



Equilibrium, Thermodynamics and Kinetic studies of Malachite green by activated *Glossocardia linearifolia* Stem

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ABSTRACT

The present work deals with the adsorption of Malachite Green (MG) dyes carried out in the presence of activated *Glossocardia linearifolia* stem carbon. Various parameters like, the effect of initial concentration, contact time, dose of adsorbent, temperature and pH were also studied. The result shows that when the amount of adsorbent increases, the percentage removal of dye increases. The applicability of Freundlich adsorption and Langmuir adsorption isotherm had also been tested. Adsorption kinetic data have been tested using pseudo second order, intra-particles models and the Elovich model. The thermodynamics parameter such as ΔG^0 , ΔH^0 and ΔS^0 were calculated. The adsorption capacities of Activated *Glossocardia linearifolia* Stem (AGLS) carbon were calculated using batch process.

KEYWORDS : Adsorption, Kinetics, Thermodynamics, Malachite Green, Activated *Glossocardia linearifolia* Stem

1. Introduction

Dyes are commonly used in many industries, like textile, food, paper, plastic, cosmetics and coloring industries. These industries commonly use synthetic dyestuff as a colorant. Discharge of dye containing waste water into the environment, contaminates surface water and ground water. However 7×10^5 tones of dye stuff are produced annually [1,2]. The dyes in the waste water even at very low concentration affect the aquatic life and human health by polluting the environment. The toxicity and carcinogenicity have lead to exploration of possible detoxicants [3, 4]. Dyes can causes allergic, dermatitis, skin irritation, cancer, mutation, etc. In general dyes are poorly biodegradable and some of the dyes produce aromatic amine which is highly carcinogenic in nature [5, 6]. Many investigators have studied the feasibility of using inexpensive alternative materials like peral millet husk, date pits, saw dust, buffing dust of leather industries, coir pith, crude oil, residue, tropical grass, olive stone almond shell, pine bark, wool waste, coconut shell etc [7, 8]. The present study seeks to evaluate the efficiency of a activated *Glossocardia linearifolia* Stem adsorbent in the removal of malachite green dyes from dye solution.

2. Experimental methods**2.1 Adsorbent**

Glossocardia linearifolia stem was collected from agricultural area. The stem was washed with distilled water to remove the impurities and dried for four hours at 125°C in a hot air oven. The dried material was ground well to a fine powder and then activated around 600°C then sieved and stored in plastic bags and kept ready for use.

**Glossocardia Linearifolia****2.2 Adsorbate**

The stock solution of malachite green concentration 1000 mg/L was prepared by dissolving 1 g of malachite green in 1000 ml of double

distilled water. Different concentration of dyes solution range from (50 to 250 mg/L) were prepared from the stock solution by appropriate dilution

2.3 Batch adsorption experiments

Batch adsorption was tested by adding 25 mg of AGLS to 50 ml of the dye solution of different initial concentration (50 to 250 mg/L) at a particular pH. The experiment was carried out using a wrist action shaker for the period of 180 min and 120 rpm using 250 ml stopper glass flasks at 30°C to 60°C . The residual concentrations of dyes in each sample after adsorption at different time intervals were determined by UV-Visible spectrophotometer. The equilibrium q_e (mg/g) was calculated by the following mass balance principle.

$$q_e = \frac{(C_0 - C_e)V}{M} \dots\dots\dots (1)$$

Where C_0 and C_e are the initial and equilibrium concentrations (mg/L) of dyes respectively, V is the volume (L), M is the weight (g) of the adsorbent. The removal efficiency of the adsorbents on dyes was calculated by using the following expression.

$$R\% = \frac{C_0 - C_t}{C_0} \times 100 \dots\dots\dots (2)$$

2.4 Kinetic Experiments

The batch kinetic [9] experiments were basically similar to those used in the adsorption equilibrium method. The dyes samples were taken at specific time intervals and the concentration of dyes was measured similarly. The all kinetic experiments were carried out at 30, 40, 50 and 60°C with initial dye concentration of 50, 100, 150 200 and 250 mg/ L The amount of adsorption at time t , q_t (mg/g) was calculated by.

$$q_t = \frac{(C_0 - C_t)V}{M} \dots\dots\dots (3)$$

Where C_t (mg/L) is the liquid phase concentration of dye at any time.

