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RESEARCH ARTICLE

JORDANIAN NATURAL ZEOLITE USE AS SORBENT FOR REMOVAL LEAD FROM AQUEOUS SOLUTIONS

***,[1]Abdal-Kareem M.A. Dawagreh, [1,2]Mohammad R. Abdallah, Qasaimeh, [1]Nada M. Al-Ananzeh, [1]Hussam E Alkhasawneh and [1]Mohammad Hailat**

[1]Department of Chemical Engineering, Al-Huson University College, Al- Balqa' Applied University, PO Box (50), Al-Huson 21510 , Irbid, Jordan
[2]Environmental Engineering Department, Al-Lith Engineering College, Umm Al-Qura University, Saudi Arabia

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ABSTRACT

In this work the potential of zeolites for lead ions adsorption from industrial aqueous solutions was studied. This was done by using batch sorption experiments, where zeolites made in direct contact with the contaminant under study until equilibrium is attained. Effects of different parameters on the sorption process were examined, such as sorbent concentration, contact time and PH. Lead ions uptake increases with increasing the initial concentration of the zeolites. The equilibrium data were well represented by Freundlich isotherm model. Also, an increase in the PH was found to enhance the removal of these ions. The volcanic Tuff which also called "zeolites" is a locally available material that can be utilized of the removal of contaminants, such as heavy metals, from wastewater streams. Its availability and cheapness make it a good candidate among other sorbents in the adsorption science. In Jordan, zeolites are found in the following locations: zeolitic tuffs are located at Jabal Aritayn (30km NE of Azraq), Tlul Al-shahba (20km E of Al safawi), Tal-Rimah (35km NE of Al Mafraq) and other small deposits in the South Jordan (Tell Burma, Tell Juhaira and Wadi El-Hisa) and in Central Jordan (Makawer, Al-Zara, Wadi Heidan and Wadi Al-Walah). Phillipsite, chabazite and faujasite are the most abundant zeolite minerals found in the Jordanian zeolitic tuff. The zeolite content in these tuffs varies from (20% to 50%). processing routes, zeolite concentrates with grades up to 90% were achieved. The objective of this work is to study the possibility of the use of zeolites for the removal of heavy metals from industrial aqueous solution. The uptake capacity of zeolites and the effect of different parameters on the adsorption process, such as the PH, and concentrations, will be considered.

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INTRODUCTION

Zeolites are a group of hydrated aluminum-silicates of the alkali or alkaline earth metals (sodium, potassium, magnesium, calcium) characterized by low mining cost, availability, bulk density and high resistance to alteration (Mercer and Ames, 1978). Zeolites have a three-dimensional crystalline framework of tetrahedral silica or alumina anions strongly bonded at all corners, and they contain channels filled with water and exchangeable cations. The structure is suitable for ion exchange due to isomorphism: the replacement of Al_3^+ with Si_4^+ in the structure, giving rise to a deficiency of positive charge in the framework. This is balanced by mono and divalent exchangeable cautions such as Na^+, Ca_2^+, K^+ and Mg_2^+. The main consequence of this structure type is represented by the reversibility of the hydration and cation exchange processes that preserve the original network. Thus, they present specific properties (e.g., adsorption-desorption capacity, ion exchange capacity, catalytic properties), which confer to this group of minerals important and diverse possibilities of usage. Ion exchange is defined as a process in which an insoluble substance removes ions of positive or negative charge from an electrolytic solution and releases other ions of like charge into the solution in chemically equivalent amounts (Benefield *et al.,* 1982). Cation selectivity refers to the preference order of zeolite for cations based on the various factors which determine the selectivity; ion size, valence and

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****Corresponding author: Abdal-Kareem M.A. Dawagreh***
Department of Chemical Engineering, Al-Huson University College, Al- Balqa' Applied University, PO Box (50), Al-Huson 21510 , Irbid, Jordan.

