



Kinetic, Thermodynamic and Equilibrium Study on Removal of Lead(II) from Aqueous Solution Using Fly Ash

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Abstract

Batch experiments have been carried out to evaluate the removal of lead(II) ion from aqueous solutions by using fly ash under various conditions of metal ion concentration, contact time, pH, temperature and particle size. The adsorption data have been discussed using Freundlich and Langmuir adsorption isotherm models. The kinetics of adsorption have been discussed using Lagergren equation, pseudo second-order rate equation and intra-particle diffusion models. Thermodynamic parameters such as change in Gibbs free energy ΔG , change in enthalpy ΔH , and change in entropy ΔS have been evaluated. The adsorption process has been found to be spontaneous, feasible and endothermic in nature.

Keywords: Fly ash, adsorption, lead(II) ion, Langmuir isotherm, Lagergren first-order equation, pseudo-second-order equation, Intraparticle diffusion model.

Introduction

Lead (Pb) is one of the most toxic heavy metals and is widely used in many industries. The effluents from these industries cause pollution to soil and water. Among various methods to remove heavy metals from water, adsorption method is effective and economical. A large number of substances along with fly ash have been used as adsorbents¹⁻⁶.

In the present study, the effect of various experimental parameters such as initial Pb(II) concentration, contact time, temperature, pH and particle size have been investigated to evaluate the effectiveness of raw fly ash (a material produced from the combustion of coal in thermal power plant) as a potential and low cost adsorbent. Adsorption kinetics, isotherms and thermodynamic parameters have also been discussed.

Material and Methods

The fly ash used in this study was of JSPL, a coal burning power plant at Raigarh (C.G). Scanning electron micrographs, FTIR and XRF analysis were obtained from SAIF – IIT Bombay. These data were used to characterize the fly ash and its morphology. Stock solution of Pb(II) was prepared from A.R quality Pb(NO₃)₂.

Adsorption experiments were carried out by shaking 1.0 g of fly ash with 25 ml aqueous solution of Pb(II) of given concentration in different glass bottles. The progress of adsorption was determined by centrifuging the solution at different time intervals and analyzing by spectrophotometer. The influence of pH (2.0, 4.0, 6.5 and 8.0), initial Pb(II) concentration (100\150,

200 and 250 mgL⁻¹), contact time (20,40,60,80,100,120 and 140 min.), temperature (303K, 313K and 323K) and particle size (45 μ ,75 μ and 150 μ) were evaluated during the present study. For equilibrium study initial Pb(II) concentration used were 25,50,75,100,125,150,175,200,225 and 250 mgL⁻¹.

The amount of Pb(II) ions adsorbed in milligram per gram was determined by using the following mass balance equation⁷.

$$q_e = V (C_i - C_e) / m$$

where C_i and C_e are Pb(II) ion concentration in mgL⁻¹ before and after adsorption respectively, V is the volume of adsorbate in litre, and m is the weight of the adsorbent in grams. The percentage of removal of Pb(II) ion was calculated from the following equation⁷:

$$\text{Removal \%} = 100 (C_i - C_e) / C_i$$

Results and Discussion

Characterisation of fly ash: Table-1 shows the chemical composition of the fly ash sample obtained from XRF studies:

Table-1
Chemical composition of the fly ash

Constituent	wt (%)
SiO ₂	43.170
Al ₂ O ₃	13.248
Fe ₂ O ₃	41.198
CaO	1.090
MgO	0.727
TiO ₂	1.262

Different fly ash contain the same basic chemical elements but in different proportions. Table 1 shows that SiO₂ and Al₂O₃ contents make up about 56.42 % of fly ash. Fe₂O₃ and CaO contents compose about 42.29 %. This fly ash can be classified as class F⁸ as CaO content is less than 10% and SiO₂, Al₂O₃ and Fe₂O₃ content is greater than 70% .

Figure-1 is the SEM image of fly ash. It is clear that finer fly ash particles (45μ) are primarily spherical whereas the coarser particles (150μ) are mainly composed of irregular and porous structure. Figure- 1(c) is the SEM image of fly ash(45μ) after adsorption.

The FTIR spectra of fly ash before and after adsorption is shown in figure-2. It shows a broad band around 3500 cm⁻¹, which is attributed to surface -OH group of silanol groups (-Si-OH) and adsorbed water molecules on the surface. A peak around 1622.19 cm⁻¹ is attributed to bending mode (δ O-H) of water molecule⁸.

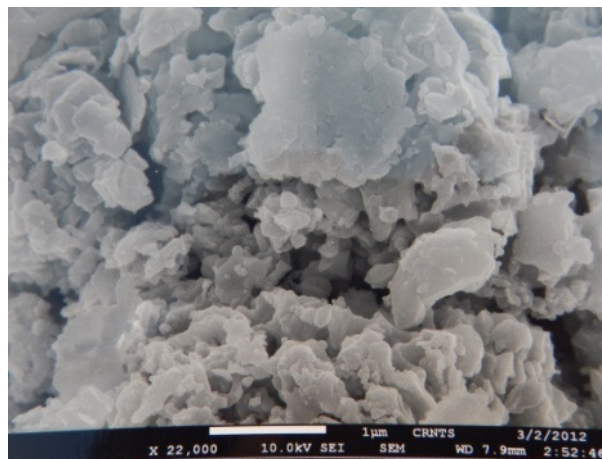


Figure-1(b)
 Before adsorption (150μ)

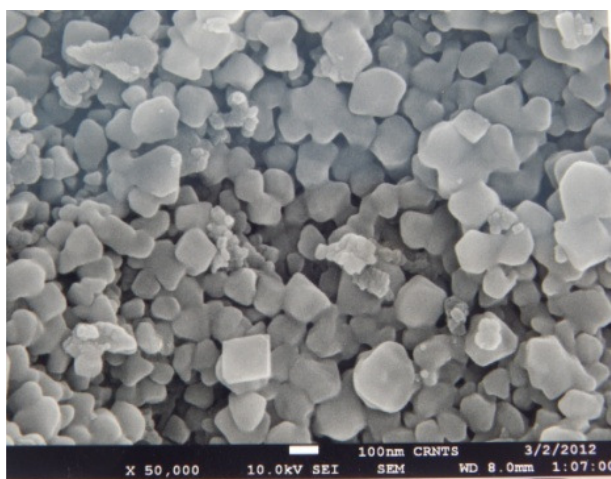


Figure-1(a)
 Before adsorption (45μ)

