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### Removal of Pb (II) from aqueous solution by adsorption using activated carbon developed from Apricot stone: equilibrium and kinetic

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## Removal of Pb (II) from aqueous solution by adsorption using activated carbon developed from *Apricot stone*: equilibrium and kinetic

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### ABSTRACT

Activated carbons were developed from Apricot stone, by chemically treating with sulfuric acid. Batch adsorption experiments were performed to find out the effective lead removal at different metal ion concentrations, adsorbent size, and dosage. Adsorption of Pb<sup>2+</sup> ion was strongly affected by pH. Apricot stone exhibited the highest lead adsorption capacity at pH 6.0. Isotherms for the adsorption of lead on Apricot stone were evaluated with the Langmuir, Freundlich adsorption isotherm models. The equilibrium data fitted well to the Langmuir and Freundlich isotherm models. Adsorption kinetics data were modeled using the pseudo-first and pseudo-second-order models. The results indicate that the second-order model best describes adsorption kinetic data. The estimated maximum capacities of lead ions adsorbed by Apricot stone activated with sulfuric acid were 21.38 mg g<sup>-1</sup>. This high uptake showed Apricot stone activated carbon as among the best adsorbents for Pb(II).

*Keywords:* Lead removal; Adsorption; Isotherm; Activated carbon; Apricot stone

### 1. Introduction

Methods applied for removal of heavy metals from wastewater include chemical precipitation [1], solvent extraction [2] ultra-filtration [2], biochemical treatment [3], ion exchange [4], and adsorption [5–8]. Of these, adsorption, which is considered as a third stage of waste treatment, proved its advantage over the other processes because of its cost-effectiveness and the high-quality of treated effluent it produces. Adsorption is the process by which a solid adsorbent can attract a component in water to its surface and form

an attachment via a physical or chemical bond, thus removing the component from the fluid phase.

Activated carbon is the most effective adsorbent used; however, other different cheaper adsorbents are used or under investigations. Of these, peat was used to adsorb different heavy metals [9] and marine algae can adsorb Cd<sup>2+</sup> and Pb<sup>2+</sup> [10]. Different low-cost materials include clays, maize cob, deoiled soya, bagasse, palm fruit bunch which were used to remove dyes and heavy metals from water [11,12]. Also, microorganisms were found to be effective in the removal of heavy metal [13].

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Most of the activated carbons are produced by a two-stage process carbonization followed by activation. The first step is to enrich the carbon content and to create an initial porosity and the activation process helps in enhancing the pore structure. Basically, the activation is two different processes for the preparation of activated carbon: physical activation and chemical activation. In fact, there are two important advantages of chemical activation in comparison with physical activation. One is the lower temperature in which the process is accomplished. The other is that the global yield of the chemical activation tends to be greater since burn off char is not required. Among the numerous dehydrating agents, sulfuric acid in particular is the widely used chemical agent in the preparation of activated carbon.

The aim of this investigation was to determine the adsorption behavior of lead (II) on activated carbon developed from Apricot stone. The effects of the initial lead concentration, temperature, and pH on adsorption were investigated systematically, and multilinear mathematical model representing the adsorption behavior of the lead (II) was proposed within the experimental conditions for the activated carbon samples. Most of the experiments in this study were performed by using distilled water containing different lead concentrations. The thermodynamic parameters such as  $\Delta G$ ,  $\Delta H$ , and  $\Delta S$  from the adsorption measurements in the present study are very useful in elucidating the nature of adsorption

## 2. Materials and methods

### 2.1. Preparation of activated carbon

Apricot fruits are widely consumed fresh and in processed forms as juice and jams. Apricot stone, a by-product of the Apricot juice industry, is therefore an inexpensive. The *Apricot stone* were washed with distilled water, dried at 105°C, and crushed in a mortar. Sulfuric acid as a chemical reagent added during the activation process helps produce more effective adsorbent. The activation procedure was accomplished as: *Apricot stone* was impregnated with 50%  $H_2SO_4$  then this mixture of  $H_2SO_4$  and untreated *Apricot stone* was heated at 200°C for 2 h and then filtered and the resulting chemical loaded *Apricot stone* was placed in a furnace and heated ( $10^\circ C min^{-1}$ ) to the final carbonization temperature of 650°C for 30 min [11]. In all experiments, the heating rate was kept constant. After cooling, the activated carbon was washed to get it acid free and its pH was checked. The carbonized adsorbents were dried and rewashed many times until its pH reaches 4. After this, the

