

Studies on the removal of Pb(II) from wastewater by activated carbon developed from *Tamarind wood* activated with sulphuric acid

C.K. Singh^a, J.N. Sahu^a, K.K. Mahalik^a, C.R. Mohanty^b,
B. Raj Mohan^c, B.C. Meikap^{a,*}

^a Department of Chemical Engineering, Indian Institute of Technology (IIT), Kharagpur,
Kharagpur Technology, 721302, West Bengal, India

^b Orissa State Pollution Control Board, Bhubaneswar, Orissa, India

^c Department of Chemical Engineering, National Institute of Technology, Surathkal, India

Received 10 May 2007; received in revised form 4 August 2007; accepted 16 August 2007

Available online 22 August 2007

Abstract

The low-cost activated carbon were prepared from Tamarind wood material by chemical activation with sulphuric acid for the adsorption of Pb(II) from dilute aqueous solution. The activated carbon developed shows substantial capacity to adsorb Pb(II) from dilute aqueous solutions. The parameters studied include physical and chemical properties of adsorbent, pH, adsorbent dose, contact time and initial concentrations. The kinetic data were best fitted to the Lagergren pseudo-first-order and pseudo-second order models. The isotherm equilibrium data were well fitted by the Langmuir and Freundlich models. The maximum removal of lead(II) was obtained 97.95% (experimental) and 134.22 mg/g (from Langmuir isotherm model) at initial concentration 40 mg/l, adsorbent dose 3 g/l and pH 6.5. This high uptake showed Tamarind wood activated carbon as among the best adsorbents for Pb(II).

© 2007 Elsevier B.V. All rights reserved.

Keywords: Activated carbon; Chemical activation; Lead(II); Wastewater treatment; Adsorption

1. Introduction

The removal Pb(II) from industrial effluents is a major problem due to the difficulty in treating such wastewaters by conventional treatment methods. The presence of lead in wastewater is dangerous to aquatic flora and fauna even in relatively low concentration and stringent environmental regulations attracts the attention of chemists and environmental engineers for its control. The major sources containing lead are the wastewaters from process industries engaged in lead–acid batteries, paint, oil, metal, phosphate fertilizer, electronic, wood production and also combustion of fossil fuel, forest fires, mining activity, automobile emissions, sewage wastewater, sea spray, etc. Apart from this, lead is also used in storage batteries, insecticides, plastic water

pipes, food, beverages, ointments and medicinal concoctions for flavoring and sweetening.

Lead poisoning causes damage to liver, kidney and reduction in hemoglobin formation, mental retardation, infertility and abnormalities in pregnant women. Due to hazardous nature of lead(II), it directly or indirectly may cause anemia, headache, chills, diarrhea and poisoning leading to the dysfunction of kidneys, reproductive system, liver, brain and central nervous system also.

Various methods have been proposed for the treatment of wastewaters containing lead wastes such as chemical precipitation [1,2], electrochemical reduction [3], ion exchange [4], biosorption [5] and adsorption [6]. The choice of treatment depends on effluent characteristics such as concentration of lead, pH, temperature, flow volume, biological oxygen demand, and the economics involved and the social factor like standard set by government agencies. However, adsorption onto the surface of activated carbons is the most widely used method [7,8]. Activated carbon is a black solid substance resembling granular or powder charcoal and is carbonaceous material that has

* Corresponding author. Tel.: +91 3222 283958(O)/2283959(R);
fax: +91 3222 282250.

E-mail addresses: bcmeikap@che.iitkgp.ernet.in,
bcmeikap@iitkgp.ac.in (B.C. Meikap).

Nomenclature

b	Langmuir constant (l/g)
C_e	equilibrium lead concentration (mg/l)
C_0	initial lead concentration (mg/l)
k	measure of adsorbent capacity (l/g)
K_{ad}	equilibrium rate constant of pseudo-first-order adsorption (min^{-1})
K_L	Langmuir constant (mg/g)
K_2	equilibrium rate constant of pseudo-second order adsorption (min^{-1})
$1/n$	sorption intensity, dimensionless
q_e	amount of lead adsorbed at equilibrium (mg/g)
t	time (min)
T	temperature ($^{\circ}\text{C}$)
V	volume of the solution (l)
W	weight of adsorbent (g)

highly developed porosity, internal surface area and relatively high mechanical strength. They are widely used as adsorbents in wastewater and gas treatments as well as in catalysis. Despite its prolific use in industries, activated carbon remains an expensive material. In recent years, research interest in the production of low-cost alternatives to activated carbon has grown.

In practice, coal and agricultural byproducts of lignocelluloses materials are two main sources for the production of commercial activated carbons. Agricultural wastes have emerged as a better choice. Though raw agricultural wastes can be used as adsorbents without further treatment, activation could enhance their adsorption capacity. The production of activated carbons from agricultural wastes converts unwanted, surplus agricultural waste, of which billions of kilograms are produced annually, to useful valuable adsorbents, such as nutshells [9], oil palm waste [10], rice husk, peanuts [11], sawdust [12], and canes from some easy-growing wood species [13]. 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. The effect of different chemical reagents on the production and quality of activated carbon was studied extensively by different researchers [9,11,14].

There are two important advantages of chemical activation in comparison to 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, sulphuric acid in particular is the widely used chemical agent in the preparation of activated carbon. Knowledge of different variables during the activation process is very important in developing the porosity of carbon sought for a given application. Chemical activation by H_2SO_4 improves the pore development in the carbon structure,

and because of the effect of chemicals, the yields of carbon are usually high [15]. In this work it has been reported that the results obtained on the preparation of activated carbon from Tamarind wood with sulphuric acid activation and their ability to remove lead from wastewater. The influence of several operating parameters, such as contact time, initial concentration, particle size and adsorbent dose, etc. was investigated in batch mode. The kinetic data were fitted to different models and the isotherm equilibrium data were fitted to Langmuir and Freundlich.

