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Adsorption of Malachite Green Dye on Termite Soil

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ABSTRACT:

A number of adsorption materials have been found to remove the toxic heavy metals and dyes from an aqueous solution and hence they are employed in tertiary water treatment process. Activated carbons, plant bio masses, animal wastes and many soils are found to be non-toxic, abundant, eco-friendly adsorbents and adaptable to laboratory conditions. The aim of this research work is to evaluate the efficiency of Termite soil to adsorb Malachite green dye (MG dye) from an aqueous solution. The main objectives of this investigation are to find a best fitting isotherm and kinetic model to describe the present system. The equilibrium study data, isotherm constants, kinetic data and the FTIR spectral studies would be helpful to understand the mechanism of adsorption of MG dye from an aqueous solution by the Termite soil.

KEYWORDS: Malachite green, Termite soil, Adsorption isotherm, NaoH, Hcl.

1. INTRODUCTION

Water pollution is a major global problem which requires ongoing evaluation and revision of water resource policy at all levels. When toxic substances enter lakes, streams, rivers, oceans, and other water bodies, they get dissolved or lie suspended in water or get deposited on the bed. Waste water from chemical manufacturing or chemical processing industries contribute much to the water pollution. Industrial waste water usually contains specific and readily identifiable chemical compounds. Dyes are used primarily in the production of consumer products, including paints, textiles, printing inks, paper and plastics. Among the different pollutants of aquatic ecosystems, dyes are large and important group of industrial chemicals. Textile dyes are generally aromatic compounds and may consists of some heavy metals embodied in their structure. Adsorption is a process that occurs

when a liquid or gas called adsorbate accumulates on the surface of a solid or liquid forming a molecular or atomic film. Adsorbents are used usually in the form of spherical pellets, rods mouldings or monoliths with hydro dynamic diameters between 0.5 and 10 mm. They must have high abrasion resistance, high thermal stability and small pore diameters which results in high exposed surface area and hence high surface capacity for adsorption.

Dudova M, Klika Z, Capkova P et al (2011) have detailed out The adsorption of Malachite green on montmorillonite from acid solutions. *Mohd Rafatullah, Othman Sulaiman,* (2011) have studied the thermodynamic parameters for the adsorption of Methylene Blue on different low cost adsorbents. *Xavier A & Usha D,* (2011) removed of Methylene Blue dye from textile and leather industrial waste, using activated carbon as adsorbents namely, Commercial Activated Carbon (CAC), Rose Apple Carbon (RAC), Coconut shell carbon (CSC) and Saw dust carbon (SDC). Xiulihan etal (2011) has done studies on the adsorption potential of lotus leaf to remove Methylene Blue from aqueous solution was investigated in batch and fixed bet column experiments, which followed Langmuir, Freundlich, Temking and Koble Corrigan isotherm models adsorption behaviour. *Gaikuad, Misal SA et al* (2010) worked on the removal of Malachite green from aqueous solution by adsorption on silica gel has been carried out. *Barati A, Alikhani H, Hekmat A, Zendehdel M,* (2010) have studied dye removal from waste water of several classes dye. Malachite green has wide cause some harmful effects in humans. *Marcelo J, Avena et al* (2009) did studies on kaolinite and four soil samples to determine the effect of Methylene Blue dimerization on the measured surface area. *Hongs, Wenc, et al* (2009) investigated the effect of temperature on the equilibrium for removal of Malachite green dye from aqueous solution using bentonite. *Oualid Hamdaoui et al* (2007) investigated on the removal of Methylene Blue from aqueous solution by wheat bran in batch conditions. *Arvind L, Thakkar, William L,* (2006) used potato starch to remove Methylene Blue from aqueous solution of a monomer or dimer, depending upon its concentration.

A number of adsorption materials have been found to remove the toxic heavy metals and dyes from an aqueous solution and hence they are employed in tertiary water treatment process. Activated carbons, plant bio masses, animal wastes and many soils are found to be non-toxic, abundant, eco-friendly adsorbents and adaptable to laboratory conditions. The aim of this research work is to evaluate the efficiency of Termite soil to adsorb Malachite green dye (MG dye) from an aqueous solution. The main objectives of this investigation are to find a best fitting isotherm and kinetic model to describe the present system.The equilibrium study data, isotherm constants, kinetic data and the FTIR spectral studies would be helpful to understand the mechanism of adsorption of MG dye from an aqueous solution by the Termite soil.

2. EXPERIMENTAL METHODS

2.1. Materials and methods

Analar grade Malachite green dye of Sigma Aldrich was used.It is a cationic dye of amorphous nature having molecular formula $C_{23}H_{25}N_2Cl$, molecular weight 364.911g/mol. Methylene blue is a hetero cyclic aromatic chemical compound. Though it has many uses in a range of different fields such as biology and chemistry, it also has many adverse effects including hypertension, pre-cordial pain, dizziness, mental confusion, headache, fever, staining of skin, injection site necrosis (SC), fecal discoloration, nausea, vomiting, abdominal pain, discoloration of urine, bladder irritation, anemia. It causes tumors to form in the liver, thyroid and mammary glands of mammals.

