto activated carbons prepared from agricultural waste bagasse by $ZnCl_2$ activation. *Chemosphere.* 45 : 51-58.

Wang, X.S., Zhoub, Y., Jiang, Y. and Sun, C. 2008. The removal of basic dyes from aqueous solutions using agricultural by-products. *J. Hazard. Mater.* 157 : 374-385.

Wang, Y., Boyjoo, Y. and Choueib, A. 2005. comparative study of dye removal using fly ash treated by different methods. *Chemosphere.* 60 : 1401-1407.

Wang, Y., Boyjoo, Y., Choueib, A. and Zhu, Z.H. 2005. Removal of dyes from aqueous solution using fly ash and red mud. *Water Res.* 39 : 129-138.

Weber, T.W. and Chakravorti, P.K. 1974. Pore and solid diffusion models for fixed bed adsorbent. *J. Am. Inst. Chem. Engrs.* 20 : 228-252.

Wu, F.C., Tseng, R.L. and Juang, R. 1999. Pore structure and adsorption performance of the activated carbons from plum kernels. *J. Hazard. Mater.* 69 : 287-302.

Yalc, N., and Sevinc, V. 2000. Studies of the surface area and porosity of activated carbons prepared from rice husks. *Carbon.* 38 : 1943-1945.