

Adsorption characteristics of ferro-fol drug with bentonite clay, kinetic and thermodynamic studies

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Abstract

A batch adsorption system was applied to study the adsorption of different concentration from Fe (11) from aqueous solution by bentonite clay at temperature range (303-323) K.

The adsorption capacities and rate of Fe (11) ions on to clay were evaluated. Langmuir and freundlich adsorption models were applied to describe the isotherms and isotherm constants.

Equilibrium data agreed very well with the Langmuir model.

The kinetic experiment data correlated well with the first order kinetic model using Lagergreen's equation, Thermodynamic parameters for the sorption process was studied. Results also showed that bentonite was favourable adsorbed.

Introduction :

iron is the fourth most abundant element in the earth's Crust, it is present in a variety of rock and soil minerals both as Fe (111) and Fe (11). is required for proper transport and storage of oxygen means of hemoglobin and myoglobin while it's oxidized forms, methemoglobin and metmoglobin, which contain Fe (111), will not bind oxygen⁽¹⁾ Iron plays an essential role in photosynthesis and is the limiting growth nutrient for phytoplanktons in some both Fe (11) and Fe parts of the ocean⁽²⁾ .

(111) are important in the biosphere, serving as an active center of range of protons such ⁽³⁾ as oxidases, redresses and dehydrates .

Ferro-fol drug is a haematinic preparation for prophylaxis of iron and folic acid deficiency during pregnancy and lactation, also this drug has been widely used for the treatment and prevention Excess iron in the body of anemia.⁽⁴⁾ .

causes liver and kidney damage (haemochromatosis). About lmg of iron is lost each day through sloughing of cells from skin and mucosal surfaces.

Among the many methods available for the removal of trace Iron namely : chemical precipitation, physical treatment such as ion exchange coagulation, solvent extraction and membrane process, adsorption has been shown to be economically feasible alternative.

Because of their low cost and local viability⁽⁵⁾ natural materials, such as clay minerals are good sorbents and inexpensive have received much attention

in sorption of heavy metals from contaminated solution.

Among the natural materials bentonite^(6, 7) is some of the most useful heavy metal sorbents proposed. Clay can remove pollutants from aqueous solution owing to their high cation exchange capacity and high specific surface area associated with their small particles size⁽⁸⁾.

Removal of metal cations by clay minerals is controlled by parameters such charge characteristics of the clay.

As⁽⁹⁾ charge In addition to the clay charge properties, metal uptake is also influenced by the characteristics of the metal ion itself such as its ionic radius, charge size and hard-soft acid-base properties⁽¹⁰⁾ Many researchers have investigated the influence of other factors on metal uptake by clays such as metal Concentrations⁽¹¹⁾ pH⁽¹²⁾, Presence Of competing ions⁽¹³⁾, temperature⁽¹⁴⁾ and ionic Strength⁽¹⁵⁾

The application of sorption⁽¹⁶⁾ In technique using clay has been tested . addition mixed sorbent systems, such as alumina and clay used to remove the colour of dye⁽¹⁷⁾ mixture of bentonite and wood- sawdust also used to study the sorption capacity and kinetics of basic red 46 from⁽¹⁸⁾ aqueous solution .

This work concentrates on the study of ferrous ion adsorption on to bentonite clay. The langmuir and freundlich equations were used to fit the equilibrium isotherm. The adsorption rate was determined quantitatively.

Materials and methods

The cheap, widely - available sorbent used in this study is bentonite clay supplied by Geological Surveying Company. Bentonite has molecular formula $\text{Mg}_2\text{Al}_2(\text{Si}_4\text{O}_{10})_2(\text{OH})_2[\text{Na}, \text{Ca}]$. the clay composition (by weight) is: SiO_2 (56.77), Al_2O_3 (15.67), CaO (4.48), MgO (3.42), K_2O (0.60), Na_2O (1.11), Fe_2O_3 (5), L.O.I (12.49). '

The sorbent used in the experiments was ferrous-folic drug (150mg $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) and (0.5 mg of folic acid) supplied by Egyptian pharmaceutical industries CO.A.R.E (ElPlCO). This drug a haematonic preparation for prophylaxis of iron and folic acid deficiency during pregnancy and lactation, its capsules are formulated to release most of the iron in the upper small intestine where absorption is the greatest and not in the stomach where gastric irritation may be caused.

Batch adsorption experiments Stock solution (100ppm) of drug was

prepared. The stock solution was then diluted using distilled water to give standard solutions of the appropriate concentrations that ranging from then 10 ml of (8.667-2.376 mg.L⁻¹).Then aliquots of these standard solutions were placed in around bottom 250 ml flasks and equilibrated using a magnetic stirrer, with (0.1 g) of bentonite clay at constant temperature. Thereafter, the solutions were filtered and the concentrations of Fe (11) in supernatant were analyzed at 248|nm using an atomic absorption spectrophotometer (GBC-933 plus) model.

The amounts of adsorption were calculated based on the difference of Fe (11) concentrations in aqueous solutions before and after adsorption, the volume of aqueous solution 10 ml and the weight of the clay (0.1gm) according to: Adsorption capacity

$$Q_e = (C_o - C_e) V / W \text{ --- (1)}$$

Where C_o is the initial Fe (11) concentration (mg.L⁻¹) C_e is the final or equilibrium Fe (11) concentration (mg.L⁻¹), V is the volume of the Fe (11) solution (ml), and w is the weight of the clay (g).

