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Utilization of sugarcane bagasse for removal of basic dyes from aqueous environment in single and binary systems

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ABSTRACT

In the present study a biodegradable and low cost sorbent was investigated for its sorptive capacity for various basic dyes in both single and binary dye solutions. As wastewaters from textile industries may contain a variety of dyes, therefore, it is of great interest to have a sorbent capable of removing dyes either singly or simultaneously. The agricultural by-product has shown its potential to remove Basic Blue 3 (BB3), Methylene Blue (MB) and Basic Yellow 11 (BY11) in both single and binary systems. The effect of pH, contact time and initial concentration, dosage and particle size was studied in batch experiments at room temperature. The optimum pH for the removal of studied dyes was observed in the pH range 4–9 and the sorption reached equilibrium at 60 min. The optimum sorbent dosage was recorded at 0.10 g. The sorption process fitted well in the pseudo-second order kinetic model and Langmuir isotherm was applicable for all the dye systems studied. Maximum sorption capacities were 23.64 mg g^{-1}, 28.25 mg $g^{-1}$and 67.11 mg g^{-1} for BB3, MB and BY11, respectively, in single dye system. However, a decrease in the maximum sorption capacity was observed in the binary systems and this might be resulted from the competition of the same binding sites.

Keywords: Sorption; Low cost sorbent; Sugarcane bagasse; Basic dyes; Isotherms; Kinetics

1. Introduction

Almost every industry uses dyes or colouring substances to colour their products. Effluent discharged from dyeing industries are highly coloured and cause problems to the environment. It can be toxic, mutagenic and carcinogenic to aquatic life as well as affecting the aquatic biota and food web [1]. The presence of dyes in water sources is aesthetically unacceptable and may be visible at concentration as low as 1 ppm [2]. Many dyes are difficult to degrade, as they are generally stable to light, oxidizing agent and are resistant to aerobic digestion [3]. Due to the serious environment impacts of the present of dyes in industrial effluent, there is a need to remove these pollutants before it is discharge into the aqueous environment.

Membrane separation, aerobic and anaerobic degradation using various microorganisms, chemical oxidation, adsorption using various kind of adsorbent are some of the conventional ways to remove colour from industrial waste [4–6]. Among all, the sorption process by activated carbon has been shown to be one of the most efficient methods to remove dyes from effluents [7]. Activated carbon is the most widely used adsorbent in the industries

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due to its capability of adsorbing many types of dyes with a high adsorption capacity. However, it remains as an expensive adsorbent and has high regeneration cost while being exhausted [8]. Thus, these drawbacks increase the importance of finding more economical sorbent for the removal of various dyes from the effluents.

In recent years, various non-conventional and low-cost materials such as agricultural wastes have been studied for the removal of dyes from aqueous solution. These include orange peel [9,10], rice husk [8,11,12], pineapple stem, spent tea leaves [13,14] and wheat straw [15]. However, studies of dye removal in mixture dye system were very limited for recent years. In the present work, adsorption performance of sugarcane bagasse in the removal of cationic dyes, BB3, MB and BY11 was studied in both single and binary systems. Several effective parameters such as initial dye concentration, contact time, pH, sorbent dosage, sorption kinetics and isotherm studies were conducted to evaluate the sorption capacity of sugarcane bagasse (SB).

2. Experimental

2.1. Materials

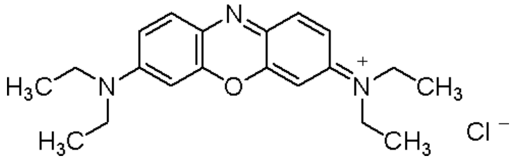
Sugarcane bagasse was collected and cut into small pieces. Then the bagasse was boiled for 3 h to remove the sugar residue within it. It was rinsed several time with tap water and dried overnight at 60°C. The dried bagasse was ground and sieved through a 3 mm sieve and labeled as NSB.

2.2. Sorbates

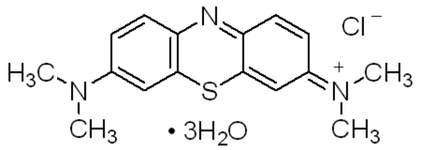
Synthetic dye solutions of BB3, MB and BY11 were the sorbates used in this study and all the dye structures are showed in Fig. 1. The cationic dye BB3 (C.I. = 378011, 25% dye content), MB (C.I. = M9140, 82% dye content) and BY11 (C.I. = B7133, 20% dye content) were used without further purification. All synthetic dyes used in this study were purchased from Sigma-Aldrich Pte. Ltd. (USA). Standard dyes solutions of 1000 mg L^{-1} were prepared as stock solutions and subsequently diluted when necessary.

