Adsorption of Direct Blue 5 Dye by Activated Carbon as Adsorbent - Modeling and Kinetics

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"Abstract"

In this study, a study has been carried on adsorption of direct black dye which is used in textile industries of Sanganer (Jaipur). The amount adsorbed on granular activated carbon at equilibrium was measured and the equilibrium data were tested for Langmuir Frendlich, Temkin and Dubinin- Radushkevich isotherms for their applicability. The experimental data satisfied Langmuir, Freundlich and Temkin adsorption isotherm having correlation coefficients R^2 > 0.98. The maximum monolayer coverage from Langmuir isotherm was 17.51 mg/g. The heat of adsorption process was estimated from Temkin isotherm to be 3.796 J/mol. The calculations of thermodynamic parameters indicated that the adsorption is spontaneous and exothermic. The adsorption is found to increase with increase in temperature. The adsorption studies with granular activated carbon fit the second order kinetic model with R^2= 0.9997.

Keywords: isotherm, kinetics, adsorption, equilibrium.

"1.Introduction"

There is practically no human activity that does not produce waste products and in addition there is a direct relationship between the standard of living in a society or country and the amount of waste products produced. Approximately, 23% of the world's population live in developed countries, consume 78% of the resources and produce 82% of the waste products. In addition, it has to be pointed out that the volume of residual waste increases in an exceptional way with regards to a country's level of industrialization. Textile mills are major consumers of water and consequently one of the largest groups of industries causing intense water pollution. The extensive use of chemicals and water result in generation of large quantities of highly polluted wastewater. Textile processing employs a variety of chemical, depending on the nature of the raw material and products. It is estimate that about 10% are lost in industrial wastewater. The wastewater generated by the different production steps (i.e. sizing of fibers, scouring, desizing, bleaching, washing, mercerization, dyeing and finishing) has high pH and temperature. It also contains high concentration of organic matter[1], non-biodegradable matter, toxic substances, detergents and soaps, oil and grease, sulfide, soda and alkalinity. Textile industry is confronted with the challenge of both color removal for aesthetic reasons and effluent salt content. Textile processing is one of the most important industries in the world and it employs a variety of chemicals, depending on the nature of the raw material and product some of these chemicals are different types of enzymes, detergents, dyes, acids, sodas and salts. The Ministry of Environment and Forests, Government of India has prohibited the handling of benzidine based dyes vide the notification published in the Gazette in January, 1990. As per this notification; handling of all the 42 benzidine based dyes are prohibited from 1993 onwards (Central Coir Research Institute notice Dated 15/10/2007) is given as Table-1.

Table 1
List of banned dyes

Name of dye	Name of dye	Name of dye
Acid Orange 45	Acid Red 85	Acid Black 29
Acid Black 94	Azoic Diazo Compo.112	Azoic Diazo Compo.112
Direct Yellow 24	Direct Orange 1	Direct Orange 8
Direct Red 1	Direct Red 10	Direct Red 13
Direct Red 17	Direct Red 28	Direct Red 37
Direct Red 44	Direct Violet 1	Direct Violet 12
Direct Violet 22	Direct Blue 2	Direct Blue 6
Direct Green 1	Direct Green 6	Direct Green 8
Direct Green 8:1	Direct Brown 1	Direct Brown 1:2
Direct Brown 2	Direct Brown 6	Direct Brown 25
Direct Brown 27	Direct Brown 31	Direct Brown 33
Direct Brown 51	Direct Brown 59	Direct Brown 79

Adsorption has been used extensively in industrial process for separation and purification[2-4]. The removal of colored and colorless organic pollutants from industrial wastewater is considered as an important application of adsorption processes. At the present, there is a growing interest in using low cost, commercially available materials for the adsorption of dyes. Many studies have been conducted to evaluate adsorption of dyes onto a wide range of natural and synthetic, organic and inorganic adsorbents. The main adsorbents used for dye adsorption are activated carbon, clay, wood chips, fly ash etc. The adsorption treatment required less investment cost, easy to operate and is insensitive to toxic pollutants. The adsorption treatment is applicable to all types of dyes and has low regeneration cost[5]. The disadvantages of adsorption treatment include that some adsorbent requires activation and adsorption capacity is affected by changing temperature and pH[6]. The adsorptive methods of decolorization use a sorbet medium that physically removes the dye ions and other contaminants from the effluent through physicochemical process of adsorption.