For batch kinetic studies, 0.1 g of clay were equilibrated under optimum conditions as mentioned earlier. the clay and 10 ml of Fe (11) solutions were placed in 250 ml round bottom flasks and stirred by a magnetic stirrer. The sorption time was varied between 5 to 30 minute at constant four temperatures (303-31 3-318-323k). At predetermined times the solutions in the beakers were separated from the clay by filtration. After filtration, the concentrations of Fe (11) in supernatant were determined at wavelength 248.3nm.

Results and discussion

Adsorption isotherms: The equilibrium data were analyzed in accordance with the langmuir and freundlich sorption isotherms.

These isotherms allow to describe adsorption phenomena of metals from aqueous solution on to clay sample. The experimental values of isotherm were used in the linear forms of Langmuir equation, which is valid for monolayer sorption on to surface with finite number of identical sites, is given by ⁽¹⁵⁾

$$C_e / Q = 1 / K_L + a / K_L \cdot C_e \text{ .----- (2)}$$

Where C_e: The equilibrium or final concentration of Fe (11) (mg.L⁻¹) or p.p.m

Q: The amount of Fe (11) adsorbed per unit weight of clay at equilibrium concentration (mg/g). and a, K_L are the Langmuir constants related to the affinity of binding and is a measure of sites (L.mg⁻¹),(L.gm⁻¹) an the energy of adsorption. Adsorption isotherms parameter is shown in table (1).

The widely used empirical freundlich equation based on sorption on endogenous surface, which assumes that different sites with several adsorption energies are involved is given by⁽¹⁹⁾

$$Q_e = K_f C_e^{1/n} \text{ ----- (3)}$$

Where K_f (mg.g-1) and n are freundlich distribution coefficients indicating to the total adsorption capacity of solid and in density, respectively. Equation (3) can be linearized in logarithmic form and freundlich constant can be determined

$$\log Q_e = \log K_f + 1/n \log C_e \text{ ----- (4)}$$

K_f and n can be determine from a linear plot of $\log Q_e$ against $\log C_e$. The calculated results of the Langmuir and freundlich isotherm constants are given in table (2). It is found that the adsorption of Fe (11) on the clay correlated well $r > 0.99$ with Langmuir equation as compared to the freundlich equation under the concentration range studied.

fig (1) shows the type of adsorption isotherms is L-type according to the Giles classification⁽²⁰⁾.

Fig (2) shows the relation between C_e / Q_e and C_e for the adsorption of Fe (11) ion on bentonite and the experimental adsorption isotherm values fitted into the linearized forms of Langmuir equation. This linear plots show that the adsorption obeys Langmuir isotherm model. From the slope and intercept of Langmuir isotherms the numerical values of langmuir isotherms constants were summarized in table (2).

The highest maximum adsorption value (a) will place at 313k and equal to 92.79 (L. mg⁻¹) and the lowest at 323k and equal to 33.65 (L.mg⁻¹).

Values of K_l (constant related to bonding- energy of the adsorbent) was highest at 313k and equal to 80.645 (L.gm⁻¹) and lowest at 313k and equal to 29.76 (L.mg⁻¹). It has been reported that the effect of isotherm shap with a view to predict if an adsorption system is "favorable" or "unfavorable" The essential features of Langmuir isotherm can be expressed in tends of a dimensionless constant separation factor or equilibrium parameter, R_L which is used to predict If an adsorption system is "favorable" or "unfavorable" The⁽²¹⁾. R_L . is defined by: separation factor ,

$$R_L = 1 / 1 + K_L C_o \text{ ----- (5)}$$

Where C_o is the initial Fe (11) and k_L is concentration (mg.L⁻¹) an k_L Langmuir's constant or the equilibrium Table (3) listed the constant (L/gm-1). a calculated values of R_L . Based on the effect of separation factor on isotherm shap the R_L values are in the range of $0 < R_L < 1$, which are indicate that the adsorption of Fe(11) on bentonite is favourable. Thus, bentonit is favourable adsorbed. Bentonite clay can be considered to be microporous, therefore pores are large enough to let Fe

(11) ions through. The mechanism of ion adsorption on porous adsorbents may involve three steps⁽²²⁾: (i) diffusion of the ions to the external surface of adsorbent; (ii) diffusion of ions into the pores of adsorbent; (iii) adsorption of the ions on the internal surface of adsorbent. The first step of adsorption may be affected by metal ion concentration. The last step is relatively a rapid process.

Linear plot of $\log Q_e$ versus $\log C_e$ shows that the adsorption follows Freundlich isotherm model (fig (3)) with correlation coefficient $r > 0.9$. K_f and n were calculated from the intercept and slope of the plot (table 2)

Kinetics of adsorption

The rate of adsorption of Fe(II) by bentonite⁽²³⁾ was determined using Lagergren's equation :

$$\ln (q_e - q_t) = \ln q_e - K_{ad} t \text{-----(6)}$$

Where q_t is the amount of Fe(II) adsorbed (mg/g) at time (t).

