REMOVAL OF Fe^{+3} AND Pb^{+2} IONS FROM AQUEOUS SOLUTIONS BY ADSORPTION USING BANANA PEELS

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ABSTRACT: The present study was carried out evaluate the possibility of using banana peels for the removal of Fe^{+3} and Pb^{+2} from aqueous solutions. This paper incorporates the effects of dose, concentration and pH. Adsorption of heavy metals on adsorbents was found to increase on decreasing initial concentration, the sorption capacity strongly increased with pH in the range 2-8. The results showed that the removal of heavy metals, such as Fe^{+3} and Pb^{+2} from aqueous solution was efficient using banana peels as bioadsorbent. The adsorption percentage of Fe^{+3} and Pb^{+2} ions by banana peels were very high. The Langmuir isotherm model was the best to describe the experimental data. The maximum sorption capacity was found to be 79.28 and 96.58 mg/g for Fe^{+3} and Pb^{+2}, respectively. Over all, the present findings suggest that banana peels are friendly environmental bioadsorbent, efficient and low cost biosorbent which represents an excellent potential for Fe^{+3} and Pb^{+2} removals from aqueous solutions.

Key words: Banana peels, Biosorption, heavy metals, Industrial wastewater, IR, SEM.

INTRODUCTION

Human health, social and economic progress, and the success of the global ecosystem depend on a reliable supply of high-quality water. Considering that water is a basic requirement of all living beings, rapid urbanization and development have substantially increased the demand for water. In addition, water is used in a variety of residential and industrial processes, including petroleum refineries as well as agricultural, medicinal, and pharmaceutical procedures. These activities introduce many toxic pollutants and waste substances into water (**Anser et al., 2020**).

Heavy metals (such as zinc, cadmium, chromium, lead, copper, nickel, platinum, vanadium, titanium, and silver) are present in substantial concentrations in wastewaters generated by electroplating, metal surface treatment, and printed circuit board (PCB) fabrication operations (**Barakat, 2011**). Metal finishing waste is often a slurry (sludge) containing metal hydroxides, such as magnesium, ferric, zinc, and nickel hydroxides.

Water is used in the iron and steel industry for cooling and separation of by-products. Water is also utilized in the electric arc furnaces for slag handling, impurity removal, and air pollution control as well as scale breaking in hot-rolling operations. As a result, the wastewater from the steel manufacturing process is highly contaminated, carrying a wide range of heavy metal ions (**Beh et al., 2012**).

Iron is one of the major constituents of the lithosphere and comprises approximately 5% of it. It is routinely detected in municipal waste effluent, particularly in cities where iron and steel are manufactured. Iron readily complexes with sulphates in the sediments of many surface levels of water. The primary concern about the presence of iron in drinking water is its

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objectionable taste. The taste of iron in drinking water can be easily detected even at low concentrations of about 1.8 mg/l (**Sadon *et al.,* 2012**).

The scope of research is so large that recently subject reviews are regularly published on using a particular crop processing waste. Thus, in particular, information on the use of buckwheat waste (**Shaikhiev *et al.,* 2020**) wheat waste (**Farooq *et al.,* 2010**), banana peel (**Akpomie *et al.,* 2020**), tea waste (**Hussain *et al.,* 2018**), coffee waste (**Anastopoulos *et al.,* 2017**), fruit peels (**Bhatnagar *et al.,* 2015**) and waste (**Othman *et al.,* 2013**) and others as sorption materials is summarized. The world literature provides information about using onion by-products as sorption materials for various pollutants, but this information is sketchy.

Plant waste materials which are presented in large quantities from many processes and almost costless, may be successful alternative sorbent material to be used as effective adsorbents (**Deans and Dixon, 1992**) such as fruit peels, tree bark, peanut skin and growing plants (tobacco and tomato root tissues) were used to remove different metals ions from polluted water (**Cobbette, 2000**).

Several studies have shown that banana peels could quickly remove heavy metals from river water and is at least as effective, and in some cases even better than existing methods (**Rodriques *et al.,* 2006**). Evidently, banana peels are rich in negative charged molecules, so they attract the heavy and positively charged metal pollutants in water, and they are quite effective, also, for every treatment with the peel powder, around 65 percent of the water was decontaminated (**Cobbette, 2000**).

Banana peel, an agro wastes is discarded all over the world as useless material. It is causing byproduct management problems though it has some compost, cosmetics and sorbent potentiality. It is an abandoned, readily available, low cost and cheap, environment friendly bio-material. Considering the above criteria, banana peel was selected to prepare the biosorbent. A step was taken for preparing biosorbent and used for removal of lead from water (**Hossain *et al.,* 2012**). In this study, banana peel was used for removal of ferric and lead ions from aqueous

solutions by the batch method. Effective parameters such as initial pH of the metal solutions, initial metal concentrations and banana peel dosage were investigated. The adsorbent-adsorbate equilibrium behavior has been investigated using Langmuir isotherms.

