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Effect of Temperature on the Pb (II) Removal from Single Aqueous Solutions by a Locally Natural Mordenite: Equilibrium and Kinetic Modeling

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Abstract: Zeolites have been shown to be effective adsorbents for the removal of heavy metals from aqueous solutions. In this study, mordenite (a natural zeolite) has been used for the removal of lead ions to evaluate its potential use as a low-cost adsorbent. Batch experiments have been conducted to evaluate the equilibrium and process kinetics at different temperatures. The equilibrium experimental data for various temperatures studied conformed to the six adsorption isotherm equations: the Langmuir, Freundlich, Sips, Redlich-Peterson (RP), Dubinin-Radwhkevich (DR), and Flory-Huggins (FH). Constants were determined for each of the isotherms. The apparent thermodynamic parameters were calculated and the obtained values supported the conclusion that the lead ions adsorption onto mordenite was a spontaneous, exothermic process of physical nature. The kinetic experimental data fitted the pseudo-second-order, parabolic diffusion and Elovich equations successfully. The process of lead ions adsorption onto the Na-mordenite was diffusion-controlled. The value of apparent activation energy also confirmed a physical mechanism for the adsorption of lead ions onto Na-mordenite.

Keywords: Lead (II), mordenite, isotherm, kinetics, modeling

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INTRODUCTION

The increasing levels of heavy metals in the environment pose a serious threat to human health, living resources and ecological systems. Lead is one of the most common pollutants found in industrial effluents and can damage the nervous system, kidneys, and reproductive system, particularly in children. Lead has been found in at least 1026 of 1467 National Priorities List sites identified by the US Environmental Protection Agency (EPA). The EPA requires lead in drinking water not to exceed 0.015 mg/L (1). Consequently, various processes such as chemical precipitation, ion exchange, membrane separation, and activated carbon adsorption, etc., have been employed to remove lead ions from aqueous solutions. However, most of these methods suffer from some disadvantages including incomplete metal removal, toxic sludge generation, and high costs when applied to dilute effluents on a large scale. There is a crucial need for the development of methods that are not only cost effective, but can be easily implemented.

In this context, natural zeolites have started gaining interests for removing undesirable heavy metal ions from industrial and processing effluent water, mainly due to their valuable sorption characteristics provided by combination of ion-exchange and molecular sieve properties which can be relatively modified (2, 3). Zeolites are naturally occurring hydrated aluminosilicate minerals. Most common natural zeolites are formed by alteration of glass-rich volcanic rocks (tuff) with saline water. The structures of zeolites consist of three-dimensional frameworks of SiO_4 and AlO_4 tetrahedra. Zeolite utilization as ion-exchangers for environmental protection is stimulated by the non-toxic nature, their availability in the many parts of the world, the low cost, and the regenerative nature (3). Up to now, more than 150 zeolites have been synthesized and 40 naturally occurring zeolites are known. Among these natural zeolites, clinoptilolite and chabazite for example has received extensive attention due to their attractive selectivities for certain heavy metal ions. Ouki and Kavannagh evaluated the potential of the natural clinoptilolite and chabazite as adsorbents to remove the heavy metals (Pb, Cd, Cu, Zn, Cr, Ni, and Co) from wastewater (2). Inglezakis et al. investigated the ion exchange of Pb^{2+}, Cu^{2+}, Fe^{3+}, and Cr^{3+} on natural clinoptilolite (4). Chojnacki et al. utilized the natural clinotilolite as adsorbent for mercury removal (5). The selectivity series of natural clinoptilolite were determined by Erdem et al. as follows: $Co^{2+} > Cu^{2+} > Zn^{2+} > Mn^{2+}$ (6). Camilo et al. investigated the sorption characteristics of heavy metal ions by a natural zeolite and found the selectivity of the studied metals to be $Cu^{2+} >> Zn^{2+} > Ni^{2+}$ (7). Sprynskyy et al. investigated the selection mechanism of Pb^{2+}, Cu^{2+}, Ni^{2+}, and Cd^{2+} adsorption onto clinoptilolite and showed that the maximum adsorption capacity towards Cd^{2+} was 4.22 mg/g at an initial concentration of 80 mg/L and towards Pb^{2+}, Cu^{2+} and Ni^{2+} 27.7, 25.76 and 13.03 mg/g at 800 mg/L, respectively (2). Akgül et al. studied the removal of silver (I) from aqueous solutions with clinoptilolite and found the adsorbent was suitable for silver removal from aqueous media (8). Despite the great interest in ion exchange with clinoptilolite, only few reports exist on the investigation of utilizing the mordenite to remove the heavy metal ions from aqueous solutions.

