Adsorptive removal of heavy metal ions from industrial effluents using activated carbon derived from waste coconut buttons

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Abstract

Activated carbon (AC) derived from waste coconut buttons (CB) was investigated as a suitable adsorbent for the removal of heavy metal ions such as Pb(II), Hg(II) and Cu(II) from industrial effluents through batch adsorption process. The AC was characterized by elemental analysis, fourier transform infrared spectroscopy, X-ray diffraction, scanning electron microscopy, thermal gravimetric and differential thermal analysis, surface area analyzer and potentiometric titrations. The effects of initial metal concentration, contact time, pH and adsorbent dose on the adsorption of metal ions were studied. The adsorbent revealed a good adsorption potential for Pb(II) and Cu(II) at pH 6.0 and for Hg(II) at pH 7.0. The experimental kinetic data were a better fit with pseudo second-order equation rather than pseudo first-order equation. The Freundlich isotherm model was found to be more suitable to represent the experimental equilibrium isotherm results for the three metals than the Langmuir model. The adsorption capacities of the AC decreased in the order: Pb(II) > Hg(II) > Cu(II).

Key words: coconut buttons; heavy metal ions; adsorption isotherm; wastewater; desorption

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Introduction

The annual global output of coconut stands at 5.5 million tones. The leading producers are Indonesia (27%), Philippines (23%) and India (22%), followed by Sri Lanka, Mexico, Vietnam, New Guinea and Brazil. Based on the 2006–2007 data of the Coconut Development Board, India has 1.94 million hectares (ha) coconut cultivation, yielding 15,840 million nuts. In India, Kerala is the main coconut growing state with an area of 897,800 ha followed by Tamil Nadu (370,600 ha) and Karnataka (385,400 ha). It was observed that a considerable loss in the yield of coconut resulted from the phenomenon of shedding of buttons. The loss varied from 55% to 95%, and depended on factors such as nutritional deficiencies, unfavorable weather conditions, defective pollination and abscission. Hence a significant amount of coconut buttons are discarded as agricultural waste. Efforts were made to convert this cheap and abundant agricultural waste into activated carbon (AC), a value-added adsorbent for the treatment of industrial effluents and also to minimize the disposal of solid wastes.

Heavy metals present in ground water and surface water are a threat to human and local ecosystem. Therefore, tremendous efforts are being made to decrease the con-

centration of heavy metals in the effluent wastewaters to permissible discharge levels set by various pollution control and regulatory authorities in various countries. A wide range of processes such as chemical precipitation, ion exchange, membrane filtration, electrolytic method, coagulation, reverse osmosis and adsorption are being used for the removal of heavy metal ions from industrial effluents. Among these processes, the most promising efficient technique has been identified as adsorption with a suitable adsorbent. Activated carbons are unique and versatile adsorbents because of the availability of extensive surface area, its micro porous structure and high adsorption capacity. But the high cost restricts their use. Hence the production of activated carbons from cheaper materials is a need and has gained significant attention for wastewater treatment in developing countries. Various researchers have developed activated carbons from renewable and cheaper precursors like indigenous agro wastes (Garcia et al., 2003; Namasivayam and Sangeetha, 2006; Baccara et al., 2009; Anirudhan et al., 2009; Shi et al., 2010). The manufacture of activated carbons generally involves two steps: pyrolysis and physical and/or chemical activation of the resulting carbonized substances. The physical activation involves oxidation and gasification of the char at high temperatures. In chemical activation process the carbonization and activation are carried out in a single step by

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the thermal decomposition of the raw material impregnated with certain chemicals such as $ZnCl_2$, NH_4Cl, $BaCl_2$, $MgCl_2$, NaOH, KOH, K_2CO_3, K_2HPO_4 and H_3PO_4. During activation, the textural properties of the materials may also change. The adsorption capacity of materials has been shown to be increased by the activation process of lignocellulosic precursors (Namasivayam and Sangeetha, 2004). Physical activation permits better control over the creation of a desired micro porous structure. Activation at higher temperatures enlarges the diameter of the pores, where by the pore volume increases significantly. Steam activation has shown the best potential for producing activated carbons with high surface area and good pore ratio (Ahmedna et al., 2000).

Previous studies for the preparation of activated carbons from agricultural wastes has been studied by earlier workers (Anirudhan et al., 2009; Shi et al., 2010), did not consider the production of activated carbon from coconut buttons. This study contributes a dual purpose. Firstly, the unwanted surplus agricultural waste can be converted into useful value-added adsorbent; secondly, the use of agricultural byproducts provides a potential adsorbent for the removal of heavy metal ions from wastewater.

In the present work, steam activation method was used to prepare the activated carbon from coconut buttons. Steam activation is a cost-effective method. It gives better activation and enhances the widening of the narrow micro-pore network at higher temperatures (Kumar et al., 2006). The objective of this study was to demonstrate the adsorption performance of the activated carbon, prepared from coconut buttons using steam activation method, for the removal of heavy metals from water and wastewater. The adsorption of Pb(II), Cu(II) and Hg(II) was studied in a batch system with respect to the initial pH, contact time, initial metal ion concentration and adsorbent dose.

1 Materials and methods

1.1 Materials

The precursor used for the preparation of adsorbent was coconut buttons, easily available in the areas where coconut trees are grown. All chemicals were of analytical reagent grade. All metal ions were used in their chloride form and were obtained from Fluka, Switzerland, while H_2SO_4, $NaHCO_3$, NaCl, HCl and NaOH, used were purchased from E. Merck, India. The metal solutions of various concentrations from 10 to 300 mg/L were prepared with distilled water from a stock solution of 1000 mg/L.

