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Adsorption Study for Chromium (VI) on Iraqi Bentonite

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Abstract

The subject of this research involves studying adsorption to remove hexavalent chromium Cr(VI) from aqueous solutions.

Adsorption process on bentonite clay as adsorbent was used in the Cr(VI) concentration range (10-100) ppm at different temperatures (298, 303, 308 and 313)K, for different periods of time.

The adsorption isotherms were obtained by obeying Langmuir and Freundlich adsorption isotherm with R^2 (0.9921-0.9060) and (0.994-0.9998), respectively.

The thermodynamic parameters were calculated by using the adsorption process at four different temperatures the values of ΔH, ΔG and ΔS was [(+6.582 – +6.547) $kJ.mol^{-1}$, (-284.560 – -343.070) $kJ.mol^{-1}$ and (+0.977 – +1.117) $kJ.K^{-1}.mol^{-1}$] respectively. This data indicates the spontaneous sorption process.

The kinetic study of adsorption process was studied depending on three kinetic equations:

1- Lagergren equation
2- Morris-Weber equation
3- Reichenberg equation

Keywords:

Introduction

Chromium can exist in several chemical forms, which have oxidation states ranging from 0-VI. However, only trivalent and hexavalent chromium are stable enough to occur in the environment[1,2].

Chromium is a naturally occurring element found in rocks, animals, plants, soil and in volcanic dust and gases[3]. Chromium (VI) in the form of CrO_4^{2-} or $Cr_2O_7^{2-}$ and $HCrO_4^-$, is largely employed in the chemical industry for chrome plating, the manufacture of dyes and pigments, leather tanning, wood preserving, battery, rust and corrosion inhibitors, textiles, electroplating, metal processing, steel fabrication and canning industries[4-9]. Although chromium is used in many industries and is known to be essential trace, but it is found to be toxic an causing cancer at high levels[10].

Toxicological studies are shown that the degree of toxicity of some elements depends on the chemical form in which the element presents, Cr(VI) is more toxicity; by contrast Cr(III) is negligible because it often forms insoluble hydroxides at neutral pH[11,12]. The USA EPA is require (0.05 and 0.1 mg/L) of Cr(VI) in drinking water and inland water, respectively[11,13].

Cr(VI) is known as carcinogens, mutagens, tetratogens in biological systems, beside that it can pose health risks such as liver damage, dermatitis, castro intestinal ulcers, diarrhea, vomiting and nausea[12,14].

The problem of removing pollutants from water are grown with rapid industrialization the discharge of toxic into water courses is a serious pollution problem.

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Adsorption is the tendency for accumulation of a substance to take place at a surface or at an interface the accummenence of adsorption is due to the atoms in any surface being subject to unbalanced forces of attraction perpendicular to the surface plane and these forces possessing a certain unsaturation[15-17].

Langmuir Isotherm is a good representation of chemisoprtion, and usually represents the low P portion of other isotherms[18].

Langmuir behavior assumes a rapid reversible adsorption, and interaction only between sorbate molecules and a surface site[19,20].

The Langmuir equation could be expressed as:

$$\left[\frac{\mathbf{C_e}}{\mathbf{Q}}=\frac{\mathbf{1}}{\mathbf{k}}+\frac{\mathbf{a}}{\mathbf{k}}\cdot\mathbf{C_e}\right] \;\ldots\ldots\ldots\ldots..(1)$$

where Q is the amount of adsorbate (mg/g), C_e is the equilibrium concentration (mg/L), and a, k are constants related to adsorption capacity and energy of adsorption respectively or sometimes called Langmuir constants.

Freundlich Isotherm is one of the most important isotherms that deals with sorption at solid-liquid interface. Most of surface are heterogeneous, so the change in potential energy is regular, and the adsorption sites are not equivalent in energy[21,22], hence the multilayer formation is highly expected.

Freundlich equation could be written as follows[23,24]:

$$Q = k_F\, C_e^{1/n} \;\ldots\ldots\ldots.(2)$$

$$\log Q = \log k_F + 1/n \log C_e \;\ldots\ldots(3)$$

where Q is the adsorbate quantity (mg/g), C_e is the concentration of adsorbate at equilibrium (mg/L), k_F, and n is the sorption capacity and an empirical parameter, respectively and also called Freundlich constants.

The major objective of this work is to minimize the concentration of Cr(VI) in aqueous solutions to the acceptable levels, by removing it via and an adsorption process is used and the optimum conditions for adsorption process have been found. The detailed kinetic and thermodynamics studies have been employed to investigate the mechanism, rate and order of the adsorption process.

Materials and Methods

The bentonite clay was supplied from the state company for geological survey and mining-Iraq, the bentonite had the following composition with particle size less than 75µm.