Activated carbon is the most commonly used method of dye removal by adsorption and is very effective for adsorbing cationic, mordant, and acid dyes and to a slightly lesser extent, dispersed, direct, vat, pigment and reactive dyes[7]. Activated carbon can be produced by heating a raw material like wood, lignite or coal in an atmosphere of CO_2, CO, O_2 or H_2O. It readily adsorbs most dissolved organic compounds because crushed carbon has a large surface area due to a large number of pores and this large surface area is effective in adsorbing the organic compounds. It is ineffective in removing disperse; vat and pigment dyes from their pure solutions Activated carbon adsorbents are applicable within a wide range of pH. It has got porous texture which gives it a large surface area. Its chemical nature can easily be modified by chemical treatment in order to increase its properties. However, it is very expensive.

"2.Materials and Methods"

2.1 Characterization of Dye

The dye used in the study is Direct Blue 5 being utilized in textile industry of Sanganer. The characteristics of dye have been summarized as Table-2.

Table 2
Characterization of direct blue

Particulars	**Result**
pH of 1% solution	7.98
Purity,%	88.76
λ_{max}	566 nm

The granular activated carbon of CDH make was used for the adsorption studies. The specifications of granular activated carbon are summarized as Table-3.

Table 3
Specifications of GAC (CDH)

Particulars	**Granular activated carbon**
pH	8.67
Moisture, %	5%
Ash, %	2.5
Bulk density, g/mL	408
Loss on drying, %	10
Arsenic, %	0.0005
Iron, %	0.05
Zinc, %	0.15

2.2 Experimental procedure

The stock solution of blue dye of 1000 mg/L concentration was prepared and diluted to desired concentration of 62.5, 125, 250 and 500 mg/L. The desired dosage of activated carbon was added to 100 ml dye solution and adsorption treatment was carried out in 250 ml conical flasks. The study was undertaken at different stirring time, dosages and temperature. The samples for analysis was then filtered with Whatman No.1 filter paper and analyzed for remaining concentration of dye with the help of UV-VIS spectrophotometer. The activated carbon was washed with distill water before addition to dye solution as it was found that the color leached in the first wash. It was washed again till no color leach in the wash.

2.3 Analysis of Maxima of Blue 5 Direct dye

The maximum absorbance of Blue 5 direct dye was determined with the help of UV-VIS spectrophotometer. The wavelength corresponding to maximum absorbance of dye gives the maxima of the dye. The absorbance of different concentration of blue dye solution was determined at its maxima and standard graph was prepared for further studies.

2.4 Adsorption isotherm models

One of the initial models for the adsorption of a species onto a simple surface was put forth by Irving Langmuir in 1916. Langmuir assumed that a surface consists of a given number of equivalent sites where a species can physically or chemically stick. Physical adsorption through Vander Waals interactions is called physisorption, whereas chemical adsorption through the formation of a covalent bond is called chemisorption. It is important to realize that the processes of adsorption and the opposite process (desorption) is dynamic; a rate law can be written for each process, and when the rates become equal an equilibrium state will exist characterized by a constant fractional coverage of the original sites Isotherm curves are the curves plotted at constant temperature that describe the equilibrium state of a process.

“3.Results and Discussion”

3.1 Characteristics of Effluent of Textile industry

Waste water sample collected from Sanganer Area from the outlet of textile industry and analyzed for pollution parameters. The recipe of dye as reported by industry person was direct dye (Red 5 and Blue 5 dye in ratio 1:5). The effluent characteristics of Sanganer area obtained from one of the industrial units is given in Table-4.

Table 4
Characteristics of effluent

Parameter	Value
Chemical oxygen demand, mg/L	3000
pH	10.46
Dissolved oxygen, mg/L	5.07
Total solids, mg/L	12,380
Total suspended solids, mg/L	120
Color, Pt-Co Unit	530
Flow rate, m^3/h	6.0

3.2 Effect of various initial concentration of dye solution

1g of activated carbon was added to blue dye solution of various initial concentrations ranging from 62.5 mg/L to 500 mg/L at room temperature (30 ºC) for different contact time. The samples were then collected at different time intervals and analyzed for its concentration. The effect of fly ash as adsorbent at various initial concentrations is shown in Figure-1.

