Advances in Environmental Research, *Vol. 3*, *No. 3* (*2014*) *217-230*
DOI: http://dx.doi.org/10.12989/aer.2014.3.3.217

Equilibrium and kinetic studies for the removal of cationic dye using banana pith

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(*Received November 10*, *2013*, *Revised April 18*, *2014*, *Accepted May 30*, *2014*)

Abstract. The large quantity of green cull bananas has the potential of being used industrially and, thereby, to improve banana economics and eliminate the large environmental problem presented by banana waste. Wastewaters from textile, cosmetics, printing, dying, food colouring, and paper-making industries are polluted by dyes. The adsorption of basic dye by waste banana pith was investigated by varying dye concentrations, adsorbent dose, particle size and agitation rate. The adsorption capacity was found to be maximum value of removal by using 0.1 g of sorbent with particle size 1mm at mixing speed 200 rpm for initial concentration 25 mg/l to reach value of approximate 89%. The Langmuir, Temkin and Freundlich adsorption models were used for mathematical description of the adsorption equilibrium and it was found that experimental data fitted very well to these models except Langmuir model. Adsorption of dye was applied on (pseudo-first and pseudo-second-order kinetics), and the experimental data was more fitted to pseudo second order. The results of this study showed that banana pith could be employed as effective and low-cost materials for the removal of dyes from aqueous solutions.

Keywords: banana pith; isotherms; kinetics; adsorption

1. Introduction

The amount of waste has been steadily increasing due to the increasing human population and urbanization. Waste materials are generated from manufacturing processes, industries and municipal solid wastes. Waste management one of the major environmental concerns in the world. Human activities and changes in lifestyles and consumption patterns have resulted in an increase in solid waste generation rates. Kan (2009) Waste management is the collection, transport, processing, recycling or disposal, and monitoring of waste materials. Biomass refers to the non-food part of plants. Various biomass resources include woody and herbaceous species, wood waste agricultural and industrial residues, waste paper, municipal solid waste bio-solids, waste from food processing, animal waste aquatic plants and algae, and so on. Cardak (2009), Demirbas (2009a, b), Balat (2010), (Yenikaya *et al*. 2009). Major organic components of biomass can be classified as cellulose, hemicelluloses, and lignin. Major agricultural residues include crop residue,

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http://www.techno-press.org/?journal=aer&subpage=7 ISSN: 2234-1722 (Print), 2234-1730 (Online)

Methylene blue (MB) supplied by Sigma–Aldrich (M) Sdn Bhd, It was used as an adsorbate and was not purified prior to use. Distilled water was used to prepare all the solutions and reagents. MB was chosen in this study because of its known strong adsorption onto solids. MB has a molecular mass of 373.9 g/mol, which corresponds to methylene blue hydrochloride with three groups of water. High-quality banana pith fibres were obtained from agricultural fields in Egypt. The banana pith was washed, cut into small pieces, and then dried by using liquid nitrogen by using lifelizer to remove moisture from internal capillaries to be grind to different particle size. Finally, sieved to differentiate different particle size to be used in the following experiments.

2.2 Batch equilibrium studies

Adsorption tests were performed in a set of Erlenmeyer flasks (250 ml) where 100 ml of methylene blue solutions with initial concentrations of 5-25 mg/l were placed in these flasks. Equal mass of 0.1 g of the prepared banana pith with particle size of 2 mm was added to each flask and kept in an orbital shaker at ambient temperature for 24 h to reach equilibrium. The concentrations of MB in the supernatant solutions before and after adsorption were determined using a double beam UV–Vis spectrophotometer (Shimadzu, Japan) at 655 nm. The amount of adsorption at equilibrium, q_e (mg/g), was calculated by

$$q_e = (C_0 - C_e)V/W \tag{1}$$

where C_0 and C_e (mg/l) are the liquid-phase concentrations of dye at initial and equilibrium, respectively. V is the volume of the solution (l) and W is the mass of dry adsorbent used (g).

2.3 Batch kinetic studies

The procedure of kinetic tests was basically identical to those of equilibrium tests. Adsorption tests were performed in a set of Erlenmeyer flasks (250 ml) where 100 ml of methylene blue solutions with initial concentrations of 5-25 mg/l were placed in these flasks. Equal mass of 0.1 g of the prepared activated carbon with particle size of 200 μm was added to each flask and kept in an orbital shaker at ambient temperture. The aqueous samples were taken at time intervals and the concentrations of MB were similarly measured. The amount of adsorption at time t, q_t (mg/g), was calculated by

$$q_t = (C_0 - C_t)V/W \tag{2}$$

where C_0 and C_t (mg/l) are the liquid-phase concentrations of dye at initial and at any time t, respectively. V is the volume of the solution (l) and W is the mass of dry adsorbent used (g).