Table 1  
Main characteristic of the adsorbent

Parameters	ASAC
Pore volume ( $cm^3 g^{-1}$ )	0.192
BET surface area ( $m^2 g^{-1}$ )	393.2
Iodine number ( $mg g^{-1}$ )	154
Methylene blue number ( $mg g^{-1}$ )	91
Bulk density	0.81
Acidic surface functional groups ( $mmol g^{-1}$ )	0.78

activated carbon was deacidified by use of water and ammonical solution in later stages. Deacidification was done in following manner. After 20–25 times, washing carbon was treated with liquid aqueous ammonia solution in such a fashion that it does not affect the surface properties. Then, activated carbon was dried to 60°C [11]. The carbonized material was sieved to 250–125  $\mu m$  size and used for adsorption experiments. The specific surface area of the activated carbon was measured by  $N_2$  gas adsorption at  $-196^\circ C$  (COULTER-SA3100). BET equation was used to calculate the specific surface area. The physical characteristics of the adsorbent are given in Table 1.

### 2.2. Method of experiment

Batch reactor tests were carried out to study the effect of various operating variables on the adsorption rate. Adsorption of Pb(II) on developed activated carbon was conducted containing different weighted amounts of each sample with 100 ml solution of 10, 30, 50, 80, and 100 mg/l of initial concentration. Analytical grades of  $Pb(NO_3)_2$ , HCl, and NaOH were purchased from Merck (India) Lt. Lead ions were prepared by dissolving its corresponding nitrate salt in deionized water. The pH of solutions was adjusted with 0.1N HCl and NaOH. All the experiments were conducted five times, and the average values have been reported. Also, blank experiments were conducted to ensure that no adsorption was taking place on the walls of the apparatus used.

### 2.3. Adsorption experiments

Batch adsorption experiments were carried out in a series of stoppered reagent bottles. A weighed amount (0.1 g) of adsorbent was introduced into reagent bottles (200 ml) containing various concentrations with 100 ml aqueous solutions of copper or cadmium. The solution pH was adjusted to the desired value by adding HCl or NaOH. Then, the bottles were shaken at room temperature ( $20 \pm 2^\circ C$ ) using a mechanical shaker for a

prescribed time to attain the equilibrium. The solutions were filtered and the concentrations of metal ions were determined by AAS method. The effects of concentration (10–100 mg/l), contact time (2–180 min), solution pH (1.5–9), and adsorption dose (0.2–2 g) were studied. Blank solutions were treated similarly (without adsorbent) and the recorded concentration by the end of each operation was taken as the initial one.

The percent removal of heavy metal from solution was calculated by the following equation:

$$\text{Removal (\%)} = \frac{(C_0 - C_e)}{C_0} 100 \quad (1)$$

where  $C_0$  is initial concentration of heavy metal,  $C_e$  is the equilibrium concentration (mg/L) of lead. Isotherm studies were recorded by varying the initial concentrations of heavy metal solutions from 10 to 100 mg/L for lead. A known amount of different activated carbons is then added into solutions followed by agitating the mixture at 150 rpm till equilibrium. The adsorption capacity  $q_e$  (mg/g) after equilibrium was calculated by mass balance relationship equation as follows

$$q_e = \frac{(C_0 - C_e) \times V}{m} \quad (2)$$

where  $V$  is the volume of the solution (L) and  $W$  is the mass of adsorbate (g).

#### 2.4. Adsorption thermodynamics

The thermodynamic effect was studied by agitating 20 mg l<sup>-1</sup> of Pb(II) solution with 0.1 g of adsorbent at temperature 30, 35, 40, and 45°C for a time period of 20 min. Adsorption of Pb(II) on the walls of glass flasks and centrifuge tubes, which was found negligible, was determined by running blank experiments. Each experiment was carried out in duplicate and the average results are presented in this work.