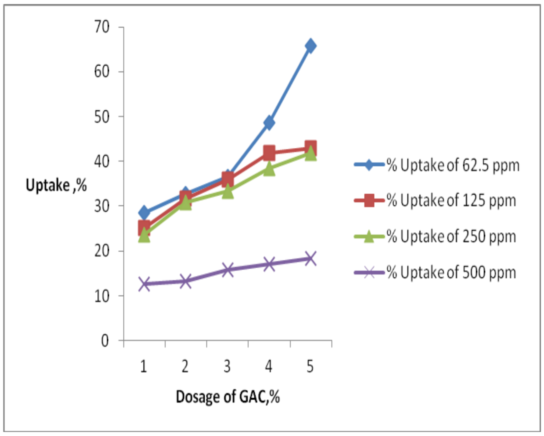

Figure 1. Uptake of blue dye of various initial concentrations

3.3 Effect of Stirring time on Color removal Efficiency of Adsorbent

The contact time between the pollutant and the adsorbent is of significant importance in the wastewater treatment by adsorption. A rapid uptake of pollutants and establishment of equilibrium in a short period signifies the efficacy of that adsorbent for its use in wastewater treatment. 1g of activated carbon was added to Blue 5 dye solution of 62.5 mg/L concentration and stirred for different time periods. The uptake was determined at different time as shown in Figure 2.

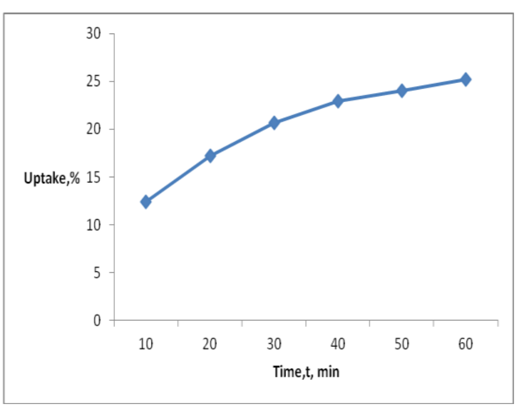

Figure 2. Effect on % color removal with time

It can be observed from Figure 2 that as the stirring time increases from 10 min to 60 min the color removal increases from 12.38 % to 25.18 %.

3.4 Effect of retention time on color removal efficiency of adsorbent

1 g of activated carbon was added to 100 ml of Blue 5 dye solution of 62.5 mg/L concentration and stirred for 60 minutes, the stirring was stopped and then blue dye was given retention with activated carbon and samples withdrawn after each hour to study effect of retention time on color reduction. The results for Blue 5 dyes are presented in Figure 3.

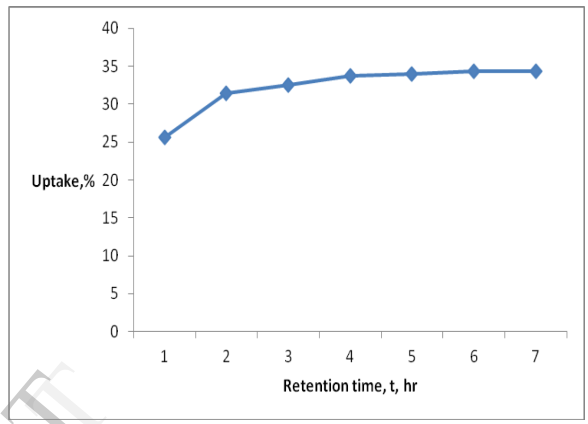

Figure 3. Retention of dye with time

The retention increased color removal from 25.58 % to 34.30 % as shown in Figure 3 in one hour period only.

ASORPTION ISOTHERMS

Adsorption isotherm[8-10] describes the relation between the amount or concentration of adsorbate that accumulates on the adsorbent and the equilibrium concentration of the dissolved adsorbate. Equilibrium studies were carried out by agitating a series of beakers containing 100 mL of Blue 5 dye solutions of initial concentration 62.5 mg/L with 1 g of granular activated carbon at 30^0 C with a constant agitation. Agitation was provided for 7.0 h, which is more than sufficient time to reach equilibrium. The equilibrium can be seen from the curve in Figure 3.

Formula used for calculation of q_e is as follows:

$$q_e = \frac{C_0 - C_e}{m} \times V \quad \ldots\ldots\ldots\ldots\ldots\ldots..1$$

3.5 Langmuir Isotherm

The Langmuir equation or Langmuir isotherm or Langmuir adsorption equation relates the coverage or adsorption of molecules on a solid surface to gas pressure or concentration of a medium above the solid surface at a fixed temperature.

