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Biosorption of Malachite Green from Aqueous Solution Using Alocasia Macrorrhizos Leaf Powder

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ABSTRACT: In the present study biosorption capacity of leaves of Alocasia Macrorrhizos was explored for the Biosorption of cationic dye Malachite green (MG) from water. The factors affecting the rate processes involved in the uptake of the dye for adsorbent was contact time(min.), initial dye concentration(mg/L), dosage(gm), pH and temperature(K) were studied Equilibrium data were analyzed using Langmuir, Freundlich and Temkin isotherm models.. The Langmuir isotherm model showed good fit to the equilibrium adsorption data and the maximum adsorption capacity obtained was 100 mg/g at 303 K. Kinetic data were studied using pseudo-first and pseudo-second order kinetic models. The kinetic studies shows that the Biosorption of malachite green dye on to Alocasia Macrorrhizos leaf powder follows pseudo- second order kinetics and the mechanism of adsorption was described by intra-particle diffusion model. Various thermodynamic parameters such as change in enthalpy, entropy and Gibb's free energy were also determined. It was found that the biosorption is endothermic in nature. Present investigation suggests that Alocasia Macrorrhizos may be utilized as a low cost adsorbent for removal of malachite green from aqueous solution.

KEY WORDS: MG, Kinetics, Isotherm, biosorption, initial concentration, pH.

I.INTRODUCTION

Several processes have been applied for the treatment of dyes from wastewater such as chemical, biological and physical process. However, the commercial activated carbon is quite expensive and has limited their application. Due to economic reasons, the discovery towards alternative adsorbents to replace the costly activated carbon is highly recommended. The present investigation is an attempt to explore the possibility of using Alocasia macrorrhizos leaf powder to remove malachite green in aqueous solution, since the raw material is harmless, cheaper, and plentiful. To study the suitability of the available biosorption kinetic models for the removal of Malachite Green dye using Alocasia Macrorrhizos leaf powder as biosorbent. To study the biosorption capacity and intensity using isotherms for the removal of Malachite Green dye using Alocasia Macrorrhizos leaf powder as biosorbent. Kinetics of the adsorption with respect to the initial dye concentration and temperature was also investigated. The pseudo-first-order, pseudo-second-order and intra particle diffusion models were used to test the kinetic data. Obtained data were fitted properly in the pseudo-second-order kinetic model. The thermodynamic parameters such as change in free energy (ΔG°), enthalpy (ΔH°) and entropy (ΔS°) were also determined and the results obtained confirmed that the sorption process is feasible, spontaneous, and endothermic[3]. Liquid phase batch operations were useful for the study of effect of contact time, temperature, pH, initial concentration of malachite green[8]. Biosorbent dose, time and pH were important for biosorption in the single factor analysis.

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The biosorption process was found to follow the pseudo second-order kinetic model, indicating that chemical biosorption was the rate-controlling mechanism through sharing or exchange ions between biosorbent and malachite green in the biosorption process[10]. The use of low-cost, abundantly available, highly efficient and eco-friendly adsorbent wood apple shell (WAS) has been reported as an alternative to the current expensive methods of removing of malachite green dye from aqueous solution. The effects of different variables, adsorbent dosage, initial dye concentration, pH, contact time, temperature etc. were investigated and optimal experimental conditions were ascertained. Thermodynamic study concluded the spontaneous and endothermic nature of the adsorption. Present investigation and comparison with other reported adsorbents concluded that, wood apple shell may be applied as a low-cost attractive option for removal of malachite green from aqueous solution[2].

With increasing the initial concentration of malachite green, the pore-diffusion coefficient increased while the film-diffusion coefficient decreased[4]. The positive value of ΔH^0 showed that adsorption of malachite green onto the wood adsorbent was endothermic. The negative values of ΔG^0 at various temperatures indicate the spontaneous nature of the adsorption process[9]. The adsorption kinetics indicated that the adsorption proceeds according to pseudo-second-order model. The adsorption of malachite green was found to be exothermic and it was accompanied by decrease in the entropy. Column studies were performed and the regeneration of the adsorbent was done easily using environmentally benign polyethylene glycol-400[6]. The positive value of ΔH^0 and negative values of ΔG^0 show the sorption process is endothermic and spontaneous. The positive value of ΔS^0 shows the increased randomness at the solid-liquid interface during the biosorption of dyes onto loquat seed[1]. The temperature variation study showed that the malachite green adsorption was endothermic and spontaneous with increased randomness at the solid solution interface[5]. Prawn waste has become one of the most significant wastes in aquaculture, from prawn waste had been utilized as an adsorbent for uptake of cationic dye (i.e., malachite green) from aqueous solution at 28 °C. [7].

The results generated by this work can be used for determination of optimum conditions for adsorption of mixture of dyes in aqueous solutions. Dyes are present in mixture form in various Industrial effluents like Textile Industries, Sewage water, Water treatment plants. This work can have use in Design of adsorption columns for dyes removal.

II. MATERIALS AND METHODS (EXPERIMENTAL PROCEDURE)

Preparation of Biosorbent

Alocasia Macrorrhizos leaves collected were washed twice with distilled water to remove dirt and completely dried in sunlight for 20 days and then in oven to remove the excess moisture content. The dried leaves were then cut into small pieces and powdered. The powder was screened to 150 μ m size and directly used as biosorbent without any pretreatment.