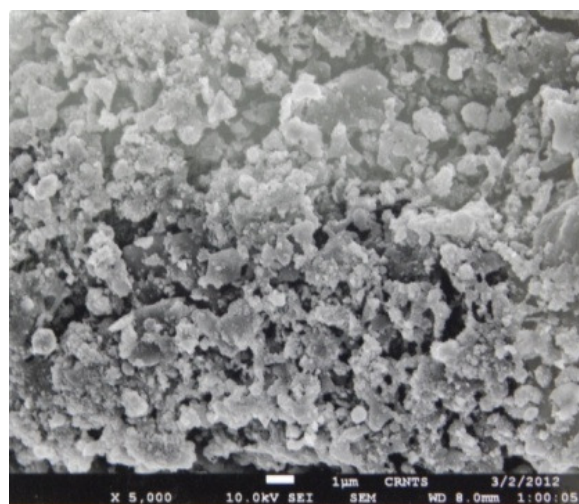


Figure-1(c)
 After adsorption

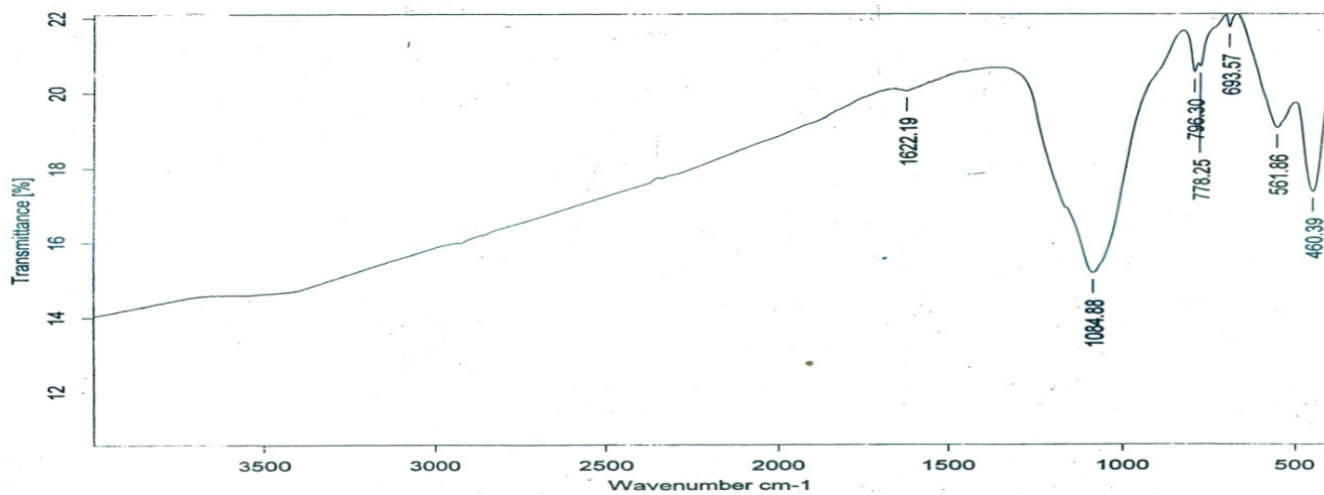


Figure-2
 (a) FTIR Before adsorption

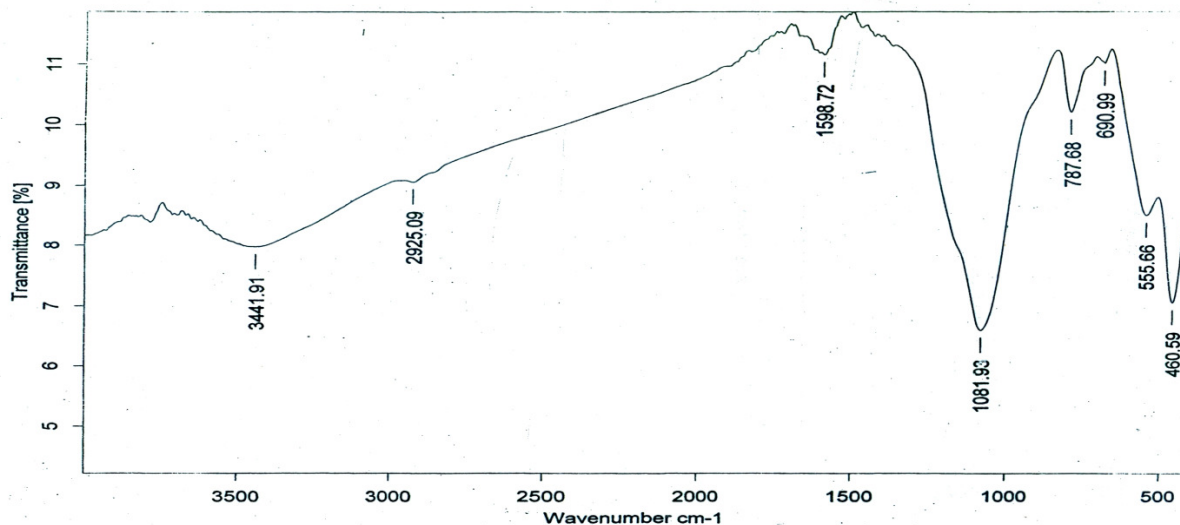


Figure-2
(b) FTIR After adsorption

The main broad band at 1084.88 cm⁻¹ in the fly ash before adsorption, corresponds to asymmetric stretching vibrations of Si-O-Si and Al-O-Si becomes sharper and shifts toward lower frequency 1081.93 cm⁻¹ as a result of the formation of new reaction products. The bands located at 796.30 cm⁻¹ and 460.39 cm⁻¹ are ascribed to bending vibrations of Si-O-Si and O-Si-O bonds, implying the presence of quartz. The bands located at 693.57 cm⁻¹ and 561.86 cm⁻¹ indicate the presence of mullite^{8,9}. After adsorption new bands appeared in 3444.91 cm⁻¹, 2925.09 cm⁻¹, 1598.72 cm⁻¹, 787.68 cm⁻¹ which indicate that adsorption has taken place.