1.2 Preparation of adsorbent

A sufficient quantity of coconut buttons were grilled and washed with deionised water to remove sand and other particles. After being dried in the sun, the material was taken in a beaker. About 100 g of dry coconut buttons were suspended in 300 mL concentrated sulphuric acid and the suspension was stirred for 2 hr. The treated coconut buttons were then separated from the acid solution by filtration and the product was then washed with distilled water to remove

residual acid and soluble compounds. It was then dried at 110°C in an oven for two hours.

About 50 g of acid treated coconut buttons were then put in a purpose-made graphite tube and placed at the centre of muffle furnace (Matri, India). Steam, produced from a steam generator, was passed into the graphite tube at a rate of approximately 3 mL/min (liquid water). The sample was then heated at 10°C/min to 400°C and held at that temperature for 1 hr. After cooling the furnace to room temperature, the carbonized material was washed with distilled water until a constant pH was reached. The washed carbon was then dried in an oven at 110°C and cooled. The yield of carbon was found to be 40.0%. Finally the material was ground and sieved to –80+230 mesh size (average diameter of 0.096 mm) and stored in a desiccator for further use.

1.3 Equipments and characterization methods

The specific surface area of activated carbon was measured using N_2 adsorption data at 77 K with a Quantasorb surface area analyzer (model $Q_{s/7}$). The total pore volume was determined from the amount of N_2 adsorbed at a relative pressure of 0.95. The Brunauer-Emmett-Teller (BET) surface area of the sample was calculated using BET isotherm equation by assuming the area of a N_2 molecule to be 0.162 mm². The pore size distribution was estimated by the density functional theory (DFT) method. The t-plot method was applied to calculate the micro pore volume. The micro pore volume was estimated to be the liquid volume of N_2 at a relative pressure of 0.993. Aggregate density ($\rho_{aggregate}$) is the density of the small clusters of matter inside the carbon particle. The density was experimentally measured using a benzene replacement method (Hanzawa et al., 1998). The value was found to be 1.66 g/mL. Carbon sphere density (ρ_{sphere}) is defined as the mass of carbon in one particle divided by the volume of the particle. The value of the density was calculated using the expression:

$$\rho_{sphere} = \frac{\rho_{aggregate}}{1 + \upsilon_{micro} \times \rho_{aggregate}} \tag{1}$$

where, υ_{micro} is the microvolume (0.14 mL/g). The value was found to be 1.34 g/mL. Boehm's titration is usually employed to determine the total acid groups of the adsorbent in the case of activated carbons (El-Sheikh et al., 2004). A weighted amount of adsorbent (0.2 g) was put in an Erlenmeyer flask. A volume of 50 mL of 0.1 mol/L NaOH solution was added. To attain equilibrium, the suspension was shaken for 2 hr at room temperature. After filtration, the residual NaOH concentration was determined by titration with 0.1 mol/L hydrochloric acid solution, using phenolphthalein as indicator. The quantity of consumed NaOH was converted to cation exchange capacity (CEC) and expressed in mequiv./g (Puziy et al., 2002).

A Nicolet 400 D FT-IR spectrophotometer was used to analyse the surface functional groups. A Rigaku diffractometer (USA) with Ni filtered CuKα radiation source was used to record the X-ray diffraction analysis of the

adsorbent. The surface morphology of the adsorbent was studied with the help of a scanning electron microscope (Philips XL-3 CP, the Netherlands) operated at 12 kV. The thermal characteristics of the original coconut buttons and activated carbon were examined by Metler Toledo Star thermogravimetric analyzer (Switzerland). The pH of point of zero charge (pH$_{pzc}$) was determined by the potentiometric method of Schwarz et al. (1984). The apparent density of the adsorbent was determined by specific-gravity bottle using nitrobenzene as displacing liquid. GBC Avanta A5450 atomic absorption spectrophotometer (AAS) was used to determine the metal concentrations. A temperature controlled water bath shaker (Labline, India) with a temperature variation of ± 1°C was used for batch studies.

1.4 Adsorption experiments

Adsorption kinetics and equilibrium isotherms were studied from batch techniques. Batch experiments were carried out in 100 mL Erlenmeyer flasks at 30°C and 200 r/min. A weighed amount (0.1 g) of adsorbent was added to 50 mL of metal solutions of varying concentrations ranging between 10 and 300 mg/L each time and shaken continuously for 4 hr to attain equilibrium. The solutions were centrifuged and the concentrations of metal ions were determined by AAS method. The uptake of metal ions was calculated by the difference in their initial and final concentrations. Effects of pH (2–9), contact time (5–240 min), initial concentration of metal ions (10–300 mg/L) and adsorbent dose (0.05–0.30 g) on the uptake of metal ions from 50 mL solution of wastewater were studied and the optimum values were used in the experiments.

1.5 Desorption experiments

After adsorption experiments with a 25 mg/L metal ion solution, the metal ion loaded carbon was separated by centrifugation. The carbon was washed gently with deionised water to remove any unadsorbed metal ion. The spent adsorbent was agitated with 50 mL of 0.1 mol/L HCl solution for 4 hr. The desorbed metal ions were estimated as before. The regenerated adsorbent sample was reused in the next cycle of the adsorption experiment. The adsorption-desorption experiments were conducted for three cycles.

