



Chicken bone ash as an efficient metal biosorbent for cadmium, lead, nickel, and zinc from aqueous solutions

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ABSTRACT

Chicken bone ash (CBA) is the combustion residues of chicken bone and it has been investigated as a metal biosorbent for cadmium, lead, nickel, and zinc from aqueous solutions. The effect of various factors as contact time, amount of CBA, solution pH and initial concentration on adsorption process such of metal ions was studied at room temperature to optimize the conditions for maximum adsorption. Also, successful application in removing these metal ions was achieved by using real wastewater samples. This study indicates that CBA has the potential to become an effective and economical adsorbent for the removal of these metal ions.

Keywords: Chicken bone ash; Biosorbent; Heavy metals adsorption

1. Introduction

There are some metals that concern us because of occupational or residential exposure; some of them are known as heavy elements or heavy metals. Heavy metals could be very harmful to humans and aquatic life [1,2]. Although some of these heavy metals are reported to be essential trace ions, enhanced intake causes several undesirable effects in humans. Furthermore, one of the major sources of pollution of water bodies is the high degree of industrialization and urbanization. The most common toxic metals found in industrial wastewater are Cr, Ni, Mn, Hg, Cd, Cu, Zn, and Pb, which are nonbiodegradable and tend to accumulate in living organisms [3,4]. Therefore, removal of these contaminants has received much

attention in recent years. Several techniques have been applied for this purpose such as ion-exchange [5], chemical precipitation [6], reverse osmosis [7], coagulation and flocculation [8], membrane separation [9], and biosorption [10–14].

Between all of these methods, recently, new technologies involving the removal of toxic metals from wastewaters have directed attention to biosorption, based on metal-binding capacities of various biomaterials [15]. The main reason to give much attention to this method is the major advantages of biosorption over conventional treatment methods such as low cost, high efficiency, minimization of chemical and/or biological sludge, no additional nutrient requirement, regeneration of the biosorbent, and the possibility of metal recovery. Biosorption can be defined as the ability of inactive/dead biomaterials to accumulate heavy metals from wastewater through metabolically

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mediated or physicochemical pathways of uptake. The biosorption process involves a solid phase (sorbent or biosorbent, biomaterial) and a liquid phase (solvent, normally water), containing a dissolved species to be sorbed (sorbate, metal ions) [16]. Previous results published in the literature concerning the adsorption of various heavy metals by several adsorbents are summarized in Table 1.

Bone ash is derived from the carbonization of the animal bones by heating them at temperatures higher than 500°C. The crushed bone, after the heating process, will form the bone charcoal which is composed of calcium hydroxylapatite (70–76 wt.%), carbon content (9–11wt.%), calcium carbonate (7–9 wt.%), etc. Bone ash has been used extensively as an adsorbent for the decolorization of cane sugar [35]. Hydroxylapatite (HA) is known as a biomaterial, which constitutes the main percent of inorganic bone component.

Even in past years, [20, 36–38] used synthetic carbonate hydroxylapatite (CHA) as an efficient adsorbent with higher capability to remove heavy metals, in comparison with HA; but, the details about its adsorption characteristics on heavy metals is deficient, which might be due to the high cost of the preparation of CHA.

Adsorption of Cd(II) and Cu(II) by CHA derived from eggshell waste [20], As and Co by bone char [39,40], Pb, fumaric and maleic acids by synthesized HA [41,42] has been done in the recent years.

In this study, we have evaluated the capability of chicken bone ash (CBA) to remove Cd(II), Ni(II), Pb(II), and Zn(II) from aqueous solution. We also investigated the efficiency of the adsorbent to sorption of heavy metals by taking samples from Zayanderood River and Esfahan steel industry, to estimate the practical application of the adsorbent (Table 2).