Preparation of Biosorbate:

The Malachite Green stock solution of 1000 mg/L was prepared by dissolving 1 g of Malachite Green in 1000 ml of distilled water which was later diluted to required concentrations. All the samples were prepared using distilled water. Concentrations of the dye solutions were determined from the absorbance of the solution at the characteristic wavelength of dye using double beam UV-Visible spectrophotometer.

Experimental Parameters:

Batch equilibrium experiments were carried out by varying contact time, pH, initial dye concentration, biosorbent dosage, and temperature. In each experiment, accurately weighed *Alocasia Macrorrhizos* leaf powder was added to 10 ml of dye solution in a 250 ml conical flask and the mixture was agitated at 230 rpm in an orbital shaker. Samples were withdrawn at regular time intervals and centrifuged. The dye concentration in the supernatant was determined using double beam UV-Visible spectrophotometer.

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III.RESULTS AND DISCUSSION

Experimental data is generated in a batch mode of operation to study the effect of contact time, pH of the solution, initial concentration of the solution, biosorbent dosage and temperature on the removal of Malachite Green from the aqueous solution (prepared in the laboratory) using Alocasia Macrorrhizos leaf powder as biosorbent. The effect of various parameters on the biosorption of MG are first analyzed by experiments and then attempted theoretically to justify and compare the observations made from graphical analysis.

Effect of Contact time (t)

The effect of contact time on biosorption of Malachite green onto Alocasia Macrorrhizos was studied at 10 mg/L. From Fig-1, it can be seen that the rate of biosorption was very rapid at initial period of contact time. Thereafter, it decreased gradually with time until biosorption was reached at the equilibrium point. The equilibrium time for Alocasia macrorrhizos – malachite green systems was 40 min, thereafter, the $Q_{eq}=91.5$ (Malachite green) and no further biosorption occurred with prolonged time.

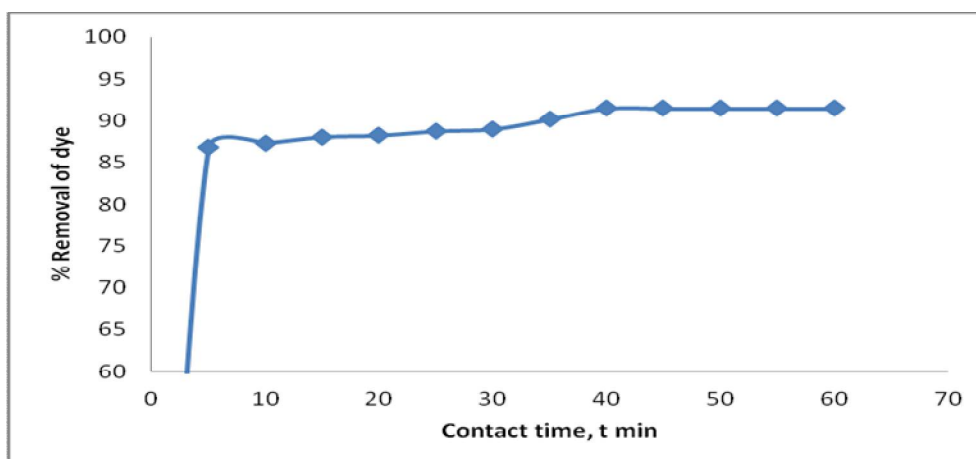


Fig-1: Effect of contact time on % removal of green dye

Effect of initial concentration of aqueous dye solution (C_0)

From the Fig-2 it showed that the dye uptake increased with an increase in initial concentration of dye while the percentage removal of dye decreased with an increase in initial dye concentration. However, the percentage removal of dye on Alocasia macrorrhizos leaf powder was decreased from 92.20 to 87.74 % for malachite green. Though an increase in dye uptake was observed, the decrease in percentage removal may be attributed to lack of sufficient surface area to accommodate much more dye available in the solution. The percentage removal at higher concentration levels shows a decreasing trend where as the equilibrium uptake of dye displays an opposite trend.

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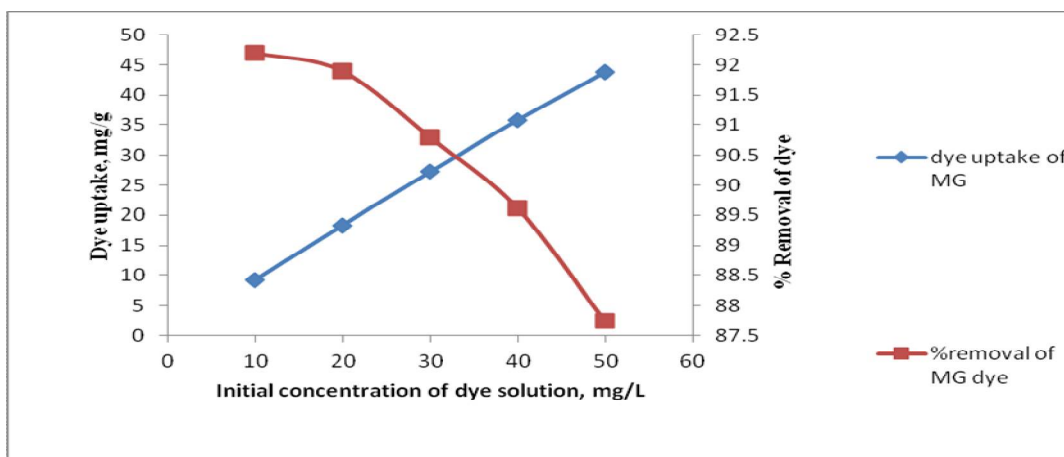


Fig-2: Effect of initial concentration of green dye on %removal and dye update

Effect of pH:

The effect of pH of aqueous solution on % biosorption of malachite green dye is shown in Fig-3. The % biosorption of malachite green dye is increased from 87.77 to 92.11 % as pH increased from 2 to 6 and beyond the pH value of 6 it was decreased. Hence optimum pH for malachite green is taken as 6.