2.3. Batch experiments

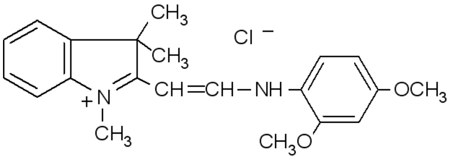
The batch experiments were carried out by mixing 0.1 g of NSB with 20.0 mL of 50 mg L^{-1} dye solutions in a centrifuge tube and shaken on an orbital shaker at 150 rpm for 4 h unless otherwise stated at room temperature (25±2°C) The supernatant was analysed for its dye concentration using Perkin Elmer Lambda 35 UV–Vis spectrophotometer. Measurements were made at the wavelength corresponding to the maximum absorption; for BB3, λ_{max} = 654 nm, MB, λ_{max} = 664 nm and for BY11, λ_{max} = 412 nm. Dilutions were made when measurements exceeded the linearity of the calibration curves. All the batch experiments were carried out in duplicates ant the results showed were the average. Control experiments without NSB were carried out to make sure that the sorption was by the sorbent and not due to the wall of the centrifuge tube.



Basic Blue 3



Methylene Blue



Basic Yellow 11

Fig. 1. The structure of Basic Blue 3, Methylene Blue and Basic Yellow 11.

For the study of the effect of pH, a series of dye solutions with the adjusted pH of 2–10 were prepared. The desired pH was obtained by adding HCl or NaOH (0.1–2.0 M). For the study of the effect of contact time, the experiments were carried out at the dye concentrations of 50, 100 150 and 200 mg L^{-1}. The samples were withdrawn and analysed for their dye concentrations at predetermined intervals. For the study of sorbent dosage, the experiments were carried out with the amount of sorbent from 0.05 g to 0.20 g. For the study of the effect of particle size, the sorbent was divided into 3 different particle sizes (<300 micron, 300–600 micron and > 600 micron) using serial sieves. The study of sorption isotherms was carried out using initial dye concentrations from 50 to 300 mg L^{-1}.

3. Results and discussion

3.1. Effect of pH

For BB3 and MB, both dyes have shown essentially the same sorption behaviour regardless the single or binary dye systems, thus only the sorption trend of BB3 was

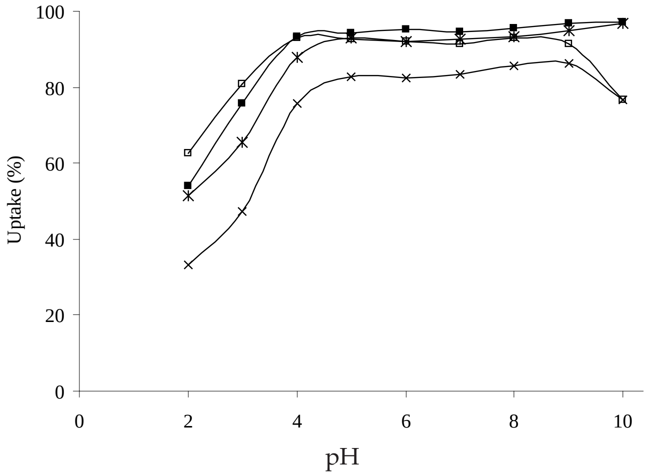


Fig. 2. Effect of pH on the sorption of BB3 and BY11 by NSB. ■ of BB3 (single), ✳ of BB3 (binary), □ of BY11 (single) and × of BY11 (binary).

presented. Fig. 2 shows the effects of pH on the uptake of BB3 and BY11 by NSB in single and binary systems. The lowest dye uptake for all the systems was recorded at pH 2. The percentage uptake increased as the pH of the dye solutions increased. A sharp increase in the percentage uptake was observed at pH 2–4. The percentage uptake remained almost constant over the pH range of 4–9. It is suggested that at a lower pH, the carboxyl groups on the biosorbent surface which are responsible for binding the dye cations predominantly protonated, therefore this led to a lower uptake of the dyes. As the pH of the dye increased, sorption became favourable due to the deprotonation of the carboxyl groups, resulting in the increase of available binding sites [16]. However there is a slight drop in the percentage uptake of BY11 in both single and binary systems at pH 10. This phenomenon might be due to the occurrence of another mode of adsorption, such as ion exchange or chelatation [17]. There is also a possibility that the NSB might be subjected to hydrolysis, which creates positively charged sites which is not favourable for removing cationic species [18].

