Treatment of Rhodamine B Contaminated Wastewater Using Lagerstroemia Speciosa Leaf: A Potential Low Cost Adsorbent

M. A. Rahman*, N. Rahman, M. T. Uddin and M. A. Islam

*Department of Chemical Engineering and Polymer Science,
Shahjalal University of Science and Technology, Sylhet 3114, Bangladesh*

Abstract

The potentiality of *Lagerstroemia speciosa* Leaf (LSL) as a low cost adsorbent was investigated for the removal of a cationic dye Rhodamine B (RB) from aqueous solution in batch and column mode. The adsorbent was characterized by Scanning Electron Microscope and Fourier Transform Infra-Red (FTIR) spectrometer. The pH at zero point charge (pH_{pzc}) of the adsorbent was determined by pH drift method and was found to be 6. The effect of operational parameters such as adsorbent dosage, pH, initial dye concentration and contact time on the adsorption of dye onto adsorbent was studied. The optimum pH for the adsorption of RB was found to be 3.1. The Langmuir and Freundlich isotherm models were used for modeling the adsorption equilibrium data. The adsorption isotherm fitted Langmuir model confirming monolayer adsorption of RB with an adsorption capacity of 66 mg/g. The kinetics of adsorption was analyzed using pseudo-first order and pseudo-second order kinetic models, and the sorption kinetics was found to follow pseudo-second-order kinetic model. In order to investigate the industrial applicability of LSL, fixed bed column studies were also carried out. The effect of adsorbent bed height on the shape of breakthrough curve was investigated. The bed capacity, percentage removal and service time increased with the increase in bed depth due to the increase in the total surface area. The experimental data were modeled using Thomas model and Bed Depth Service Time model to predict the column performance and breakthrough curves and found to be in good agreement with the experimental data.

Keywords: *Adsorption, Rhodamine B, Lagerstroemia speciosa leaf, Isotherms, Kinetics.*

1. Introduction

Today's rapid economic development of Bangladesh is marked by rapid industrialization, which leaves adverse effect to environment especially to water. Among all the pollutants, the contribution of dye to the water pollution is tremendous. Synthetic dyes are widely used in industries like dyestuffs, textile, paper, food, leather, polymer industries and also in photoelectron chemical cells. The effluent of these industries contain significant amount of color which affects the aesthetic quality of water, diminish the photosynthetic activity in aquatic life due to reduced light penetration and may also be mutagenic, toxic to some aquatic life because of the presence of aromatics, metals, chlorides etc. [1,2]. Conventional wastewater system fails to treat dye contaminated wastewater due to their high stability against light, temperature, chemical and microbial attacks[3]. Recently government has imposed strict legisla-

Corresponding author: anis.cep@gmail.com

tion on the limit of color discharge to water body. Therefore, there is a considerable urgent need to find cost effective treatment method potentin eliminating dyes from wastewater. A wide range of methods havebeen developed for theremoval of synthetic dyes from wastewater todecrease their impact on the environment. The technologiesinvolve adsorption on inorganic or organic supports, decolorization by photocatalysis, and/or by advanced oxidation processes,microbiological or enzymatic decomposition, etc. Amongst different techniques, adsorption techniques have gained favor recently due to their efficiency in the removal of pollutants too stable for conventional methods.Adsorption is a surface phenomenon which primarily deals with the utilization of surface forces,andconcentratesmaterials to be removed on the surface of solid bodies. It is a physico-chemical process whichinvolves different interactions such aselectrostaticinteraction, Van der Waals forces, ionic and covalent bondsdepending on the nature of the adsorbent and the origin of dyes. Adsorption produces high quality product,treats high volume effluent,and is a process which is economically feasible.Though activated carbon is reported to be very effective for adsorbing cationic, mordant and reactive dyes among the adsorbents[4,5], its application is limitedtoits high associated cost and disposal problem afterexhaustion.So modern investigation is commended to find a renewable, cost effective source of adsorbentto replace costly activated carbon, which is mainly a cellulosic biomass. The used biomaterial requires no regeneration as it is abundantly available in the environment. Moreover it can be incinerated to gain energy and the resulting carbon can be used as adsorbent.In this study, Rhodamine B (RB) is selected as a model dye in order to investigate the potentiality of (LSL) as an adsorbent. *Lagerstroemia speciosa* plant is commonly known as pride of India, which is a tropical plant abundantly available inBangladesh. The waste dry leaves have no practical use or commer-

