



Removal of basic dyes from aqueous solution using sugarcane bagasse: optimization by Plackett–Burman and Response Surface Methodology

Eng-Cheong Khoo^a, Siew-Teng Ong^{b,*}, Yung-Tse Hung^c, Sie-Tiong Ha^b

^aFaculty of Engineering and Science, Department of Science, Universiti Tunku Abdul Rahman, Jalan Genting Kelang, Setapak, Kuala Lumpur 53300, Malaysia

^bFaculty of Science, Department of Chemical Science, Universiti Tunku Abdul Rahman, Jalan Universiti, Bandar Barat, Kampar 31900, Malaysia

Fax: +605 4661676; email: ongst@utar.edu.my

^cDepartment of Civil and Environmental Engineering, Cleveland State University, Cleveland, OH, USA

Received 17 October 2011; Accepted 28 February 2013

ABSTRACT

In the present study, Plackett–Burman design has been used to identify the significant factors affecting the removal of basic dyes using agricultural waste, sugarcane bagasse (natural sugarcane bagasse). The effect initial dye concentration and sorbent dosage were identified to be the variables responsible for affecting the percentage uptake of Basic Blue 3 (BB3) and Methylene Blue (MB) from aqueous solution. Meanwhile, none of the studied variables were found to be significantly affecting the percentage uptake of Basic Yellow 11 dye. The interaction between the factors and their optimum levels for maximum percentage uptake of dyes were determined using Response Surface Methodology. Both models were highly significant with correlation coefficients (R^2) of 0.9947 and 0.9967 for BB3 and MB dye solutions, respectively. For BB3, the percentage uptake of 98.59 was obtained with optimized conditions at 50 mg L^{-1} initial dye concentration and 0.13 g sorbent dosage. Whereas for MB, under the optimum of 72 mg L^{-1} of initial dye concentration and 0.18 g of sorbent dosage, the percentage uptake was recorded to be 95.19. The experimental values agreed well with the predicted values with percentage errors less than 3%.

Keywords: Dyes; Low-cost adsorbent; Adsorption; Statistical experiments; Optimum conditions

1. Introduction

Dyes or coloring matter is being used in almost every industry to color their products. Effluents discharged from dyeing industry are highly colored and causes pollution to the environment. Over 10,000

different dyes and pigments were used in textile and printing industries worldwide. Without proper treatment, effluent discharged from dyeing industries can cause many problems to the environment as most of dyes are toxic, mutagenic and carcinogenic. This pollutant poses hazardous effect not only to the aquatic life, but also other living organisms [1]. In addition, many dyes are difficult to degrade, as they are

*Corresponding author.

Table 1
Comparison of maximum adsorption capacity for BB3, MB, and BY by different adsorbents

Dye	Adsorbent	Maximum adsorption capacity (mg/g)	References
BB3	Activated sludge biomass	36.5	[11]
	Ethylenediamine modified rice hull	3.29	[12]
	Quartenized sugarcane bagasse	5.58	[13]
	Natural durian peel	49.50	[14]
	NSB	23.64	[15]
MB	Beech sawdust	9.78	[16]
	Rice husk	40.58	[17]
	Modified chitosan	202	[18]
	Fly ash	5.718	[19]
	NSB	28.25	[15]
BY21	Peat	660	[20]
BY37	Modified chitosan	595	[21]
BY11	NSB	67.11	[15]

generally stable to light, oxidizing agent and are resistant to aerobic digestion [2]. For instance, basic dyes which are commonly used in coloring acrylic fiber, are generally more toxic than other classes of dyes. Due to the serious environmental impacts of dyes, it is crucial to remove these pollutants before it is being discharged into the aqueous environment.

Activated carbon had been widely used in industries for the removal of dyes from aqueous solution due to its capability to adsorb many types of dyes and applicability in wide range of pH. However, it still remains as an expensive adsorbent and has high regeneration cost when exhausted [3]. These kinds of disadvantages increase the importance of finding more economical sorbent for the removal of various dyes from the industrial effluents. In recent years, various non-conventional and low-cost materials, such as agricultural waste, have been studied for the removal of dyes from aqueous solution. These include rice husk in both natural and modified form [3–6], walnut shells [7], coffee husk [8], wheat straw [9], and fruits peels [10]. The maximum adsorption capacity of some of the reported works is presented in Table 1.

Conventional and classical methods of studying a process by maintaining other factors involved at an

unspecified constant level does not describe the combined effect of all the factors involved [22]. Besides, it is also a time-consuming method. The mentioned limitations of conventional methods can be overcome by introducing statistically experiment design such as Plackett–Burman design and Response Surface Methodology (RSM) [23]. These statistical techniques are capable to optimize all the affecting parameters collectively to fits the experimental domain in the theoretical design through a response function.

Plackett–Burman design determines the most important variables for further optimization and gave unbiased estimates of linear of all variables with maximum accuracy for a given number of observations [24]. Meanwhile, RSM determines the optimum operational conditions of the system [25]. In this present study, sorption performance of sugarcane bagasse in the removal of Basic Blue 3 (BB3), Methylene Blue (MB) and Basic Yellow 11 (BY11) from dye solution were studied. The interaction between the factors was studied and optimized using RSM three major steps: (i) design and perform the experiments; (ii) response surface modeling through regression analysis; (iii) optimization and checking the adequacy of the model.

2. Experimental

2.1. Preparation of sorbent

Sugarcane bagasse was collected and cut into small pieces. The bagasse was boiled using boiling water for 3 h to remove the sugar residue within it. After the boiling process, it was washed several times with tap water and rinsed with distilled water before dried overnight in the oven at 60°C. The dried bagasse was ground and sieved through a 3-mm sieve and labeled as natural sugarcane bagasse (NSB).