Table 1
Comparison of the adsorption capacity (q_e) of various heavy metals on several adsorbents

Entry	pH	Adsorbent	Metal ion	Adsorption capacity q_e (mg/g)	Reference
1	–	Modified corn cob	Zn(II)	79.21	[17]
2	6.0	Cattle manure vermicompost	Zn(II)	20.30	[18]
3	6.0	Acid-treated coconut shell carbon	Zn(II)	60.41	[19]
4	5.0	Carbonate hydroxyapatite	Cd(II)	111.1	[20]
5	6.0	Carbon aerogel	Cd(II)	400.80	[21]
6	7.0	Green coconut shell powder	Cd(II)	285.70	[22]
7	5.0	<i>Streptomyces lunalinharesii</i>	Cd(II)	24.8	[23]
8	6.0	<i>Acidiphilium strains</i>	Cd(II)	248.62	[24]
9	5.0	Formaldehyde treated acron waste	Pb(II)	164.2	[25]
10	5.0	HAp/Fe ₃ O ₄	Pb(II)	598.8	[26]
11	5.0	Barbula lambarenensis	Pb(II)	62.50	[27]
12	5.5	Ethylenediamine-modified yeast biomass coated with magnetic chitosan microparticles	Pb(II)	121.26	[28]
13	4.0	Tea waste	Ni(II)	15.26	[29]
14	7.0	Natural iron-oxide coated sand	Ni(II)	1.26	[30]
15	5.0	Alyssum discolor biomass	Ni(II)	13.1	[31]
		Acid treated biomass		34.7	
16	–	Spirogyra hyalina	Cd	18.181	[32]
			Pb	31.250	
17		Husk of black gram	Pb	49.97	[33]
			Cd	39.99	
			Zn	33.81	
			Ni	19.56	
18	5.5	Natural lignite	Pb(II)	39	[34]
			Zn(II)	15	
			Cd(II)	5	
19	5.6	Chicken Bone Ash (CBA)	Cd	1,924	This work*
			Ni	1,854	
			Zn	1,627	
			Pb	1,842	

*Adsorption capacity for all of the toxic metals was extracted from Langmuir equation.

Table 2
Isotherm constants for Cd(II), Pb(II), Ni(II) and Zn(II) sorption by CBA with initial concentration ranged from 0.5 to 30 mg/dm³

Metals	Freundlich isotherm constants		
	k_F	$1/n$	R^2
Cd(II)	1.002	0.9985	0.99998
Ni(II)	0.9995	0.9996	0.999998
Zn(II)	1.000	0.9995	1
Pb(II)	Langmuir isotherm constants		
	q_{\max}	b	R^2
Pb(II)	1,627	0.0006221	0.999964

2. Experimental section

2.1. Adsorbate and analytical measurements

All the chemicals and reagents used in this study were of analar grade (AR grade) and were used as supplied, without further purification. Stock solutions of Cd(II), Ni(II), Pb(II), and Zn(II) ions were prepared by dissolving Cd(NO₃)₂·4H₂O, NiCl₂·6H₂O, Pb(NO₃)₂, and ZnSO₄·7H₂O separately in 1,000 cm³ of distilled water. Then, each of the stock solution was used to prepare metal ion concentrations ranging between 0.5 and 30 mg/dm³. The pH of the solution was adjusted by adding dilute HCl. A digital pH/mV meter (Metrohm model 710) was used for pH measurements. Atomic absorption spectroscopy (Perkin-Elmer 2380) was used to determine the concentration of the unadsorbed Cd(II), Ni(II), Pb(II), and Zn(II) ions in the solution.

2.2. Adsorbent and characterization

Chicken bone was sliced into small fragments and was burnt until the meat attached on the bone was removed. After that the bone was kept in a furnace for 24 h in air at 600 °C. The bone ash was powdered and sieved through a 40-mesh Sieve.

X-ray diffraction (XRD) was measured on the X-Ray Diffractometer (Bruker D8 ADVANCE with Ni filtered Cu-K α radiation) to study the crystalline phase structures of the CBA. The specific surface area of CBA was determined using N₂ adsorption/desorption method at 77 K by standard BET method using Quantasorb (Quantachrome, USA). The Fourier transform infrared spectroscopy (FT-IR) analysis was performed using a Fourier transform infrared spectrometer (Jasco FT/IR-680 plus spectrophotometer). The powders were blended with IR-grade KBr in an agate mortar and pressed into tablets.