The following relation can represent the linear form of the Langmuir isotherm model:

$$\frac{1}{q_e} = \frac{1}{q} + \frac{1}{bqC_e} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots 2$$

Here, q_e is the amount adsorbed at equilibrium (mg/g), C_e the equilibrium concentration of the adsorbate (mg/L), and q (mg/g) and b (L/mg) are the Langmuir constants related to the maximum adsorption capacity and the energy of adsorption, respectively. These constants can be evaluated from the intercept and the slope of the linear plot of experimental data of $1/q_e$ versus $1/C_e$. Conformation of the experimental data into Langmuir isotherm model indicates the homogeneous nature of activated charcoal surface, i.e. each dye molecule/ activated charcoal adsorption has equal adsorption activation energy. The results also demonstrate the formation of monolayer coverage of dye molecules on the outer surface of fly ash.

Table 5
Adsorption Capacity

C_o, mg/L	Temp-30⁰ C		Temp-40⁰ C		Temp-50⁰ C	
	Ce, mg/L	q_e, mg/g	Ce, mg/L	q_e, mg/g	Ce, mg/L	q_e, mg/g
62.5	41.0	2.127	39.2	2.51	38	2.590
125	84.9	3.571	82	4.07	80	4.5
250	182.1	6.666	179.4	7.20	175	8.40
500	399.4	9.090	391.9	10.28	389	11.1

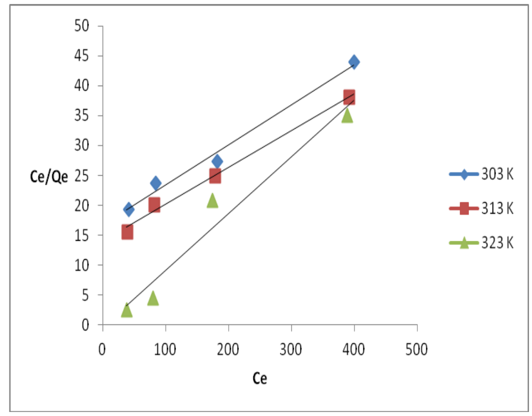

Figure 4 Langmuir Isotherm for Blue 5 dye

The maximum adsorption capacity at various initial concentrations and effect of fly ash at different temperature of fly ash is given from Table- 5 and Figure-4 respectively.

3.6 Freundlich Adsorption Isotherm

The linear form of the Freundlich isotherm model is given by the following equation:

$$\ln q_e = \ln K_f + \frac{1}{n} C_e \quad \ldots\ldots\ldots\ldots\ldots\ldots 3$$

where K_F (mg/g) (L/mg) $^{1/n}$ and 1/n are Freundlich constants related to adsorption capacity and adsorption intensity of the absorbent, respectively. Linear plot of ln q_e versus ln C_e shows that the adsorption of direct Blue 5 dye from an aqueous solution on fly ash also follows the Freundlich isotherm. Values of K_F and n are calculated from the intercept and slope, respectively. If n value is $1 < n < 2$, it suggests that the dye adsorption on fly ash is favorable. The value of n is given as Table-6 which is more than 1 and less than 2 which shows the dye adsorption by fly ash as favorable.

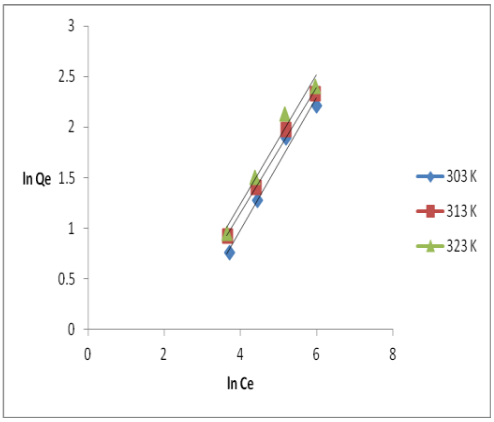

Figure 5. Freundlich Isotherm of Blue 5 dye

The predicted Langmuir and Freundlich isotherm equations for Blue 5 at room temperature are given as equation 2 & 3 respectively.

