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Research Article

ADSORPTION OF CHROMIUM (VI) FROM AQUEOUS SOLUTION BY USING MULTANI MITI

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ABSTRACT

The urbanization rate in India is very fast. It has increased from 10.84 to 28.5% up to 2001. Unregulated growth of urban area cause many problems. Due to increase in population, urbanization and industrialization many waste materials are entering into waste water streams and water streams causing dreadful diseases. One of the most toxic chemical which is entering into the surface water is hexavalent chromium. The hexavalent chromium compounds are manmade and they are widely used in different types of industries such as paint manufacturing industries, chromium plating industries and smelting industries. The waste water from those industries is discharged into natural water bodies without any treatment are changing the quality of water. Taking that factor into consideration the present study deals with the controlling technology of chromium (VI) by batch adsorption technique using multani mitti. The study is carried out with respect to contact time, concentration, dosage, effect of pH and temperature. The experimental data tested with adsorption isotherm and kinetics studies.

INTRODUCTION

Heavy metals most common pollutants found in industrial waste water. Heavy metals enter into the environment due to bioaccumulation. These metals are highly toxic and disturb the ecosystems. One such heavy metal is chromium. Generally, Chromium widely used in tannery, leather, paint manufacturing and metal plating industries. Chromium (VI) is highly toxic and leads to carcinogenic disorders. Hence it is very important that chromium should be removed from industrial waste water before being discharged in natural water bodies and aquatic environment[1].

Several commercial methods are available for the removal of heavy metal pollutants from waste water. These methods are including chemical precipitation, electroplating, chemical coagulation, ion exchange, membrane separation etc. However these methods are expensive and it may produce large amount of sludge. Adsorption of heavy metals such as chromium (VI) is an efficient technology in industrial waste water treatment. This method is cheap and applicable for small scale industries. The present studies determine the effectiveness of multani mitti. It was used to remove chromium (VI) from waste water, by performing batch adsorption. The present investigation was aimed to study the effect of pH, contact time, initial concentration of chromium, multani matti dosages and temperature[2]. To avoid the other metals interference the removal of chromium carried out by synthetic waste water along with paint manufacturing waste water.

METHODS AND MATERIALS

Selection of adsorbent

Multani mitti is selected as an adsorbent for the present study. Using multani miti as an adsorbent work has been done on Tajmahal[3-5]. Multani Mitti is known as montmorillonite and it contains grains of sand particles[6]. They contain complex multi-center crystalline structures of oxides and hydroxide of magnesium, aluminum, Zinc and silicon and it is known as fullerene mud and rich in lime[7].

Taking all these factors into consideration multani mitti is selected as an adsorbent[8]. The present work, examines the possibility of using a well-known physicochemical method like adsorption for the removal of chromium from the water. The initial screening studies have been carried by introducing a known amount of adsorbent into the aqueous solution of chromium. Multani mitti was used as adsorbents for controlling chromium in water.

Preparation of Cr (VI) solution

A stock solution of Cr (VI) (0.35g/100ml) was prepared by dissolving appropriate quantity of AR grade $K_2Cr_2O_7$ in 100 ml of distilled water from Millipore purification unit. The stock solution was further diluted with distilled water to desired concentration for obtaining the test solutions. The initial metal ion concentrations ranged from 10 mg to 85 mg/ml were prepared. Final residual metal concentration after adsorption was directly measured by spectrophotometer.

The adsorption experiments are carried out with respect to Contact time between adsorb ate and adsorbent, effect of Chromium concentration, effect of adsorbent dosage, effect of pH and temperature.

RESULTS AND DICUSSION

Effect of Contact time between Chromium and Multani miti

The experimental set up measures the effect of contact time on the batch adsorption of Cr (VI) an initial concentration of 10 mg/L, 35 mg/L and 85 mg/L is maintained. Contact time required for the maximum removal of Cr (VI) by multani mitti is represented by figure-1. There is a steep increase in the adsorption in the initial stages of the reaction as seen in the curve[9]. Maximum adsorption occurs at the 20th minute for solution which having initial concentrations 10 mg/L, 35 mg/L and maximum adsorption occurs at 10^{th} minute for solution which having higher concentration(85 mg/L). After 10 -20 minutes there is no adsorption change was observed. The time variation curve is smooth and continuous and it indicates the formation of monolayer coverage on the outer interface of the adsorbent[10-12]. From the figure- I, study reveals that as contact time increases the percentage removal increases. It indicates that initially the adsorption process is directly proportional to contact time and it attains equilibrium in 10-20 minutes.

Effect of Initial Chromium concentration on Multani miti

The adsorption of Cr (VI) at different initial concentrations ranging from 50-200 mg/L was determined. The experimental data were measured at 60 minutes to get full equilibrium. The adsorption curves of Cr (VI) from solutions containing different initial concentrations are shown in figure-2. The percentage removal of chromium (VI) is more and it is ranging between 93-98% for all concentration[13-14].

Effect of Multani mitti Dosages

To study the effect of multani miti dose (g) on the removal of Cr (VI) was done with 85 mg/L, solutions, while the amount of multani mitti added was varied between 0.2 – 2.0 gm/L. The values indicate an increase in the adsorption capacity with increase in multani mitti dosages. This is because of an increase in the number of available adsorption sites of the adsorbent which results in enhanced removal of Cr (VI)[15]. Results are given in figure-3 showed that the percentage removal of Cr (VI) is directly proportional to adsorbent dosage, and reached an optimum at 0.8 gm of sorbent [2,16].