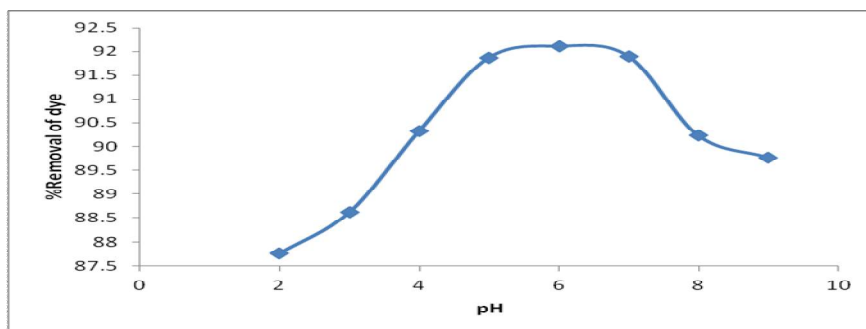


Fig-3:Effect of pH on % removal of malachite green dye

Effect of biosorbent dosage (w)

The percentage biosorption and dye uptake of malachite green dye are drawn against biosorbent dosage for 150 μ m biosorbent size Fig-4. The biosorption of malachite green dye increased from 86.87 to 96.90 (with an increase in biosorbent dosage from 0.05 to 0.2 g). Such behavior is obvious because with an increase in biosorbent dosage, the number of active sites available for malachite green dye biosorption would be more. The change in % biosorption of malachite green dye is marginal.

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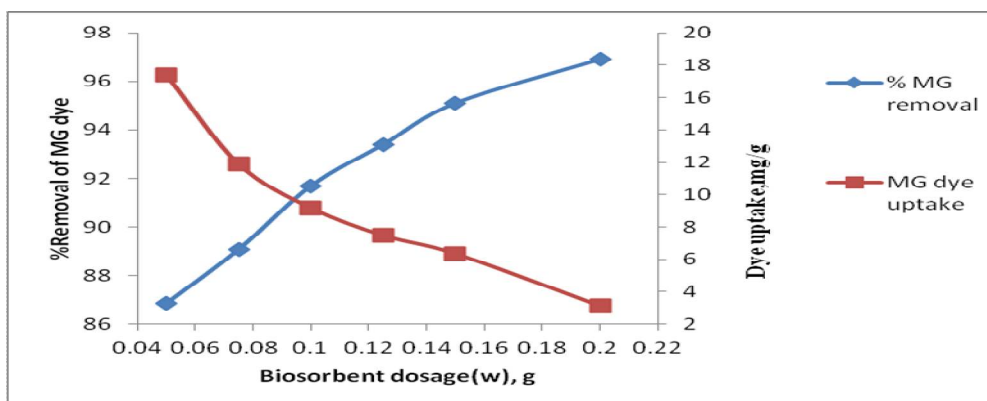


Fig-4: Effect of biosorbent dosage on %removal and uptake malachite green dye

Effect of Temperature (T)

The effect of temperature was investigated from batch experiments carried out at five different temperatures: 283, 293, 303, 313 and 323 K. With an increase in temperature, the % removal was increased from 84.68 to 95.70 % for malachite green for the initial concentration of 10mg/L, as shown in Fig-5.

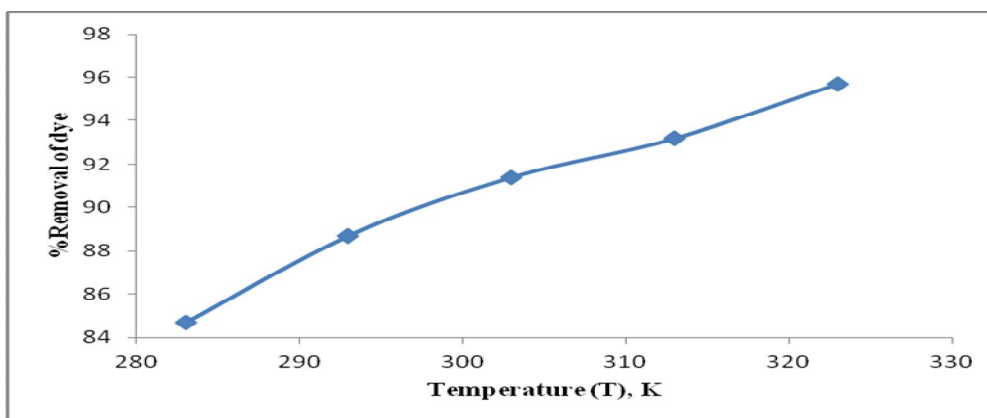


Fig-5: Effect of temperature on %removal of malachite green dye

Adsorption Isotherms:

The isotherms are characterized by certain constants, the values of which express the surface properties and affinity of the sorbent and can also be used to compare biosorptive capacity of biosorbent for different dyes. Out of several isotherm model equations, four have been applied for this study.