MATERIAL AND METHODS

Experimental and Preparation of Samples Collection and Milling

The banana was purchased to use peels plant waste was separated simply from banana 15k.g of it from local markets in the city of zagagiz the peel was washed with distilled water to remove adhering external dirt and then dried 3 days under the sun and the color of banana change was observed from yellow to brownish black. Dried peels were crushed and milling into powder by home mill then dried in a hot air oven at 110°C for 3h the moisture content was lost from powder and kept in an air tied bottle for experimental uses, All samples were stored in a dry place to use for biosorption tests experiments 1 kg of powder was obtained of dried crusts from 15 kg of bananas fruits.

Heavy Metals Standard Solutions

Heavy metals standard solutions were prepared depending on the atomic and molecular weight of the element (**Harley and Prescott, 1996**) and they were:

Ferric and Lead Standard Solutions

An adsorbate standard solution of 1000 mg/l of Ferric and Lead was prepared by dissolving 5.151 and 1.3422g and of $FeCl_3$-$7H_2O$ and Pb $(NO_3)_2$ respectively in 1000 ml of DDW.

Methods

Chemical Composition of Banana Peels

Moisture, crude fat, crude protein, ash, crude fiber, cellulose and hemicellulose contents were determined in banana peels, according to **AOAC (2005)**.

Adsorbent dosage

Effect of adsorbent dosage on the equilibrium adsorption of heavy metal ions were investigated with banana peels of 0.1, 0.25, 0.5 and 1g in for

sets of 100 ml water which contained 100 mg/l of heavy metal concentration each. The conical flasks were shaken for 24 hours with 150 rpm at room temperature. Then, samples were filtered. Fe^{+3} and Pb^{+2} concentrations in filterate were determined by Atomic Absorption Spectrometer (AAS).

Effect of pH

Effect of pH for tested heavy metals adsorption onto banana peels was investigated with 1g of banana peels in 100 ml water containing 100 mg/l of heavy metal ions and each sample was adjusted to pH between 2.0 to 8 using either 1N HCl or NaOH solution. The biomass was separated from the solution by filteration and the resulting solutions were analyzed for Fe^{+3} and Pb^{+2} by Atomic Absorption Spectrometer.

Effect of Initial Ion Concentration

Equilibrium adsorption experiments (triplicates) were conducted with 100 ml water containing 1 to 1000 mg/l of Fe^{+3} and Pb^{+2} concentrations. Banana peels of 1 g were added in each sets of experiments and shaken for 24 hours with 150 rpm at room temperature with pH=6.0.

Infrared analysis

The infrared (IR) spectrum of banana was determined using a Fourier transform infrared (FTIR) spectrophotometer (Spectrum One-B, Perkin Elmer, US) for detection of various functional groups. The purified gellan gum was ground with KBr powder and pressed into pellets for FTIR measurement in the frequency range of 4000–400 cm^{-1} (**Zhbankov** *et al.,* **1997**).

Desorption study

Desorption of banana was studied using 7 types of solvent medium including tap water, distilled water, 0.1N H_2SO_4, 0.1N HCl, 0.1N HNO_3, 0.1N NaOH and 0.1N CH_3COOH. For this purpose, 0.5 g of saturated (with heavy metals) banana was taken in 100 ml of above mentioned medium and shaken at 120 rpm for 24 hr. (**Pazos** *et al.,* **2013**).

Equilibrium study

Equilibrium adsorption experiments (triplicates) were conducted with 16 Erlenmeyer of 100 ml water containing 1 to 1000 mg/l of Fe^{+3} and Pb^{+2} concentrations. Banana peels of 0.1, 0.25 and

0.5 g were added in each sets of experiments and shaken for 24 hours with 150 rpm at room temperature without pH adjustment. Langmuir and Freudlich isotherm models were thus tested according to the method adopted by **Manikandan** *et al.* **(2016)**.

Analysis

The collected water samples from different experiments were filtered with filter paper (Whatmann1) and prepared for AAS analysis. Samples were analyzed in term of heavy metal ions by Atomic Absorption Spectrometer (Thermo scientific Ice 3000). The pH was measured by pH meter. The amount of metal ions adsorbed was determined by difference between the initial and final concentrations. The sorption efficiency (%) and amounts of adsorbed metal (q_e) by hulls were calculated using the following equations:

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

Where: C_0 and C_e (mg/l) are the liquid-phase concentrations of metal initially and at equilibrium, respectively. V is the volume of the solution (l), m is the mass of adsorbent (g) and qe (mg/g) is the amount of adsorbed metal at equilibrium (**Ossman** *et al.,* **2014**).