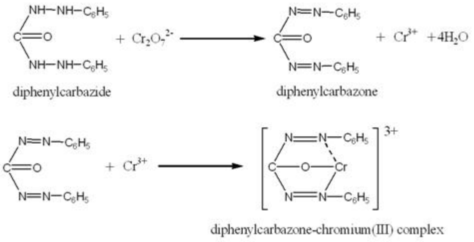
Constituent	SiO_2	Al_2O_3	CaO	MgO	K_2O	Na_2O	Fe_2O_3	L.O.I
Wt%	56.77	15.67	4.48	3.42	0.60	1.11	5.02	12.49

The molecular formula of bentonite could be written as:

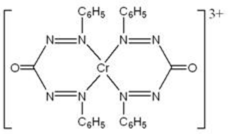
$$Mg_2Al_{10}Si_{24}O_{60}(OH)_{12}[Na,Ca]$$

The spectrophotometer method uses the selective reaction of Cr(VI) with (1,5-DPC) in basic media to yield a red-violet diphenyl carbazone complex.

The general mechanism was illustrated for oxidation of DPC with Cr(VI) was shown in the following equation[2,3].



The suggested final structure of the [Chromium-1,5-diphenylcarbazone] is as follows:



The complex was prepared using 2.5ml (1M) NaOH in volumetric flask 250 ml, and 50ml of 100ppm Cr(VI) was added to it, 7.5ml of DPC was added (with stirring) to the Cr(VI) solution followed by 2.5ml (50%) H_3PO_4 and then diluted to the mark by distilled water.

A sample of 5 ppm complex solution was taken and a spectrum scan was measured using UV-Vis Spectrophotometer and showed a λ_{max} at 543 nm

The quantity of adsorbate was calculated by using the following formula

$$Q_e = V_{sol}\,(C_o - C_e) / m \quad \ldots\ldots..(4)$$

Where:

Q_e = Quantity of adsorbate (mg/g).

V_{sol} = Total volume of adsorbate solution (L).

C_o = Initial concentration of adsorbate solutions ($mg.L^{-1}$).

C_e = Concentration of adsorbate solution at equilibrium ($mg.L^{-1}$).

m=Weight of adsorbant (g).

While the adsorption percentage was calculated by using the following formula.

$$\%Q = \frac{(C_o - C_e)}{C_o} * 100 \quad \ldots\ldots\ldots..(5)$$

Where:

%Q = Adsorption percentage.

And the adsorption coefficient was calculated by using the following formula[27]:

$$K_{ads} = \frac{Q_e}{C_o} \quad \ldots\ldots\ldots\ldots\ldots.(6)$$

Where:

K_{ads} = The adsorption coefficient.

Results and Discussion

Table (1) and Figure (1) shows the variation of C_o-C_e with the amount of adsorbent (bentonite).

Table -1- The values of (C_o-C_e) and M (quantity of sorbent)

C_o-C_e /$mg.L^{-1}$	M /g
40.18	0.050
40.27	0.075
40.38	0.100
40.30	0.150
40.25	0.200
40.22	0.300

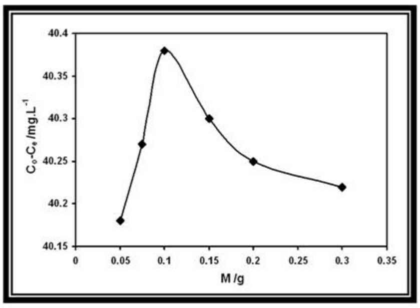


Fig. -1- The effect of clay dose on the adsorption process for 50 ppm Cr(VI) solution on bentonite at 298K

Table (2) and Figure (2) show the variation of Q_e with the contact time for 50 ppm Cr(VI) solution at 298 K to be in attachment with (0.1g) of bentonite clay.

Table -2- The values of Q_e and C_e at different time for 50 ppm Cr(VI) solution at 298K

C_e /$mg.L^{-1}$	Q_e /$mg.g^{-1}$	Time /min.
9.91	4.009	5
9.85	4.015	10
9.79	4.021	15
9.71	4.029	20
9.62	4.038	30
9.61	4.039	40
9.61	4.039	50
9.60	4.040	60

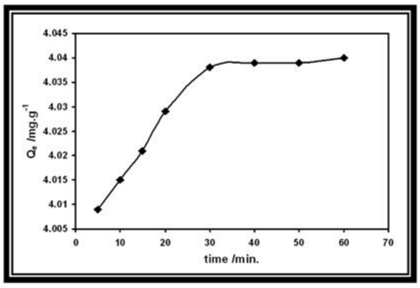


Fig. -2- The variation of Q_e with equilibrium time for 50 ppm Cr(VI) solution at 298K

The pH is an important factor that effected on the adsorption process, Table (3) and Figure (3) show the variation of %Q with different pH values for 50 ppm Cr(VI) solution at 298 K. It's clear that (pH=7) has the maxium Q% value, and this pH was chosen to all experiments of adsorption done subsequently.