2. Experimental technique

2.1. Preparation of activated carbon

The Tamarind wood was collected from local Indian Institute of Technology campus of Kharagpur, West Bengal, India, and washed with deionized water four to five times for removing dirt and dust particles. The washed wood was cut into $50.8 \text{ mm} \times 76.2 \text{ mm}$ pieces. The woods were sun dried for 20 days. The dry woods were then soaked in concentrated sulphuric acid (H_2SO_4) at normal temperature and pressure. Then this mixture of H_2SO_4 and untreated wood was heated at 200°C . Then this 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 activated carbon was deacidified by use of water and ammonical solution in later stages. Deacidification was done in following manner. After 25–30 times washing carbon was treated with liquid aqueous ammonia solution in such a fashion that it does not affect the surface properties. Then carbon was dried to 60°C . After this carbon was crushed in a small ball mill with 50 small balls for 1 h. The powder from ball mill is collected and dried to remove the moisture. Then this powder carbon was kept in airtight packet for the experimental use.

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 200 ml solution of 10, 20, and 40 mg/l of initial concentration. The aqueous lead solutions were prepared by dissolving known amount of corresponding nitrate salt ($\text{Pb}(\text{NO}_3)_2$) in 1 l ordinary water. Analytical-grade reagents were used in all cases. The stock solution was diluted as required to obtain standard solutions of concentrations ranging between 10 and 100 mg/l. The experiments were performed in a thermal shaker at controlled temperature (25°C) for a period of 24 h at 120 rpm using 250 ml Erlenmeyer flasks containing 40 ml of different lead concentrations at room temperature. Samples were taken out at regular interval and the residual concentration in the solution was analyzed using atomic-absorption spectroscopy (AAS) after filtering the adsorbent with Whatman filter paper to make it carbon free. The batch process was used so that there is no need for volume correction. The experiments were carried out at different initial concentration and adsorbent dosages. Atomic-absorption

spectrophotometry utilizes the phenomenon that atoms absorb radiation of particular wavelength. By atomic-absorption spectrophotometer, the metals in water sample can be analyzed. It consists of four basic structural elements; a light source (hollow cathode lamp), an atomizer section for atomizing the sample (burner for flame, graphite furnace for electrothermal atomization), a monochromator for selecting the analysis wavelength of the target element, and a detector for converting the light into an electrical signal. It detect concentration of Pb(II) in ppm level in the solution and volume of sample required is only 1 ml for one analysis.

Batch adsorption experiments were performed by contacting 0.4 g of the selected carbon samples with 200 ml of the aqueous solution of different initial concentration (10, 20, and 40 mg/l) at natural solution pH. Continuous mixing was provided during the experimental period with a constant agitation speed of 120 rpm for better mass transfer with high interfacial area of contact. The remaining concentration of Pb(II) in each sample after adsorption at different time intervals, was determined by atomic-absorption spectroscopy. The lead concentration retained in the adsorbent phase was calculated according to

$$q_e = \frac{(C_0 - C_e)V}{W} \quad (1)$$

where C_0 and C_e are the initial and equilibrium concentrations (mg/l) of lead solution respectively; V is the volume (l); and W is the weight (g) of the adsorbent. Two replicates per sample were done and the average results are presented.

The effect of adsorbent dosages (1–5 g/l) on the equilibrium adsorption of Pb(II) on the selected carbon was investigated by employing different initial concentrations (10, 20, and 40 mg/l) at pH 6.5. For these experiments, the flasks were shaken, keeping the temperature (25 °C) constant and agitation speed (120 rpm) for the minimum contact time required to attain equilibrium, as determined from the kinetic measurements detailed above.

The BET surface areas of the Tamarind wood activated carbon were measured in BET-Flowsorb-2300 analyzer on the basic principle of monolayer adsorption. Nitrogen and helium mixture nearly 30–35% was used to form the monolayer. In this method first sample quantity is optimized after several trial runs so that surface area falls within 0.5–2.5 m². The sample is dried in an air oven at 105 °C and free from any gases or vapors for which 200–250 °C for 15 min is adequate. Surface area in Flowsorb-2300 is displayed in terms of quantity of sample contained in sample tube holder and the displayed number is converted to specific surface area by dividing the weight of the sample.

3. Result and discussion

3.1. Lead adsorption

3.1.1. Contact time study

The effect of contact time for three different concentrations of Pb(II) (10, 20, and 40 mg/l) with activated carbon dose (2 g/l) at natural pH of solution (pH 6.5) is shown in Fig. 1. The lead percent adsorption increases with time until the equilibrium is attained between the amounts of lead adsorbed on the activated

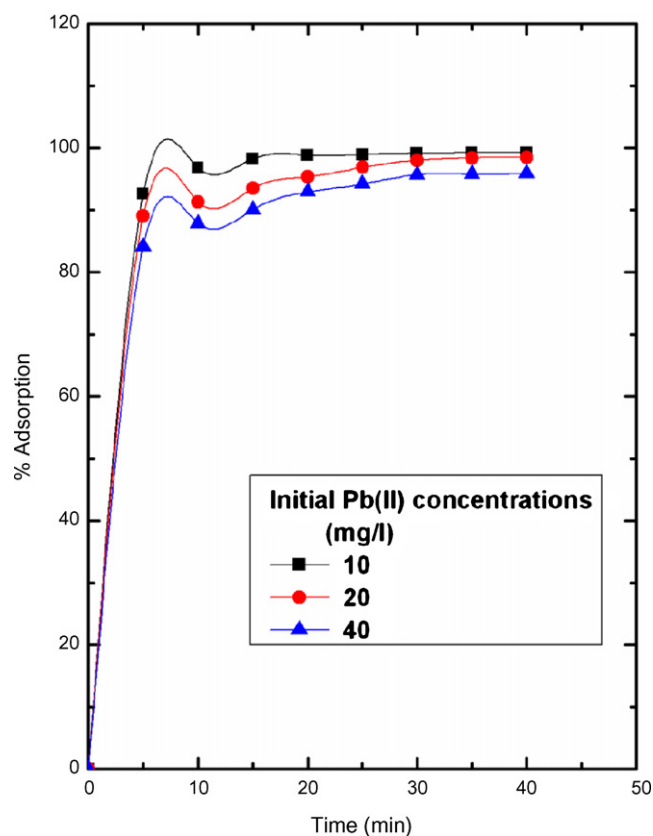


Fig. 1. Effect of contact time on lead removal.