3. Results and discussion**3.1 Effect of contact time and initial dye concentration**

The experimental results of adsorption of various dye concentrations with contact time are shown in **Fig.1**. This figure shows that the % removal initially increases and reaches the limiting value. So that the equilibrium was established at 50 minutes. Hence all the remaining experiments were carried out at 50 minutes. The equilibrium data given in **Table.1**, reveals that, the percentage

removal decreases with increase in initial dyes concentration, due to the number of available active sites which remains constant but the initial concentration of dyes increases, so that % removal decreases [10, 11].

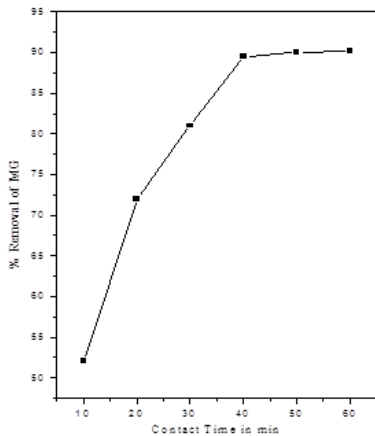


Fig.1. Effect of Contact time on the removal of MG dye onto AGLS [MG]= 50mg/L; Temp 30°C; Adsorbent dose= 25mg/50ml

Table.1. Equilibrium parameters for the adsorption of MG dye onto AGLS

3.2 Effect of adsorbent dosages

The effect of the AGLS doses was studied at 60°C by varying the amount of adsorbent dose 50-250 mg for the initial concentration of 50 mg/L Fig. 2 reveals that increase in percentage removal of MG dye with increase in dose of adsorbent. This is due to the increase in adsorbent surface area and the availability of more adsorption sites.

C0 (mg/L)	Temperature (C)			
	30	40	50	60
50	0.1803	0.1753	0.3067	0.1593
100	0.0991	0.0960	0.1811	0.0866
150	0.0683	0.0661	0.1285	0.0594
200	0.0521	0.0504	0.0995	0.0452
250	0.0421	0.0407	0.0812	0.0365

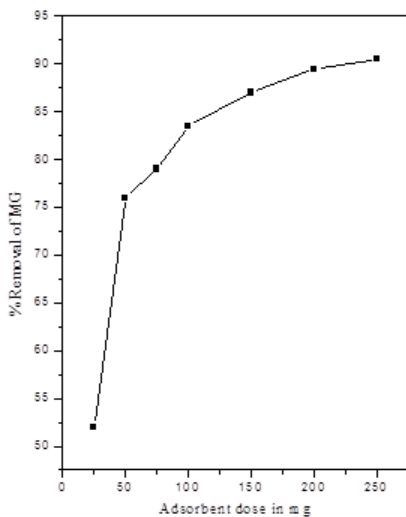


Fig.2- Effect of Adsorbent dose on the removal of MG dye onto ACP [MG]=50mg g/L; Contact time=50 m in; Temp p 30°C;

3.3 Effect of pH

The solution pH is one of the most important factors that control the adsorption of MG dye. To examine the effect of pH on the % removal of MG dye the pH of initial solution were varied from 2.0 to

10.0 by adding NaOH or HCl in to them. The % removal increases as the pH increases up to 6.5. there after the % removal decreases. At pH 6.5 the optimum % removal takes place. So the remaining experiments were carried out at pH 6.5. The experimental result was shown in Fig. 3.