cial importance. The use of LSL as an adsorbent is not reported previously by any researchers. Therefore, this is a novel source of adsorbent which is cheap, renewable, and abundantly available in the environment. Rhodamine B is a xanthene dye which is widely used in different industries and is reported to be carcinogenic for containing four N-ethyl linkages in its structure[6]. It also causes skin irritation, respiratory diseases, eye damage and in its worst form it can cause cancer [7,8]. Thus removal of RB from industrialeffluent is of significant commercial and environmental importance. The influence of principal operational parameters like initial concentration, contact time; equilibrium and kinetic studies; effect of bed height in assessing column performance were carried out for thesorption process to justify thepotentiality of the LSL for the removal of RB from its effluent.

2. Materials and Methods

2.1 Preparation of Adsorbent

LSLs were collected from local area in Sylhet. The collected leaves were washed followed by boilingin distilled water (for three days) to completely remove lignin, chlorophyll and other pigment components of the leaves. Subsequently, the boiled leaves were dried in a hot air oven (Memmert, Model: 500) at 100°C for about 24 hours and then crushed in a blender to powdered form.

2.2 Adsorbate

Stock solution of 100 mg/LRB (chemical formula-$C_{28}H_{31}ClN_2O_3$, molecular weight-479.02 g/mol) was prepared by dissolving 0.1 g analytical grade RB (Loba Chemic Pvt. Ltd., India) in 1L of distilled water, which was further diluted to desired extent to obtain desired concentration. The absorption spectrum and concentration of RB was analyzedby UV-vis spectrophotometer (Model: UV-2600) at 554 nm (maximum wavelength of absorbance). The pH values of solutions were adjusted using 0.1 N HCl and NaOH solution (Merck Specialities Private Ltd., Mumbai).

2.3 Characterization of Adsorbent

Scanning Electron Microscopy (SEM) (Model: JEOL JSM-6490LA,) was observed by taking he surface morphology (texture and porosity) of the prepared adsorbent. The infrared spectrums of the LSL adsorbent before and after dye uptake were obtained using Fourier transform infrared (FTIR) spectroscopy (IRPrestige-21, Shimadzu, Kyoto, Japan) operating in the range of 4000-400 cm-1. FTIR characterization was performed in order to identify chemical functional groups present on the LSL that might be involved in the RB uptake procedure. Point of zero charge,pHPZCof LSL adsorbent was determined by pH drift method[9]. 0.20 g of adsorbent was introduced in differentErlenmeyer flask of 250 ml containing 200 ml of 0.01 M NaCl solution. pH values of thesolutions were adjusted in range of 3 to 12 by adding 0.1 M HCl and NaOH solutions. These flasks were then shaken in a flask shaker machine (Stuart Scientific Co. Ltd. UK, Model:SF1) for 48 hours and the final pH of thesolutions weremeasured. The point of intersection of the plot of pHfinalminus pHinitialversus pHinitialwas recorded as pHPZCof LSL.

2.4 Batch Adsorption Study

Adsorption isothermstudies were carried out by contacting 0.20 g of LSL with 200 ml of RB solution (at pH 3.1) of different initial concentrations (50, 100, 150, 200, 250, 300 mg/L) in 200 ml Erlenmeyer flasksat 25ºC. The samples were then shakenat a constant oscillation of 350 osc/min for 4 hours. After equilibrium, the equilibriumadsorption density, q_e(mg/g),was determined by the following equation

$$q_e = \frac{(C_0 - C_e)V}{W} \qquad (1)$$

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 in liter and W is the mass of theadsorbent in gram. Adsorption kinetics experiment as well as the effect of contact time on the sorption process were carried out by bringing 0.2 g LSL in contact with 200 ml RB solution (at pH 3.1) of different initial concentrations (100,200, 300 mg/L) in different 200 ml Erlenmeyer flasks. The samples were then shaken at a constant oscillation speed of 350 osc/min for 3 hours. Samples were pipetted out at different time intervals, centrifuged and then theconcentration in the supernatant solution was analyzed as before.