3.2. *Effect of contact time and initial concentration*

The effect of contact time on the uptake of BB3 and BY11 by NSB in single dye solutions at different initial concentrations is shown in Fig. 3. The uptakes of all dye systems increased rapidly at the first 15 min and gradually attained equilibrium after 60 min. The equilibrium sorption was achieved after 60 min, thus, a contact time of 4 h with dye solutions was applied in all the subsequent parameters studied. The rapid uptake at the initial stage suggested a strong attractive force between the dye molecules and the adsorbent. Similar observations were previously reported in the removal of acid dyes by shells [19,20]. In addition, as the initial dye concentration increased, the percentage of dye uptake decreased. This was due to the increase of the ratio of the dye's cations to the dosage of the adsorbent.

3.3. *Sorption kinetics*

The kinetics of the dye adsorption on NSB was examined using the following equations [21,22,23]:

Pseudo-first order

$$\log\left(q_e - q_t\right) = \log q_e \frac{k_1 t}{2.303} \tag{1}$$

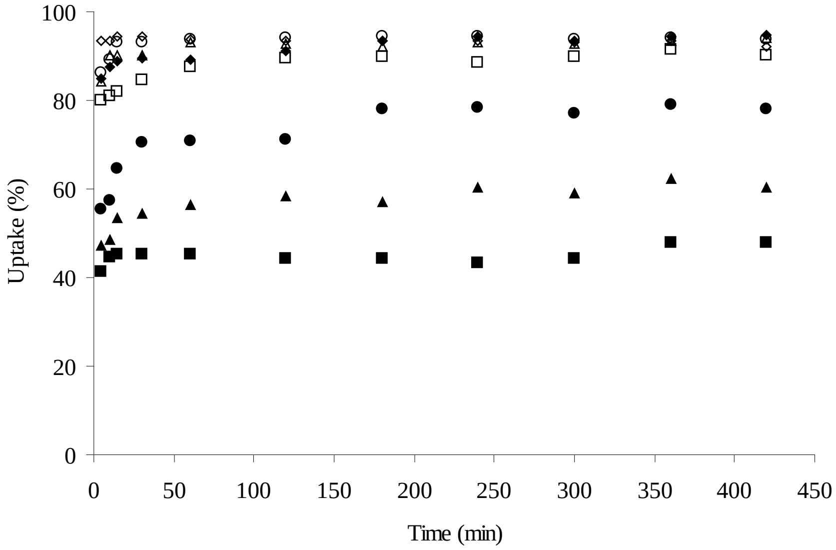


Fig. 3. Effect of initial concentration and contact time on the sorption of BB3 and BY11 by NSB. ■, ▲, ●, ◆ – 50, 100, 150 and 200 mg/L of BB3, and □, △, ○, ◊ – 50, 100, 150 and 200 mg/L of BY11.

and pseudo-second order

$$\frac{t}{q_t} = \frac{1}{h} + \frac{t}{q_e} \tag{2}$$

where q_e = the amount of dyes sorbed at equilibrium (mg g^{-1}), q_t = the amount of dyes sorbed at time t (mg g^{-1}), k_1 = the rate constant of pseudo-first order sorption (min^{-1}), h ($k_2q_e^2$) = the initial sorption rate (mg g^{-1} min^{-1}) and k_2 = the rate constant of pseudo-second order kinetics (g mg^{-1} min^{-1}).

By comparing both kinetic models based on the correlation coefficients, pseudo-second order kinetics showed a better correlation of the experimental data for all the systems studied. In addition, a wide range of variations were recorded between the sorption capacities determined from the pseudo-first order kinetic model than those obtained experimentally (Table 1). Therefore, the dye systems under study are more appropriately described by the pseudo-second order model which is based on the assumption that the rate limiting step may be chemical sorption or chemisorption involving valency forces through sharing or exchange of electron between sorbent and sorbate [23].