2.2. Preparation of Adsorbents, Malachite green solution

2.2.1. Preparation of Adsorbents

Termite soils were collected from Thennangudi village, Thanjavur district. They were washed with distilled water. Size of the Particles present in the soil is graded with the help of sieves. Percentage of the different particle sizes were given in the Table1 and shown as Pie diagramme in Fig. 1, Major portion of termite soil was found to be below the size of 0.75 mm. Particle size below 0.75 mm was taken for the further study as an adsorbent which is designated as Termite Soil Adsorbent (TSA).

2.2.2. Preparation of Malachite green dye solution.

Analar grade Malachite green dye of Sigma Aldrich was used. The stock solution of dye was prepared by dissolving an appropriate amount of accurately weighed dye in double distilled water to a concentration of 1000 mg/L. The experimental solutions were prepared from the stock solution by proper dilution.

2.3. Adsorption Dynamic experiments.

2.3.1. Batch Equilibrium method

In order to estimate the adsorption characteristics of TSA for the adsorption of MG, batch sorption studies were carried out. Known weight of adsorbent is added to 50 mL of prepared dye solution of known concentration. The contents were shaken thoroughly using a rotary mechanical shaker with a speed of 200 rpm. The solution was then centrifuged at preset time intervals by keeping all other factors constant.

2.3.2. Effect of dosage

Effect of adsorbent dosage was studied by varying the adsorbent dose from 15 to 75 mg for 100mg/L of 50 mL MG dye solution. The various dosage consisting of the adsorbent respectively mixed with the dye solution and the mixture was agitated in a mechanical shaker for 100 minutes. The adsorption capacities for different doses were determined by keeping all other factors constant.

2.3.3. Effect of initial dye concentration

In order to determine the effect of initial concentration, experiments were conducted with different initial concentration 100,150 and 200 mg/L of the dye solution for different contact time ranging from 5 to 100 min, keeping all other factors constant.

2.3.4. Desorption Studies

Desorption studies were carried out to recover the dye and to regenerate the spent adsorbent for repeated use. After performing adsorption experiments with 1000 mg/L of dye with 1000 mg of adsorbent dose at pH 7, adsorbents were separated by centrifugation and washed with water to remove the surface deposited dye. 100 mg of the adsorbent loaded with dye was shaken with 50 mL of aqueous solution having the pH values ranging from 2 to 10 for 1 hour. After vigorous agitation, solution is centrifuged. The concentration of the dye in centrifugate was recorded to evaluate the percentage regenerated dye.

Result and Discussion

3.1. Effect of various parameters.

3.1.1. Effect of adsorbent Dose.

The effect of different dosage of absorbent on adsorption can be inferred from Table II and Fig.1 As the amount of adsorbent increases from15to 75 mg for 50 ml of 100 mg/L dye solution, the percentage removal of dye found to increase from 64.64% to 82.5%. The increase in adsorption with increase in dosage may be attributed to increased adsorbent surface with the availability of more adsorption sites. 50 mg of adsorbent is chosen for further studies.

3.1.2. Effect of Contact Time with respect to initial concentration.

The effect of contact time on the percentage removal of dye is shown under various other fixed operating conditions. The experimental results of adsorption of dye on TSA for various concentrations of dye solution (100, 150, 200 mg/L) with contact time at 305 K were presented in Table 2 and depicted in Fig. 2. It is observed that the percentage removal increased rapidly with an increase in contact time. After 80 minutes no noticeable change in the percentage removal was observed. Equilibrium was established in 40 to 80 min for all the studied concentrations.

3.1.3. Effect of initial concentrations

The study on the effect of initial concentration showed that the percentage of the removal of dyes decreased with the increase of initial concentrations of adsorbate solution as given in Table III and shown in Fig. 2.

The percentage removal of MG dye at the equilibrium was found to decrease from 77.24 to 60.45 as the initial concentration of MG dye increased from 100 mg/L to 200 mg/L. This is because the ratio of available adsorbent surface to the concentration of solute decreases with the increase of initial concentration and hence the percentage of removal decreases with the increase of initial concentration.

However the amount of dye adsorbed on the unit mass of the adsorbent increased with an increase in the initial concentration of the adsorbate solutions. The amount of MB dye adsorbed (mg/g) was found increase from 77.24 to 120.89 as the initial concentration of MB dye increased from 100 to 200 mg/L which is shown in the Table IV.

3.2. Adsorption isotherm

The theoretical amount of dye adsorbed at equilibrium q_e (mg/g), was calculated by the following mass balance relationship.

$$q_e = (c_o - c_e)\frac{v}{w} \quad \text{---------- (1)}$$

Where C_o and C_e (mg/L) are the initial and equilibrium phase concentration of dyes respectively, V the volume of the solution (L) and W is the weight of the TSA (g)

In the present study, Langmuir and Freundlich adsorption isotherm models were used to analyse the experimental data.

3.2.1.Freundlich isotherm

The Freundlich isotherm model is the earliest known equation describing the adsorption process. It is an empirical equation and can be used for non-ideal sorption that involves heterogeneous adsorption. The Freundlich isotherm can be derived assuming a logarithmic decrease in the adsorption with the increase in the fraction of occupied sites and is commonly given the following non-linear equation.

$$q_e = k_f c_e^{1/n} \quad \text{---------- (2)}$$

Where k_f is defined as distribution co-efficient and represents the quantity of dye absorbed onto absorbent for unit equilibrium concentration. C_e is the equilibrium concentration of the dye solution. n indicates the adsorption intensity. Linearised logarthimic form of the Freundlich equation is given below.

$$\log q_e = \log k_f + \frac{1}{n}\log C_e \quad \text{---------- (3)}$$

The applicability of the Freundlich adsorption isotherm was analysed using the experimental data by plotting log q_e versus log C_e. The data obtained from linear Freundlich isotherm plot for the adsorption of the Methylene Blue onto TSA is presented in Table V. The Freundlich isotherm is shown in Fig. 3. The regression coefficient (r^2) values are close to 1, which indicates the perfect fitting of the data.