Mordenite (a naturally occurring zeolite) is also abundant in nature and has a typical chemical formula of $(Ca, Na_2, K_2)_4\ Al_8Si_{40}O_{96} \cdot 28H_2O$. The morphology indicates an open structure of easy access, formed by open channels of 8- and 12-membered rings (2.6 × 5.7 and 6.7 × 7.0 Å, respectively). The rather low Si/Al ratio (5:1) may render mordenite advantageous for binding heavy metal ions from aqueous solutions (9). Wang et al. have investigated the Cu^{2+} and Ni^{2+} sorption characteristics using mordenite and found the mordenite suitable for the removal of Cu^{2+} and Ni^{2+} from aqueous solutions (10, 11). Equilibrium and kinetics are two aspects in the adsorption process. In this study, the aim is to evaluate the usefulness of mordenite as low cost sorbent material for the removal of lead ions from single aqueous solution. The effect of temperature on lead ions adsorption equilibrium and kinetics was investigated in detail.

MATERIALS AND METHODS

Naturally occurring mordenite was provided for study by China University of Mining and Technology. The mordenite was lightly ground and sieved. Fine particles with a diameter $<150\ \mu m$ were used to prepare the Na-mordenite. To prepare homoionic Na-mordenite, 30 g of mordenite was mixed with 500 ml of 1 M NaCl. After 24 h end-over-end shaking, the mordenite suspension was centrifuged at 3000 rpm and the supernatant was replaced with fresh 1 M NaCl solution. This procedure was repeated three times, followed by a three-fold washing with 500 ml de-ionized water. The Na-mordenite was dried at 60°C overnight and stored in polyethylene flask for further experiments.

The stock solution of lead (II) (2000 mg/L) was prepared in deionized water using lead nitrate (analytical grade). All working solutions were prepared by diluting the stock solution with deionized water.

XRD measurement was done with X-Ray Diffraction (D/Max-3B) using Cu Kα radiation. Sears' method was chosen to estimate the surface areas of the Na-mordenite (12). The copper bisethylenediamine complex method was used to determine the cation exchange capacity (CEC) of the Na-mordenite (13).

The adsorption experiments were carried out in 100 ml Erlenmeyer flasks by mixing together a constant adsorbent with a constant volume of the aqueous solution of lead ions. The contents in the flasks were agitated by placing them in constant temperature water bath thermostat for a known time interval. The mixture was then centrifuged and lead ions in the aqueous solutions were determined with Atomic Absorption Spectroscopy. The following conditions were maintained for different sets of experiments:

Effect of pH: Na-mordenite 2 g/L; lead ions concentration 40 mg/L; temperature 20°C; contact time 90 min; stirring speed 150 rpm; pH 2.0 to 6.0 at unit intervals

Kinetics: Na-mordenite 2 g/L; lead ions 40 mg/L; temperature 20, 30, 40°C; pH 6.0; stirring speed 150 rpm; contact time 5, 15, 30, 45, 60, 90 min

Equilibrium and Thermodynamics Na-mordenite: 2 g/L; temperature 20, 30, 40, 50°C; contact time 90 min; stirring speed 150 rpm; pH 6.0; lead ions concentration 20, 40, 60, 80, 100 mg/L

The amount of adsorbed lead ions (q, mg/g) and removal efficiency (%) were calculated as follows:

$$q = \frac{V(C_0 - C_e)}{M} \tag{1}$$

$$\text{Removal\%} = 100 \times \frac{C_0 - C_e}{C_0} \tag{2}$$

where V is the solution volume (L), M the amount of sorbent (g), and C_0 and C_e (mg/L) the initial and equilibrium lead ions concentrations, respectively.

