Original Article

Sorption of Lead (II) Ions on Activated Coconut Husk

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ABSTRACT

Background: In recent years, various toxic chemicals/compounds have been widely detected at dangerous levels in drinking water in many parts of the world posing a variety of serious health risks to human beings. One of these toxic chemicals is lead, so this paper aimed to evaluate of efficiency coconut husk as cheap adsorbent for removal lead under different conditions.

Methods: In the spring of 2015, batch studies were performed in laboratory (Branch of Hamadan, Islamic Azad University,) to evaluate the influences of various experimental parameters like pH, initial concentration, adsorbent dosage, contact time and the effect of temperature on the adsorption capacity of coconut husk for removal lead from aqueous solution.

Results: Optimum conditions for Pb (II) removal were pH 6, adsorbent dosage 1g/100ml of solution and equilibrium time 120 min. The adsorption isotherm was also affected by temperature since the adsorption capacity was increased by raising the temperature from 25 to 45 °C. The equilibrium adsorption isotherm was better described by Freuindlich adsorption isotherm model.

Conclusion: It is evident from the literature survey that coconut-based biosorbents have shown good potential for the removal of various aquatic pollutants. Coconut husk-based activated carbon can be a promising adsorbent for removal of Pb from aqueous solutions.

Keywords: Biosorption, Coconut Husk, Pb (II), Water Pollution.

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INTRODUCTION

Water contamination with heavy metals is a serious problem due to toxicity of heavy metals [1].The presence of heavy metals in water source is becoming a major environmental and public health issue [2]. The heavy metals are special because of their persistency and toxicity in the environment. About 20 metals are classified as heavy metals, which half of them are harmful to human health because of toxicity [3]. As one of the important toxic heavy metals, lead in human causes severe hurts to, liver, nervous system, reproductive system, kidney and brain. Severe exposure to lead has been related with sterility, stillbirths, abortion and neo-natal deaths. Industrial activities, such as battery manufacturing, metal plating and finishing, printing and pigment, ammunition, soldering material, ceramic and glass industries, iron and steel manufacturing units produce large quantities of wastewater containing lead [4]. Common purifier methods for the removal of toxic metal comprise membrane separation [5], electrochemical precipitation, fertilization and adsorption, emulsion per traction, ion exchange differ with respect to cost, complexity and efficiency [4].

From last few decades, biosorption method has appeared as an economic and efficient alternative for water and wastewater treatment utilizing natural, cheap, renewable and abundant. At present, biosorption method has been enriched by a vast amount of papers published in different journals [6]. Among different purifying methods, adsorption technique is one of the fantastic techniques used for removing heavy metals from wastewater [7]. Biosorption is an emerging technique for wastewater treatment utilizing biomaterials such as agricultural wastes [6]. Biomass and other waste materials may also offer a cheap, available and renewable additional supply of activated carbon. These waste materials have low or no economic cost and often present a disposal problem. Converting these low-cost by-products into activated carbon makes it valuable and solving waste disposal and most attractive provide a potentially commercial activated carbon [8].

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In recent years, some researchers have been searching for more cost-effective methods to obtain the carbon. As carbon can be achieved theoretically from any carbonaceous materials that are rich of element carbon, some low-value and easily available agricultural by-products, such as peanut shell, rice husk, cassava peel, sawdust, olive kernels and wheat straw have been utilized as the precursors of activated carbon to remove heavy metals. Newly, other materials such as algal bloom waste and durian peel have also been explored in order to prepare activated carbon [9]. Babassu coconut mesocarp, an abundant agricultural lignocellulosic by-product, is a fibrous residue left after producing the nuts [10]. Available abundantly, high adsorption capacity, cost effectiveness and renewability are the major factors making coconut husk as commercial alternatives for water and wastewater treatment [6].

To make better use of this cheap and vast agricultural waste, it is proposed to convert coconut husk (CH) into activated carbon. Conversion of coconut husk to activated carbon will supply a double purpose. First, unvalued agricultural waste is converted to useful, valuable adsorbent and second, the use of agricultural by-products represents a high potential source of adsorbents, which will enter to solving part of the wastewater treatment problem in many countries [11].

In the present research, efficiency of activated coconut husks for the removal of Pb from aqueous solution has been studied and the results have been analyzed.