Q_e is the amount of Fe(II) adsorbed (mg/g) at equilibrium time, t is the time taken for adsorption (min) and K_{ad} is the rate constant of adsorption (min^{-1}). Straight line plots of $\ln (q_e - q_t)$ against (t) were used to determine the rate constant K_{ad} at different concentrations. The kinetics of Fe(II) adsorption on bentonite follow the first order rate expression that used for reversible reaction with an equilibrium being established between liquid and solid phases⁽²⁴⁾.

Kinetic data of Fe(II) adsorption on bentonite listed in table (4) and shown in fig (4). Table (5) shows values of K_{ad} . There are increasing in the rate of adsorption with increasing of the initial concentration of Fe(II).

Activation Energy calculation

The activation energy of the sorption process was calculated via introducing Arrhenius equation⁽²⁰⁾ as shown in equation (7):

$$\ln K = \ln A - E_a / RT \text{---- (7)}$$

where A (min^{-1}) is Arrhenius parameter.

which is a temperature independent factor. E_a is the activation energy (J.mol^{-1})

R is the gas constant ($8.314 \text{ J.K}^{-1} \text{ mol}^{-1}$) and T is the temperature in Kelvin.

Straight line plots of $\ln K_{ad}$ and $1/T$ (fig. (5)) to determine the activation energy for the adsorption of Fe (II) on bentonite.

The value of E_a has a very important role in estimating the dynamics of the sorption process, and it represents the energy barrier for a process to occur. The E_a values are shown in table (6), and the E_a value for the initial concentration of Fe (II) equal to 2.376 mg.L^{-1} was higher than E_a for

the other concentration. This behaviour due to the adsorption of ions on the lowest potential energy caly sites which require high E_a value (27) KJmol^{-1} while E_a is -a t Co equal equal to (3.6) KJmol^{-1} a to 8.667(mg.L^{-1}) that due to the chemisorptions of Fe (11) on highest potential energy sites of clay.

Thermodynamic Properties calculation

The values of maximum adsorption (b) for the adsorption of Fe (11) on bentonite at four temperature (303, 313, 318, 323) K are reported in table (7) by using other f f Langmuir ⁽²⁰⁾ models shown in old O equation (8):

$$C_e / Q = 1 / b + C_e \text{ ---- (8)}$$

Which give good fit for various temperatures. The thermodynamic function ΔH , ΔG , and ΔS were calculated using the following formulaso:

$$b = a \cdot \exp (-\Delta H / R t \text{ --- (9)}$$

$$\ln b = \ln a - (\Delta H / R) \cdot 1 / t \text{ ----- (10)}$$

$$\ln a = \Delta S / R \text{ ---- (11)}$$

$$\Delta G = \Delta H - T \Delta S \text{ --- (12)}$$

Fig (6) showed a linear relationship between (log b) and (1/T) and the value of ΔG , ΔS , and ΔH are reported in table (8).

The negative values of the ΔH indicate that Kinetics of adsorption That the exothermic chemisorption process has Been established on the surface of clay.

the values of ΔG were negative, so that sorption of Fe (11) on bentonite clay was - considered a spontaneous process ⁽²⁵⁾

the negative values of ΔS ($-84 \text{ J.mol}^{-1}\text{K}^{-1}$) is due to the increasing in the order of the rearrangement of Fe (11) ions on the surface of clay.

Conclusions

in this study the adsorption isotherms could be well fitted by the Langmuir equation, also the rate of adsorption could be described by the first-order equation.

The sorption process was exothermic and spontaneous with the increasing in the order of the rearrangement of the system. There are increasing in the rate of adsorption with the increasing of the initial concentration of Fe (11) ions. The activation energy of sorption was higher at initial concentration the equal to 2.376 (mg.L^{-1}) than a other concentration.

The results showed that the bentonite clay is a favourable absorbers for the Fe (11) adsorption.

Table (1): Adsorption isotherms parameters Ce and Q. of Fe (II) on bentonite at different temperatures.

Co mg/L	303K		313K		318K		323K	
	Ce mg/L	Q mg/L	Ce mg/L	Q mg/L	Ce mg/L	Q mg/L	Ce mg/L	Q mg/L
8.667	0.294	0.8373	0.299	0.8368	0.31	0.8357	0.33	0.8337
7.334	0.1655	0.7168	0.1600	0.7174	0.153	0.7180	0.1902	0.7091
6.00	0.037	0.5963	0.02	0.598	0.039	0.5961	0.049	0.5951
4.598	0.019	0.4579	0.018	0.458	0.021	0.4577	0.028	0.457
3.521	0.014	0.3497	0.012	0.3509	0.018	0.3503	0.024	0.3497
2.376	0.006	0.237	0.001	0.2375	0.004	0.2372	0.007	0.2374

Table (2): Freundlich and Langmiur parameter at different temperatures.

T(K)	r	n	K_L L.gm⁻¹	K_f mg.g⁻¹	a L.gm⁻¹	r
303	0.9921	3.3557	80.645	1.2727	92.79	0.9582
313	0.9927	4.4385	61.3497	1.1128	72.223	0.9479
318	0.9924	3.51	46.9484	1.2357	54.667	0.9319
323	0.9929	3.1078	29.762	1.2712	33.654	0.9368

Table (3): R_L values based on Langmuir equation.