2.5 Column Experiment

Fixed bed adsorption experiments were conducted in a plastic cylindrical column of 3.2 cm internal diameter. The up flow of solution to the columnwas supported from a reservoirby a diaphragm pump (DENG YUAN-Diaphragm pump, Model: 2500). The adsorbent bed was supported by two 1 cm high layer of glass woolplaced attwo end of the adsorbent bed.

To investigate the impact of bed height on the fixed bed adsorption process 16.10, 19.85, 26.25 g of the adsorbent was placed in the column to produce the bed height of 5.5, 7.5, 10.5 cm respectively. Rhodamine B solution of 300 mg/Lwas pumped upward through the column at a constant flow rate of 15ml/min, andthe effluent collected from the exit of the column at different time intervals were analyzed for final concentration of RB till the effluent RB concentration reached a value of 99% of its initial concentration.

3. Results and Discussion

3.1 SEM and FTIR Characterization of Adsorbent

The surface topography of theLSLadsorbenthas been analyzed by SEM. SEM image (Fig.1.) shows rough and corrugated surface of the adsorbent. The adsorption of the RB occurs on this surfaceof the adsorbent. FTIR analysis of free and loaded adsorbent (Fig. is not shown) depicts that the shift in maximum absorbance occurs for O–H stretching, C–H group stretching, C=O stretching of unionized carboxylate stretching of carboxylic acid, C=O stretching of carboxylic acid withintermolecular hydrogen bond, aromatic C=C stretching and C–O stretching. It indicates that the
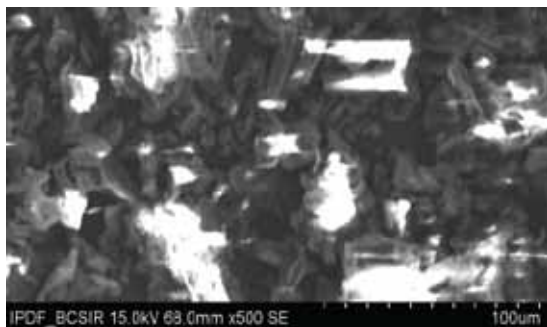
Fig. 1: SEM micrograph of LSL

Fig. 2: Determination of point of zero charge

functional groups present in the LSL, such as hydroxyl, carboxylate, carbonyl groups could be potential adsorption sites for uptake of RB from its aqueous solution.

3.2 Determination of Point of Zero Charge

The pH at which the sum of all the surface positive charges balances the sum of all the negative charges is called the point of zerocharge, pH_{pzc}. The choice of working solution pH is strongly dependent on the pH_{pzc}. At a pH of the solution below pH_{pzc}, the surface of the sorbent becomes positively charged and attracts anions from the solution. When the solution pHis greaterthan pH_{pzc}, the surface of the adsorbent is negatively charged and attract cations.