RESULTS AND DISCUSSION

Chemical Composition of Banana Peels

The chemical composition of banana peels is summarized in Table 1. Data showed that banana peels composition was, organic matter 92%, ash content 3.8%; crude protein 5.4%; crude fat 0.1%; lignin 36.1%; hemicellulose 5.6% and cellulose 44.8%. The obtained results are in accordance with those of **María** *et al.* **(2018). Basso** *et al.* **(2002)** who reported that there was a correlation between lignin content of several lignocelluloses and their ability to remove heavy metals from aqueous solutions, therefore the high content of cellulose and lignin observed in the tested banana peels favour biosorption of metal ions. Moreover, lignocellulosic materials are very porous, have a very high specific surface area and affinity for water (**Pehlivan** *et al.,* **2008**), which improve the performance of these materials as sorbents. Cellulosic surface becomes partially negatively charged when

immersed in water so that possesses columbic interaction with cationic species, which contributes to the high binding abilities of these materials, especially divalent metal cations (**Laszlo and Dintzis, 1994**). The oxygen of each carbonyl (present in fats, lignin, protein and pectin) and hydroxyl group (present in cellulose and lignin) are considered a strong Lewis base because of the presence of its vacant double electrons, which could bind to a metal cation forming a complex of coordination these were obtained by **Hanan and Ahmed (2013)**.

Effect of adsorbent dosage

The detailed results of the kinetic experiments with varying adsorbent concentration were presented in Fig. 1. It is clearly show that the amount of metal ions adsorbed varied with the banana peels concentration. The degree of adsorption increased with an increase in the amount of adsorbent in solution and equilibrium. The higher degree of adsorption can be related to the increase in adsorption sites with the increase in the adsorbent masses (**Benguella and Benaissa, 2002**). Fig. 1 shows the percentage removal of lead and ferric at a concentration of 100 mg/l. It is evident that the percentage removal of lead increases from 45% to 85% with the increasing adsorbent concentration. The best economical adsorbent concentration was chosen to be 1% (w/v). The observed trend can be attributed to the fact that, initially with an increase in adsorbent dosage, more number of active sites are available which coupled with higher surface area of the adsorbent facilitates the increase in the extent of adsorption. This means that almost all ions are bound to the adsorbent and equilibrium is reached between the ions bound to the adsorbent and those remaining unadsorbed in the solution in a faster time interval (**Gutha *et al.*, 2011; Dubey and Shiwan, 2012**). Another reason to reduced removal efficiency at the higher loading of the adsorbents is the overlapping of adsorption sites due to the possible aggregation of the adsorbent particles leading to a decrease in the total surface area. Similar trend was observed by **Boota *et al.* (2009)**.

Effect of contact time

Fig. 2 shows the variation in the percentage removal of heavy metals with contact time using 1 g/100 ml of treated banana peels adsorbent at 6.0 pH. It is observed that for Fe^{3+}, and Pb^{2+} ions, the percentage removal is nearly 94% at 1000 ppm even throughout the 120 min. contact times. It is observed that in all cases the percentage removal is comparatively lower for 20 min. contact time, with increasing removal efficiencies at higher contact time up to 120 min and then gradually decrease at 180 minutes. The maximum removal obtained at 120 minutes. It is evident from the results that the contact time required to attain equilibrium is dependent on the initial concentration of heavy metals. For the same concentration, the percentage removal of heavy metal increases with increase of contact time till equilibrium is attained. The optimal contact time to attain equilibrium with banana peels adsorbent is 120 minutes.