carbon and the remaining in solution. The figure shows that the adsorption of Pb(II) increases with time from 0 to 15 min and more and then becomes almost constant up to the end of experiment. It can be concluded that the rate of lead binding with activated carbon is more at initial stages, which gradually decreases and becomes almost constant after an optimum period of 40 min.

A comparison at different activated carbon dose (1.0 and 2.0 g/l) with a constant pH 6.5 has been made and presented in Fig. 2 for the adsorption efficiency. It has been found that under identical conditions higher doses of activated carbon enhances the removal efficiency of lead. This is quite obvious that at higher dose activated carbon adsorb more quantity of Pb(II), which resulted in increased efficiency.

3.1.2. Initial concentration

The effect of lead concentration in the solution on the adsorption has been shown in Fig. 3. It can be seen from the figure that with increased Pb(II), there was decrease in percentage of adsorption of lead.

3.1.3. Physical properties

The smaller the particle sizes of a porous carbon are, the greater the rate of diffusion and adsorption. Intra-particle diffusion is reduced as the particle size reduces, because of the shorter mass transfer zone, causing a faster rate of adsorption. Since we have prepared our carbon in a powdered form so it has a great efficiency of removal. Particle size distribution analysis

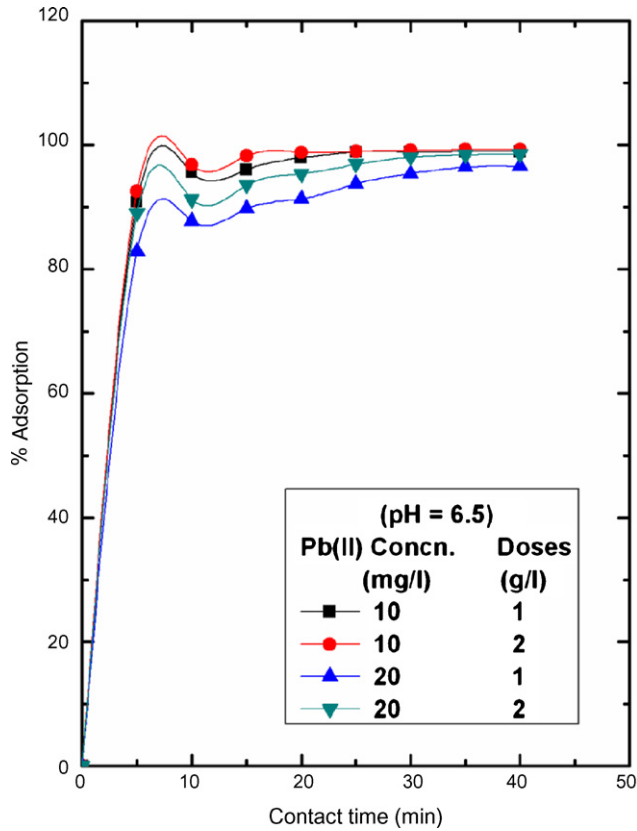


Fig. 2. Comparison of effect of contact time on percentage removal of Pb(II) at different activated carbon doses and initial concentrations at constant pH.

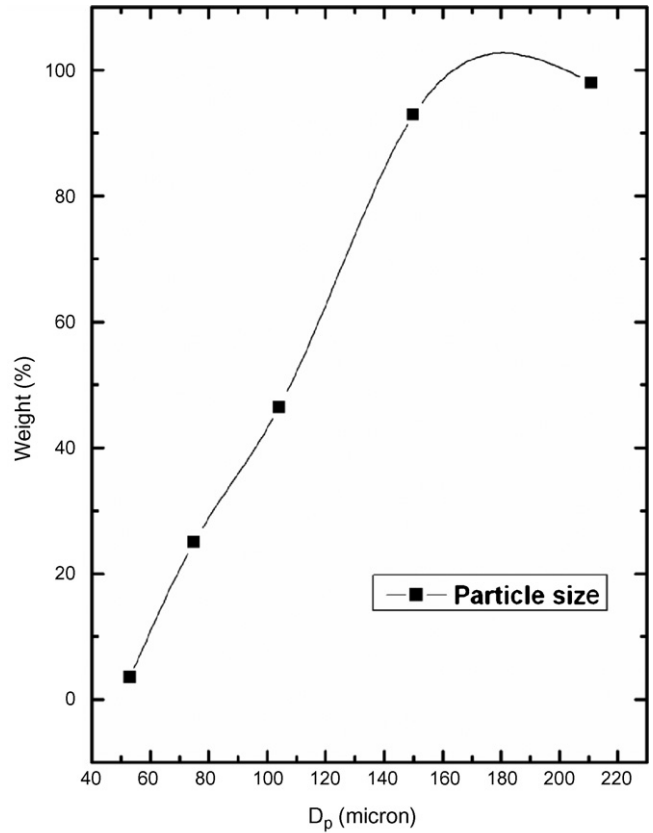


Fig. 4. Particle size analysis by sieves for Tamarind wood activated adsorbent and cumulative weight percentage.