The point of zero charge of LSL adsorbent has been determined to be 6 from pH_{final} minus $pH_{initial}$ versus $pH_{initial}$ plot as shown in Fig. 2. Due to the zwitterionic nature of the dye solution, the working solution pH is kept at 3.1 which is the pK_a value of RB [10]. At pH<3.1, the RB is in its cationic form (RBH^+) and the adsorbent surface is positive as the pH< pH_{pzc}, thus a repulsion force leads to decreased adsorption. At pH>3.1 the adsorbent surface becomes less positively charged up to pH 6 and above pH 6 the surface negativity continues to increase. Also, at pH>3.1, the total negative charge of RB zwitterion continues to increase, causing increased repulsion between adsorbent and adsorbate leading to decreased adsorption density [11].
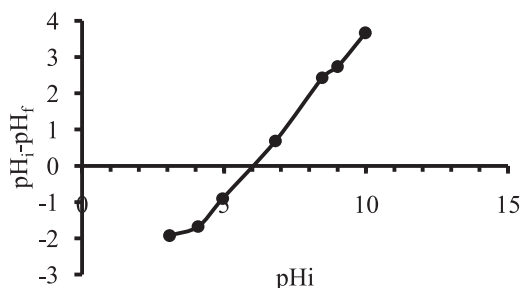
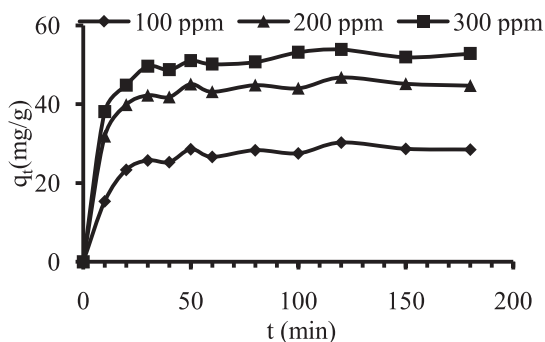### 3.3 Effect of Initial Dye Concentration and Contact Time

The dependency of the uptake of RB on LSL on the initial concentration of dye and how it changes with time is presented in Fig. 3. The adsorption density increases from 28.5 mg/g to 52.8 mg/g as the initial concentration increases from 100 to 300 mg/L, as shown in the Figure 3. This tendency is due to the high concentration gradient i.e. high mass transfer driving force at higher initial dye concentration leading to increased adsorption density. The same behavior was reported in the literature [9,12].

As it can be observed from the Figure 3, the adsorption is very rapid in the first 20 min corresponding to 85 ± 5 % dye removaland it gradually decreases till equilibrium is reached at 180 min. The higher sorption rate at the preliminary stage may be attributed to the availability of the vacant active sites and high concentration gradient at the initial period. After that, the number of occupied active site increases which in turn increases the repulsion between adsorbate on the surface and in the solution and also the concentration gradient decreases, leading to gradual decrease in adsorption capacity at subsequent stages [13].

3.5 Adsorption Isotherms

The equilibrium dependency of the amount of dye adsorbed on its liquid phase concentration at constant temperature is known as adsorption isotherm. It is an important parameter to estimate the adsorbate-adsorbent interaction, which is critical in optimizing the required amount of adsorbent for a certain degree of

Fig. 3: Effect of initial concentration and contacttimeon adsorption of RB onto LSL (C_o = 100, 200, 300 mg/L, W = 0.2 g, V = 200 mL)

Fig. 4: Langmuir isotherm for adsorption of RB onto LSL

removal. Among the available different isotherm models, two famous models namely Langmuir and Freundlich models have been tested in this study for the sorption process.

The linearized form of Langmuir model [14] can be written as

$$\frac{C_e}{q_e} = \frac{1}{K_L q_\infty} + \frac{1}{C_e q_\infty} \qquad (2)$$

Where, q_∞ is the maximum monolayer adsorption capacity (mg/g) and K_L is the constant related to the free energy of adsorption (L/mg). The values of q_∞ and K_L were cal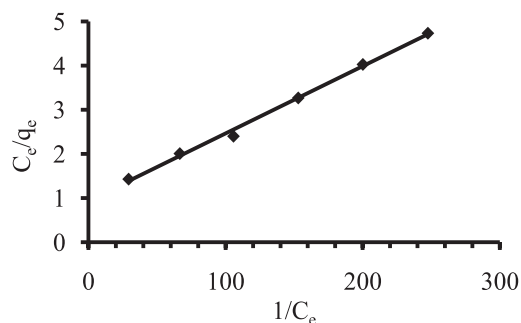culated from the slope and intercept of the linear plots of $\frac{C_e}{q_e}$ versus $\frac{1}{C_e}$ as shown in Fig. 4.