The values of 'n' are greater than 1, which indicates the favaourability of adsorption with moderate intensity.

3.2.2. Langmuir isotherm

The theoretical Langmuir isotherm is valid for adsorption of a solute from liquid solution as monolayer adsorption on a surface containing a finite number of identical sites. Langmuir isotherm model assumes uniform

energies of adsorption onto the surface without transmigration of adsorbate in the plane of the surface. Therefore, the Langmuir isotherm model was chosen for estimation of the maximum adsorption capacity corresponding to complete monolayer coverage on the adsorbent surface. The Langmuir non-linear equation is

$$q_e = q_e - \frac{Q_m b C_e}{1 + b C_e} \quad \text{---------- (4)}$$

Where C_e is the equilibrium concentration (mg/L) and q_e is the amount adsorbed at equilibrium (mg/L), Q_m is constant which reflect a complete monolayer coverage mgg^{-1}, b is adsorption equilibrium constant (Lmg^{-1}) that is related to the apparent energy of sorption. The linearised form of Langmuir isotherm is

$$\frac{C_e}{Q_e} = \frac{1}{Q_m b} + \frac{C_e}{Q_m} \quad \text{---------- (5)}$$

The linear plots of C_e/Q_e versus C_e suggest the applicability of Langmuir isotherm. The values of Q_m and b were determined from the slope and intercepts of plot. Values of Q_m and b obtained in the present system were given in Table 4. Langmuir Isotherm plot is shown in Fig. 4.

The r^2 values indicate the perfect fitting of data with Langmuir isotherm when compared with Freundlich isotherm. The essential characteristics of Langmuir equation can be expressed in terms of dimensionless separation factor or equilibrium parameter R_L which is defined as $R_L = 1/ (1+bC_i)$ where b is the Langmuir constant and C_i is the initial concentration of the solution. The values of R_L indicate the type of adsorption. The significance of R_L values were presented in Table-VI. The R_L values are found between 0 and 1, which indicates the favorable adsorption of dye for the studied concentration range.

3.3. Kinetics of adsorption

Several steps control the mechanism of adsorption process such as chemical reaction, diffusion control and mass transfer. The kinetics of dye adsorption onto TSA is required for selection of optimum operating conditions for the full-scale batch process. The kinetic parameters which are helpful for the prediction of adsorption rate, give important information for designing and modeling the adsorption processes.

Thus the kinetics of Malachite green onto TSA was analyzed using pseudo first order, pseudo second order and intra-particle diffusion kinetic models.

3.3.1 Test for kinetics models

Best fitting kinetic model for a system can be determined by using the statistical tool percentage of sum of error squares (SSE) (Abechi et al., 2011). This can be evaluated by the following formula; the sum of error squares is given as follows;

$$SSE\ (\%) = \sqrt{\sum[(q_e)_{exp} - (q_e)_{cal}]^2 / N}$$

Where N is the number of data points, (q_e) exp is the experimental q_e and (q_e) cal is the calculated q_e.

3.3.2 Pseudo first order equation

The adsorption kinetic data were described by the Lagergren p ---------- (6) which the earliest known equation is describing the adsorption rate based on the adsorption capacity. The linear form of equation is generally expressed as follows.

$$\ln(q_e - q_t) = \ln q_e - k_1 t$$

Plots of log (q_e-q_t) versus t drawn were shown in fig-5 to 7. The rate constant k_1 and theoretical q_e can be determined from slope and intercept of the plot respectively. Results ---------- (7) Table-VII. The coefficient of regression (r^2) values is not close to 1 .This shows deviatic ler rate.

3.3.3. Pseudo second order equation.

The adsorption kinetic may be described by the pseudo second order model. The integrated and linearised Eq.(8) for the boundary conditions q_t = 0-q_t at t = 0-t equation is given below.

$$\frac{t}{q_t} = \frac{t}{q_e} + \frac{1}{k_2 q_e^2} \quad \text{---------- (8)}$$

If the second order kinetics is applicable then the plot of t/q_t versus t should show a linear relationship. Obtained results were shown in Table VIII. Concerned of plots are shown in Fig-8 to 10 values of k_2 and equilibrium adsorption capacity q_e were calculated from the slope and intercept. Both k_1 and k_2 values found to decrease when initial concentration of dye increased. This indicates that pore diffusion limit the rate of adsorption. The r^2 values infer that data were best fitted with pseudo second order kinetics.

The SSE % values were given in Table VII and VIII. It was concluded that the second order kinetic model was more appropriate rather than the first order kinetic model.