2.2. Preparation of dye solutions

Synthetic dye solutions of BB3, MB and BY11 were the sorbates used in this study, and all the dye structures were shown in Fig. 1. The cationic dyes, BB3 (C.I. = 378,011, 25% dye content), MB (C.I. = M9140, 82% dye content) and BY11 (C.I. = B7133, 20% dye content) were purchased from Sigma-Aldrich Pte. Ltd. and used without further purification. Standard dye solutions of 1,000 mg L⁻¹ were prepared as stock solutions and subsequently diluted when necessary.

2.3. Characterization studies

Field emission scanning electron microscopy (FESEM) analysis was carried out to study the surface morphology of NSB. The micrograph was taken using

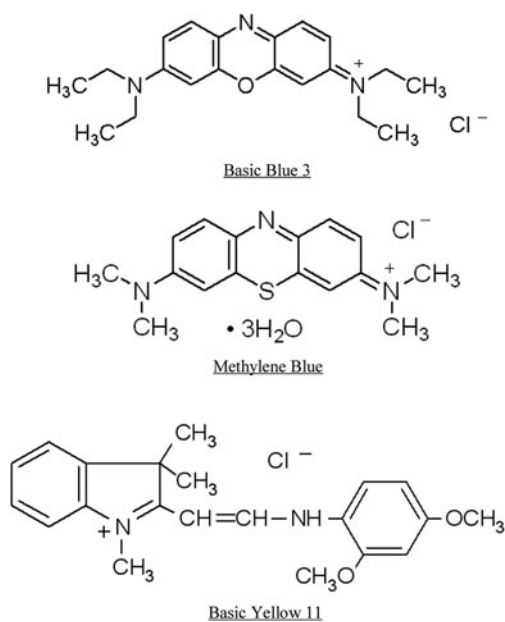


Fig. 1. Chemical structure of BB3, MB and BY11.

FESEM JSM 6701F (JEOL) operated at emission current of 2.00 kV with working distance of 6.0 mm. The functional groups NSB were determined using Perkin–Elmer System 2000 FTIR Spectrometer. The sample disk was prepared by mixing NSB with KBr. The mixture was then ground and compressed into a pellet before it was analyzed to obtain the spectrum.

2.4. Plackett–Burman design

Plackett–Burman design was used to evaluate the relative importance of various factors that influence the percentage uptake of dyes. The purpose of applying this method is to identify which factor(s) has a significant effect on the percentage uptake of the studied dyes. In this study, five assigned variables (pH, contact time, initial dye concentration, sorbent particle size, and sorbent dosage) were screened in 12 experimental designs. All the experiments were carried out in duplicate under room temperature ($25 \pm 2^\circ\text{C}$). The results presented are the means with relative standard deviation of less than 5%. Table 2 shows the design and results for the removal of BB3 and MB. All the experiments were carried out in duplicate and the means of the percentage uptake of dyes were taken as response. The experimental design and statistical analysis of the data were done by using Design Expert Version 7.1.3.

2.5. Optimization of percentage uptake of dye using RSM approach

In this study, central composite design (CCD) model was used. The variables used for the studied

dye solutions at five coded levels ($-\alpha$, -1 , 0 , $+1$, $+\alpha$) were shown in Table 3. The CCD matrix for two coded independent variables and the observed response for BB3 and MB are shown in Table 4. All the experiments were conducted in duplicate and the mean values of the duplicates were taken as the response (percentage uptake of dyes). Cubic equation used for optimization of the percentage uptake of dye is shown as following:

$$\begin{aligned}
 Y = & \beta_0 + \sum_{i=1}^2 \beta_i X_i + \sum_{i=1}^2 \beta_{ii} X_i^2 + \sum_{i=1}^2 \beta_{iii} X_i^3 \\
 & + \sum_{i=1}^1 \sum_{j=i+1}^2 \beta_{ij} X_i X_j \\
 & + \sum_{i=1}^1 \sum_{j=i+1}^2 \beta_{ijj} X_i X_j^2 + \sum_{i=1}^1 \sum_{j=i+1}^2 \beta_{ijj} X_i^2 X_j
 \end{aligned} \quad (1)$$

where β_0 , β_i , β_{ii} , β_{iii} , β_{ij} , β_{ijj} , and β_{ijj} are the constant coefficients, and X_i and X_j are the independent variables. All the experimental design and statistical analysis of the data were done by using Design Expert Version 7.1.3.

3. Results and discussion

3.1. Characterization of NSB

The surface morphology of NSB before and after MB dye adsorption is shown in Fig. 2(a and b), respectively. From the SEM, it was observed that there was only a minimal difference in the surface morphology before and after MB dye adsorption. Similar results were obtained for NSB after BB3 and BY11 adsorption. Besides, it is also shown that the investigated low cost adsorbent is a nonporous material, due to the absence of pores and cavities. This finding agreed well with the previously reported work on some of the low-cost adsorbents [10].

The FTIR analysis was used to study the changes in the functional groups of NSB before and after sorption. Fig. 3 showed that there is no significant difference in the functional groups present in NSB before and after sorption. NSB show absorption in the region of $3,200\text{--}3,600\text{ cm}^{-1}$ indicates the existence of free and intermolecular bonded hydroxyl groups. The peak observed at $2,905\text{ cm}^{-1}$ can be assigned to stretching vibration of C–H group. Peaks around $1,720\text{ cm}^{-1}$ were due to the C=O group and around $1,620\text{ cm}^{-1}$ corresponded to C=C or asymmetric and symmetric stretching C=O vibrations. A strong C–O band was observed at $1,051\text{ cm}^{-1}$ was due to –OCH₃ group, showing the presence of lignin structure in the NSB

Table 2
Plackett-Burman design and results for the removal of BB3 and MB from dye solution