Effect of initial Pb(II) ion concentration: Figure-3 shows the graph between percentage removal of Pb(II) ion with different initial concentration. It is clear that as initial Pb(II) ion concentration increases, the percentage removal of Pb(II) ion decreases from 81.2% at 100 mgL⁻¹ to 66.4% at 250 mgL⁻¹. It may due to the fact that adsorbents possess a limited number of active sites and these sites become saturated at certain concentration¹⁰.

It is evident from figure-4 that adsorbed amount at equilibrium, q_e (mgg⁻¹) increases with increase in initial concentration of Pb(II) ion from 2.03 mgg⁻¹ at 100 mgL⁻¹ to 4.14 mgg⁻¹ at 250 mgL⁻¹. The necessary driving force to overcome the mass transfer resistance of Pb(II) ion between the aqueous and the solid phase is possibly provided by the initial concentration of metal ion. The increase in Pb(II) ion concentration also increases the interaction between Pb(II) ions in the aqueous phase and the fly ash surface. This also resulted in higher uptake of Pb(II) for the given mass of fly ash¹⁰.

Effect of contact time: The removal of Pb(II) ion by adsorption on fly ash from aqueous solution increases with time (figure-5) till equilibrium attained in 120 min. It is evident from the graph that the time of saturation is independent of concentration.

Initially, the adsorption rate is fast. It may be due to more number of active sites on adsorbent surface. After that as active sites become less available, the adsorption is likely to be an attachment controlled process. Similar findings have been reported by other investigators¹¹⁻¹².

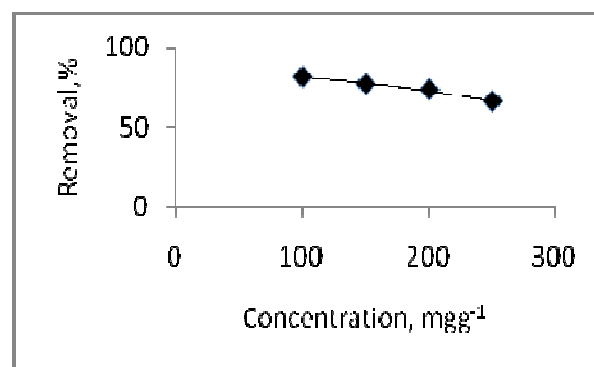


Figure-3
Effect of initial conc. on Pb(II) adsorption

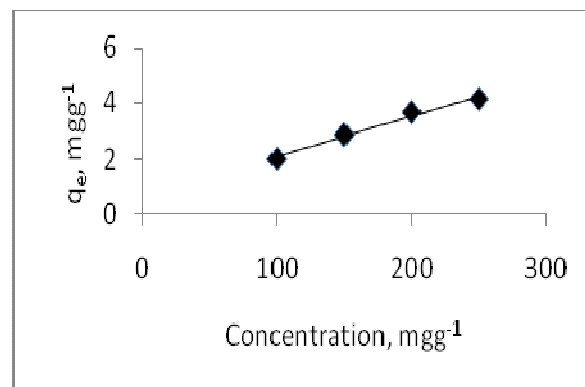


Figure-4
Effect of initial conc. on Pb(II) adsorption

Effect of pH: The pH of the solution has a significant impact on the uptake of heavy metals. Figure-6 shows the amount adsorbed (mgg^{-1}) at different pH. The amount of Pb(II) adsorbed on fly ash increases from 1.78 mgg^{-1} (71.2 %) to 2.35 mgg^{-1} (94.3 %) by increasing pH of solution from 2.0 to 8.0. Adsorption capacity ϕ and adsorption energy b also increases with increase of pH.

Studies have shown¹³ that fly ash particles are positively charged at low pH and negatively charged at high pH. With increase in pH, fly ash surface becomes more negatively charged. Thus, more favourable electrostatic attraction forces

enhance cationic metal ion adsorption. In alkaline medium both adsorption and precipitation take place.

Effect of temperature: Figure-7 shows the effect of temperature on adsorption. The result shows that adsorption of metal ion by the fly ash increases from 2.03 mgg^{-1} (81.2 %) to 2.38 mgg^{-1} (95.2 %) by increasing temperature from 303K to 323K indicating the process to be endothermic. The rate constant of adsorption are 1.8×10^{-2} , 2.1×10^{-2} and 2.3×10^{-2} per min at 303K, 313K and 323K respectively which indicate that the rate of adsorption also increases with temperature.