Table -3- The adsorption percentage at different pH values, using bentonite at 298K for 50 ppm Cr(VI) solution

Q%	pH
79.62	3
79.85	5
80.76	7
79.54	9
79.37	11

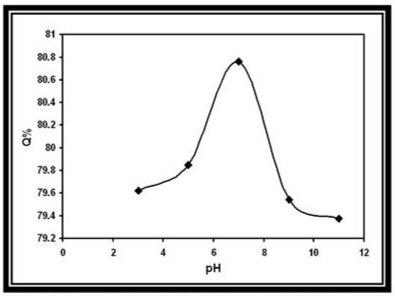


Fig. -3- The adsorption percentage vs. pH, using bentonite at 298K for 50 ppm Cr(VI) solution

The equilibrium adsorption isotherms are of fundamental importance in the design of adsorption systems[(22]. Table (4) and Figure (4) show the Langmiur data and Linear relationship between C_e/Q_e and C_e of a series Cr(VI) solutions at (298, 303, 308 and 313)K.

Table -4- The values of C_o, C_e and C_e/Q_e for the adsorption of series Cr(VI) solutions at different temperatures

298K			303K			308K			313K		
C_o/ $mg.L^{-1}$	C_e/ $mg.L^{-1}$	C_e/Q_e/ $g.L^{-1}$	C_o / $mg.L^{-1}$	C_e / $mg.L^{-1}$	C_e/Q_e/ $g.L^{-1}$	C_o / $mg.L^{-1}$	C_e / $mg.L^{-1}$	C_e/Q_e/ $g.L^{-1}$	C_o $mg.L^{-1}$	C_e $mg.L^{-1}$	C_e/Q_e/ $g.L^{-1}$
10	2.10	2.65	10	2.22	2.85	10	2.31	3.00	10	2.40	3.15
20	4.08	2.55	20	4.17	2.63	20	4.25	2.69	20	4.36	2.78
30	5.95	2.47	30	6.08	2.54	30	6.17	2.59	30	6.27	2.64
40	7.80	2.42	40	7.93	2.47	40	8.03	2.51	40	8.11	2.54
50	9.62	2.38	50	9.65	2.39	50	9.84	2.45	50	9.92	2.49
60	11.35	2.33	60	11.41	2.34	60	11.55	2.38	60	11.64	2.40
70	12.96	2.27	70	13.05	2.29	70	13.14	2.31	70	13.25	2.33
80	14.58	2.22	80	14.67	2.24	80	14.80	2.26	80	14.86	2.28
90	16.10	2.17	90	16.22	2.19	90	16.37	2.22	90	16.43	2.23
100	17.56	2.13	100	17.61	2.13	100	17.70	2.15	100	17.84	2.17

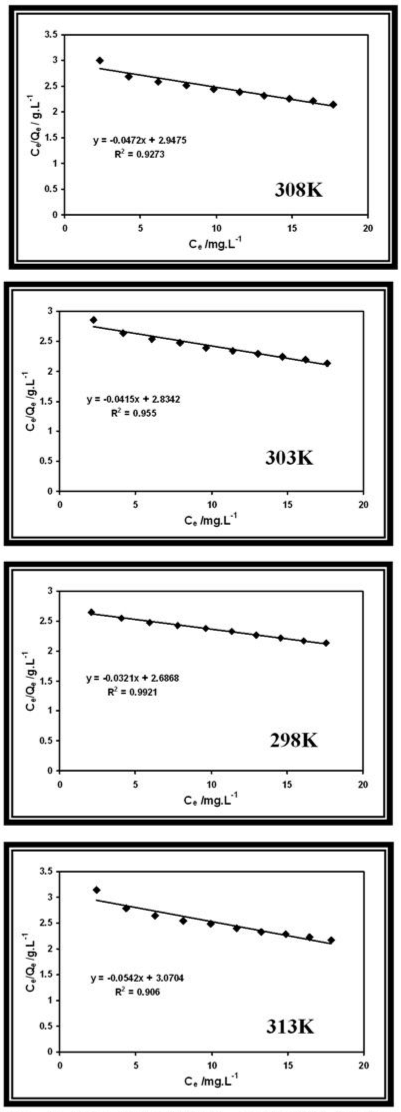


Fig. -4- The plot of C_e/Q_e against C_e for the adsorption of series Cr(VI) solutions at different temperatures

Table (5) Show the Langmuir constant at different temperatures

Table -5- The values of Langmuir constant at temperature range (298-313)K

T (K)	K_L /L/mg	a	R^2
298	0.3721	0.0119	0.9921
303	0.3528	0.0146	0.9550
308	0.3392	0.0160	0.9273
313	0.3256	0.0170	0.9060

Table (6) and Figure (5) show the data and linear relationship of Freundlich isotherm between Log Q and Log C_e for the adsorption of Cr(VI) solutions at the temperature range (298-313)K.