3.4. The intra-particle diffusion model

The adsorbate species are most probably transported from the bulk of the solution into the solid phase through film diffusion followed by intra-particle diffusion process which is often the rate limiting step in many adsorption processes, especially in a rapidly stirred batch reactor. So the intra-particle diffusion is another kinetic model should be used to study the rate of dye adsorption onto TSA. The intra-particle diffusion model is commonly expressed by Webber Morris equation as given below.

$$q_t = k_{dif} t^{1/2} + c \quad \text{---------- (9)}$$

Where c = the intercept
k_{dif} = The intra-particle rate constant in (mg/g/min)
q_t = Quantity adsorption at the time

The values of q_t were found to be linearly correlated with values of $t^{1/2}$ and pore diffusion rate constant k_{diff} directly evaluated from the slope of the regression line. Webber morris plots are shown in Fig.11 to 13

Rate constants for the pore diffusion were given in Table IX. k_{diff} increases with the increase of initial concentration of solution. This indicates that pore diffusion is the rate determining step in this adsorption process.

3.5. Desorption Studies

The results obtained were given as bar diagramme in Fig.14. It is seen that there is no much effect of pH of the desorbing solution. This may be due to physisorption. A maximum of 70 % desorption of methylene blue dye from the spent material was achieved with a solution of pH 12 and minimum of 50 % desorption was achieved with a solution of pH 2.

3.6. FT-IR spectroscopy analysis

FTIR spectrogram of TSA and TSA loaded with MG dye were shown in Fig.15 and 16 respectively. Both FT-IR seems to be similar. No much difference is observed which confirms the physisorption mechanism.

4. Conclusion

Ability of Termite Soil as an adsorbent to remove Malachite green from aqueous solution was investigated. The percentage of removal increased with an increase in contact time and also with dosage of Termite Soil adsorbent (TSA).The adsorption equilibrium data was fitted with Freundlich isotherm and Langmuir isotherm. The r^2 value indicated the best fitting with Langmuir isotherm explaining monolayer adsorption.Freundlich & Langmuir constants were calculated. It reveals,the dimensionless separation factor R_L values indicated the favourable adsorption. The kinetic data were fitted with pseudo first order and pseudo second order equation. The r^2 values indicated the best fitting of pseudo second order equation. Rate constant for pore diffusion indicated that pore diffusion was found to be the rate controlling step. 0.2 N HCl is found to be best desorbing agent among the water, 0.2 N NaOH and 0.2 N HCl solutions.FTIR results revealed the physisorption mechanism.Increase of temperature increased the adsorption.Above result revealed that Termite Soil adsorbent was a potential adsorbent for removal of Methylene Blue dyes.

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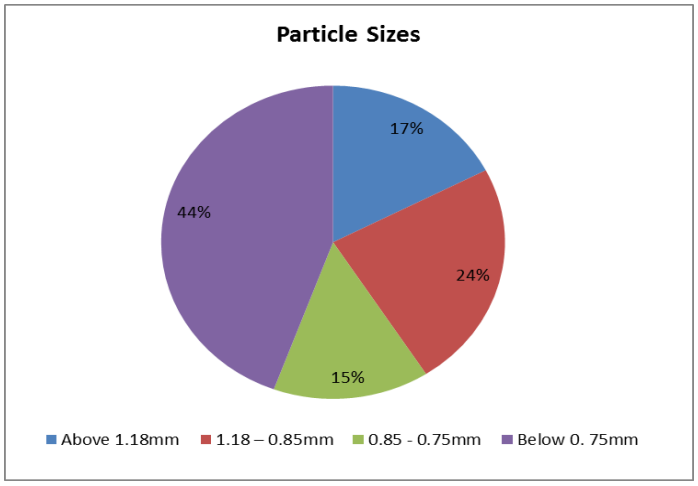
Caption for Figures And Tables.

Flow Chart-I



Figures:

Figure 1.

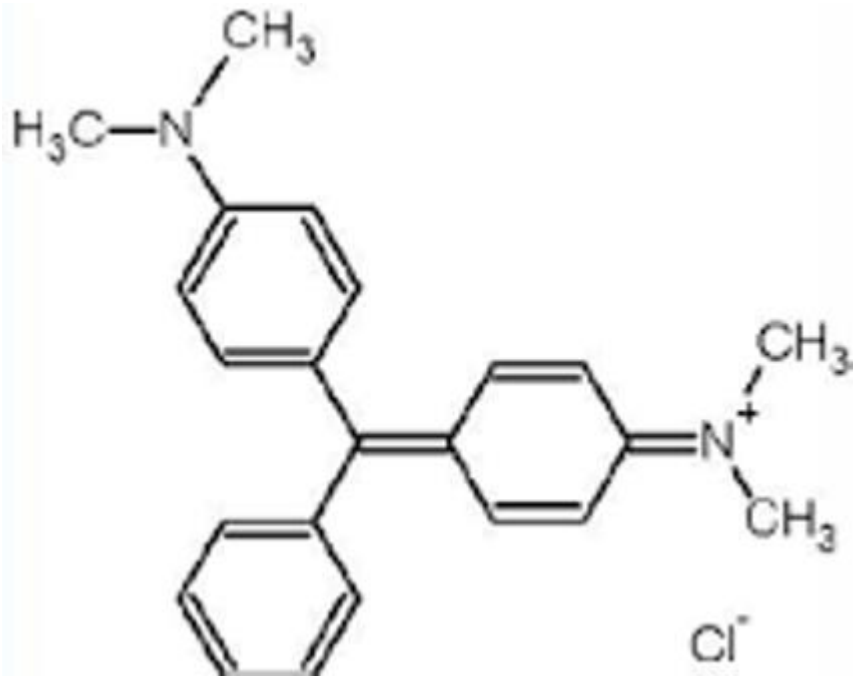
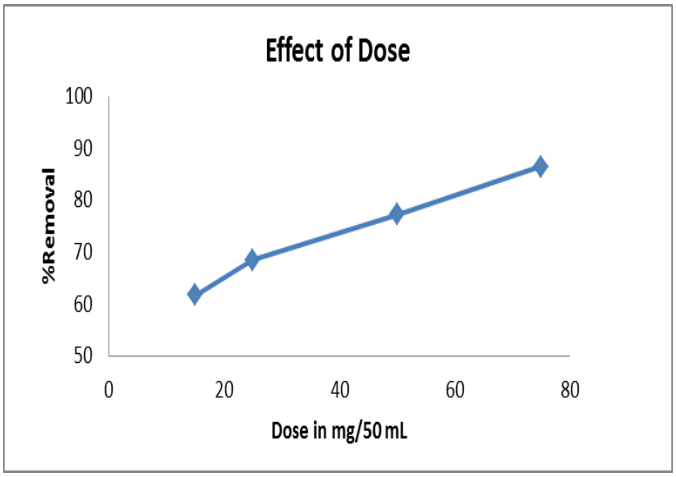