hydration energies are important factors in determining the selectivity of a given ion in a specific system. The zeolite prefers or is highly selective for certain cations and does not prefer or is less selective for others (Colella, 1996) has demonstrated that zeolites can be used in radioactive wastewater treatment due to their high ion exchange selectivity and resistance to degradation from radiation. (Pansini, 1996) found that zeolite removes heavy metals from natural and industrial wastewaters. (Mercer *et al.,* 1970) have used natural zeolite to remove ammonium ions from wastewater. (Blanchard *et al.,* 1984) have demonstrated that the preferred method for removing traces of NH4+ from fresh hatchery water is a possible method to satisfy the standard for water reuse. The proposed design has used an ion exchange using natural clinoptiololite. (Semmens, 1978) have indicated that clinoptiololite has a good selectivity for the ammonium ion. The efficiency order is as follows: Pb_2+ > NH_4^+ > Cd_2^+, Cu_2^+, Sr_2^+ > Zn_2^+ > Co_2^+ The above heavy metals as well as ammonium in wastewater can be treated by passing it through clinoptiololite columns (Simes and Hidin 1978) has suggested that clinoptiololite is highly selective for barium and lead, but is less selective for copper, cadmium and zinc. Zeolites in Jordan were first discovered (Dwairi, 1987). The Jordanian phillipsitic tuff has a good efficiency in ammonium removal from wastewater, as well as Hungarian clinoptiololite, but, phillipsite is more selective for ammonium ions (Dwairi, I. 1991). In addition, (Dwairi, 1992) has found that the Jordanian phillipsite is of good efficiency for cesium fixation and immobilization from nuclear waste in comparison with phillipsitic rich tuff from California and moderately rich tuffs from Arizona and Nevada.

(Attili, 1992) has evaluated the Jordanian phillipsitic tuff for applications in water softening, removal of ammonium and toxic heavy metal ions in laboratory and pilot plant scale (Al-Rashdan, 1994) has evaluated the effect of Jordanian phillipsitic zeolitic tuff from Jabal Aritayn on the removal of ammonium and phosphate from domestic wastewater. It was concluded that phillipsitic zeolitic tuffs have shown acceptable efficiency in the removal of pollutants from the effluent. (Ibrahim *et al*., 2002) have evaluated zeolites from Tell Rimah for industrial wastewater treatment. The results have indicated that Tell Rimah zeolitic tuff showed high selectivity for the removal of Cu, Cr, Ni and Zn from electroplating factory effluents and could be used for removing Pb and Fe from the wastewater of the battery factory. (Ibrahim, 1987) found that faujasite tuff from Jordan has a good selectivity for lead from wastewater. (Al Dwairi, 2007) studied the characterization of Jordanian zeolitic tuff and its potential use in wastewater treatment. He studied the mineralogy and geochemistry of all zeolitic tuff localities in Jordan and evaluated Jordanian natural zeolitic tuffs obtained from Hannoun (HNZ) and Mukawir (MNZ) volcanoes for removing selected heavy metals (Zn and Pb) and total organic carbon from domestic wastewater. The results show that HNZ and MNZ are suitable for domestic wastewater treatment (Ibrahim *et al*., 2009) have used natural phillipsite-faujasite tuff from Jordan to remove paraquat from synthetic wastewater. They found that natural zeolites are suitable to remove paraquat. (Al Dwairi, 2009) evaluated the Jordanian phillipsitic tuff from Al-Ataita volcano for applications in wastewater treatment. He concluded that Al-Ataita phillipsitic tuff shows good removal percentages of ammonium from municipal wastewater. Zeolite is an amazing crystalline mineral capable of adsorbing and absorbing many different types of gases, moisture, petrochemicals, heavy metals, low-level radioactive elements and a multitude of various solutions. The channels in the zeolite provide large surface areas on which chemical reactions can take place. The cavities and channels within the crystal could occupy up to 50% of its volume. zeolites can adsorb or absorb large amounts of materials, such as ions or gas molecules.zeolites are hydrated aluminosilicates characterized by high surface areas and high cation exchange capacities.