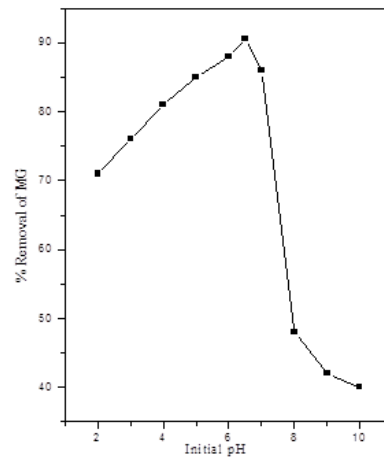


Fig.3- Effect of Initial pH on the removal of MG dye onto ACP [MG]=50mg g/L; Contact time=50 m in; Temp p 30°C; Adsorbent dose=25mg g

3.4 Adsorption isotherm studies

To quantify the sorption capacity of the adsorbent for the removal of dyes, the most commonly used isotherms, are the Freundlich and Langmuir isotherms and hence these were used in this study.

3.4.1 Freundlich isotherm

Linear form of Freundlich isotherm model [12] is represented by the equation

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \dots\dots\dots(4)$$

Where q_e is the amount of dyes adsorbed per unit weight of the adsorbent (mg/L) K_f is (mg/g(L/mg)) the measure of adsorption capacity and $1/n$ is the adsorption intensity. The value of K_f and n are calculated from the intercept and slope of the plot of $\log q_e$ Vs $\log C_e$ respectively. The constant values of K_f and n are given in Table.2. In general the K_f value increases with increase in adsorbate dose. The magnitude of the exponent $1/n$ gives an indication of the favorability of adsorption. The value of $n > 1$ represents favorable adsorption condition [7] (or) the value of n are in the range of 1 to 10 confirms the favorable condition for adsorption. The adsorption coefficient K_f of dyes on activated AGLS was found to be between from 5.8107 - 6.1062 L/g. The K_f value indicates that the saturation time for adsorption of dyes is attained quickly due to the high affinity of activated Glossocardia linearifolia stem towards the adsorbate. The values of n were in the range of 1.8611 to 2.9265 (mg/L) for MG dyes adsorption. So Freundlich isotherm is suitable for this adsorption. The K_f values also indicate the feasibility of multilayer adsorption were possible. This reveals that the activated Glossocardia linearifolia stem was more efficient for the removal of MG dye.

3.4.2 Langmuir isotherm

The Langmuir isotherm model [13] is based on the assumption that maximum adsorption corresponds to a saturated monolayer of solute molecules on the adsorbent surface. The linear form of the Langmuir isotherm equation can be described by

$$C_e/q_e = (1/Q_m \cdot b) + (C_e/Q_m) \dots\dots(5)$$

Where C_e (mg/L) is the equilibrium concentration of the dye, q_e (mg/g) is the amount of dye per unit weight of adsorbent, Q_m and b are Langmuir constants related to adsorption capacity and rate of adsorption respectively. Q_m is the amount of dye at complete monolayer coverage (mg/g) which gives the maximum adsorption

capacity of the adsorbent and b (L/mg) is the Langmuir isotherm constant that relates to the energy of adsorption or rate of adsorption. The linear plot of C_e/q_e against the equilibrium concentration C_e shows that the Langmuir model is fitted. The Langmuir constant Q_m and b were determined from the slope and intercept of the Langmuir plot and these values are given in **Table.2**. The feasibility of the Langmuir isotherm can also be expressed in terms of the dimensionless constant separation factor R_L [14, 15] by the equation.

Table.2. Langmuir and Freundlich isotherm parameters for adsorption of MG dye onto AGLS

MG (mg / L)	Ce (mg / L)				Qe (mg / g)				% Removal of MG dye			
	30°C	40°C	50°C	60°C	30°C	40°C	50°C	60°C	30°C	40°C	50°C	60°C
50	4.97 20	4.58 24	4.24 96	4.04 32	90.0 56	90.8 3	91.5 008	91.91 36	90.0 56	90.8 3	91.5 00	91.9 13
100	17.2 69	15.6 20	13.3 97	11.8 04	165. 46	168. 7	173. 204	176.3 90	82.7 30	84.3 8	86.6 02	88.1 95
150	37.4 79	33.8 03	30.2 72	27.1 55	225. 04	232. 3	239. 454	245.6 89	75.0 13	77.4 6	79.8 18	81.8 96
200	73.2 36	68.5 76	30.2 72	58.8 19	253. 52	262. 8	339. 454	282.3 61	63.3 81	65.7 1	84.8 63	70.5 90
250	119. 95	114. 05	63.6 79	102. 74	260. 08	271. 8	372. 641	294.5 04	52.0 17	54.3 7	74.5 28	58.9 00