MATERIALS AND METHODS

Preparation of Activated Carbon

The coconut (CH) was bought in the spring of 2015, and then coconut husk was separated, cleaned and thoroughly washed with distilled water, and then dried in an oven. This dried coconut husk was then treated with H_2O_2 solution for 24 h to oxidize adhering organic impurities and washed well with double distilled water to remove the excessive hydrogen peroxide then dried at 110 °C for 1 h under vacuum [12]. Dried coconut shell fibers were activated at 700 °C for 2 h. The material was grounded and sieved to desired particle scales such as <106, 106–125, 125–180, 180–212, 212–250, 250–300, >300 BSS mesh. Finally, granules of activated coconut husk (ACH) thus obtained were stored in separate vacuum desiccators until required [13]. All experiments have been done in the laboratory of Branch of Hamadan, Islamic Azad University

Chemicals

A 1000 mgL^{-1}Pb (II) stock solution was prepared by dissolving a weighed quantity of Pb $(NO_3)_2$ in distilled water. All samples and solutions for adsorption and analysis were prepared by suitable dilution of the freshly prepared stock solution. The pH measurements were made using a Metrohm pH meter. Test solutions pH_s were adjusted using reagent grade dilute H_2SO_4 (0.1N) and NaOH (0.1N).

Batch Equilibrium Studies

Study of Adsorption Isotherms

Experiments were conducted in batch mode. One hundred ml samples of aqueous solutions of metal ions at different initial concentrations (100–800 mgL^{-1}) and at modified pH were transferred into 250 ml Erlenmeyer flask. Specified amounts of the coconut husk were added to these solutions. After 24 h, solutions were filtered and Pb ion concentrations in the filtrate were measured. Concentrations of Pb ions in samples were determined ICP-OES. Concentration of metal remained in the sorbent phase q_e (mgg^{-1}) was calculated from the expression:

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

Where C_0 and C_e are the initial and final (equilibrium) concentrations of the metal ion in solution (mgL^{-1}), W is the mass of the sorbent (g) and V is the solution volume (L).

All experiments were carried out at a constant situation. Throughout the study, the initial metal concentrations from 100 to 800 mgL^{-1}, the pH quantities varied from two to six, the initial biomass Concentrations from 2.5 to 4 g 100 mL^{-1}, the temperature from 25 to 45 °C, and the contact time from 10 to 120 minutes [14].

Langmuir Isotherm

Langmuir isotherm supposes monolayer adsorption onto a surface containing a limited number of adsorption sites of uniform strategies of adsorption with no transmigration of adsorbate in the area of surface [15]. Equation for linear form of Langmuir isotherm is given by the following equation:

$$\frac{C_e}{q_e} = \frac{1}{Q_0\ b} + \frac{1}{Q_0} C_e \qquad (2)$$

Where C_e is the equilibrium concentration of the adsorbate (mgL^{-1}), q_e is the amount of adsorbate adsorbed per unit mass of adsorbent (mgg^{-1}), Q_o and b are Langmuir constants related to adsorption capacity and rate of adsorption, respectively [15].

Freundlich Isotherm

Freundlich isotherm in the other hand assumes heterogeneous surface energies, in which the energy term in Langmuir equation differs as a function of the surface coverage [11]. The famous equation for Freundlich isotherm is presented by the following equation:

$$q_e = K_F C_e^{1/n} \quad (3)$$

where qe is the measure of adsorbate adsorbed per unit mass of adsorbent (mgg-1), C_e is the equilibrium concentration of the adsorbate (mgL^{-1}), *K*F and *n* are Freundlich constants with *n* giving an indication of how suitable the adsorption process.

RESULTS

Adsorption of heavy metal ions onto the surface of a biological material was affected by following factors: biomass concentration, pH, metal ion concentration, time and temperature.

Effect of Biomass Concentration

Coconut husk (CH) quantity for removal of Pb ions was investigated by adding different amounts of CH in the 1–4 g at a temperature of 25±0.1 ˚C. The number of sites accessible for biosorption depends upon the amount of the adsorbent. The effect of the coconut husk concentration on the metal removal efficiency is shown in Figure 1. The Pb ions removal was increased linearly with the inceasing value of biosobant up to the biomass concentration of 4 g100 mL-1. Beyond this dosage, the increase in removal efficiency was lower.

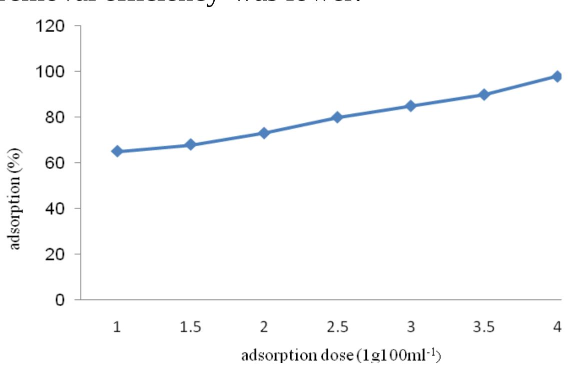

Figure 1. Effect of adsorbent dose on the adsorption of Pb (pH: 6.0; temperature: 25 °C and equilibrium time: 120 min).

