Research Paper

REMOVAL OF BASIC DYE (METHYLENE BLUE) USING LOW COST BIOSORBENT: WATER HYACINTH

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INTRODUCTION

Environmental pollution has recently become a serious problem worldwide [1]. Colour is the first contaminant to be recognized in fresh water and has to be removed from wastewater before discharging into water bodies [2]. Dyes are widely used in Indian industries, viz., textile, paper, plastic, leather, printing, food, cosmetics, etc., to colour their final product. There are more than ten thousand types of commercially available dyes with over seven hundred tons of dyestuff produced annually worldwide [3]. In textile industries, about 1000 L of water is used for every 1000kg clothes processed. It is estimated about 10-15% of the dyes were lost in industrial effluent [4]. Discharge of such coloured effluents impart colour to the receiving water bodies (rivers, stream and lakes) and causes many significant problems such as increasing the toxicity and chemical oxygen demand. It also reduces light penetration which is a derogatory effect on photosynthesis phenomena [5]. However, wastewater with dyes is very difficult to treat, since the dyes are recalcitrant organic molecules, are resistant to aerobic digestion and are stable to light, heat and oxidizing agent [6].

The most commonly used methods for colour removal are biological and chemical precipitation. However, these processes are effective and economical only in cases where solute concentrations are relatively high [7]. The physicochemical processes are used to treat dye laden wastewaters. These processes are costly and cannot effectively be used to treat the dye wastewater at a wide range. The advantages and disadvantages of selected methods of dye removal from wastewaters are presented in Table 1.

Table 1 : Advantages and limitations of the methods used for dye removal

Physical / Chemical Methods	Process	Advantages	Disadvantages	References
Fentons reagent	Oxidation by using H_2O_2-Fe	Effective Decolourisation of all dyes	High Volume of Sludge Generation	[8]
Ion exchange	Ion exchange resin	Regeneration of adsorbent	Not effective for all dyes	[9]
Ozonation	Oxidation using Ozone gas	Effluent volume remains same	High Cost and half-life (20 minutes)	[10]
Photochemical	Oxidation using H_2O_2-UV	Zero sludge production	Formation of byproducts	[9]
Adsorption – Activated Carbon	Contaminant removal based on solid support	Excellent removal of wide varieties of dyes	Very expensive	[11]
Membrane filtration	Separation of particles	Effective removal for all dyes	Production of high concentrated sludge, Expensive	[12]
Sonolysis	Ultrasound technique	No sludge production	Dissolved Oxygen requirement at high rate.	[13]
Electrocoagulation	Anode and cathode treatment process	Effective removal of dyes	High cost	[14]
Chemical Coagulation	Addition of coagulants	Good Colour removal, Economic	Sludge production	[11] [15]

Currently, biosorption process is one of the effluent methods of removing pollutants from wastewater and provides an attractive alternative treatment, since it is inexpensive and readily available. Consequently, many studies have been made on the possibility of biosorbents using Apple Pomance [16], Sugarcane bagasse [17], Neem leaf powder [18], Bannana Pith [19], Wheat Shell [5], Palm tree flower [20], Indian rose wood sawdust [21], Rice husk [22], Peanut shell [23], Coir pith [24], Sunflower seed husk [25], Vintex negundo stem [26], orange peal [27], Eucalyptus bark [28] and Wheat bran [29] for the removal of different dyes from aqueous solutions at different operating conditions.

The present investigation deals with a series of experiments to assess the suitability of Water Hyacinth (WH) as a biosorbent in removal in removal of Methylene Blue (MB). Methylene Blue (a basic and cationic dye) is the most commonly used substance for dying cotton, wood and silk. MB is not regarded as acutely toxic, but it can cause various harmful effects such as nausea, vomiting, diarrhea, gastritis, chest pain, severe headache and methenoglobinemia like syndrome [30-32].MB was selected in order to evaluate the capacity of WH for the removal of dye from aqueous solution.