2.3. Adsorption experiment procedure

About 10 cm³ of the adsorbate solutions of several concentrations, ranging between 0.5 and 30 mg/dm³, were placed in flasks. Then CBA was added to each flask. The flasks were sealed with stopper and shaken several times at 200 rpm at 25 °C. After shaking each of the solution at a given time, it was immediately centrifuged. The metal ion concentrations in the solutions were determined by AAS. The amount of metal ions adsorbed at equilibrium was q_e , which represents the metal ion uptake, and was calculated according to Eq. (1):

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

where q_e is the adsorption amount (mmol/g), C_0 and C_e are the initial and final concentrations, respectively, of metal ions in aqueous solution (mmol/dm³), V is the volume of solution (dm³), and m is the weight of the adsorbent (g).

The sorption percentage [removal (%)] of metal ions from aqueous solution is computed as follows in Eq. (2):

$$\text{Removal (\%)} = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (2)$$

2.4. Real sample preparation

Real samples were taken from Zayanderood River and a factory located 65 km southwest of Esfahan. The samples were filtered through a 0.45 μ m filter paper and transferred to a 100 cm³ polyethylene bottle.

3. Results and discussion

3.1. Characterization of the adsorbent

XRD pattern of the CBA revealed that it was well crystallized (see Fig. 1). It also showed that HA was the main inorganic component in CBA. The specific surface area of CBA, determined by the BET method, was 40 m²/g.

The FT-IR spectrum of CBA showed peaks at 961–1,041 cm⁻¹ and 569–606 cm⁻¹ representing the characteristic, well-crystallized apatite phase (see Fig. 2). The peak at 961 cm⁻¹ corresponds to ν_1 stretching mode and 1,041 cm⁻¹ represents ν_3 vibration mode of phosphate groups. The sharp peaks at 569–606 cm⁻¹ are assigned to the bending mode of phosphate. The absorption band at 1,457 cm⁻¹ is due to the vibration

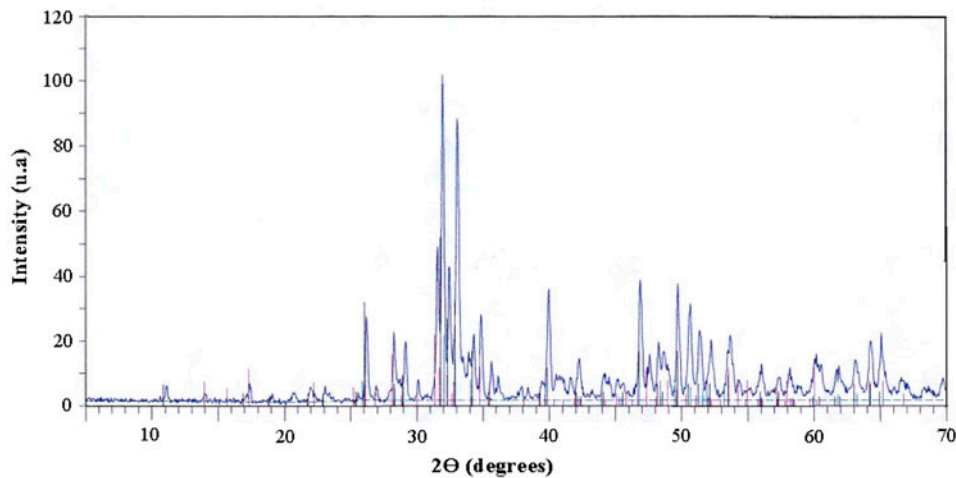


Fig. 1. XRD patterns of CBA.