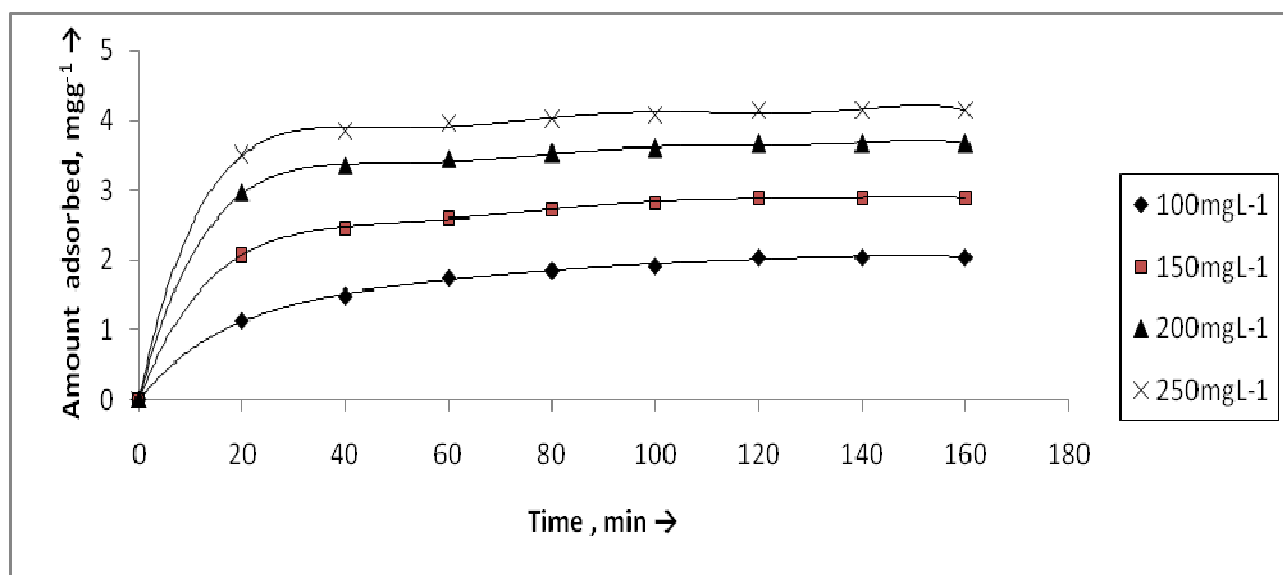


Figure-5
 Effect of contact time on adsorption of Pb(II) ion on fly ash

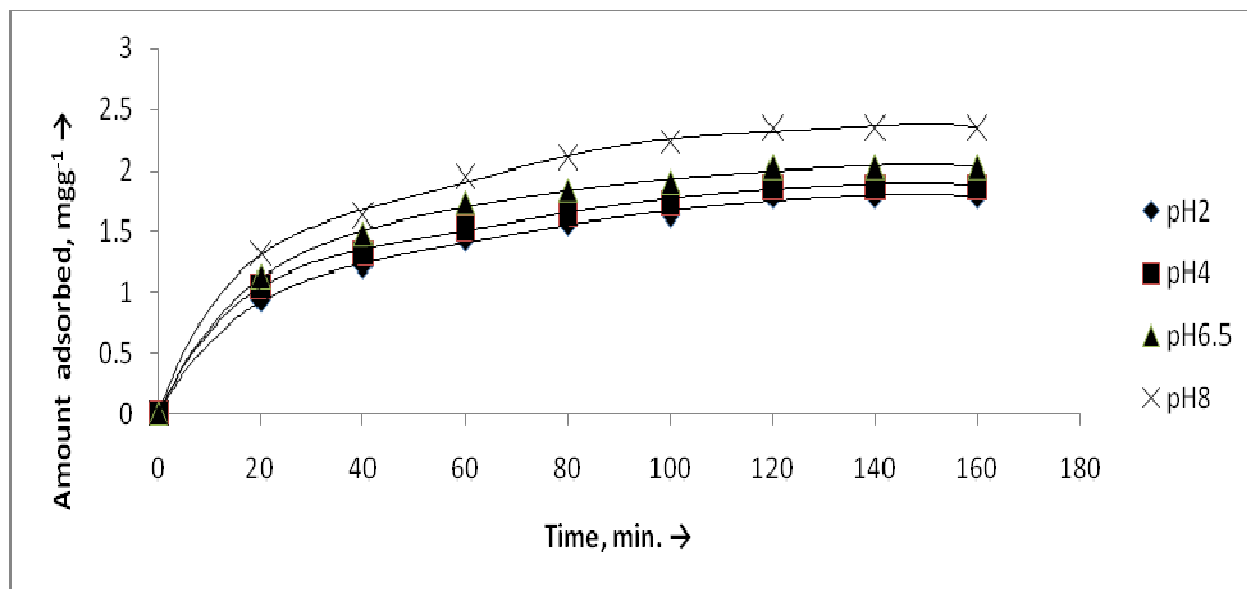


Figure-6
 Effect of pH on adsorption of Pb(II) ion on fly ash

Effect of particle size: Figure-8 shows the effect of particle size of fly ash on adsorption of Pb(II) ion. The amount of Pb(II) adsorbed on fly ash increases from 1.81 mgg⁻¹ (72.4%) to 2.03 mgg⁻¹ (81.2 %) by decreasing particle size of fly ash from 150 μ to 45 μ. This increase in amount of Pb(II) adsorbed on fly ash is due to increase in surface area of fly ash particles with decreasing particle size.

Adsorption Isotherm: The linear form of the Langmuir and Freundlich isotherm¹⁴ have been used to analyze the experimental data. The Langmuir isotherm is given by the following equation:

$$C_e/q_e = 1/\phi \cdot b + C_e/\phi$$

where C_e (mgL⁻¹) is equilibrium concentration of Pb(II) and ϕ and b are Langmuir constants. ϕ is related to adsorption capacity and b is related to adsorption energy. The plot of C_e/q_e versus C_e shown in figure-9 is linear which suggests that Langmuir isotherms is applicable. Values of ϕ and b have been calculated from slope and intercept respectively and are given in Table- 2. It is evident from the result that the values of ϕ and b increase on increasing the temperature.