Dye	Experimental run	Variables					Observed response (%)	Predicted response (%)
		pH	Contact time (min)	Particle size (μm)	Initial dye concentration (mg L^{-1})	Sorbent dosage (g)		
BB3	1	3.00	5.00	150.00	50.00	0.05	79.12	66.30
	2	9.00	5.00	850.00	200.00	0.05	29.24	31.11
	3	9.00	240.00	850.00	50.00	0.05	84.33	79.52
	4	3.00	240.00	150.00	200.00	0.20	85.89	80.10
	5	3.00	240.00	850.00	50.00	0.20	96.11	100.70
	6	3.00	5.00	850.00	50.00	0.20	88.44	86.43
	7	9.00	5.00	850.00	200.00	0.20	73.78	64.77
	8	9.00	5.00	150.00	50.00	0.20	97.12	112.43
	9	3.00	240.00	850.00	200.00	0.05	23.54	32.91
	10	3.00	5.00	150.00	200.00	0.05	25.51	32.17
	11	9.00	240.00	150.00	50.00	0.05	93.32	93.05
	12	9.00	240.00	150.00	200.00	0.20	95.66	92.57
MB	1	3.00	5.00	150.00	50.00	0.05	80.57	66.30
	2	9.00	5.00	850.00	200.00	0.05	29.36	30.98
	3	9.00	240.00	850.00	50.00	0.05	85.03	79.52
	4	3.00	240.00	150.00	200.00	0.20	86.79	80.52
	5	3.00	240.00	850.00	50.00	0.20	96.38	101.36
	6	3.00	5.00	850.00	50.00	0.20	89.31	87.44
	7	9.00	5.00	850.00	200.00	0.20	73.71	65.08
	8	9.00	5.00	150.00	50.00	0.20	98.83	113.79
	9	3.00	240.00	850.00	200.00	0.05	23.23	32.48
	10	3.00	5.00	150.00	200.00	0.05	25.28	32.49
	11	9.00	240.00	150.00	50.00	0.05	93.03	93.61
	12	9.00	240.00	150.00	200.00	0.20	96.11	92.93

Table 3
Experimental range and levels of independent variables for BB3 and MB

Factors	Factor code	Range and levels (coded)				
		-1.414	-1	0	+1	+1.414
Initial dye concentration (mg L^{-1})	A	50.00	71.97	125.00	178.03	200.00
Dosage (g)	B	0.05	0.07	0.13	0.18	0.20

[26]. The peak at 610 cm^{-1} was due to the bending modes of aromatic compounds.

3.2. Evaluation of variables affecting percentage uptake of dye

Plackett-Burman design was used to screen the relative importance of the various factors that influence the percentage uptake of dye. The dye concentrations were analyzed using Perkin Elmer Lambda

35 double-beam UV/visible spectrophotometer with 1.0 cm light path cuvette (quartz cell) at the wavelength corresponding to the maximum absorption; for BB3, $\lambda_{\text{max}} = 654\text{ nm}$, for MB, $\lambda_{\text{max}} = 664\text{ nm}$ and for BY11, $\lambda_{\text{max}} = 412\text{ nm}$. Dilutions were made when measurements exceeded the linearity of the calibration curves. Table 5 showed the analysis of variance (ANOVA) for BB3, MB and BY11. From the result, the $\text{Prob} > F$ value of 0.0027 for both BB3 and MB dye solutions indicate that the models were significant.

Table 4
The CCD matrix for two coded independent variables and the observed response for BB3 and MB

Dye	Experimental run	Coded values of variables		Observed response (%)	Predicted response (%)
		A	B		
BB3	1	−1.414	0	96.97	98.59
	2	+1.414	0	71.57	71.99
	3	0	−1.414	43.98	45.00
	4	0	+1.414	94.63	95.66
	5	−1	−1	79.95	78.50
	6	1	−1	50.61	50.02
	7	−1	1	97.36	95.92
	8	1	1	87.39	86.80
	9	0	0	82.13	83.70
	10	0	0	82.88	83.70
	11	0	0	85.08	83.70
	12	0	0	85.11	83.70
	13	0	0	83.30	83.70
MB	1	+1.414	0	57.78	58.86
	2	0	−1.414	47.13	47.75
	3	0	+1.414	91.92	92.55
	4	−1.414	0	94.65	94.81
	5	−1	1	95.49	95.19
	6	−1	−1	77.50	77.21
	7	1	−1	44.19	43.24
	8	1	1	79.28	78.33
	9	0	0	80.31	80.28
	10	0	0	82.40	80.28
	11	0	0	79.85	80.28
	12	0	0	80.93	80.28
	13	0	0	78.89	80.28

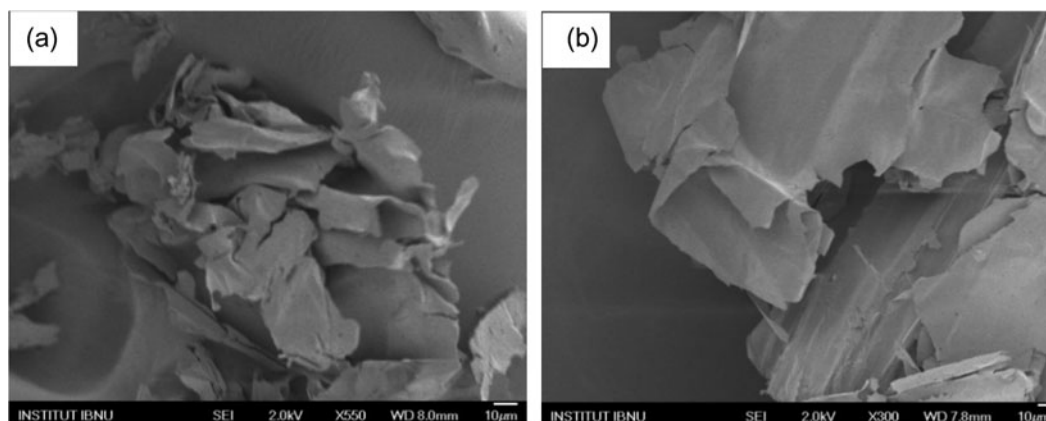


Fig. 2. FESEM images of NSB before adsorption (a) and after MB adsorption (b).