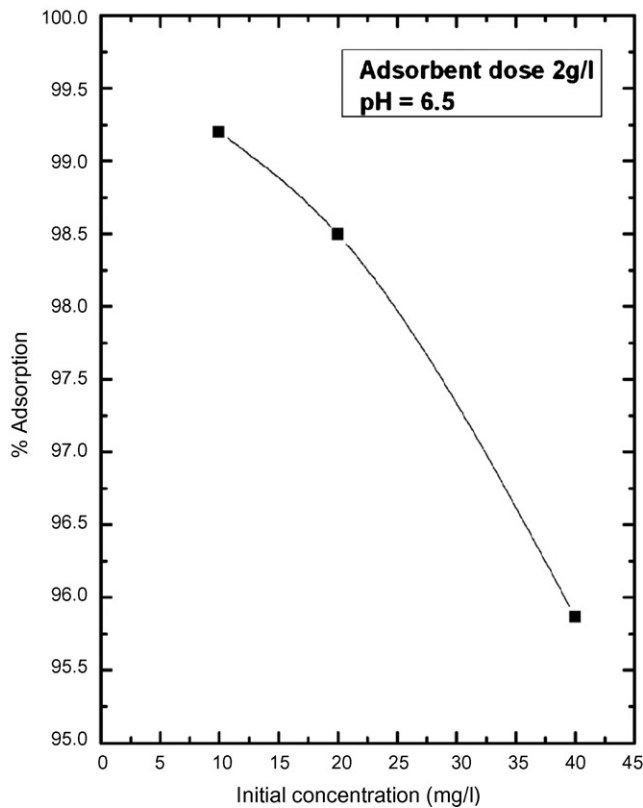


Fig. 3. Effect of initial concentration of lead at adsorbent dose 2 g/l.

was done manually. Initial sample of weight 145 g was taken for analysis. This sample was passed through different sieves and amount of fine and coarse were measured. For the mesh size 72, 100, 150, 200 and 300 data were obtained are shown in Fig. 4.

Density of the activated carbon plays an important role for its adsorption capacity. If two carbons differing in bulk density are used at the same weight per liter, the carbon having higher bulk density will be able to remove more efficiently. Average bulk density can be calculated by water displacement method. In this method, volume of water displaced is observed by a particular amount of carbon. The data were obtained for this experiment is shown in Fig. 5 we can see that average bulk density is 1.5625 g/ml. The BET surface area of activated carbon prepared from tamarind was measured and it was found 612 m²/g and total pore volume of 0.508 cm³/g. This shows the prepared activated is reasonably good for adsorption.

3.1.4. Chemical properties

Activated carbon pH may influence the removal efficiency. Distinctly acidic activated carbon may react with the material to be removed and may hamper the surface properties of the activated carbon. For our experiment the pH of carbon was 6.5.

Ash content of the carbon is the residue that remains when the carbonaceous portion is burned off. The ash consists mainly of minerals such as silica, aluminum, iron, magnesium and calcium. Ash in activated carbon is not required and considered to be an impurity.

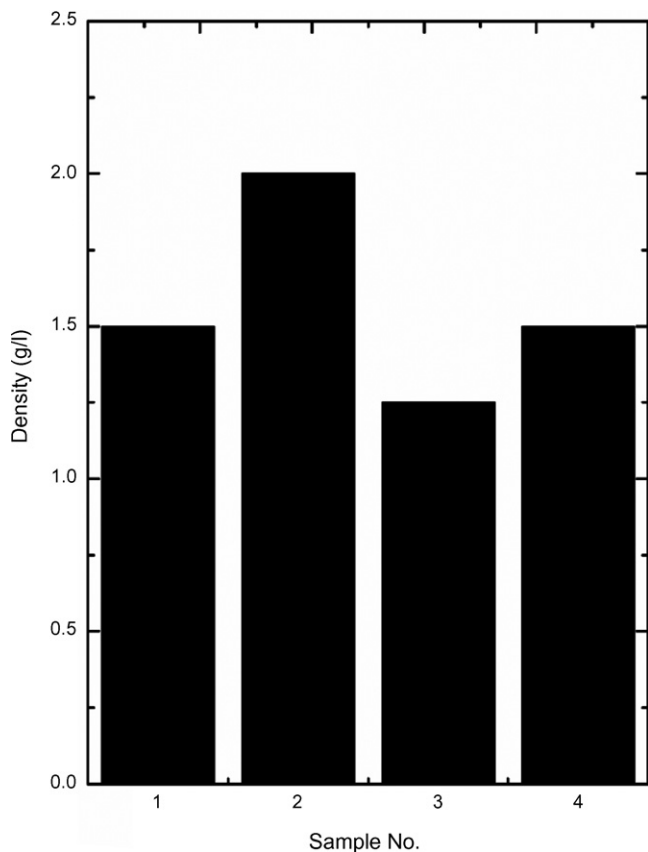


Fig. 5. Density of powder Tamarind wood activated carbon of different sample.

3.1.5. Kinetic models

To investigate the mechanism of lead adsorption, two kinetic models were considered as follows.

Lagergren proposed a pseudo-first-order kinetic model. The integral form of the model is

$$\log(q_e - q) = \log q_e - \frac{K_{ad}}{2.303} t. \quad (2)$$

where q is the amount of lead sorbed (mg/g) at time t (min), q_e is the amount of lead sorbed at equilibrium (mg/g), and K_{ad} is the equilibrium rate constant of pseudo-first-order adsorption (min^{-1}). This model was successfully applied to describe the kinetics of many adsorption systems.

The adsorption kinetics may also be described by a pseudo-second order reaction. The linearized-integral form of the model is

$$\frac{t}{q} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t. \quad (3)$$

where K_2 is the pseudo-second order rate constant of adsorption.