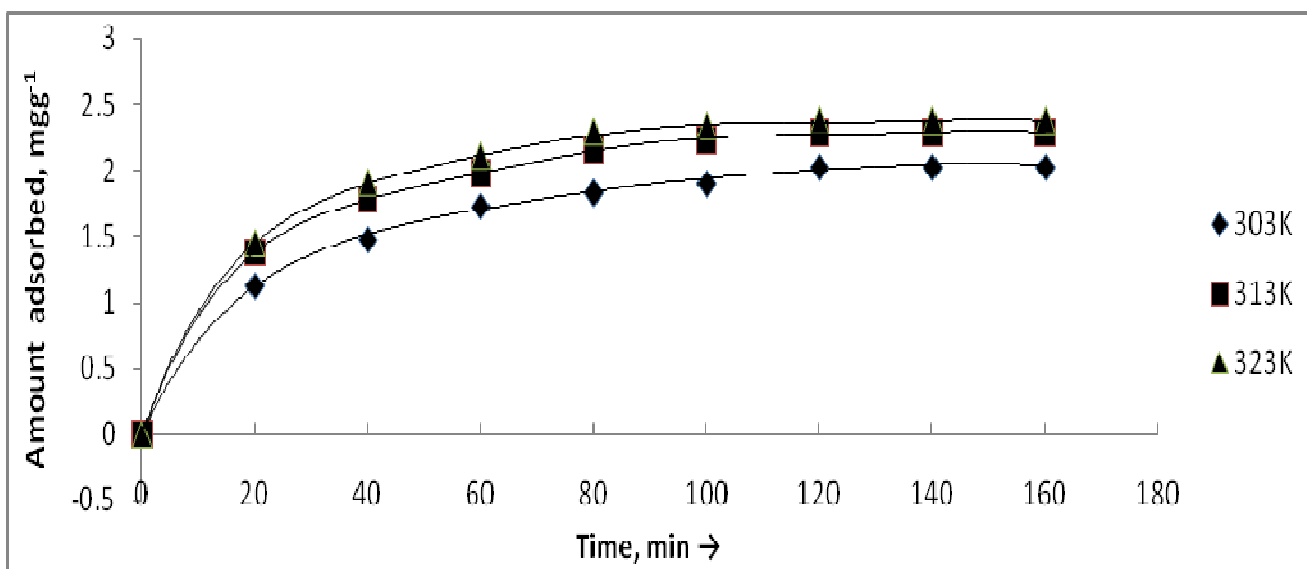


Figure-7
 Effect of temperature on adsorption of Pb(II) ion on fly ash

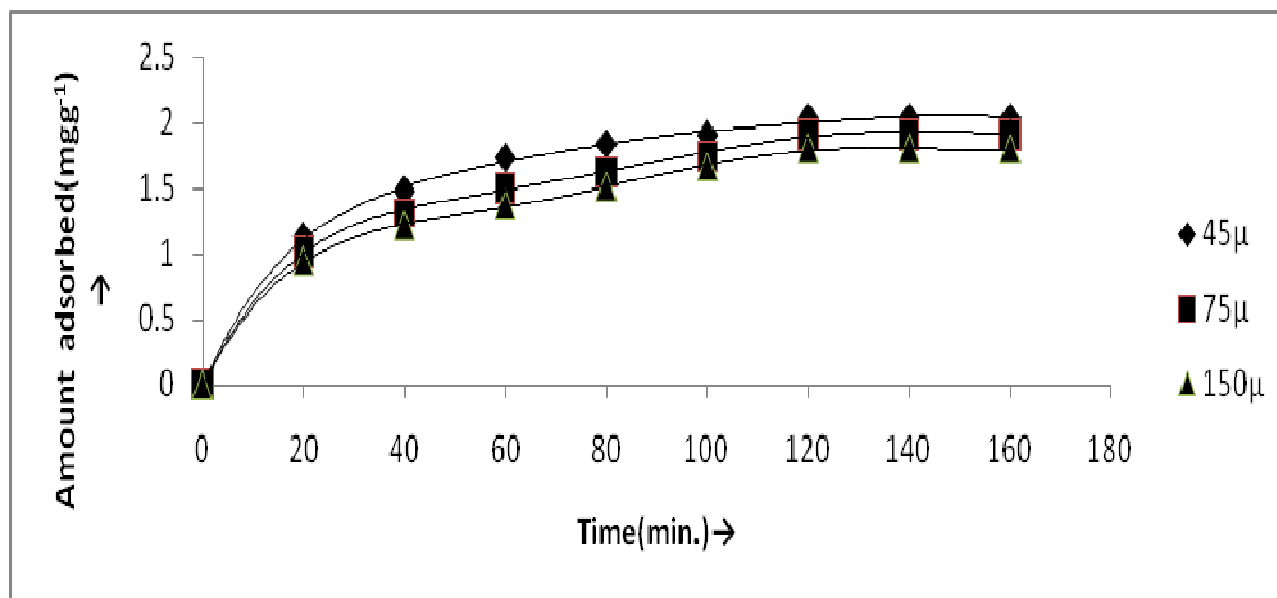


Figure-8
 Effect of particle size on adsorption of Pb(II) ion on fly ash

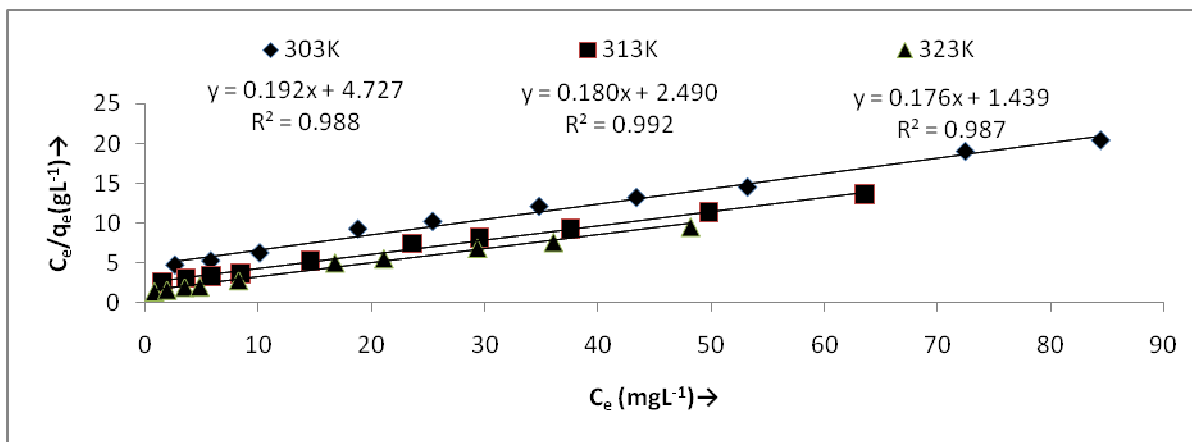


Figure-9
 Langmuir adsorption isotherm for the adsorption of Pb(II) ion on fly ash