Effect of initial concentration

The effect of different initial concentrations of Fe^{+3} and Pb^{+2} on the removal efficiency of Fe^{+3} and Pb^{+2} is shown in Fig. 3. Results confirmed that the adsorption of Fe^{+3} and Pb^{+2} by banana peels was studied at various iron and lead concentrations in the extent from 50 mg/l to 1000 mg/l. Adsorption efficiencies decreased with the increasing of the iron and lead concentration at constant adsorbent amount 10 g/l. **Farai *et al.* (2014)** notified that the equilibrium curve shows that the overall percent removal of metal from solution decreases with an increase in initial concentration. This may be attributed to lack of sufficient surface area to accommodate much more metal available in the solution. This demonstrate that the adsorbed amount of both heavy metals by the banana peels dependent upon availability of binding sites for Fe^{+3} and Pb^{+2}. The removal efficiency of studied metals was the highest at 50 mg/l for Fe^{+3} and Pb^{+2}. Similar trend was confirmed by **Ali *et al.*, (2014) and Javadian *et al.* (2015)**. Many studies reported that increasing the initial Pb^{+2} concentration above 500 mg/l led to decline the percent adsorption of Pb^{+2} indicating saturation of all the binding sites on banana peels, surface beyond a particular concentration and establishment of equilibrium between adsorbate and adsorbent (**Malkoc, 2006; Ozturk *et al.*, 2004**). Also observed that extent of compound adsorbed increases with an increase in initial concentration but at the same time removal efficiency (expressed in percentage removal) decreases because of faster saturation of adsorbent surface. Thus, in order to treat water with high metal concentration, dilution of the solution or increase in the biosorbent dosage would be required (**Banerjee *et al.*, 2012**).
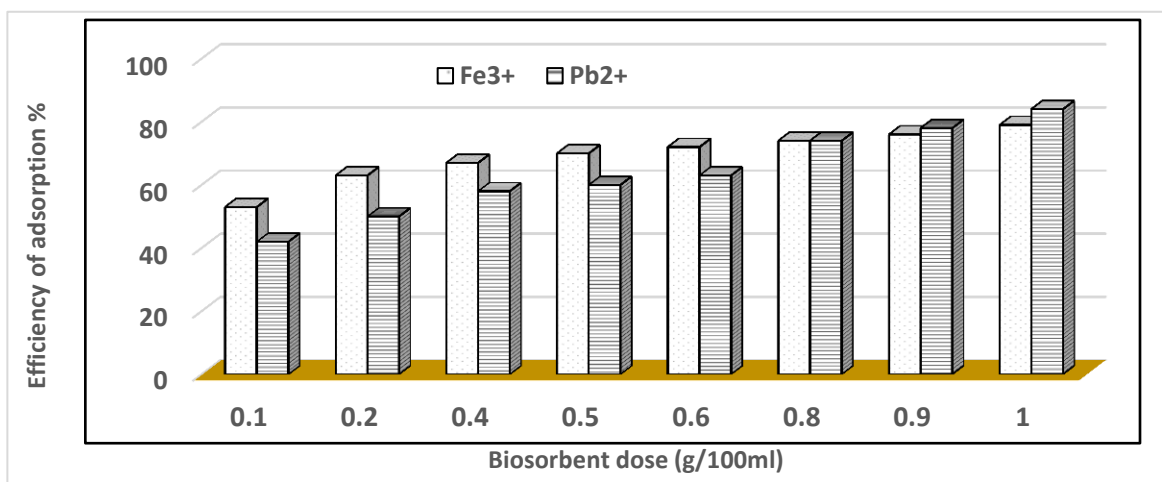
Fig. 1. Effects of adsorbent dosage on Fe^{+3} and Pb^{+2} (100 mg/l) adsorption on banana peels at 30°C, 150 rpm shaking, pH 6.0 and the contact time 1h
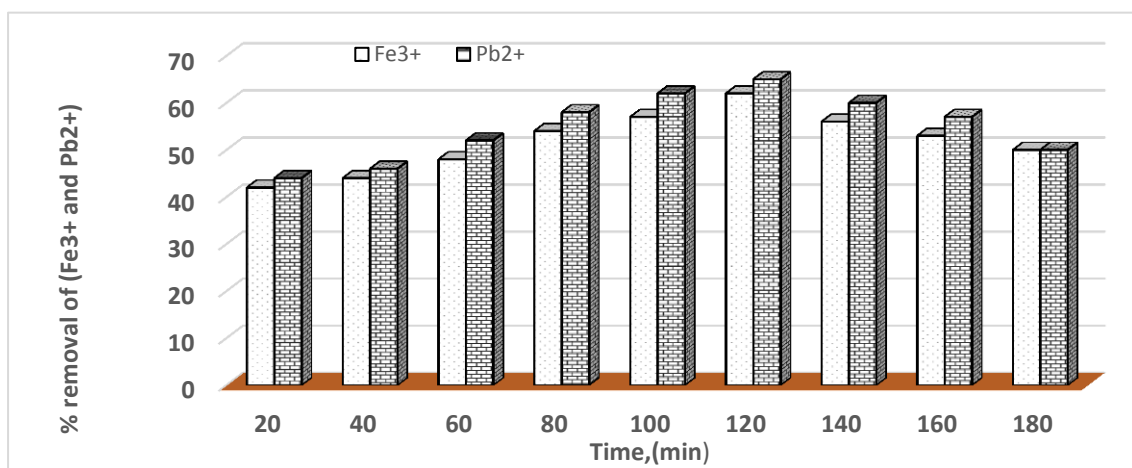

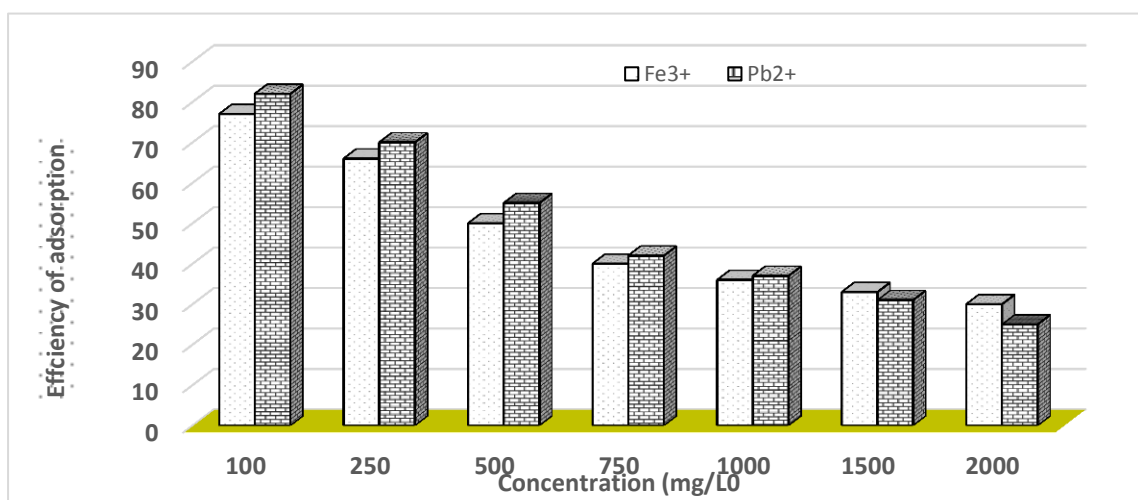**Fig. 2. Effect of contact time on % removal of lead and ferric ion by banana peels adsorbent**