Effect of pH on Adsorption Studies

The pH of the aqueous solution is an important controlling parameter in the adsorption p rocess[17]. In the present work, adsorption of chromium Cr(VI) on the clay adsorbent was studied over the pH range of 2.0 to 8.0 for a constant amount of clay (multani matti) 1 gm, and chromium (VI) concentration of 35 mg/L at room temperature. At high acidity the clay mineral surface will be completely covered with H_3O^+ ions and Cr (VI) ions can hardly compete with them for adsorption sites. With increase in pH, the competing effect of hydronium ions decreases and positively charged Cr (VI) ions adsorb on the free binding sites of multani mitti. This is a common observation for all cases of adsorption of metal cations on solid surface in media of different acidity and basicity[18-19].

Effect of Temperature on Adsorption Studies

Temperature has an important effect on the process of adsorption. The percentage of Chromium adsorption is studied as a function of temperature. The results obtained are presented in figure- 5 at temperatures of 0^oC, 30^oC, 40^oC, $50\ ^oC$, 60^oC and 80^oC. The decrease in percentage of adsorption with rise in temperature may be due to desorption caused by an increase in the available thermal energy. Higher temperature induces higher mobility of the adsorbate causing desorption [20].

Adsorption Isotherm studies

Temkin Adsorption Isotherm

Temkin adsorption isotherm is studied by taking into account he interactions between adsorbents and metal ions to be adsorbed and is based on the assumption that the free energy of sorption is a function of the surface coverage[21]. The linear form of the Temkin isotherm is represented as:

$$q_e = B \ln A + B \ln C_e \quad \text{...... (1)}$$

Where C_e is the equilibrium concentration of the adsorbate in mg/L, q_e is the amount of adsorbate adsorbed at equilibrium (mg/g), $RT/b_T = B$ where T is the temperature (K) and R is the ideal gas constant (8.314 J $mol^{-1}K^{-1}$) and A and b_T are constants. Figure-6 (a) & (b) showing relation between C_e and q_e, from the figures and table-2 it was observed that the adsorption of Chromium by multani mitti is following Temkin adsorption isotherms. The straight line of the graph indicates that adsorption is practically proportional to concentration. The adsorption capacity increases, with increase in temperature. Adsorption (both physical and chemical) processes are exothermic in nature.

Freundlinch Adsorption isotherm

The Freundlich equation is an empirical relationship describing the adsorption of solute from liquid to a solid surface. The Freundlich equation is expressed as

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

The linear form of Freundlich equation is as follows

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

Where k_f is the Freundlich adsorption capacity and *n* is the adsorption intensity. A plot of log q_e versus log C_e gives a linear line with slope of 1/n and intercept of log k_f given in table-2. From the figure-7 and table-2 it is observed that the adsorption of aqueous Chromium solution at different temperature perfectly fits in to Freundlich isotherm. The r^2 value and ASS, values are more compared to Langmuir isotherms. Freundlich adsorption isotherms are linear at lower and higher concentrations at all temperatures.

The Langmuir adsorption isotherm is the best known linear model to determine the adsorption parameters. Langmuir model represented by following equation

$$q_e = \frac{k_L C_e}{1 + b_L C_e} = \frac{q_0 b C_e}{1 + b_L C_e} \quad \text{.........(4)}$$

The linearized form of Langmuir adsorption isotherm is as follows

$$C_e = \frac{1}{q_0 b_L} + \frac{1}{q_0} C_e \quad \text{..........(5)}$$

Where q_e is the amount adsorbed at equilibrium (mg/g), Q_0 is the monolayer adsorption capacity (9mg/g), C_e the equilibrium concentration of adsorbate (mg/L) and b_L is the Langmuir constant related to energy adsorption. The figure-8 shows the plot of C_e/q_e against C_e. Values of Q_0 and b_L are presented in table-2. The figure-8 and table-2, indicates that the adsorption isotherm studies of removal of chromium by multani mitti does not fit for the Langmuir isotherm.

Adsorption Kinetics model

In order to check the kinetic parameters of removal of Chromium using multani mitti, the experimental data were tested with pseudo first order kinetic model, pseudo second order kinetic model and Elovich kinetic model. This adsorption kinetics studies are used to investigate the mechanism and the rate controlling steps of adsorption. The mechanism of adsorption involves the chemical reaction of functional groups present on the surface of the adsorbent and the adsorbate, temperature and pH[22].

Pseudo first orde r kinetic model

This model explains that the rate of change of solute uptake with time is directly proportional to difference in saturation concentration and the amount of solid uptake with time [23]. The plot of log (q_e – q_t) versus t should give a straight line with slope of –k/2.303 and intercept log q_e. Where qe and q_t are the amount of chromium adsorbed at equilibrium and at contact time t. From the slope and intercept of the plot the adsorption rate constant and equilibrium adsorption capacity were calculated[22]. From the graph (Figure- 9) and statistical report (Table-1) it was observed the present study removal of Chromium by Multani mitti is not following pseudo first order kinetic model.

Pseudo Second order kinetic model

The pseudo second order kinetic order equation expressed as

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad \text{........(6)}$$

Where k_2 is the rate constant of pseudo second order adsorption (g/mg min) and q_e is the equilibrium adsorption capacity (mg/gm)[24]. Figure-8 shows the pseudo second order plot for the adsorption of Chromium using multani mitti at different initial concentration at room temperature. When the pseudo second order kinetic is applicable, the plot of t/q_t against t should give a linear relationship from which K_2 and q_e determined respectively from the intercept and slope of the plot. The figure-10 shows the plot of pseudo second order kinetic model with linear relation. The r^2 values are higher than the values of pseudo first order kinetic model. It is observed form the figure-10 that the adsorption of Chromium by multani mitti perfectly fits to the pseudo second order kinetic model[25].