The goodness of fit of the models to experimental data was checked by comparison of the correlation coefficient (r) and sum of squared errors (*SSE*), which is defined as:

$$SSE = \sum\left[\frac{(q_{\exp} - q_{theo})^2}{{q_{\exp}}^2}\right] \tag{3}$$

where q_{theo} is adsorption capacity obtained by calculating from the corresponding model (mg/g) and the $q_{\exp}$ is the experimental data of the adsorption capacity (mg/g).

RESULTS AND DISCUSSION

Adsorbent Properties

The XRD pattern of the adsorbent used in this study is shown in Fig. 1. The result suggests that the sample mainly consists of mordenite. An inconsistency was found in the evaluation of a specific surface area of zeolites in the literature. Some workers have reported that these materials have a surface of several hundred m^2/g, while others reported several m^2/g (5). This inconsistency could be linked with differences in the properties of zeolites, their origins, and the method of determination. In the present work, specific surface area

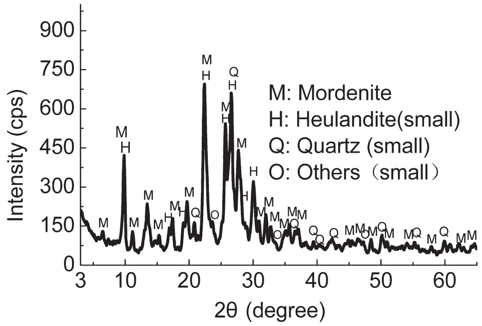

Figure 1. XRD pattern of the sample.

of the Na-mordenite was found to be rather low, 7.0 m^2/g. The CEC of Na-mordenite applied in this study was 28 meq/100 g.

Effect of pH

Solution pH seems to be the most important factor in the sorption process. The effect of initial solution pH on the adsorption of lead ions was studied by contacting 0.25 g Na-mordenite with 50 ml lead ions solution (40 mg/L). The pH values ranging from 2 to 6 were studied in the experimental run. Lead ions removal efficiency increased from 48.8 at pH 2 to 84.7% at pH 6 (Fig. 2). The decrease in removal efficiency at lower pH values may be either due to the competition between lead ions and H^+ (H_3O^+) ions for the same binding sites or be ascribed to the dissolution of aluminum and silicon from the framework at extreme pH conditions. In the subsequent studies, experiments were conducted in the solution pH value of 6 to avoid any possible hydroxide precipitation.

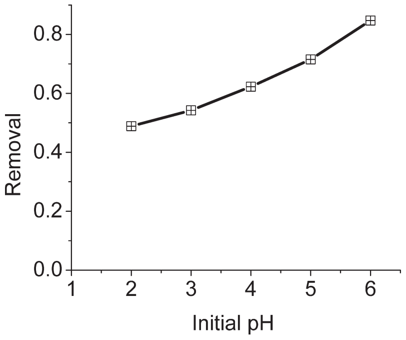

Figure 2. Effect of pH removal efficiency.

Equilibrium Studies

The analysis of the isotherm data is of particular importance to develop an equation which accurately represents the results and could be used for design purpose. In this study, the experimental data of lead ions onto Na-mordenite at various temperatures were fitted to the Langmuir, Freundlich, Sips, Redlich-Peterson (RP), Dubinin-Radwhkevich (DR), Flory-Huggins (FH) equations (Table 1) by non-linear or linear regression technique to determine the best fitting isotherm(s) and the corresponding parameters.