Table -6- The values of C_e, Log C_e and Log Q for the adsorption of series Cr(VI) solutions at different temperatures

298K			303K		
C_o / mg.L^{-1}	Log C_e	Log Q_e	C_o / mg.L^{-1}	Log C_e	Log Q_e
10	0.322	-0.102	10	0.346	-0.109
20	0.610	0.201	20	0.620	0.199
30	0.774	0.381	30	0.783	0.378
40	0.892	0.507	40	0.899	0.506
50	0.983	0.606	50	0.984	0.605
60	1.054	0.687	60	1.057	0.686
70	1.112	0.756	70	1.115	0.755
80	1.163	0.815	80	1.166	0.815
90	1.206	0.868	90	1.210	0.867
100	1.244	0.916	100	1.245	0.915
308K			313K		
C_o/ mg.L^{-1}	Log C_e	Log Q_e	C_o/ mg.L^{-1}	Log C_e	Log Q_e
10	0.363	-0.114	10	0.380	-0.119
20	0.628	0.197	20	0.639	0.194
30	0.790	0.377	30	0.797	0.375
40	0.904	0.504	40	0.909	0.503
50	0.922	0.603	50	0.996	0.602
60	1.062	0.685	60	1.065	0.684
70	1.118	0.754	70	1.122	0.753
80	1.170	0.814	80	1.172	0.813
90	1.214	0.867	90	1.215	0.866
100	1.247	0.915	100	1.251	0.914

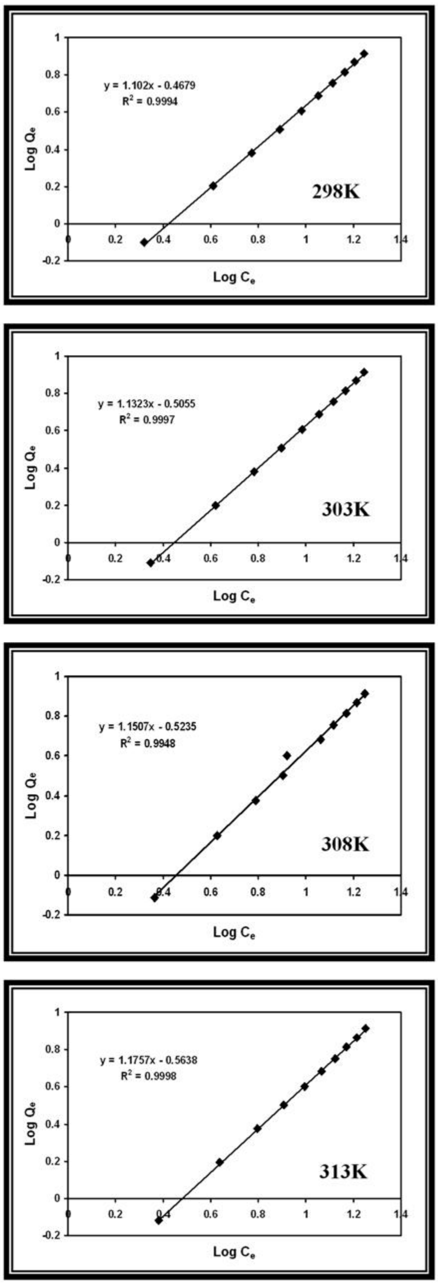


Fig. -5- Freundlich linear relationship between Log Q_e vs. Log C_e for a series Cr(VI) solutions at different temperature

Table (7) Show the Freundlich constants at different temperatures

Table -7- The values of Freundlich constants at different temperatures

T (K)	Log k_f	k_f / mg/g/(mg/L)^n	1/n	n	R^2
298	-0.4679	0.3404	1.1020	0.9074	0.9994
303	-0.5055	0.3122	1.1323	0.8831	0.9997
308	-0.5331	0.2930	1.1523	0.8678	0.9997
313	-0.5638	0.2730	1.1757	0.8505	0.9998

Table (8) and Figure (6) show that the adsorption percentage of Cr(VI) decrease as temperature increase, where as the adsorption process is an exothermic process, so absorption was involved with adsorption and all processes and all processes increase with temperature increase.

Table -8- The adsorption percentage and Q_e values at different temperature

T/K	Q_e / mg.g^{-1}	Q%
298	4.038	80.76
303	4.035	80.70
308	4.016	80.30
313	4.008	80.16

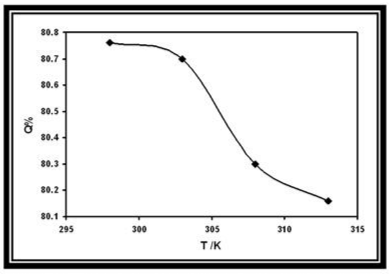


Fig. -6- The effect of temperature on the adsorption percentage

The thermodynamic functions, ΔH, ΔG and ΔS were calculated using the following formulas[28,29].

