



Removal of lead by activated carbon and citrus coal from drinking water

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ABSTRACT

Heavy metals are one of the most important environmental pollutants. Among heavy metals lead (Pb) which is toxic, well recognized as an environmental pollutant. The main objective of this study was to examine the adsorption efficiency of granular activated carbon for lead (II) removal from aqueous solution compared with citrus charcoal. The removal efficiency was controlled by solution pH, initial ion concentration, height and number of column and its arrangement. The results showed that with increasing the initial concentration of lead the removal efficiency was decreased. The removal rate for citrus coal was more than the granular activated carbon. The optimum time of 120 min was considered. The maximum removal of lead for GAC and citrus coal was observed at pH = 6.5. The maximum removal of lead was observed in the citrus coal/ GAC (98.5%). The efficiency of citrus coal for Pb adsorption was higher than the GAC. Citrus Charcoal which considered as a low cost material is mainly produced from waste of citrus. Drinking water standards were obtained using Citrus coal/GAC column in series.

Keywords: Heavy metals; Lead; Granular activated carbon; Citrus coal

1. Introduction

Heavy metals are one of the most important environmental pollutants. Rapid population growth and industrialization has led to introduction of heavy metals in to the environment [1]. Presences of heavy metals in environment (water, soil and food chain) are harmful to human health [2]. Heavy metals are non-biodegradable and therefore accumulated in the environment, causing various diseases and disorders [3]. Detrimental effects of heavy metals are also established on all living species [4].

Among heavy metals lead (Pb) which is toxic, well recognized as an environmental pollutant [5]. Industrial activities, such as mining and metal processing, can lead to heavy metal contamination in soil and water sources (surface and

groundwater), causing toxic effect upon entering the food chain eventually threaten human health [6]. The major environmental sources of Pb and its compounds are paints, battery industries, lead smelters and lead pipes [5]. Effluents containing Lead (II) discharged into water supplies sources owing to the expansion of industries [7]. The maximum concentration level (MCL) for lead (II) in drinking water is lower than 15 ppb (0.015 mg/L) that has been mandated by environmental protection agency [7]. The strict limitation on discharging of lead to the natural water sources are potential health effects of lead on children and adults [7]. Therefore, lead content in water, industrial wastewater and contaminated environment must be reduced to a minimum ppb level [4]. Golestan province is located in the Caspian Sea. Agriculture is widely practiced in this area due to sufficient precipitation over the catchment. Extra application of chemical fertilizers has resulted in soil and water contami-

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nation with heavy metals. Many local studies have reported lead concentration up to 0.2 and 3 mg/L in water and sediments, respectively.

Treatment processes for heavy metals in contaminated environments include membrane filtration, chemical precipitation, ion exchange, carbon adsorption and co-precipitation/adsorption [3]. In developed countries, removal of heavy metals from contaminated environments is achieved by advanced processes such as ion exchange, ultrafiltration or electrochemical deposition. However, these processes are not cost effective and may not be suitable for developing countries [8]. Cost effective alternative technologies or sorbent are needed for treatment of metal contaminated environments [3]. In this regard, adsorption has become one of the appropriate treatments [9]. The sorbent may be of mineral, organic or biologic origin, zeolites, biomass, industrial by product and polymeric materials [9]. The main disadvantages of industrial sorbent are their high cost [10]. Cost is an important parameter determined for selecting the sorbent materials. Natural sorbents are superior than their industrial types due to lower costs [3]. In comparison with other technologies, sorption processes have some advantages, especially in water treatment, such as quite simple design and operation, flexibility and finally free by products effluents which are suitable for reuse purposes [4]. In the recent years, there has been great interest in the removal of toxic heavy metals by adsorption [11]. Several studies have examined heavy metals removal from aqueous solution by various sorption including zeolites [12], red mud [13], chitosan [14], waste biosorbent [15], carbon nanotubes [16], graphene oxide nanocomposite, granular bentonite [7], coal combustion ash and activated carbon [17] and TiO₂ nanoparticle [18].