3. Results and discussion

3.1 Effect of contact time and initial dye concentration on adsorption equilibrium

The adsorption of MB on banana pith at different initial concentrations ranged from (5-25 mg/l), was studied as a function of contact time ranging from 5-180 min in order to determine the equilibrium time. Fig. 1 shows time course of the adsorption equilibrium of MB onto adsorbent. It can be observed that the dye uptake increased with time and, at some point in time, reached a

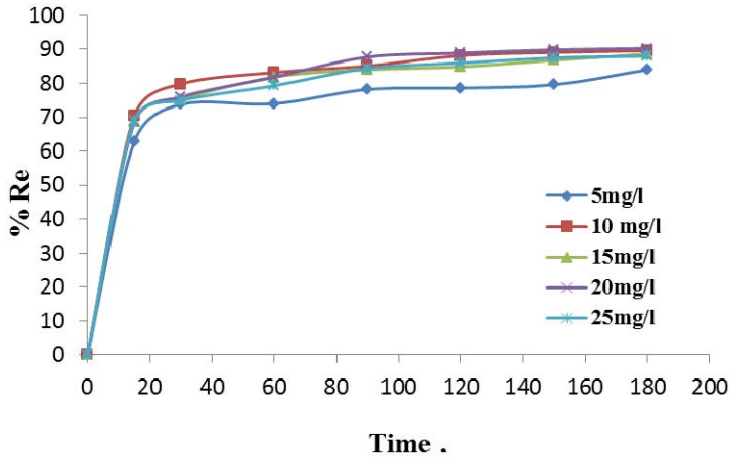


Fig. 1 Influence of dye concentration on adsorption of MB by banana pith (sorbent dose: 0.1 g; particle size: 1 mm; stirring speed; 200 r.p.m; contact time: 3 h)

constant value where no more dye was removed from the solution. At this point, the amount of dye being adsorbed onto the adsorbent was in a state of dynamic equilibrium with the amount of dye desorbed from the adsorbent. The time required to attain this state of equilibrium was termed the equilibrium time and the amount of dye adsorbed at the equilibrium time reflected the maximum dye adsorption capacity of the adsorbent under these particular conditions. It indicated that the contact time needed for MB solutions with initial concentrations of 5-25 mg/L to reach equilibrium was 120 min. However, the experimental data were measured at 180 min to make sure that full equilibrium was attained. The rapid adsorption observed during the first 60 min is probably due to the abundant availability of active sites on the adsorbent surface, and with the gradual occupancy of these sites, the sorption becomes less efficient, also by increasing MB concentration the percentage removal increase to reach 90% after 180 min for 25 mg/l dye concentration.

3.2 Effect of adsorbent dose

The adsorption of methylene blue onto banana pith powder was studied by varying the adsorbent quantity (0.05, 0.1, 0.2, 0.3 and 0.4 g) in the test solution while keeping the initial dye concentration 25 mg/L, ambient temperature with 1mm particle size and 200 rpm constant at all different time intervals until 180 min. As one was expected, the percentage of dye removal with increasing adsorbent dosage increased and equilibrium time was decreased with adsorbent dose Fig. 2. The adsorption ratios of dyes increased from 52 to 82%, in MB at the first 15 min. and increasing of dye removal still increase to reach nearly constant values after 120 min. Increase in the adsorption with adsorbent dose can be attributed to increased adsorbent surface area and availability of more adsorption sites. But unit adsorption decreased with increase in adsorbent dose. For banana pith, unit adsorption was decreased from 82.4 mg/g to 52.4 mg/g as the adsorbent dose was increased from 0.05 to 0.4 g/100 ml in the test solution. This may be attributed to overlapping or aggregation of adsorption sites resulting in decrease in total adsorbent surface area available to methylene blue and an increase in diffusion path length. Equilibrium time was lesser at higher