### 3. Results and discussions

#### 3.1. Effect of agitation time and initial concentration on Pb (II) adsorption

Fig. 1 shows the effect of agitation time and Pb(II) concentration on removal. This figure shows that the adsorption of Pb (II) increases with an increase in agitation time and attains equilibrium in 10 min for 10 mg l<sup>-1</sup> Pb(II) and 20 min for 30, 50, 80, and 100 mg l<sup>-1</sup> Pb(II). It is very clear from the results that the agitation time required for maximum uptake of metal ions

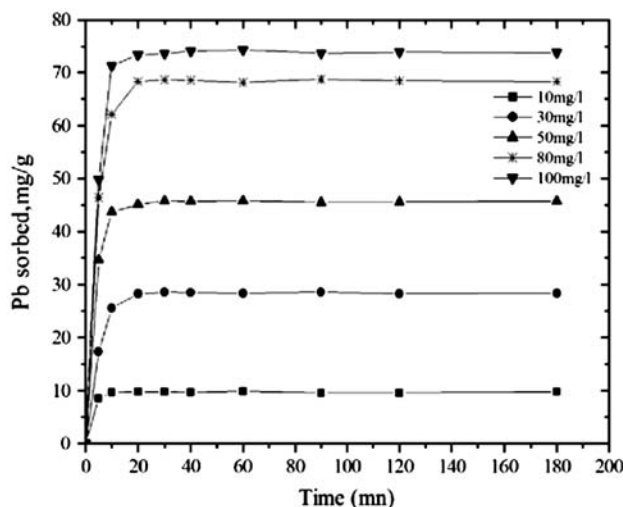


Fig. 1. Effect of contact time for the adsorption of lead ions onto activated carbon from Apricot stone at  $T=20^\circ\text{C}$ ,  $\text{pH}=6$ ,  $S/L=1\text{ g/L}$ .

by carbon was dependent on the initial Pb(II) concentration. This result is interesting because equilibrium time is one of the parameters for economical wastewater treatment plant applications. According to these results, the agitation time was fixed at 5 h for the rest of the batch experiments to make sure that equilibrium was reached. It also shows that increases in metal ion concentration increased the amount of metal ion uptake per unit weight of carbon (mg/g).

#### 3.2. Effect of adsorbent concentrations

Fig. 2 shows the effect of adsorbent concentration on the % removed at equilibrium conditions. It was observed that the amount of lead adsorbed varied with varying adsorbent concentration. The amount of lead adsorbed increases with an increase in adsorbent concentration from 0.2 to 2 g. The percentage of lead removed was increased from 86.51 to 98.13% for an increase in activated carbon concentration from 0.2 to 2 g. This is due to the greater availability of the exchangeable sites or surface area at higher concentration of the adsorbent.

For the complete removal of Pb (II) from 1,000 ml of 50 and 100 mg l<sup>-1</sup> a maximum carbon concentration of 1.5 and 2.0 g l<sup>-1</sup>, respectively, is required.

#### 3.3. Effect of particle size on Pb (II) removal

The percent removal of Pb (II) decreased with increasing adsorbent particle size. The percent removal for particle size ranges of 750–500, 500–250  $\mu\text{m}$ , and 250–125  $\mu\text{m}$  was 76.47, 81.65, and 86.74%, respectively.

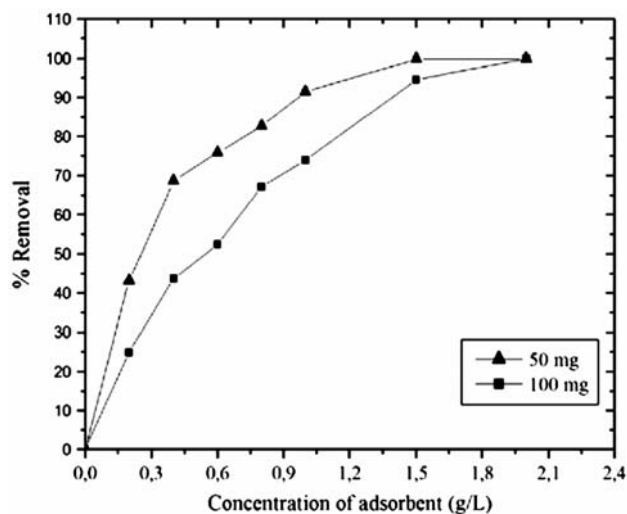


Fig. 2. Effect of adsorbent concentration for the adsorption of lead ions onto activated carbon from Apricot stone at 20°C, initial concentration 50 and 100 mg/l, pH=6.