2 Results and discussion

2.1 Adsorbent characterization

The N$_2$ adsorption isotherm data are shown in Fig. 1. Adsorption measurements were followed by desorption measurements under the same conditions. This isotherm is of type IV based on the IUPAC classification and is typical of mesoporous materials. In these materials, capillary condensation normally takes place, leading to hysteresis loops. Micropore filling occurs at relatively low temperatures.

From the N$_2$ adsorption-desorption isotherm it was clear that the adsorbed volume for the steam-activated
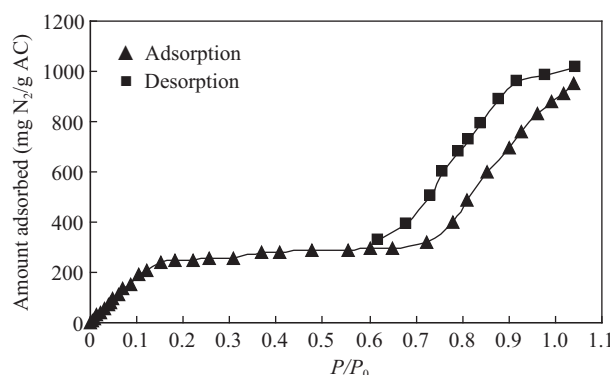

Fig. 1 Nitrogen adsorption isotherm at 77 K on activated carbon (AC).

carbon increases with increasing P/P_0, indicating the wider pore size distribution. The isotherm indicates the micro-mesoporous structure of the carbon. The various porous carbons prepared from pinewood (Tseng et al., 2003), corncob (Wu et al., 2001), bagasse (Juang et al., 2002), and plum kernel (Juang et al., 2000) via steam activation produced increased surface area and the fraction of mesopore volume with increased activation temperature. Similar trends were observed in the carbons prepared from eucalyptus wood and activated in the atmosphere containing CO$_2$ and a small amount of O$_2$ (Hu and Vansant, 1995). But, carbons when activated with KOH, ZnCl$_2$, or H$_3$PO$_4$ did not proportionally enhance the fraction of mesopore volume with the increase of activation temperature (Wu et al., 2005).

Figure 2 shows the pore size distribution of activated carbon and the porosity parameters are given in Table 1. The pore size distribution seems to provide especially useful information on porous solids (Seaton et al., 1989).

The FT-IR spectrum of the activated carbon is given in Fig. 3. FT-IR studies of various carbon materials have identified the presence of several surface functionalities with C=O (carboxylic, anhydride, lactone and ketone) at 1750–1630 cm^{-1}; C=C at 1600–1450 cm^{-1}; C–O stretching and O–H bending (lactonic, ether, phenol, etc.) at 1440–1000 cm^{-1}; CH (900–600 cm^{-1}); OH, NH, CH stretching in alkyl or aryl at 3488–3100 cm^{-1} and S=O at 1118 cm^{-1} (Zawadzki, 1989; Aik and Guo, 2001; Peng et al., 2003; Mohan et al., 2006; Domingo-Garc*ı*a et al., 2002; Rao et

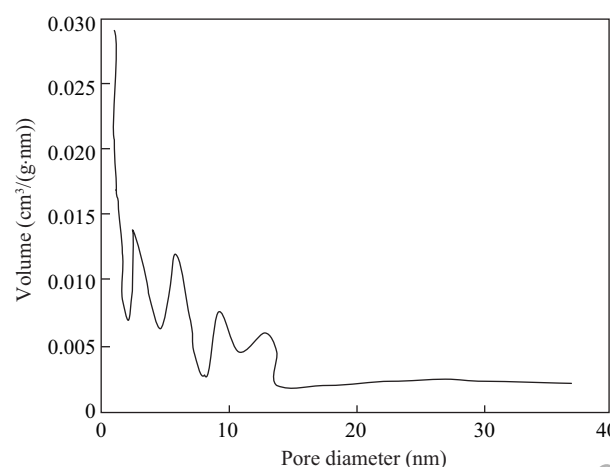**Fig. 2** Pore size distribution of AC.

Table 1 Surface and physical properties of activated carbon

| Parameter	Value
Specific surface area (m^2/g)	479.0
Micropore volume (mL/g)	0.14
pH_{pzc}	5.2
Apparent density (g/mL)	1.06
Aggregate density (g/mL)	1.66
Sphere density (g/mL)	1.34
Total acidity (mequiv./g)	1.0
Carbon (%)	70.82
Hydrogen (%)	2.60
Nitrogen (%)	3.08
Oxygen (%)	20.41
Particle size (mesh size)	−80 + 230 (average particle diameter 0.096 mm)

Fig. 4 XRD pattern of AC.
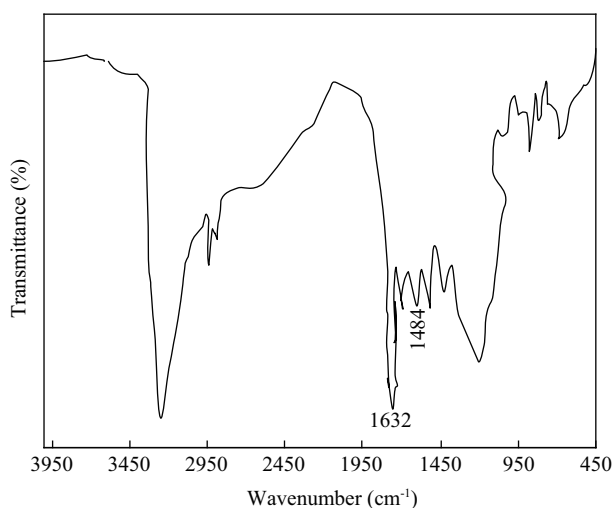
Fig. 3 FT-IR spectrum of AC.