Fe (II) Initial conc. (mg.L⁻¹)	303 K	313 K	318 K	323 K
	R_L	R_L	R_L	R_L
8.667	1.428 x 10 ⁻³	1.880 x 10 ⁻³	2.452 x 10 ⁻³	3.862 x 10 ⁻³
7.334	1.688 x 10 ⁻³	2.218 x 10 ⁻³	4.081x 10 ⁻³	4.562 x 10 ⁻³
6.00	2.062 x 10 ⁻³	2.271 x 10 ⁻³	3.537x 10 ⁻³	5.568 x 10 ⁻³
4.598	2.6895 x10 ⁻³	3.533 x 10 ⁻³	4.611x 10 ⁻³	7.255 x 10 ⁻³
3.521	3.51 x 10 ⁻³	4.608 x 10 ⁻³	6.010 x 10 ⁻³	9.452 x 10 ⁻³
2.376	5.192 x 10 ⁻³	6.813 x 10 ⁻³	8.885x 10 ⁻³	1.394 x 10 ⁻³

**Table (4): Kinetic data of adsorption different concentration of Fe (II) on bentonite at different temperatures
sorption time from 5,10,15,25,30 min. (PART A)**

Temp (K). 303													
Conc. (mg.L ⁻¹)	Conc. (mg.L ⁻¹) 7.334			Conc. (mg.L ⁻¹) 6.00			Conc. (mg.L ⁻¹) 4.598			Conc. (mg.L ⁻¹) 3.521			Conc. (mg.L ⁻¹) 2.375
	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e
1.358	7.168	7.1325	0.0355	5.963	5.929	0.034	4.579	4.539	0.04	3.507	3.471	0.036	2.37
0.597		7.1435	0.0245		5.935	0.028		4.551	0.028		3.449	0.022	
0.078		7.1515	0.0165		5.942	0.021		4.558	0.021		3.4985	0.0085	
0.033		7.1605	0.0075		5.953	0.01		4.568	0.0105		3.5003	0.0067	
0		7.168	0		5.963	0		4.579	0		3.507	0	

Temp (K). 313													
Conc. (mg.L ⁻¹)	Conc. (mg.L ⁻¹) 7.334			Conc. (mg.L ⁻¹) 6.00			Conc. (mg.L ⁻¹) 4.598			Conc. (mg.L ⁻¹) 3.521			Conc. (mg.L ⁻¹) 2.375
	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e
0.543	7.135	7.1484	0.0251	5.980	5.94	0.04	4.58	4.544	0.036	3.509	3.418	0.091	2.375
0.492		7.1535	0.02		5.958	0.022		4.556	0.024		3.431	0.078	
0.219		7.1575	0.016		5.964	0.016		4.562	0.018		3.478	0.031	
0.017		7.169	0.0045		5.9675	0.0125		4.57	0.01		3.491	0.018	
0		7.1735	0		5.98	0		4.58	0		3.509	0	

**Table (4): Kinetic data of adsorption different concentration of Fe (II) on bentonite at different temperatures
sorption time from 5,10,15,25,30 min. (PART B)**

Temp (K). 318													
Conc. (mg.L ⁻¹)	Conc. (mg.L ⁻¹) 7.334			Conc. (mg.L ⁻¹) 6.00			Conc. (mg.L ⁻¹) 4.598			Conc. (mg.L ⁻¹) 3.521			Conc. (mg.L ⁻¹) 2.375
	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e
1.350	7.1804	7.159	0.0214	5.917	5.897	0.11	4.577	4.419	0.158	3.503	3.408	0.091	2.372
0.65		7.1635	0.0169		5.810	0.107		4.48	0.09		3.429	0.078	
0.22		7.1675	0.0129		5.864	0.053		4.497	0.08		3.463	0.031	
0.09		7.1745	0.0059		5.872	0.045		4.532	0.045		3.483	0.018	
0		7.1805	0		5.917	0		4.577	0		3.503	0	

Temp (K). 333												
t	Conc. (mg.L ⁻¹) 6.00			Conc. (mg.L ⁻¹) 4.598			Conc. (mg.L ⁻¹) 3.521			Conc. (mg.L ⁻¹) 2.376		
	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t	q _e	q _t	q _e -q _t
235	5.951	5.824	0.127	4.57	4.443	0.127	3.497	3.401	0.096	2.374	2.354	0.02
245		5.831	0.12		4.488	0.082		3.424	0.073		2.358	0.016
255		5.886	0.065		4.511	0.059		3.437	0.06		2.362	0.012
265		5.899	0.052		4.539	0.031		3.458	0.039		2.366	0.0085
275		5.951	0		4.57	0		3.497	0		2.374	0

Table (5): Values of K_{ad} at different temperature.

Co mg.L ⁻¹	303 K Kad min ⁻¹	313 K Kad min ⁻¹	318K Kad min ⁻¹	323 K Kad min ⁻¹
8.667	0.1928	0.2008	0.2041	0.2105
7.334	0.080	0.0903	0.0946	0.0982
6.00	0.0574	0.0636	0.0647	0.066
4.598	0.0663	0.0855	0.0906	0.096
3.521	0.0323	0.059	0.0631	0.0666
2.376	0.0486	0.0799	0.0851	0.0956

Table (6): Values of Ea at different initial concentrations of Fe (II).