Langmuir Isotherm

A basic assumption of the Langmuir theory is that biosorption takes place at specific homogeneous sites within the biosorbent. It is then assumed that once a solute particle occupies a site, no further biosorption can take place at that site. The rate of biosorption to the surface should be proportional to a driving force. The driving force is the concentration in the solution, and the area is the amount of bare surface.

The Langmuir relationship is hyperbolic and the equation is:

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$$Q_{eq} = \frac{Q_{max} b C_{eq}}{1 + b C_{eq}}$$

The above equation can be rearranged to the following linear form:

$$\frac{C_{eq}}{Q_{eq}} = \frac{1}{b Q_{max}} + \frac{1}{Q_{max}} C_{eq}$$

Fig-6.is plot of $[C_{eq}]$ versus $[C_{eq}/Q_{eq}]$, which is a straight line with slope $1/Q_{max}$ and an intercept of $1/bQ_{max}$.

Table-1 Langmuir isotherm parameters

Dye	Equation	Q_{max} (mg/g)	b, L/mg	R^2
Malachite green	$(C_{eq}/Q_{eq}) = 0.010C_{eq} + 0.078$	100	0.128	0.994

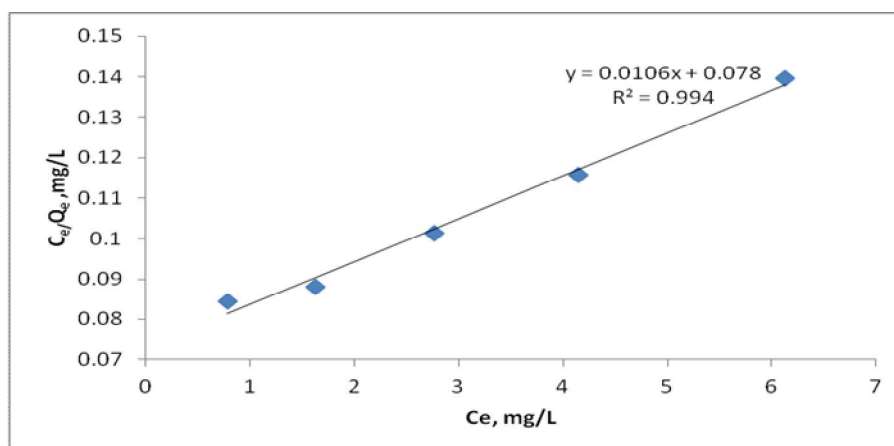


Fig-6: Langmuir isotherm for % biosorption of malachite green dye

Freundlich isotherm:

Freundlich studied the sorption of a material onto animal charcoal. He found that if the concentration of solute in the solution at equilibrium, C_{eq} , was raised to the power m , the amount of solute adsorbed being Q_{eq} , then C_{eq}^m / Q_{eq} is a constant at a given temperature. This fairly satisfactory empirical isotherm can be used for non-ideal sorption and is expressed by the following equation:

$$Q_{eq} = K_f C_{eq}^{1/m}$$

The equation is conveniently used in the linear form by taking the logarithm of both sides as:

$$\log (Q_{eq}) = (1/m) * \log (C_{eq}) + \log (K_f)$$

Freundlich isotherm is derived assuming heterogeneity surface. K_f and m are indicators of biosorption capacity and biosorption intensity. The value of 'm' should lie in between 1-10 for favorable biosorption.

Fig-7 is a plots of $\log [C_{eq}]$ versus $\log [Q_{eq}]$, which is a straight line with a slope of $1/m$ and an intercept of $\log (K_f)$.

From the value of biosorption intensity, it can be concluded that Freundlich isotherm indicates for favorable biosorption.

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Table-2: Freundlich isotherm parameters

Dye	Equation	K _f	m	R ²
Malachite green	$\log Q_{eq} = 0.759 \log C_{eq} + 1.075$	11.8850	1.3175	0.988

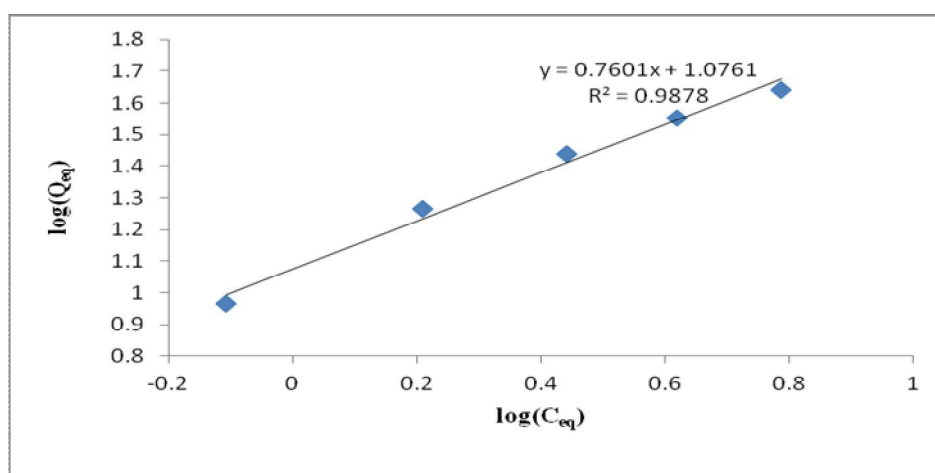


Fig-7: Freundlich isotherm for % biosorption of malachite green dye

Temkin Isotherm:

The Temkin isotherm has generally been applied in the following form (Aharoni and Sparks, 1991):

$$q_{eq} = \frac{R * T}{b_T} \ln [A_T * C_{eq}]$$

This can be written as,

$$q_{eq} = \frac{R * T}{b_T} \ln [C_{eq}] + \frac{R * T}{b_T} \ln [A_T]$$

Fig-8 shows a plot of $\ln [C_{eq}]$ versus Q_{eq} , which is a straight line with slope of RT/b_T and intercept of $RT/b_T \ln [A_T]$.