al., 2006; Zhu et al., 2009). The spectrum of the activated carbon in Fig. 3 had several bands assigned to C=O (1632 cm^{-1}), C=C (1484 cm^{-1}), C–O (1382 cm^{-1}), C–H (760 cm^{-1}), O–H, N–H, C–H stretching (3426 cm^{-1}) and S=O (1116 cm^{-1}). The good sorption properties of the adsorbent can be attributed to the presence of the C=O and S=O groups which can coordinate with heavy metal ions. The XRD pattern of activated carbon (Fig. 4) indicates the amorphous nature of the material lacking any crystallinity.

Figure 5 shows SEM photographs of the activated carbon at 5000 and 2000 times magnification. The sample has a rough texture with a large number of pores in different sizes and shapes. Shallow cavities could be observed on the external surface. These cavities and pores on the surface
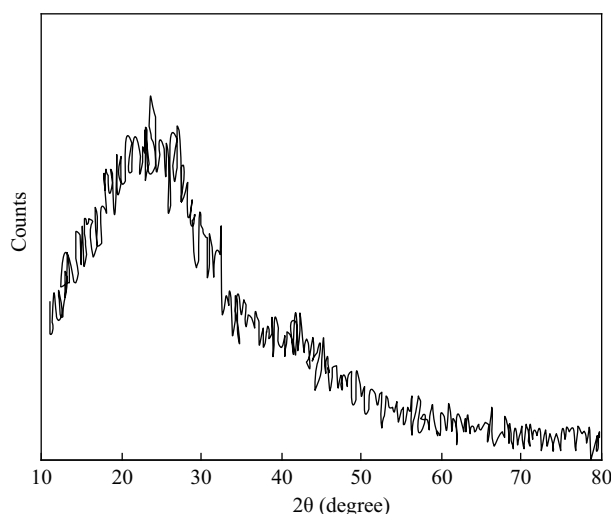
increase the contact area and facilitate pore diffusion during adsorption (Tay et al., 2009).

Pyrolysis behaviour of dried coconut buttons and activated carbon were studied using TG analysis (Fig. 6). The TGA and DTG curves of coconut buttons, which was used as the raw material for the preparation of activated carbon, show weight losses at three different stages. The first weight loss (10.20%) by heating the materials upto 150°C has been due to the elimination of moisture. The second stage of decomposition from 150–380°C (T_1 = 299°C), corresponds to a weight loss of 50.07%, was due to vapourisation of volatile matters and tars, which results in the carbonization of coconut buttons. In the third stage from 380–580°C (T_2 = 473°C), a weight loss of 76.4% of the initial dry weight was observed. This transition is the characteristic of pyrolitic depolymerisation process in lignin and internal thermal rearrangement of the carbohydrate anhydride residues resulting in char formation; eventually provide pseudo-graphitic layers with microporous interstices (Pollard et al., 1995). In the TGA and DTG curve of activated carbon the first weight loss around 80°C observed was probably caused by the thermodesorption of water vapour. The activated carbon is thermally stable up to 600°C. At higher temperatures, some partial decomposition takes place due to decomposition of graphene structure (Kim and Yie, 2005). Enhancement in the thermal stability is observed upon activation. TG analysis exhibited that steam activation resulted in activated
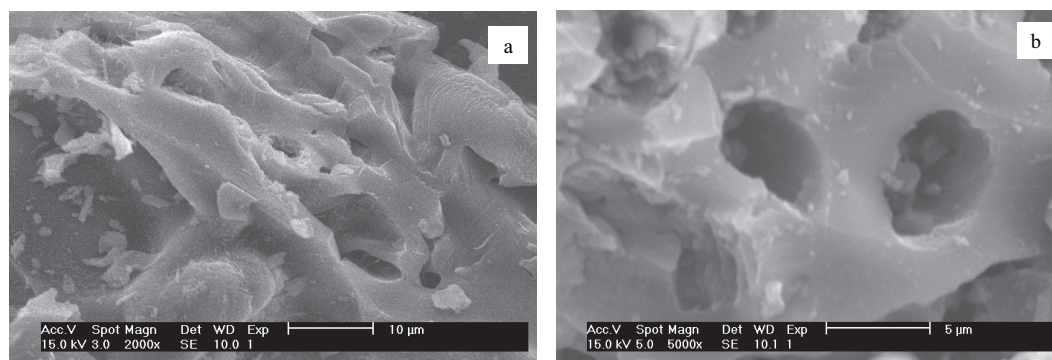
Fig. 5 SEM photographs of AC at ×2000 (a) and ×5000 (b) magnifications.
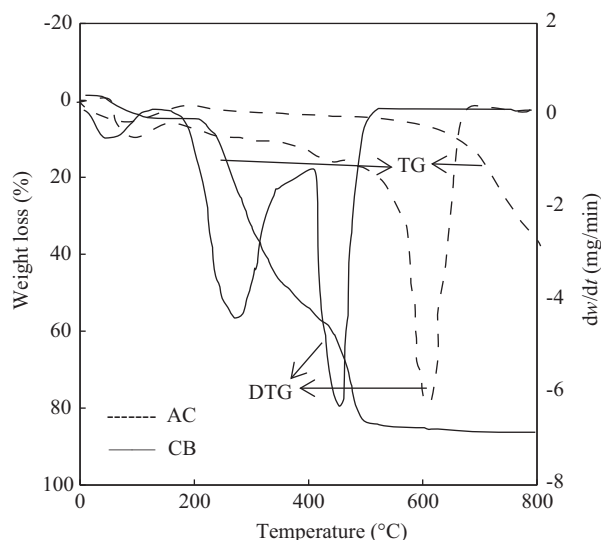
Fig. 6 TG and DTG curves of coconut buttons (CB) and activated carbon (AC).