The adsorption of Pb (II) attains equilibrium in 120, 90, and 60 min for particle size ranges of 750–500, 500–250  $\mu\text{m}$ , and 250–125  $\mu\text{m}$  with surface areas 212, 278, and 393  $\text{m}^2\text{g}^{-1}$ , respectively. This is because adsorption is a surface phenomenon, so the smaller adsorbent particle size offers a comparatively larger and more accessible surface area and hence, higher adsorption occurs at equilibrium.

### 3.4. Effect of pH

The pH of the solution has a significant impact on the uptake of heavy metals, since it determines the surface charge of the adsorbent, the degree of ionization, and speciation of the adsorbate. In order to establish the effect of pH on the biosorption of lead (II) ion, the batch equilibrium studies at different pH values were carried out in the range of 1.5–9 (Fig. 3). Fig. 3 shows that heavy metal removal capacity increased very sharply with an increase in pH from 4.5 to 7 and significantly decreased by reducing the pH values and slightly decreased at higher pH values. According to Low et al. [12], little sorption at lower pH could be ascribed to the hydrogen ions competing with metal ions for sorption sites. This means that at higher  $\text{H}^+$  concentration, the biosorbent surface becomes more positively charged, thus reducing the attraction between adsorbent and metal ions. In contrast as the pH increases, more negatively charged surface become available, thus facilitating greater metal uptake [13]. At a higher pH, the lead ion precipitated as their hydroxides which decreased the rate of adsorption and subsequently the percent removal of metal ion.

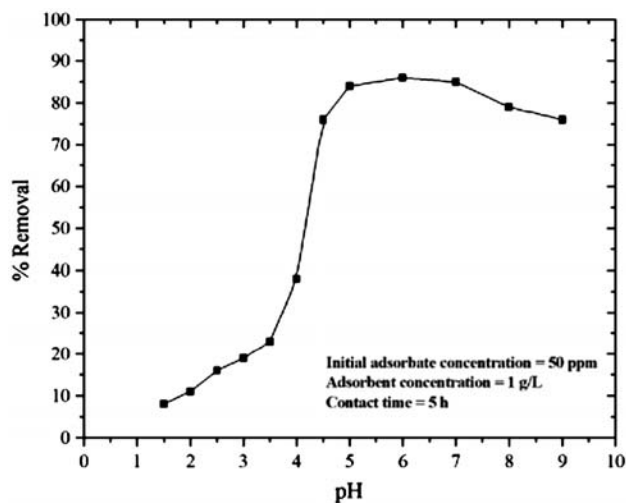


Fig. 3. Effect of pH on removal of lead and zinc from aqueous solution by activated carbon from Apricot stone at  $T=20^\circ\text{C}$ .

### 3.5. Kinetics of adsorption

The kinetic results for lead are shown Fig. 4 where the metal sorbed on activated *Apricot stone* is plotted against the equilibration time. The experimental data were analyzed with the utilization of a pseudo-second-order adsorption kinetic model [14]. The first-order rate equation of Lagergren is one of the most widely used for the sorption of a solute from liquid solution [15] and is represented as:

$$\ln(q_e - q_t) = \ln q_e - K_{\text{lad}} t \quad (3)$$

Where  $q_e$  is the mass of metal adsorbed at equilibrium (mg/g),  $q_t$  is the mass of metal adsorbed at time  $t$  (mg/g),  $K_{\text{lad}}$  is the first-order reaction rate constant (1/min).

The pseudo-first order considers the rate of occupation of adsorption sites to be proportional other number of unoccupied sites. A straight line of  $\ln(q_e - q_t)$  vs.  $t$  indicates the application of the first-order kinetic model. In addition, a pseudo-second-order equation based on adsorption equilibrium capacity may be expressed in the form:

$$\frac{t}{q_t} = \frac{1}{K_{2ad} q_e^2} + \frac{1}{q_e} t \quad (4)$$

Where  $K_{2ad}$  is the second-order reaction rate equilibrium constant (g/mg min). The plots of  $t/q_t$  vs.  $t$  (Fig. 4) were used to determine the rate constants and correlation coefficients. Table 2 shows the pseudo-second-order rate constants and correlation coefficients for removal of lead from aqueous