The applicability of the above two models can be examined by each linear plot of $\log(q_e - q)$ vs. t , and (t/q) vs. t , respectively, and are presented in Figs. 6 and 7. To quantify the applicability of each model, the correlation coefficient, R^2 , was calculated from these plots. The linearity of these plots indicates the applicability of the two models. However, the correlation coefficients, R^2 , showed that the pseudo-second order model, an indication of a chemisorptions mechanism, fits better the experimental data

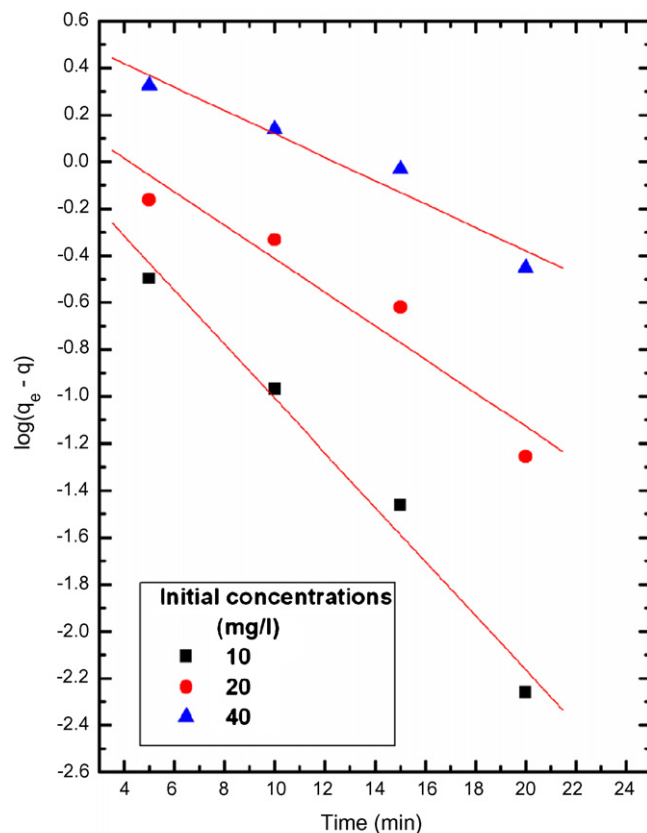


Fig. 6. Kinetics of lead removal according to the Lagergren model at initial lead concentration of 10, 20, and 40 mg/l.

($R^2 > 0.999$) than the pseudo-first-order model (R^2 is in the range of 0.948–0.982).

3.2. Adsorbent dose study

The effect of adsorbent dosage on the percentage removal of lead has been shown in Figs. 8 and 9. It can be seen from the figure that initially the percentage removal increases very sharply with the increase in adsorbent dosage (1–5 g/l) but beyond a certain value, the percentage removal reaches almost a constant value. This trend is expected because as the adsorbent dose increases the number adsorbent particles increases and thus more Pb(II) is attached to their surfaces. It is also seen that the percentage adsorption increases from 87.55 to 97.06. A maximum removal of 97.95% was observed at adsorbent dosage of 3 g/l at pH 6.5 for an initial lead concentration of 40 mg/l. Therefore, the use of 3 g/l adsorbent dose is justified for economical purposes.

3.2.1. Adsorption isotherms

Several models have been used in the literature to describe the experimental data of adsorption isotherms. The Freundlich and Langmuir models are the most frequently employed models. In the present work both models were used.

The lead sorption isotherm followed the linearized Freundlich model as shown in Fig. 10. The relation between the metal uptake capacity ' q_e ' (mg/g) of adsorbent and the residual

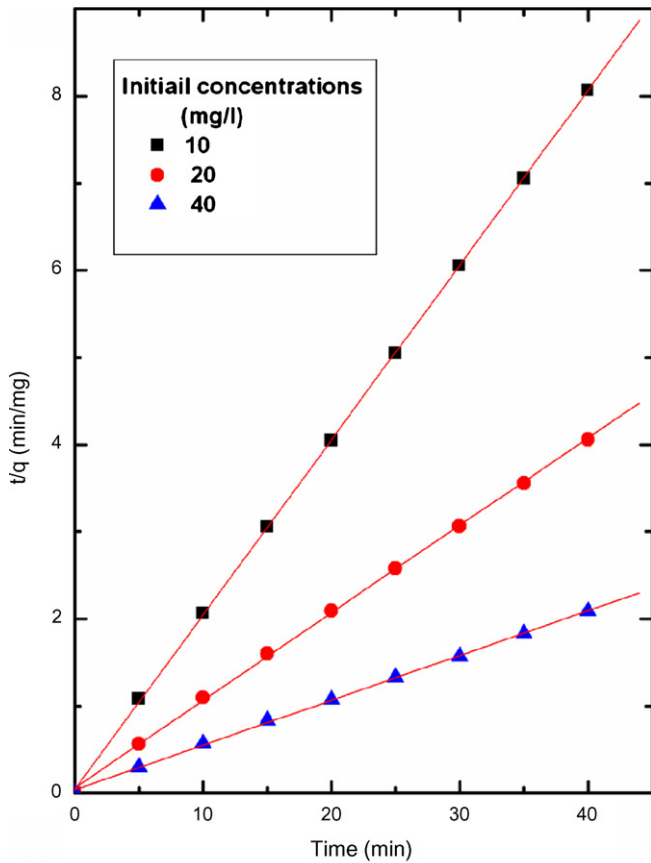


Fig. 7. Kinetics of lead removal according to pseudo-second-order model at initial lead concentration of 10, 20, and 40 mg/l.

metal ion concentration ‘ C_e ’ (mg/l) at equilibrium is given by

$$\ln q_e = \ln k + \frac{1}{n} \ln C_e. \tag{4}$$

where the intercept $\ln k$ is a measure of adsorbent capacity, and the slope $1/n$ is the sorption intensity. The isotherm data fit the Freundlich model well ($R^2 = 0.9756$). The values of the constants k and $1/n$ were calculated to be 14.378 and 0.9243. Since the value of $1/n$ is less than 1, it indicates a favorable adsorption.