Table-2
 Adsorption isotherm constants for adsorption of Pb(II) on fly ash

Langmuir Isotherm Results				Freundlich Isotherm Results		
Temp.(K)	Correlation coefficient, R ²	φ	b	Correlation coefficient, R ²	K _f	n
303	0.988	5.21	0.041	0.987	0.497	2.05
313	0.992	5.56	0.072	0.967	0.755	2.19
323	0.987	5.68	0.122	0.962	1.062	2.40

A dimensionless separation factor (R_L) gives important information about the nature of adsorption and is defined as¹⁵

$$R_L = 1/1+b.C_i$$

where C_i is the initial concentration in mgL⁻¹ and b is Langmuir constant (L/mg) related to adsorption energy. If 0 < R_L < 1, it indicates the adsorption process to be favourable and if R_L > 1 the process is unfavourable. Also it can be explained that when b > 0, adsorption system is favourable¹⁶. The calculated values are given in table- 3 . The values 0 < R_L < 1 and b > 0 suggest that the process is favourable.

Table-3
 Dimensionless separation factor (R_L)

C _i (mgL ⁻¹)	R _L		
	303 K	313 K	323 K
25	0.494	0.356	0.247
50	0.328	0.217	0.141
75	0.245	0.156	0.098
100	0.196	0.122	0.076
125	0.163	0.100	0.061
150	0.140	0.084	0.052
175	0.122	0.073	0.045
200	0.109	0.065	0.039
225	0.098	0.058	0.035
250	0.089	0.052	0.032

The Freundlich equation has also been used for the adsorption of lead (II) on fly ash which is represented as
 $\log q_e = \log K_f + 1/n \log C_e$

where q_e is the amount of lead ion adsorbed (mgg⁻¹), C_e is the equilibrium concentration of lead ion in solution (mgL⁻¹) and K_f and n are constants for the adsorption capacity and intensity of adsorption respectively. Plots of log q_e versus log C_e has been shown in figure-10 and values of K_f, n and R² (correlation coefficient) value have been obtained and given in table-2. Comparing R² value obtained from Langmuir plots and Freundlich plots clearly shows that the experimental data fits better in Langmuir equation.

Adsorption kinetics: The Lagergren first order¹⁷, pseudo-second-order¹⁸ and Intraparticle diffusion kinetic models¹⁹ have been employed to discuss the adsorption kinetics.

The Lagergren first order kinetic model: The Lagergren first order rate equation is represented as :

$$\log (q_e - q_t) = \log q_e - k_1.t/2.303$$

where q_e and q_t are the amounts of Pb(II) adsorbed (mgg⁻¹) at equilibrium and at time t, respectively. k₁ is the Lagergren rate constant (min⁻¹). Plots of log(q_e - q_t) versus t has been shown in figure- 11. Values of q_e and k₁ at different initial concentrations have been calculated from the slope and intercept respectively. These values have been given in table- 4.

The pseudo-second-order kinetic model: The adsorption data have been applied to pseudo-second-order kinetic model also. The equation is represented as:
 $t/q_t = 1/K_2 \cdot q_e^2 + t/q_t$

where K_2 is the rate constant of second order adsorption (g/mg/min.). Plots of t/q_t versus t has been shown in (figure-12). Values of K_2 and q_e have been calculated from the slope of the graph. These values have been given in table- 4.

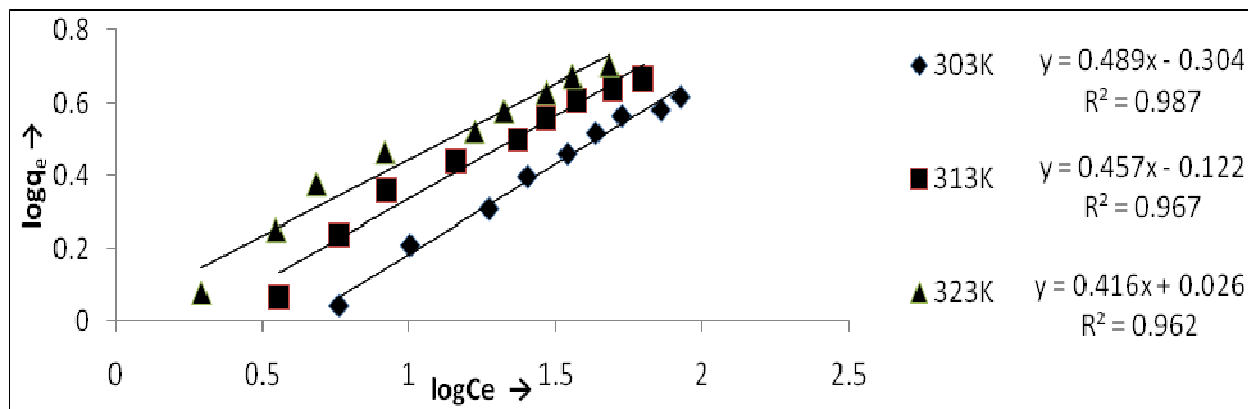


Figure-10
 Freundlich adsorption isotherm for adsorption of Pb(II) ion on fly ash