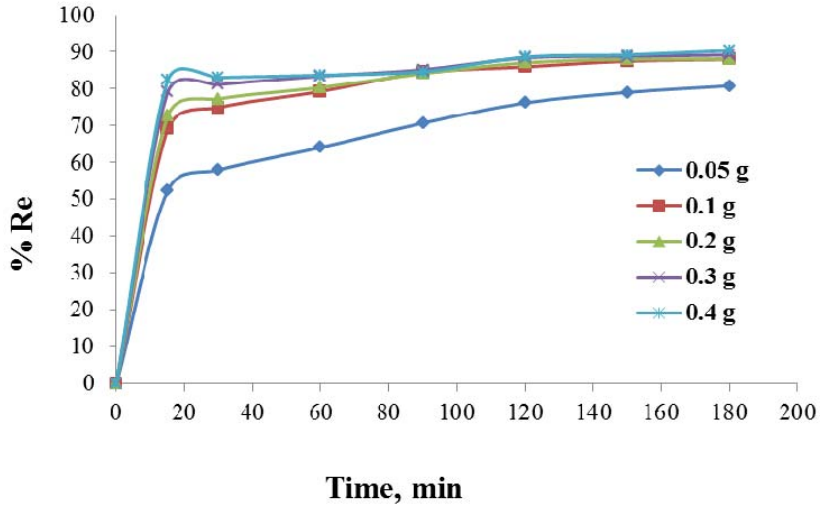


Fig. 2 Effect of sorbent dose on adsorption of MB by banana pith (dye concentration: 25 mg/l; particle size 1mm; stirring speed 200 r.p.m; contact time: 3 h)

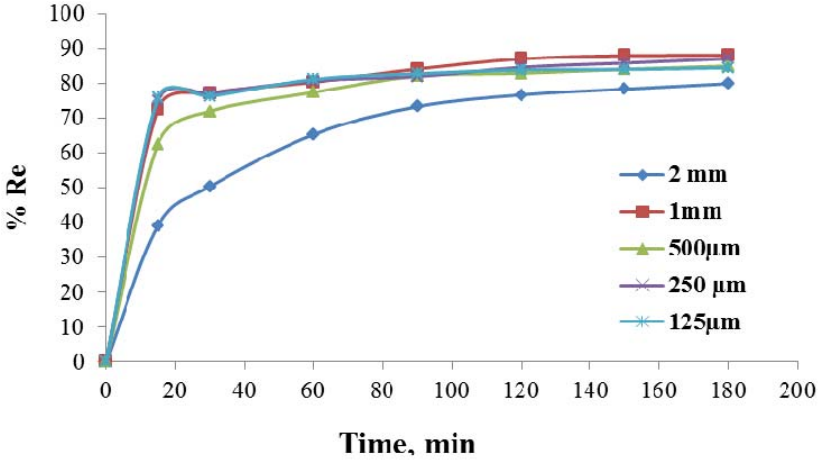


Fig. 3 Effect of sorbent particle size on adsorption of MB by banana pith (dye concentration: 25 mg/l; stirring speed: 200 r.p.m ; sorbent dose: 0.1 g; contact time: 3 h)

adsorbent doses. Finally, from the results it can be concluded that there is a slight different between the sorbent doses except 0.05 g. There for (0.1 g) of sorbent was chosen as an optimum amount for the latter experiments.

3.3 Effect of sorbent particle size

The adsorption of MB on banana pith at different particle sizes was studied as a function of contact time in order to determine the equilibrium time, while keeping the initial dye concentration 25 mg/L, ambient temperature with 0.2 g of the sorbent and 200 rpm constant at all different time

intervals until 180 min. Fig. 3 shows the effects of sorbent particle size on biosorption percentages of dyes. The ratios of dyes sorbed increased as the sorbent particle size was decreased from 76.7% at 2 mm to 84% at 125 μm after 120 min, as the rapid increase in dye removal at the first 30 min, after that small increase was observed to reach equilibrium at 120 min. The ratios of dyes sorbed had neared the maximum values; this may be due to the increase of surface area by decreasing particle size which increases the contact surface between sorbent molecule and dye solution which improve the dye sorption process. The sorbent particle size in 1 mm was used in all other parameter experiments.

3.4 Effect of agitation rate

The variation of dyes uptake with agitation rate is shown in Fig. 4. Uptake of methylene blue increase as the agitation rate increased from 100 to 400 r.p.m in the test solution while keeping the initial dye concentration 25 mg/l, 0.1 g of sorbent, ambient temperature and 1mm particle size constant at all different time intervals until 180 min. Increasing agitation rate decreases the film resistance to mass transfer surrounding the sorbent particles thus increasing sorption. However, uptake of MB after little time becomes independent of agitation rate; this is probably associated with the rapid uptake of the dye. The minimal effect of agitation rate on the dye sorption by sorbent indicated also that external mass is not the sole rate-limiting factor in a well agitated system, so it can conclude that agitation rate is not effective factor for dye removal for time more than 60 min and 200 rpm will be the best agitation rate to be stable at lower time and to save power consumption by using high speeds without efficient difference in percentage dye removal.