Elovich Model

The Elovich model equation is generally expressed as[26-27]

$$\frac{dq_t}{d_t} = \propto \exp(-\beta q_t) \quad \text{...... (7)}$$

Where:
α is the initial adsorption rate ($mg \cdot g^{-1} \cdot min^{-1}$)
β is the desorption constant ($g \cdot mg^{-1}$).
If the adsorption of aqueous Chromium solution by multani mitti fits to the Elovich model, a plot of q_t versus ln (t) should give a linear relationship with a slope of (1/β) and an intercept of 1/β ln (αβ). The results of Elovich plot for the adsorption of Chromium solution by multani mitti at various initial concentrations are given in figure-11. Form the table-1 and figure-11 it is observed that the Elovich model does not fit for the adsorption of Chromium solution by multani mitti. The Elovich model mainly applicable for chemical adsorption kinetics[28].

Intra particle diffusion model analysis

The adsorption of aqueous Chromium solution by multani mitti fits into pseudo second order kinetic model. To find out the adsorption mechanism the data is tested with intra particle diffusion model. Adsorption is a multi-step process invo lving transport of the solute molecules from the aqueous phase to the surface of the solid particulates followed by diffusion into the interior of the pores[22]. Assuming that the rate is controlled by pore and intra particle diffusion, the amount adsorbed (q_t) is proportional to the $t^{1/2}$, as shown below

$$q_t = k_{id}\, t^{1/2} + I \quad \text{....... (8)}$$

Where q_t is the amount of Chromium adsorbed (mg/g) at time t (min), and I is the intercept (mg/g). k_{id} and I values are obtained from the slopes and intercept of the linear p lot. Figure-12 represents the plots of q_t versus $t^{1/2}$ for adsorption of Chromium solution by multani mitti at various initial Chromium concentrations. From the figure-12 and table-1 it can conclude that the adsorption mechanism is not fits to intra particle diffusion model.

Thermodynamic calculations

Thermodynamic parameters were calculated from the variation of the equilibrium constant, *K*, at different temperature by using following equation.

$$k_c = \frac{q_e}{C_e} \quad \text{.......(9)}$$

K_c (L/g) values were obtained using the Khan and Sing method[29] by plotting ln (q_e/C_e) versus q_e and extrapolating to zero q_e. The intercept of the straight line with the vertical axis gives the values of K_c. The Gibbs free energy change of the adsorption process is related to K_c as in equation given below[30].

$$G^o = - RT \ln K_c \quad \text{....... (10)}$$

The changes in enthalpy (ΔH^o) and entropy (ΔS^o) for Chromium adsorption were calculated from the slope and intercept of the plot of ln *Kc* against 1/*T* according to the van't Hoff equation[30].

$$\ln k_c = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \quad \text{......(11)}$$

Plotting lnKc versus 1/T for multani mitti shows straight lines (Fig. 14). From their slope and intercept, H^0 and S^0 are determined. The negative values of G^0 indicate a favourite and spontaneous process[31].
The negative values of H^0 show that chromium adsorption by multani mitti is exothermic process and it can be concluded that the nature of the adsorption process is physical[32], either cation exchange or hydrogen bonding[31-33]. The positieve value of S^0 indicates an increase in randomness at the solid-solution interface during the adsorption process. In oreder to find out the efficiency of adsoption process, the dimensionless

equilibrium parameter, parameter, R_L was calculated by using following equation (Altundogan et al., 2000; Mahramanlioglu et al., 2002; Singh and Pant, 2004; Agarwal et al., 2004; Bhakat et al., 2006; Mhunia et al., 2007).

$$R_L = \frac{1}{1 + bC_o} \qquad \text{..........(12)}$$

Where C_o= Initial Concentration (mg/mL)
b= Langmuir isotherm constant.

Values of the dimensionless equilibrium parameter, R_L, of different temperatures are calculated for the initial concentrations of Chromium (VI) and are given in table-4. The parameters explain the differences in the shapes of the isotherm. The R_L values shows between 0 and 1 indicate favorable adsorption. The R_L value above 1 indicates unfavorable. The observed R_L values for Chromium (VI) are between 0 to 1 and it indicative of the favourable adsorption

Table 1: Kinetic parameters for adsorption of Chromium by Multani mitti

S.No	Parameters	Chromium concentration (10 mg/L)	Chromium concentration (35 mg/L)	Chromium concentration (85 mg/L)
	Pseudo first order kinetic model			
01	R^2	0.929	0.958	0.999
	ASS	0.012	0.013	0.000
	K_1	0.172	0.108	0.967
	Pseudo Second order kinetic model			
02	R^2	0.985	0.951	0.999
	ASS	0.494	0.101	0.039
	K_2	0.419	0.879	0.003
	Elovich model			
03	R^2	0.832	0.928	0.599
	ASS	15.26	94.93	24.38
	α	4.947	9.508	14.71×10^{21}
	β	0.454	0.112	0.655
	Intraparticle diffusion model			
04	$_R2$	0.423	0.630	0.190
	ASS	52.69	489.6	49.30
	kid	0.210	0.976	0.115
	I	4.93	11.93	79.77