For each variable under studied, it is considered to be significant if it is having a value of $\text{Prob} > F$ less than 0.05. As such, both sorbent dosage and initial dye

concentration appeared to be the significant variables in affecting the uptake of BB3 and MB. This agrees well with some of the findings in conventional batch

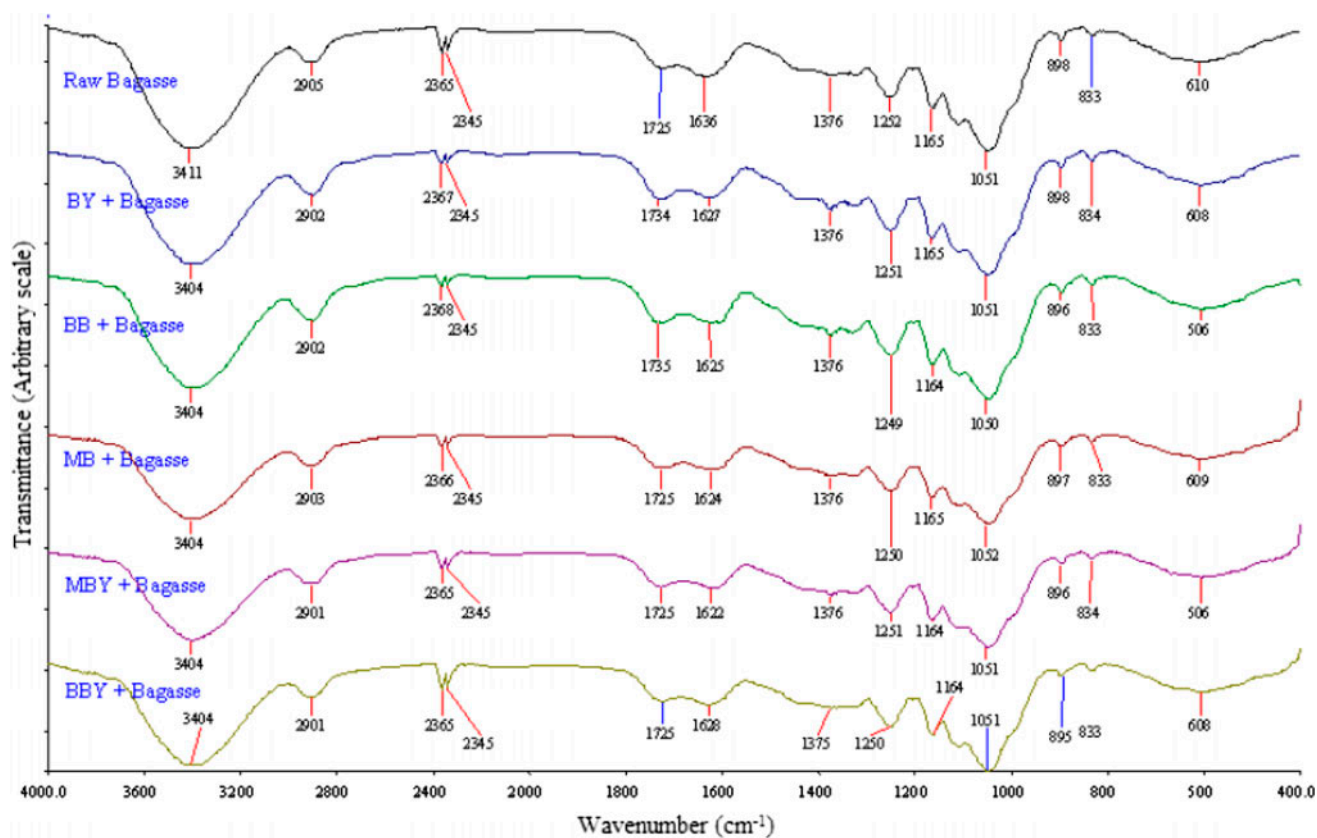


Fig. 3. FT-IR spectra of NSB (before and after sorption).

studies whereby both sorbent dosage and initial dye concentration were reported to be influential factors in dye uptake [12,25,27].

As for BY11, the value of $\text{Prob} > F$ is 0.5652 showed that the model was not significant. From the same table, it is also evident that all the model terms for BY11 were insignificant with $\text{Prob} > F$ higher than 0.05. In addition, BB3 and MB have greater percentage uptake over BY11. This might due to the different size of the adsorbates (Fig. 1). BB3 and MB have lower molecular size, and thus, they were more easily bind to the sorption sites in the binary solution compared with BY11, which was larger. The molecular conformation and dimension of adsorbate affect the adsorption, where planar molecules access and pack in the slit-shaped pores more efficiently as compared to nonplanar molecule [28].

3.3. Verification of Plackett–Burman design models

To validate the models, function of desirability was applied using Design-Expert Version 7.1.3. The experimental conditions to be verified were selected based on the highest desirability. Table 6 shows the

experimental conditions as well as the predicted and experimental values for BB3 and MB. The experimental values obtained agreed well with the predicted values for both models with relative small errors between the predicted and the actual values, which were 0.50 and 2.09%, respectively for BB3 and MB. Model of BY11 was not validated due to its insignificance.