3.5 Adsorption isotherm modelling

Analyses of isotherm data are important in order to develop an equation which accurately represents the results and could be used for design purposes. Several isotherms are available for such analysis. Most often three models for equilibrium description of the adsorption behavior of

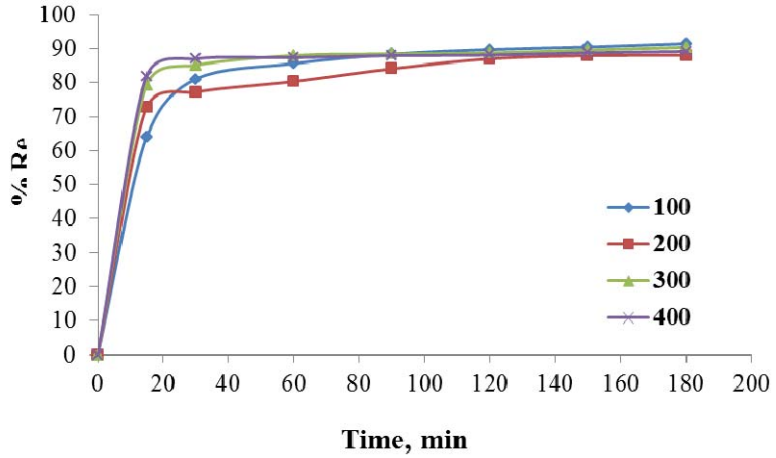


Fig. 4 Effect of agitation rate on adsorption of MB by banana pith (dye concentration: 25 mg/l; sorbent dose: 0.1 g; particle size: 1 mm; contact time: 3 h)

the systems are applied.

3.5.1 Langmuir model

Langmuir's isotherm model suggests that the uptake occurs on homogeneous surface by monolayer sorption without interaction between adsorbed ions. The linear form of Langmuir isotherm equation is represented by the following equation

$$q_e = \frac{K_L \cdot C_e}{1 + .a_L C_e} \qquad \frac{C_e}{q_e} = \frac{1}{K_L} + \frac{a_L}{K_L} \cdot C_e \tag{3}$$

Where q_e is the amount of dye adsorbed per unit mass of sorbent particles at equilibrium; C_e is the equilibrium phase concentration of dye and a_L, K_L are Langmuir constant. Therefore, a plot of C_e/q_e versus C_e, gives a straight line of slope $\frac{a_L}{K_L}$ and intercept $\frac{1}{K_L}$. Fig. 5 shows the experimental data that were fitted by the linear form of Langmuir model. The values of a_L *and* K_L are listed in Table 1 with their uncertainty and their determination coefficients, R^2.

3.5.2 Freundlich model

Freundlich isotherm describes the adsorption equation for non-ideal adsorption that involves heterogeneous adsorption. This empirical isotherm is expressed by the following equation

$$q_e = K_F \cdot C_e^{\frac{1}{n_F}} \tag{4}$$

The equation is conveniently used in the linear form by taking the logarithm of both sides as

$$\log q_e = \log K_F \cdot \frac{1}{n_F} . \log C_e \tag{5}$$

Where q_e is the amount of dye adsorbed per unit mass of sorbent particles at equilibrium; C_e is the

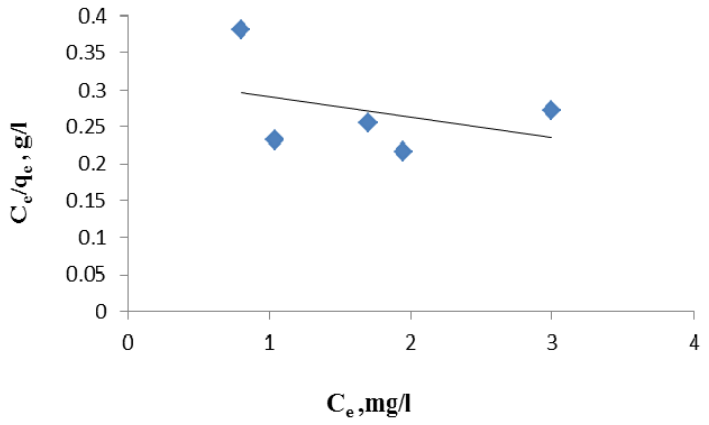


Fig. 5 Langmuir isotherm of MB adsorption onto banana pith

Table 1 Isotherm parameters for removal of methylene blue by peanut hulls powder

Isotherm	Parameters	
Freundlich model	$1/n_f$	0.833
	K_f	17.37
	R^2	0.9132
Temkin model	A	1.775
	B	6.6741
	R^2	0.974
Langmuir model	K_L	3.129
	a_L	-0.088
	R^2	0.1423