Effect of PH

The effect of the solution pH on the adsorption of Pb ions onto coconut husk was assessed at different values, ranging from 2 to 6, with a stirring time of 120 min. In these experiments, the initial metal concentration and dos of adsorbent were set at 100 mgL^{-1} and 1 g, respectively, for all batch tests in this experiment. As shown in Figure 2, the metal uptake increased with the increasing pH in the range of 2 to 6. At pH values of about 6, sorption capacities achieved maximum values.

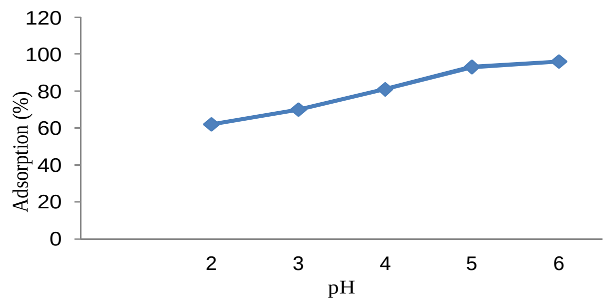

Figure 2. The adsorption of Pb onto CH as a function of the equilibrium pH (adsorbent dose: $1g100mL^{-1}$; equilibrium time: 120 min and temperature: 25 °C).

Effect of Contact Time

Experiments were carried out for various contact times with a fixed adsorbent dose, pH and concentration. Typical biosorption kinetics exhibits a fast initial uptake, followed by a slower process. The highest level of heavy metal removal took place within the first 90 min (Figure 3). After this time, the amount of bound metal ions did not change with rapid slope during the course of the process.

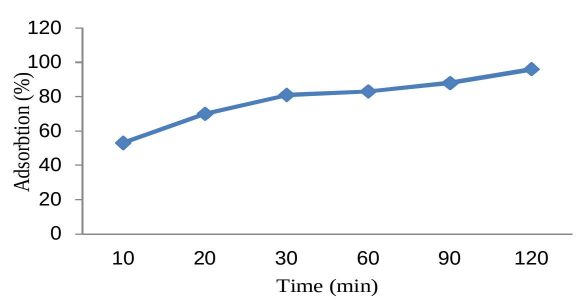

Figure 3. The adsorption of Pb onto CH as a function of the time (adsorbent dose: $1g100ml^{-1}$; equilibrium pH: 6 and temperature: 25 °C).

Effect of Temperature

To study the effect of temperature on the Pb (II) adsorption over CH three temperatures 25, 35 and 45 °C were experimented (Figure 4). The results showed that adsorption process increased with adding temperature from 25 to 45 °C. The quantities adsorbed passed through a maximum at 45 °C and then started to decrease with the temperatures increasing to 50 °C.

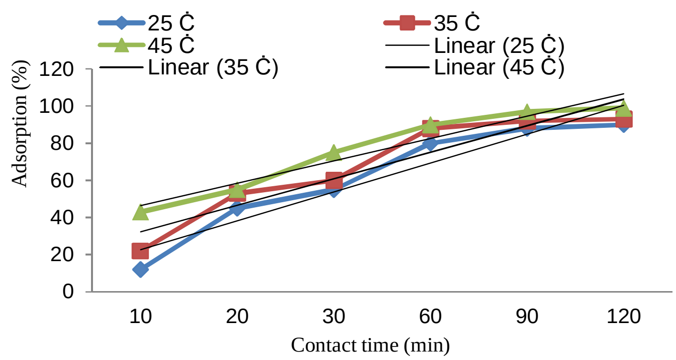

Figure 4. The adsorption of Pb onto CH as a function of the temperature (adsorbent dose: 1g100ml^{-1}; equilibrium time: 120 min and pH: 6).

Effect of Initial Concentration of Pb

The percentage removal of Pb (II) ion by the adsorbent initially decreased slowly with adding Pb ions concentration to 600 mgL^{-1} and then declined rapidly when Pb ions concentration reached to 800 mgL^{-1} (Figure 5).