The well-known linearized form of Freundlich isotherm [15] can be expressed as

$$ln\, q_e = ln\, K_f + \frac{1}{n} ln\, C_e \qquad (3)$$

Where, K_f (mg g^{-1}(mg L^{-1})$^{1/n}$) is the Freundlich isotherm constant, which is an indicative of the extent of adsorption and n (dimensionless) is a measure of the adsorption intensity or surface heterogeneity that represents bond distribution, can be measured from slope and intercept of the linear plots of $ln\, q_e$ versus $ln\, C_e$ (Figure is not shown).

The correlation coefficients (r^2) and the isothermal parameters obtained from the application of the Langmuir and Freundlich

models are summarized in Table 1. The closeness of the value of r^2 to unity was used to justify the fitness of the experimental data to the isotherm models and Langmuir model was found to be best fitted for the studied system with maximum monolayer adsorption capacity of 66 mg/g. The adsorption capacity of various biomasses for the removal of RB are reported and compared with that of the present study in Table 2. It is observed that the maximum adsorption capacity of LSL adsorbent is relatively higher than other low cost bio materials investigated so far. Therefore, LSL can be effectively used for treating RB contaminated wastewater.

3.6 Adsorption Kinetics
Designing of an adsorption column along with the time to reach equilibrium and the equilibrium density can be predicted, provided the rate at which adsorption occurs i.e. the kinetics of adsorption is well known. Several kinetic models are available. Among them pseudo first order and pseudo second order models are used to analyze the time dependent batch adsorption data obtained from the Fig. 3.

The linearized-integral form of the most popular pseudo first order model [16] by Lagergren is

$$ln(q_e - q_t) = ln\, q_e - k_1 t \qquad (4)$$

The integrated form of the pseudo second order model [17] can be linearized as

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad (5)$$

$$h = k_2 q_e^2 \qquad (6)$$

Table-1: Langmuir and Freundlich isotherm parameters for the adsorption of RB onto LSL

Langmuir isotherm parameters			Freundlich isotherm parameters		
q_∞(mg/g)	K_L(L/mg)	r^2	K_f	n	r^2
66	0.0163	0.9963	5.054	2.282	0.9508

Table-2: Comparison of the maximum monolayer adsorption capacity of RB on to various adsorbents.

| Adsorbents	Adsorption density,q_∞ (mg/g)	References
Coconut coir pith	14.9	[18]
Exhausted coffee ground	5.255	[19]
Waste banana pith	13.10	[20]
Orange peel	3.23	[21]
Jackfruit peel	4.361	[22]
Acid treated mango leaf powder	2.98	[23]
LSL	66	This study

Where k_1 (min^{-1}) is the rate constant of pseudo-first-order sorption and k_2 (g/mg-min) is the rate constant of pseudo-second order sorption. Here h (mg/g-min) represents the initial adsorption rate.

A plot of $ln(q_e-q_t)$ against t should give a linear relationship if the kinetic data follows pseudo first order kinetics. The values of k_1 and q_e can be calculated from the slope and intercept of the obtained straight line (figure is not shown). A linear plot of $\frac{t}{q_t}$ versus t confirms the conformity of the kinetic data to the pseudo second order kinetics. The slope and intercept of the straight line (Fig. 5.) gives the value of q_e and k_2.

The kinetic parameters obtained from the application of the kinetic models are summarized in Table 3. From the table it can be seen that, the experimental data fits to the pseudo second order model with r_2^2 value higher than 0.99 at all concentration, larger than that of the pseudo first order model. Hence, it can be concluded that adsorption kinetics of RB on LSL is fitted and described by the pseudo second order in a far better way than the pseudo first order.