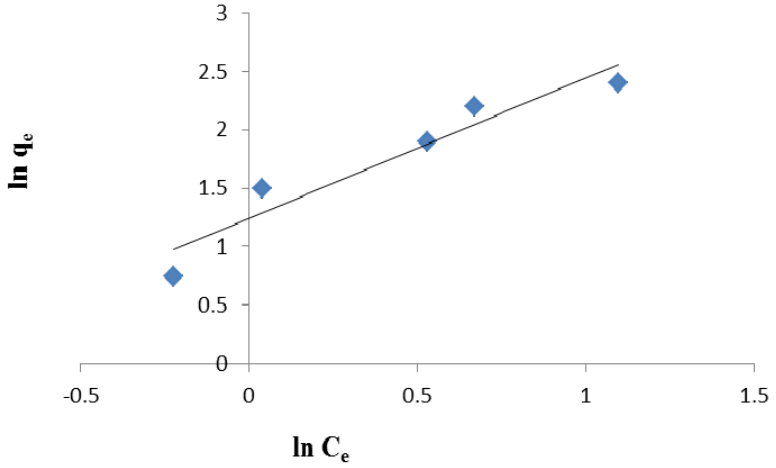


Fig. 6 Freundlich isotherm of MB adsorption onto banana pith

equilibrium phase concentration of dye, Freundlich constants, K_F and $1/n_F$, are related to adsorption capacity and intensity of adsorption, respectively. The values of n_F and K_F can be calculated from the slope and intercept of the plot of log q_e versus log C_e derived from Eq. (4) as shown in Fig. 6. The magnitude of the exponent $1/n_F$ gives an indication of the favourability of adsorption.

The applicability of the Freundlich sorption isotherm was also analyzed, using the same set of experimental data, by plotting log (q_e) versus log (C_e). The constant values obtained from linear Freundlich isotherm plot for the adsorption of MB onto banana pith particles are presented in Table 1. The applicability of linear forms of Freundlish model to banana pith was proved by the high correlation coefficients $R^2 > 0.9$. This suggests that the Freundlish isotherm provides a good model of the sorption system. The $1/n$ is lower than 1.0, indicating that MB is favorably adsorbed by banana pith.

3.5.3 Tempkin model

Tempkin adsorption isotherm model was used to evaluate the adsorption potentials of banana pith for MB dye as shown in Fig. 7. The derivation of the Tempkin isotherm assumes that the fall in the heat of sorption is linear rather than logarithmic, as implied in the Freundlich equation. The Tempkin isotherm has commonly been applied in the following form

$$q_e = \left(\frac{RT}{b}\right)\ln(A\ C_e) \tag{6}$$

Where $RT/b = B$. T is the absolute temperature in Kelvin and R is the universal gas constant, 8.314 J (mol K)$^{-1}$. The constant b is related to the heat of adsorption. A plot of q_e versus ln C_e yielded a linear line.

Examination of the data shows that the Tempkin isotherm fitted well the MB adsorption data for banana pith. The linear isotherm constants and coefficients of determination are presented in Table 1. The correlation coefficients R^2 obtained from Tempkin model were comparable to that obtained for Freundlich equation, which explain the applicability of Tempkin model to the adsorption of MB onto banana pith.

Finally, as shown in Table 1 which summarizes the results of the isotherm constants for the three different equilibrium isotherms tested. On the basis of the correlation coefficients (R^2), Langmuir model yields ($R^2 = 0.1423$) while Freundlich model ($R^2 = 0.9132$) and Tempkin isotherm was ($R^2 = 0.974$) which seemed to represent the equilibrium adsorption data with better fit for Freundlich and Tempkin isotherms.

3.6 Kinetic models applied to the adsorption of MB onto banana pith

It is important to be able to predict the rate at which contamination is removed from the aqueous solutions in order to design an adsorption treatment plant. Several kinetic models are available to understand the behavior of the adsorbent and to examine the controlling mechanism of the adsorption process and also to test the experimental data. The conformity between experimental experimental data and the model-predicted values was expressed by the correlation coefficients