Table 2: Isotherm kinetics for adsorption of Chromium (VI) by Multani mitti

S.No	Parameters	Temperature °C					
		0	**30**	**40**	**50**	**60**	**80**
	Temkin Adsorption Isotherm						
01	R^2	0.999	0.999	0.999	0.999	0.995	0.999
	ASS	0.140	1.943	2.588	2.926	20.26	0.731
	a_T	-1.880	-0.991	-1.113	-2.509	-2.124	-1.326
	b_T	0.988	0.961	0.963	0.979	0.988	0.978
	Langmuir Adsorption Isotherm						
02	R^2	0.677	0.720	0.794	0.999	0.993	0.644
	ASS	0.012	0.009	0.006	0.017	0.0002	0.012
	Q_0	1.25	1.25	1.26	1.23	1.32	1.24
	b_L	-0.0029	-0.0028	-0.0029	-0.0022	-0.0036	-0.0028
	Freundlich Adsorption Isotherm						
03	R^2	0.999	0.999	0.999	0.999	0.998	0.998
	ASS	0.0005	0.0002	0.0001	0.0001	0.0008	0.0007
	Log k_f	-0.203	-0.198	-0.200	-0.159	-0.220	-0.176
	1/n	1.103	1.096	1.097	1.070	1.103	1.089

Table 3: Thermodynamic parameters of Chromium adsorption by Multan mitti

S. No	Temperature	ΔG^{o} (KJ/ mol)	ΔS^{o} (KJ/ mol)	ΔH^{o} (KJ/ mol)
01	273	-10.151×10^{3}	4.780	-76.53
02	303	-11.336×10^{3}		
03	313	-11.710×10^{3}		
04	323	-12.532×10^{3}		
05	333	-12.477×10^{3}		
06	353	-13.200×10^{3}		

Table 4: Effect of Parameter RL values at different concentration and different temperatures

S.No	Temperature	Concentration of Chiromium (mg/L) and R_L values		
		10 mg/mL	35 mg/mL	85 mg/mL
01	0	0.1002	0.0286	0.0117
02	30	0.1002	0.0286	0.0117
03	40	0.1003	0.0286	0.0118
04	50	0.1002	0.0286	0.0118
05	60	0.1004	0.0286	0.0118
06	80	0.1010	0.0289	0.0119

Effect of Adsorbent Dosage on Other Parameters

The batch adsorption studies are carried out with paint manufacturing industry wastewater with multani miiti. The adsorptive removal of chromium was calculated along with other physical and chemical parameters of paint industry waste water to find out removal efficiency of multani mitti. The experimental results of before and after adsorption studies were shown in Table-4.

S.No	Parameter	Before Treatment	After Treatment
01	pH	6.4	6.6
02	Conductivity	8.13	7.55
03	TDS	5.26	4.93
04	Salinity	6.5	6.1
05	Turbidity	240 NTU	200 NTU
06	Acidity	360	160
07	Alkalinity	1440	760
08	Total Hardness	760	260

The efficiency of multani mitti used in present studies are allowed to check the removal capacity of other physical parameters such as pH, Electro conductivity (EC), Turbidity, Salinity, Total Dissolved Solids(TDS), Acidity, Alkalinity and Total Hardness of water. From the figure- 15 (a) & (b), a rapid change was observed in Hardness, Alkalinity and Acidity after batch adsorption studies. This experiment conforms that multani mitti dose not only has tha capacity to remove chromium (VI), it may decrease acidity, alkalinity, hardness, salinity and conductivity of water (Antunes et al., 2003).

CONCLUSION

Multani mitti is used for adsorption of Chromium (VI) from industrial waste water studies reveals a low cost technique which can be used by small scale industries. Adsorption with multani mitti is not only cheaper but requires less maintenance and supervision.