The Langmuir equation relates solid phase adsorbate concentration (q_e), the uptake, to the equilibrium liquid concentration (C_e) as follows:

$$q_e = \left(\frac{K_L b C_e}{1 + b C_e} \right). \tag{5}$$

where K_L and b are the Langmuir constants, representing the maximum adsorption capacity for the solid phase loading and the energy constant related to the heat of adsorption respectively. It can be seen from Fig. 11 that the isotherm data fits the Langmuir equation well ($R^2 = 0.98081$). The values of K_L and b were determined from the figure and were found to be 134.22 mg/g and 0.01202 l/mg, respectively. The outcome values of parameters k , n , K_L , b , R^2 for all the experiments with pH of solution equal to 6.5 for maximum removal of Pb(II) are presented in Table 1.

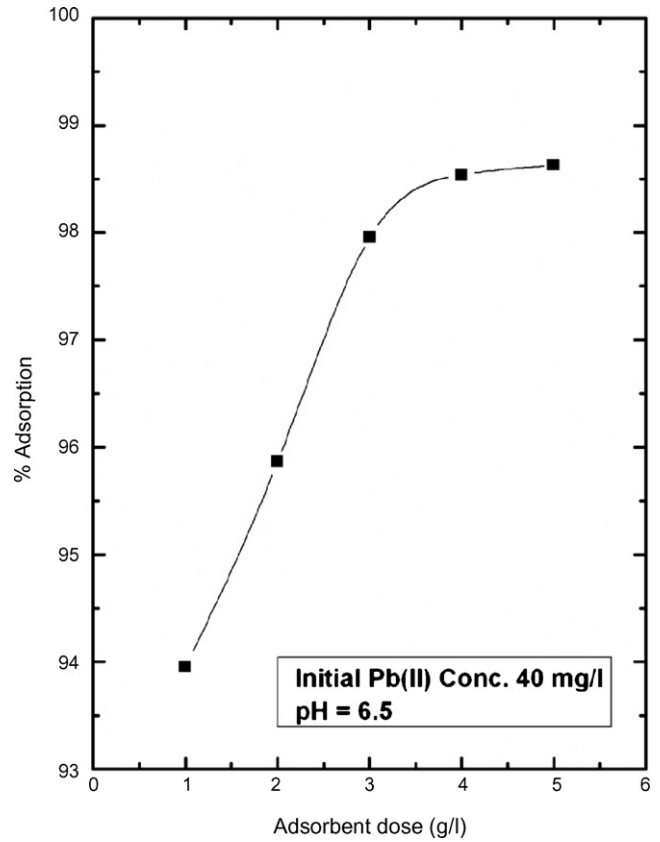


Fig. 8. Effect of adsorbent dose on Pb(II) removal at solution pH 6.5.

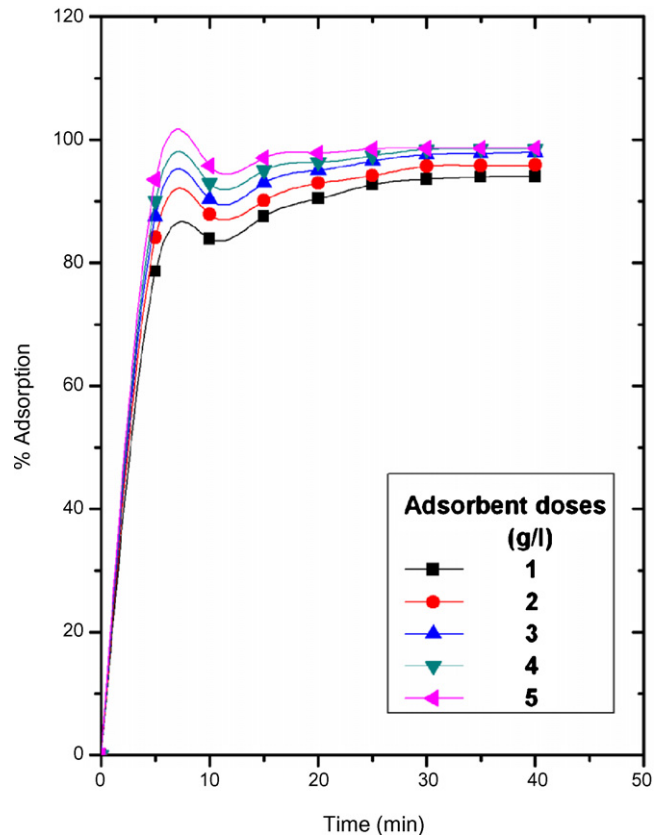


Fig. 9. Effect of adsorbent doses with time at 40 mg/l initial concentration of Pb(II).