MATERIALS AND METHODS

Biosorbent

The Water hyacinth was collected from the local Vallankulam lake, Coimbatore. The parts of WH (Petiole, Leaves and Roots) were separated and washed thoroughly with tap water several times in order to remove soil particles. It is chopped into small pieces to ease of drying. The WH was dried in the oven at 60^0C for 48 hours until the moisture content remained same. The biosorbent is then blended by using dry blender and sieved for 5-10 minutes. The biosorbent of size 45µm [19] was stored in a bottle for further use.

Adsorbate

The basic dye, Methylene Blue (CI name : basic blue 9, Class: Thiazine, CI No.: 52015, Chemical formula : $C_{16}H_{18}N_3SCl$, Molecular weight : 319.9 g/mol and λ_{max} : 665 nm) was selected because of its known strong adsorption onto solids. A stock solution of

500mg/l was prepared and suitably diluted to the required concentrations.

Experimental Methods

The equilibrium experiments were carried out by batch process at 150 rpm by adding 0.4 g of WH powder to 100 ml dye solution. The dye concentration was varied from 100-300 mg/l for three different temperatures (30°C, 40°C and 50°C). The percentage removal of dye and amount of MB adsorbed by WH was obtained from the following expressions,

$$\% \, Removal = \frac{(C_o - C_e)}{C_o} X \, 100 \quad (1)$$

$$q_e = \frac{(C_o - C_e)V}{W} \quad (2)$$

Where C_o(mg/l) is the initial MB concentration, C_e(mg/l) is the equilibrium MB concentration, q_e(mg/g) is the equilibrium amount of dye sorbed by WH, V(L) is the volume of sample and W(g) is the dry weight of the WH powder. The equilibrium data obtained were fitted using six different isotherm models, namely the Langmuir, Freundlich, Temkin and Dubinin – Radushkerich models.

RESULTS AND DISCUSSIONS:

Adsorption isotherms are important for the description of how adsorbates will interact with an adsorbent and are critical in optimizing the use of adsorbent [33]. Thus, to have an insight into the sorption behavior of Methylene Blue onto Water Hyacinth, the correlation equilibrium data using theoretical equations are essential for interpretation and prediction of sorption data. The analysis of sorption data and fitting them to isotherm models are very important to select a suitable model that can be used for designing a system [34-35]. Adsorption isotherm study was carried out on six isotherm models, namely the Langmuir, Freundlich, Temkin and Dubinin – Radushkerich models. The applicability of the isotherm equation to describe the sorption process was judged by the correlation coefficients, R^2 values [36]. The parameters of these isotherm models were presented in Table 2.

The amount of MB dye adsorbed at equilibrium is shown in Table 3. The q_e increased with initial concentration (C_0) and was highest with the highest concentration (C_0 – 300 mg /l) for all the temperatures, while the percentage removal of MB decreases with increase in concentration and the highest removal was at the initial concentration of 100 mg/l [37] for temperatures varying from 30° C to 50°C.

Table 2 : Isotherm Parameters for MB sorption onto WH

Isotherm Model	Isotherm Parameters	Temperature		
		30°C	40°C	50°C
Langmuir	q_{max} (mg/g)	17.58	25.35	46.35
	K_L	0.0134	0.0122	0.0086
	R_L	0.53	0.58	0.71
	R^2	0.981	0.987	0.988
Freundlich	n	2.76	2.23	1.84
	K_F	1.78	1.75	1.71
	R^2	0.9363	0.9358	0.9926
Temkin	B_T (J/mol)	641.54	396.9	250.50
	B_T	3.9267	6.5561	10.72
	A_T	0.124	0.0855	0.0735
	R^2	9.443	0.968	0.9877
Dubinin-Radushkevich	B_D (mol^2 / KJ^2)	0.0086	0.0087	0.0093
	E (KJ/mol)	7.62	7.58	7.33
	Q_D (mg/g)	69.98	36.1	16.68
	R^2	0.9456	0.9717	0.995

Table 3: Amount of MB Adsorbed at Equilibrium for three different temperatures.