3.4. Data analysis by RSM

The effects of initial dye concentrations and sorbent dosage on the percentage uptake of BB3 and MB dye solutions were studied using RSM. The modified cubic models describing the correlation between the 2 variables and the percentage uptake for both solutions were shown as follows:

BB3 dye solution:

$$\begin{aligned} \% \text{ Uptake} = & 114.943 - 1.377A + 259.837B \\ & + 9.032AB + 0.004A^2 - 2376.894B^2 \\ & - 0.0292A^2B \end{aligned} \quad (2)$$

Table 5
Regression analysis (ANOVA) of Plackett–Burman for the removal of BB3, MB and BY11 from dye solution

Dye	Source	Degree of freedom	Sum of square	Mean square	F-value	Prob > F
BB3	Model	5	8521.37	1704.27	14.46	0.0027
	pH	1	466.75	466.75	3.96	0.0937
	Contact time (min)	1	611.18	611.18	5.19	0.0630
	Particle size (µm)	1	549.18	549.18	4.66	0.0742
	Initial dye concentration (mg L ⁻¹)	1	3495.94	3495.94	29.67	0.0016
	Sorbent dosage (g)	1	3398.31	3398.31	228.84	0.0017
	Residual	6	707.07	117.84		
	Total	11	9228.44			
MB	Model	5	8744.01	1748.80	14.53	0.0027
	pH	1	462.60	462.60	3.84	0.0976
	Contact time (min)	1	581.11	581.11	4.83	0.0703
	Particle size (µm)	1	582.25	582.25	4.84	0.0701
	Initial dye concentration (mg L ⁻¹)	1	3628.53	3628.53	30.16	0.0015
	Sorbent dosage (g)	1	3489.52	3489.52	29.00	0.0017
	Residual	6	721.92	120.32		
	Total	11	9465.94			
BY11	Model	5	1696.75	339.35	0.84	0.5652
	pH	1	148.16	148.16	0.37	0.5664
	Contact time (min)	1	169.97	169.97	0.42	0.5400
	Particle size (µm)	1	0.41	0.41	1.02 × 10 ⁻³	0.9756
	Initial dye concentration (mg L ⁻¹)	1	275.78	275.78	0.68	0.4397
	Sorbent dosage (g)	1	1102.43	1102.43	2.74	0.1491
	Residual	6	2416.88	402.81		
	Total	11	4113.63			

Table 6
Plackett–Burman model validation

Dye solution	Factors					Percentage uptake (%)	
	pH	Contact time (min)	Particle size (µm)	Initial dye concentration (mg L ⁻¹)	Sorbent dosage (g)	Predicted	Experimental
BB3	8.76	193.00	178.00	194.00	0.19	87.49	85.98
MB	8.76	193.00	178.00	56.00	0.06	90.65	88.56

MB dye solution:

$$\begin{aligned} \% \text{ Uptake} = & 91.717 - 0.807A + 298.411B \\ & + 5.829AB + 0.002A^2 - 1836.390B^2 \\ & - 0.017A^2B \end{aligned} \quad (3)$$

Where A = initial dye concentration and B = sorbent dosage.

Cubic equation was used because the quadratic model was not significant while cubic model was found to be aliased. Tables 7 and 8 showed the ANOVA table for BB3 and MB, respectively. From

these tables, both of the models were significant ($p < 0.0001$) with model F -value of 186.06 and 302.55 for BB3 and MB, respectively. Coefficient of determination (R^2) for BB3 dye solution was found to be 0.9947 while the rest (0.53%) was explained as residues. On the other hand, R^2 for MB dye solution was reported as 0.9967. Both of the R^2 values were relatively high, showing that there were good agreements between the experimental and predicted values in both models. The closer the R^2 is to unity, the stronger the model and the better it predicts the response [29]. The predicted multiple correlation coefficient (predicted R^2) value of 0.9236 is reasonable agreement

Table 7
Regression analysis (ANOVA) for the removal of BB3 from dye solution

Source	Degree of freedom	Sum of square	Mean square	F-value	P
Model	6	3148.85	524.81	186.06	<0.0001
A	1	707.14	707.14	250.70	<0.0001
B	1	1283.11	1283.11	454.90	<0.0001
AB	1	93.76	93.76	33.24	0.0012
A ²	1	4.42	4.42	1.57	0.2574
B ²	1	310.88	310.88	110.22	<0.0001
A ² B	1	38.05	38.05	13.49	0.0104
Residual	6	16.92	2.82		
Lack of fit	2	9.73	4.87	2.71	0.1805

Notes: R²: 0.9947, Adjusted R²: 0.9893, Predicted R²: 0.9236, Adequate precision: 43.483 and C.V.: 2.10%.

Table 8
Regression analysis (ANOVA) for the removal of MB from dye solution

Source	Degree of freedom	Sum of square	Mean square	F-value	P
Model	6	3267.99	544.66	302.55	<0.0001
A	1	1292.27	1292.27	717.84	<0.0001
B	1	1003.46	1003.46	557.41	<0.0001
AB	1	73.18	73.18	40.65	0.0007
A ²	1	23.04	23.04	12.80	0.0117
B ²	1	185.57	185.57	103.08	<0.0001
A ² B	1	13.21	13.21	7.34	0.0351
Residual	6	10.80	1.80		
Lack of fit	2	3.96	1.98	1.16	0.4014

Notes: R²: 0.9967, Adjusted R²: 0.9934, Predicted R²: 0.9694, Adequate precision: 52.771 and C.V.: 1.76%.

with the adjusted multiple correlation coefficient (adjusted R²) value of 0.9893 for BB3 dye solution. Meanwhile, the value of predicted R² (0.9694) also reasonably agreed with the adjusted R² (0.9934) for MB dye solution.

