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Removal of Rhodamine 6G from Aqueous Solution by Adsorption on Bio Adsorbent Prepared from Hyptis Suaveolens (Vilayti Tulsi): Kinetic, Equilibrium and Thermodynamic Study

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Abstract

The adsorption capacity of bio adsorbent, prepared from Hyptissuaveolens (VilaytiTulsi), for Rhodamine 6G removal from aqueous solution was investigated in the present study. The effect of ph, initial dye concentration, time, adsorbent dosage and temperature was investigated. The present adsorption follows pseudo second order kinetics. Langmuir isotherm and Freundlich isotherm was used for present study. The maximum adsorption capacity under optimum condition was found to be 48.78 mg g^{-1}. The thermodynamics study shows endothermic, spontaneous adsorption process.

Keywords: Rhodamine 6G, Adsorption, Dye removal, Bio adsorbent.

Introduction:

Different chemical materials were used in various industrial processes. Dyes are the mostly used organic chemical in the industries such as paper and pulp, lather, textile etc. Residual part of dyes comes in effluent of such industries creating environment problem [1, 2]. Most of the organic dyes are highly toxic and carcinogenic [3-5]. The presence of dyes can seriously affect the light penetration and damage the aquatic life [6]. There are different methods available for removal of hazardous dyes such as photo degradation and photo catalysis [7-10], electrochemical degradation [11], bio degradation [12, 13], chemical coagulation and adsorption. Among all adsorption have been the most common method employed for dye removal [14-16]. Activated carbon is the most suitable adsorbent but it is quite expensive so alternate cheaper adsorbent is a need [17-18].

Literature survey shows that different adsorbent have been reported for dye removal some of them are nanomaterials [19] such as magnetite@graphene oxide [20] Carbon nanotubes [21], $CoFe_2O_4$/rGO nanocomposite [22] while some are prepared from natural material such as Moroccan natural phosphate [23], activated carbon prepared from Prosopisspicigera L. wood [24], rice husk [25, 26], Clitoriafairchildiana pods [27], Prunusamygdalus L. [28] etc.

Rhodamine 6G is a fluorescent basic dye mainly used to coloured wool, cotton, silk etc. It is toxic and carcinogenic in nature [8, 29], so this dye was selected for present study.

Experimental:

Preparation of Adsorbent

Fully grown plants of Hyptissuaveolens (*VilaytiTulsi*) were collected around Vaijapur city. The steams and braches were cut into small pieces, washed with distilled water for 2-3 times, dried under shed. The adsorbent was prepared as per the procedure mentioned in our earlier publication [30].

Preparation of sorbet

Rhodamine 6G (Rh 6G), a cationic dye supplied by LobaChem India with Colour Index (C.I.) 45,160 and molecular formula $C_{28}H_{31}N_2O_3Cl$ was used for present study. A stock solution of 1000 mg L^{-1} was prepared by dissolving accurately weigh dye quantity in double distilled water. Dilution with double distilled was carried out to get desired experimental concentration.

Adsorption Studies

For adsorption studies, in 250 mL stoppered glass bottle 50 mL dye solution of desired concentration and pH was taken at room temperature. 0.1 g adsorbent was added, the solution was stirred by mechanical shaker. At predetermined time interval, the small fraction were withdrawn, the dye solution was separated from adsorbent by centrifugation at 4,000 rpm. The absorption of supernatant solution was measured. The standard curve of dye was prepared with 1-9 mgL^{-1} solution at 526 nm using Elico double beam spectrophotometer SL-210. 0.1 M HCl and 0.1 M NaOH was used to control initial pH. Different adsorbent dosage (0.05 to 0.3 g) and 50 mL of 50 mgL^{-1} dye solution was shaken for 30 min to study effect of adsorbent dosage. Effect of temperature was studied by adding 0.1 g adsorbent in 50 mL dye solution of 25 mgL^{-1}, 50 mgL^{-1}, 75 mgL^{-1} and 100 mgL^{-1} concentration at 313, 323 and 333 ^{0}K in thermostat rotatory shaker. The following equation was used to determine the solid phase dye concentration at time.

$$q_t = \frac{(c_0 - c_t)v}{w} \qquad (1)$$

whereq_t is adsorption amount at time t, C_o and C_t are dye concentration initial and at time t in mg L^{-1} respectively, V is volume of solution in L and W is weight of adsorbent in g. The adsorption capacity of adsorbent was determined by using Langmuir and Freundlich isotherm.