Fig. 3. Effects of initial metal concentration on Fe^{+3} and Pb^{+2} adsorption on banana peels, (banana peels concentration was 1g/l, pH =6.0 Contact time was 1h.).

Effect of pH on Metals Adsorption

The effect of solution pH on metals adsorption was shown in Fig. 4. The biosorption capacity of the banana peels for the metals increased as pH increased, and then it decreased at high pH. The maximum uptakes were 216.09 mg/g for Pb^{2+} and 84.47 mg/g for Fe^{3+} at pH 5.0, 5.5 and 6.0, respectively. The solution pH, which affects the biosorption performance of biosorbent, is an important controlling parameter in the process (**Han *et al.*, 2006**). The effect of pH on the metal biosorption can be achieved through the competition between H_3O^+ ions and metal ions for the biosorptive sites. At low pH, little biosorption occurs, as the large amount of H_3O^+ exists in solution and the competition biosorption takes place (**Cruz *et al.*, 2004**). Moreover, the protonation state of ligands on the surface will endorse reaction with metal ions (**Krishnani *et al.*, 2008**). As pH increased, these negatively charged functional groups would be exposed in cations to accumulate in the double layer surrounding the polyelectrolyte (**Lamelas *et al.*, 2006**). Hence the biosorption on the gellan surface was increased, which was corresponding to the maximum biosorption capacity obtained at pH 6.0 for Pb^{2+} and 6.0 for Fe^{3+}, respectively. At high pH, the precipitation of metals occurs by the formation of metal hydroxides, so it is impossible for the biosorption (**Veglio and Beolchini, 1997**).

Effect of shaking speed

The effect of shaking speed on adsorption of lead and ferric were studied over the range of 30-200 rpm for 2h with 100 ml water containing 10 mg/l lead and ferric and 0.5 g of banana peel. Fig.5 indicates that the percent adsorption increased with an increased of shaking speed and obtained a maximum 88% and 90% adsorption at near 120 rpm. This shaking speed was employed for other experiment. At low and high speeds, the copper removal was lower than optimum. Low speed could not spread the particles properly in the water for providing active binding sites for adsorption of lead and ferric. It is resulted an accumulation of banana peel in the bottom of water and buried the active binding sites. On the other hand, the high speed vigorously spreading the particles of banana peel in the water and did not allow sufficient time to bind with lead and ferric ions.

Characterization of banana peels

Fourier transform infrared (FTIR) spectrometer was obtained to characterize surface functional groups of banana peels before and after adsorption process. Fig. 6 represent Fourier transform infrared spectrum of rice husk which compared with IR spectrum of heavy metals; Fe^{2+} and Pb^{2+} sorbed species. The bands absence and appearance of other bands were taken as a strong evidence for adsorption process for Fe^{2+} and Pb^{2+}.

The IR analysis of the banana peels was shown in Fig. 6. The peaks assignments of the banana peels were as follows: 3400–3200 cm^{-1} was related to the stretch vibration of O— H or hydrogen bond existing in all polymers. The signals at 3000– 2800 cm^{-1} were related to the stretch vibration of CH_2 bond, and the signals at 1850–1600 cm^{-1} were due to the stretch vibration of C=O bond. The signals at 1400–1370 cm^{-1} were attributed to the vibration of C—O bond and the signals at 900–1150 cm^{-1} for C—O—C bond (**Sun *et al.*, 2009**). There were shifts in wave numbers of peaks at 3399.72 cm^{-1}, 2936.10 cm^{-1}, 1640.02 cm^{-1}, 1418.17 cm^{-1} and 1063.64 cm^{-1} after loading Pb^{2+} and Fe^{3+} (Fig. 6).

The FTIR spectra revealed the presence of many functional groups such as hydroxyl, carboxyl and glycosidic bond on the banana peels surface. In comparing between the banana peels and the metal- banana peels, it can be observed that there were some shifts in wave number of dominant peaks associated with the metal-laden banana peels. These shifts in wave number were corresponding to the metal binding process taking place on the surface of biosorbent (Pavasant *et al.*, 2006). The O — H stretching group and the C =O stretching group were involved with metal biosorption. The wave numbers of the other groups such as CH_2 group, C— O —C group and C =O group were also changed.