The lack-of-fit observed for BB3 and MB models were 0.1805 and 0.4014, respectively. The insignificance in lack-of-fit implies that the model was valid. The coefficient of variance (C.V.) of BB3 model was reported as 2.10%. Meanwhile, the C.V. for MB model was 1.76%. The lower of the C.V. value, the greater is the precision and reliability of the experiments carried out [30]. Adequate precision indicates the signal to noise ratio and a ratio greater than 4 is desirable. A ratio of 43.483 and 52.771 obtained for BB3 and MB, respectively, showed an adequate signal. Thus, both models can be used to navigate the design space. Table 7 shows all the coefficients were significant ($p < 0.05$) except A²; however, it cannot be eliminated in order to support the hierarchy model. For MB model, all the coefficients were significant. BY11 dye

was not studied using RSM due to the insignificance of the model and all the studied factors at Plackett–Burman study.

The interaction between two factors and their optimum levels can be easily understood and located using response surface plot [30]. Fig. 4 shows the 3D surface plot for BB3 dye solution of interaction between initial dye concentration and sorbent dosage. The maximum percentage uptake of BB3 dye was observed when initial dye concentration was at minimum point, while sorbent dosage was at maximum point within the studied range. As the initial dye concentration increased, the percentage uptake decreased. Decrease in percentage uptake at high initial dye concentration might be due to insufficient of available binding sites and increase in the ratio of the BB3's cations to the dosage of the adsorbent. Similar observation was observed in the removal of Cd²⁺ [31]. As the sorbent dosage in the plot increase, the percentage uptake increased. The higher uptake observed can be related to the increase in the availability of adsorption

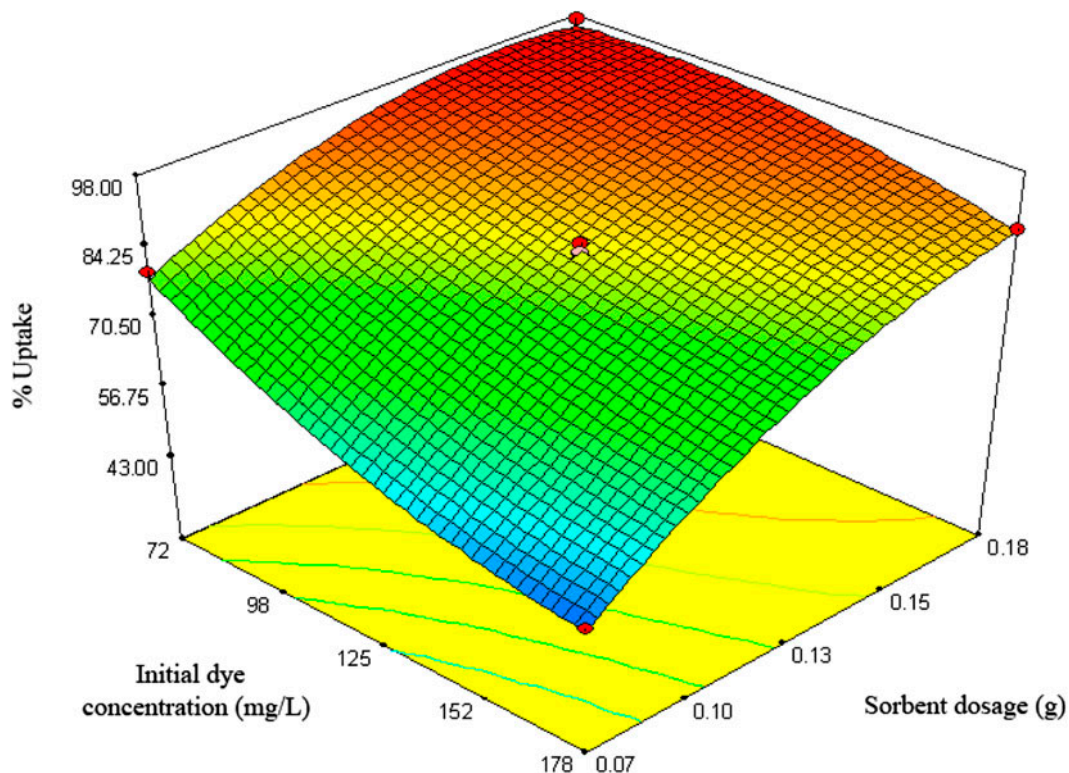


Fig. 4. 3D surface plot for uptake of BB3 in single dye solution as a function of initial dye concentration and sorbent dosage.

sites [25]. The optimum values of experiment factors obtained were 50 mg/L of initial dye concentration and 0.13 g of sorbent dosage. At these optimum operational values, the model predicted 98.59% uptake.

Fig. 5 represented the 3D surface plot for MB dye solution of interaction between initial dye concentration and sorbent dosage. A similar observation with BB3 dye solution was obtained. The maximum percentage uptake of MB dye was observed when the initial dye concentration was at minimum, while sorbent dosage was at maximum. Optimum operational values were reported as 72 mg/L of initial dye concentration and 0.18 g of sorbent dosage. The model predicted 95.19% uptake at these optimum operational values.

3.5. Verification of RSM models

In order to determine the validation of the model equations for both studied dye solutions, experiments were conducted based on the experimental conditions with the highest desirability which generated by Design Expert v.7.1.3 software. Table 9 shows the experiments conditions as well as the predicted and experiments results. From the experimental results

obtained, the percentage uptake for BB3 and MB dye solutions were 95.96 and 95.05%, respectively. Both of the results were close to the predicted results with percentage errors of 2.63% and 0.14% for BB3 and MB, respectively. Hence, both models can be concluded as valid.