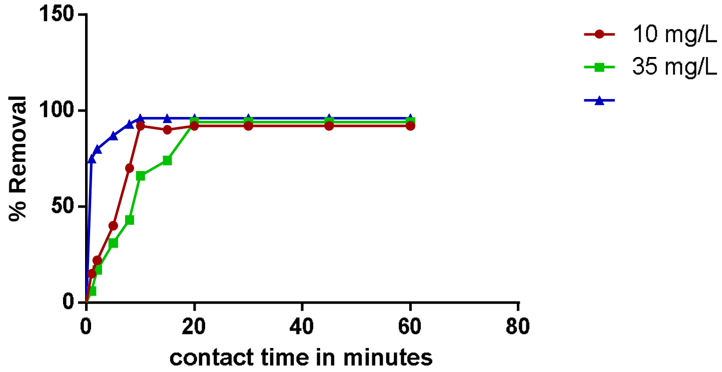


Fig. 1: Variation of contact time between Chromium and Multani miti

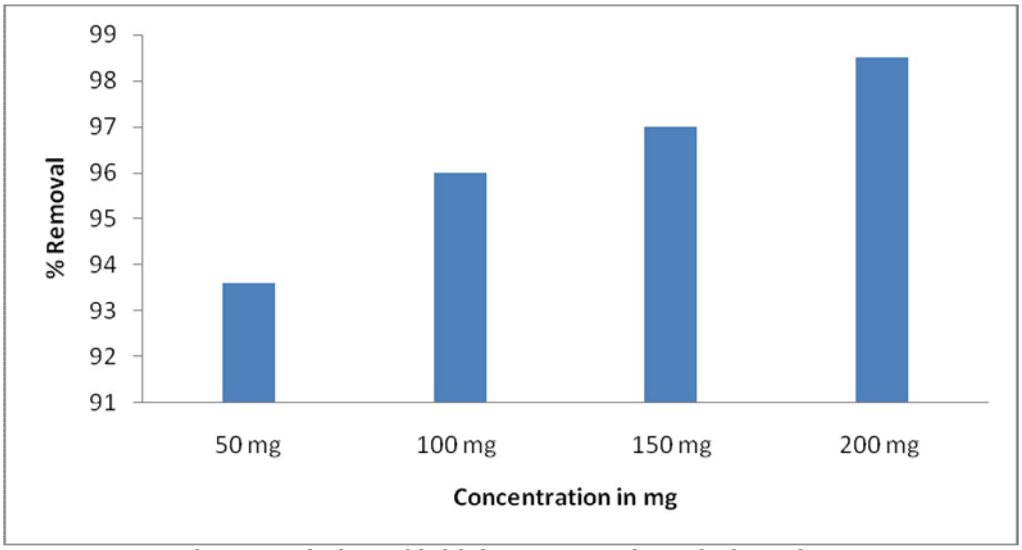


Fig. 2: Variation of initial concentration of Chromium

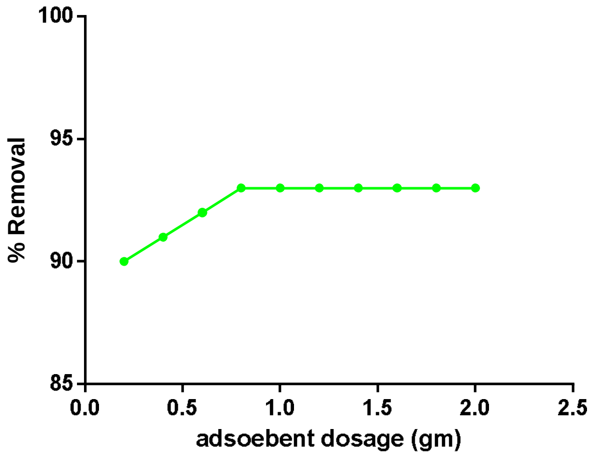


Fig. 3: Variation of Multani miti dosages

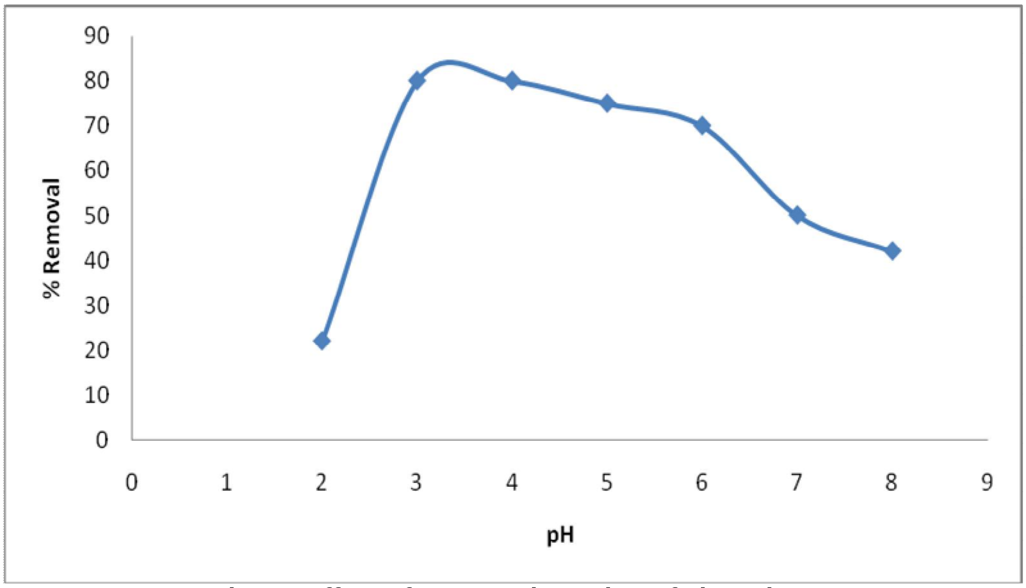


Fig. 4: Effect of pH on Adsorption of chromium