The values of *qe*, k_2 and h against C_0 in the corresponding linear plots of the pseudo-second order equation were regressed to obtain the expressions for these values in terms of the initial dye concentration. Each of these parameters can be expressed as a function of C_0 for BB3, MB and BY11 [23].

$$Q_e = \frac{C_0}{A_qC_0 + B_q} \tag{3}$$

$$k_2 = \frac{C_0}{A_kC_0 + B_k} \tag{4}$$

$$h = \frac{C_0}{A_hC_0 + B_h} \tag{5}$$

where A_q, B_q, A_k, B_k, A_h and B_h are constants related to the respective equations. The values of these constants are shown in Table 2. Thus, the generalised predictive models for BB3, MB and BY11 sorbed at any initial concentration and contact time within the given range with relationship of q_e, C_o and t can be represented as follows by substituting the various values in the pseudo-second order kinetic equation.

For BB3–NSB system:
in single dye solution

$$q_t = \frac{C_0t}{0.261C_0 - 1.385 + (0.033C_0 + 3.550)t} \tag{6}$$

in binary dye solution

$$q_t = \frac{C_0t}{0.318C_0 - 3.478 + (0.033C_0 + 3.698)t} \tag{7}$$

For MB–NSB system:
in single system

$$q_t = \frac{C_0t}{0.218C_0 - 8.076 + (0.022C_0 + 3.999)t} \tag{8}$$

Table 1
Sorption capacities and correlation coefficients based on pseudo-first and pseudo-second order kinetics

Dye systems		Initial concentration (mg L^{-1})	Pseudo-first order		Pseudo-second order		Experimental
			Sorption capacities (mg g^{-1})	R^2	Sorption capacities (mg g^{-1})	R^2	Sorption capacities (mg g^{-1})
BB3 (single)		100	7.005	0.722	15.926	0.999	15.783
		200	22.798	0.336	18.450	0.996	17.013
MB (single)		100	3.946	0.753	18.051	0.999	17.828
		200	21.662	0.754	21.598	0.998	21.003
BY11 (single)		100	1.682	0.324	18.797	1.000	18.854
		200	6.717	0.731	36.232	0.999	35.381
Binary BB3–BY11	BB3	100	7.432	0.693	15.093	0.999	14.815
		200	23.818	0.688	18.939	0.999	19.012
	BY11	100	10.088	0.636	12.092	0.994	10.631
		200	25.633	0.468	15.674	0.999	15.953
Binary MB–BY11	MB	100	4.264	0.520	17.094	0.999	17.163
		200	20.361	0.816	21.882	0.998	21.159
	BY11	100	8.827	0.479	12.642	0.997	12.749
		200	25.316	0.560	15.699	0.998	15.284

Table 2
Empirical parameters for predicted q_e, k_2 and h from C_0

Dyes		A_q (g mg^{-1})	B_q (g L^{-1})	A_k (mg min g^{-1})	B_k (mg^2 min g^{-1} L^{-1})	A_h (g min mg^{-1})	B_h (g min L^{-1})
BB3 (single)		0.033	3.550	1.053×10^2	-4.145×10^3	0.261	–1.385
MB (single)		0.022	3.999	1.054×10^2	-5.209×10^3	0.218	–8.076
BY11 (Single)		-3×10^{-4}	5.435	65.342	-4.001×10^3	0.092	–8.265
Binary BB3–BY11	BB3	0.033	3.598	1.216×10^2	-4.839×10^3	0.318	–3.478
	BY11	0.050	3.361	42.190	-2.806×10^2	0.175	10.200
Binary MB–BY11	MB	0.023	3.952	113.620	-5.821×10^3	0.204	–5.658
	BY11	0.048	3.396	57.763	-1.606×10^3	0.114	15.171

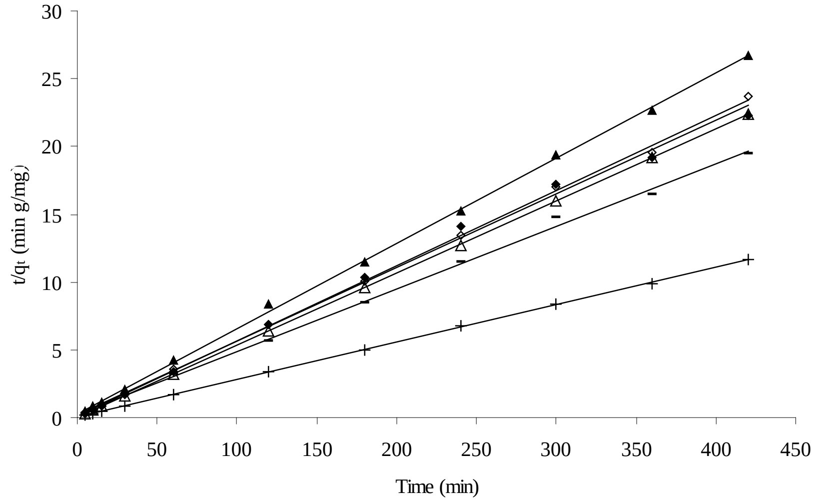


Fig. 4. Pseudo second-order kinetics of BB3, MB and BY11 in single system. ▲, ◆ – 100 and 200 mg/L of BB3; △, ◇ – 100 and 200 mg/L of MB; and ×, + – 100 and 200 mg/L of BY11.