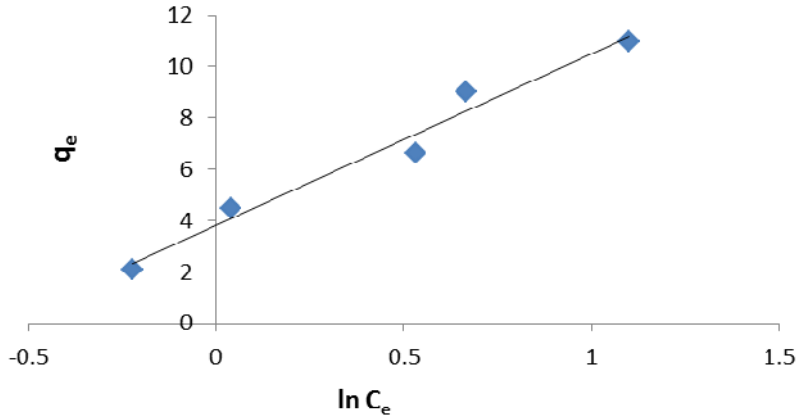


Fig. 7 Tempkin isotherm of MB adsorption onto banana pith

(R^2, values close or equal to 1, the relatively higher value is the more applicable model). In the present investigation, the adsorption data were analyzed using two kinetic models: the pseudo-first-order and pseudo-second-order kinetic models. (Karaoğlu *et al*. 2010) The Lagergren rate equation is one of the most widely used adsorption rate equations for the adsorption of solute from a liquid solution. The pseudo-first-order kinetic model of Lagergren may be represented by

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \tag{7}$$

Integrating this equation for the boundary conditions $t = 0$ to $t = t$ and $q = 0$ to $q = q_t$, give

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303}t \tag{8}$$

where q_e and q_t are the amounts of MB adsorbed (mol/g) at equilibrium and at time t (min), respectively, and k_1 is the rate constant of adsorption (min^{-1}). The values of q_e and k_1 can be determined from the intercept and the slope of the linear plot of $\ln(q_e - q_t)$ versus t.

The pseudo-second-order model based on equilibrium adsorption is expressed by the following equation

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \tag{9}$$

Rearranging the variables in Eq. (9) gives

$$\frac{dq_t}{(q_e - q_t)^2} = k_2 dt \tag{10}$$

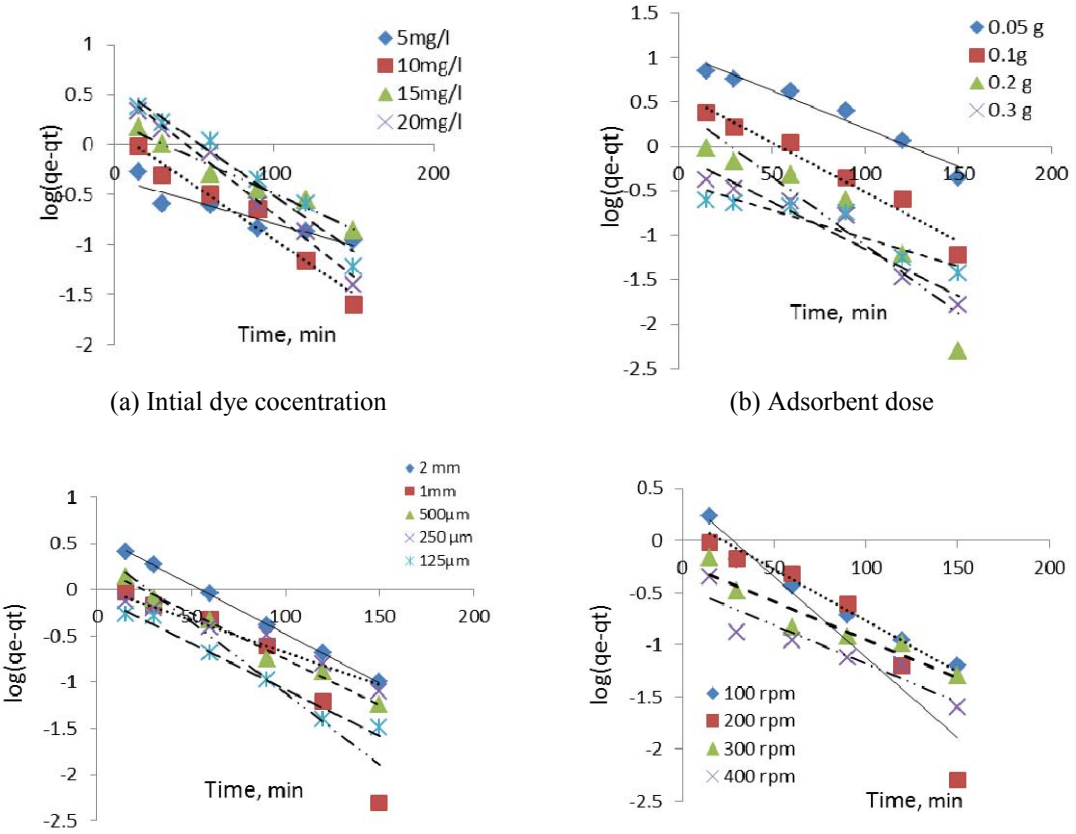
Taking into account, the boundary conditions $t = 0$ to $t = t$ and $q = 0$ to $q = q_t$, the integrated linear form of Eq. (10) can be rearranged to obtain