Zeolite characteristics important to a wide range of applications

Industrial wastewater streams are normally polluted with toxic materials such metals, phenols and dyes. These materials have harmful effects on the human and animals, plants and aquatic life. Heavy metals are among the most serious contaminants that are usually produced during metal processing, finishing electroplating and paper milling. Examples of these metals are lead and zinc that are among the toxic metals affecting the environment. It is of great importance to adopt new technologies to remove them from wastewater to reach acceptable limits set by governmental regulations. According to World Health Organization, the limit of lead is 0.001 ppm (World Health Organization, 1984) (Brandt *et al*., 1993). Adsorption is the adhesion of atoms, ions, or molecules from a gas, liquid, or dissolved solid to a surface (Brandt *et al.,* 1993). This process creates a film of the adsorbate on the surface of the adsorbent. This process differs from absorption, in which a fluid (the absorbate) permeates or is dissolved by a liquid or solid (the absorbent) .Note that adsorption is a surface-based process while absorption involves the whole volume of the material. Ingestion of metals such as lead (Pb), cadmium (Cd), mercury (Hg), arsenic (As). copper (Cu) and chromium (Cr) may pose great risks to human health. Trace appearance such as lead and cadmium will interfere with essential nutrients of similar such as calcium (Ca^{+2}) and zinc (Zn^{+2}) (Cussler, 1997). The heavy metal which we are interested in is lead (Pb^{+2} ions), so its definition, sources and effects will be disused in the following paragraphs. Lead: is the most significant of all the heavy metals because it is both very toxic and common. It gets into water from corrosion of plumbing materials, where lead has been used freely since roman times (Rollman *et al*., 1981). Also, it can gets into water from different industries such as: welding process of lead to make the water and sewage pipes, welding process in keeping foods in plates or in cans, inks of printers, pencils and in the painting industries, and as many other applications and a industries. Because of size and charge similarities, lead can substitute for calcium and included in bone. Children are especially susceptible to lead because developing skeletal systems require high calcium levels (Kokotailo *et al*., 1978).

Experimentation

Material Preparations

The materials used in this study are mainly Lead, and Zeolite. A stock solution of Pb ions was prepared from $Pb.H_2O$ with concentration of 1000 ppm. The stock solution was used to prepare 76 samples of 10ppm Lead solutions. Zeolite was in the form of a solid rock, and was brought from the Jabal Aritayn (30km NE of Azraq). It was crushed to fine particles and screened. The zeolite used for adsorption were retained less than 2 mm to powder. This size is appropriate since it provides the required surface area for adsorption, and does not float to give good contact between the sand and the solution. zeolite was dried at 110°C, for 48 hours.

Batch adsorption Experiment

Cretin amount of Zeolite (Z) was transferred to flasks containing 500ml of Pb^{+2} an aqueous solutions of Pb^{+2} ions of predetermined concentrations. The initial Pb^{+2}ions concentration were 10 ppm. The samples bottels were placed in a magnatic stirrer to enable the mixture to be agitated at room temperature. Sampels are withdrawn at known time periods in order to study the kinetics of the sorption process. Otherwise, the sampels were allowed to come to equilibrium by storage 48 hr with agitation followed by separation by centrifugation in Garber device followed by filtration, the suspensions were then removed and separate the Zeolite particels from the solutions. Finally the filtrate was collected and analyzed, for residual Lead concentration. The standard conditions used through out the study were: temperature of 25 °C, PH=8.5, Z=25g=50 mg/ml and Pb ions concentration =10ppm.

Analysis of Zeolite

The zeolite have been analyzed by X-ray fluorescence technique XRF ARL 9900 wavelength dispersive – Simultaneous XRF Power 4.2KW (production 2010).Shown the result in the table (1):

Analysis of Lead

After filtration, the filtrate is taken to the Jordan Atomic Energy Commission (JAEC) lab and analyzed for residual concentration of Lead using Inductively Coupled Plasma Optical Emission Spectroscopy (**ICP- OES**) model "The ICAP 6300 Series ICP".

Analysis of Data

After measuring the residual Lead ions concentrations **C,** the adsorbate uptake is calculated using the following equation:

$$q = \frac{C_0 - C_t}{M} \times V \qquad \text{..............................} \quad (1)$$

Where,

q: is the Lead uptake (mg adsorbate / g of adsorbent).
C_o: is the initial concentration.
C_t: is the final concentration after adsorption process or after fixed time period.
V:is the volume of suspension (ml) .
M: is the mass of sorbent (mg).

