



Application of Fin System on Adsorption of Methylene Blue Dye using Adsorbent Coating Layer: Mathematical Formulae

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Abstract. Dye solution treatment using a fin system is a new adsorption treatment technique that can replace expensive conventional adsorption treatment units that are currently mostly used in industry. The fins function as the media for an adsorbent coating and are immersed in a tank containing dye wastewater. By installing the coated fins in the tank, the dye concentration can be easily reduced up to 90%. The purpose of the present study was to develop simple mathematical equations for the fin system using an adsorbent coating layer (ACL). The equilibrium adsorption data fitted to the Langmuir isotherm, indicating that the adsorption surface is homogeneous and in monolayer state. A mathematical formula derived from the isotherm model of adsorption was developed that can be used on large-scale removal of methylene blue dye from industrial wastewater since the error percentage between the calculated and the experimental values was less than 15%.

Keywords: *adsorbent coating layer (ACL); adsorption; fin system; mathematical equations; methylene blue dye; treatment.*

1 Introduction

Water contamination has become a serious environmental threat to the world owing to rapid industrialization and urbanization [1]. A number of industries (e.g. plastics, textile, cosmetics, leather, food, etc.) use dyes for coloring their products, which produces wastewater containing high concentrations of coloring agents absorbed by organic substances. The textile industry is known as the most chemically intensive industry in the world, as it uses more than 2,000 types of chemicals and 7,000 types of dyes, thus being the number one polluter of fresh water after agriculture [2]. The carcinogenic and mutagenic characteristics of dyes cause severe harm to aquatic life and human beings (e.g. dysfunction of kidneys, reproductive system, liver, brain and central nervous system) [3]. Removal of coloring substances from wastewater includes various techniques such as adsorption, precipitation, coagulation, filtration, and chemical oxidation [4].

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Adsorption is one of the preferred methods in industry as it is a good alternative for the removal and recovery of dyes from wastewater due to its cost efficiency, good removal efficiency and convenience in operation when compared to other methods [5]. An adsorbent coating layer (ACL) has been devised for the removal of MB dye in wastewater [6]. It is formed from the combination of bentonite, water-based paint and solvent at a specific ratio (0.3:2.5:2). It can be categorized as an effective adsorbent due to its high removal percentage of dye at 99% recovery [6].

Dye solution treatment using a fin system is a new adsorption treatment technique that can replace expensive conventional adsorption treatment units that are currently mostly used in industry. The concept of the fin was taken from heat exchangers and heat sinks in order to increase the adsorption surface area. The fins function as the media for an adsorbent coating layer and can be immersed in a tank containing dye wastewater in simple and easy way. The present study describes the development of an adsorbent coating layer (ACL) for a fin system and its mathematical formulae derived from the treatment of wastewater containing methylene blue dye from industrial applications.

2 Experiment

2.1 Materials and Methods

2.1.1 Adsorbate

Methylene blue (MB) was purchased from Modern Lab Sdn. Bhd. Malaysia and used without further purification. The preparation of MB stock solution at 1000 mg L⁻¹ concentrations was carried out by dissolving 1 g of MB powder in 1 L of distilled water. The stock solution was also diluted at certain concentrations (100-1000 mg L⁻¹) for further experimentation.

2.1.2 Adsorbent Coating Preparation

The adsorbent coating was prepared by mixing water-based paint, bentonite clay and also water. The bentonite was supplied by Modern Lab Sdn. Bhd. Malaysia and used as received without any adjustments. The water-based paint or binder consists of calcium carbonate, polyvinyl alcohol, formalin, ammonia and hydrosol, and was procured from Sissons Paints Sdn. Bhd. The mixtures were stirred homogeneously using an agitation mixer until the components were completely dispersed.

2.2 Instrument

The equilibrium dye concentrations were analyzed using a UV-Vis spectrophotometer (HASH UV-Vis model DR 5000 Spectrophotometer) at a wavelength of 664 nm and giving the reading in terms of absorbance. A calibration curve was plotted in order to know the concentration of the unknown dye solution.

2.3 Adsorption Equilibrium and Isotherms

Modelling of experimental adsorption isotherm data is an important way to anticipate the mechanisms of adsorption that lead to an improvement in the area of adsorption [7]. Two types of isotherm models, incorporating the Langmuir and Freundlich isotherms respectively, were examined in this work. The equilibrium data acquired from the experiment were fitted and analyzed to these two models. The suitability of the isotherm equation for the equilibrium data was compared with the values of the correlation coefficient, R^2 , where a higher value, near to 1, means a good fit to the isotherm model. Studying the adsorption isotherm model was critical in order to know which equation was most suitable to be developed for the fin system. Afterward, the equation from the isotherm was combined with other adsorption equations to produce new mathematical formulae for the fin system by utilizing ACL in treating MB dye wastewater.