in binary system

$$q_t = \frac{C_0 t}{0.204C_0 - 5.658 + \left(0.023C_0 + 3.952\right)t} \tag{9}$$

For BY–NSB system:
in single system

$$q_t = \frac{C_0 t}{0.092C_0 - 8.265 + \left(-0.0003C_0 + 5.435\right)t} \tag{10}$$

in BB3–BY binary system

$$q_t = \frac{C_0 t}{0.175C_0 + 10.2 + \left(0.050C_0 + 3.361\right)t} \tag{11}$$

in MB–BY binary system

$$q_t = \frac{C_0 t}{0.114C_0 + 15.171 + \left(0.048C_0 + 3.396\right)t} \tag{12}$$

The theoretical model derived for BB3, MB and BY11 sorption in single dye solutions by NSB was applied to the uptake of these dyes. Fig. 5 shows the comparison between the calculated and the experimental values and it is clear that the theoretically generated curves agreed well with the experimental data.

3.3. Sorption isotherm

Adsorption isotherms are the basic requirements for the design of adsorption systems [24]. In the present work, the equilibrium data for BB3, MB and BY11 in both single and binary systems were modeled with Langmuir, Freundlich and BET models. The linearised Langmuir equation is written as:

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_a q_m} \tag{13}$$

whereas the linearised form of Freundlich is shown as follows:

$$\log q_e = \log K_f + \frac{\log C_e}{n} \tag{14}$$

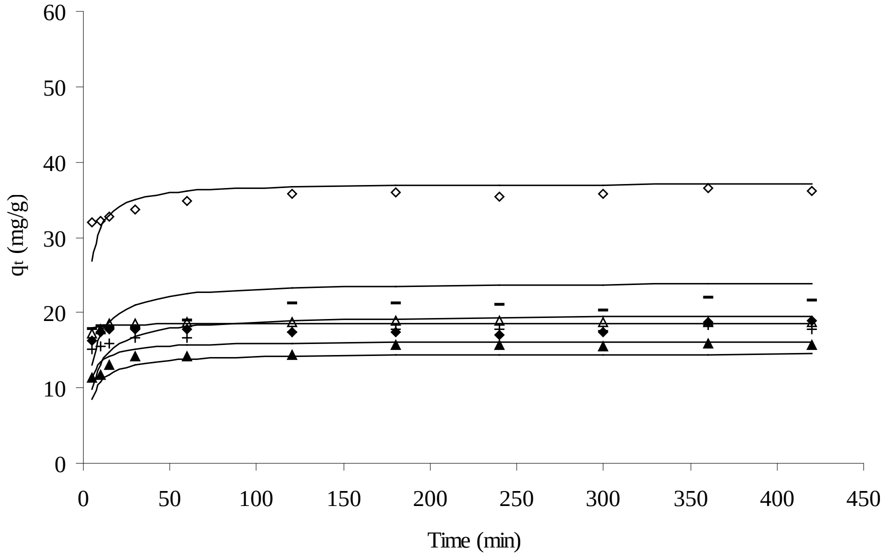


Fig. 5. Comparison between the measured and pseudo-second order model time profiles for BB3, MB and BY11 sorption from single dye solutions. ▲, ◆ – 100 and 200 mg/L of BB3 (experimental); +, – – 100 and 200 mg/L (experimental) of MB; and △, ◇ – 100 and 200 mg/L of BY11 (experimental). Line – theoretical.