RESULTS AND DISCUSSION

Effect of Adsorbent

The effect of sorbent addition on the removal of Pb ion from aqueous solutions was investigated at $25c^0$. Different amounts of the zeolite were used at initial concentration of Pb (l0ppm). Other conditions were not altered in this run. The results are given in Figure (1). The residual concentrations of Pb^{+2} ions further decreased upon increasing adsorbent concentration from (50, 70, 90) mg/ml. The reason for this is the increase of the number of active sites available for metals binding upon addition of the zeolite, in other words that was due to the greater availability of exchangeable/ adsorbtion sites at higher sorbent concentration.

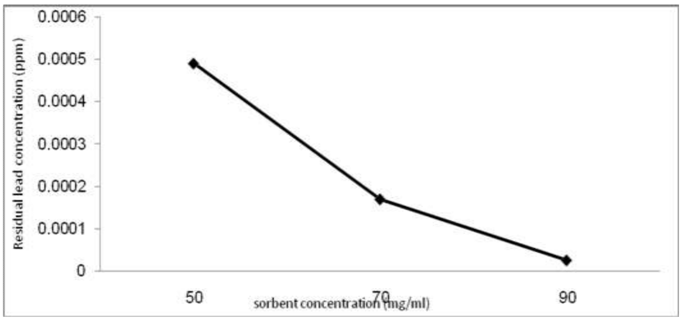

Figure 1. Effect of sorbent concentration on the amount of lead removal

when different sorbent concentrations 50, 70, and 90 mg/mI, It is seen that when increasing the contact time of the adsorption process, the residual Lead concentration decreasing . as shown in figures:

Effect of PH

An examination of the effect of the PH on metal sorption was considered in this part. The PH was adjusted to give three values 3, 7, *and 12.* of the metal. Solutions of 10ppm Pb^{+2} were used, while other conditions were fixed. Figure (2) shows that increasing the PH resulted in an increase the values uptake. This is expected due to the negative charges prevailing at the surface of the sorbent at high PH values, which give rise to the attraction forces between the Zeolite and Pb^{+2} ions. At considerably low PH, the opposite occurs, where repulsive forces take place between Pb^{+2}ions and the positively charged surface of the sorbent thus lowering the uptake.

This is in agreement with earlier findings reported in the literature. For example, Lothenbach (1996) studied the effect of PH on the uptake of Cd^{+2},$Cu^{+2}$and Pb^{+2} ions by al zeolite and reported that the uptake of these metal ions increased with increasing solution PH. On the one hand, the low uptake of Pb^{+2} ions at lower PH may be attributed to strong competition by H^{+}ions for the available exchangeable sites, but on the other hand it was reported that the Zeolite surface becomes positively charged at lower PH values because of amphoteric dissociation and consequently would not favour the adsorption of positively charged ions as Pb^{+2} ions.

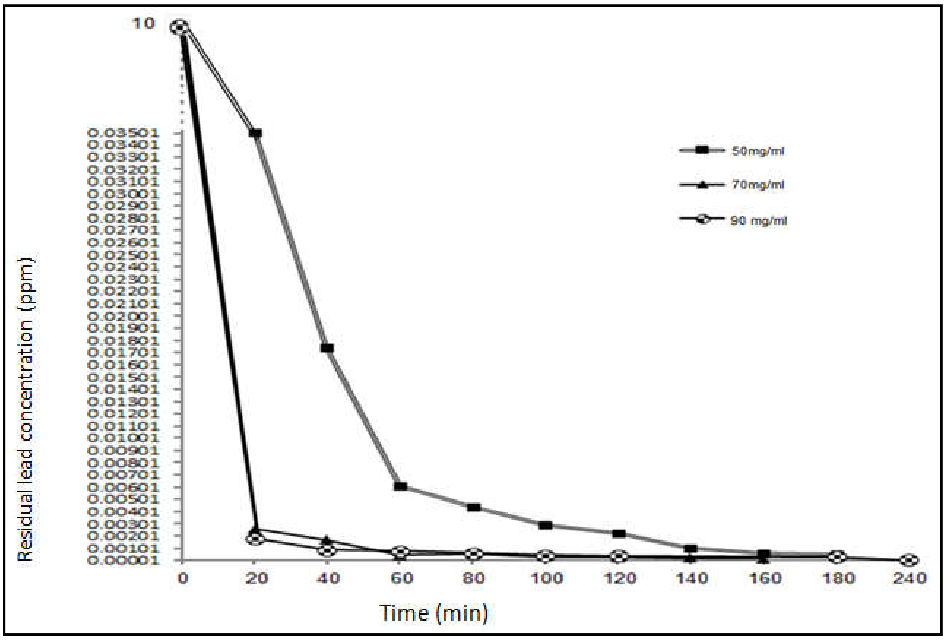

Figure (1-a) residual lead ccncentration (ppm) versus conduct time (min) at different adsorbent concentration using 10 ppm initial lead concentration