Generally, the empirical Langmuir isotherm model assumes monolayer adsorption (the adsorbed layer is one molecule in thickness), where adsorption can only occur at a finite (fixed) number of definite localized sites that are identical and equivalent, with no lateral interaction and steric hindrance between the adsorbed molecules, even on neighboring sites, and homogeneous adsorption [8]. The Langmuir isotherm equation is expressed by the following Eq. (1):

$$q_e = \frac{q_{max} K_L C_e}{1 + K_L C_e} \quad (1)$$

The Freundlich isotherm is an empirical equation for multilayer and heterogeneous adsorption sites. The Freundlich isotherm model can be expressed with the following Eq.(2):

$$q_e = K_F C_e^{1/n} \quad (2)$$

where C_e is the equilibrium concentration of solute in the bulk solution (mg L^{-1}), q_e is the amount of solute adsorbed per unit weight of adsorbent (mg g^{-1}), K_L is the Langmuir adsorption constant (L mg^{-1}), q_{max} is the monolayer adsorption capacity of the adsorbent (mg g^{-1}), K_F is the Freundlich isotherm constant (mg g^{-1}) (L mg^{-1})^{1/n}, and n is the adsorption intensity.

2.4 Methods of Validation

2.4.1 Constant Volume of MB Dye Solution

The experimental results from the study of a constant volume of MB dye solution were used to validate the mathematical equations. Three initial concentrations, 100, 200 and 300 mg L⁻¹ of MB dye solution at 2, 4 and 6 h, were selected as reference values (Table 1). The adsorbent mass was calculated using a mathematical formula derived beforehand by inserting the values of C_i, V, K_L, q_{max} and C_t and R into Eqs. (9) to (12). The stainless steel plates were coated according to the mass of adsorbent calculated and dried at 70 °C for 10 h in order to validate the calculated value. The coated fins were immersed in MB solutions and the dye removal capacity percentage was measured at various time intervals. Then, the experimental result was compared with the reference values (Table 1) for the conforming calculated adsorbent mass of ACL to produce a targeted dye removal percentage.

Table 1 Standard table of concentrations at different times, C_t (mg L⁻¹).

Time (h)	Initial concentration (mg L ⁻¹)									
	100		200		300		400		500	
	C _t	R	C _t	R	C _t	R	C _t	R	C _t	R
0	100.00	0	200.00	0	300.00	0	400.00	0	500.00	0
1	60.87	39	119.35	40	201.14	33	298.62	25	369.95	26
2	37.09	63	92.72	54	197.81	34	268.66	33	342.37	32
3	16.37	84	86.54	57	187.31	37	257.73	36	339.51	32
4	15.96	84	79.41	60	169.28	44	249.64	38	332.38	34
5	15.45	85	72.28	64	167.86	44	246.64	38	327.15	35
6	15.16	85	68.95	66	165.00	45	242.04	39	322.87	35
7	15.08	85	59.44	70	159.30	47	239.66	40	309.56	38
8	15.04	85	57.06	71	155.02	48	239.18	40	308.13	38
23	12.46	88	49.45	75	152.64	49	238.23	40	307.18	39
24	12.45	88	48.03	76	151.69	49	237.28	41	305.75	39
Time (h)	Initial concentration (mg L ⁻¹)									
	600		700		800		900		1000	
	C _t	R	C _t	R	C _t	R	C _t	R	C _t	R
0	600.00	0	700.00	0	800.00	0	900.00	0	1000.00	0
1	509.75	15	576.32	18	677.13	15	791.25	12	929.15	7
2	505.94	16	565.38	19	670.95	16	771.75	14	927.72	7
3	476.46	21	560.15	20	662.86	17	757.01	16	915.36	8
4	472.66	21	555.40	21	660.49	17	751.31	17	910.60	9
5	469.33	22	553.54	21	651.45	19	750.83	17	903.95	10
6	465.53	22	550.64	21	644.79	19	747.50	17	894.44	11
7	459.34	23	546.36	22	639.09	20	745.13	17	875.42	12
8	452.69	25	542.56	22	632.91	21	742.27	18	851.17	15
23	450.31	25	540.66	23	622.92	22	741.32	18	850.69	15
24	449.36	25	539.23	23	621.97	22	737.99	18	849.26	15