Results and Discussion:

Effect of pH

Initial pH of the dye solution is the most dominant parameter that affect the adsorption capacity of the adsorbent [31]. The initial pH of solution affect the adsorption procedure as it affect the ionization of dyes and adsorbent surface [14]. To study the effect of pH, 50 mL solution of 50mg L^{-1} dye concentration was shaken with 0.1g adsorbent for 30 min. An increase in pH from 4 to 7.5 increases the adsorption of Rhodamine 6G (16.14 to 20.19 mg g^{-1}) further increase in pH from 7.5 to 10 slightly decreases the adsorption. The optimum pH is 7.5. Fig.1 shows the effect of pH on adsorption.

Effect of Adsorbent dose

To test the effect of adsorbent dose, 50 mg L^{-1} dye concentration was stirred with varying adsorbent amount (0.05 to 0.3 g) at optimum pH for 30 min. the result was shown in Fig.2. It has been observed that due to increase in adsorption site the % removal of dye increases but unit adsorption was decreased from 39.7 mg g^{-1} to 7.18 mg g^{-1} as amount of adsorbent was increased from 0.05 g to 0.3 g.

Effect of dye concentration

To study the effect of initial dye concentration, 50mL dye solution with varying concentration (25 mg L^{-1} to 100 mg L^{-1}) was stirred with 0.1 g adsorbent at optimum pH (7.5). The results are shown in Fig.3, as the initial dye concentration increases the percentage removal of dye decreases, but the unit adsorption increases from 10.27 mg g^{-1} to 48.72 mg g^{-1}.

Adsorption dynamics

The pseudo first order kinetic model

The pseudo first order kinetic model expression is given by Lagergren [32] as follows

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \qquad (2)$$

Where q_t and q_e are amount of dye adsorbed at time t and equilibrium, respectively k_1 is the rate constant. Fig. 4 shows the Lagergren pseudo first order plot for adsorption of Rh 6G at various initial concentration. The values of k_1 and q_e were calculated from slope and intercept from plot of log (q_e-q_t) versus t and represented in table1. The experimental data with Lagergren pseudo first order plot provide poor correlation coefficient (R^2) values similar inapplicability was also observe by Lata et al [33].

The pseudo second order kinetic model

The pseudo second order Lagergren equation is expressed as [34]

$$\frac{t}{q_t} = \frac{1}{q_e^2 k_2} + \frac{t}{q_t} \qquad (3)$$

Plot of t/q_tversus t was shown in the Fig.5. From the slopes and intercepts, values of equilibrium adsorption capacity (q_e) and second order rate constant (k_2) were determined and expressed in table 1. From the values of regression coefficient it can be concluded that the present system follows pseudo second order Lagergren model. The adsorbent and adsorbate both affect the adsorption process of present study [35].