Table 3: Temkin Isotherm parameters

Dye	Equation	A _T , L/mg	b _T	R ²
Malachite green	$Q_{eq} = 0.058 \ln C_{eq} - 0.677$	8.54×10^{-6}	43433	0.986

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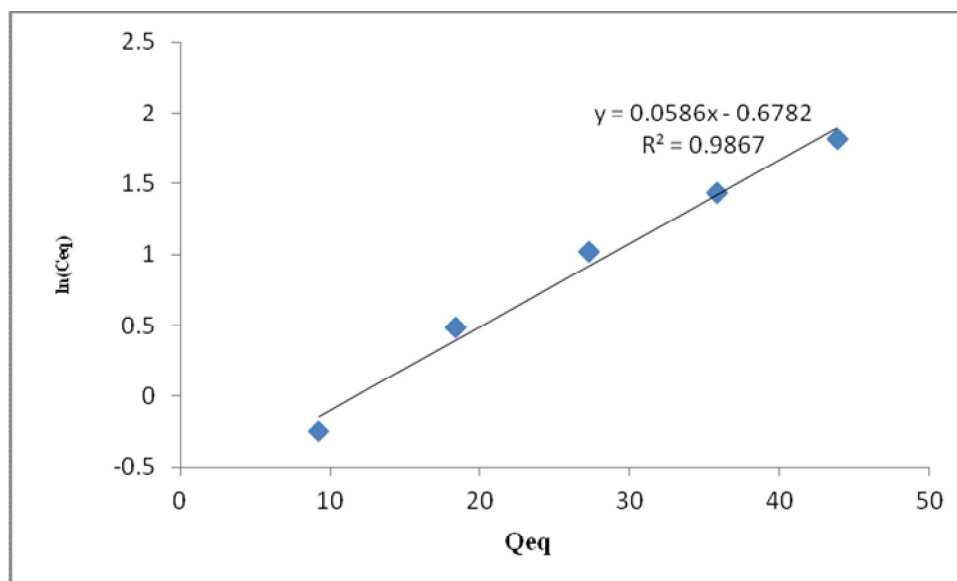


Fig-8: Temkin isotherm for % biosorption of malachite green dye

The correlation coefficients obtained from the Langmuir, Freundlich and Temkin models were 0.994, 0.988 and 0.986 for malachite green and Langmuir equation was observed to be more suitable for the experimental data of malachite green dye.

kinetics:

The kinetics of solute uptake is required for selecting optimum operating conditions for full-scale batch process. The kinetics of the biosorption data was analyzed using two kinetic models, pseudo-first order and pseudo-second order. These models correlate solute uptake, which is important in predicting the reactor volume.

Pseudo First order kinetics:

The order of biosorbate – biosorbent interactions have been described using kinetic models. Traditionally, the Lagergren first order model finds wide application. In the case of biosorption preceded by diffusion through a boundary, the kinetics in most cases follows the Lagergren first order rate equation:

$$(dQ_t/dt) = K_1 (Q_{eq} - Q_t)$$

Where Q_{eq} and Q_t are the amounts of dyes adsorbed at equilibrium time and any time t and K_1 is the rate constant of the pseudo first order biosorption.

The above equation can be presented as

$$\int (dQ_t / (Q_{eq} - Q_t)) = \int K_1 dt$$

Applying the initial condition $q_t = 0$ at $t = 0$, we get

$$\log (Q_e - Q_t) = \log Q_e - (K_1/2.303) t$$

Plot of time t versus $\log (Q_e - Q_t)$ gives a straight line for first order kinetics, facilitating the computation of biosorption first order rate constant (K_1).

In the present study, the kinetics were investigated with 100 ml of aqueous solution ($C_0 = 10\text{mg/L}$) for the contact time of 5 to 60 min. Lagergren first order plots are drawn in Fig.9.

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Table.4 First order equation and coefficients

Kinetics	Dye	Model equation	R ²	Rate constant, K ₁
First order	Malachite green	$\log(Q_{eq}-Q_t) = 0.081t - 0.228$	0.767	0.186 min ⁻¹