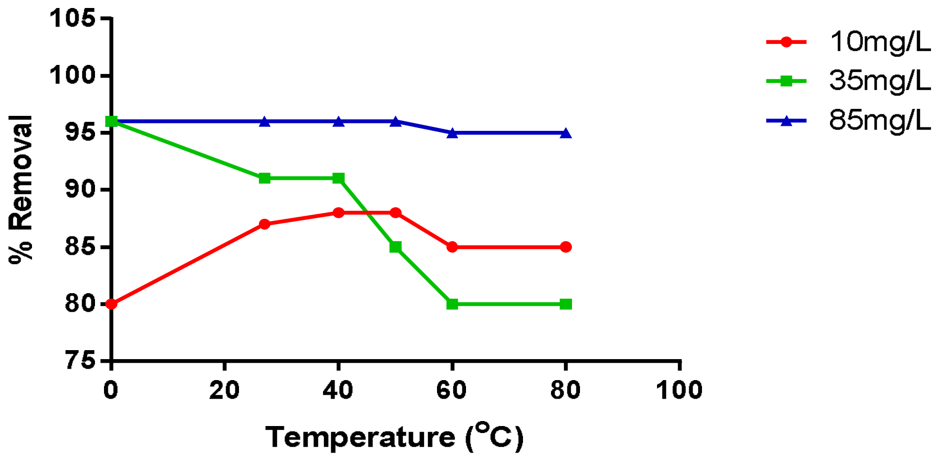
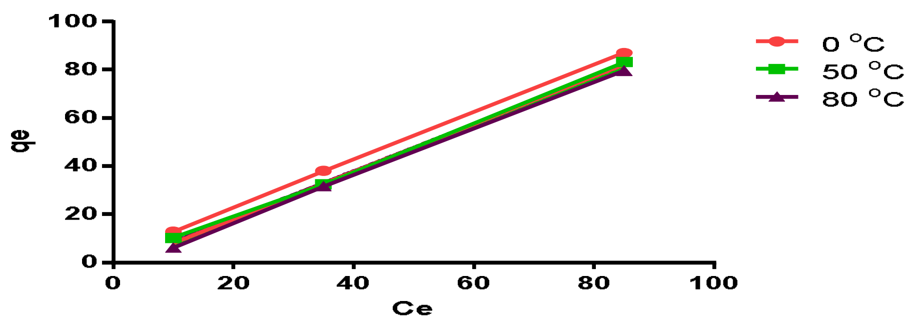


Fig. 5: Effect of Temperature



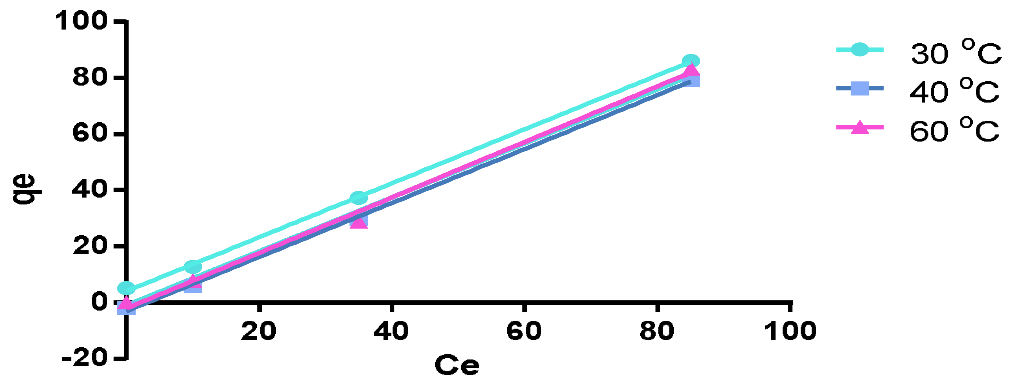


Fig. 6(a) & (b): Temkin adsorption isotherm plots of Chromium removal at different temperature

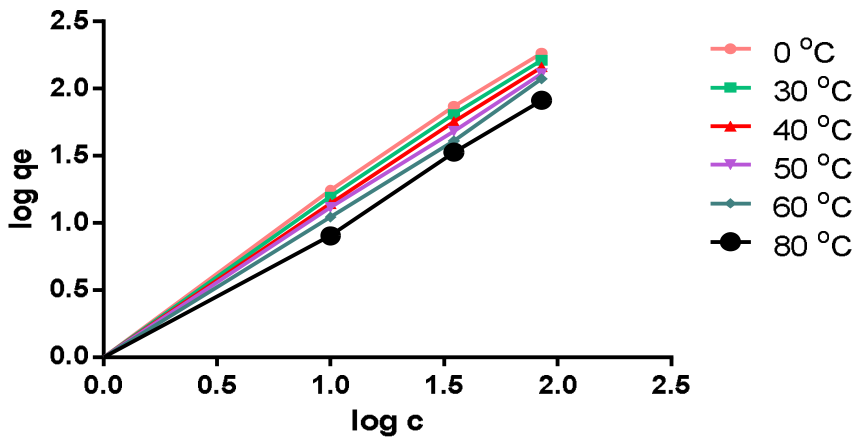


Fig. 7: Freundlich adsorption isotherm plots of Chromium removal at different temperature