Scanning Electronic Microscopy (SEM)

Micro porous structure of banana peels with particle sizes of 150-212 μm was observed at a resolution of 500x (Fig. 7). The micrograph of biosorbent shows some cavities in the surfaceûs structure capable of uptaking heavy metal ions as well as an irregular and porous microstructure
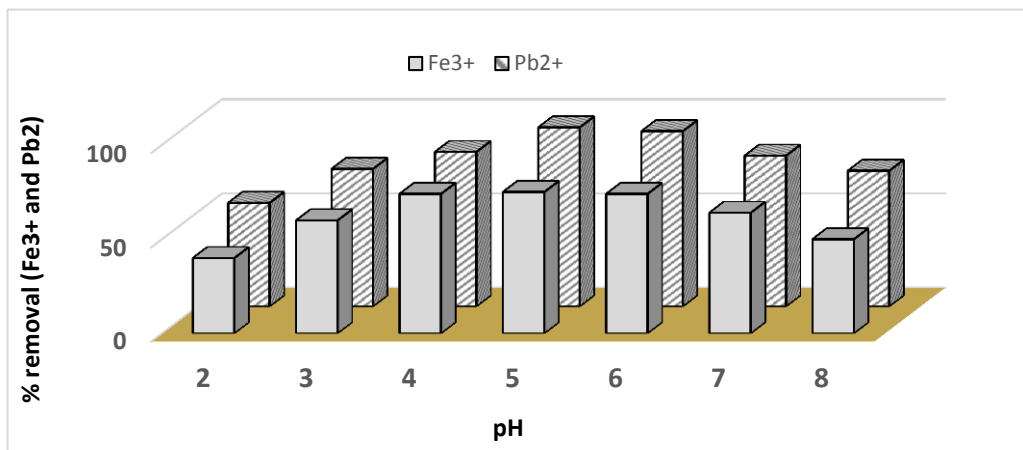
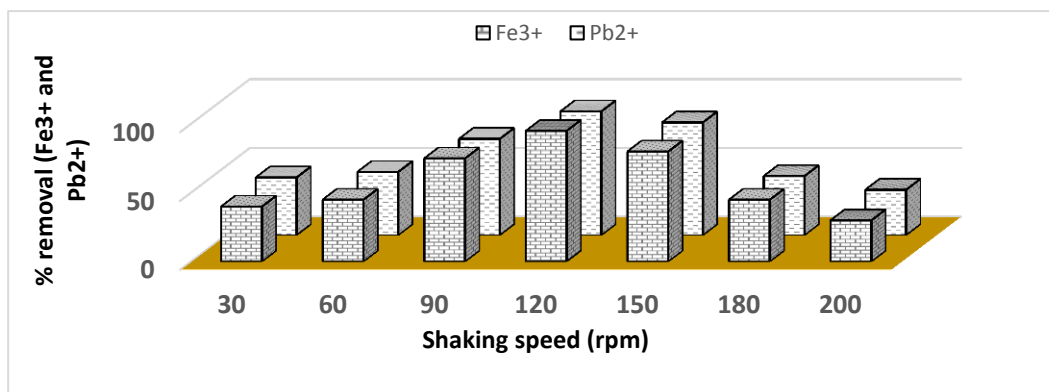**Fig.4. The influence of pH on the efficacy of heavy metal banana peels**

Fig. 5. Effect of shaking speed on removal of lead and ferric (t: 24h; Co: 10 mg/l; d: 5 g/l; T: 20°C; pH: 6.0)
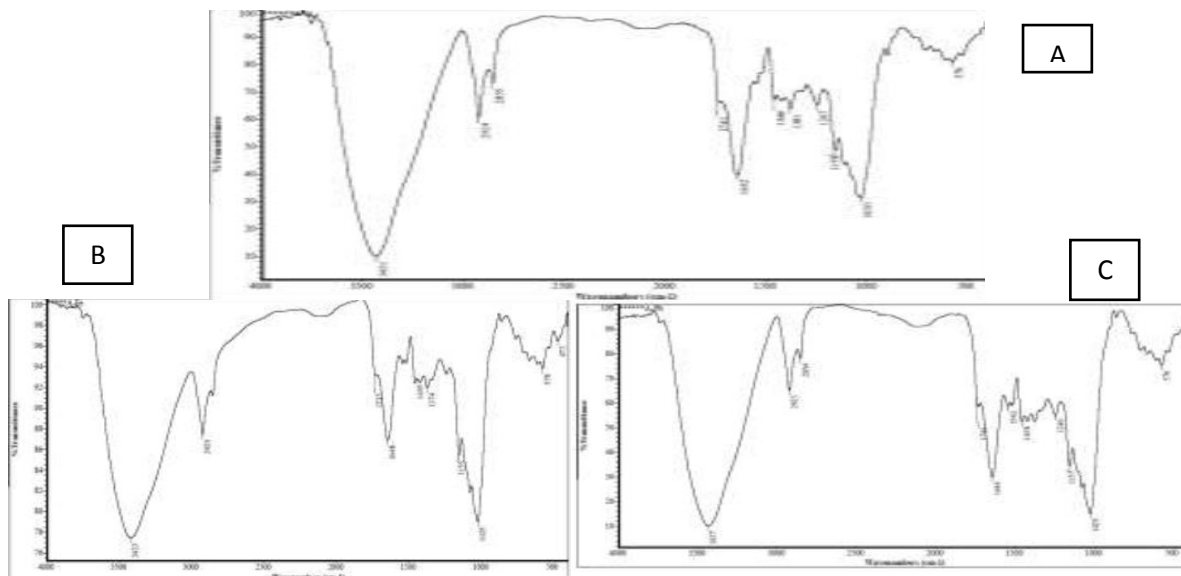