4. Conclusion

The optimization and the modeling of percentage uptake of BB3, MB, and BY11 dye solutions were conducted using Plackett–Burman design and RSM. Plackett–Burman design was successfully applied to identify the most significant factors which influence the percentage uptake of the studied dye solutions. Initial dye concentration and sorbent dosage were the factors identified to be affecting the percentage uptake of BB3 and MB. None of the studied factors was found to be significantly affecting the percentage uptake of BY11 dye. The optimum operational condition was determined through RSM. An uptake greater than 95% was achieved for BB3 at initial dye concentration of 50 mg/L and sorbent dosage of 0.13 g. Meanwhile, the optimal conditions for MB uptake were identified as 72 mg/L of initial dye concentration

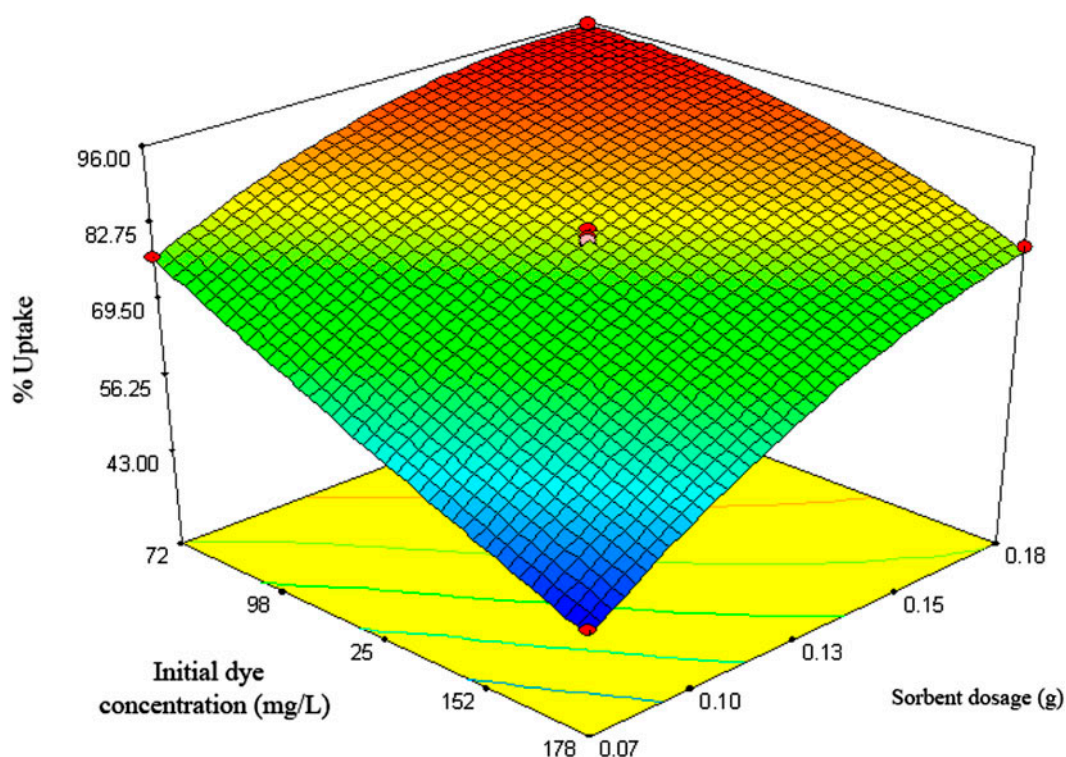


Fig. 5. 3D surface plot for uptake of MB in single dye solution as a function of initial dye concentration and sorbent dosage.

Table 9
Validation of model equations for BB3 and MB dye solution

Dye solution	Factors		Percentage uptake (%)		Percentage error (%)
	Sorbent dosage (g)	Initial dye concentration (mg L ⁻¹)	Predicted	Observed	
BB3	0.13	50	98.59	95.96	2.63
MB	0.18	72	95.19	95.05	0.14

and 0.18 g of sorbent dosage. With these operational conditions, the percentage uptake of MB was obtained as 95.19%. The observed values agreed well with the predicted values in the model validation test with percentage errors of 2.63 and 0.14% for BB3 and MB dye solutions, respectively.

Acknowledgments

The authors are thankful for the support by the International Foundation for Science, Stockholm, Sweden and the Organization for the Prohibition of Chemical Weapons, The Hague, the Netherlands via grant number W/4386-1 and teaching assistantship for E.C. Khoo by Universiti Tunku Abdul Rahman (UTAR).

References

- [1] A.R. Gregory, S. Elliot, P. Kludge, Ames testing of direct black 3B parallel carcinogenic, *J. Appl. Toxicol.* 1 (1991) 308–313.
- [2] G. McKay, A. Sweeny, Principles of dye removal from textile effluent, *Water Air Soil Pollut.* 14 (1980) 3–11.
- [3] R.P. Han, D.D. Ding, Y.F. Xu, W.H. Zou, Y.F. Wang, W.F. Li, L.N. Zou, Use of rice husk for the adsorption of Congo red from aqueous solution in column mode, *Bioresour. Technol.* 99 (2008) 2938–2946.
- [4] R.P. Han, Y.F. Wang, W.H. Yu, W.H. Zou, J. Shi, H.M. Liu, Biosorption of methylene blue from aqueous solution by rice husk in a fixed-bed column, *J. Hazard. Mater.* 141 (2006) 713–718.
- [5] S.T. Ong, W.N. Lee, P.S. Keng, S.L. Lee, Y.T. Hung, S.T. Ha, Equilibrium studies and kinetics mechanism for the removal of basic and reactive dyes in both single and binary systems using EDTA modified rice husk, *Int. J. Phys. Sci.* 5 (2010) 582–595.