Fig. 7 Effect of pH on the adsorption of metal ions onto AC. Conditions: adsorbent dose 2 g/L; temperature 30°C; equilibrium time 3 hr; concentration 25 mg/L.

carbon material with higher thermal stability compared to the precursor, coconut buttons.

The pH dependent surface charge is widely used to predict the adsorption characteristics of adsorbents. The pH_{pzc} of AC was found to be 5.2.

2.2 Effect of pH on the adsorption of metals

Effect of pH on the removal of Pb(II), Hg(II) and Cu(II) was evaluated at a pH range 2.0–9.0. Figure 7 shows the removal of Pb(II), Hg(II) and Cu(II) by activated carbon as a function of pH. The percentage removal was found to increase gradually with increasing pH and reaches an optimum value of pH 6.0 for Pb(II) and Cu(II), and 7.0 for Hg(II). The solution pH affects the surface charge of the adsorbent, the degree of ionization and the speciation of the surface functional groups. The point of zero charge (pH_{pzc}) of the adsorbent was 5.2. Below 5.2 the surface of the adsorbent is positively charged and above this pH the surface of the adsorbent becomes negative. The perusal of metal speciation diagram (Lindsay, 1979) indicates that in the highest sorption range (6.0–7.0), the predominant species of metal in solution are M^{2+} and $M(OH)^{+}$ ions. The increase in metal removal with increase in pH values can be explained on the basis of a decrease in competition between proton and the metal cations for the same functional groups and by the decrease in positive charge of the adsorbent which results in a lower electrostatic repulsion between the metal cations and the surface.

2.3 Effect of ionic strength

The effect of ionic strength on the adsorption of metal ions was investigated by conducting batch experiments with varying concentrations of 0.001, 0.005, 0.01, 0.05 and 0.1 mol/L NaCl. When the ionic strength increased from 0.001 to 0.1 mol/L, the percentage removal decreased from 98.7% to 69.8% for Pb(II), 95.8% to 60.3% for Hg(II) and 90.6% to 49.2% for Cu(II), at an initial concentration of 25 mg/L. The inverse relation of ionic strength on adsorption capacity suggests the possibility of ion exchange mechanism in the process of adsorption. The Na^{+} ions used in the
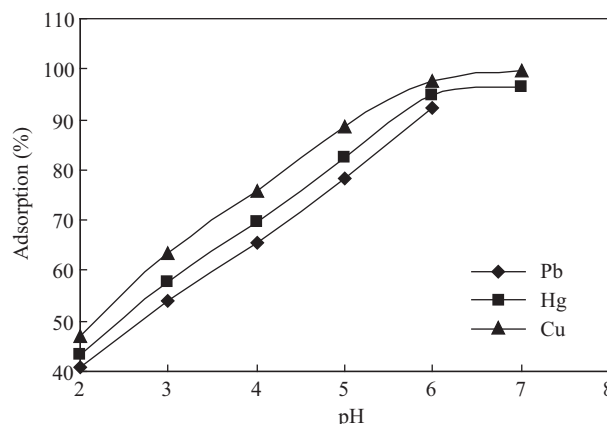
salt compete with metal ions for the cation exchange sites of the activated carbon. According to McBride (1997), ions that form outer-sphere surface complexes show decreasing adsorption with increasing ionic strength. The decrease in adsorption with increase in ionic strength suggests that the metal ions are adsorbed on the activated carbon by forming outer-sphere complexes.