C_0 (mg/l)	C_e (mg/l)			% Removal			q_e (mg/g)		
	30°C	40°C	50°C	30°C	40°C	50°C	30°C	40°C	50°C
100	66.7	58	46.9	33.3	42.0	53.1	8.3	10.5	13.3
150	105.75	91.5	78	29.5	39.0	48.0	11.1	14.6	18.0
200	155	131	114	22.5	34.5	43.0	11.3	17.3	21.5
250	199	176.5	146.48	20.4	29.4	41.4	12.8	18.4	25.9
300	244.5	222.6	188.6	18.5	25.8	37.1	13.9	19.4	27.9

Langmuir Isotherm: The Langmuir isotherm is a well known linear model for monolayer adsorption adsorption and most frequently used to determine the adsorption parameters. The Langmuir isotherm can be written in the form

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_L \, q_m} \quad \text{-------------- (1)}$$

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad \text{---------------- (2)}$$

Where K_L is the equilibrium constant or Langmuir constant related to the affinity of binding sites in (L/mg) and q_m represents the adsorption capacity when the surface is completely covered with dye molecules. K_L and q_m were calculated from the slope and intercept of the linear plot of C_e/q_e Vs C_e .From table 2 ot shows that the Langmuir equation represents a good adsorption process, the R^2 values were higher than 0.981, which indicate a very good mathematical fit. The reason is that the surface is homogeneous [38]. The maximum adsorption capacities for MB on to water hyacinth at 30°C, 40°C and 50°C were found to br 17.58 mg/l, 25.35 mg/l and 46.35 mg/l respectively. Maximum adsorption capacities of water hyacinth increased with increase in temperature.

The dimensionless constant separation factor (R_L) is an important feature of Langmuir isotherm, which is deified by the following relationship,

$$\mathbf{R_L = 1 / (1+K_L \, C_o)}$$

Based on the values of R_L, the isotherm type indicates to be unfavourable (R_L >1), Linear (R_L=1), favourable (1>R_L >0) or irreversible (R_L =0) [39].The results presented in Table 3 shows that the adsorption of methylene blue onto water hyacinth is favourable.

Table 4. Removal of Methylene Blue using different adsorbents

Sl.No.	Adsorbent	q_{max} (mg/g)	Reference
1	Vintex Negundo stem	183.88-207.88	26
2	Sunflower seed husk	4.757 – 23.196	25
3	Indian rosewood saw dust	11.8 – 51.4	22
4	Rice husk	40.58	7
5	Neem leaf powder	8.76-19.61	18
6	Wheat shell	16.56-21.50	38
7	Coir pith	5.87	24
8	WH Root powder	8.04	40
9	Water Hyacinth	17.58 – 46.35	This Study

The adsorption capacities of water hyacinth for methylene blue varied in the rage of 17.58 mg/g to 46.35 mg/g. Table 4 shows the previously reported adsorption capacities of some adsorbent for methylene blue. By comparing with pervious works it can be stated that our findings holds good.

Freundlich Isotherm : The Freundlich isotherm [20] is an empirical equation used to describe heterogeneous systems. It states that the adsorption process takes place on heterogeneous surface and the adsorption capacity is related to the concentration of methylene blue dye at equilibrium is given by

$$q_e = k_F\, C_e^{1/n} \qquad (3)$$

The linear form of Freundlich isotherm is expressed by

$$\log q_e = \ln K_F + \frac{1}{n}\ln C_e \qquad (4)$$

Where q_e is the amount adsorbed (mg/g) at equilibrium, C_e is the final concentration of dye solution (mgl), K_F and n are Freundlich constants. The plot against ln q_e Vs ln C_e yields a straight line with slope and intercepts values by which n and K_F can be calculated.

Freundlich constant (K_F) gives the adsorption capacity of an adsorbent and is a constant which shows the strength of the relationship between adsorbent and adsorbate. The value of K_F of water hyacinth for methylene blue at 30°C, 40°C and 50°C are on the order of 1.78, 1.75 and 1.71 respectively. The values of n in the rage of 1 to 10 represent good adsorption. In the present study, the exponent (n) was 1 < n < 10, indicating favorable adsorption. The R^2 value varies between (0.936 to 0.9926) which indicates a good correlation between the parameters.