While the linearised form of BET model is given as follows:

$$\frac{C_e}{(C_i - C_e)q_e} = \left(\frac{1}{Bq_m}\right) + \left(\frac{B-1}{Bq_m}\right)\left(\frac{C_e}{C_i}\right) \quad (15)$$

where C_e = equilibrium concentration of the dye (mg L^{-1}), q_e = amount of dye sorbed at equilibrium (mg g^{-1}), q_m = maximum sorption capacity (mg g^{-1}), K_a = constant related to the energy of the sorbent (mg^{-1}), n = Freundlich constant for intensity, K_f = Freundlich constant for sorption capacity, C_i = initial concentration of dye (mg L^{-1}), and B = BET constant expressive of the energy of interaction with surface.

The Langmuir isotherm curves of BB3, MB and BY11 by NSB in single and binary system are shown in Figs. 6 and 7, respectively. The calculated results of the Langmuir, Freundlich and BET isotherms constants and regression coefficients are shown in Table 3. These three models are based on different assumptions: Langmuir model implies monolayer sorption onto a surface with a finite number of identical sites, Freundlich model is based on sorption on a heterogeneous surface while BET model is the modified Langmuir model which has the same assumptions except that it allows multilayer adsorption. For all the systems under study, the Langmuir isotherm was found to provide reasonable fitting. The applicability of this isotherm to sorption of different types of dyes by agricultural waste has been reported previously [25]. The maximum capacity of NSB for all studied dye systems is included in Table 3. A drop in maximum capacity for binary system was observed as compared to the single

Table 3
Langmuir, Freundlich and BET constants for the sorption of all studied dye systems

Dyes		Langmuir			Freundlich			BET	
		q_m (mg g^{-1})	K_a (l mg^{-1})	R^2	K_f	n	R^2	B	R^2
BB3 (single)		23.641	0.077	0.975	7.268	4.554	0.953	–28.821	0.944
MB (single)		28.249	0.106	0.997	7.231	3.663	0.961	–762.000	0.971
BY11 (single)		67.110	0.060	0.984	6.276	1.791	0.933	–0.178	0.021
Binary BB3–BY11	BB3	21.834	0.059	0.976	6.077	4.303	0.953	–14.894	0.946
	BY11	18.416	0.039	0.959	4.535	4.120	0.949	–7.065	0.890
Binary MB–BY11	MB	26.178	0.097	0.988	7.065	3.851	0.923	–87.100	0.961
	BY11	21.142	0.028	0.927	5.664	4.760	0.757	5.762	0.781

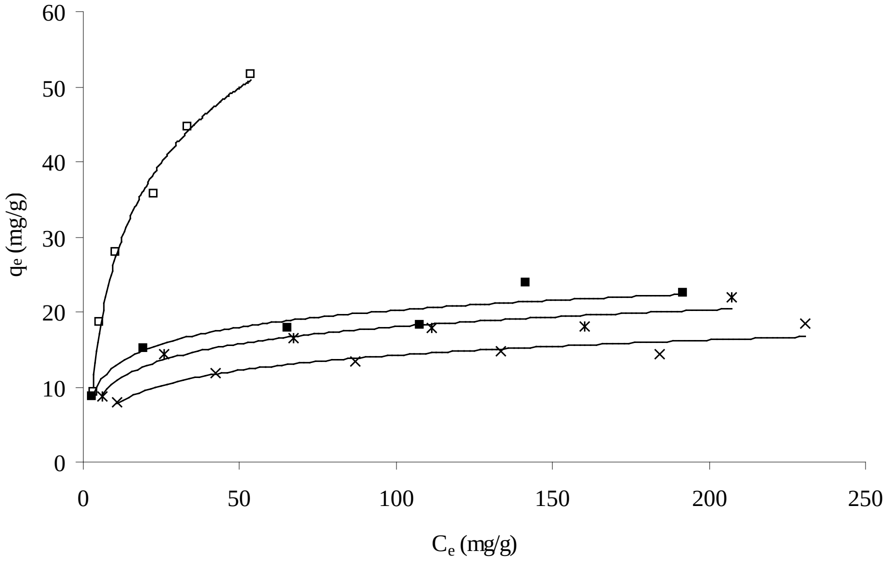


Fig. 6. Langmuir isotherm curves of BB3 and BY11 in single and binary systems. ■, □, ✳, × – single BB3, single BY11, binary BB3 and binary BY11.