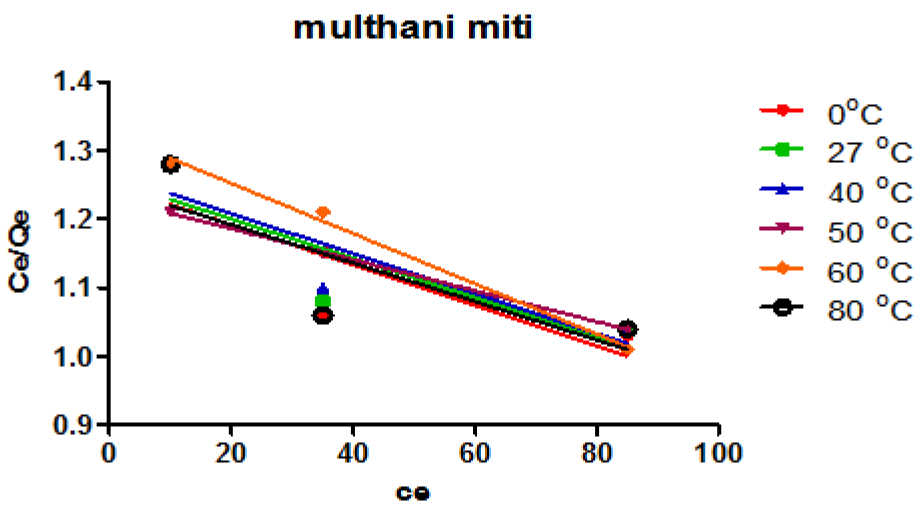


Fig. 8: Langmuir adsorption plot for chromium removal at different temperature

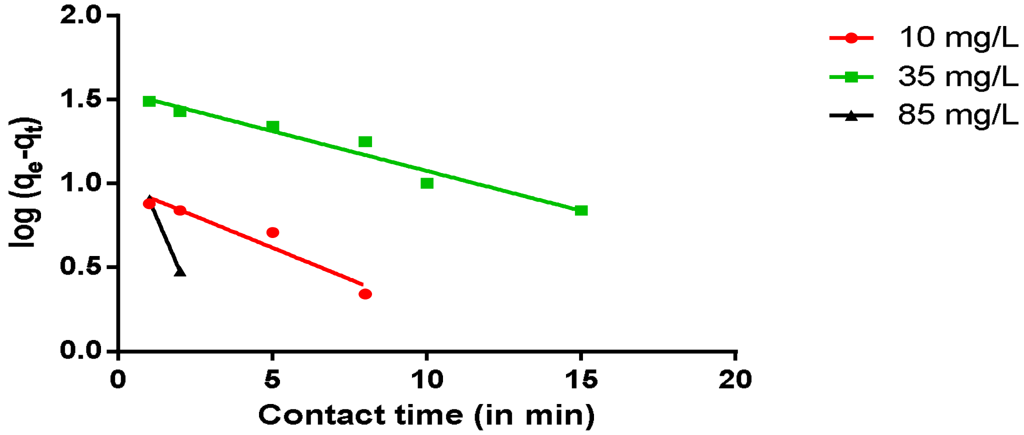


Fig. 9: Pseudo first order kinetic model

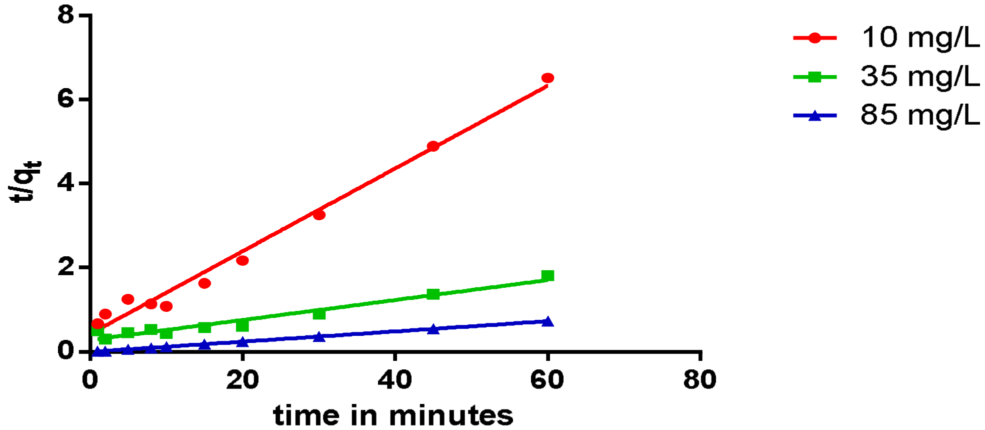


Fig. 10: Pseudo Second order kinetic model

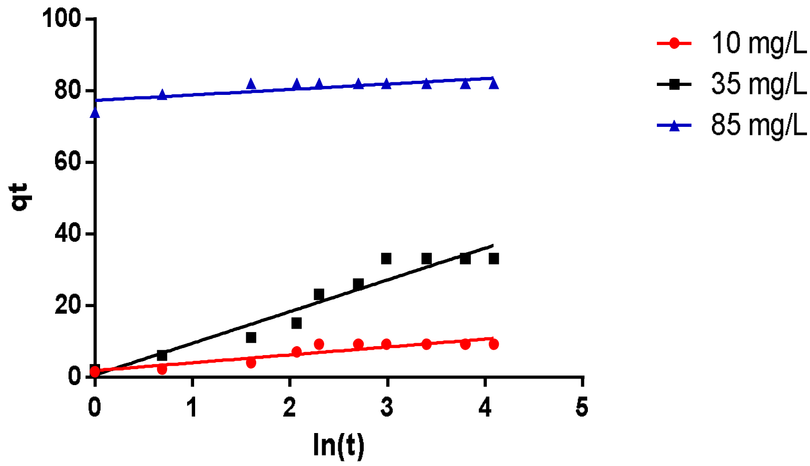


Fig. 11: Elovich Model

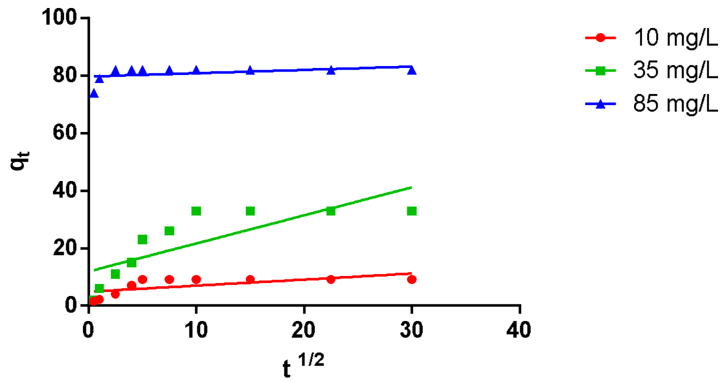


Fig. 12: Intra particle diffusion model