Temkin and Pyzhev Isotherm : Temkin equation represents the heat of adsorption and adsorbent – adsorbate interaction on the surface [41]. The Temkin isotherm is given by,

$$q_e = \left(\frac{RT}{b_T}\right)\ln(A_T C_e) \qquad (5)$$

Or

$$q_e = B_T \ln(A_T\, C_e) \qquad (6)$$

Where B_T = RT / b_T, b_T is the adsorption potential of the adsorbent ($Jmol^{-1}$), A_T is the Temkin isotherm constant (L/mg), R is the molar constant (8.314 J/mol-K), T is the temperature in ^{0}K. A linear for of equation is given by,

$$q_e = B_T \ln A_T + B_T \ln C_e \qquad (7)$$

The constants A_T and B_T can be determined from the plot q_e vs ln C_e. The values of Tempkin constants and correlation coefficient R^2 are presented in Table 2.

Fig.(7) to Fig. (9) shows the Temkin isotherm model for the methylene blue dye adsorption on to the adsorbent (water hyacinth) from which the relevant isotherm parameters are listed in Table 2. It is observed that the value of R^2 at 30^{0}C, 40^{0}C and 50^{0}C were 0.9443, 0.968 and 0.9877 which gives a close fit to the methylene blue adsorption on water hyacinth, but the values are slightly lower than the Langmuir and Freundlich isotherms [34].Further, it is observed from Table 2 that the adsorption heat of Methylene Blur adsorption on water hyacinth at 30^{0}C, 40^{0}C and 50^{0}C were 641.54 J/mol, 396.9 J/mol and 250.50 J/mol respectively.

Dubinin – Radushkerich (DR) Isotherm : DR isotherm is generally applied to express the adsorption mechanism with a Gaussian energy distribution onto a heterogeneous surface [42,43]. The utility of the equation lies in the fact that the temperature dependence is reflected in the adsorption potential [44,45]. The DR isotherm is given by,

$$ln q_e = \ln q_D - 2B_D\, RT \ln\left(1 + \frac{1}{C_e}\right) \qquad (8)$$

Where, B_D is a constant related to the mean free energy (E) of adsorption per mole of adsorbate as it migrates to the surface of the adsorbent from infinite distance.

$$E = \sqrt{\frac{1}{2\,B_D}} \qquad (9)$$

q_D is the Dubinin – Radushkerich isotherm constant related to the degree of adsorbate adsorption by the adsorbent surface or the theoretical saturation capacity. It was applied to estimate the porosity, apparent free energy and the characteristics of adsorption. A linear plot between $RTl_n(1+1/C_e)$ Vs l_n q_e was developed in order to find the isotherm parameters. The DR isotherm parameters were presented in Table 2. The linear regression coefficient (R^2) values are in the rage of 0.9456 to 0.995, which represents that the experimental data fitted well with DR isotherm model. It was stated that if the value of E is below 8 KJ/mol, the adsorption process can be considered as physical adsorption and if the value range between 8-16 KJ/mol, it is chemical adsorption. From, Table 2 it can be observed that the value of E ranges between 7.33 to 7.62 KJ/mol. It clearly shows that the physical adsorption plays a vital role in the adsorption process of methylene blue dye adsorption onto the adsorbent (water hyacinth) [34].

Graphical Representation of Isotherm:

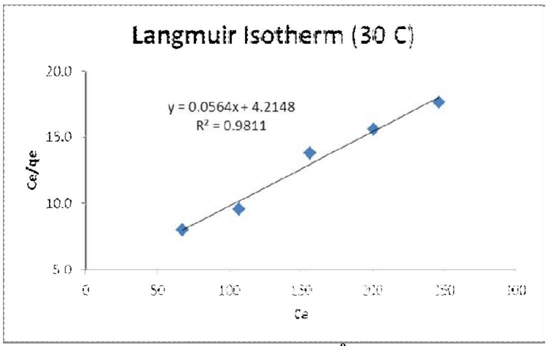

Fig.(1) Langmuir Isothem at 30^0 C

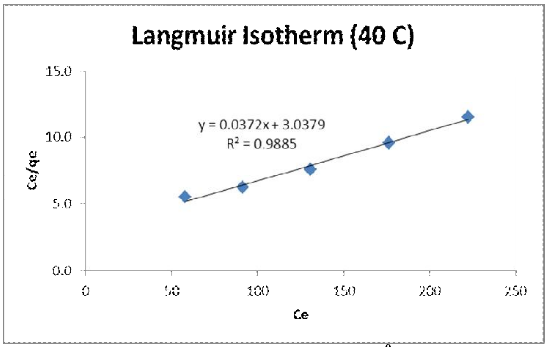

Fig.(2) Langmuir Isothem at 40^0 C

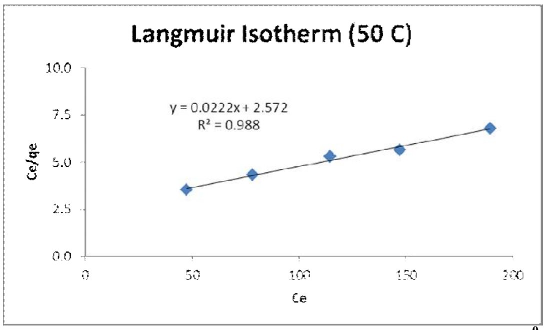

Fig.(3) Langmuir Isothem at 50^0 C

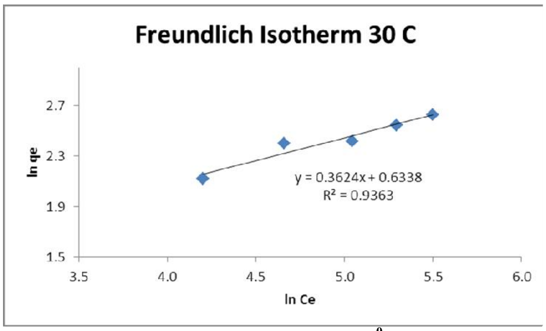

Fig.(4) Freundlich Isothem at 30^0 C

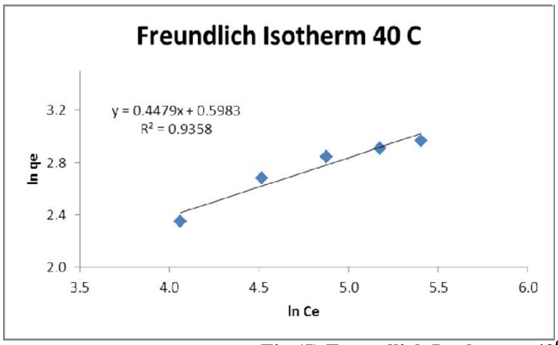

Fig.(5) Freundlich Isothem at 40^0 C

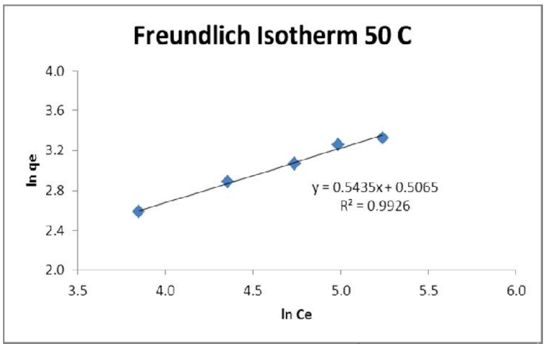

Fig.(6) Freundlich Isothem at 50^0 C

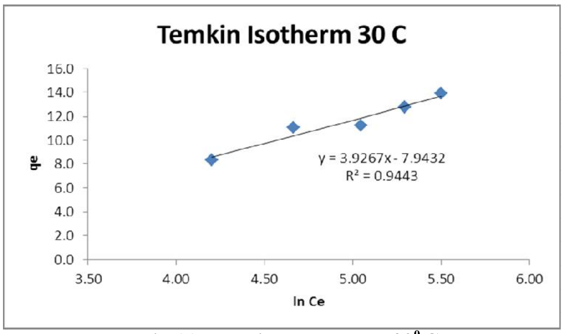

Fig.(7) Temkin Isotherm at 30^0 C

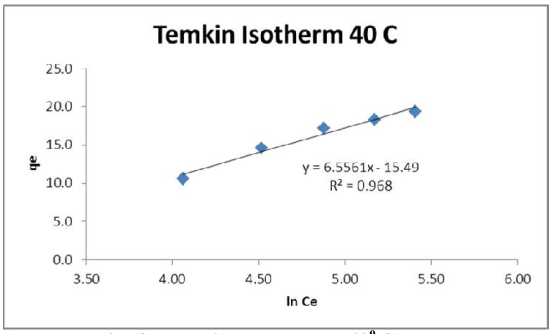

Fig.(8) Temkin Isotherm at 40^0 C

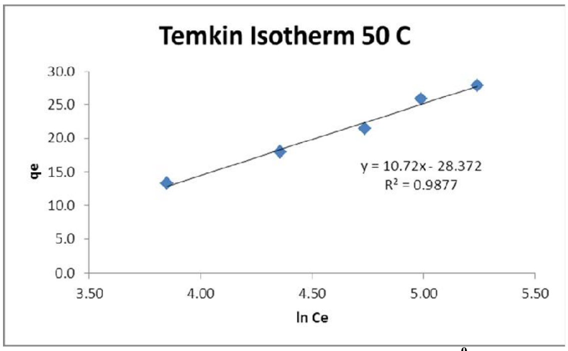