Figure 2.



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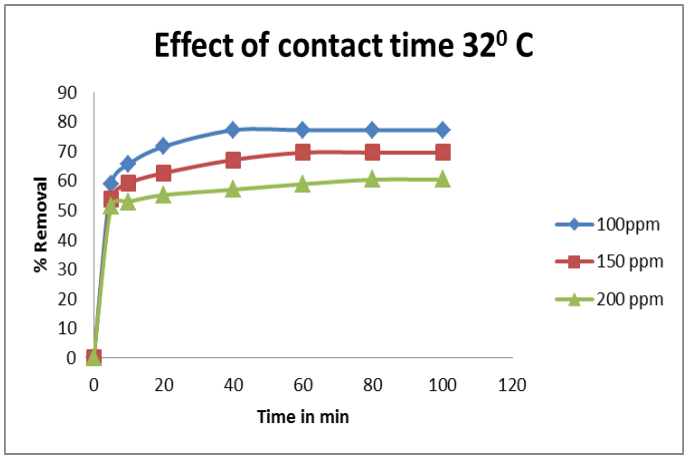

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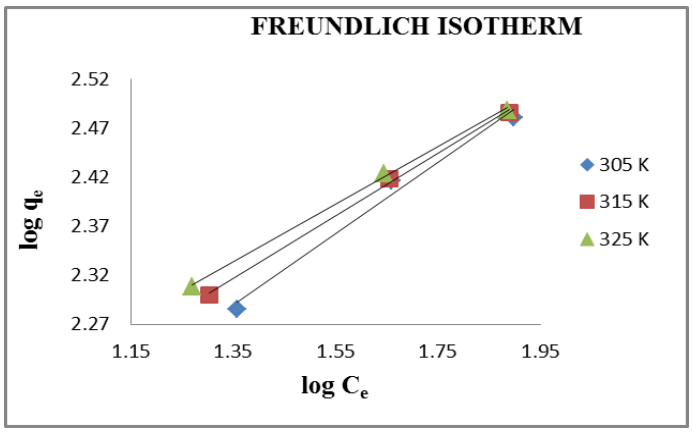

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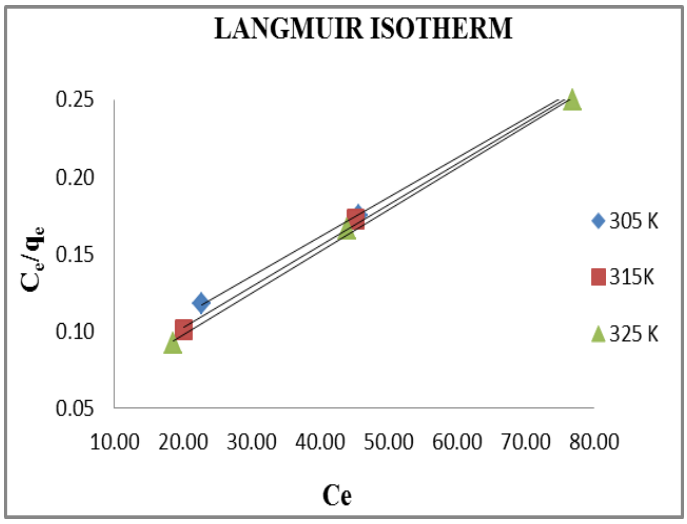

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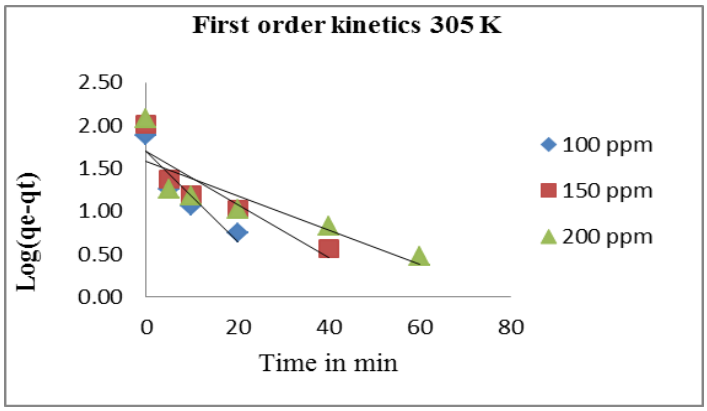