$b = a \cdot \exp(-\Delta H/RT)$ ……..…(7)
$\ln b = \ln a - (\Delta H/R) \cdot 1/T$ ……...(8)
$\ln a = \Delta S/R$ ……………(9)
$\Delta G = \Delta H - T\Delta S$……..………(10)

Where b is the maximum adsorption quantity for series Cr(VI) solutions at different temperature and could be obtained from the plot of C_e/Q_e vs. C_e (Fig.(4)).

The equation (8) was used to calculate the ΔH, by plotting ln b vs. 1/T. Table (9) and Figure (7) showed a linear relationship and the slope represents -ΔH/R, where R is the gas constant (8.314J.mol^{-1}.K^{-1}) , and ΔH_{ads} were calculated.

Table -9- The values of the reciprocal of temperature and ln b

T (K)	1/T * 10^{-3}	b	ln b
298	3.35	2.68	0.98
303	3.30	2.83	1.04
308	3.24	2.94	1.08
313	3.19	3.07	1.12

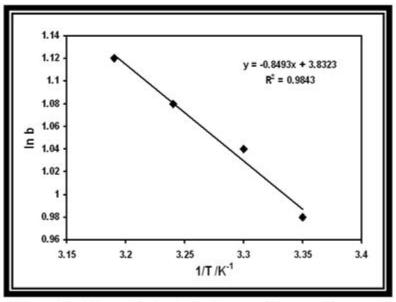


Fig. -7- The plot of ln b vs. the reciprocal of temperature

Table -10- Show the thermodynamic functions of the adsorption process

T (K)	ΔH*10^{-3} (kJ.mol^{-1})	ΔG (kJ.mol^{-1})	ΔS*10^{-3} (J.mol^{-1}.K^{-1})
298	6.55225	- 284.560	+ 977
303		- 307.680	+ 1037
308		- 325.167	+ 1077
313		- 343.070	+ 1117

From the Table (10), It's clear that ΔH has the positive values, which indicate the ideal and the maximum value of a physic-sorption process. All values of ΔG were negative so the adsorption of Cr(VI) on the bentonite was considered a spontaneous process, also ΔS was had the positive values, and it was increased with the temperature increase, the disorder of the system increase, i.e. the system becomes more disordered, which due to the absorption process and diffusion of Cr(VI) ions in the bentonite pores.

The kinetic of the adsorption process was studied via introducing three models, also the rate constant was calculated.

1- Lagergren Model

The rate constant k_{ads} was determined from the following pseudo-1^{st}-order rate expression, which could be expressed as follows[30]:

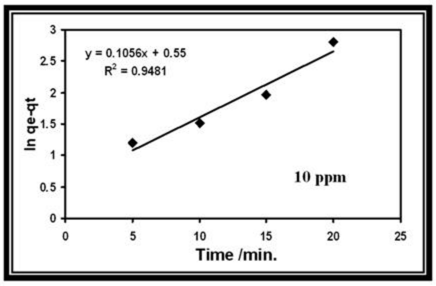
$$\ln q_e\text{-}q_t = \ln q_e\text{-}k_{ads}\, t \;\ldots\ldots\ldots(11)$$

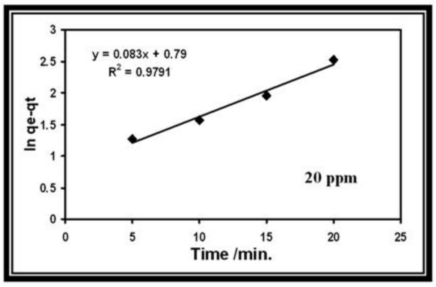
where q_t and q_e are the amount of Cr(VI) adsorbed at time t /min., and at equilibrium time respectively, k_{ads} /min.$^{-1}$ is the rate constant and values are given in Table (11) at 298K.

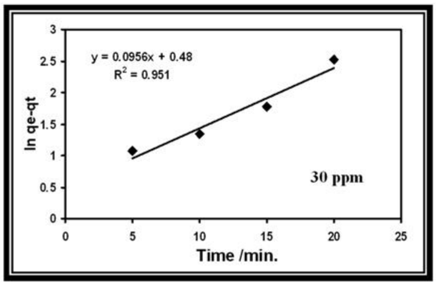
The linear relationship was obtained via plotting ln q_e-q_t vs. t /min., as shown in Figure (8).