Fig.(9) Temkin Isotherm at 50^0 C

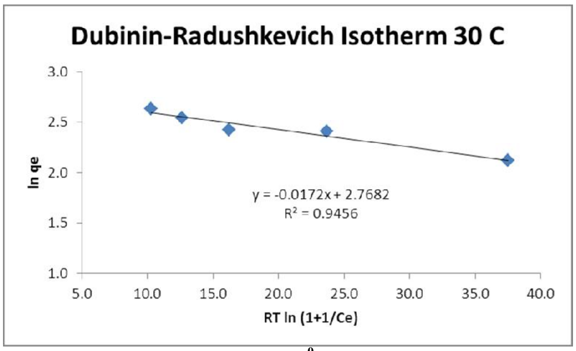

Fig. (10) D-R Isotherm 30^0C

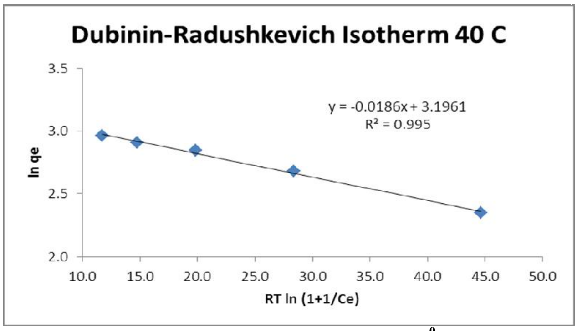

Fig. (11) D-R Isotherm 40^{0}C

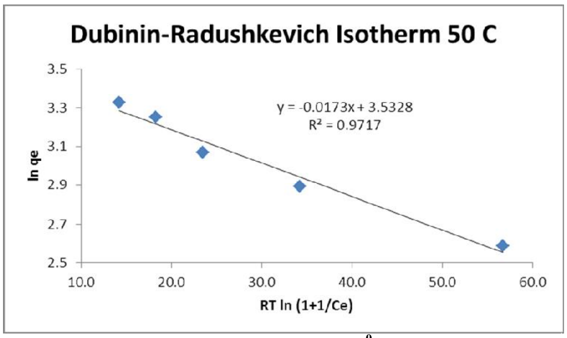

Fig. (12) D-R Isotherm 50^{0}C

CONCLUSION

Adsorption of methylene blue onto water hyacinth has been modeled using Langmuir, Freundlich, Temkin and Dubinin – Radushkerich isotherm equations. The experimental results shows a best-fit with the tested isotherm models and D-R model suggests that all the dye is removed from aqueous medium by adsorption process.

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