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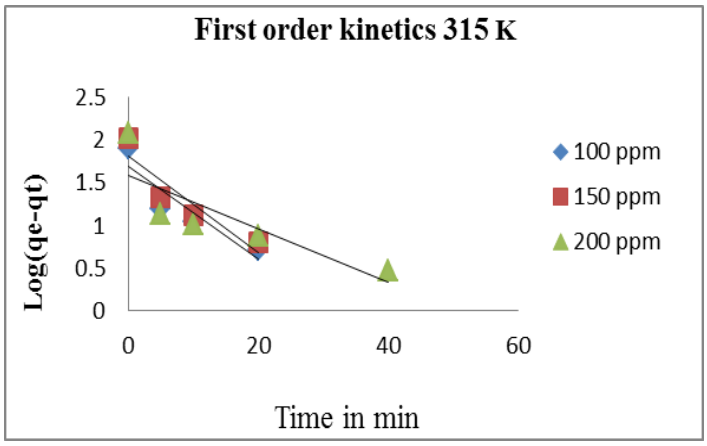

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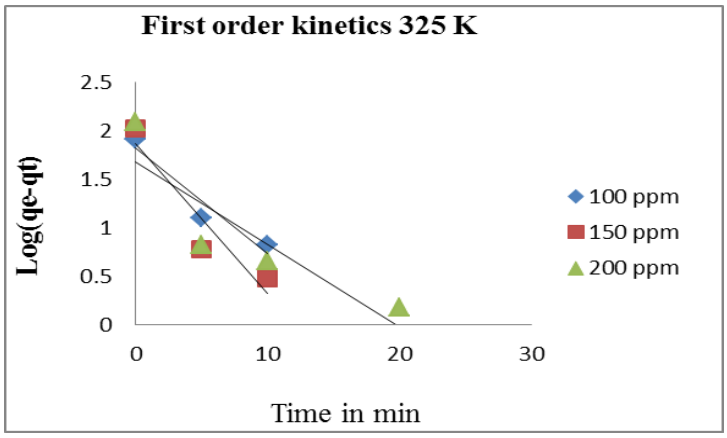

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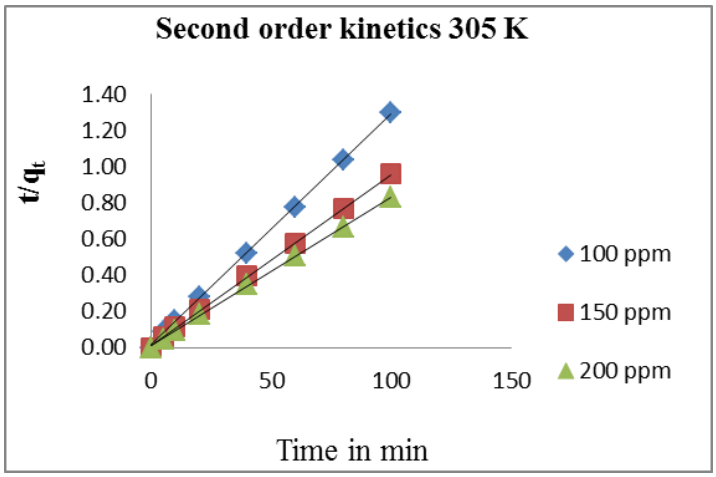

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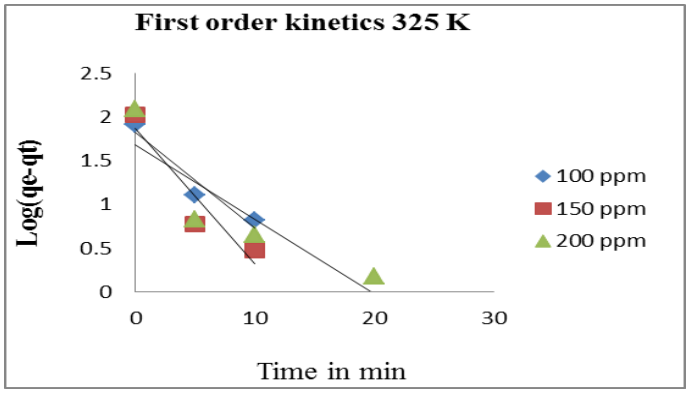