$$R_L = \frac{1}{1 + bC_0} \dots\dots\dots(6)$$

Where C_0 (mg/L) is the initial concentration of adsorbent and b (L/mg) is Langmuir isotherm constant. The parameter R_L indicates the nature of the isotherm.

- $R_L > 1$ unfavorable
- $R_L = 1$ Linear
- $0 < R_L < 1$ Favorable
- $R_L = 0$ Irreversible

The R_L values lies between 0 to 1 indicate the favorable adsorption for all initial concentration. The calculated R_L values were given in **Table.3**. The calculated R_L values were with in the range of 0.0365 to 0.1811. So the adsorption of MG follows the Langmuir isotherm.

Table.3. Dimensionless separation factor (R_L) for adsorption of MG dye onto AGLS

Temperature (°C)	Langmuir parameter		Freundlich parameter	
	Q_m	b	K_f	n
30	286.1309	0.0909	5.8107	2.9265
40	299.1008	0.0941	5.8979	2.8731
50	494.764	0.0452	5.1320	1.8611
60	323.577	0.1055	6.1062	2.7871

3.5 Thermodynamic study

Thermodynamic parameter such as change in free energy (ΔG°) (KJ/mol), Enthalpy (ΔH°) (KJ/mol) and entropy (ΔS°) (JK/mol) were calculated by $K_0 = C_{solid} / C_{liquid} \dots\dots\dots(7)$

$$\Delta G^\circ = -RT \ln K_0 \dots\dots\dots(8)$$

$$\log K_0 = \frac{\Delta S^\circ}{2.303R} - \frac{\Delta H^\circ}{2.303RT} \dots\dots\dots(9)$$

Where K_0 is the equilibrium constant, C_{solid} is the solid phase concentration at equilibrium (mg/L). C_{liquid} is the liquid phase concentration at equilibrium (mg/L). T is temperature in Kelvin and R is the gas constant (8.314 J mol⁻¹K⁻¹). A graph was drawn between log K_0 vs 1/T. The ΔH° and ΔS° values obtained from the slope and intercept of Van't Hoff plots. These values were given in **Table.4**. The negative ΔG° values indicate that the spontaneous nature of

adsorption and also the magnitude of ΔG° indicate the adsorption is physical adsorption (ie, less than 70 KJ/mol) The value of ΔH° is positive, this indicates the endothermic nature of adsorption. The positive ΔS° indicates the increased randomness during the adsorption. This also supports the possibility of physical adsorption [16, 17]

Table: 4. Thermodynamic parameter for the adsorption of MG onto AGLS

CO (mg/L)	ΔG°				ΔH°	ΔS°
	30°C	40°C	50°C	60°C		
50	-5550.835	-5968.794	-6381.572	-6729.448	6.4266	39.57 32
100	-3946.587	-4389.473	-5011.674	-5567.706	12.668	54.70 5 89
150	-2769.424	-3213.115	-3692.364	-4178.738	11.488	47.01 2 77
200	-1382.059	-1692.702	-4629.514	-2424.078	17.709	63.65 8 56
250	-203.415	-456.772	-2883.101	-996.3114	14.963	50.62 9 70

3.6 Adsorption kinetics

The kinetics studies were done by using pseudo second order [18] Elovich [19, 20] and intra-particle diffusion [21] models.