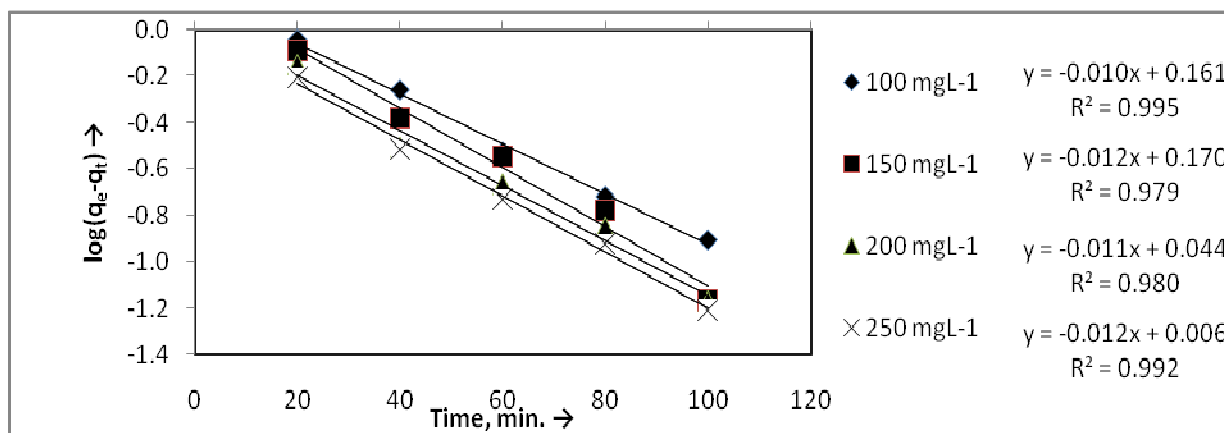


Figure-11
 Lagergren first-order kinetic plot for adsorption of Pb(II) ion on fly ash

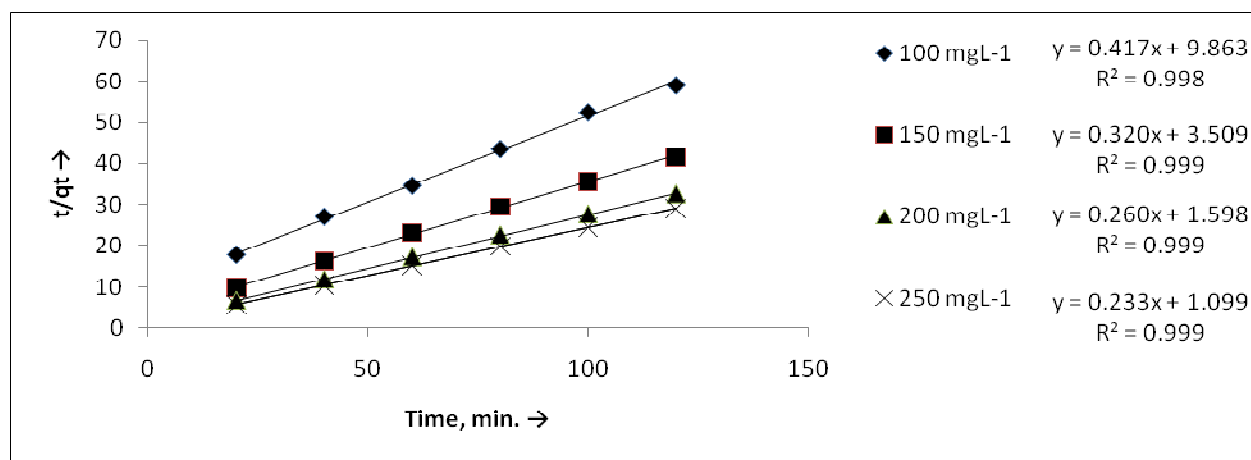


Figure-12
 Pseudo-second-order kinetic plot for adsorption of Pb(II) ion on fly ash

The Intraparticle diffusion model: The kinetic results were analyzed by the Weber and Morris intraparticle diffusion model¹⁹ to elucidate the diffusion mechanism. The model is expressed as:
 $q_t = K_d \cdot t^{1/2} + I$

where I is the intercept which reflects the boundary layer effect and K_d is the intra-particle diffusion rate constant. Plot of q_t versus $t^{1/2}$ has been shown in figure-13. The value of K_d and I have been calculated from the slope and intercept respectively and are given in table- 4. If the regression of q_t versus $t^{1/2}$ is linear and passes through the origin then Intraparticle diffusion is considered to be the sole rate-limiting step. As the linear plots did not pass through the origin, it is evident that intraparticle diffusion is not the only rate limiting step and some boundary layer effect is indicated.

It is evident from the table- 4 that the kinetic data are better explained by pseudo-second-order kinetic model as it shows high correlation coefficient ($R^2 > 0.99$) at all the studied concentration in comparison to the other kinetic models. Moreover, $q_{e(cal)}$ values agree better with the experimental data in the case of pseudo-second-order kinetic model.

Thermodynamic treatment of the adsorption process: In order to study the feasibility of the adsorption process, the thermodynamic parameters such as free energy, enthalpy and

entropy changes can be estimated from the following equations²⁰.
 $K_c = C_s/C_e$
 $\Delta G = -RT \ln K_c$
 $\log K_c = \Delta S/2.303 R - \Delta H/2.303 RT$

where C_e is the equilibrium concentration in solution in mgL^{-1} and C_s is the equilibrium concentration on the adsorbent in mgL^{-1} and K_c is the equilibrium constant. The Gibbs free energy (ΔG) was calculated from the above equation. The values of ΔH and ΔS have been calculated from the slope and intercept of the plot between $\log K_c$ versus $1/T$ (not shown). All these values are listed in table- 5.

The values of activation energy (E_a) and sticking probability (S^*) have been calculated from the experimental data. They were calculated using modified Arrhenius type equation related to surface coverage(θ) as follows²¹

$$\theta = (1 - C_e/C_i)$$

$$S^* = (1 - \theta)e^{-E_a/RT}$$

The sticking probability, S^* , is a function of the adsorbate/adsorbent system under consideration. It must satisfy the condition $0 < S^* < 1$. It depends on the temperature of the system. The values of E_a and S^* has been calculated from slope and intercept of the plot of $\ln(1-\theta)$ versus $1/T$ (not shown) respectively and have been shown in table-5.