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}t \tag{11}$$

where k_2 is the second-order constant (g/(mol min)) and can be determined from the slope and intercept of the linear plot of t/q_t versus t. (Prasad and Santhi 2012). Table 2 lists the results of the kinetic parameters calculated using the pseudo-first-order and pseudo-second-order models. The curve fitting plots of log ($q_e - q_t$) versus t does not show good results for the sorption period Fig. 8. The correlation coefficients, R^2, given in Table 2, of the pseudo-first-order model are low and underestimated the adsorbed amount of the dye at equilibrium for MB. The calculated q_e values from the first-order kinetic model do not give reasonable values, which are too low compared with experimental q_e values.

This finding suggests that the sorption of MB on banana pith is not diffusion-controlled and the process does not follow the pseudo-first-order adsorption rate expression of Lagergren.

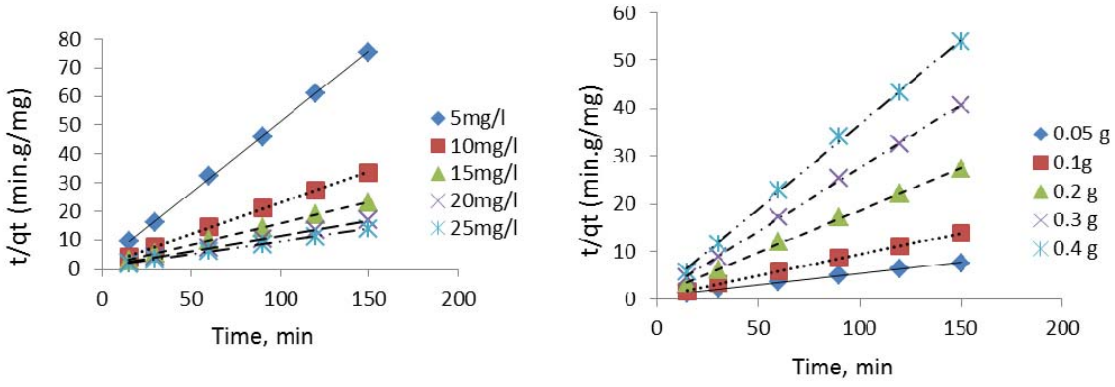
The plots of t/q_t versus t give a straight line for all parameters studied as seen from Figs. 8 and 9 confirming the applicability of the pseudo second-order equation. The rate of pseudo-second-order reaction is dependent on the amount of solute adsorbed on the surface of adsorbent and the adsorbed amount at equilibrium. Values of k_2 and equilibrium adsorption capacity q_e were



(a) Intial dye cocentration (b) Adsorbent dose

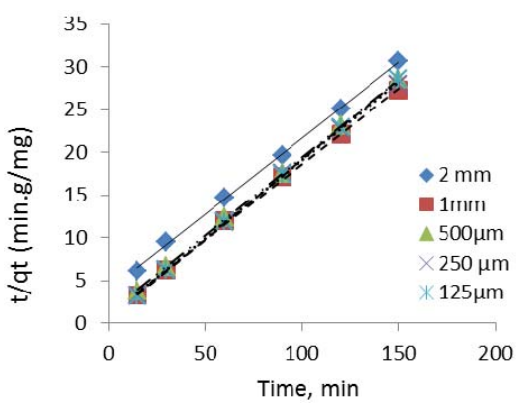
(c) Particle size (d) Mixing speed

Fig. 8 Pseudo-first-order kinetics for adsorption of MB onto banana pith at different parameters

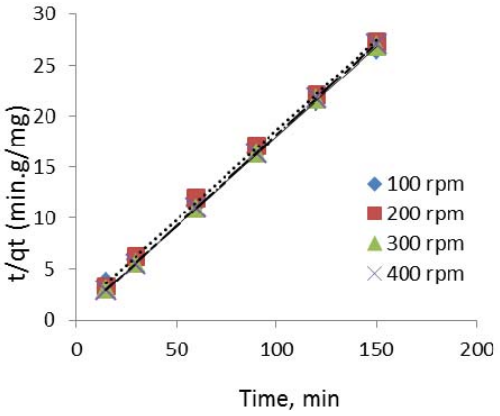


(a) Intial dye cocentration (b) Adsorbent dose

Fig. 9 Pseudo-second-order kinetics for adsorption of MB onto banana pith at different parameters