Co mg.L ⁻¹	8.667	8.667	8.667	8.667	8.667	8.667
Ea KJ.mol ⁻¹	3.602	8.566	8.596	15.22	27.159	27.474

Table (7): Maximum adsorption at different temperatures.

b	Log b	1 / T x 10 ⁻³
80.645	1.9065	3.3
61.35	1.79	3.19
46.95	1.672	3.145
29.76	1.474	3.095

Table (8): Thermodynamic parameter ΔG , ΔS and ΔH of Fe (II) adsorption on bentonite clay.

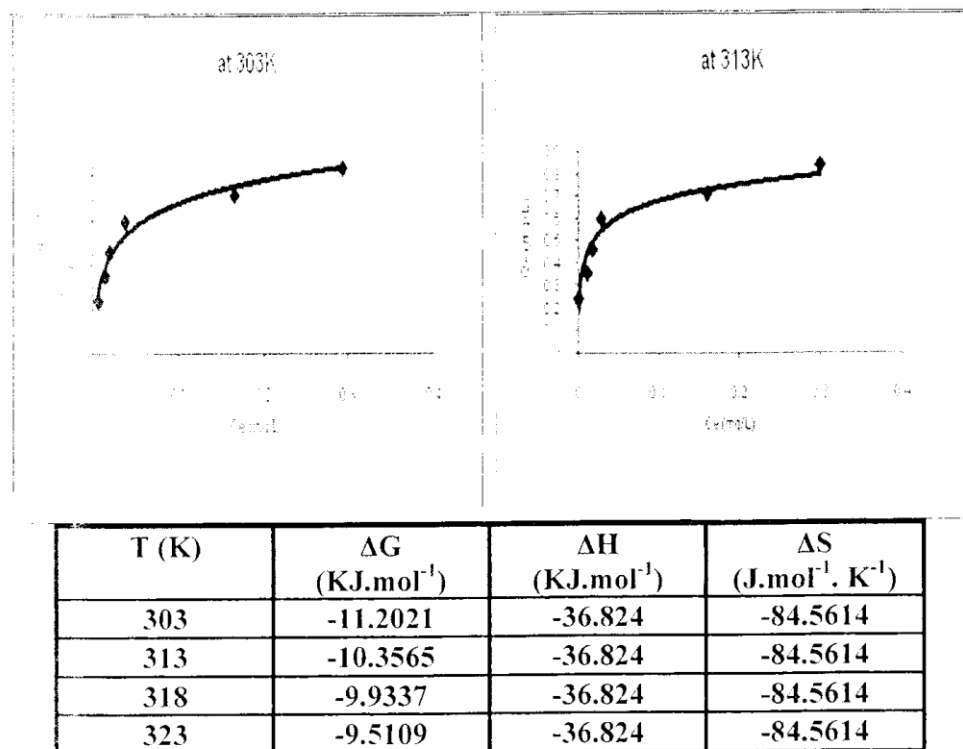


Fig (1):-Adsorption isotherm of Fe(II) ion on bentonite clay at different temperatures.