2.4.2 Varying Volume of MB Dye Solution

The purpose of this experiment was to verify the validity of the mathematical equations for the fin system with a large-scale application volume of dye wastewater. Three different volumes of MB solutions (0.5, 10 and 50 L) were measured at the same initial concentration (100 mg L^{-1}). The adsorbent mass was calculated by inserting the values of C_i (100 mg L^{-1}), K_L (0.16 L mg^{-1}), q_{\max} (277.8 mg g^{-1}), C_i (12.45 mg L^{-1}) and R (88 %) into Eqs. (9) to (12). Again, the plates were coated according to the adsorbent mass calculated and dried at 70°C for 10 h.

The coated fins were immersed in the MB solutions and aerated at the same pump power (220~240 V/50 Hz/5.6 W). The concentration of the MB solution was measured every 1 h for 24 h. The experimental results of the three volumes were compared to the conforming calculated adsorbent mass of the ACL to see if it can be applied in large-scale treatment of dye wastewater.

3 Results and Discussion

3.1 Isotherm Model Studies

It is essential to find the best fitted isotherm model of ACL for MB dye removal in order to establish the appropriate mathematical equations for the fin system. Two types of isotherm models, i.e. the Langmuir and Freundlich models, were plotted and analyzed. Determination of the adsorption isotherm of MB was performed at 30°C and a range of concentrations ($100\text{-}1000 \text{ mg L}^{-1}$).

The adsorption isotherms were determined according to the linear form of the Langmuir and Freundlich models, and different parameters were determined, i.e. maximum adsorption amount (q_{\max}), Langmuir constant (K_L), adsorption intensity (n), Freundlich isotherm constant (K_F) and correlation coefficient (R^2). These values were acquired from the intercept and the slope of a linear plot of the experimental data.

Figure 1 (a) and (b) show the plots of the Langmuir and Freundlich models for adsorption of MB onto ACL at 30°C . The correlation coefficient value, R^2 , of the Langmuir isotherm was 0.9871, whereas for the Freundlich isotherm, the R^2 value was 0.655. The R^2 of the Langmuir model was close to 1, which suggests that the Langmuir model fitted to the experimental adsorption data better compared to the Freundlich model. As a consequence, the result demonstrates that the adsorption of MB by ACL takes place in a monolayer adsorption manner [9].

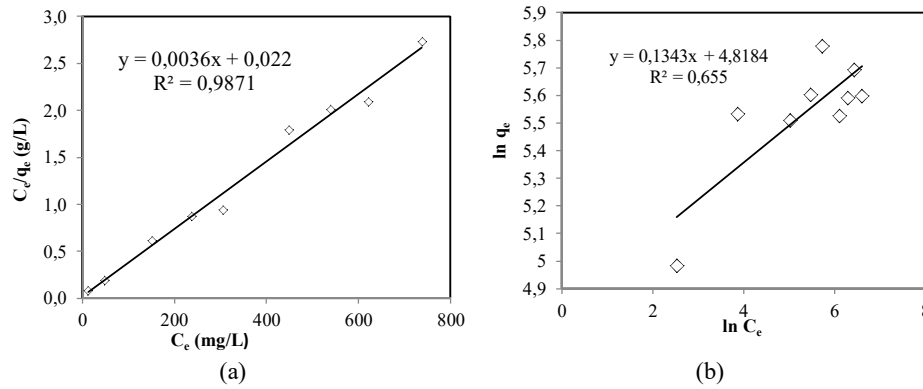


Figure 1 Plots of (a) Langmuir and (b) Freundlich isotherms for MB adsorption onto adsorbent coating layer at different concentrations (at 30 °C).

Table 2 Langmuir and freundlich models constant and correlation coefficients for sorption of mb onto adsorbent coating layer at 30 °C.