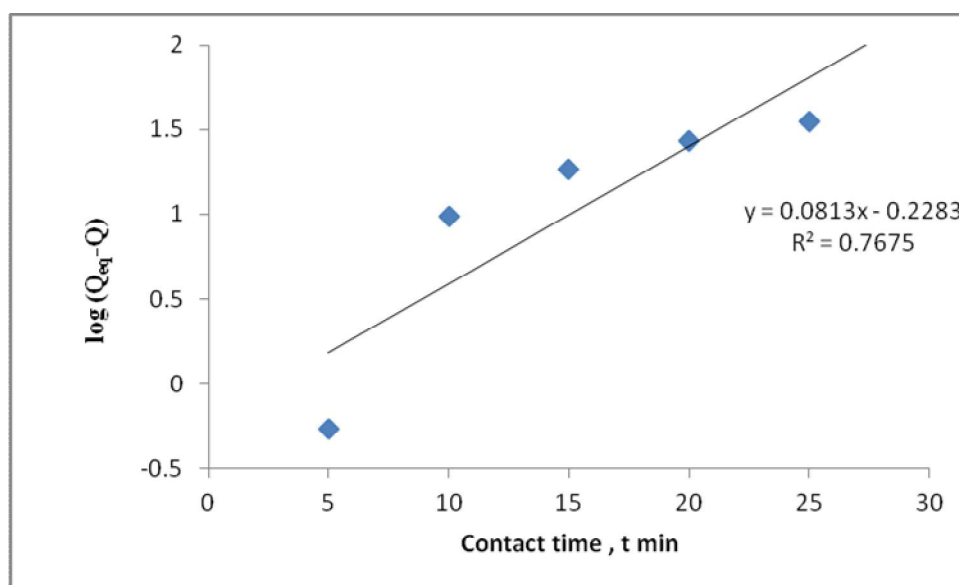


Fig-9: first order kinetic for % biosorption of malachite green dye

Pseudo second order kinetic equation:

If the experimental results do not follow the above equation, pseudo second order kinetic equation: $(dq_t/dt) = K_2 (Q_e - Q_t)^2$ is applicable, where 'K₂' is the second order rate constant.

The other form of the above equation is:

$$(dq_t / (Q_e - Q_t)^2) = K_2 dt$$

Let $Q_e - Q_t = x$

$dQ_t = dx$

$1/x = K_2 x + C$

$C = 1/Q_e$ at $t = 0$ and $x = q_e$

Substituting these values in above equation, we obtain:

$$1/(Q_{eq} - Q_t) = K_2 t + (1/Q_{eq})$$

Rearranging the terms, we get the linear form as:

$$(t/Q_t) = (1/ K_2 Q_{eq}^2) + (1/Q_{eq}) t.$$

If the pseudo second order kinetics is applicable, the plot of time t versus (t/Q_t) gives a linear relationship that allows computation of K_2 .

In the present study, the kinetics are investigated with 100 ml of aqueous solution ($C_0 = 10\text{mg/L}$) in the agitation time intervals of 5 min to 60 min. Pseudo second order plot of time 't' versus (t/Q_t) drawn in Fig-10.

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Table 5. Pseudo Second order kinetics

kinetics	Dye	Model equation	R ²	Rate constant, K ₂
Pseudo Second order	Malachite green	$t/Q_t = 0.095 t + 2.339$	0.976	0.0038 gm.(mg.min) ⁻¹

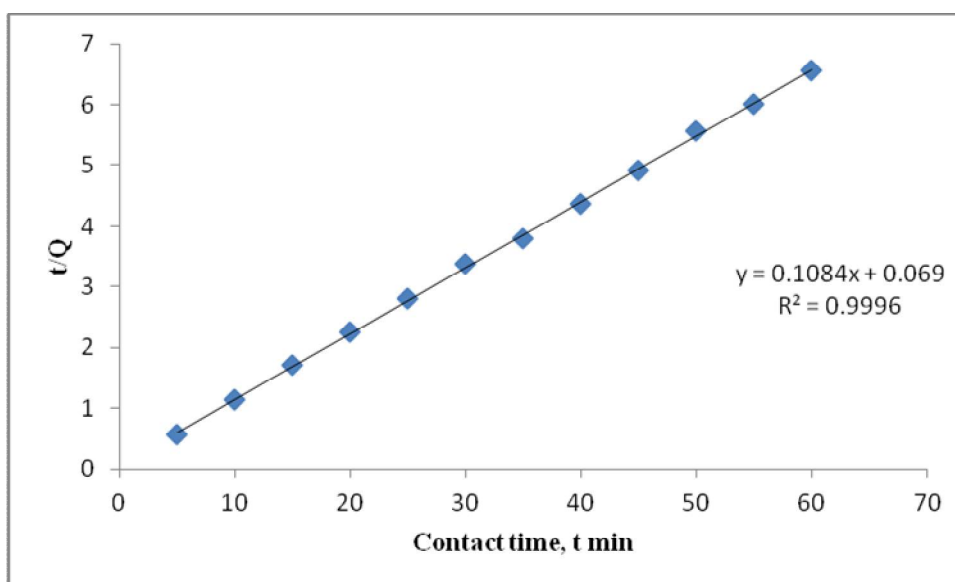


Fig-10:second order kinetics for %biosorption of malamchite dye

Intra-particle diffusion model:

The malachite green is most probably transported from the bulk of solution into the solid phase by intra-particle diffusion, which is often the rate limiting step in many biosorption processes. The possibility of intra-particle diffusion is explored by using the following equation (Furusawa and Smith (1992))

$$Q_t = K_{dif} t^{0.5} + C$$

Where

C is the intercept and K_{dif} is the intra-particle diffusion rate constant.