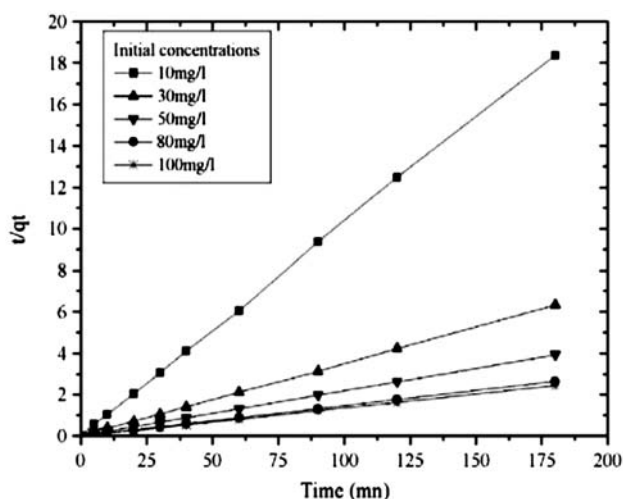


Fig. 4. Kinetics of lead removal according to pseudo-second-order model at initial lead concentration of 10, 30, 50, 80, and 100 mg/l; pH=6; S/L=1 g/1L.

solutions. The calculated correlation coefficients are less than 0.963 for the first-order kinetic model, whereas the values of the correlation coefficient are greater than 0.999 for the pseudo-second-order kinetic model. Therefore, the adsorption kinetics could be well explained and approximated by the pseudo-second-order kinetic model for the activated carbon from Apricot stone. These results suggest that the pseudo-second-order mechanism is predominant and that chemisorption might be the rate-limiting step that controls the adsorption process. Kinetic parameters for the adsorption of  $Pb^{2+}$  ions onto activated carbon by Apricot stone at various initial concentrations.

### 3.6. Adsorption isotherms

In order to successfully represent the equilibrium adsorptive behavior, it is important to have a satisfactory description of the equation state between the two phases composing the adsorption system. Two kinds of several isotherms equations were tested to fit the experimental data [16]:

$$\text{Langmuir isotherms : } C_e/q_e = 1/q_{\max}b + (C_e/q_{\max}) \quad (5)$$

$$\text{Freundlich isotherm : } \ln q_e = \ln K + (1/n) \ln C_e \quad (6)$$

where  $q_e$  is the amount adsorbed at equilibrium (mg/g) and  $C_e$  is the equilibrium concentration of metal ions in solution (mg/L). The other parameters are different isotherm constants, which can be determined by regression of the experimental data. In the Langmuir equation,  $q_{\max}$  (mg/g) is the measure of adsorption capacity under the experimental conditions and  $b$  is a constant related to the energy of adsorption. Freundlich treatment gives the parameters,  $n$ , indicative of bond energies between metal ion and the adsorbent and  $K$ , related to bond strength. The linearized Langmuir, Freundlich isotherms of lead are shown in Figs. 5 and 6. The essential feature of the Langmuir isotherm can be expressed in terms of dimensionless separation parameter  $R_L$ , which is indicative of the isotherm shape that predicts whether an adsorption system is favorable or unfavorable.  $R_L$  is defined as [17]:

$$R_L = 1/(1 + bC_0) \quad (7)$$

where  $b$  is the Langmuir constant. The parameter  $R_L$  indicates the shape of the isotherm as follows:

- $R_L$  value type of isotherm
- $R_L > 1$  unfavorable
- $R_L = 1$  linear
- $0 < R_L < 1$  favorable
- $R_L = 0$  irreversible

According to McKay et al. (1982) [18],  $R_L$  values between 0 and 1 indicate favorable adsorption. The  $R_L$  values were found to be between 0.0017 and 0.0172 (Table 3) for concentrations of 10, 30, 50, 80, and 100 mg/l Pb(II).