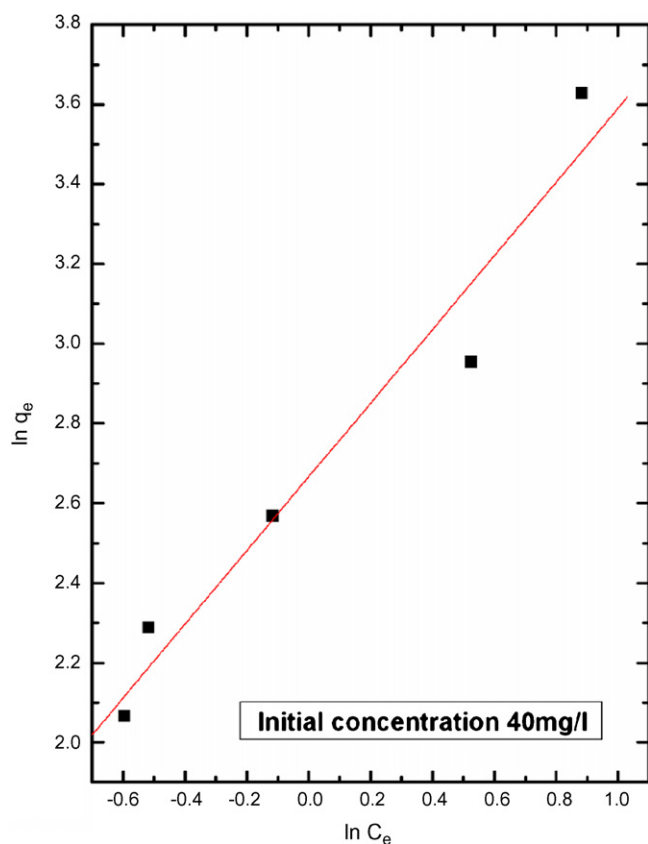


Fig. 10. Freundlich adsorption isotherm.

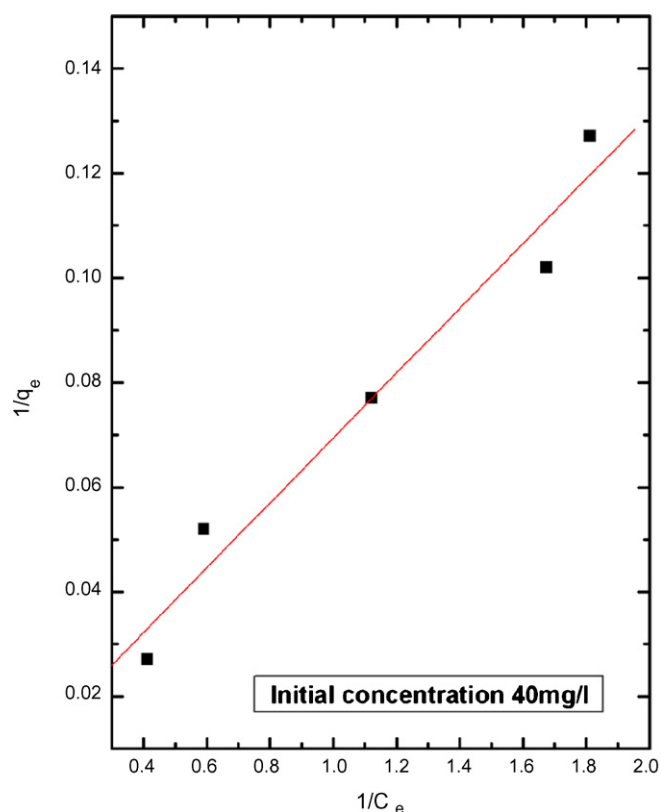


Fig. 11. Langmuir adsorption isotherm.

Table 1

Adsorption isotherms parameter at pH 6.5 for lead(II) removal

K_L (mg/g)	b (l/g)	R^2	k (l/g)	$1/n$
134.22	0.012	0.9808	Langmuir	14.378
		0.9756	Freundlich	

Table 2

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

Adsorbent	Lead(II)	Reference
Red mud	88.2	Gupta et al. [16]
Coir	18.9	Conrad and Hansen [17]
Crushed concrete fines	37	Coleman et al. [18]
Fly ash	15.08	Cho et al. [19]
Peat	95.2	Ho and Mckay [20]
<i>Oryza sativa</i> L. husk	8.6	Zulkali et al. [21]
Turkish low rank coal	13.58	Arpa et al. [22]
Bituminous coal	8.89	Rawat et al. [23]
Coconut shell activated carbon	21.88	Goel et al. [24]
Palm shell activated carbon	95.2	Issabayeva et al. [25]
Sawdust	3.19	Yu et al. [26]
Bone powder	55.3	Abdel-Halim et al. [27]
Nile rose plant power (water hyacinth)	27.4	Abdel-Halim et al. [27]
Qafuor tree commercial carbon	27.3	Abdel-Halim et al. [27]
Tamarind wood activated carbon	134.22	Present study

A comparison of the maximum capacity of Tamarind wood activated carbon with those of some other adsorbents reported in literature is given in Table 2. 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.

4. Conclusion

Removal of lead from wastewater is possible by using abundantly available low-cost adsorbents. The present investigation shows that Tamarind wood activated carbon is an effective adsorbent for the removal and recovery of lead from wastewater. A detailed experimental investigations have been carried out for initial concentrations of Pb(II) of 10, 20, and 40 mg/l. The adsorption capacity of activated carbon is greater than that of a commercial carbon link carbon. From the kinetics studies it is observed that adsorption of Pb(II) is very rapid in the initial stage and decreases while approaching equilibrium. The kinetics of Pb(II) adsorption followed the pseudo-second order rate expressions and demonstrated that intra-particle diffusion plays a significant role in the adsorption mechanism. The Freundlich and Langmuir isotherm models are found to be linear, indicating the applicability of classical adsorption isotherm to this adsorbate–adsorbent system. Experimental results are in good agreement with Langmuir adsorption isotherm model, and have shown a better fitting to the experimental data. The BET surface area was found $612 \text{ m}^2/\text{g}$ and total pore volume of $0.508 \text{ cm}^3/\text{g}$. The data reported may be very useful for designing an economically cheap treatment process using batched or stirred-tank

flow reactors for the removal of Pb(II) from Pb(II) containing wastewater from chemical and allied process industries.