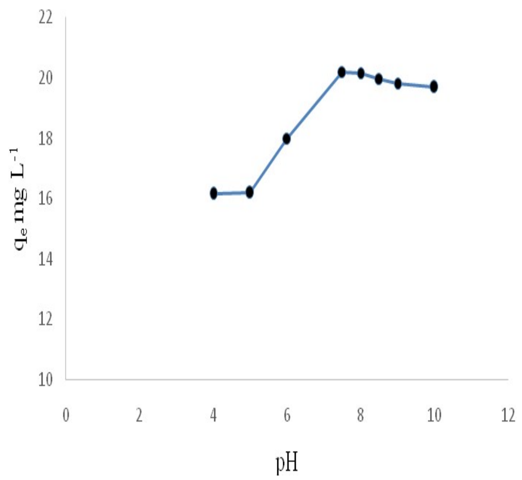

Fig.1: Effect of pH on dye removal

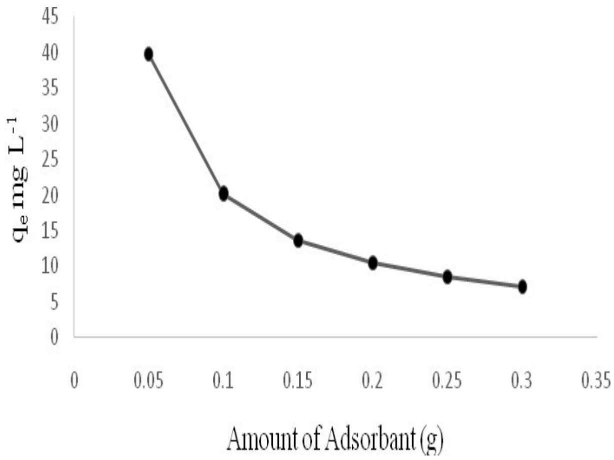

Fig. 2: Effect of Adsorbent dose

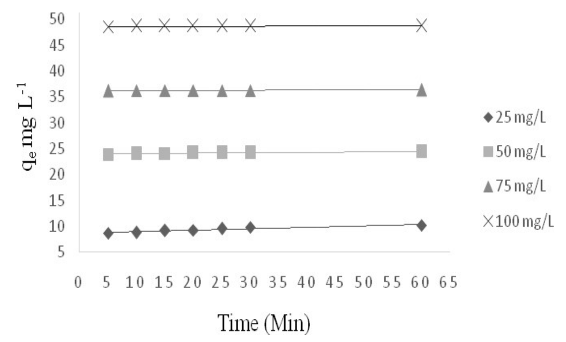

Fig. 3: Effect of initial dye concentration on adsorption

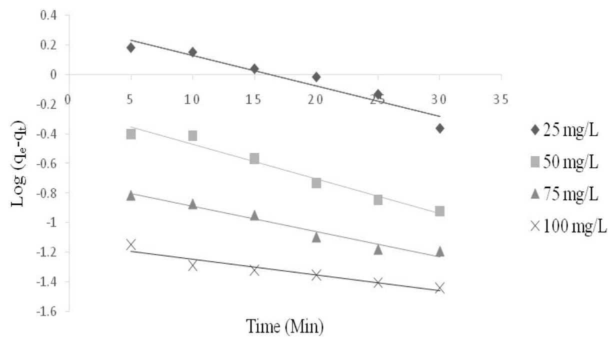

Fig. 4: The pseudo first order kinetic

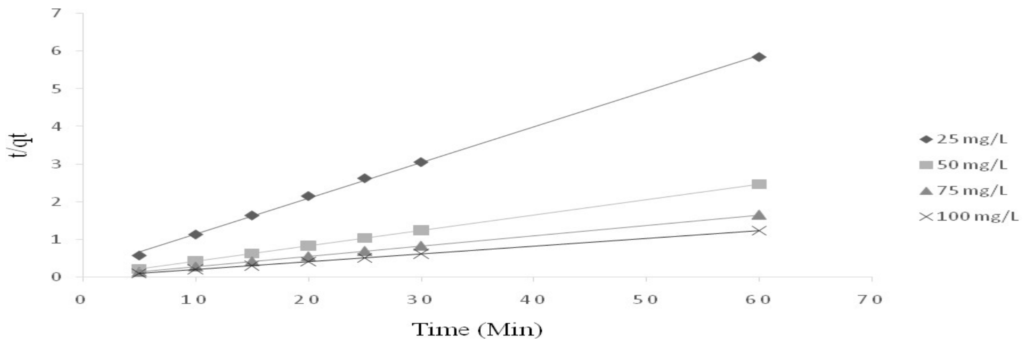

Fig. 5: The pseudo second order kinetic

Table 1: Rate constants for pseudo first-order and pseudo second-order adsorption