Figure 11

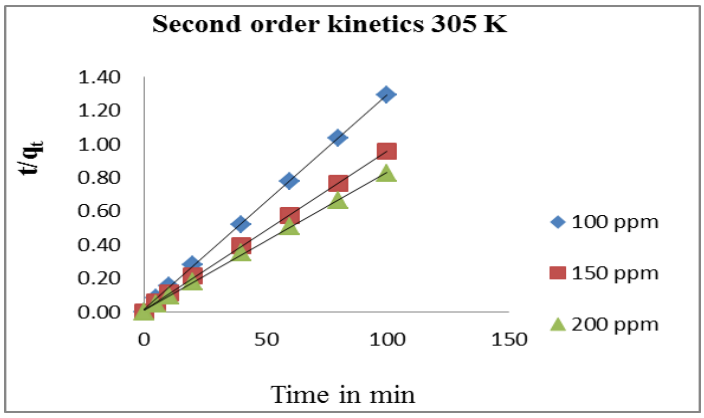


Figure 12

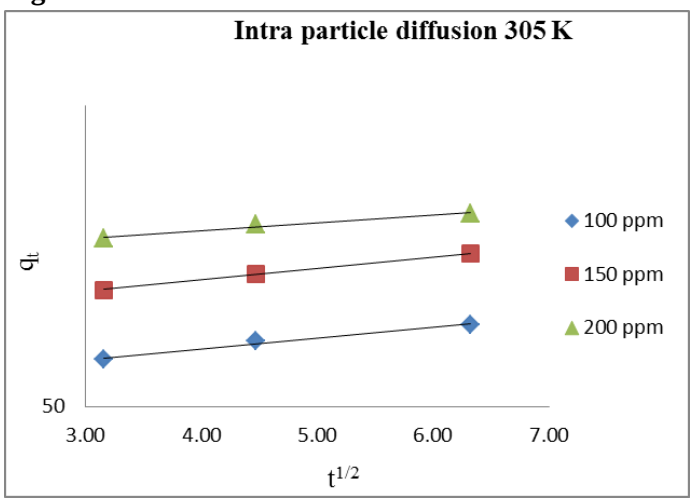


Figure 13

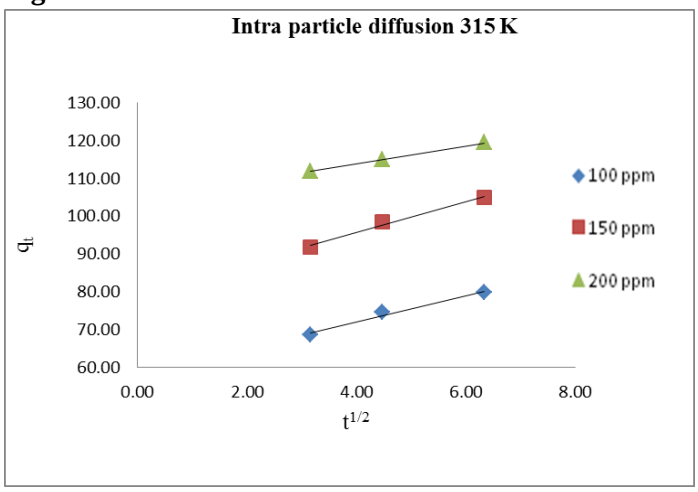


Figure 14.

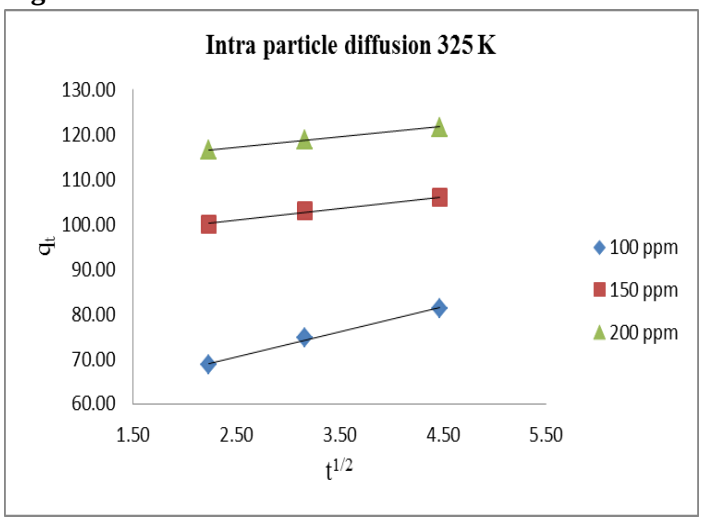


Figure 15.

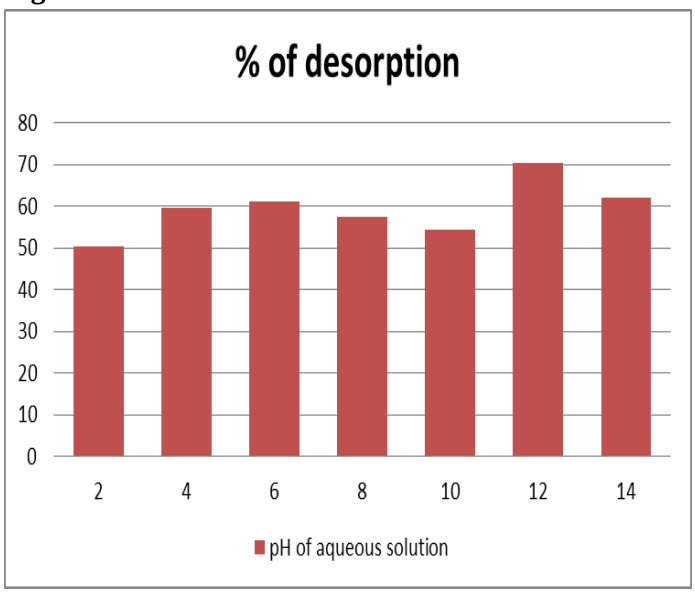


Figure 16.