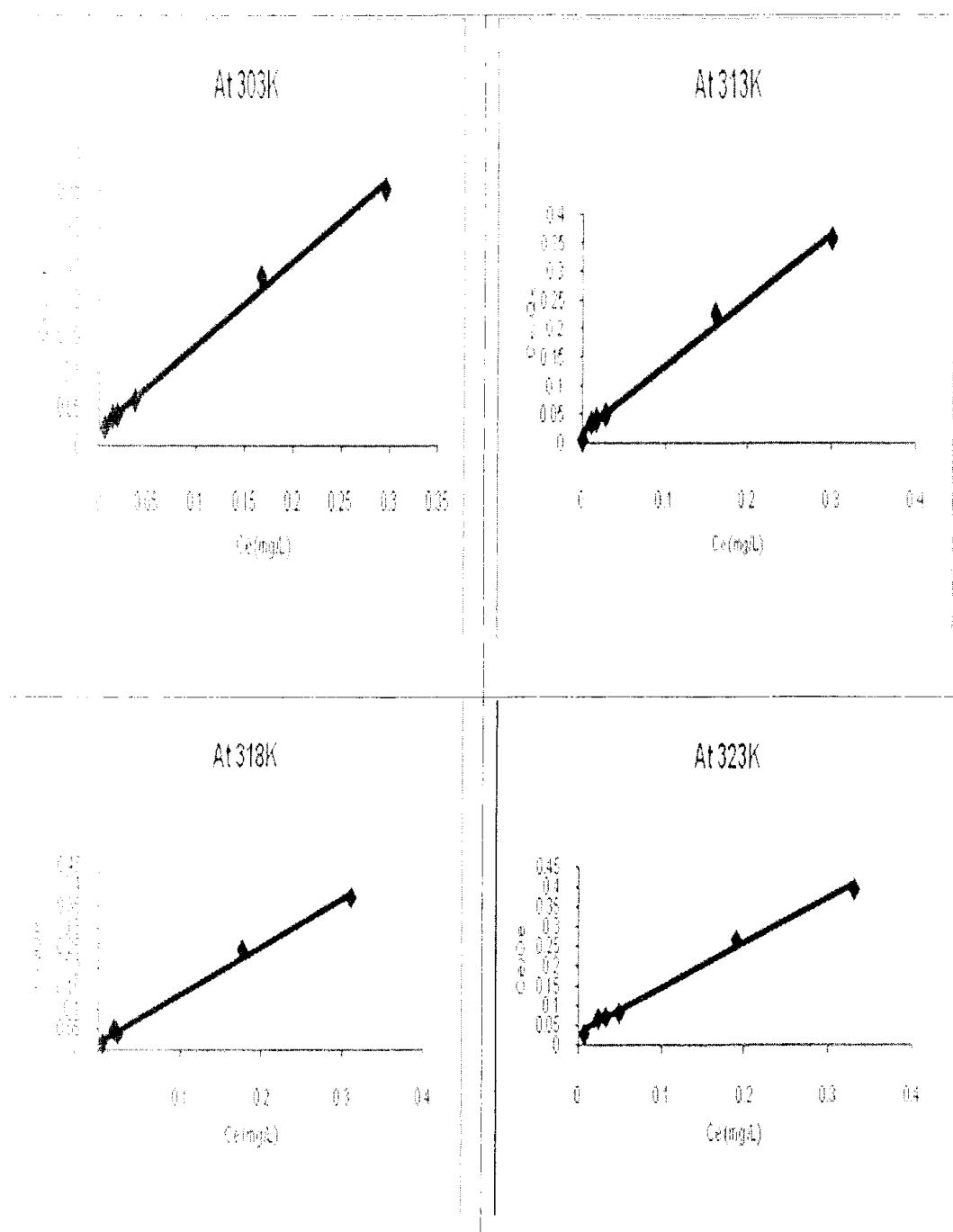


Fig (2):- The relation between (C_e/Q_e) and C_e for the adsorption of Fe (II) ion on bentonite .