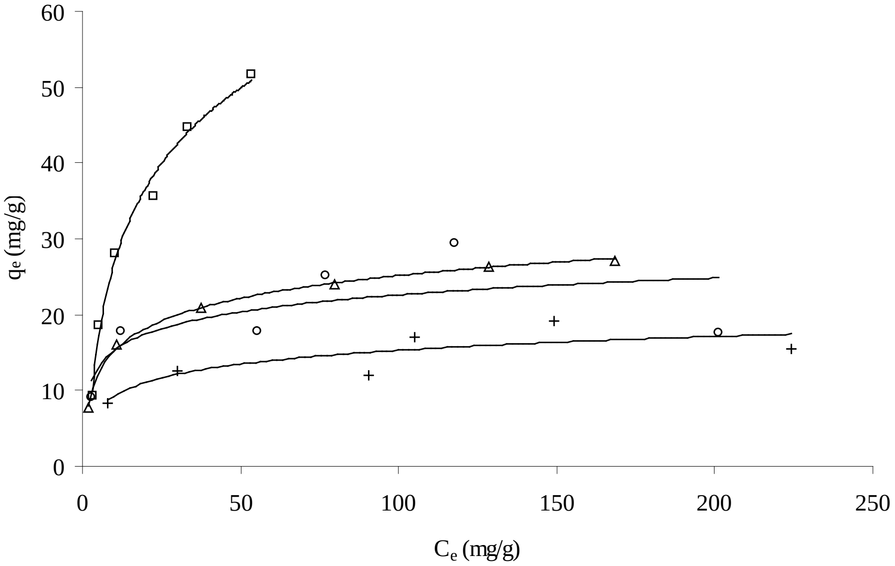


Fig. 7. Langmuir isotherm curves of MB and BY11 in single and binary systems. △, □, ○, + – single MB, single BY11, binary MB and binary BY11.

system. This might be due to the competition between two dyes for the sorption site.

The essential character of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor, R_L [26], which is defined by the relationship:

$$R_L = \frac{1}{(1 + K_a C_0)} \tag{16}$$

where K_a = Langmuir constant (ml mg^{-1}) and C_0 = initial dye concentration. Referring to the effect of the separation factor on the isotherm shape, the R_L values are in the range of $0 < R_L < 1$, which indicates that the adsorption of studied dyes on NSB is favourable. Table 4 shows the R_L values for all the dye systems studied.

Table 4
Values of R_L for all the studied dye systems

C_0 (mg/L)	BB3 (single)	MB (single)	BY11 (single)	Binary BB3–BY11		Binary MB–BY11	
				BB3	BY11	MB	BY11
50	0.199	0.159	0.249	0.254	0.339	0.172	0.417
100	0.111	0.086	0.142	0.145	0.204	0.094	0.263
150	0.077	0.059	0.100	0.102	0.146	0.065	0.192
200	0.078	0.045	0.077	0.078	0.113	0.049	0.151
250	0.047	0.036	0.062	0.064	0.093	0.040	0.125
300	0.040	0.030	0.052	0.054	0.079	0.033	0.106

3.4. *Effect of sorbent dosage*

The effect of the sorbent dosage on the percentage uptake of BB3, MB and BY11 by NSB in single and binary systems are shown in Fig. 8 and Fig. 9, respectively. The percentage uptake for all studied dyes in single and binary systems showed an increased percentage uptake as the sorbent dosage increased from 0.05 g to 0.20 g. The trend followed the expected pattern where increasing of the sorbent dosage increases the percentage uptake of dye. The increase in the adsorption with the adsorbent dose can be attributed to the increased adsorbent surface area and availability of more adsorption sites [27–29]. The optimum dosage was found to be 0.10 g of NSB for all the dye systems. Only minimal increment in the percentage uptake was observed when the sorbent dosage was above 0.10 g.

3.5.*Effect of particle size*

The effect of particle size on the percentage uptake of BB3, MB and BY11 by NSB in both single and binary systems is shown in Table 5. The results show that the sorption decreases as the particle size increases. The lower uptake was due to the decrease in the surface area over the weight of the sorbent as the size increased. Small particles move faster in the solution therefore there is more shear on their surface. It follows that the boundary layer thickness is thinner than that on the larger particles which resulted a faster rate of sorption. A similar trend were reported in the removal of dyes by modified rice hull [16,30].