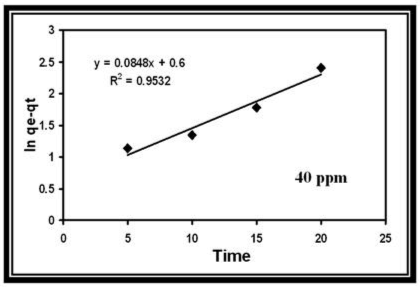
Table -11- The rate constants of adsorption process for a series Cr(VI) solutions at 298K

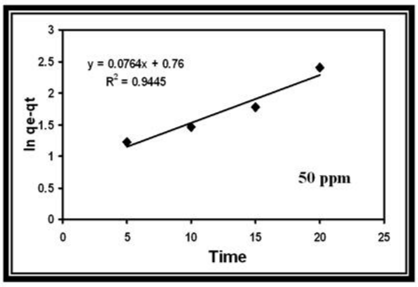
C_o (mg.L^{-1})	k_{ad} (min.$^{-1}$)	C_o (mg.L^{-1})	k_{ad} (min.$^{-1}$)
10	0.1056	60	0.0890
20	0.0830	70	0.0934
30	0.0956	80	0.1198
40	0.0848	90	0.0756
50	0.0784	100	0.1192

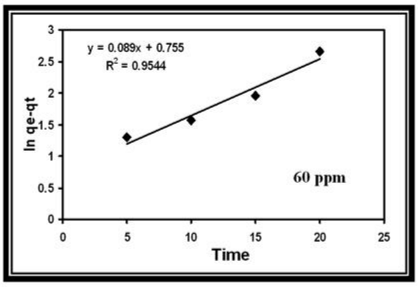


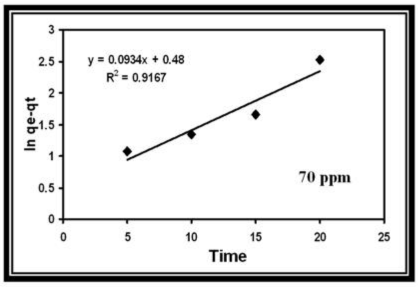












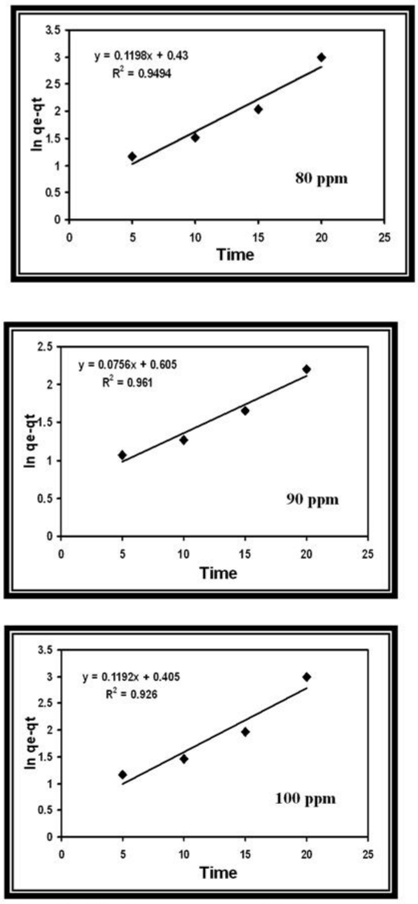


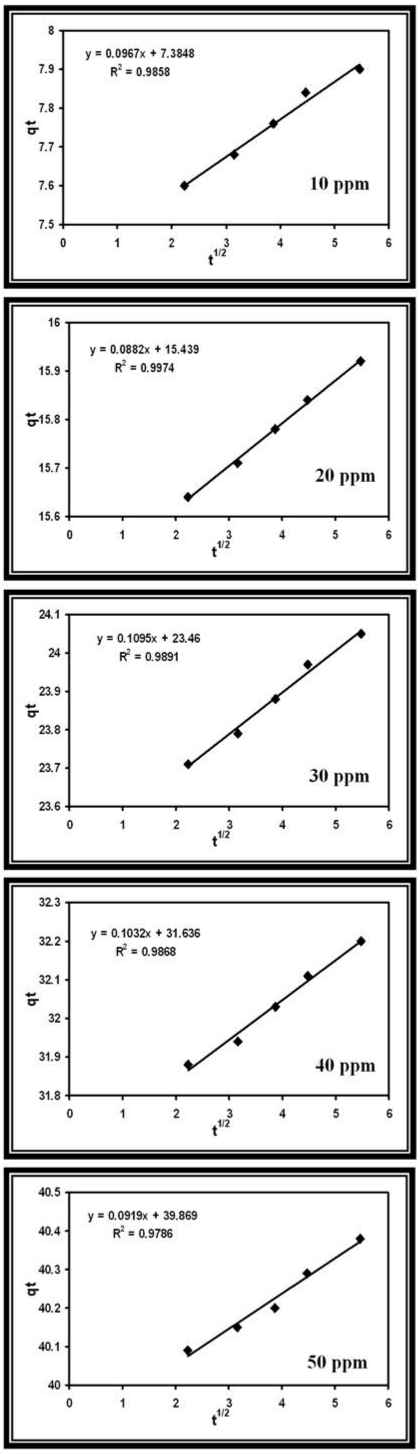
Fig. -8- The Lagergren model for a series Cr(VI) solutions at 298K

2- Morris-Weber Model

This kinetic model was used to estimate the rate limiting step of any adsorption process, the equation of this model could be expressed as follows$^{(31,32)}$.

$$q_t = k_d \sqrt{t} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(12)$$

Where q_t is the quantity of adsorbed material at any time /mg.g^{-1}, k_d is the diffusion constant, and t is the time of diffusion /min., the plotting of q_t vs. $\sqrt{t}$ was accomplished at 298K. Figure (9) shows the plot of q_t vs. $\sqrt{t}$ for different Cr(VI) concentrations at 298K.