In the present study, the kinetics are investigated with 100 ml of aqueous solution ($C_0 = 10\text{mg/L}$) at different biosorbent dosages for the contact time of 5 to 60 min. The plot is shown in Fig-11.

$$Q_t = 0.100t^{0.5} + 8.416, R^2 = 0.932 \text{ (Malachite green)}$$

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Table.6: Intra-particle diffusion rate constant

S.No	Intra particle diffusion model	Slope, K_{id} (mg/g-min ^{0.5})	Intercept, I	R ²
1	Malachite green	0.100	8.416	0.932

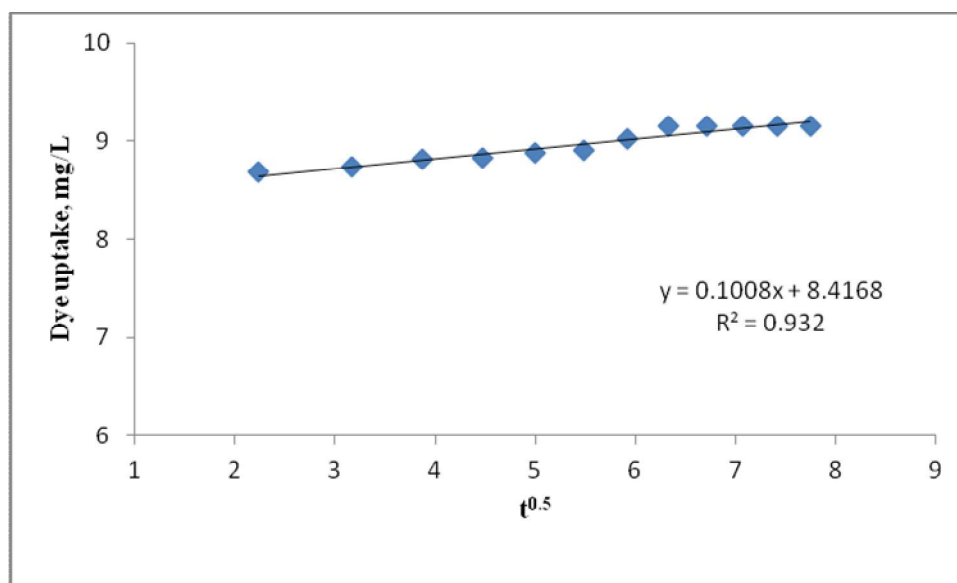


Fig-11: intra particle diffusion model for biosorption of malachite green dye

Thermodynamic studies:

Thermodynamic parameters such as Enthalpy change (ΔH°), Free energy change (ΔG°) and Entropy change (ΔS°) can be estimated using equilibrium constants changing with temperature. The free energy change of the sorption reaction is given by the following equation:

$$\Delta G^\circ = -RT \ln K_a$$

Where,

- ΔG° - Standard free energy change in J/mol,
- R - Universal gas constant, 8.314 Jmol⁻¹ K⁻¹,
- T - Absolute temperature, K
- K_a - Equilibrium constant

The free energy change indicates the degree of spontaneity of the Biosorption process and the negative value reflects more energetically favorable biosorption.

The equilibrium constant may be expressed in terms of enthalpy change of biosorption as a function of temperature as follows:

$$\frac{d \ln K_a}{dT} = \frac{\Delta H^\circ}{RT^2}$$

According to above equation, the effect of temperature on the equilibrium constant K_a is determined by the sign of ΔH° . Thus, when ΔH° is positive, i.e., when the biosorption is endothermic, an increase in temperature (T) result in an increase in K_a . Conversely, when ΔH° is negative, i.e., when the biosorption is exothermic, an increase in temperature (T) result in a decrease in K_a implied a shift of the biosorption equilibrium to the left.

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The integrated form of Eq becomes

$$\ln K_a = \frac{-\Delta H^0}{R \cdot T} + Y$$

This equation can be rearranged to

$$\Delta G^0 = -R \cdot T \ln K_a = \Delta H^0 - Y \cdot R \cdot T$$

$$\Delta G^0 = \Delta H^0 - T \cdot \Delta S^0$$

Where, $R \cdot Y = \Delta S^0$, free energy change with temperature.

The equilibrium constant can be represented as follows:

$$\ln [K_a] = \frac{-\Delta H^0}{RT} + \frac{\Delta S^0}{R}$$

This shows clearly that the biosorption process is composed of two contributions, enthalpy change and entropy, which characterize whether the reaction is spontaneous. The ΔG^0 , ΔH^0 and ΔS^0 values of malachite green dye ions at different temperatures 303K-323 K are given in Table.7

$$\ln [K_a] = -3332(1/T) + 13.36(\text{Malachite green})$$

Thermodynamic parameter for the biosorption process of malachite green is computed from the graph of $\ln [K_a]$ versus $1/T$, which is shown Fig-12.

Values of ΔG^0 obtained for malachite green is -6.633KJ/mol. Negative values of ΔG^0 show the feasibility and spontaneity of the biosorption process.

The enthalpy changes ΔH^0 of biosorption for malachite green are 27.702 KJ.mol⁻¹ and entropy change ΔS^0 for malachite green are 111.07J.mol⁻¹.K⁻¹. The positive value for ΔH^0 confirms that the overall biosorption of malachite green by *Alocasia macrorrhizos* leaf powder is an endothermic process.