- [6] S.T. Ong, P.S. Keng, C.K. Lee, Basic and reactive dyes sorption enhancement of rice hull through chemical modification, *Am. J. Appl. Sci.* 7 (2010) 447–452.
- [7] H. Aydin, G. Baysal, Y. Bulut, Utilization of walnut shells (*Juglans regia*) as an adsorbent for the removal of acid dyes, *Desalin. Water Treat.* 2 (2009) 139–147.
- [8] L.S. Oliveira, A.S. Franca, T.M. Alves, S.D.F. Rocha, Evaluation of untreated coffee husks as potential biosorbents for treatment of dye contaminated waters, *J. Hazard. Mater.* 155 (2008) 507–512.
- [9] R.M. Gong, S.X. Zhu, D.M. Zhang, J. Chen, S.J. Ni, R. Guan, Adsorption behaviour of cationic dyes on citric acid esterifying wheat straw: Kinetic and thermodynamic profile, *Desalination* 230 (2008) 220–228.
- [10] S.T. Ong, P.S. Keng, S.T. Ooi, Y.T. Hung, S.L. Lee, Utilization of fruits peel as a sorbent for removal of methylene blue, *Asian J. Chem.* 24 (2012) 398–402.
- [11] H.C. Chu, K.M. Chen, Reuse of activated sludge biomass: I. Removal of basic dyes from wastewater by biomass, *Process Biochem.* 37 (2002) 595–600.
- [12] S.T. Ong, C.K. Lee, Z. Zainal, Removal of basic and reactive dyes using ethylenediamine modified rice hull, *Bioresour. Technol.* 98 (2007) 2792–2799.
- [13] S.Y. Wong, Y.P. Tan, A.H. Abdullah, S.T. Ong, The removal of basic and reactive dyes using quarterised sugarcane bagasse, *J. Phys. Sci.* 20 (2009) 59–74.
- [14] S.T. Ong, S.Y. Tan, E.C. Khoo, S.L. Lee, S.T. Ha, Equilibrium studies for Basic Blue 3 adsorption onto durian peel (*Durio zibethinus* Murray), *Desalin. Water Treat. J.* 45 (2012) 161–169.
- [15] S.T. Ong, E.C. Khoo, S.L. Hii, S.T. Ha, Utilization of sugarcane bagasse for removal of basic dyes from aqueous environment in single and binary systems, *Desalin. Water Treat. J.* 20 (2010) 86–95.
- [16] F.A. Batzias, D.K. Sidiras, Dye adsorption by calcium chloride treated beech sawdust in batch and fixed-bed systems, *J. Hazard. Mater. B114* (2004) 167–174.
- [17] V. Vadivelan, K. Vasanth Kumar, Equilibrium, kinetics, mechanism, and process design for the sorption of methylene blue onto rice husk, *J. Colloid Interface Sci.* 286 (2005) 90–100.
- [18] M.Y. Chang, R.S. Juang, Equilibrium and kinetic studies on the adsorption of surfactant, organic acids and dyes from water onto natural biopolymers, *Colloids Surf. A: Physicochem. Eng. Aspects* 269 (2005) 35–46.
- [19] K. Vasanth Kumar, V. Ramamurthi, S. Sivanesan, Modeling the mechanism involved during the sorption of methylene blue onto fly ash, *J. Colloid Interface Sci.* 284 (2005) 14–21.
- [20] S.J. Allen, G. McKay, J.F. Porter, Adsorption isotherm models for basic dye adsorption by peat in single and binary component systems, *J. Colloid Interface Sci.* 280 (2004) 322–333.
- [21] G.Z. Kyzas, N.K. Lazaridis, Reactive and basic dyes removal by sorption onto chitosan derivatives, *J. Colloid Interface Sci.* 331 (2009) 32–39.
- [22] K. Ravikumar, S. Ramalingam, S. Krishnan, K. Balu, Application of response surface methodology to optimize the process variables for Reactive Red and Acid Brown dye removal using novel adsorbent, *Dyes Pigm.* 70 (2006) 18–26.
- [23] A. Rajendran, M. Thirugnanam, V. Thangavelu, Statistical evaluation of medium components by Plackett-Burman experimental design and kinetic modeling of lipase production by *Pseudomonas fluorescens*, *Indian J. Biotechnol.* 6 (2007) 267–278.
- [24] R.H. Myers, D.C. Montgomery, *Response Surface Methodology*, second ed., John Wiley and Sons, New York, NY, 1995.
- [25] V.K. Garg, R. Kumar, R. Gupta, Removal of malachite green dye from aqueous solution by adsorption using agro-industry waste: A case study of *Prosopis cineraria*, *Dyes Pigm.* 62 (2004) 1–10.
- [26] U.K. Garg, M.P. Kaur, V.K. Garg, D. Sud, Removal of nickel (II) from aqueous solution by adsorption on agricultural waste biomass using a response surface methodological approach, *Bioresour. Technol.* 99 (2008) 1325–1331.
- [27] D.A. Fungaro, M. Bruno, L.C. Grosche, Adsorption and kinetic studies of methylene blue on zeolite synthesized from fly ash, *Desalin. Water Treat. J.* 2 (2009) 231–239.
- [28] Y. Guo, S. Kaplan, T. Keranfil, The significance of physical factors on the adsorption of polyaromatic compounds by activated carbons, *Carbon* 46 (2008) 1885–1891.
- [29] K. Chauchan, U. Trivedi, K.C. Patel, Application of response surface methodology for optimization of lactic acid production using date juice, *J. Microbiol. Biotechnol.* 16 (2006) 1410–1415.
- [30] J.K. Kim, B.R. Oh, H. Shin, C. Eom, S.W. Kim, Statistical optimization of enzymatic saccharification and ethanol fermentation using food waste, *Process Biochem.* 43 (2008) 1308–1312.
- [31] J.I.S. Khattar, Shailza Optimization of Cd²⁺ removal by the cyanobacterium *Synechocystis pevalekii* using the response surface methodology, *Process Biochem.* 44 (2009) 118–121.