2.4 Effect of contact time and initial concentration

The effect of contact time on the uptake of Pb(II), Cu(II) and Hg(II) is shown in Fig. 8. Adsorption experiments were carried out with varying initial metal ion concentrations from 25 to 100 mg/L using 2 g/L activated carbon. The results show that the increase in the contact time increased the adsorption of metal ions, and it remained constant after the equilibration time of 3 hr. The contact time required for the maximum uptake of metal ions by activated carbon was independent of the metal ion concentration. Therefore, the optimum agitation period of metal adsorption is about 3 hr. The amount of heavy metal adsorbed by activated carbon at equilibrium increased from 12.45 to 44.40 mg/g for Pb(II), 12.21 to 40.77 mg/g for Hg(II) and 11.92 to 36.40 mg/g for Cu(II) when changing the initial concentration from 25 to 100 mg/L. This indicates that the initial metal concentration plays an important role in the adsorption capacity of metals on the activated carbon surface.
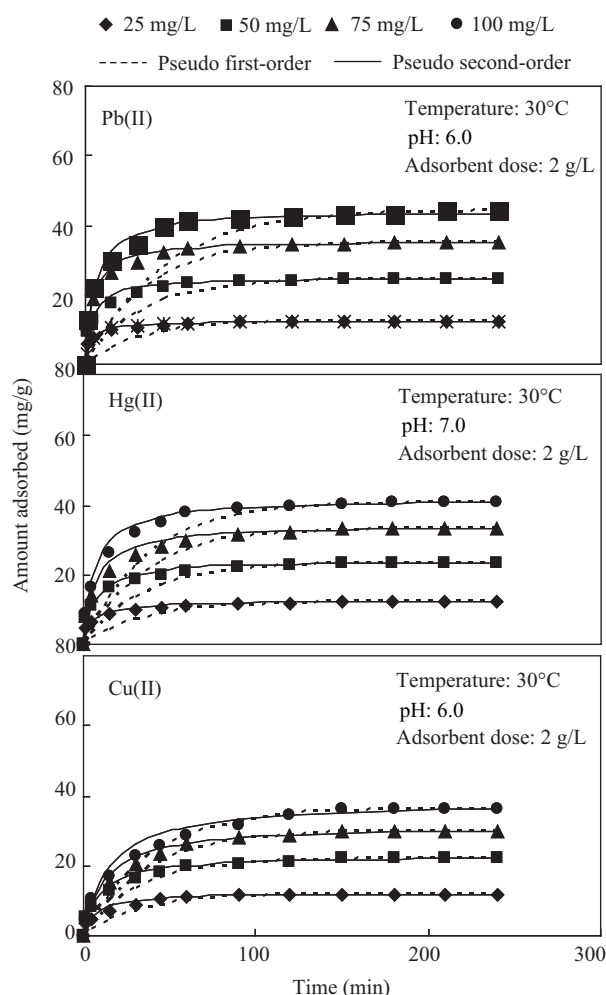
2.5 Adsorption kinetics

In order to examine the controlling mechanism of the adsorption process pseudo first- and second-order kinetic equations (Ho and Mckay, 1999) were used to test the experimental kinetic data. The pseudo first-order rate kinetic equation of Lagergren(1898) is expressed as:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303}t \tag{2}$$

where, q_e (mg/g) and q_t (mg/g) are the amounts of metal ions adsorbed at equilibrium and at time t (min) respectively, and k_1 (min^{-1}) is the rate constant of adsorption. The values of k_1 and q_e were evaluated from the linear plots of $\log(q_e-q_t)$ versus t for different concentrations of metal ions. The comparison of the experimental results

Fig. 8 Comparison of experimental contact time data and the fittings to pseudo first-order and second-order kinetic models for the adsorption of Pb(II), Hg(II) and Cu(II) onto AC at different concentrations.

and calculated values of the contact time for different concentrations of metal ions is shown in Fig. 8. The calculated q_e values do not agree with the experimental q_e values.

The pseudo second-order rate equation is expressed as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{3}$$

where, k_2 (g/(mg·min)) is the rate constant of second-order adsorption. The straight line plots of t/q_t against t have

also been analysed. The values of q_e, k_2 and the correlation coefficients R^2 for the metal ions at different concentrations were calculated from these plots and are shown in Table 2. The values of R^2 for the pseudo second-order kinetic model are greater than 0.990. The half-life time was found to vary with the initial concentration, indicating the assumption of a second-order adsorption mechanism rather than first-order one. The half-time increased from 5 to 10 min for Pb(II), 5 to 15 min for Hg(II) and 10 to 25 min Cu(II) when the initial metal concentration increased from 25 to 100 mg/L. The calculated q_e values from pseudo second-order equation are in good agreement with the experimental values (Fig. 8). Therefore the sorption reaction belongs to the pseudo second-order kinetic model and that the overall rate of the metal adsorption process appears to be controlled by the chemical process via ion exchange and/or complexation process.

2.6 Equilibrium isotherms

Adsorption isotherms are useful for finding out the adsorption capacity of the adsorbent, the solute-solution interaction and the degree of accumulation of adsorbate on the surface of the adsorbent. In the present study, the adsorption of metals onto activated carbon was studied by two adsorption isotherm models: Langmuir and Freundlich isotherms.

Langmuir isotherm:

$$\frac{C_e}{q_e} = \frac{1}{Q^0 b} + \frac{C_e}{Q^0} \tag{4}$$

Freundlich isotherm:

$$\ln q_e = \ln K_F + \ln C_e \tag{5}$$

where, q_e (mg/g) is the amount of adsorbed metal ions per unit weight of adsorbent at equilibrium concentration, C_e (mg/L). The Q^0 (mg/g) and b (L/mg) are the Langmuir constants related to the maximum monolayer capacity and energy of adsorption, respectively. The K_F and $1/n$ are Freundlich constants related to adsorption capacity and intensity of adsorption respectively. Langmuir isotherm assumes a monolayer adsorption surface without any lateral interaction between adsorbed molecules. Freundlich model is suitable for describing multilayer adsorption with interaction between adsorbed molecules. The Langmuir

Table 2 Kinetic parameters for the adsorption of Pb(II), Hg(II) and Cu(II) ions onto activated carbon

Metal ion	Concentration (mg/L)	Pseudo first-order		Pseudo second-order		
		K_1 ($\times 10^{-2}$ min^{-1})	R^2	K_2 ($\times 10^{-2}$ g/(mg·min))	q_e (mg/g)	R^2
Pb(II)	25	3.18	0.954	2.43	12.62	0.999
	50	3.08	0.950	0.98	25.01	0.999
	75	2.88	0.934	0.72	35.63	0.999
	100	2.21	0.874	0.41	45.05	0.999
Hg(II)	25	2.88	0.963	1.69	12.45	0.999
	50	2.82	0.977	0.68	24.15	0.998
	75	2.76	0.970	0.43	34.42	0.998
	100	2.68	0.935	0.39	41.83	0.999
Cu(II)	25	3.13	0.963	1.24	12.28	0.999
	50	3.09	0.964	0.55	22.77	0.998
	75	2.73	0.983	0.31	31.27	0.997
	100	2.61	0.976	0.18	38.60	0.994

and Freundlich parameters were obtained by plotting C_e/q_e versus C_e and $\ln q_e$ versus $\ln C_e$, respectively.