3.6.1 The pseudo-second-order kinetic model

The linear form of pseudo second order equation is expressed as

$$\frac{dq_t}{dt} = K_2(q_e - q_t)^2 \dots\dots\dots(10)$$

where k_2 (g/mg min) is the pseudo second order rate constant .For the boundary conditions $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_t$, integrated form of Eq. (10) becomes:

$$(q_e - q_t) = \frac{1}{q_e} + K_2 t \dots\dots\dots(11)$$

This is the integrated rate law for a pseudo second order reaction. Equation (11) can be rearranged to obtain Eqs (12) which has a linear form:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \dots\dots\dots(12)$$

If the initial adsorption rate (h)(g/mg min) is

$$h = K_2 q_e^2 \dots\dots\dots(13)$$

Then Eqs. (12) and (13) becomes:

$$\frac{1}{q_t} = \frac{1}{h} + \frac{1}{q_e} t \dots\dots\dots(14)$$

Where k_2 (g/mg min) is the pseudo second order rate constant, q_e is the amount of dye adsorbed per unit mass of adsorbent (mg/g) at equilibrium, q_t is the amount of dye adsorbed at time "t". A graph is drawn between t/q_t versus "t", the q_e and k_2 can be calculated from the slope and intercepts of the graph. The correlation coefficient value (r) were also calculated. These values are given in **Table.5**. The correlation coefficient value (r) for the pseudo second order was greater than 0.9900 (ie, > 0.9900) and also q_e value calculated from the model was almost equal to the experimental value. So the adsorption of MG follows pseudo second order model.

Table.5. The kinetic Parameters for the adsorption of MG on to AGLS

Co	Temp °C	pseudo second order				Elovich model			Intra-Particle diffusion		
		q _e	K ₂	γ	h	α	β	γ	Kid	γ	Intercept
50	30	100.080	0.00128	0.9919	12.8452	110.5234	0.0691	0.9918	1.6225	0.9942	0.1832
	40	100.2021	0.00135	0.9921	13.5907	146.8037	0.0724	0.9920	1.6443	0.9958	0.1724
	50	100.2511	0.001443	0.9932	14.5075	198.9961	0.0758	0.992	1.6650	0.9960	0.1625
	60	100.8469	0.00142	0.9936	14.4698	187.4683	0.0745	0.9927	1.6635	0.9975	0.1646
100	30	184.2176	0.000672	0.9941	22.7999	177.5959	0.0367	0.9926	1.5748	0.9976	0.1887
	40	187.3143	0.00676	0.9926	23.7098	209.8361	0.0371	0.9929	1.5943	0.9979	0.1821
	50	190.9157	0.000718	0.9929	26.1605	280.9139	0.0379	0.9935	1.6238	0.9940	0.1725
	60	194.6650	0.00070	0.9911	26.8841	279.7291	0.0370	0.9939	1.6310	0.9950	0.1737
150	30	254.8363	0.000421	0.9909	27.3184	155.4677	0.0246	0.9941	1.4945	0.9960	0.2095
	40	260.6738	0.00044	0.9910	30.2472	201.5094	0.0250	0.9948	1.5291	0.9961	0.1983
	50	267.7725	0.000462	0.9913	33.1244	249.6425	0.0251	0.9950	1.5585	0.9920	0.1903
	60	272.2697	0.00038	0.9922	28.1846	307.9750	0.0268	0.9991	1.5751	0.9913	0.1750
200	30	297.9219	0.00027	0.9927	24.5051	85.6862	0.0183	0.9994	1.3339	0.9931	0.2583
	40	305.3655	0.00028	0.9937	26.8841	105.9773	0.0186	0.9960	1.3751	0.9915	0.2436
	50	314.0891	0.00029	0.9950	29.4047	127.8459	0.0186	0.9985	1.4105	0.9920	0.2329
	60	321.6071	0.00032	0.9959	33.6923	171.4670	0.0190	0.9986	1.4557	0.9936	0.2176
250	30	322.7917	0.00019	0.9957	19.5742	49.6622	0.0150	0.9980	1.1362	0.9938	0.3205
	40	333.8572	0.00019	0.9980	21.1575	55.8777	0.0148	0.9983	1.1731	0.9958	0.3101
	50	348.8991	0.000179	0.9982	21.8066	57.7381	0.0148	0.9927	1.1899	0.9988	0.3105
	60	353.7305	0.00020	0.9985	25.2662	73.5208	0.0145	0.9929	1.2486	0.9986	0.2878