Langmuir Isotherm

The Langmuir isotherm is applicable to homogeneous adsorption where each lead ion/Na-mordenite sorption process has equal sorption activation energy. The Langmuir constants, $q_{L,m}$, b_L were determined using non-linear regression

Table 1. Summary of equilibrium models

Isotherms	Isotherms	Linear form	Equation no.	References
Langmuir	$q_e = q_{L,m}b_LC_e/1+b_LC_e$	$C_e/q_e = 1/q_{L,m}b_L + C_e/q_L$	(4)	(14)
Freundlich	$q_e = K_FC_e^{1/n_F}$	$\log q_e = \log K_F + 1/n_F \log C_e$	(5)	(15)
Sips	$q_e = q_Sb_SC_e^{1/n_S}/1+b_SC_e^{1/n_s}$	$\ln [q_S/q_e - 1] = -\ln b_S - 1/n_S \ln C_e$	(6)	(16)
Radlich-Peterson	$q_e = K_{RP}C_e/1+a_{RP}C_e^{n_{RP}}$	$\ln [K_{RP}C_e/q_e - 1] = \ln a_{RP} + n_{RP} \ln C_e$	(7)	(17)
Dubinin-Radwhkevich	$q_e/q_{DR} = \exp(-\beta\varepsilon^2)$	$\ln q_e = \ln q_{DR} - \beta\varepsilon^2$	(8a)	(18)
	$\varepsilon = RT \ln [1+(1/C_e)]$		(8b)	
Flory-Huggins	$\log \theta/C_0 = \log K_{FH} + n_{FH} \log (1-\theta)$		(9a)	(19)
	$\theta = 1 - C_e/C_0$		(9b)	

q_e (mg/g), C_e (mg/L): the sorption capacity and concentration of solute at equilibrium, respectively. $q_{L,m}$ (mg/g): the Langmuir monolayer sorption capacity. b_L (L/g): Langmuir constant related to the adsorption energy. K_F ((mg/g) (L/mg)$^{1/n}$) and n_F (dimensionless): the Freundlich constants. q_S (mg/g), b_S (L/mg) and n_S (dimensionless): the Sips isothermal constants. K_{RP} (L/mg), a_{KP} (L/mg) and n_{RP}: the Radlich-Peterson model constants. q_{DR} (mg/g), β (mol^2/kJ^2): the Dubinin-Radwhkevich constants. ε (kJ/mol): Polanyi potential. K_{FH} (L/mg), n_{FH} (dimensionless): the Flory-Huggins constants. θ (dimensionless): the degree of surface coverage. R (8.314 J/mol K): universal gas constants. T (K): absolute temperature.

analysis and are shown in Table 2. The equilibrium sorption capacity, $q_{L,m}$, was found to be increased from 13.95 to 24.48 mg/g for an increase in the reaction temperatures from 20 to 50°C.

The equilibrium parameter R_L, which is defined as $R_L = 1/(1 + b_L C_0)$, where C_0 is the initial concentration of lead ions, in the range $0 < R_L < 1$, reflects the favorable adsorption process (20). The values of R_L were found in the range $0 < R_L < 1$ and shown in Fig. 3, indicating that the adsorption

Table 2. Various isotherm models parameters for lead ions sorption onto Na-mordenite at different temperatures

	Temperature (K)			
	293	303	313	323
Langmuir				
$q_{L,m}$ (mg/g)	13.95	18.55	21.56	24.48
b_L	0.0132	0.0194	0.0182	0.0175
r	0.9850	0.9943	0.9948	0.9934
SSE	0.0218	0.0090	0.0109	0.0158
Freundlich				
K_F	0.5088	1.0799	1.1367	1.2110
n_F	1.6554	1.8594	1.8022	1.7664
r	0.9924	0.9991	0.9997	0.9994
SSE	0.0086	0.0006	0.0003	0.0005
Sips				
q_S	74.76	111.17	148.06	169.70
b_S	0.0062	0.0089	0.0071	0.0066
n_S	1.540	1.719	1.688	1.660
r	0.9919	0.9991	0.9997	0.9996
SSE	0.2018	0.3393	0.2423	0.2091
Redlich-Peterson				
K_{RP}	16.67	14.17	12.43	24.61
a_{RP}	32.21	12.63	10.42	19.79
n_{RP}	0.3985	0.4687	0.4531	0.4382
r	0.9924	0.9991	0.9998	0.9998
SSE	0.0087	0.0006	0.0003	0.0005
Dubinin-Radushkevitch				
q_{DR} (mg/g)	60.38	78.43	92.81	104.13
β	-0.0054	-0.0049	-0.0051	-0.0051
Es (kJ/mol)	9.57	10.08	9.96	9.95
r	0.9927	0.9993	0.9993	0.9986
Flory-Huggins				
Log K_{FH}	6.16	5.54	5.52	5.46
n_{FH}	3.56	3.42	3.54	3.61
r	0.9920	0.9993	0.9997	0.9998
ΔGa^0 (kJ/mol)	-34.5	-32.1	-33.1	-33.7