Due to the flexible and cost advantages over other sorbents, activated carbon is widely employed for water and wastewater treatment [11]. Citrus trees are well grown in Iran's climate and are considered native species in the northern region. From the waste of citrus tree can be produced charcoal (lemon coal or citrus coal) with low cost. In this study adsorption efficiency of industrial activated carbon was compared with citrus charcoal for lead (II) removal from aqueous solution.

The objectives of this study are:

- Compare lead adsorption by granular activated carbon (GAC) and citrus charcoal
- Study the effects of initial pH of the solution, initial concentration of lead, contact time and the column height on the adsorption of lead
- Investigate the effects of the columns number and resize mesh of activated carbon on removal efficiencies

2. Materials and methods

This study was experimental that the two adsorbents were studied for the removal of lead (II) from aqueous solution. A downward flow and fixed bed reactor was used (Fig. 1). The influent and effluent tank has a volume of 4 L, height and diameter of column was 50 and 6 cm respectively. Column volume was equal to 1.13 L. Hydraulic retention times (HRT) were 30, 60, 90, 120 and 180 min. The flow rate according the HRT was equal to 2.26, 1.13,

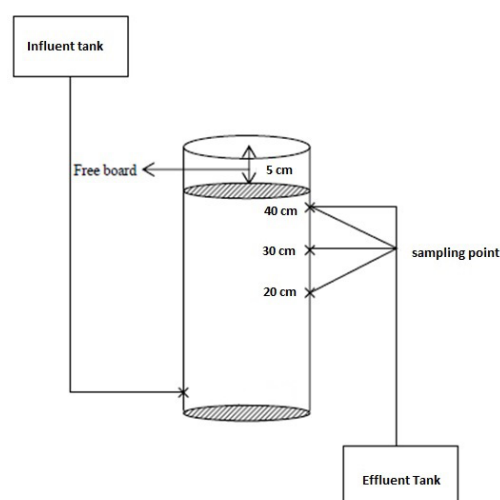


Fig. 1. The schematic of reactor.

0.75, 0.56 and 0.37 L/s respectively. Fig. 1 shows the schematic of reactor.

2.1. Adsorbent preparation

Adsorbents used in this study are including granular activated carbon (GAC) and citrus coal (lemon charcoal). Granular activated carbon (GAC) used in this study with mesh 0.2–0.5, 1–3 and 1–5 mm and surface area equal to 500–1500 m²/g that was provided in the industrial production from Norit Netherlands BV Company. For the preparation of citrus coal initially wood of citrus to form of compressed put in a sealed aluminum container, so no oxygen cannot enter, followed by the container was put into the electric furnace for 2 h to at 700°C. After this time the aluminum container was removed from the furnace. After cooling, the carbon obtained after crushing with a porcelain mortar through the mesh sieves 50 and 12 mesh was sieve and preparations citrus coal in the range of 0.5–0.2 mm.

2.2. Preparation of lead solution and detection

For preparation of standard solution of lead, use the 1000 ppm of stock solution that was made in Spain (Scharlau Brand). The lead-free water (distilled water) was used to prepare the solution. To provide the desired concentration of stock solution by dilution was carried out. The remaining amount of lead detected by atomic absorption spectrometry Varian 240 model.

2.3. Studied variables

Input lead concentration in reactor for both adsorbents was 1.5, 3, 5 and 8 mg/L. Initial pH was including 3.5, 4.5, 6.5 and 8.5 that was adjusted by a molar solution of NaOH and nitric acid. The contact time in the adsorption column was 30, 60, 90, 120 and 180 min. Samples were taken from the 20, 30 and 40 cm of heights of the adsorption column both adsorbent.

3. Results and discussion

In this study adsorption efficiency of granular activated carbon (GAC) and citrus charcoal for lead (II) removal from aqueous solution was conducted. The studied variables were including initial lead concentration, pH, contact time and heights of the adsorption column.

3.1. Effect of initial lead concentration

The effects of initial concentrations of lead (II) for both adsorbents are presented in Fig. 2. Maximum removal of lead was observed in the 1.5 mg/L for both adsorbents. With increasing the initial concentration of lead the removal efficiency was decreased. Therefore concentration of 1.5 mg/L was considered as the optimum. Removal rate for citrus coal was more than the granular activated carbon.

Increase in ions concentration decreased removal percentage. This might be due to the fact that escalated competing ions react with surface active group which ultimately reduce surface active sites and their saturation [19].