3.6.2 The Elovich equation

The Elovich model equation is generally expressed as

$$\frac{dq_t}{dt} = \alpha \exp(-\beta q_t) \dots\dots\dots(15)$$

where α is the initial adsorption rate (mg g⁻¹ min⁻¹) and β is the desorption constant (g mg⁻¹) during any one experiment. To simplify the Elovich equation. Chien and Clayton assumed t>>t and by applying boundary conditions q_t = 0 at t= 0 and q_t = q_t at t = t Eq.(15) becomes:

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t \dots\dots\dots(16)$$

If dyes adsorption fits the Elovich model, a plot of q_t vs. ln (t) yields a linear relationship with a slope of (1/β) and an intercept of (1/β) ln (αβ). The Elovich parameters α, β and correlation coefficient (γ) were given in **Table.5**. The initial adsorption rate (α) and desorption constant (β) increases with increase in initial concentration of MG dyes and also the correlation coefficient (γ) were greater than 0.9900 (ie, γ > 0.9900). The Elovich model can also suitable for the MG adsorption onto AGLS.

3.6.3 Intra-particle diffusion model

According to Weber and Morris suggested the following equation for intra-particle diffusion model

$$q_t = K_{id} t^{1/2} + C \dots\dots\dots(17)$$

Where k_{id} is the intra-particle diffusion constant (mg/g min), and q_t is the amount of the dye adsorbed at time “t”. According to Weber and Morris model, a graph is drawn between q_t and t^{1/2}, the line was passing through the origin. But here the intercept value indicates the lines were not passing through origin. This was due to that the intra-particle diffusion takes place along with some other process. This may be boundary layer adsorption or instantaneous adsorption.

3.7. Effect of the ionic strength on the adsorption of Malachite Green

The effect of sodium chloride on the adsorption of malachite green on to AGLS is shown in **Fig.4**. In a low solution concentration of NaCl had less influence on the adsorption capacity. The partial neutralization of the positive charge on the adsorbent surface and a consequent compression of the electrical double layer by the Cl⁻

anion cause the increase in the adsorption of the malachite green at higher ionic strength. The chloride ions can also enhances adsorption of malachite green ion onto activated AGLS by pairing of their charges and hence reducing the repulsion between the malachite green molecules adsorbed on the surface and there by the AGLS to absorb more of positive malachite green dye [16, 17, 22, 23]

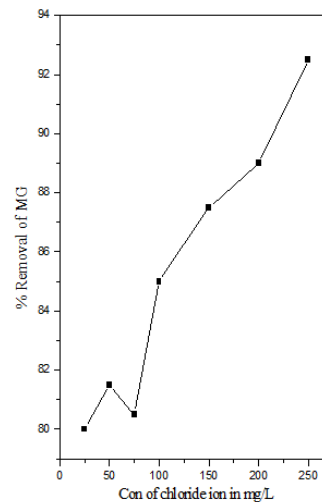


Fig-4- Effect of Other ions on the removal of MG onto ACP [MG]=50mg/L, Contact time=50 min; Temp 30°C, Adsorbent dose=25mg/50ml

Conclusion

The adsorption characteristics of MG dye onto activated *Glossocardia linearifolia* stem are strongly affected by the initial dye concentration, initial pH and the adsorbent dose. The pH 6.5 was favorable for the optimum adsorption of MG dye by AGLS. The R_L values and other adsorption parameters indicate both Langmuir and Freundlich isotherms are favorable for AGLS adsorption. The pseudo second order, Elovich and intra-particle kinetic model were found to applicable for the adsorption of MG onto AGLS reaction model. The thermodynamics parameters ΔG°, ΔH° and ΔS° values indicate that the adsorption is endothermic and physical adsorption.

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