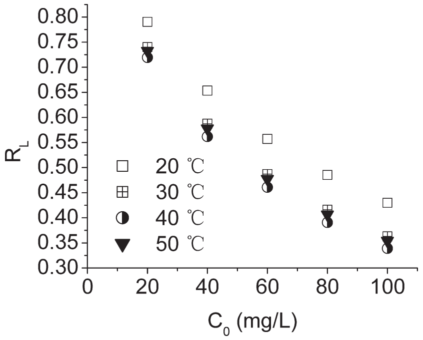

Figure 3. Plots of R_L against C_o at various temperatures.

process was favorable and the Na-mordenite exhibited a good potential for lead ions.

Freundlich Isotherm

The Freundlich equation is another widely used sorption isotherm. The calculated constants K_F, n_F are also given in Table 2. Value of K_F derived from the Freundlich equation is an indicator of the adsorption capacity of a given adsorbent. The results obtained in the present study indicate that the adsorption capacity (K_F) increased with the rise in the reaction temperatures ranged from 20 to 50°C. The exponents n_F are larger than unity at various temperatures, providing an indication of favorable process (21).

Sips Isotherm

The Sips isotherm equation is characterized by the heterogeneity factor, n_S, and it can be employed to describe the heterogeneity system. The values of the Sips isotherm constants derived from the non-linear method are given in Table 2 as well. The heterogeneity factors, n_S, are larger than unity at various temperatures, thereby indicating the heterogeneous nature of the surface of Na-mordenite. From the values of SSE of point of view, the experimental data do not obey better the Sips equation compared to the Langmuir, Freundlich and Redlich-Peterson isotherms.

Redlich-Peterson Isotherm

Redlich-Peterson equation was also used to fit the experimental data and the values of related parameters together with correlation coefficients calculated using a non-linear method are summarized in Table 2. The constant, n_{RP}, is less than unity, suggesting the non-linearity of the lead ions/Na-mordenite interaction (22).

Dubinin-Radushkevitch Isotherm

To distinguish the mechanisms involved in lead ions adsorption by the adsorbent, the Dubinin-Radushkevitch (DR) isothermal model, which is based on the Polanyi theory, was employed in this study. In its widely used form, this model relates the fractional coverage to the Polanyi potential (ε) as given in Eqs. 8 (a,b).

The values of Dubinin-Radushkevitch isotherm parameters are listed in Table 2. The maximum lead adsorption capacities, q_{DR}, from 20 to 50°C were 60.38, 78.43, 92.81, and 104.13 mg/g, respectively. The relatively high value predicted for the Na-mordenite/lead ions adsorption system is probably due to the shape of the equilibrium curve (22). To estimate the nature of interaction between lead ions and the binding sites, mean adsorption energy ($E_s = (2\beta)^{-0.5}$) per mole of the adsorbate, which is the energy required to transfer one mole of lead ions to the adsorbent surface from infinity in solution, was determined (Table 2).

The E_s value indicates the nature of interaction between adsorbate and adsorbent, as indicated in Table 3 (22).