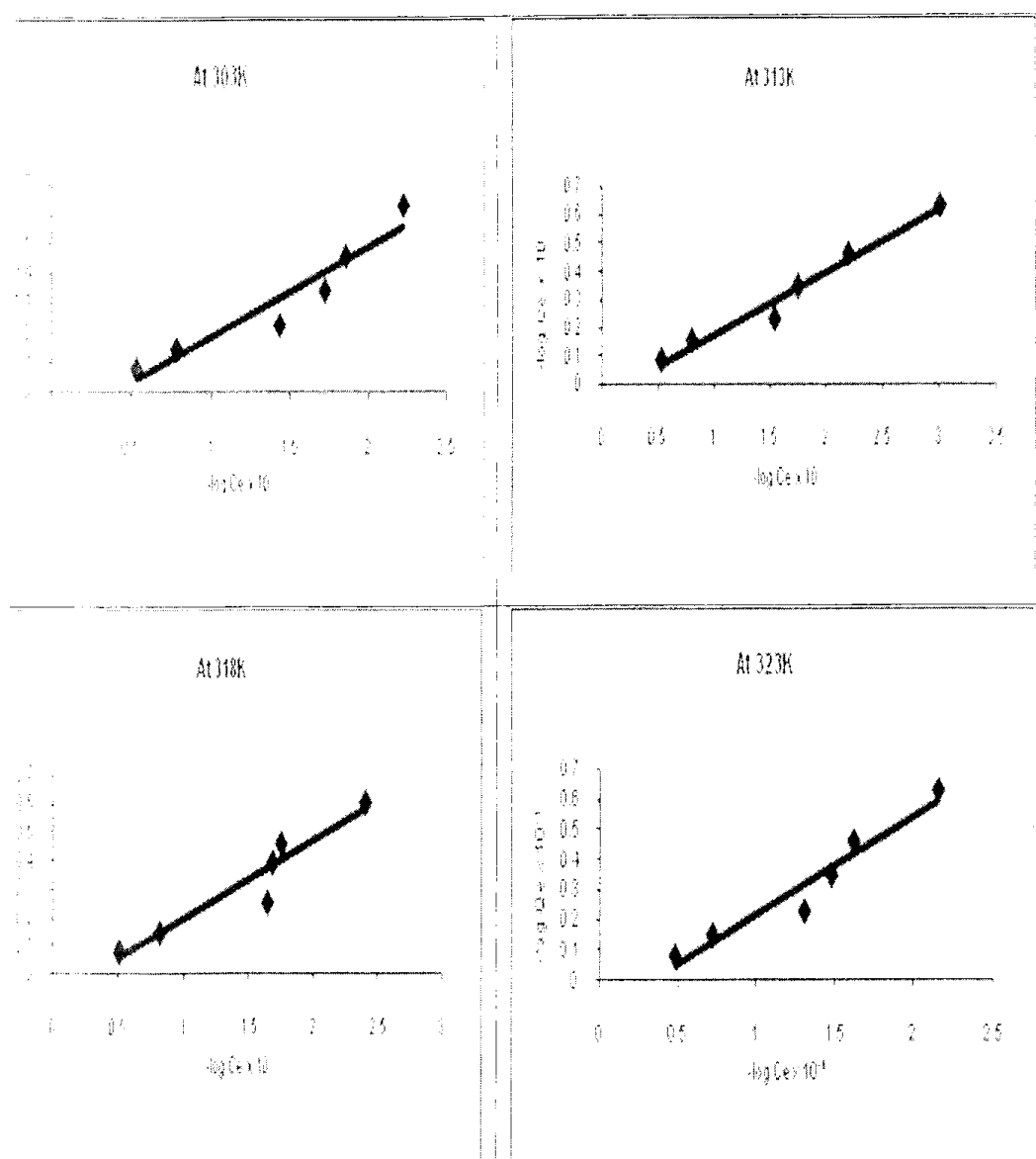


Fig (3):- Freundlich plots for bentonite/ Fe (II) ion system at different temperatures.

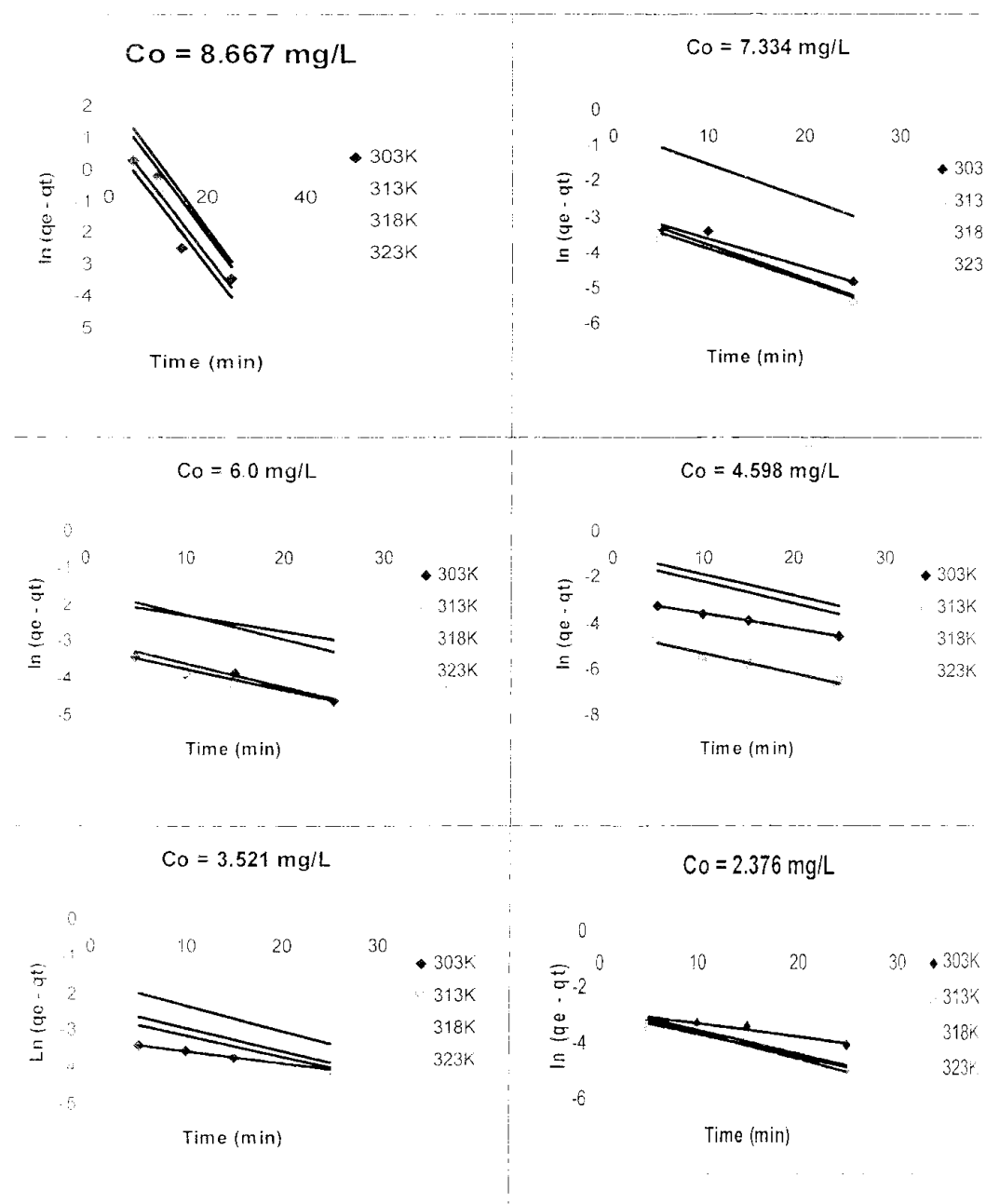


Fig (4):- Relation between $\ln(q_e - q_t)$ and time (min).