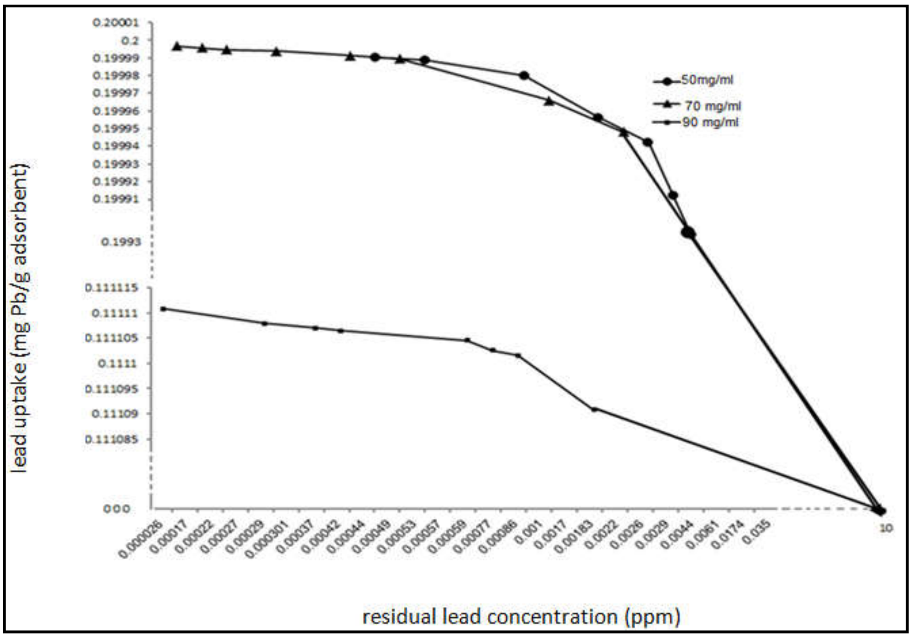

Figure (1-b) lead uptake (mg Pb/g adsorbias.vab.ac.be. Institute of Adsorption Science (IAS)

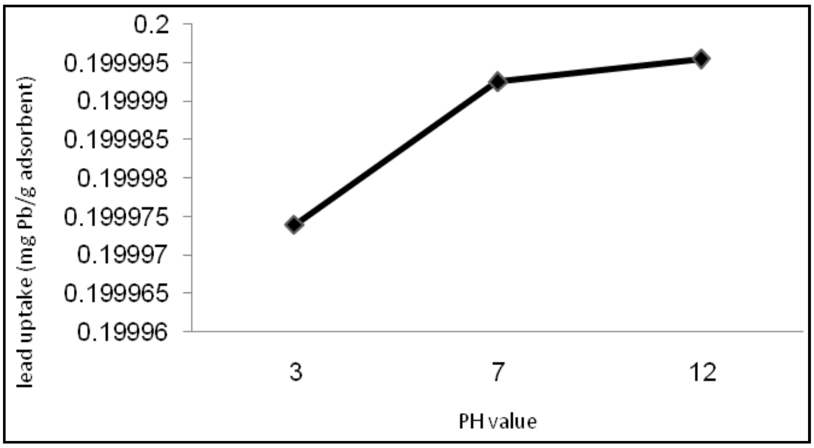

Figure (2) lead uptake (mg Pb/g adsorbent) versus PH for Pb ions adsorption on natural zeolit