Conc. C_0 (mg L^{-1})	pseudo first-order			pseudo second-order		
	q_e (mg g^{-1})	K_1 (min^{-1})	R^2	q_e (mg g^{-1})	K_2 (min^{-1})	R^2
25	2.186	0.0476	0.9244	10.537	0.0472	0.999
50	1.7290	0.0536	0.9680	24.390	0.2302	1
75	5.2432	0.0391	0.9612	36.363	0.5817	1
100	13.749	0.0244	0.9124	48.780	1.4008	1

Adsorption equilibrium study

Two isotherm, Langmuir isotherm and Freundlich isotherm was used for present study.

Langmuir isotherm

Langmuir isotherm is represented by following equation [36]

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{bq_m} \quad (4)$$

Where q_e is the amount adsorbed at equilibrium (mg g^{-1}), Ce is the equilibrium dye solution concentration (mg L^{-1}), q_m is Langmuir constant (related to adsorption capacity) (mg g^{-1}) and b is Langmuir constant (related to energy of adsorption) (L mg^{-1}). Fig 6 shows plot of Ce/q_e versus C_e, the isotherm parameters are given in table 2.

Freundlich isotherm

Freundlich isotherm is represented by following equation [36, 37]

$$\log q_e = (1/n) log C_e + \log k_f \qquad (5)$$

Where k_f is adsorption capacity, n is adsorption intensity, C_e is equilibrium dye concentration in solution and q_e is equilibrium dye concentration in solid. Fig 7 shows plot of log q_e versus log C_e, the isotherm parameters are given in table 2.

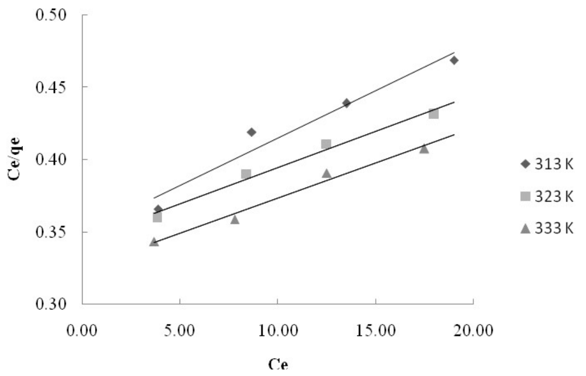

Fig. 6: Langmuir isotherm

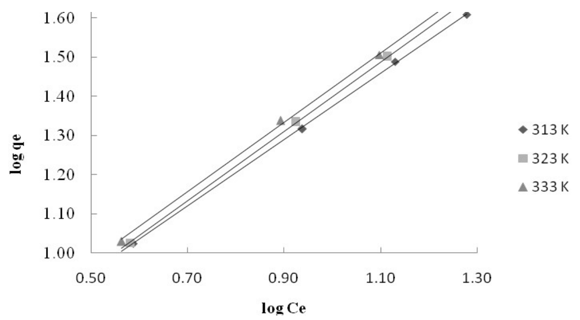

Fig. 7: Freundlich isotherm

A dimensionless constant called as equilibrium parameter or separation factor RL can be an essential parameter for Langmuir model is given by following equation [36, 38]

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

Where b is Langmuir constant (related to energy of adsorption) (L mg^{-1}), C_0 is initial dye solution concentration mg L^{-1}. In the present study the value of R_L (table 2) lie in between 0 to 1 indicate favourable adsorption.