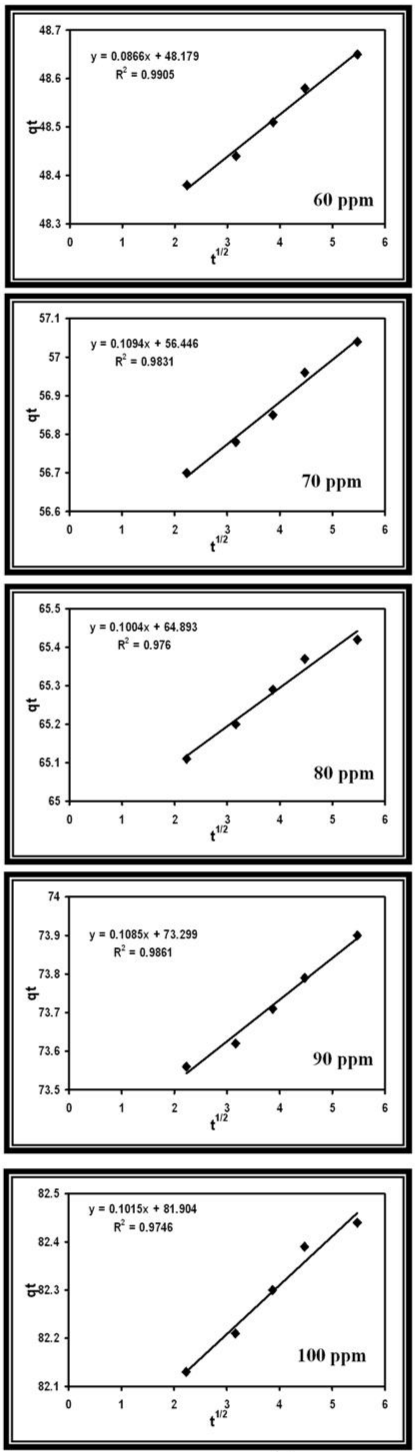


Fig. -9- The plot of q_t vs. $\sqrt{t}$ for a series Cr(VI) solutions at 298K

3- Reichenberg Model

This kinetic model was proposed to discuss the behavior of many adsorption process in solution and Reichenberg had introduced following formula(33):

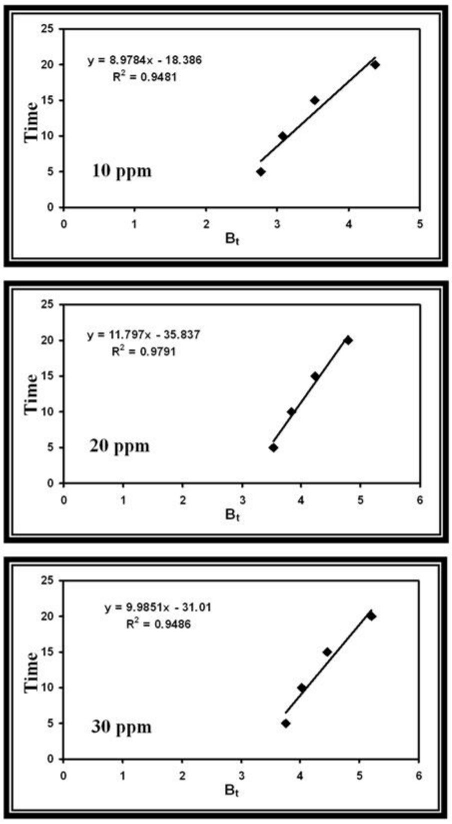
$F = [1-6/\pi^2]\ e^{-Bt}$ ………….(13)

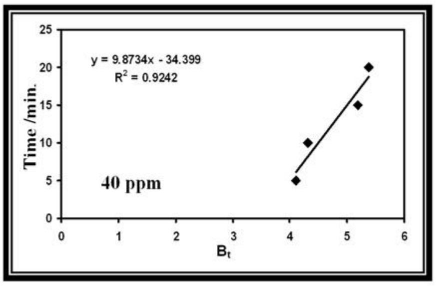
$B_t = -0.4977 - \ln(1-F)$ ………(14)