4. Conclusion

The results of this study show that NSB has the potential to remove BB3, MB and BY11 in both single and binary systems. The sorption of all dye systems was pH-dependent and the optimum sorption range was from pH 4 to 9. The percentage uptake of all dye systems increased with increasing the contact time and sorbent dosage while

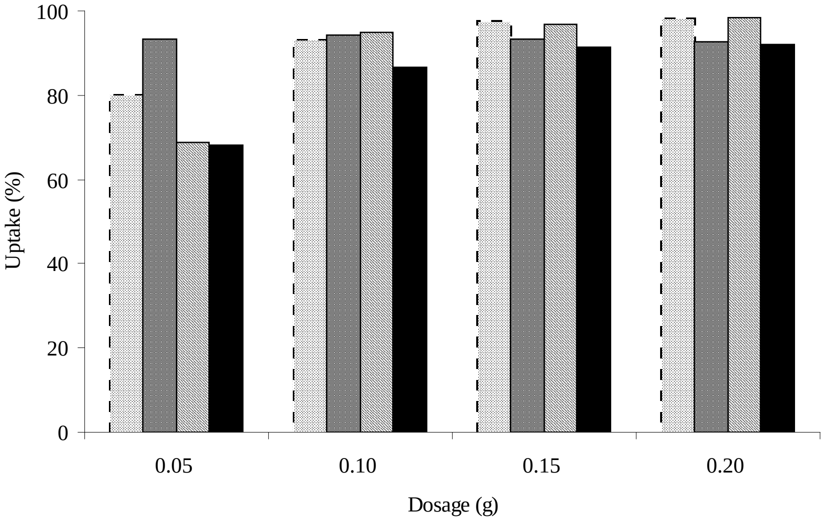


Fig. 8. Effect of sorbent dosage on the sorption of BB3 and BY11 in single and binary systems. ░, ▦, ▧, ■ – single BB3, single BY11, binary BB3 and binary BY11.

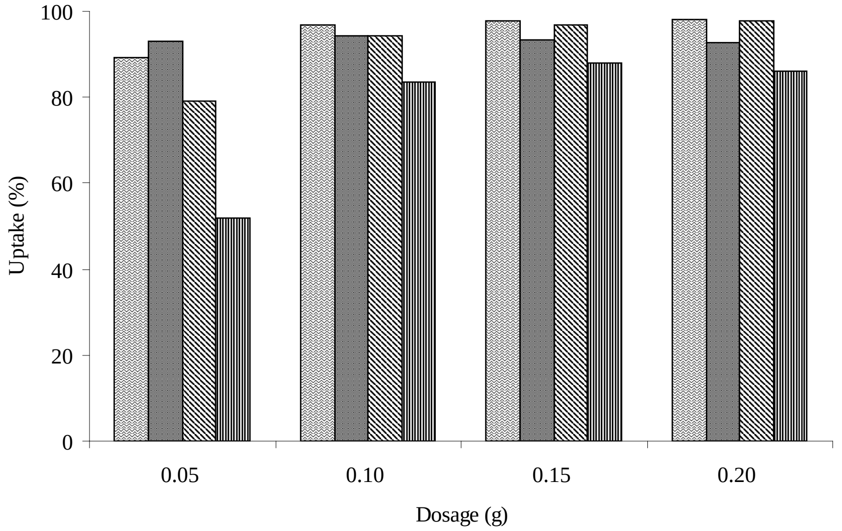


Fig. 9. Effect of sorbent dosage on the sorption of M3 and BY11 in single and binary systems. ▩, ■, ▧, ▥ – single MB, single BY11, binary M3 and binary BY11.

Table 5
Percentage uptake of all studied dye systems based on particle size

Particle size Systems		% Uptake		
		< 300 micron	300 – 600 micron	> 600 micron
BB3 (single)		96.62%	92.99%	91.61%
MB (single)		95.99%	94.81%	94.12%
BY11 (single)		96.70%	95.71%	92.30%
Binary BB3–BY11	BB3	96.65%	91.33%	87.18%
	BY11	89.36%	81.43%	76.53%
Binary MB–BY11	MB	97.26%	95.00%	89.99%
	BY11	87.49%	82.59%	75.24%

decreased with increasing the initial concentration and particle size. The optimum sorbent dosage was found to be 0.10 g of NSB. The analysis of the data shows that pseudo-second order kinetic models gave a better correlation of the experimental data as compared to the pseudo-first kinetic model. Therefore, it indicates that the rate limiting step may be chemisorption. Langmuir isotherms agreed very well for all the studied dye systems with maximum monolayer sorption of 23.64 mg g^{-1}, 28.25 mg g^{-1} and 67.11 mg g^{-1} for BB3, MB and BY11, respectively, in the single dye system. A decrease in the maximum sorption capacity was observed for all the dyes in the binary systems and this might be attributed to the competition for the binding sites.

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