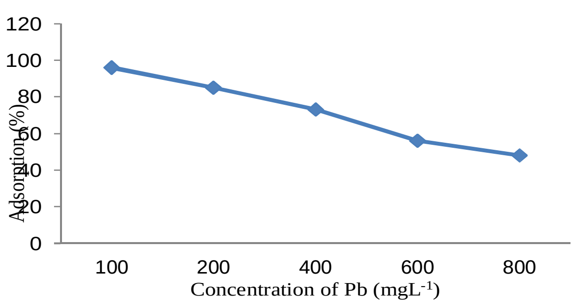

Figure 5. The adsorption of Pb onto CH as a function of the initial concentration (adsorbent dose: 1 g/100ml^{-1}; equilibrium time: 120 min and pH: 6).

Adsorption Isotherms

The adsorption isotherm demonstrates how the adsorption molecules release between the liquid phase and the solid phase when the adsorption process reaches an equilibrium phase. To find the acceptable model that can be used for design isotherm model, different isotherm models were analyzed. Adsorption isotherm study was carried out on two isotherm models: the Langmuir and Freundlich isotherm models. Correlation coefficients R^2 size is used to describe the applicability of the isotherm equation. For the Langmuir isotherm, when C_e/q_e is traced against C_e, a straight line with slope of $1/Q_o$ is earned (Figure 6). The Langmuir constants b and Qo were calculated from Eq [3].

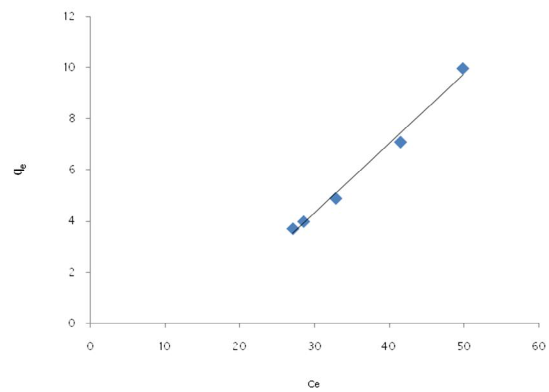

Figure6. Langmuir adsorption isotherm of Pb (II) onto CH at 25 ◦C

For the Freundlich isotherm, the plot of ln q_e against ln C_e affords a straight line with slope of $1/n$, as presented in Figure 7, which showed that the adsorption of Pb (II) on the CH was desirable. Therefore, Freundlich constants *K*F and *n* were calculated from Eq [16].

The correlation coefficient, R^2 of 0.99 showed that the adsorption process of Pb ions on the prepared activated carbon was well fitted to the Freundlich isotherm.

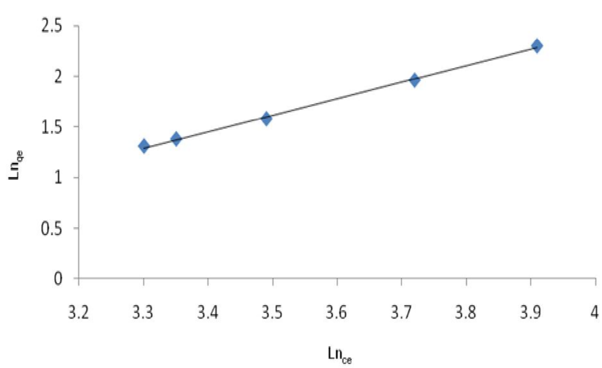

Figure 7. Freundlich adsorption isotherm of Pb (II) onto CH at 25 ◦C

Table 1. Adsorption capacities for some activated carbon.

Adsorbents	Adsorption capacity (mgg^{-1})	Sources
Pineapple stem	119.05	[17]
Coconut husk	99	[18]
Coffee husks	90.1	[19]
Garlic peel	82.64	[20]
Fallen phoenix tree's leaves	80.9	[21]
Raw date pits	80.3	[22]
Ground hazelnut shells	76.9	[23]
Coconut husk based activated carbon	66	[24]
Charcoal	62.7	[25]
Activated Rosa canina seeds (500 ◦C)	47.2	[26]
Olive pomace	42.3	[25]
Rice husk	40.59	[27]
Cherry sawdust	39.84	[13]
Activated date pits (900 ◦C)	17.3	[22]
Activated olive stones with 40 wt.% PbCl2 at 873K	16.1	[28]
Activated date pits (500 ◦C)	12.9	[22]
Hazelnut shell-activated carbon 750 ◦C	8.82	[16]
Coir pith carbon	5.87	[29]
Apricot stones-activated carbon 750 ◦C	4.11	[25]
Walnut shell-activated carbon 750 ◦C	3.53	[16]
Almond shell-activated carbon 750 ◦C	1.33	[16]
Fir wood based activated carbon	1.21	[30]
Corncob based activated carbon	0.84	[31]
This work	65	