3.2. Effect contact time

The effects of contact time for both adsorbent are presented in Fig. 3. Maximum removal of lead was observed at 180 min for both adsorbents. With increasing the contact time removal efficiency was increased. In most times the removal rate of citrus coal was more than the granular activated carbon. No difference was observed between 120 and 180 min. Therefore, 120 min was selected as the optimum contact time.

3.3. Effect of pH

The effects of various pH for both adsorbent are presented in Fig. 4. Maximum removal of lead was observed at pH = 6.5 for both sorbents. An increase in adsorption rate was obtained from pH 3.5 to 6.5; however, at higher pH levels adsorption rate reduced. Our results were in agreement with the data obtained by Fernandez-Nava et al. for Pb (II) adsorption onto bentonite [12]. In adsorption processes, pH of the aqueous solution is an important operational parameter because it affects the solubility of metal ions [1]. In another study conducted by Momcilovic et al. in a study for removal of lead (II) from aqueous solution reported the optimum pH lower than 6.7 was obtained for lead adsorption by pine cone activated carbon [20].

From Fig. 4, it is shown that increase in pH solution from 3.5 to 6.5 linearly escalated Pb removal efficiency. This may be due to the formation of soluble hydroxyl complexes [1]. Removal efficiency declined above pH 6.5. At pH > 6.5, the presence of oxygen containing functional groups conferred negative charge on the adsorbent surface and resulted repulsive interaction between the metal ions and adsorbent [1].

Lower metal adsorption in acidic pH can be explained by the prevailing of H_3O^+ ions that occupy the majority of the binding sites [21].

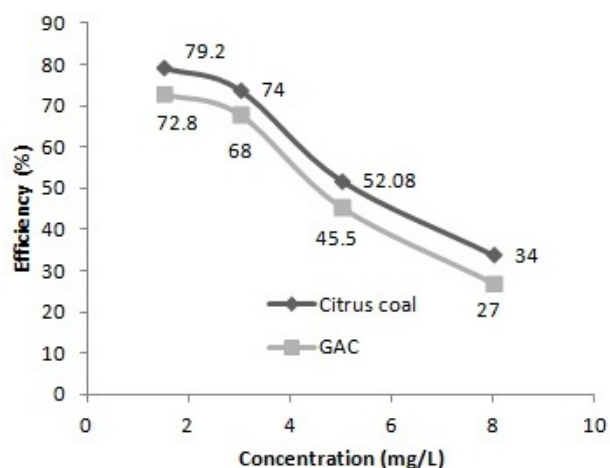


Fig. 2. The effect of initial concentration of lead.

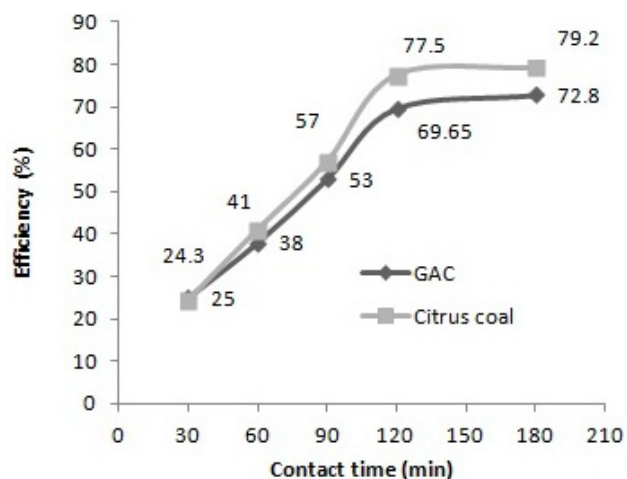


Fig. 3. The effect of contact time (pH = 6.5, $C_0 = 1.5$ mg/L).