The positive values of entropy change reflects the affinity of the biosorbent for malachite green and increased randomness at the solid liquid interface with some structural changes in the biosorbate and biosorbent. Also positive values of ΔS^0 correspond to an increase in the degree of freedom of the adsorbed specie

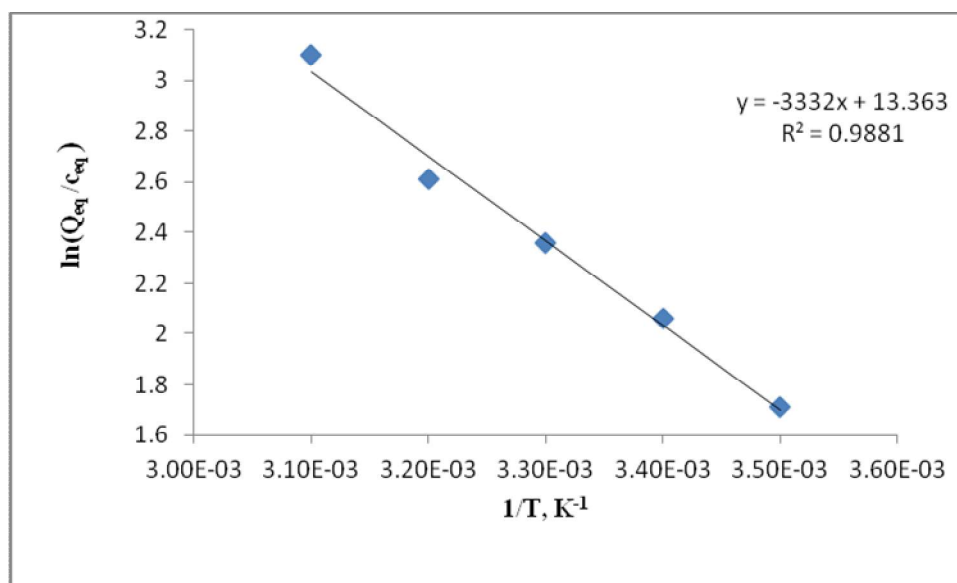


Fig-12: Van't hoff plot

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Table.7: Thermodynamic parameters for Adsorption of Malachite Green

S.NO	Temperature, K	ΔH , KJ mol ⁻¹	ΔS , J mol ⁻¹ K ⁻¹	ΔG , KJ mol ⁻¹
1	283	27.702	111.07	-3.823
2	293	27.702	111.07	-4.933
3	303	27.702	111.07	-6.633
4	313	27.702	111.07	-7.153
5	323	27.702	111.07	-8.263

IV. CONCLUSIONS

The aim of this investigation is to determine the suitability of *Alocasia Macrorrhizos* leaf powder as a biosorbent for the removal of malachite green dye from aqueous solutions. The equilibrium, kinetic and thermodynamics studies are carried out for biosorption of Malachite green dye experimentally and theoretically from the analysis of the experimental data the following conclusions were made:

1. The biosorption performance was strongly affected by parameters such as contact time, initial dye concentration, pH, biosorbent dosage and temperature.
2. The percentage biosorption of Malachite green was increased with an increase in contact time.
3. The equilibrium time for Malachite green was 40 min.
4. With an increase in the initial concentration the percentage removal was decreased and dye uptake was increased.
5. The dye uptake was decreased and percentage removal was increased with an increase in the biosorbent dosage from 0.05-0.2g
6. The plot of p^H versus percentage removal of dye showed that the significant biosorption took place at p^H value of 6 for Malachite green .
7. The percentage biosorption of Malachite green was increased with an increase in the temperature.
8. The dye removal data of Malachite green follows the Langmuir model with the best fit.
9. The kinetics of the biosorption of Malachite green on *Alocasia Macrorrhizos* leaf powder can be better described with pseudo second-order kinetics.
10. The study reveals the endothermic nature of Biosorption as ΔH is positive, irreversible nature of Biosorption as ΔS is positive and spontaneity of Biosorption as indicated by negative ΔG for Malachite green dyes.
11. From kinetic data it was concluded that Malachite green dye is endothermic in nature. Hence the above said *Alocasia Macrorrhizos* leaf powder is effective and efficient biosorbent and is capable of removing Malachite green .

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NOMENCLATURE

C	Concentration of solute in the solution at any time t, mg/L
C_0	Initial concentration of dye, mg/L
C_{eq}	Concentration of Biosorbate in equilibrium, mg/L
Q	Dye uptake, mg/g
Q_{eq}	Equilibrium dye uptake, mg/g
t	Contact time, minutes
W	Weight of biosorbent per liter of aqueous dye solution, g/L
V	Volume of aqueous dye solution, ml.
d_p	particle size of the biosorbent, μm .
T	Temperature of the dye solution, K
R^2	Correlation coefficient
Q_{max}	maximum sorption capacity, mg/g (q_{eq} for a complete monolayer)
b	Langmuir isotherm constant, L/mg.
K_f	Freundlich isotherm coefficient.
m	measure of adsorption intensity (between 1-10 favorable adsorption)
A_T, b_T	Temkin isotherm constants, L/mg.
K_1	pseudo first order rate constant, (min^{-1})
K_2	pseudo second order rate constant, ($\text{g} / \text{mg} \cdot \text{min}$)
R	Universal gas constant, (8.314 J/ mol K)
K_d	Intra particle diffusion rate constant, $\text{mg} \cdot \text{g}^{-1} \cdot \text{min}^{-0.5}$
ΔG°	Gibb's free energy of adsorption (KJ/mol)
ΔH	Change in enthalpy (KJ /mol)
ΔS	Change in entropy (J/mol K)
K_a	Equilibrium constant.