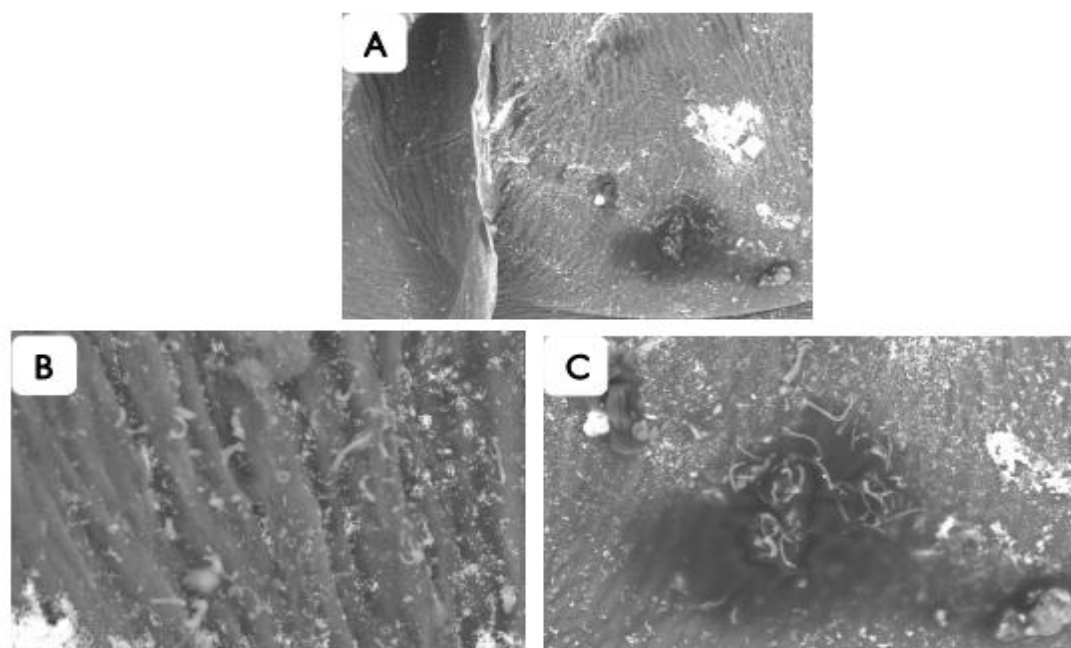**Fig.6. FTIR spectra of dried banana peel powder**

Fig (7.a): Scanning Electronic Microscopy SEM of banana peels before adsorption, Fig (7.b): SEM of banana peels after adsorption of Fe^{2+} and Fig (7.c). SEM of banana peels after adsorption of Pb^{2+}

Desorption Performance

Desorption of metals or regeneration of adsorbents from used-adsorbents are crucial to repeat the use of adsorbents, recover of precious metals and reduce the cost of operation in any water treatment system. The adsorbed metals on adsorbents can not be completely reversible as reported by several observations on literature (**Ainsworth *et al.*, 1994**). Eight solvent were used for desorption/regeneration experiment and the results are presented in Fig. 8.

Among the solvents the tap water, distilled water, CH_3COOH and NaOH were resulted a limited amount of metals ion recovery (<30%). The highest (94%) recovery was found with the use of $0.1N$ H_2SO_4.

The other two acids 0.1 N HCl and 0.1 N HNO_3 were shown lower efficiencies (54 and 72%). In the desorption system H^+ released from the acids which replaced metal cation (Fe^{+3} and Pb^{+2}) on the surface of the gellan (**Karthikeyan *et al.*, 2007**). The recovery and reuse processes for banana peels can be continued upto 10 times with minor deviation in removal efficiency.