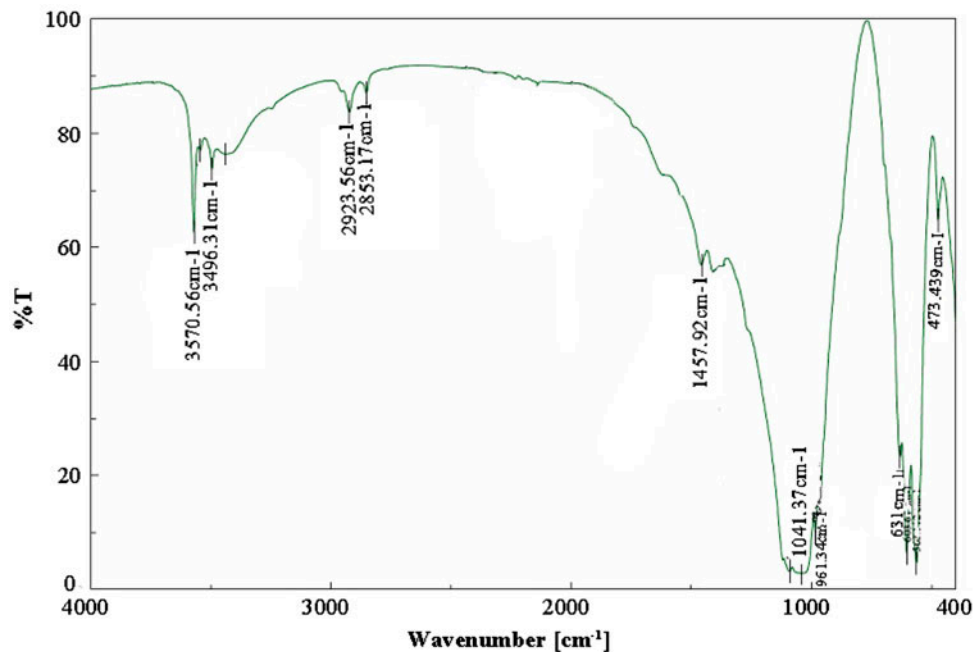


Fig. 2. FT-IR spectrum of CBA.

mode of carbonate groups. The adsorption around $3,570\text{ cm}^{-1}$ was OH associated by hydrogen bond stretching, which is the typical band for HA.

3.2. Effect of pH

pH is one of the most important parameters in controlling the uptake of heavy metals from wastewater and aqueous solutions. Most of the industrial wastewaters are acidic [43,44] and the basic conditions could promote hydroxide precipitation, this introduced uncertainty into the adsorption investigation [45,46].

In order to consider the effect of pH on the percentage of adsorption with the given amounts of each metal ions, the acidic pH was chosen. After a contact time of 24 h, it was observed that removal capacity was 100% at three pH values (1.5, 3, and 5.6).

3.3. Effect of adsorbent dose

An increase in the adsorbent concentration resulted in an increase in metal ions' uptake by CBA. It was found that, when the initial metal concentration was 30 mg/dm^3 , the maximum uptake took place at

0.05 g of adsorbent for all metal solutions. In the case of Pb(II) solution, the complete adsorption took place even at 0.005 g of the adsorbent. However, under this situation, the percentage removal of Cd(II), Zn(II), and Ni(II) was 75.45, 74.34, and 23.28%.

3.4. Effect of contact time

The maximum metal removal capacity was attained after about 5 min of contact for lead and nickel and for cadmium, it happened after 20 min of contact. Not only the contact time was longer for zinc solution, but also the maximum adsorption was smaller than the other metals even after 24 h.

3.5. Adsorption isotherms

Adsorption isotherms describe how adsorbates interact with adsorbents. The equilibrium sorption isotherm is important in the design of sorption systems. Equilibrium relationships between the adsorbent and the adsorbate are described by sorption isotherms, usually by the ratio between the quantity adsorbed, and that remaining in the solution at a fixed temperature at equilibrium. In most cases, the equilibrium of the process is described by Langmuir and Freundlich isotherm models. The Langmuir isotherm model is a theoretical model for monolayer adsorption that was calculated according to Eq. (3):

$$q = \frac{q_{\max} b C_e}{(1 + b C_e)} \quad (3)$$

where q is the amount of metal adsorbed, mg/g; q_{\max} is the maximum metal uptake value corresponding to sites saturation, mg/g; C_e is the equilibrium metal concentration in solution, mg/dm³; and b is the ratio of adsorption/desorption rates.

The Freundlich isotherm model is an experimental model and it is usually expressed as follows in Eq. (4):

$$q = k_F \times C_e^{1/n} \quad (4)$$

where k_F and n are constants related to sorption capacity and sorption intensity, other symbols are as previously described.