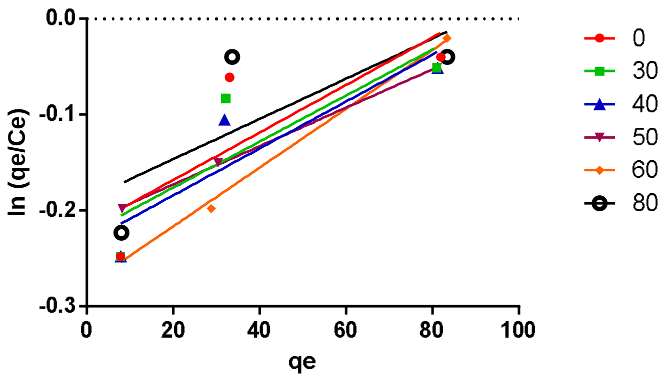


Fig. 13: Relationship between ln (q_e/C_e) and q_e

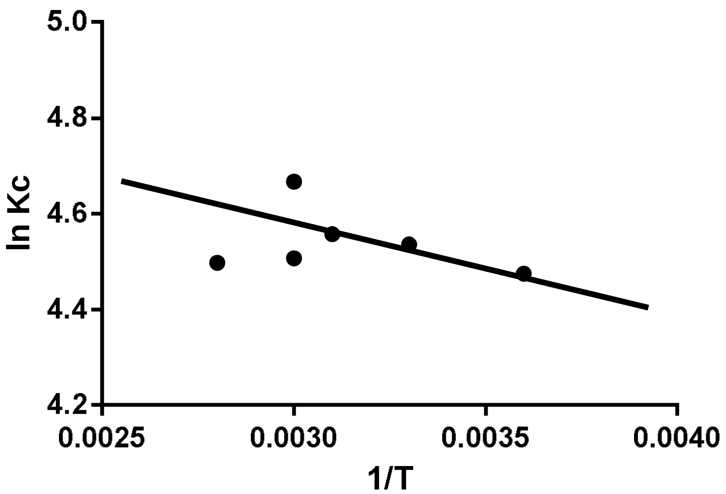
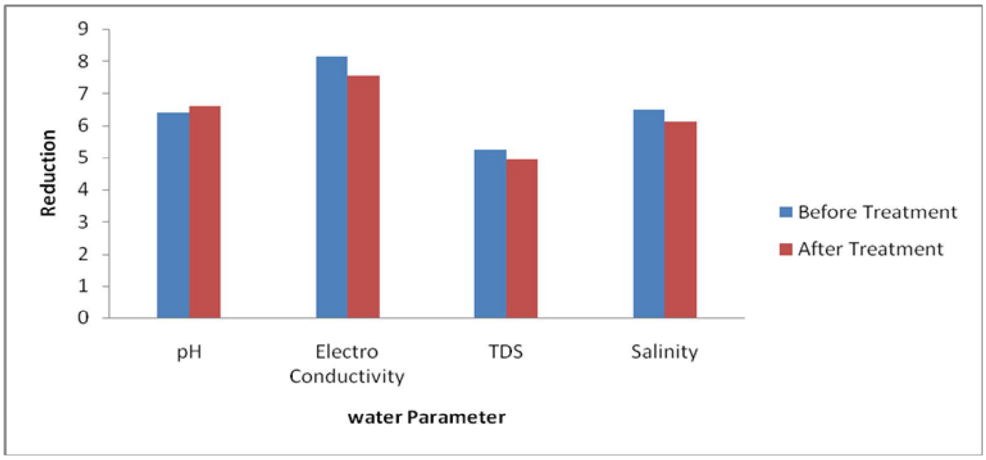


Fig. 14: plots of ln K_c and 1/T of chromium adsorption at different temperature



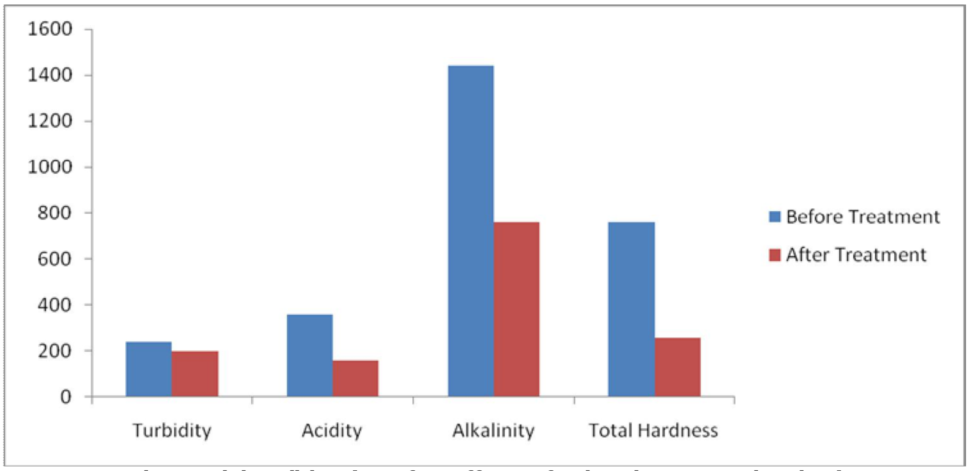


Fig. 15 (a) & (b): plots for effect of adsorbent on physical and chemical parameters of paint industry waste water

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