References

- [1] M. Swiderska-Broz, Contribution of humic acids to the removal of some heavy metals by chemical precipitation, *Environ. Prot. Eng.* 13 (3–4) (1987) 29–38.
- [2] M.M. Husein, J.H. Vera, M.E. Weber, Removal of lead from aqueous solutions with sodium caprate, *Sep. Sci. Technol.* 33 (12) (1998) 1889–1904.
- [3] S.W. Lin, R.M.F. Navarro, An innovative method for removing Hg^{2+} and Pb^{2+} in ppm concentrations from aqueous media, *Chemosphere* 39 (11) (1999) 1809–1817.
- [4] D. Petruzzelli, M. Pagano, G. Tiravanti, R. Passino, Lead removal and recovery from battery wastewaters by natural zeolite clinoptilolite, *Solvent Extr. Ion Exch.* 17 (3) (1999) 677–694.
- [5] A. Saeed, M. Iqbal, M.W. Akhtar, Removal and recovery of lead(II) from single and multimetal (Cd, Cu, Ni, Zn) solutions by crop milling waste (black gram husk), *J. Hazard. Mater.* 117 (1) (2005) 65–73.
- [6] S. Doyurum, A. Celik, Pb(II) and Cd(II) removal from aqueous solutions by olive cake, *J. Hazard. Mater.* 138 (1) (2006) 22–28.
- [7] N. Kannan, J. Balamurugan, Removal of lead ions by adsorption onto coconut shell and dates nut carbons—a comparative study, *Indian J. Environ. Prot.* 25 (9) (2005) 816–823.
- [8] N. Kannan, S.M. Devi, Studies on removal of copper(II) and lead(II) ions by adsorption on commercial activated carbon, *Indian J. Environ. Prot.* 25 (1) (2005) 28–37.
- [9] J.W. Kim, M.H. Sohn, Production of granular activated carbon from waste walnut shell and its adsorption characteristics for Cu^{+2} ion, *J. Hazard. Mater.* 85 (2001) 301–315.
- [10] C.A. Lua, J. Guo, Preparation and characterization of chars from oil palm waste, *Carbon* 36 (11) (1998) 1663–1670.
- [11] K. Perisamy, C. Namasivam, Removal of lead by adsorption on to peanut hull carbon from water and lead plating industry wastewater, *Chemosphere* 32 (1996) 769–789.
- [12] F. Marquez-Montesinos, T. Cordero, J. Rodriguez-Mirasol, J.J. Rodriguez, Powdered activated carbons from *Pinus caribaea* sawdust, *Sep. Sci. Technol.* 36 (14) (2001) 3191–3206.
- [13] M.C. Basso, E.G. Cerrella, A.L. Cukierman, Activated carbons developed from a rapidly renewable biosource for removal of cadmium(II) and nickel(II) ions from dilute aqueous solutions, *Ind. Eng. Chem. Res.* 41 (2) (2002) 180–189.
- [14] G.A. Macías, M.A. Díaz, V. Gómez, M. González, Preparation and characterization of activated carbons made up from different woods by chemical activation with H_3PO_4 , *Smart Mater. Struct.* 12 (2003) 24–28.
- [15] J. Guo, W.S. Xu, Y.L. Chen, A.C. Lua, Adsorption of NH_3 onto activated carbon prepared from palm shells impregnated with H_2SO_4 , *J. Colloid Interface Sci.* 281 (2) (2005) 285–290.
- [16] V.K. Gupta, M. Gupta, S. Sharma, Process development for the removal of lead and chromium from aqueous solutions using red mud—an aluminium industry waste, *Water Res.* 35 (5) (2001) 1125–1134.
- [17] K. Conrad, H.C.B. Hansen, Sorption of zinc and lead on coir, *Bioresour. Technol.* 98 (1) (2007) 89–97.
- [18] N.J. Coleman, W.E. Lee, I.J. Slipper, Interactions of aqueous Cu^{2+} , Zn^{2+} and Pb^{2+} ions with crushed concrete fines, *J. Hazard. Mater.* 121 (1–3) (2005) 203–213.
- [19] H. Cho, D. Oh, K. Kim, A study on removal characteristics of heavy metals from aqueous solution by fly ash, *J. Hazard. Mater.* 127 (1–3) (2005) 187–195.
- [20] Y.S. Ho, G. McKay, The sorption of lead(II) ions on peat, *Water Res.* 33 (2) (1999) 578–584.
- [21] M.M.D. Zulkali, A.L. Ahmad, N.H. Norulakmal, *Oryza sativa* L. husk as heavy metal adsorbent: optimization with lead as model solution, *Bioresour. Technol.* 97 (1) (2006) 21–25.
- [22] C. Arpa, E. Basyilmaz, S. Bektas, O. Genc, Y. Yurum, Cation exchange properties of low rank Turkish coals: removal of Hg Cd and Pb from waste water, *Fuel Process. Technol.* 68 (2) (2000) 111–120.
- [23] N.S. Rawat, Ranjana, D. Singh, Characteristic adsorption of aqueous lead(II) on bituminous coal, *Indian J. Environ. Prot.* 13 (3) (1993) 193–197.
- [24] J. Goel, K. Kadirvelu, C. Rajagopal, K.V. Garg, Removal of lead(II) by adsorption using treated granular activated carbon: batch and column studies, *J. Hazard. Mater.* 125 (1–3) (2005) 211–220.
- [25] G. Issabayeva, M.K. Aroua, N.M.N. Sulaiman, Removal of lead from aqueous solutions on palm shell activated carbon, *Bioresour. Technol.* 97 (18) (2006) 2350–2355.
- [26] B. Yu, Y. Zhang, A. Shukla, S.S. Shukla, K.L. Dorris, The removal of heavy metals from aqueous solutions by sawdust adsorption—removal of lead and comparison of its adsorption with copper, *J. Hazard. Mater. B* 84 (2001) 83–94.
- [27] S.H. Abdel-Halim, A.M.A. Shehata, M.F. El-Shahat, Removal of lead ions from industrial waste water by different types of natural materials, *Water Res.* 37 (7) (2003) 1678–1683.