(c) Particle size



(d) Mixing speed

Fig. 9 Continued

Table 2 Comparison of the pseudo-first-order, pseudo-second-order adsorption rate constants and calculated and experimental q_e values obtained at different initial MB concentrations for different parameters studied

Dye system	$q_{e,exp}$ (mg/g)	Pseudo first order model			Pseudo second order model		
		K_1	q_{e1}	R^2	K_2	q_{e2}	R^2
		min^{-1}	mg/g		g/mg.min	mg/g	
(1) Different concentrations (W = 0.1 g; n = 200 rpm; P = 1 mm)							
C_0 = 5 mg/l	2.1	0.010	0.45	0.862	0.109	2.043	0.9996
C_0 = 10mg/l	4.48	0.025	1.375	0.962	0.042	4.583	0.9996
C_0 = 15 mg/l	6.65	0.016	1.655	0.9713	0.0285	6.693	0.9998
C_0 = 20 mg/l	9.025	0.029	3.712	0.9872	0.0158	9.381	0.9996
C_0 = 25 mg/l	11	0.025	4.03	0.9641	0.014	11.325	0.9994
(2) Sorbent amount (n = 200 rpm; P = 1 mm; C_0=25mg/l)							
w_0 = 0.05 g	20.21	0.0198	11.392	0.9591	0.00336	21.186	0.9945
w_0 = 0.1 g	11	0.0256	4.033	0.9641	0.014	11.325	0.9994
w_0 = 0.2 g	5.505	0.0355	2.654	0.8812	0.184	5.663	0.9992
w_0 = 0.3 g	3.712	0.0242	0.792	0.9233	0.0739	3.768	0.9993
w_0 = 0.4 g	2.825	0.0145	0.394	0.8608	0.1149	2.82	0.9988
(3) Particle size (W = 0.2 g; n = 200 rpm; C_0 = 25 mg/l)							
P_0 = 2 mm	5	0.024	3.765	0.9996	0.0083	5.6179	0.9992
P_0 = 1 mm	5.5	0.0355	2.654	0.8812	0.032	5.663	0.9992
P_0 = 500 μm	5.313	0.0228	1.756	0.9865	0.0278	5.477	0.9998
P_0 = 250 μm	5.45	0.0161	1.047	0.09702	0.0465	5.462	0.9994
P_0 = 125 μm	5.28	0.023	0.844	0.977	0.0653	5.350	0.9999

Table 2 Continued

Dye system	$q_{e,exp}$ (mg/g)	Pseudo first order model			Pseudo second order model		
		K_1	q_{e1}	R^2	K_2	q_{e2}	R^2
		min^{-1}	mg/g		g/mg.min	mg/g	
(4) rpm (W = 0.2 g; P = 1 mm; C_0 = 25 mg/l)							
rpm_0 = 100	5.72	0.023	1.667	0.9649	0.0326	5.663	0.9992
rpm_0 = 200	5.51	0.0355	2.654	0.8812	0.1527	5.580	1
rpm_0 = 300	5.65	0.0168	0.613	0.9078	0.0872	5.663	1
rpm_0 = 400	5.75	0.0168	0.357	0.8635	0.1527	5.580	1

calculated from the intercept and slope of the plots of t/q_t versus t, respectively. The straight lines with extremely high correlation coefficients for R^2 the pseudo-second-order kinetic model compared to those for the pseudo-first-order onto banana pith strongly suggest that all the adsorption systems are a pseudo-second-order model.

The calculated q_e values also agree very well with the experimental data in the case of pseudo-second-order kinetics.

4. Conclusions

The removal of methylene blue from simulated wastewater using banana pith has been investigated under different experimental conditions in batch mode. The adsorption of methylene blue was dependent on adsorbent surface characteristics, adsorbent dose and methylene blue concentration in the wastewater. Maximum dye was removed within 30 min of the start of every experiment. The agitation rate slightly influences the removal kinetics. The adsorption process was very fast, and it reached equilibrium in 2 h of contact. The equilibrium solid-phase concentration of methylene blue (q_e, w/w) decreased with increasing adsorbent (banana pith) concentration is mainly attributed to the unsaturation of the adsorption sites through the adsorption process. Both Tempkin and Freundlich isotherms provide good correlations for the adsorption of methylene blue compounds onto banana pith. The pseudo-second-order kinetic model was found to represent the experimental data better than pseudo-first-order model and intraparticle diffusion with a better fit ($R_2 > 0.99$). The data may be useful for designing and fabricating an economically cheap treatment process using for the removal of methylene blue from dilute industrial effluents.

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