	Langmuir		Freundlich
q_{max}	277.8	n	7.45
K_L	0.16	K_F	123.77
R^2	0.9871	R^2	0.655

3.2 Mathematical Formula for Fin System

The amount of ACL on the fins was calculated during the batch experiment by developing simple mathematical equations. These equations can be utilized by any industry that uses methylene blue as well as cationic dyes in their handling. From the equation, the required amount of ACL to be coated can be calculated as well as the minimum time durations for the dye removal. The assumptions of the mathematical equations for the fin system are as follows:

1. Monolayer adsorption
2. Constant temperature (30 °C)
3. Constant q_{max} (277.8 mg g⁻¹)
4. Constant K_L (0.16 L mg⁻¹)
5. Constant R, T, C_i (Table 1)

The main equation involved in this work is the mass balance equation of adsorption. The equation is as follows:

$$q_e = \frac{(C_i - C_f)V}{m_b} \tag{3}$$

where C_i is the initial concentration of the solution (mg L⁻¹), C_f is the final concentration of the solution (mg L⁻¹), q_e is the maximum adsorption capacity of

the adsorbent (mg g^{-1}), m_b is the mass of bentonite (g), and V is the volume of wastewater (L).

Since the ACL fitted well to the Langmuir isotherm, the non-linear equation of the isotherm was preferred in this work, as presented in Eq. (1) above. Eq. (3) = Eq. (1)

$$\frac{(C_i - C_f)V}{m_b} = \frac{q_{\max} K_L C_e}{1 + K_L C_e} \quad (4)$$

where K_L is the Langmuir adsorption constant (L mg^{-1}), q_{\max} is the monolayer adsorption capacity of the adsorbent (mg g^{-1}), and C_e is equilibrium concentration of the solute in the bulk solution (mg L^{-1}).

By re-arranging Eq. (4),

$$m_b = \frac{(C_i - C_f)V(1 + K_L C_e)}{q_{\max} K_L C_e} \quad (5)$$

By replacing

$$(C_i - C_f) = RC_i \quad (6)$$

By inserting Eq. (6) into Eq. (5),

$$m_b = \frac{(RC_i V)(1 + K_L C_e)}{q_{\max} K_L C_e} \quad (7)$$

Since we need the concentration at a time before equilibrium is reached, Eq. 8 was developed. Assume that, at equilibrium,

$$C_e = C_t \quad (8)$$

By inserting Eq. (8) into Eq. (7),

$$m_b = \frac{(RC_i V)(1 + K_L C_t)}{q_{\max} K_L C_t} \quad (9)$$

where C_t is the concentration of the adsorbate at a certain time (mg L^{-1}) and R is the dye removal percentage (%) (in between 0 and 1). An adsorbent/binder/solvent ratio of 0.3:2.5:2.0 was chosen, being the optimum value of the adsorbent coating formulation established by a previous researcher [6]. Thus, as shown in Eqs. (10) and (11), the correlation equation of ACL mass becomes,

$$m_p = \left(\frac{2.5}{0.3}\right) m_b \quad (10)$$

$$m_w = \left(\frac{2.0}{0.3}\right) m_b \quad (11)$$

Total mass of ACL needed,

$$m_t = m_b + m_p + m_w \quad (12)$$

where m_p means mass of water-based paint (g), m_w is mass of water (g), and m_t is total mass of ACL (Eq.(12)). In order to determine the total number of fins, the total surface area of the fins was calculated. The thickness of the ACL was assumed 0 as the value is very small, i.e. 87 μm . The equation of the total surface area of the fin is as follows in Eq.(13):

$$S = 2 (l \times w) \quad (13)$$

In the current study, 4.8 g of ACL (0.3:2.5:2) and 0.0112 m^2 (2 x 0.07 x 0.08) of total surface area were selected. The mass of ACL per fin can be calculated by using the following Eq.(14):

$$m_f = \left(\frac{4.8}{0.0112} \right) S \quad (14)$$

Hence, the total number of fins needed can be found by using the following Eq.(15):

$$n_f = \frac{m_t}{m_f} \quad (15)$$

where S is the total surface area (m^2), l and w are the length and width of the fins in contact with the dye solution (m), m_f is the ACL mass on the fins (g), m_t is the total mass of ACL required (g), and n_f is the total number of fins needed. The percentage error of the calculation and the experimental value for the validation study was calculated using Eq. (16):

$$\left| \text{percentage error (\%)} = \left(\frac{\text{exp value} - \text{calc value}}{\text{exp value}} \right) \times 100 \right| \quad (16)$$