The linearized Langmuir and Freundlich isotherms of lead are shown in Figs. 5 and 6. The estimated model parameters with correlation coefficient ( $R^2$ ) for the different models are shown in Table 4. From the

Table 2

Kinetic parameters for the adsorption of  $Pb^{2+}$  ions onto activated carbon by Apricot stone at various initial concentrations

Metals	$C_0$ (mg/l)	First-order kinetic model		Second-order kinetic model	
		$K_{pf}$ ( $l \text{ min}^{-1}$ )	$R^2$	$K_{sf}$ ( $g \text{ mg}^{-1} \text{ min}^{-1}$ )	$R^2$
$Pb^{2+}$	10	0.073	0.623	0.246	0.999
	30	0.046	0.532	0.052	0.999
	50	0.038	0.874	0.025	0.999
	80	0.032	0.912	0.028	0.999
	100	0.026	0.745	0.588	0.999

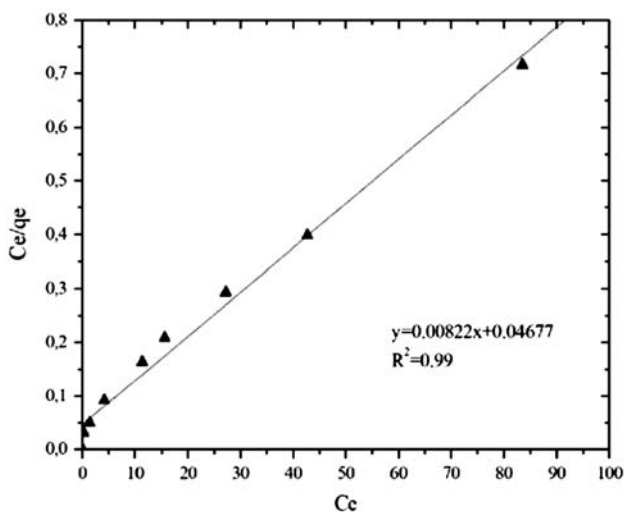


Fig. 5. Langmuir plots for the adsorption of lead (II) ions onto activated carbon from Apricot;  $T = 20 \pm 2^\circ\text{C}$ ,  $S/L = 1 \text{ g/L}$ ,  $\text{pH} = 6.0$ .

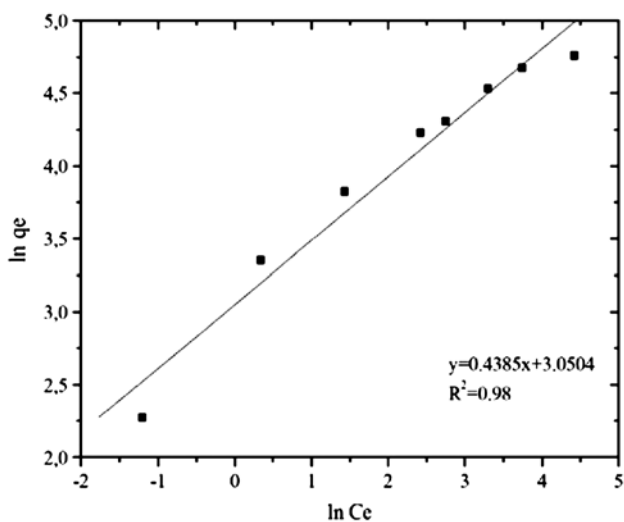


Fig. 6. Freundlich plots for the adsorption of lead (II) ions onto activated carbon from Apricot;  $T = 20 \pm 2^\circ\text{C}$ ,  $S/L = 1 \text{ g/L}$ ,  $\text{pH} = 6.0$ .

values of  $R^2$  summarized in Table 4 and it may be concluded that both equations fit reasonably well the experimental data, although the equation Langmuir provides a better fit than the Freundlich one.

The calculated adsorption capacity for lead was  $21.38 \text{ mg g}^{-1}$ . The Freundlich model is characterized by  $1/n$  the heterogeneity factor; hence, it is applicable to sorption on heterogeneous surfaces, that is, surface with non-energetically equivalent sites. The values of  $1/n$  between  $0.1 < 1/n < 1.0$  represent good adsorption of metal onto the sorbent [19].

Table 3  
Different values of  $R_L$  at different concentrations of lead

Metals	Concentrations (mg/l)	$R_L$
$\text{Pb}^{2+}$	10	0.0172
	30	0.0058
	50	0.0035
	80	0.0021
	100	0.0017

Table 4  
Freundlich and Langmuir constants

Metals	Freundlich constants			Langmuir constants		
	$K$ (l/g)	$1/n$	$R^2$	$q_{\text{max}}$ ( $\text{mg g}^{-1}$ )	$b$ (l/mg)	$R^2$
$\text{Pb}^{2+}$	21.123	0.4385	0.984	21.123	5.69	0.999

Values of the adsorption capacity of other adsorbents from the literature are given in Table 5 for comparison. The value for lead sorption observed in this work is in good agreement with values found by other researchers. Differences of metal uptake are due to the properties of each adsorbent such as structure, functional groups, and surface area.