DISCUSSION

As presented in Figure 1, the Pb uptake was increaseD when concentration of bisorbent reacheD to the biomass concentration of 4 g/100ml. Increasing the biosorbent dosage caused a wise in the biomass surface area and in the number of potential binding sites [24]. The effect of amount of adsorbent on the rate of uptake of Cr (III) was carried out at 0.5, 1, and 2 g and the uptake increased with increase in amount of the adsorbent material [32].

pH can affect protonation of functional groups (i.e.carboxyl, phosphate and amino groups) on the biosorbent phase, as well as the chemistry of the metal (i.e. its solubility). When the pH decreased, concentrations of protons enhanced and make competition between H^+ and metal ions on binding sites. Protonated active sites were unable of binding the bind metal ions because free ions remain in the solution [14]. Since biosorption is reversible process, decreasing pH would result in deprotonation. This feature is applied in regeneration of biosorbents. Another explanation is that with adding pH, solubility of complexes of metals decreases [33]. Description of mechanism of biosorption and parameters affecting its performance is necessary for the facility of the operation conditions for biosorption itself and for recycling the solid phase [34].

It is clear from the Figure 3 that the uptake of Pb (II) increases slowly with the lapse of time and reaches to saturation in 2 h. At the initial contact, time due to high amounts of available adsorbent surface the rate of adsorption was fast. The lesser yield of adsorption after passing time could be due to two reasons. First, saturation of sites reduced the availability of active surface sites on the adsorbent. Second, the remaining empty surface sites were difficult to be occupied due to repulsive force of the adsorbed metal ions on the solid phase [35]. Similar results were observed where the effect of contact time on removal of Pb (II) and Hg from aqueous solution by rice husk ash showed adsorption increased with increasing contact time [36].

The equilibrium uptake of metal ions to coconut shells was affected by temperature and increased with the increasing temperature up to 45 °C. It could offer superiority of physical sorption over chemical sorption in this part. High temperature may damage active binding sites and cause decrease adsorption. The process involved the conduction of metal ions from the bulk liquid to the solid phase and the sorption of metal ions onto the biosorbent surface similar to previous study [14].

The uptake of the Pb ions was studied separately over CH at initial Pb concentration ranging from 100 to 800 mgL^{-1}. The rate of the removal of Pb is faster at lower concentration (100 mgL^{-1}) and beyond this, it decreased. At lower concentrations, Pb would interact with the binding sites and thus 100% adsorption happens. As at higher concentrations, binding sites are saturated more Pb ions are left un-adsorbed in solution. This indicates that energetically less favorable sites become involved with increasing ion contents in the aqueous solution [3].

Adsorption isotherm studies are of fundamental importance in determining the adsorption capacity of Pb (II) onto Coconut husk (CH) and to diagnose the nature of adsorption. The correlation coefficient, R^2 of 0.99 showed that the adsorption data of Pb (II) on the prepared activated carbon was well fitted to the Freundlich isotherm. This implies that the adsorption of Pb (II) ions occur on a heterogeneous surface. The activated carbon prepared in this study had a relatively high adsorption capacity of 65 mgg^{-1} if compared to some data obtained from other papers. The large adsorption volume of the activated carbon prepared in this work could be due to its relatively large surface area and its special mesoporous structure. The differences between adsorption capacities of coconut husk with other adsorbents listed in Table 1, might be due to the variation in the original nature of the precursors, the processes used to produce the adsorbents as well as other conditions applied during the adsorption processes.

CONCLUSION

Coconut shells are an environmentally friendly potential biosorbent for heavy metals. This study examined the efficiency of this sorbent in removal of Pb (II) ions from aqueous solution. Biosorption is affected by various factors, such as biomass concentration, pH and temperature. This study demonstrated that under optimum conditions (pH=6.0, biomass concentration $1g100L^{-1}$; temperature=25 °C, and contact time=120 min), maximum biosorption capacities of 65 mgg^{-1} was obtained in the Langmuir model for Pb (II) ions. The experimental values are evaluated according to the Langmuir and Freundlich isotherms. Both models fit the experimental data but the Freundlich is more suitable. Comparison between this paper and other biosorbents show that coconut husk biomass was an efficient biosorbent for this metal ion. Because carbon is easily prepared from the agricultural by product such as coconut shells it would be useful for the economic treatment of polluted water containing heavy metals.

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