3.7 Column Experiment

The industrial applicability of LSL adsorbent is assessed by performing fixed bed column study. The key investigating factors in column
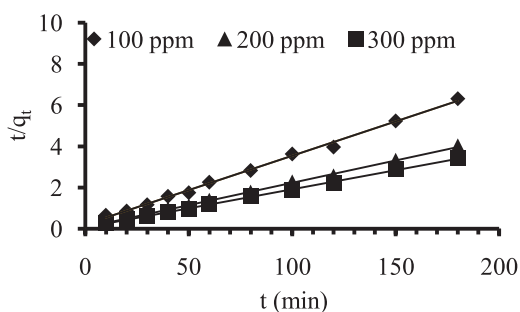
Fig. 5: The pseudo second order kinetic plot for the adsorption of RB onto LSL (C_o = 100, 200, 300 mg/L, W = 0.2 g, V = 200 mL)

performance are the time for breakthrough appearance and the shape of the breakthrough curve, C_t/C_o versus t where C_t and C_o(mg/L) are the effluent RB concentration at any time and initially respectively. The effect of bed height on column performance is extensively investigated in present study. Two well-known models namely Thomas model and Bed Depth Service Time (BDST) model have been implied to the experimental breakthrough data to estimate the column performance.

Thomas model is based on a system with constant flow rate and no axial dispersion, and whose behavior matches the Langmuir isotherm and pseudo second order kinetics [24]. The linearized form of Thomas model takes the following expression

Table-3: Comparison of the pseudo first order, pseudo second order adsorption rate constants, and experimental and model values of q_e obtained at different initial RB concentrations

C_0 (mg/L)	$q_{e_{exp}}$ (mg/g)	Pseudo first order model			Pseudo second order model		
		k_1 (min^{-1})	$q_{e_{model}}$ (mg/g)	r_1^2	k_2 (g/mg-min)	$q_{e_{model}}$ (mg/g)	r_2^2
100	28.48	0.0063	10.99	0.5306	5.59x10^{-3}	29.94	0.9966
200	46.904	0.0135	9.17	0.6686	7.15x10^{-3}	46.08	0.9987
300	52.384	0.015	6.48	0.7050	5.05 x10^{-3}	54.05	0.9991

$$ln\left(\frac{C_0}{C_t}-1\right)=\frac{k_{TH}q_e X}{Q}-k_{TH}C_0 t \qquad (7)$$

Where k_{TH} is the Thomas rate constant (ml/min-mg), q_e the maximum solid-phase concentration of the solute (mg/g), V is the effluent volume (ml), X is the mass of adsorbent (g), and Q is the flow rate (ml/min). The value of the kinetic parameters (k_{TH}, q_e) are evaluated from the slope and intercept of the straight line corresponding to equation (7).

On the other hand, BDST model claims a linear relationship between bed height Z (cm) and service time t (min), in terms of process concentrations and adsorption parameters. It assumes that the rate of adsorption is controlled by the surface reaction between adsorbate and the unused capacity of the adsorbent [25]. The linear relationship can be given as

$$t=\frac{N_0 Z}{C_0 v}-\frac{1}{K_a C_0}ln\left(\frac{C_0}{C_b}-1\right) \qquad (8)$$

Where C_b (mg/L) is the breakthrough dye concentration, N_o (mg/L) the sorption capacity of bed, v (cm/min) the linear velocity, and K_a (L/mg-min) is the rate constant. The value of the constants (N_o, K_a) are determined from the slope and intercept of the plotted equation (8).

The experimental breakthrough curve is shown in Fig. 6. and the corresponding breakthrough time (t_b), equilibrium time (t_e), adsorption capacity ($q_{e_{exp}}$) are tabulated in Table 3. Breakthrough time is considered at $\frac{C_t}{C_0}=0.05$.

The result shows that both bed capacity and exhaustion time increases with increasing bed depth due to the increase in the total surface area, binding sites and residence time as the bed depth increases. The same findings have been found by several other researchers [26,27]. The slope of the breakthrough curve decreased with increasing bed height, which resulted in a broadened mass transfer zone [28]. The adsorption capacity, $q_{e_{model}}$ obtained from the straight line of the Thomas model (Fig. 6.) resembles the experimental one, $q_{e_{exp}}$ with high correlation coefficient ($r^2 > 0.9$) and are reported in Table 4. along with the value of k_{TH}. The linear relationship ($r^2 = 0.999$) between the bed height and service time (t_b) implied by the BDST model is shown in Fig. 7. The model parameters are also enlisted in Table 4. This model can be used to predict the service time for a certain bed height at the prevailing initial concentration and flow rate, irrespective of any experiment.