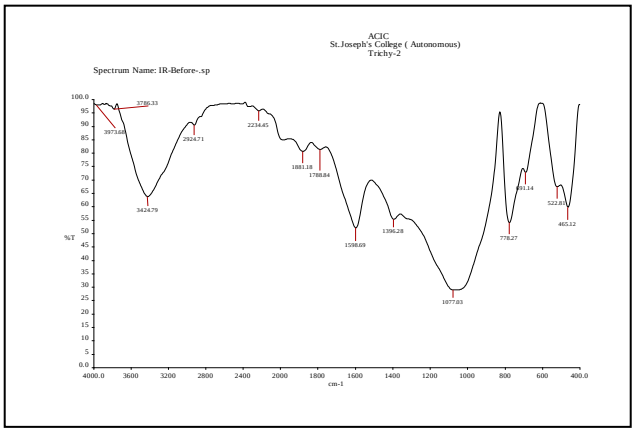
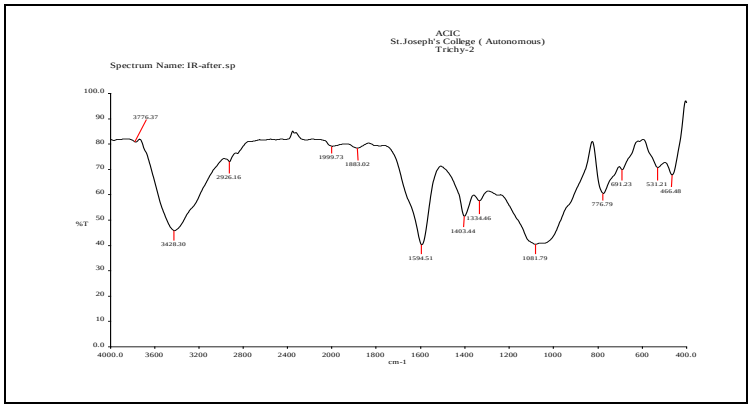


Figure 17.



Tables.

Table I

Particle size (mm)	**%**
Above 1.18	17.4
1.18 – 0.850	23.5
0.850 - 0.75	14.7
Below 0.75	44.4

Table II

Adsorbent Dosage (mg / 50ml)	**% of Removal**
15	64.64
25	72.38
50	77.24
75	82.50

Table III

Time in minutes	% of Removal		
	100 mg/L	150 mg/L	200 mg/L
0	0.00	0.00	0.00
5	58.96	53.73	51.49
10	65.67	59.20	52.99
20	71.64	62.69	55.22
40	77.24	67.16	57.09
60	77.24	69.63	58.96
80	77.24	69.63	60.45
100	77.24	69.63	60.45

Table IV

Time in minutes	Quantity adsorbed (mg/g)		
	100 mg/L	150 mg/L	200 mg/L
0	0	0	0
5	58.96	80.59	102.98
10	65.67	88.80	105.97
20	71.64	94.02	110.44
40	77.24	100.74	114.17
60	77.24	104.44	117.91
80	77.24	104.44	120.89
100	77.24	104.44	120.89

Table V

Langmuir Isotherm							**Freundlich Isotherm**		
Temperature (K)	**Q_m (mg/g)**	**B (L/mg)**	**R_L**			**R^2**	**n**	**k_f (mg/ g)**	**R^2**
			100 mg/L	**150 mg/L**	**200 mg/L**				
305	338.98	0.0500	0.17	0.14	0.14	0.999	2.76	60.117	0.983
315	344.82	0.0591	0.12	0.11	0.10	0.998	3.14	77.090	0.998
325	344.82	0.0674	0.09	0.09	0.09	0.998	3.40	86.297	0.998

Table VI

R_L	**Type of isotherm**
$R_L = 1$	Linear
$0 < R_L < 1$	Favourable
$R_L = 0$	Irreversible

Table.VII

Conc. (mg/g)	**Temp. (K)**	**First order Kinetics**				
		k_1 (1/min)	**$q_{e(cal)}$ (mg/g)**	**$q_{e(exp)}$ (mg/g)**	**R^2**	**SSE %**
100	305	0.0461	48.42	77.24	0.872	9.60
	315	0.1221	50.12	79.85	0.843	9.91
	325	0.1958	168.27	81.34	0.927	28.97
150	305	0.0691	49.66	104.44	0.837	18.26
	315	0.1290	63.97	104.85	0.855	13.62
	325	0.2510	187.50	105.97	0.889	27.17
200	305	0.1198	53.73	120.89	0.737	22.38

	315	0.1300	58.73	122.39	0.694	21.22
	325	0.3362	120.50	123.13	0.790	0.87

Table.VIII

Conc. (mg/g)	Temp. (K)	Second Order Kinetic Parameters				
		k_2 $\times 10^{-4}$ (g/mg.min)	$q_{e(exp)}$ (mg/g)	$q_{e(cal)}$ (mg/g)	R^2	SSE %
100	305	0.006	77.24	83.33	0.999	2.03
	315	0.009	79.85	83.33	0.999	1.16
	325	0.029	81.34	83.33	0.999	0.66
150	305	0.007	104.44	111.11	0.999	2.22
	315	0.011	104.85	111.11	0.999	2.09
	325	0.081	105.97	111.11	1.000	1.71
200	305	0.009	120.89	125.00	0.999	1.37
	315	0.014	122.39	125.00	0.999	0.87
	325	0.064	123.13	125.00	1.000	0.62

Table IX

		Intra Particle Diffusion	
		k_p (mg/g.min)	R^2
Conc. (mg/g)	**Temp. (K)**	2.33	0.986
	315	2.36	0.981
	325	2.56	0.995
150	305	2.64	0.999
	315	3.49	0.986
	325	3.62	0.990
200	305	3.77	0.977
	315	4.08	0.999
	325	5.63	0.999