In the case of the Freundlich model, the energetic distribution of sites is heterogeneous, due to the diversity of sorption sites or the diverse nature of the metal ions adsorbed, free or hydrolyzed species. The Langmuir model supports a monolayer sorption with a homogenous distribution of sorption sites and

sorption energies, without interactions between the adsorbed molecules or ions.

The surface adsorbent properties determine the sorption mechanisms. The most commonly reported mechanisms for metal ion sorption are ion exchange, electrostatic interaction, chelation, precipitation, and complexation.

The plots of q vs. C_e for various initial concentrations were found to be linear (see Fig. 3), but according to the values of the Langmuir and Freundlich constants given in Table 1, the adsorption data for Cd(II), Ni(II), and Zn(II) removal fitted well with the Freundlich isotherm. The Freundlich constant $1/n$ is also a measure of the deviation from linearity of the adsorption, that means, if $1/n$ is equal to unity, the adsorption is linear. When $1/n < 1$, it shows that the increased adsorption modifies the sorbent in a manner that increases the sorption capacity, such as forming new sites. If $1/n > 1$ or, becomes too large, ($1/n \gg 1$), the adsorption bond will be weak, and the value of q will be vary drastically with small changes in C_e . It can be said that as $1/n$ value of CBA is less than 1, chemical rather than physical adsorption seems to prevail [47].

Based on the correlation coefficients (R^2) obtained (Table 1), the adsorption of Pb(II) conformed to Langmuir isotherm. Small b values indicate that Pb(II) ions were attached strongly to CBA. As it was noted briefly before, in the Langmuir model, adsorption occurs uniformly on active sites of the adsorbent and once an adsorbate occupies a site, no further adsorption can take place at that site.

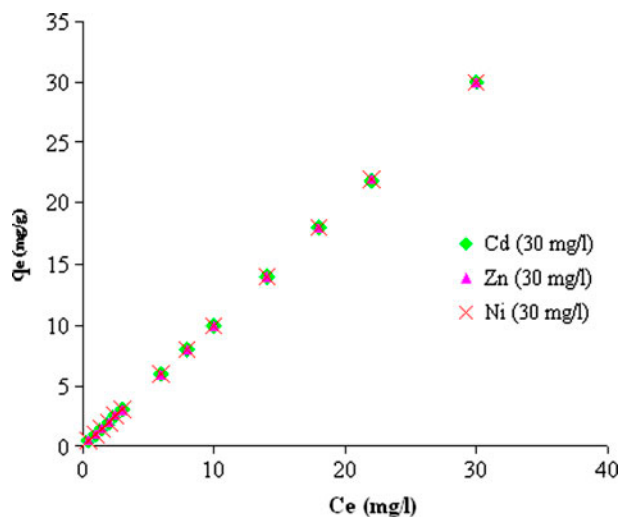


Fig. 3. Equilibrium data of heavy metal adsorption on CBA.

3.6. Application of CBA for removal of Cd(II), Pb(II), Ni(II) and Zn(II) from wastewater

Zayanderood is the only permanent river in central Iran and the main feeding source for regional aquifers that plays a key role in providing Esfahan province with drinking water. Because of the importance of this river in terms of its various uses and the fact that it receives waste materials from heavy metal industries, such as Esfahan steel company, Mobarake steel complex, Polyacryl and other chemical factories, power plants, oil refinery, and military industries, the wastewater sample was used to assess the removal efficiency of Cd(II), Pb(II), Zn(II), and Ni(II) by CBA from this river (sample 1) and from one of the noted factory (sample 2). The removal efficiency reached 100% for Cd(II), Pb(II), and Zn(II) in both samples and Ni for only sample 1. Compared with the results obtained using deionized water, not only the removal efficiency of metal ions from wastewater did not decline but also improved to 100% that means the existence of organic materials in wastewater, had no effect on adsorbing of the metals. Explanation about this increase can be related to pH of the samples. Both samples have the pH range of 7–8, on the other hand, the results obtained using deionized water were carried out in the acidic pH.

4. Conclusions

CBA has been proven to be very successful in removing heavy metals from real wastewater. The low cost, the high adsorption capacity, and the successful application in removing Cd(II), Pb(II), Ni(II), and Zn(II) from wastewater demonstrate that it has a great potential to be an efficient biosorbent.

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