The adsorption constants were calculated for concentration ranges between 10 and 300 mg/L according to the least squares fitting method and the results are presented in Table 3. The validity of the isotherm models was tested by comparing the experimental and calculated data at 30°C (Fig. 9). Based on the correlation coefficients (R^2), it is clear that the adsorption of metal ions on to activated carbon is best fitted to the Freundlich adsorption isotherm for the entire range of concentrations. The fact that the Freundlich isotherm fits the experimental data very well may be due to heterogeneous distribution of active sites on the carbon surface, since the Freundlich equation assumes that the surface is heterogeneous. The values of n for Freundlich isotherm were found to be greater than 1, indicating that metal ions are favourably adsorbed by activated carbon. According to the data in Table 3, the maximum adsorption capacity of activated carbon is in the decreasing order Pb > Hg > Cu which is in the order of increasing ionic radius of the metal ions studied (Brown et

al., 2000; Rao et al., 2007). The greater the ionic radius, the smaller is the hydrated ionic radius and greater is the affinity of the metal ions on the active sites of the adsorbent.

2.7 Comparison with other adsorbents

While the Freundlich model does not describe the saturation behaviour of the sorbents, the Langmuir constant, Q^0, represents the monolayer saturation at equilibrium or the total capacity of the adsorbent for heavy metals. The values of the adsorption capacities (Q^0) obtained in this study were 94.35 mg/g for Pb(II), 82.09 mg/g for Hg(II) and 75.78 mg/g for Cu(II). The values of Q^0 reported for the adsorption of Pb(II) were 64.79, 34.72 and 12.61 mg/g for red mud (Gupta et al., 2001), carbon aerogel (Goel et al., 2005) and rice husk ash (Feng et al., 2004), respectively. Previously reported adsorption capacities for the adsorption of Hg(II) onto some low cost adsorbents like activated carbon made from sago waste (Kadirvelu et al., 2004), rice husk ash (Feng et al., 2004) and carboxyl banana stem (Anirudhan et al., 2007) were 55.60, 6.72 and 88.98 mg/g, respectively. The values of Q^0 for the adsorption of Cu(II) were reported to be 19.50, 81.47 and 51.51 mg/g for adsorption onto activated carbon from *Phaseolus aureus* hulls (Rao et al., 2009), tannin-immobilised hydrotalcite (Anirudhan and Suchithra, 2008) and dehydrated wheat bran (Ozer et al., 2004).

2.8 Testing industrial wastewaters

The utility of the material has been demonstrated by treating with industrial wastewater. Electroplating industry and chlor-alkali industry wastewater samples were used for Cu(II) and Hg(II). Wastewater samples, collected from an electroplating industry and a chlor-alkali industry situated in the city of Cochin, India, were characterized by standard methods (APHA, 1992). Synthetic wastewater is used for Pb(II). The composition of the wastewaters is represented in Table 4. The effect of adsorbent dose on the adsorption of metal ions was studied (Fig. 10). Almost complete (100%) removal of Pb(II), Hg(II) and Cu(II) from 1 L of sample was achieved with 1.5, 2.0 and 4.0 g/L of activated
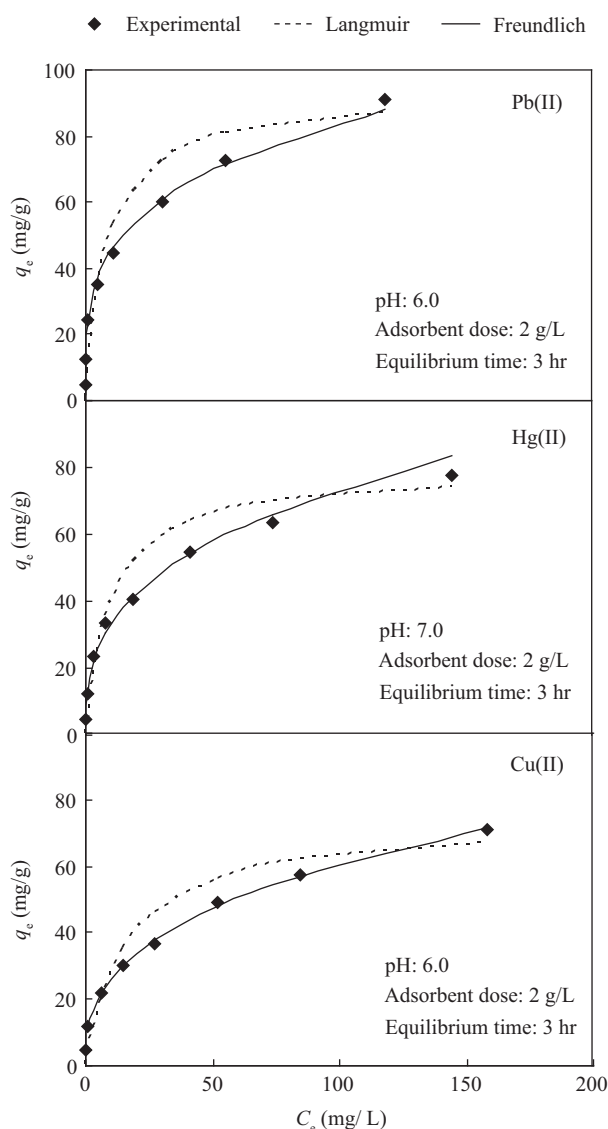
Fig. 9 Comparison of model fits of the Langmuir and Freundlich models to the experimental data for the adsorption of metal ions onto AC.
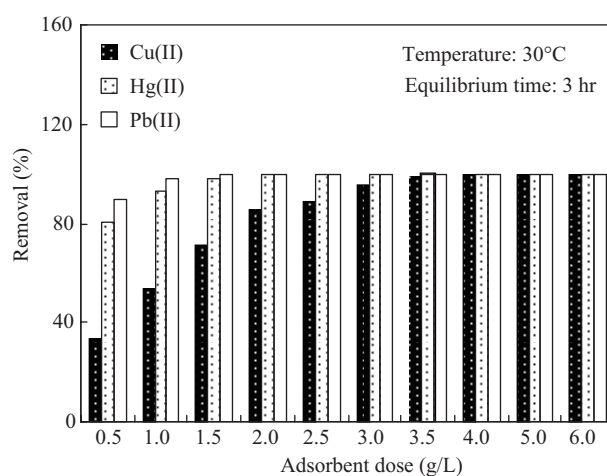
Fig. 10 Effect of adsorbent dose on the removal of Pb(II), Hg(II) and Cu(II) ions from industry effluent samples by AC.