4. Conclusion

The present study aimed at exploring the potentiality of the LSL as a novel adsorbent for the removal of fluorescent dye Rhodamine B (RB) from its aqueous solution. The SEM test proved the presence of porous rough surface favorable for adsorption. Batch adsorption was found to be strongly dependent on initial concentration and contact time. A monolayer surface coverage on energetically identical surface along with maximum adsorption

M. A. Rahman et.al/Int. J. Integ. Sci. Tech. 3 (2018) 44-52

51

Table-4: Experimental, Thomas model and BDST model parameter for sorption of RB on LSL at different bed height

Z (cm)	Experimental parameters			Thomas model parameters				BDST model parameters		
	t_b (min)	t_e (min)	q_{total} (mg)	$q_{e_{exp}}$ (mg/L)	$q_{e_{model}}$ (mg/L)	K_{TH} (ml/mg-min)	r^2	N_0 (mg/L)	K_a (L/mg-min)	r^2
5.5	93	460	990	61.5	62.15	0.066	0.9521			
7.5	168	540	1305	65.73	63.06	0.06	0.9071	19900	0.36	0.999
10.5	271	670	1800	68.58	65.9	0.0503	0.9368			
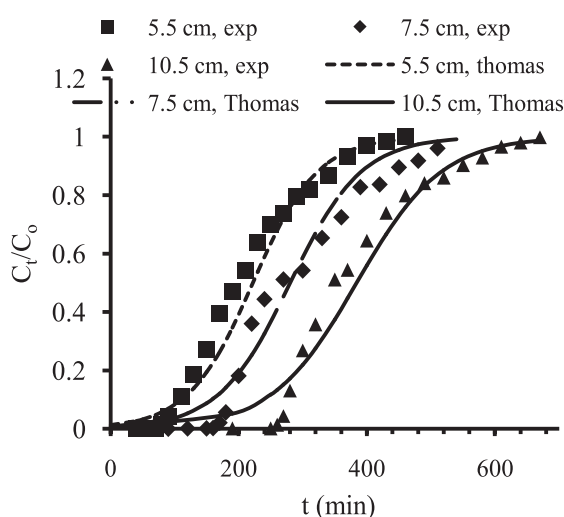
Fig. 6: The measured and modeled (Thomas model) breakthrough curvesfor sorption of RB onto LSL atdifferent bed height (C_0= 300 mg/L, X= 16.10, 19.85, and 26.24 g, Q= 15 ml/min
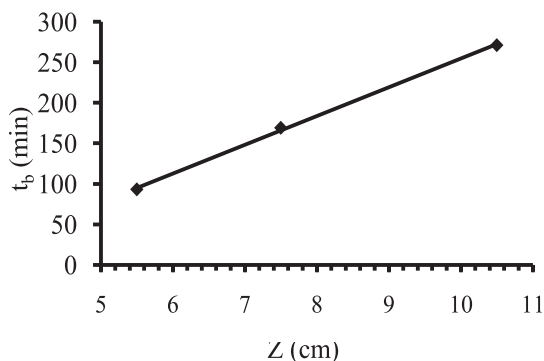
Fig. 7: BDST model plot for sorption of RB onto LSL at different bed height (C_0= 300 mg/L, X= 16.10, 19.85, and 26.24g, Q = 15 ml/min)

capacity of 66 mg/g was confirmed, as Langmuir isotherm model best fitted the equilibrium adsorption data. Compared to other biosorbents investigated to date, LSL has significantly high number of active sites. Adsorption kinetics were well described by pseudo second order kinetic model. Variation of bed height had a significant effect on the column performance. The bed adsorption capacity and service time were found to be increased with bed depth due to enhanced residence time and contact area. The experimental data was modelled using Thomas model to predict the breakthrough curve and the model parameters were found to be in good agreement with the experimental one. The linear dependency of the exhaustion time on the bed height was implied by the BDST model for estimating service time at different bed height, provided other parameters remain unchanged. All the above findings demonstrate that the LSL can be predominantly used as a promising, low cost adsorbent for dye removal from aqueous solution.

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