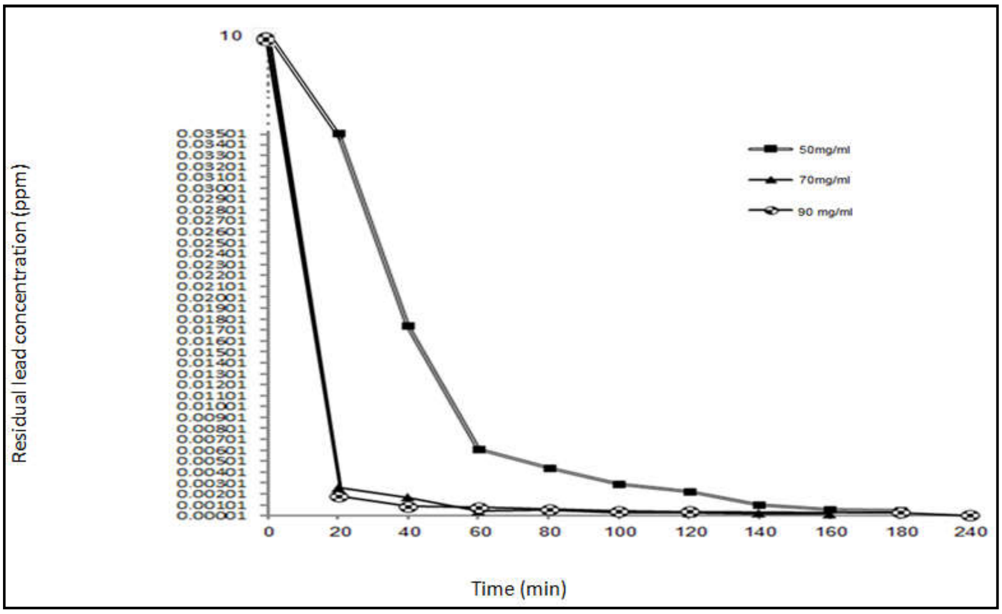

Figure (2-a) residual lead ccncentration (ppm) versus conduct time (min) at different adsorbent concentration using 10 ppm initial lead concentration

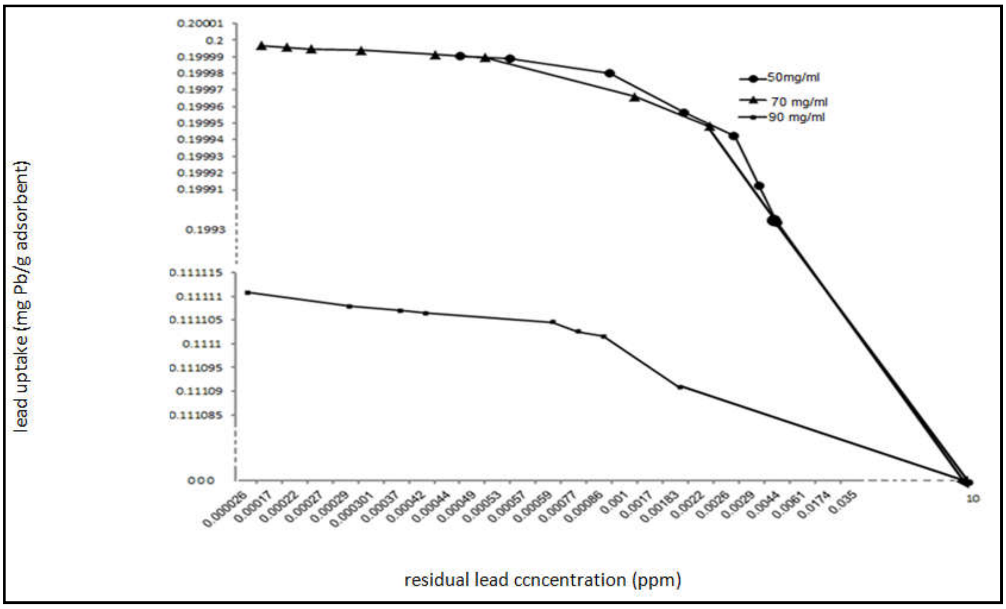

Figure (2-b) lead uptake (mg Pb/g adsorbent) versus residual lead ccncentration (ppm) conduct time (min) at different adsorbent concentration using 10 ppm initial lead concentration