3.7 Temkin Isotherm

The model assumes that the heat of adsorption which is function of temperature in the layer of molecules decrease linearly rather than logarithmic with coverage. This model is given by following equations:

$$qe = \frac{RT}{b}\ln(A_T C_e) \text{..........................4}$$

$$qe = \frac{RT}{b_T}\ln A_T + \left(\frac{RT}{b}\right)\ln Ce \text{.............5}$$

$$B = \frac{RT}{b_T} \text{.......................................6}$$

$$qe = B\ln A_T + B\ln Ce \text{.......................7}$$

A_T=Temkin isotherm equilibrium binding constant (L/kg)
b_T=Temkin isotherm constant

Temp,K	Max sorption capacity (q_0) mg/g	K_L L/mg	C_0, mg/L	R_L	R^2
		Langmuir			
303	14.9	0.0039	62.5	0.8	0.986
			125	0.66	
			250	0.5	
			500	0.33	
313	16.2	0.0044	62.5	0.78	0.993
			125	0.64	
			250	0.47	
			500	0.31	
323	17.5	0.0046	62.5	0.77	0.986
			125	0.63	
			250	0.46	
			500	0.30	

	Freundlich Isotherm		
Temp,K	1/n	*K* (mg/g) (L/g)^n	R^2
303	0.655	0.194	0.981
313	0.622	0.263	0.991
323	0.641	0.266	0.974

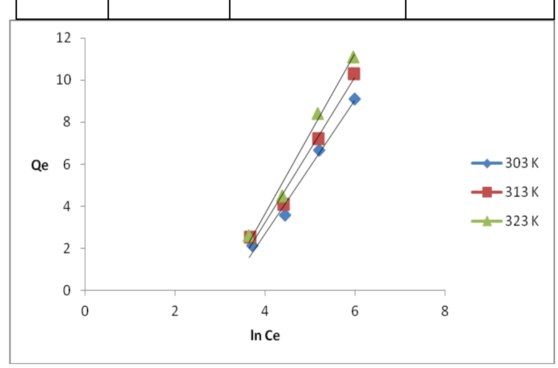

Figure 6. Temkin isotherm of blue dye

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3.8 Dubenin and Radushkevich isotherm

The equation of Dubenin and Radushkevich isotherm is given as equation 8 & 9 respectively.

$$qe = (qs)\exp\left(-B\varepsilon^2\right) \ldots\ldots\ldots\ldots\ldots\ldots\ldots 8$$

Where q_s is D-R constant

$$\varepsilon = RT\ln\left(1+\frac{1}{Ce}\right) \ldots\ldots\ldots\ldots\ldots\ldots\ldots 9$$

B is a constant which gives value of free energy E of adsorption per molecule of adsorbate when solids are transferred from infinity in solution.

$$E = \frac{1}{2B^{1/2}} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 10$$

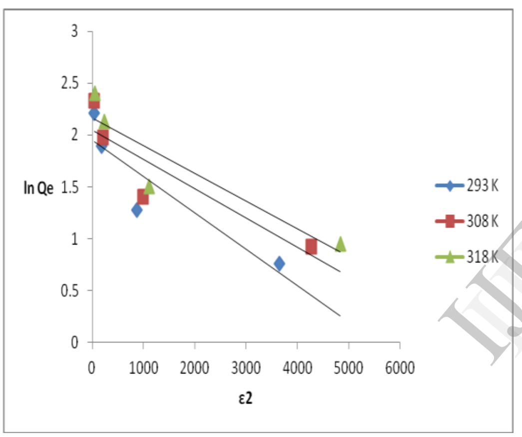

Figure 7. Dubinin-Radushkevich isotherm of blue dye

Table 6. (a)
Predicted Isotherms

Table 6(b)
Predicted Isotherms

Table 6(c)
Predicted Isotherms

Dubinin-Radushkevich Isotherm			
Qs (mg/g)	$K_{ad,}$ (mol^2/KJ^2)	E	R^2
7.016	0.0003	0.188	0.82
7.750	0.0003	0.179	0.80
8.694	0.0003	0.169	0.83

From the Figure- 4, 5, 6 & 7 and Table-6 respectively, it was observed that the Langmuir isotherm best fits the data. The lower correlation coefficient for Dubinin-Radushkevich isotherm and higher correlation coefficient of Freundlich, Temkin and confirms the non-applicability of Dubinin-Radushkevich isotherm for the direct Blue 5/GAC systems. The very much higher correlation coefficient of 0.9865, 0.9933 & 0.9866 for the Langmuir isotherm at 303, 313 and 323 K predicts the monolayer coverage of Blue 5 dye on activated carbon particles. From Table-6, it can be seen that the maximum sorption capacity q_0 (mg/g) of activated carbon for Blue 5 was 17.51 mg/g at 323 K.