Table 2: Langmuir and Freundlich isotherm parameter

Temp (^{0} K)	Langmuir isotherm parameter				Freundlich isotherm parameter		
	q_m (mg g^{-1})	b (L mg^{-1})	R_L	R^2	n	k_f (mg g^{-1})	R^2
313	205.9927	0.0149	0.7280	0.9810	1.1798	3.3500	0.9998
323	199.4389	0.0155	0.5626	0.9620	1.1285	3.2494	0.9999
333	297.9671	0.0096	0.5809	1.0000	1.1260	3.412	0.999

Effect of temperature

In the present study, increase in temperature increases dye removal percentage. Thermodynamic parameter were determined by using following equation.

$$K_0 = \frac{C_{solid}}{C_{liquid}} \qquad (7)$$

Where C_{solid} is equilibrium solid phase concentration (mg L^{-1}), C_{liquid} is equilibrium liquid phase concentration (mg L^{-1}) and K_0 is equilibrium constant. Gibb's free energy (ΔG) is represented by following equation

$$\Delta G = -RT ln K_0 \qquad (8)$$

Where K_0 is equilibrium constant, R is gas constant and T is temperature in Kelvin. The Van't Hoff equation is represented by following equation.

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

Fig. 8 represent plot of log K_0 versus 1/T, from the slope and intercept of this Van't Hoff plot the values of ΔH and ΔS were determined and represented in table 3.

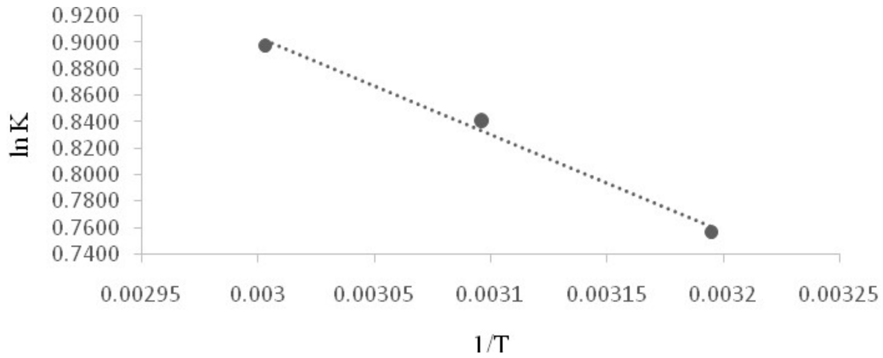

Fig. 8: log K_0 versus 1/T

The value of ΔG is -19.69 to -24.84 (kJ mole^{-1}), ΔH is 14.06 (kJ mole^{-1}) and ΔS is 59.49 (J K^{-1} mole-^{1}). On the basis of enthalpy ΔH, the adsorption process can be of three type [39]

i) Chemical adsorption (80 < ΔH < 450 kJ mole^{-1})
ii) Electrostatic interaction (20 < ΔH < 80 kJ mole^{-1})
iii) Physical adsorption (ΔH < 20 kJ mole^{-1}).

The positive value of ΔH indicate an endothermic process. The negative value of ΔG indicate spontaneous and favourable adsorption process. The positive values of ΔS indicate increase in randomness of water molecules surrounding the dye molecules.

Table 3: Thermodynamic parameter of adsorption

Temp (^{0} K)	**ΔG (kJ mole^{-1})**	**ΔH (kJ mole^{-1})**	**ΔS (J K^{-1} mole^{-1})**
313	-19.69	14.06	59.49
323	-22.58		
333	-24.84		

Conclusion:

Bio adsorbent prepared from *Hyptissuaveolens (VilaytiTulsi)* was used to study removal of Rhodamine 6G from aqueous solution under different experimental condition. It has been observed that under optimum condition 48.78 mg g^{-1} dye can be removed from aqueous solution using the bio adsorbent. The pseudo second order kinetic model was best suited for the present study. Adsorption equilibrium study shows that both Langmuir isotherm and Freundlich isotherm fits for the present study shows mono layer adsorption process. Thermodynamic study indicates an endothermic, spontaneous adsorption process. This result shows that bio adsorbent prepared from *Hyptissuaveolens (VilaytiTulsi)* can be used as a low cost adsorbent for the removal of Rhodamine 6G.

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Conflict of Interest:

The authors declare that they have no conflict of interest.

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