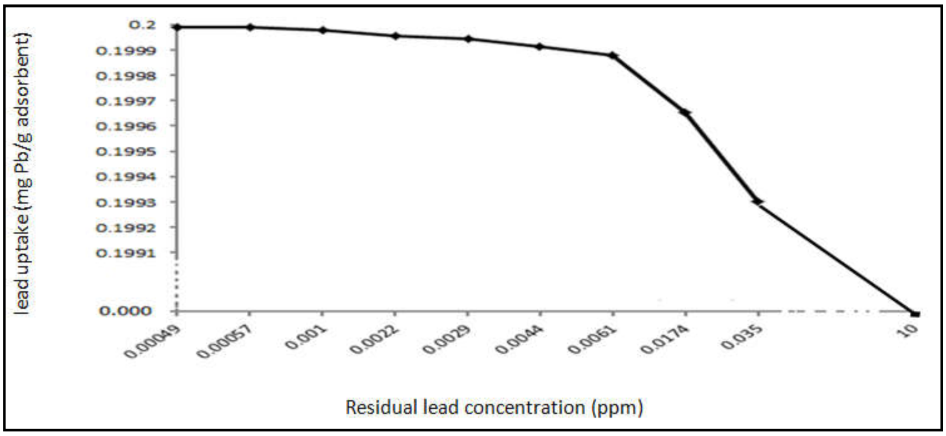

Figure (3-a) lead uptake (mg Pb/g adsorbent) versus residual lead concentration (ppm)

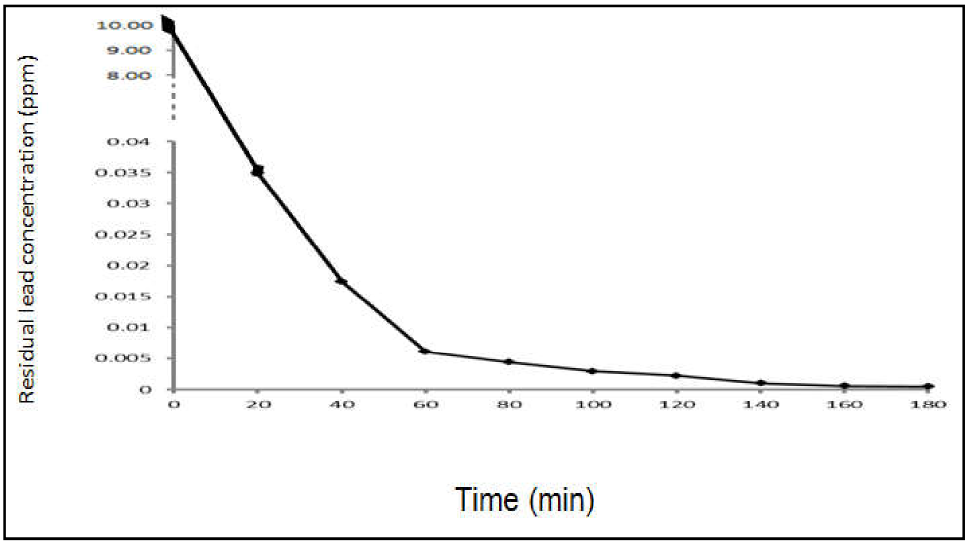

Figure (3-b) residual lead ccncentration (ppm) versus conduct time (min)

Table 1. The amount of different components in zeolite

Compound	Composition (%)
CaO	9.08
SiO_2	46.19
Al_2O_3	14.47
Fe_2O_3	13.81
MgO	8.66
SO_3	0.11
K_2O	1.47
Na_2O	3.47

Effect of Conduct Time

The kinetic of Pb^{+2} ions adsorption on Al-zeolite was studied using one initial concentration of the adsorbate (10 ppm) at *25°C,* The results are shown in figures *(3-a, 3-b).* It is seen that the rate of Pb^{+2} ions adsorption on Al-zeolite was relatively fast Equilibrium was attained in almost 20 minutes under the conditions of this experiment. The high initial uptake suggest that adsorption occurred mainly at the surface of the sorbent. It is seen that when increasing the contact time of the adsorption process, the residual Lead concentration decreasing. As shown in figures

Conclusion

The zeolite can be used as adsorbent for heavy metals as it was tested for Lead in this work as a model heavy metal. The Freundlich model fit the equilibrium data.

And after this study we can summaries our results by Saied that : The amount of Lead removal increases with increases the amount of adsorbent used, Increases the PH value, and Increases the conduct (shaking) time. The maximum removal capacity of lead ions 99.997%

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