3.9 Thermodynamic parameters

Thermodynamic parameters[11] were evaluated to confirm the adsorption nature of the present study. The thermodynamic constants, free energy change, enthalpy change and entropy change were calculated to evaluate the thermodynamic feasibility and the spontaneous nature of the process.

Enthalpy change (*ΔH*), and entropy change (*ΔS*), may be determined from Van't Hoff equation:

$$\ln K = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \ldots\ldots\ldots\ldots\ldots 11$$

By plotting ln K as ordinate and 1/T as abscissa, we will get ΔS, ΔH and by using the following equation. We can get the value of have ΔS, ΔH.

And by this equation, get the value of *ΔG*.

$$\Delta G = \Delta H - T\Delta S \ldots\ldots 12$$

Tem, K	Temkin isotherm		
	A_T (L/mg)	B	R^2
303	0.0432	3.16	0.985
313	0.0469	3.44	0.983
323	0.0480	3.79	0.987

Where, *ΔG* is the free energy change (kJ mol^{-1}), *R* is the universal gas constant (8.314 J mol^{-1} K^{-1}), K the thermodynamic equilibrium constant and T is the absolute temperature (K).

$$\Delta G = \Delta H - T\Delta S = -RT \ln K_c \quad ....13$$

$$\ln K_c = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad ....14$$

$$2.303 \log \frac{q_e}{C_e} = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad ....15$$

$$\log \frac{q_e}{C_e} = \frac{\Delta S}{R \times 2.303} - \frac{\Delta H}{RT \times 2.303}$$

The values of *ΔS, ΔH, ΔG* was obtained from a plot of log (q_e/C_e) vs. *1/T*. The plot of log (q_e/C_e) vs. *1/T* is shown in Figure-8.

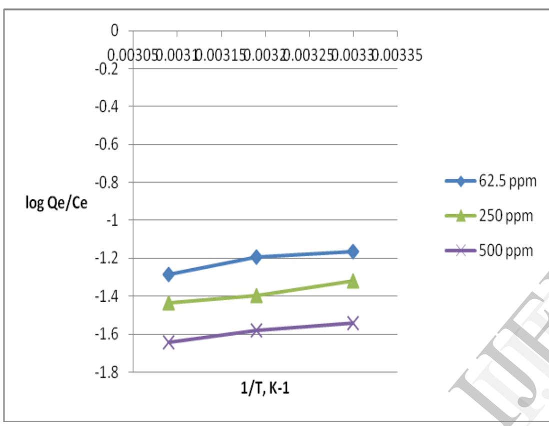

Figure 8. Graph of log (q_e/C_e) vs. 1/T

Table 7
Thermodynamic Parameters

Co, mg/L	Temperature, K	ΔG, KJ/mol	ΔH, KJ/mol	ΔS, KJ/mol
62.5	303	6.527	-10.744	-0.057
	313	7.097		
	323	7.667		
125	303	6.94	-11.543	-0.061
	313	7.55		
	323	8.16		
250	303	7.375	-10.805	-0.060
	313	7.975		
	323	8.575		
500	303	8.684	-8.89	-0.058
	313	9.264		
	323	9.844		

Heat of reaction (*-ΔH)* for physical adsorption is reported to be 4.2 to 63 kJ/ mol in literature (Fogler, 1997). The value of *–ΔH range from -8.89 to -11.543* kJ/mol from Table-7 which indicate that the nature of adsorption of Blue 5 Direct dye on GAC is physical adsorption.

The negative value of ΔH & ΔG indicate exothermic and spontaneous process of adsorption of blue dye on granular activated carbon respectively.

3.10 Kinetic parameters

(a) Pseudo First Order Reaction

The adsorption kinetics[12] can be described by a pseudo first order equation as suggested by Lagergren:

$$\frac{\partial q}{\partial T} = K_1 (q_e - q) \quad ....16$$

Where, K_1 (min^{-1}) is the rate constant of the pseudo-first order adsorption,

q (mg/ g) denotes the amount of adsorption at time t (min), and

q_e (mg/ g) is the amount of adsorption at equilibrium.

After definite integration by application of the conditions t = 0 to t = t and q = 0 to q = q_e, becomes

$$\ln(q_e - q) = \ln q_e - K_1 t \quad ....17$$

The adsorption rate constant, K_1, can be experimentally determined by the slope of linear plots ln (q_e – q) vs. t given in Figure-9.