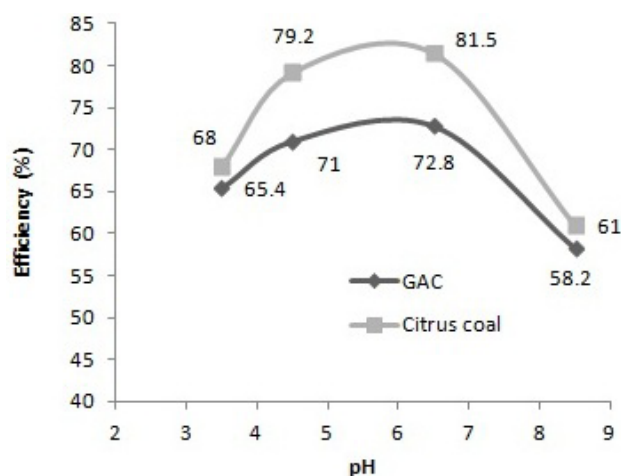


Fig. 4. The effect of pH (contact time = 120 min, $C_0 = 1.5$ mg/L).

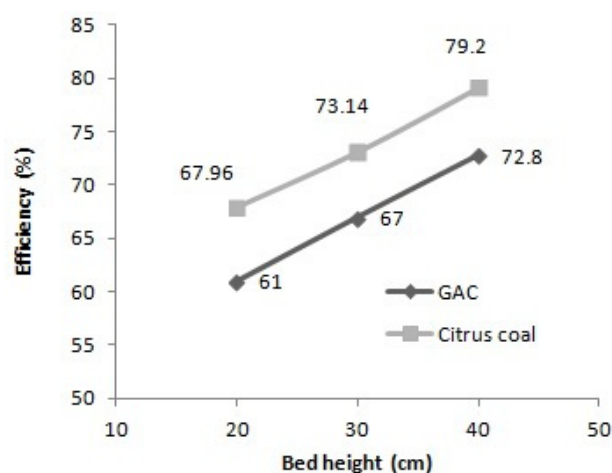


Fig. 5. The effect of column height (pH = 6.5, contact time = 120 min, $C_0 = 1.5$ mg/L).

Table 1

The effect of number of column and arrangement (pH = 6.5, contact time = 120 min)

Absorbent	C_0 (mg/L)	C_e (mg/L)	Efficiency (%)
GAC	1.5	0.408	72.8
Citrus coal	1.5	0.312	79.2
GAC + Citrus coal	1.5	0.110	92.6
Citrus coal + GAC	1.5	0.022	98.5

3.4. Effect of bed height (adsorbent mass)

The effect of bed height for both adsorbent is presented in Fig. 5. Maximum removal of lead was observed in the 40 cm for both adsorbent. Increasing bed height enhanced removal efficiency. Removal rate for citrus coal was more than the granular activated carbon.

Maximum effluent concentration declined with increase in bed height and a bed height of 40 cm was sufficient to achieve maximum adsorption efficiency. Our results are in agreement with the data obtained by Mondal [22]. In another research conducted by Goel et al. increase of bed height (adsorbent mass) enhanced lead removal efficiency of treated granular activated carbon [23].

3.5. Effect of numbers of column and arrangement

The effect of number of column and arrangement is presented in Table 1. The adsorption column solely presented low removal efficiency. When the columns were used in series removal efficiency was higher. Maximum removal of lead was observed for the citrus coal/GAC (98.5%).

4. Conclusions

The granular activated carbon and citrus coal adsorbent exhibited effectiveness in removal of Pb from aqueous solution. Removal efficiency was controlled by solution pH,

initial ion concentration, column height, numbers of column and arrangement. The efficiency of citrus coal for Pb adsorption was higher than the GAC. Citrus charcoal which considered as a low cost material is mainly produced from waste of citrus. Maximum effluent concentration declined with increase in bed height. Bed height of 40 cm was sufficient to achieve maximum adsorption efficiency. When the columns were used in series removal efficiency was higher. The efficiency of citrus coal for Pb adsorption was higher than the GAC. Maximum removal of lead was observed in the citrus coal/GAC. Drinking water standards were obtained using Citrus coal/GAC column in series.

Competing Interests

The authors declare that they have no competing interests.

Authors' Contributions

Mahdi Sadeghi proposed the study design and hypothesis and conducted data collection. Data collection were done by Akram Golbini-Mofrad and Somayeh Beirami. Data analysis and manuscript preparation were done by Mahdi Sadeghi and Ali Zafarzadeh. All authors read and approved the final manuscript.

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