Table 3　Isotherm parameters for the adsorption of metal ions onto activated carbon

Metal	Langmuir			Freundlich		
	Q^0 (mg/g)	b (L/mg)	R^2	K_F	$1/n$	R^2
Pb(II)	92.72	0.122	0.977	23.81	0.274	0.995
Hg(II)	78.84	0.104	0.978	14.96	0.345	0.995
Cu(II)	73.6	0.063	0.966	11.32	0.363	0.999

Table 4　Chemical composition of waste waters (mg/L)

Synthetic wastewater for Pb(II)		Chlor-alkai industry wastewater for Hg(II)		Electroplating wastewater for Cu(II)	
Composition	Value (mg/L)	Composition	Value (mg/L)	Composition	Value (mg/L)
Pb^{2+}	10.0	Hg^{2+}	9.9	Cu^{2+}	43.3
Ca^{2+}	60.0	Pb^{2+}	3.2	Ni^{2+}	11.7
Mg^{2+}	14.0	Cd^{2+}	0.6	Na^+	314.7
Na^+	20.0	Mg^{2+}	31.5	K^+	28.8
K^+	3.0	Ca^{2+}	36.3	Mg^{2+}	611.4
SO_4^{2-}	170.0	Na^+	260.8	Ca^{2+}	43.3
Cl-	30.0	NH_4^+	27.1	Cl^-	319.9
SO_4^{2-}	3.0	PO_4^{3-}	13.3	SO_4^{2-}	29.3
SiO_2	8.0	NO_3^-	17.1	CO_3^-	89.9
pH	6.5	Cl^-	415.7	Hardness as	
		pH	8.3	$CaCO_3$	714.4
				COD	413.3

carbon, respectively. These results are in good agreement with those obtained from batch experiments which show that the activated carbon can effectively remove the metal ions even in the presence of other ions.

2.9 Desorption studies

Desorption studies are aimed at the recovery of the adsorbed metals as well as the regeneration of the adsorbent. Desorption studies of the metal ions were carried out in three cycles with different concentrations of HCl solutions, which has been reported to be an efficient metal desorbent (Saeed and Iqbal, 2003). The desorption of metal ions was found to be more effective by using 0.2 mol/L HCl solution. The H^+ from HCl easily displaces metal ions bonded to the adsorbent during the desorption stage. After three cycles the sorption capacity of activated carbon decreased from 99.6% to 94.5% for Pb(II), 97.1% to 89.2% for Hg(II) and 95.2% to 85.2% for Cu(II). The recovery of Pb(II), Hg(II) and Cu(II) decreases from 94.3% to 86.5%, 96.5% to 88.5% and 97.4% to 90.6%, respectively.

3 Conclusions

The present investigation shows that the activated carbon prepared by steam activation of coconut buttons can be employed as a potentially low cost sorbent for the removal of Pb(II), Hg(II) and Cu(II) ions from industrial effluents. Batch adsorption test showed that the extent of metal adsorption was dependent on metal concentration, contact time, pH, ionic strength and carbon dose. The maximum removal of the metal ions is observed at the pH 6.0 for Pb(II) and Cu(II), and pH 7.0 for Hg(II). The kinetics of the adsorption is fast in the first 30 min and attained equilibrium at 3 hr. The adsorption follows pseudo second-order kinetics. The Langmuir and Freundlich adsorption models were used to represent the experimental data and the equilibrium data fitted well with the Freundlich model.

The monolayer adsorption capacity was found to be 92.72 mg/g for Pb(II), 78.84 mg/g for Hg(II) and 73.60 mg/g for Cu(II) at 30°C. The adsorption capacity of activated carbon was found to be relatively high compared with some other adsorbents reported in literature. The utility of the adsorbent was tested using industrial wastewater samples. Desorption of the metal ions was carried out using 0.1 mol/L HCl and the repeated adsorption-desorption study showed that the adsorbent AC can be effectively used as an adsorbent for the removal of heavy metal ions from industrial effluents.

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