### 3.7. Thermodynamic parameters

In environmental engineering practice, both energy and entropy factors must be considered in order to determine what processes will occur spontaneously. Gibb's free energy change,  $\Delta G^\circ$ , is the fundamental criterion of spontaneity. Reactions occur spontaneously at a given temperature if  $\Delta G^\circ$  is a negative value. The thermodynamic parameters of  $\Delta G^\circ$ , enthalpy change,  $\Delta H^\circ$ , and entropy change,  $\Delta S^\circ$  for the adsorption processes are calculated using the following equations [31]:

$$\Delta G^\circ = -RT \ln K \quad (8)$$

and

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (9)$$

Where  $R$  is universal gas constant ( $8.314 \text{ J/molK}$ ) and  $T$  is the absolute temperature in  $K$ . A plot of  $\Delta G^\circ$  obtained using  $K (=b)$  of Langmuir vs. temperature,  $T$ , was found to be linear.  $\Delta H^\circ$  and  $\Delta S^\circ$  values were obtained from the slope and intercept of this plot, respectively.  $\Delta G^\circ$ ,  $\Delta H^\circ$ , and  $\Delta S^\circ$  were obtained from the Eqs. (8) and (9) and are given in Table 6.

Table 5  
Adsorption capacities (mg/g) of lead (II) by various adsorbents

	Lead(II)	Reference
Red mud	88.2	Gupta et al. [20]
Coir	18.9	Conrad and Hansen [21]
Crushed concrete fines	37	Coleman et al. [22]
Fly ash	15.08	Cho et al. [23]
Peat	95.2	Ho and McKay [24]
Oryza sativa L. husk	8.6	Zulkali et al. [25]
Turkish low rank coal	13.58	Arpa et al. [26]
Bituminous coal	8.89	Rawat et al. [27]
Sawdust	3.19	Yu et al. [28]
Streptomyces noursei	1.6	Yan et al. [29]
Rhizopus arrhizus	13.5	Fourest et al. [30]
Apricot stone activated carbon	21.38	Present study

Table 6  
Values of thermodynamic parameters for the adsorption of Pb (II) on ASC

T(K)	$-\Delta G^\circ$ (kJ mol <sup>-1</sup> )	$\Delta H^\circ$ (kJ mol <sup>-1</sup> )	$\Delta S^\circ$ (J mol <sup>-1</sup> K <sup>-1</sup> )
293	13.978	17.324	107
313	13.721		
333	12.986		

Negative value of  $\Delta G^\circ$  indicates the feasibility of the process and spontaneous nature of the adsorption with a high preference of Pb (II) by. Positive value of  $\Delta H^\circ$  indicates the endothermic nature of the process, while positive value of  $\Delta S^\circ$  reflects the increased randomness at the solid/solution interface with some structural changes in the adsorbate and the adsorbent and an affinity of the adsorbents toward Pb(II) ions. The positive  $\Delta S^\circ$  value also corresponds to an increase in the degree of freedom of the adsorbed species [31].

#### 4. Conclusion

The present study shows that activated carbon prepared from chemically treated Apricot stone is an effective adsorbent for the removal of lead ions from aqueous solutions. The removal efficiency was controlled by solution pH, adsorbent concentration and agitation times, initial ion concentration, and particle size. In the study kinetics of sorption, the pseudo-second-order model provides better correla-

tion of the sorption data than the pseudo-first-order model, this suggests that the rate-limiting step may be chemical sorption rather than diffusion. Adsorption data fitted well with the Langmuir and Freundlich models. However, Langmuir isotherm displayed a better fitting model than Freundlich isotherm because of the higher correlation coefficient that the former exhibited. The maximal adsorption capacity ( $q_{max}$ ) of lead on Apricot stone was calculated as 21.38 mg g<sup>-1</sup>. The adsorption process was found to be endothermic, spontaneous and can be explained with the pseudo-second-order type kinetic model. The results would be useful for the design of wastewater treatment plants for the removal of lead.

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