The free energies of adsorption of lead ions onto Na-mordenite at various temperatures were 9.57, 10.08, 9.96, and 9.95 kJ/mol, respectively, suggesting that the interaction between lead ions and Na-mordenite proceeded via ion exchange. This result is in accordance with those obtained by Erdem et al. (6). They investigated the heavy metal ions (Co^{2+}, Cu^{2+}, Zn^{2+}, and Mn^{2+}) adsorption onto clinoptilolite and found the sorption energies to be 11.03, 11.95, 9.77, and 8.81 kJ/mol, respectively, at a temperature of 25°C.

Flory-Huggins Isotherm

To explain the nature of the adsorption of lead ions onto the Na-mordenite further, the adsorption isothermal model of Flory-Huggins was also employed. By plotting log (θ/C_0) versus log (1-θ), straight lines at various temperatures were obtained and the values of n_{FH} and log K_{FH} from the slope and intercept respectively were calculated and are represented in Table 2. The values of log K_{FH} ranged from 5.46 to 6.16 with a decrease in temperature from 50 to 20°C.

Table 3. The relations between the Es value and the nature of interaction between adsorbate and adsorbent

E_s value (kJ/mol)	Nature of interaction
8–16	Ion exchanges
<8	Physical adsorption

Thermodynamic parameters, such as the standard Gibbs free energy (ΔG_a^0), the adsorption enthalpy change (ΔH_a^0), and adsorption entropy change (ΔS_a^0) were determined using the following relationships:

$$\Delta G_a^0 = -2.303RT \log K_{FH} \tag{10}$$

$$\log K_{FH} = \frac{\Delta S_a^0}{2.303R} - \frac{\Delta H_a^0}{2.303RT} \tag{11}$$

where K_{FH} is the equilibrium constant of Flory-Huggins isotherm (Table 2).

The values of ΔH_a^0 and ΔS_a^0 were obtained from the slope and intercept of the Vant Hoff plots of log K_{FH} versus 1/T, shown in Fig. 4. The calculated values for ΔH_a^0 is −38.67 kJ/mol and ΔS_a^0 is −18.4 J/(mol K) for Flory-Huggins isotherm. The negative value of ΔH_a^0 suggests that the adsorption of lead ions is an exothermic process. However, the sorption capacity of lead ions increased with increasing temperature. A negative ΔH_a^0 was also reported for the sorption of lead ions onto china clay, wollastonite, bentonite and peat (23). The negative values of ΔG_a^0 as recorded in Table 2 indicate that the spontaneous nature of adsorption of lead ions onto Na-mordenite. The small negative value of ΔS_a^0 indicates that the spontaneous adsorption of lead ions with weak interaction with the surface of the adsorbent.

Kinetic Studies

Figure 5 shows that the variation of amount of lead ions adsorbed per unit mass of Na-mordenite with time at various temperatures. The result indicates that a major part of adsorption took place within the first 60 min of the process. The finding is in agreement with the result obtained by Sprynskyy et al. who investigated the selection mechanism of heavy metal adsorption on clinoptilolite (3). The rapid kinetics is of significant practical importance, as it will facilitate smaller reactor volumes ensuring high

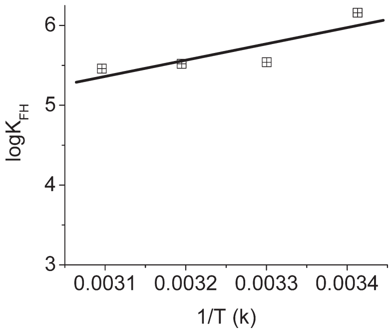

Figure 4. Plot of log K_{FH} against $1/T$.

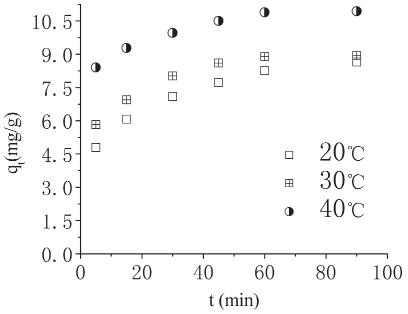

Figure 5. Variation of amount of lead ions adsorbed per unit mass of Na-mordenite with time at various temperatures (initial concentration = 40 mg/g).

efficiency and economy (24). To ensure complete equilibrium, in all equilibrium experiments performed in this work adsorbents were left shaking for 90 min.