Langmuir Adsorption Isotherm Model

The Langmuir adsorption isotherm model has been usually selected to study the adsorption principle and capacity. Constant values in model can express the surface properties and affinity of banana peels and also be used to compare the biosorptive capacities of banana peels for Fe^{+3} and Pb^{+2}. In this study, Langmuir adsorption isotherm model was used to fit experimental data. Constant values in models were listed in Table 1. Generally, when correlation coefficients (R^2) is greater than 0.95, the biosorption isotherm model is thought to be established. According to Table 1, obviously, Langmuir isotherm model described a better adsorption process. In the Langmuir isotherm model, b and q_{max} are Langmuir constants, related to the binding constant and the maximum adsorption capacity, respectively. The higher q_{max} value means stronger adsorption capability of biosorbent. In this study, the q_{max} value, the maximal metal uptakes (q_{max}) of banana peels were 79.28 and 96.58 mg/g for Fe^{+3} and Pb^{+2}, respectively. This result is in the same line with (**Ossman, 2014**) who stated that Langmuir adsorption isotherm describes the case of adsorption on banana peels very well.
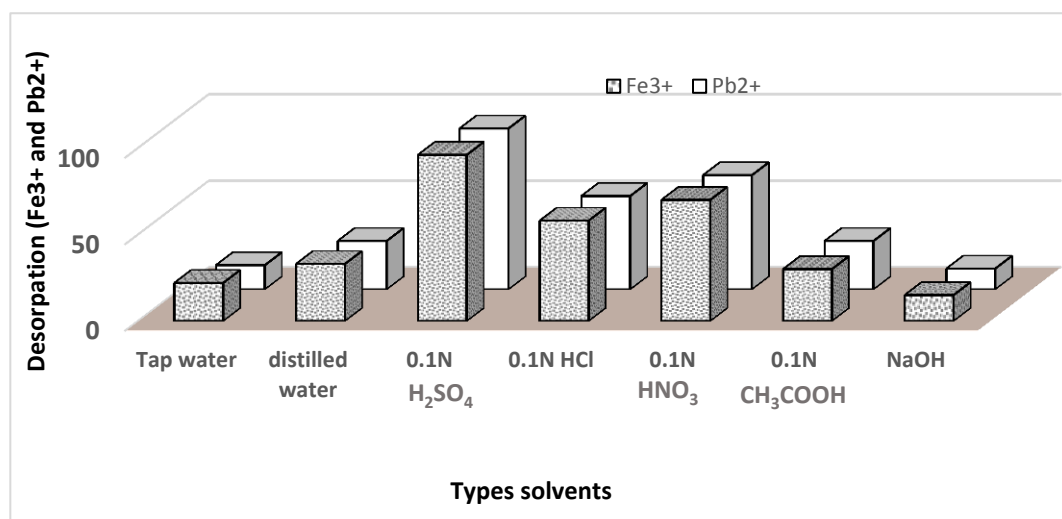
Fig. 8. Desorption of heavy metals Pb^{2+} and Fe^{3+}) (from banana peels (C_o: 10 mg/l; desorption time: 24hr.; rpm: 120; T: 20°C)

Table 1. Langmuir isotherm constants for the biosorption of Fe^{+3} and Pb^{+2} on banana peels

Heavy metals ions	Langmuir		
	b	q_{max}	R^2
Fe^{+3}	0.036	79.28	0.9801
Pb^{+2}	0.028	96.58	0.9763

Conclusions

The banana peels which are an agricultural waste substance have been converted into a carbonaceous adsorbent material. This product exhibits very good adsorption for iron and lead from aqueous solution. Adsorption of Fe^{+3} and Pb^{+2} by banana peels has been shown to depend on the pH, banana peels dosage and initial concentration. This work clearly indicates the potential of using banana peels as an excellent adsorbent for the removal of lead from aqueous solutions. One gram of banana peels is found enough to remove 83% and 77% of the iron and lead from an aqueous solution. Adsorption of ferric and lead on banana peels shows high association with Langmuir isotherm model. This result indicated that the metals adsorption capacity was in the sequence: $Fe^{+3} > Pb^{+2}$.

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إزالة ايونات الحديديك والرصاص من المحاليل المائية عن طريق الامتزاز باستخدام قشور الموز

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أجريت الدراسة الحالية لتقييم إمكانية استخدام قشور الموز لإزالة ايونات الحديديك و وايونات الرصاص من المحاليل المائية. يتضمن هذا البحث تأثيرات الجرعة والتركيز ودرجة الحموضة. وجد أن إدمصاص المعدن الثقيل على الممتزات يزداد مع تقليل التركيز الأولي ، وتزداد قدرة الامتصاص بقوة مع الأس الهيدروجيني في النطاق 2-8. أظهرت النتائج أن إزالة المعادن الثقيلة مثل ايون الحديديك و وايون الرصاص من المحلول المائي كانت فعالة باستخدام قشور الموز كممتص حيوي. كانت نسبة امتصاص أيونات الحديديك و الرصاص بواسطة قشور الموز عالية جداً. كان نموذج متساوي الحرارة Langmuir هو الأفضل لوصف البيانات التجريبية. وجد أن السعة القصوى للامتصاص كانت 79.28 و 96.58 مجم/ جم للحديد والرصاص على التوالي. بشكل عام ، تشير النتائج الحالية إلى أن قشور الموز هي مادة وامنه بيئيا، وامتصاص حيوي فعال ومنخفض التكلفة مما يمثل إمكانات عالية لعمليات إزالة ايون الحديديك و ايون الرصاص من المحاليل المائية.