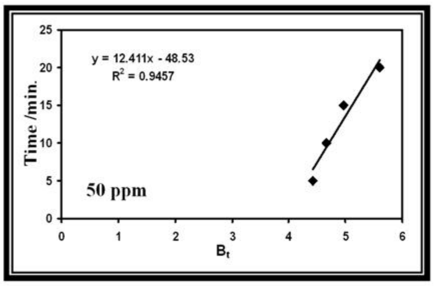
$F = q_t / q_e$ ………….(15)

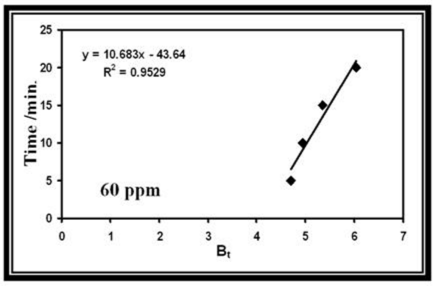
Plotting of time (min.) vs. B_t revealed a linear relationship with relatively acceptable R^2 values. Figure (10) shows the variation of B_t with time for different concentration of Cr(VI) at 298K.

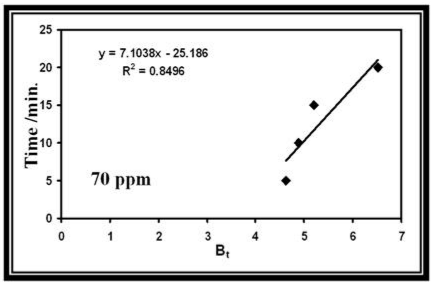
According to this model, it characterized the rate determining mechanisms which was diffusion process for Cr(VI) ions from the bulk solution to the absorbent surface and absorption occurred.

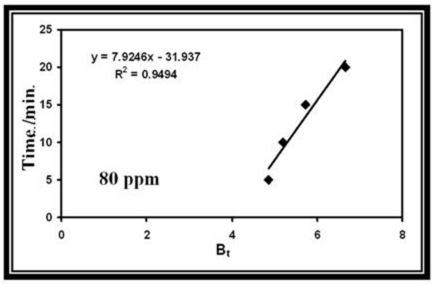


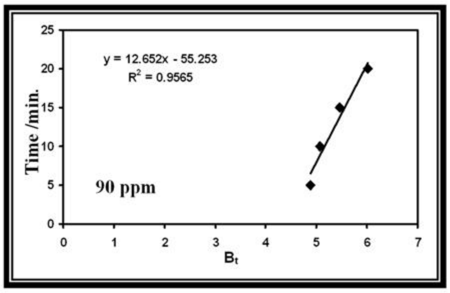












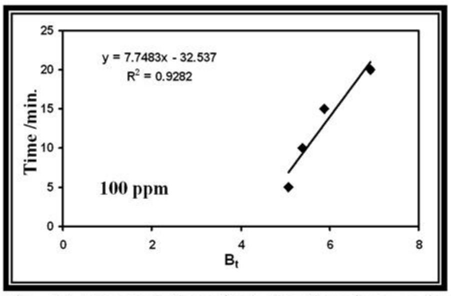


Fig. -10- The variation of Bt with time for a series of Cr(VI) solutions at 298K

Conclusion

The adsorption of Cr(VI) on bentonite was of Langmuir type at Cr(VI) concentration range (10-1000) ppm. Langmuir type indicates to the monolayer formation of Cr(VI) on the bentonite surface. The isotherms of Cr(VI) was followed both Langmuir and Ferundlich models.

The adsorption of Cr(VI) on bentonite reched to the largest value (83.54) at pH = 7.

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دراسة امتزاز الكروم السداسي على طين البنتونايت العراقي

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الخلاصة

يتضمن موضوع البحث دراسة امتزاز الكروم السداسي لازالته من المحاليل المائية. عملية الامتزاز على طين البنتونايت (كمادة مازة) لازلة تراكيز مختلفة من الكروم السداسي تراوحت بين (10-1000) ملغم/لتر بمدى من درجات الحرارة (298-313) كلفن وعند الازمان المختلفة.

تم الحصول على ايزوثرمات الامتزاز بتطبيق معادلتي لنكماير وفرندلش بنسبة خطية (R^2=0.9921-0.9060) و (R^2=0.994-0.9998) على التوالي.

تم حساب القيم الثرموديناميكية باربع درجات حرارية وكانت قيم الانثالبي محصورة بين (6.547+ — 6.582+) كيلوجول.مول$^{-1}$ وقيم الطاقة الحرة لكبس بين (284.56- — 343.07-) كيلوجول مول$^{-1}$ وقيم الانتروبي بين (977+ — 1117)جول.مول$^{-1}$. بينت القيم تلقائية عملية الامتزاز.

تمت دراسة حركيات الامتزاز اعتماداً على ثلاث معادلات حركية هي 1- معادلة لنكماير 2- معادلة موريس-ويبر 3- معادلة ريجمبرغ.