Table-4
Kinetic parameters for adsorption of Pb(II) ion on Fly ash

Conc. mgL^{-1}	Lagergren first order				Pseudo- second- order			Intraparticle diffusion		
	K_1 min^{-1}	q_{exp} mgg^{-1}	q_{cal} mgg^{-1}	R^2	K_2 $g/mg/min$	q_{cal} mgg^{-1}	R^2	K_d $mg/g.min^{1/2}$	I	R^2
100	2.30×10^{-2}	2.03	1.45	0.995	1.76×10^{-2}	2.398	0.998	0.142	0.545	0.964
150	2.76×10^{-2}	2.88	1.48	0.979	2.9×10^{-2}	3.125	0.999	0.129	1.562	0.953
200	2.53×10^{-2}	3.67	1.11	0.980	4.2×10^{-2}	3.846	0.999	0.109	2.562	0.912
250	2.76×10^{-2}	4.14	1.02	0.992	4.9×10^{-2}	4.292	0.999	0.098	3.143	0.930

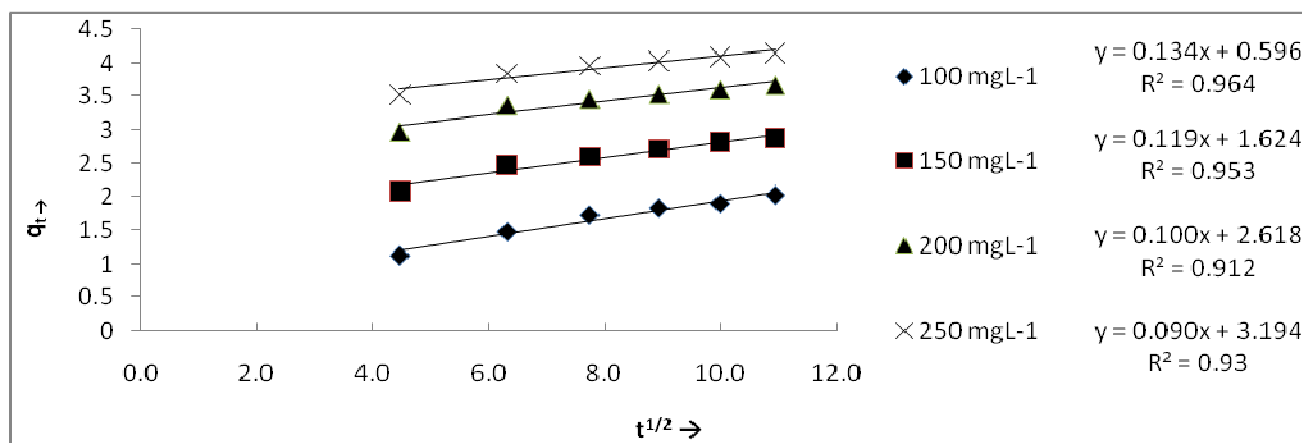


Figure-13
Intraparticle diffusion model for adsorption of Pb(II) ion on fly ash

Table-5
Thermodynamic parameters for adsorption of Pb(II) ion on fly ash

Temp. K	ΔG , kJ/mol	ΔH , kJ/mol	ΔS , J/mol	E_a , kJ/mol	S^* , J K mol ⁻¹
303	-3.686	62.13	217.51	55.64	4.68X10 ⁻¹¹
313	-6.218				
323	-8.020				

It is evident from table-5 that as ΔG values are negative, the process is spontaneous. Endothermic nature of adsorption is indicated by positive ΔH value. The positive value of ΔS shows the affinity of the adsorbent for the Pb(II) ions. The value of E_a has been found to be 55.64 kJ mol⁻¹ for the adsorption. The endothermic nature of the adsorption process is supported by positive value of E_a . This is in accordance with the positive values of ΔH . Since $S^* \ll 1$, it indicates that the probability to stick on surface of fly ash is very high²².

Mechanism: Speciation²³ of Pb(II) with varying pH has been shown in figure-14.

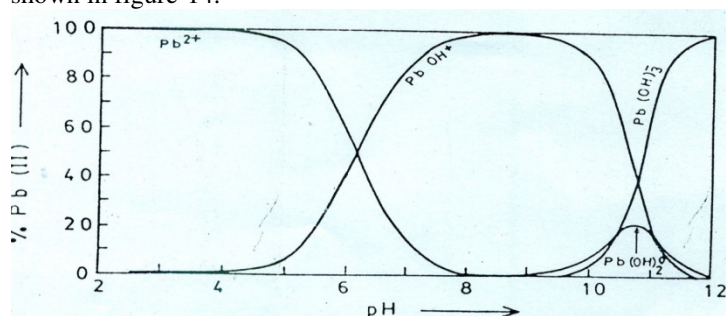
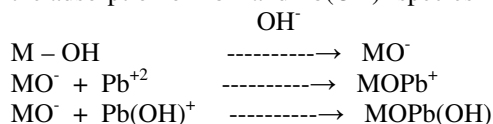


Figure-14
Speciation of Pb(II) with varying pH

It is evident that at lower pH, lead is in the form of Pb²⁺ and at ≈ 8 pH it is in the form of Pb(OH)⁺. It is probable that in acidic medium positively charged surface of adsorbent does not favour the association of cationic adsorbate species. In alkaline medium negatively charged surface offers the suitable sites for the adsorption of Pb²⁺ and Pb(OH)⁺ species^{24,25}.



Conclusion

It is evident that initial Pb(II) ion concentration, contact time, pH and temperature have marked effect on adsorption. The equilibrium data are best explained by Langmuir adsorption isotherm. Kinetics of adsorption follows second order rate equation. Thermodynamic parameters also favour the adsorption. It is expected that due to chemical composition, structure, more adsorption sites, cheap, availability in plenty etc. fly ash may prove to be an efficient adsorbent.

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