Kinetic Modeling

Mathematical models that can describe the behavior of a batch adsorption process are very useful for process optimization. Numerous kinetic equations with varying degrees of complexity have been developed in batch systems (25). In this study, four mathematical equations (Table 4) are applied to describe the kinetics of lead ions adsorption on the Na-mordenite. Conformity of the data to the linear form of each kinetic model was tested using linear regression analysis. All models except for Lagregern's equation satisfactorily described the kinetics of lead ions adsorption onto Na-mordenite at the three temperatures studied (Fig. 6 and Table 5).

The pseudo second-order equation has been successfully applied to the adsorption of metal ions, dyes, herbicides, oils, and organic substances from aqueous solutions (28). In the present study, the experimental data fitted the pseudo second-order expression very well (Fig. 6(b) and Table 5). The value of apparent activation energy, E_a (kJ/mol), can be obtained with the help of the Arrhenius equation:

$$\ln k_2 = -\frac{E_a}{RT} + \ln A \tag{12}$$

where k_2 (g/mg min) is the rate constant of pseudo-second order equation (Table 5), A the Arrhenius factor, R the universal gas constant (8.314 J/mol K), and T the absolute temperature (K). By plotting ln k_2 versus 1/T (Fig. 7), apparent activation energy (E_a) was calculated from the slope and

Table 4. Kinetic equations applied in this study

Kinetic models	Equation	Equation no.	References
Lagergren's first-order equation	$\log(q_{e,L} - q_t) = \log q_{e,L} - (k_1/2.303)\,t$	(13)	(26)
Pseudo-second order equation	$t/q_t = (1/k_2 q_{e,P}^2) + (1/q_{e,P})t$	(14)	(25)
Parabolic diffusion equation	$q_t = m + k_d\, t^{0.5}$	(15)	(27)
Elovich equation	$q_t = \alpha \ln(\alpha\delta) + \delta \ln(t)$	(16)	(32)

Note: $q_{e,L}$ and $q_{e,P}$ (mg/g) are the sorption capacity at equilibrium, q_t (mg/g) the sorption capacity at time t (min), k_1 (min^{-1}) and k_2 (g/mg min) the rate coefficients, k_d (mg/g $min^{0.5}$) the overall diffusion coefficient; m, α, δ are constants.

found to be 10.85 kJ/mol. The value is quite low, particularly for a reaction type process and suggests a principally physical sorption process (29).

The parabolic diffusion model also described the lead ions adsorption process well (Fig. 7(c) and Table 5). Conformity to the parabolic diffusion