3.3 Validation Results

3.3.1 Constant Volume of MB Dye Solution

A validation experiment was carried out to prove the validity of the developed mathematical equations (Eqs. (9) to (16)) by using a constant volume (0.5 L) of MB solution and varying the initial concentration (100, 200 and 300 mg L^{-1}). The MB dye removal percentage, R, and the concentration at a certain time, C_t , at different initial dye concentrations can be obtained from Table 1. Table 1 contains the concentration value of MB at a certain time; C_t at different removal percentages; and R. It is a standard table if these mathematical equations are used for MB dye treatment. The amount of ACL required was determined by inserting values of C_i , V, q_{max} , K_L and R into Eqs. (9) to (12). The calculated mass was coated onto a stainless steel plate, dried and immersed in 0.5 L of MB solution at 2, 4 and 6 h. The dye removal percentage (calculated using Eq. (9))

was compared with the reference values (Table 1) in order to know the MB dye removal error percentage of the calculated values compared to the experimental values. The validation result is presented in Figure 2.

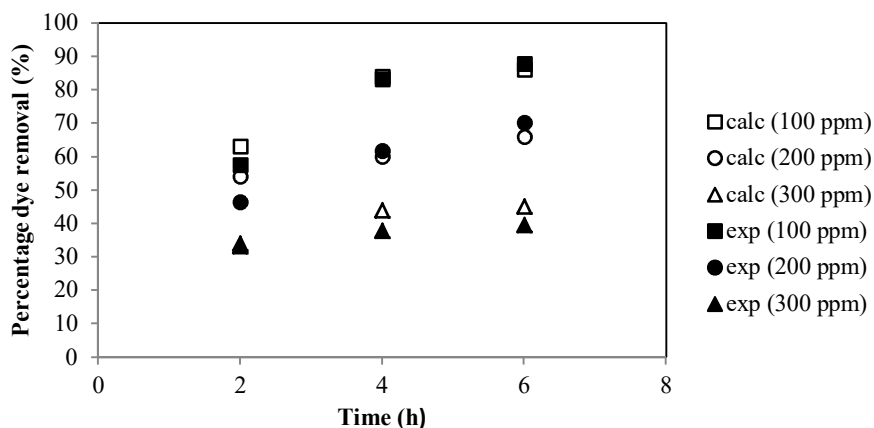


Figure 2 Graph of time of dye removal versus removal percentage at different adsorbent dosages (constant volume of MB dye: 0.5 L).

It can be seen from Figure 2 that the removal percentages achieved experimentally by applying the calculated mass of ACL for a dye concentration of 100 mg L^{-1} at 2, 4 and 6 h were 57, 83 and 88%, whereas for a concentration 200 mg L^{-1} , the dye removal percentages were 46, 62 and 70%. For a concentration of 300 mg L^{-1} , the MB dye removal percentages were 33, 38 and 39% in 2, 4 and 6 h.

The results of the removal percentages for the different adsorbent masses and initial dye concentrations are shown in Table 3. By referring to the reference results (Table 1) for a 100 mg L^{-1} concentration of dye, the removal percentages at 2, 4 and 6 h were 63, 84 and 85% respectively. However, as can be seen in Table 3, the experimental values (from calculation) were different from the reference values with an error of about 9.5, 1.2 and 3.4%. At a concentration of 200 mg L^{-1} dye solution, the reference values at 2, 4 and 6 h were 54, 60 and 66%, while for 300 mg L^{-1} dye solution, the removal percentages were 34, 44 and 45%. Besides, the error percentages for 200 mg L^{-1} were found to be 14.8, 3.2 and 5.7%, while at 300 mg L^{-1} the error percentages were 2.9, 13.6 and 13.3%.

The values based on calculation using the formula derived earlier deviated from the reference values (Table 1) due to errors that occurred experimentally. The errors may have occurred when taking the measurements, as the UV-Vis spectrophotometer is a sensitive instrument, potentially giving variations in

different readings. Therefore, it was proven that the mathematical equations for the fin system (Eqs. (9) to (16)) are valid and can be applied by industry for the treatment of MB dyes using ACL, since the error percentage is within 15%.

Table 3 Calculation of ACL Mass and removal percentage (%) at constant volume (0.5 L).