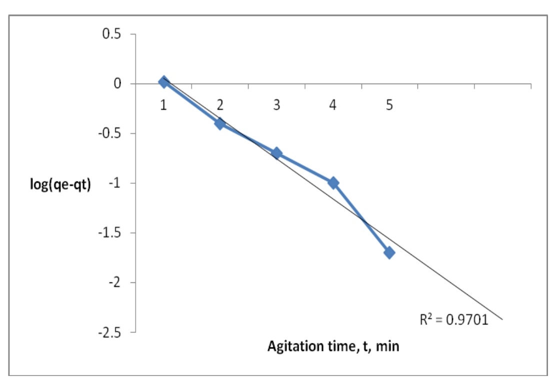

Figure 9. Pseudo first order kinetics for Blue 5 dye

Values of Langmuir adsorption rate constant (k_f) were determined from the plot of log (q_e - q_t)

against t. These values indicate that the adsorption rate was very fast at the beginning of adsorption.

K_t = 0.9285 h^{-1}
q_e = 4.199 mg/g of adsorbent

(b)Pseudo Second Order Kinetics

The pseudo-second order equation:

$$\frac{\partial q}{\partial T} = k_2 (q_e - q)^2 \qquad \ldots\ldots\ldots\ldots\ldots\ldots 18$$

Integrating, for the boundary conditions t = 0 to t = t and q = 0 to q = q_e gives

$$\frac{1}{q_e - q} = \frac{1}{q_e} + k_2 t \qquad \ldots\ldots\ldots\ldots\ldots\ldots 19$$

which has a linear form of k_2 and q_e can be obtained from the intercept and slope of plotting 1/ (q_e-q) vs. t given in Figure-10.

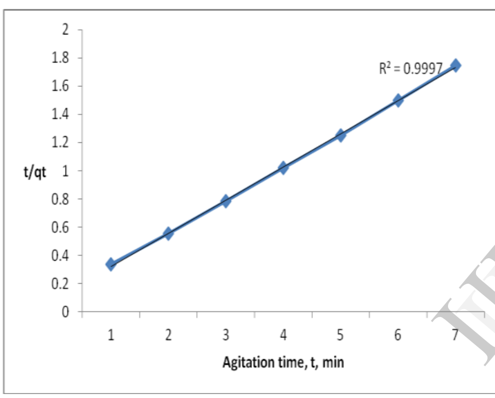

Figure 10. Pseudo second order kinetics

The comparative table of kinetics of adsorption of blue dye by fly ash showed it second order kinetics based on higher correlation coefficient given in Table-9.

Table 9
Kinetics

Pseudo I^{st} order kinetics			Pseudo II^{nd} order kinetics		
Qe, mg/g of adsorb ent	**k_1,h^{-1}**	**R^2**	**Qe, mg/ g**	**k_2, h^{-1} (mg/g of adsorbe nt)$^{-1}$**	**R^2**
4.199	0.92 85	0.97 01	4.2 57	0.6235	0.99 97

"4.Conclusions"

The efficiency of granular activated carbon for removing blue dye in batch studies was studied. The experiments were conducted with different initial concentration of blue dye solution. The values of R_L were found to be in the range of 0 to 1 clearly indicating that the adsorption process is favorable for granular activated carbon. The freundlich adsorption constant, 1/n should be between 0.1 to 1 for better adsorption. It has been found that the isotherms well fit with Langmuir constant of 0.039 L/mg at 30^0C and Langmuir constant of 0.0046 L/mg at 50^0C. The monolayer adsorption capacity of granular activated carbon was found to be 17.5 mg/g. The kinetics of removal of Blue 5 dye on GAC has been studied and it was found that the adsorption of Blue 5 dye on GAC followed pseudo second order kinetics and the value of pseudo second order rate constant has been found to be 0.6235 h^{-1}(mg/g of absorbent)$^{-1}$. Thermodynamic studies were also made for adsorption of Blue 5 dye on GAC and value of Heat of reaction (ΔH), Gibbs free energy (ΔG) and Entropy (ΔS) have been determined.
It has been concluded on the basis of the value of heat of reaction that the nature of adsorption of Blue 5 dye on GAC is physical-adsorption. The value of ΔG is slightly above the physical-adsorption value and is in the range of beginning of chemisorptions.

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