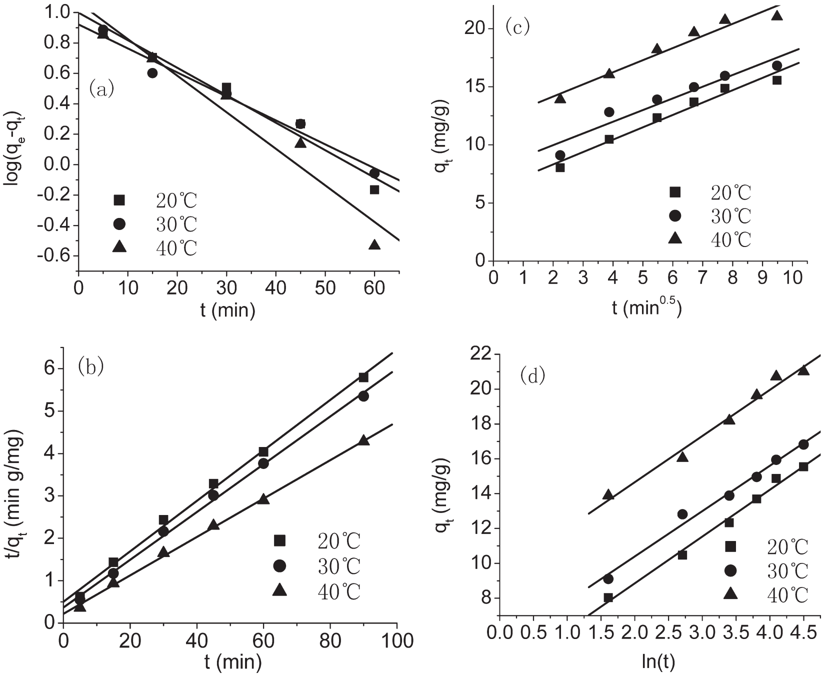

Figure 6. Linear fitting of experimental data to four models at various temperatures (a) Lagregern's first-order equation; (b) Pseudo-second order equation; (c) parabolic diffusion equation; (d) Elovich equation.

Table 5. Various kinetic models parameters for lead ions sorption onto Na-mordenite at different temperatures

	Temperature (K)		
	293	303	313
Lagregern's equation			
k_1 (min^{-1})	0.0416	0.0362	0.0553
r	0.9867	0.9866	0.9715
SSE	1.5135	2.4699	1.8396
Pseudo-second order equation			
k_2 (g/mg min)	0.0071	0.0087	0.0094
r	0.9979	0.9986	0.9992
SSE	0.0553	0.0285	0.0410
Parabolic diffusion equation			
K_d (mg/g min$^{0.5}$)	1.0625	1.0057	1.0418
r	0.9841	0.9613	0.9745
SSE	0.0092	0.0233	0.0782
Elovich equation			
α	2.6899	2.6026	2.6511
δ	1.3507	2.8271	12.8622
r	0.9955	0.9938	0.9907
SSE	0.0028	0.0032	0.0023

model suggests that the process of lead ions adsorption onto the Na-mordenite is diffusion-controlled; and either intra-particle diffusion or surface diffusion may be rate limiting.

The experimental data also fitted the Elovich model successfully (Fig. 6(d) and Table 5). The good fit of this model for the lead ions adsorption data proves its utility for the empirical prediction. The Elovich model may describe numerous processes including bulk and surface diffusion as well as

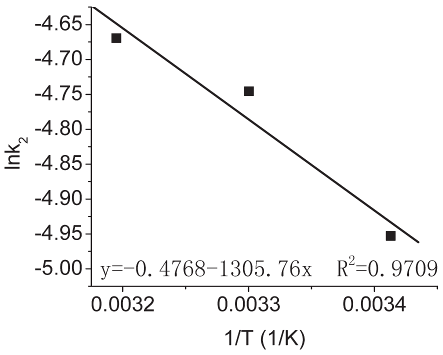

Figure 7. Plot of $\ln k_2$ versus $1/T$.

activation and inactivation of surfaces. However, prediction is still marginal at very low and very high surface coverage (30). Estimated α and δ values of the Elovich equation for lead ions adsorption by Na-mordenite were found to be temperature-dependent (Table 5). However, the chemical significance of these two constants has not been clearly resolved (30).

CONCLUSIONS

Removal of lead ions from single aqueous solutions by Na-mordenite was investigated. The Adsorption of lead ions onto Na-mordenite was pH-dependent. The equilibrium sorption data at different temperatures were fitted to the six adsorption isothermal models-Langmuir, Freundlich, Sips, Redlich-Peterson (RP), Dubinin-Radwhkevich (DR), and the Flory-Huggins (FH) isotherm. All isothems except for Sips' equation fitted the equilibrium experimental data very well on the basis of the values of correlation coefficient (r) and SSE. The sorption capacity increased with the rise in reaction temperature. The apparent thermodynamic parameters of sorption suggest that a spontaneous, exothermic sorption process of physical nature. The kinetic experimental data also fitted the pseudo-second-order, parabolic diffusion and Elovich equations successfully. The process of lead ions adsorption onto the Na-mordenite was diffusion-controlled. Compared to the various other sorbents reported in the literature, the mordenite in this study shows very good promise for practical applicability. However, more studies are needed to optimize the system from the regeneration point of view.

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