C_i (mgL^{-1})	R_{ref} (%)	T (h)	C_t (mg L^{-1})	m_b (g)	m_p (g)	m_w (g)	m_t (g)	R_{calc} (%)	error (%)
100	63	2	37.09	0.13	1.10	0.88	2.11	57	9.5
	84	4	15.96	0.21	1.75	1.40	3.36	83	1.2
	85	6	15.16	0.22	1.83	1.47	3.52	88	3.5
200	54	2	92.72	0.21	1.73	1.38	3.32	46	14.8
	60	4	79.41	0.23	1.94	1.55	3.72	62	3.3
	66	6	68.95	0.26	2.16	1.73	4.15	70	6.1
300	34	2	197.81	0.19	1.58	1.26	3.03	33	2.9
	44	4	169.28	0.25	2.05	1.64	3.94	38	13.6
	45	6	165.00	0.25	2.10	1.68	4.03	39	13.3

3.3.2 Varying Volume of MB Dye Solution

The validation experiment for a huge volume of dye wastewater is highly applicable since the textile industry uses huge amounts of water in their processing operations [10]. For this reason, the performance of ACL coated on a fin system in a large volume of wastewater was examined using the mathematical equations developed. The fins were coated with the amount of ACL calculated using Eqs. (9) to (12) at an initial MB dye solution concentration of 100 mg L^{-1} . These coated plates were immersed in three different volumes of MB solution (0.5, 10 and 50 L) and aerated at the same pump power (220~240 V/ 50 Hz / 5.6 W).

It can be seen from Figure 3 that the adsorption rate accomplished equilibrium state after 3 h for 0.5 and 10 L of MB solution with a 83.6 and 83.5% dye removal percentage. However, for 50 L of MB solution, the equilibrium state was achieved after 4 h with a dye removal percentage of 86.4%. The large volume of wastewater required a longer equilibrium time compared to the smaller volumes, i.e. 0.5 and 10 L. The main reason for the reduction of the MB dye removal rate may be poor fluid mixing causing inconsistencies in the non-consistencies of liquid inside the tank because of the same power of aeration pump was utilized. This is because mixing is sure to be poor unless the flow in the tank is turbulent. For the mixing to be effective, the fluid circulated by the impeller or aeration must sweep the whole tank so that the MB dye particles are dispersed well onto the ACL [11]. Additionally, the removal percentage results at various masses of adsorbent are summarized in Table 4.

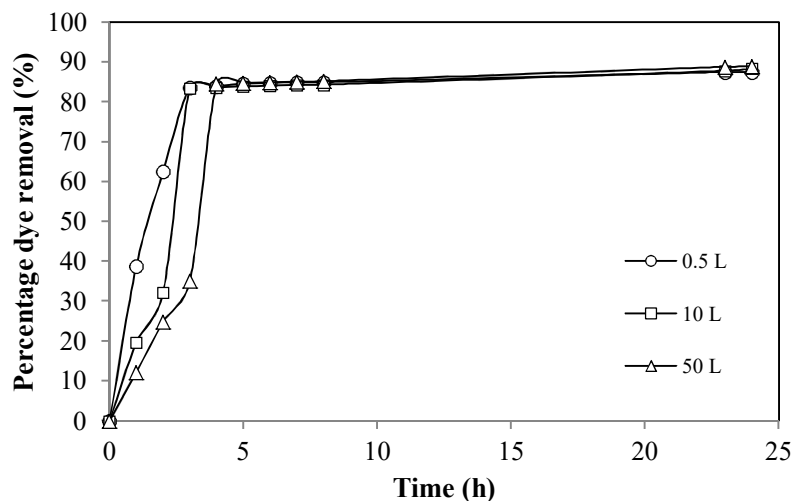


Figure 3 Graph of time of dye removal versus removal percentage at different adsorbent dosages and volumes (concentration: 100 mg L^{-1} , temperature: $30 \text{ }^\circ\text{C}$).

Table 4 Calculation of adsorbent mass and removal percentage (%) at different volumes for 24 h.

V (L)	C_i (mg L^{-1})	R_{ref} (%)	C_t (mg L^{-1})	m_b (g)	m_p (g)	m_w (g)	m_t (g)	R_{exp} (%)
0.5	100	88	12.45	0.30	2.50	2.00	4.8	83.6
10	100	88	12.45	4.76	39.70	31.73	76.19	83.5
50	100	88	12.45	23.79	198.25	158.60	380.30	86.4

4 Conclusions

The equilibrium adsorption data in this study best fitted to the Langmuir isotherm, indicating homogeneous monolayer adsorption. Mathematical equations for the fin system were effectively formulated by combining the mass balance adsorption equation and the Langmuir isotherm equation respectively. In small-scale applications, it was revealed that the mathematical formula can be used for the removal and recovery of MB dye because the error percentage between calculated and experimental values was below 15%. Furthermore, it was proven that the developed equations can be adequately applied for the treatment of MB using ACL in large-scale treatment of industrial wastewater.

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