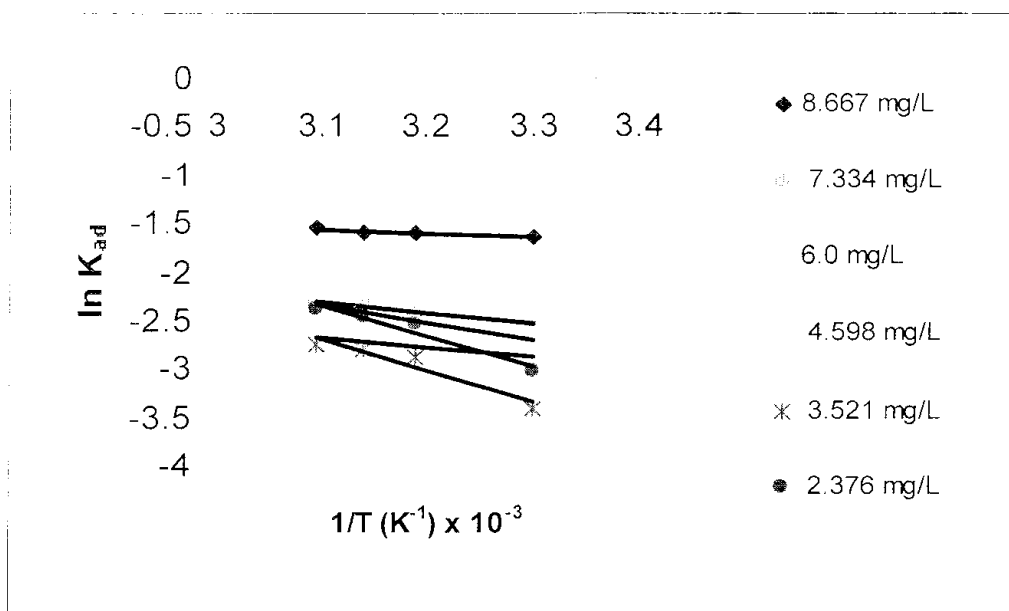


Fig (5):- Arrhenus equation of adsorption of Fe (II) ion on bentonite.

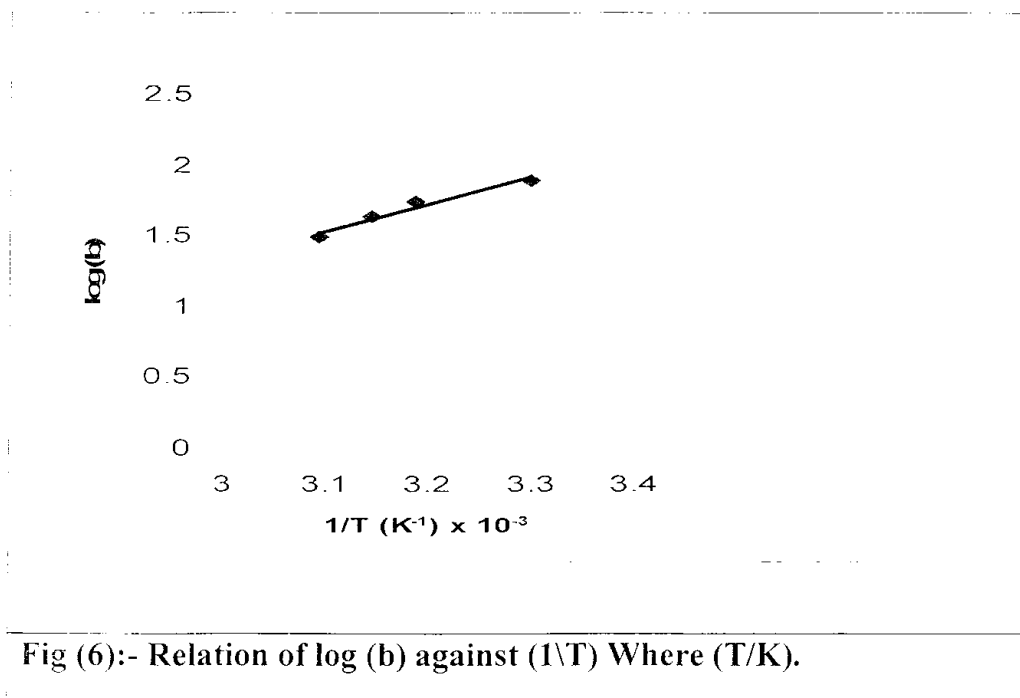


Fig (6):- Relation of log (b) against (1/T) Where (T/K).

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الخلاصة

يستخدم دواء (فيرفول) للعلاج والوقاية من مرض الانبمبا ويعتبر كمصدر للحديد
وحامض الفوليك يعطى للنساء الحوامل خلال فترة الحمل.
تمت دراسة امتزاز تراكيز مختلفة من المحلول المائي للحديد تراوحت من
(2.375.9.557) ملي غرام بالتر باستخدام طين البنتونايت عند أربع درجات حرارية
تراوحت من (303-323 كلفن) .
تم أيجاد سعة ونابت سرعة الامتزاز باستخدام طريقتي لنكمير وفرندلش، وكذلك تحديد
ثوابت الامتزاز، وتبين ان نوع متساوي درجة الحرارة هو (S) على وفق تصنيف جيل
لمتساويات درجة الحرارة. واستخدمت معادله لكار كرتين لمتابعة حركيه الامتزاز والتي تتبع
المعادلة من الرتبة الأولى. وأظهرت النتائج ان طين البنتونايت له كفاءة: واضحة لامتزاز
أيونات الحديد.