Original Article

Removal of Pb(II) ions from aqueous solution using leaves of Palm trees

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ABSTRACT

The presence of heavy metals in aquatic environments creates many problems, and if management is not done, the health of human societies and the environment will be endangered. Due to the stability of metals in aquatic environments, we must remove them from aquatic environments. Lead is a heavy metal that is very dangerous and toxic and poses a serious threat to humans and the environment. One of the most common methods of removing heavy metals from aqueous solutions is surface adsorption and it has attracted the attention of researchers due to its good performance and efficiency in terms of economic. In this study, the ability of palm tree leaves as an adsorbent to remove lead (II) ions from aqueous solutions by surface adsorption method was investigated. The effect of three parameters (pH, initial concentration of lead (II) ions, contact time between adsorbent and lead ions) on the adsorption rate of lead was investigated and also isotherm and kinetics of lead adsorption were studied. The results showed that by increasing the pH from 4 to 6, the adsorption rate of lead by the adsorbent increased and the maximum adsorption efficiency at pH was 6. Then, by increasing the pH from 6 to 8, the adsorption rate of lead by the adsorbent decreased. Investigation of the effect of initial concentration of lead ions (II) on lead adsorption rate also showed that with increasing concentration of lead ions from 20 to 300 mg/l, the adsorption rate increases. Examination of the effect of contact time on lead adsorption rate showed that with increasing contact time between adsorbent and lead ions from 15 to 60 minutes, the adsorption rate increased and then decreased in contact time of120 minutes. Examination of adsorption isotherms showed that the Langmuir model described lead adsorption by palm tree leaves better than the Freundlich model. Investigation of adsorption kinetics showed that the Ho model was more consistent with laboratory data than the Lagergren model.

Keywords: Lead, pH, initial concentration, contact time, Langmuir and Freundlich, Lagergren and Ho

Introduction

Environmental protection, in which today's generation and future generations must have a growing social life, is considered a public duty. Today, due to industrial growth; most factories, power plants and refineries deal with water and the result is water pollution. Therefore, we must be enough information about water systems, the types of pollutants, the effects of pollution, the methods of disposing of pollution, and contamination prevention methods [1]. Lead is an old metal that has been used for thousands of years for various purposes. For example, in the past, lead was used for glazing of dishes and in pharmacy and medicine. Also from lead pipes were used for convey water. Later, the use of lead was banned due to food poisoning caused by cooking food in lead boilers or consuming poisonous water. Today, lead compounds are used in the manufacture of glass, ceramics, as well as plastic stabilizers. Red lead is used to make television bulbs. About 10% of lead is converted to lead alkyl compounds, which are added to gasoline as additives. About 20% of lead products are used in the form of lead sheets, cable coatings, soldering, making ammunition, combining with precious metals such as gold, and production of pesticides. About 40 to 50 percent of lead products are used to make lead-acid batteries [2]. Lead is one of the four metals that has the most side effects on human health. Hemoglobin biosynthesis disorders and anemia, high blood pressure, kidney damage,

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miscarriage and premature infancy, nervous system disorders, brain damage, male infertility, decreased learning ability and behavioral disorders in children are some of the negative side effects of increasing the concentration of lead in the body [3]. Therefore, the removal of this pollutant from water resources is a vital issue. The maximum amount of lead according to the standards in different countries and according to the 1979 food laws in drinking water is 50 micrograms per deciliter [4]. Heavy metal removal methods include sedimentation, surface adsorption, ion exchange, electrodialysis, and membrane processes [5]. One of the methods used to remove lead contaminants is the sedimentation method. Disadvantages of this method include its high cost and also the production of large amounts of sediment that these sediments themselves must be separated by other methods [6]. So, it is important to find other methods that are both more environmentally friendly and more efficient and economical. Methods such as membrane filtration, surface absorption and ion exchange are among the topics of interest to researchers in this field [7]. Surface adsorption is method that a simple, inexpensive, and efficient operation. Surface adsorption is a process with a very rich scientific and experimental background and is very operational in terms of exploitation and design. In the surface adsorption process, the adsorbents can be recycled using simple heat transfer mechanisms (heating) or chemical washing, and the adsorbent can be reused [8]. Reusability makes cost-effective surface adsorption process economically viable and justifiable. Over the past few years, much progress has been made to improve surface adsorption performance, including the development of various adsorbents for the removal of lead ions can be noted. Research by Bousher et al (1997) on sawdust, as well as Sun and Xu (1997) on sunflower stalks showed that these materials are effective in removing metal compounds from wastewater [9, 10]. Karimi et al (2018) used corn silk to remove lead ions from contaminated water [11]. The results showed that corn silk adsorbent is effective in removing lead contaminants from water environments due to its high effective surface, having SiOH groups, maximum adsorption capacity and rapid kinetics of reaction. Saeed Faraji et al (2017) examined lead removal using pear peel by surface adsorption method [12]. The results showed that pear peel, as one of the agricultural wastes that can be obtained from compote and juice factories, is a cheap and affordable adsorbent that has the ability to remove lead metal from aqueous solution. Foroutan et al (2017) investigated the kinetic behavior of adsorbing lead ions from aqueous solutions using lotus leaves [13]. The results showed that this adsorbent is suitable for the removal of lead ions from aqueous solutions, and the second order pseudo kinematic model has a greater ability to describe the experimental data. In this study, the ability of palm tree leaves as an adsorbent to remove lead (II) ions from aqueous solutions by surface adsorption method was investigated. The effect of three parameters (pH, initial concentration of lead (II) ions, contact time between adsorbent and lead ions) on the adsorption rate of lead was investigated and also isotherm and kinetics of lead adsorption were studied.

Materials and Methods

Palm leaves were collected from Palm groves in Bushehr province (located in Iran). The leaves were then washed four times with water and twice with distilled water to remove dust and dried in an oven at 70 ° C for 36 hours. After drying, the leaves were separated from the middle stem and divided into very small pieces by hand. Then the pieces of leaf were milled and passed through the 100 mesh sieve to obtain a very homogeneous and very fine powder with a high adsorption surface. 1.59 grams of nitrate lead ($Pb(NO_3)_2$) was weighed and dissolved in one liter of distilled water. Then, from the mother solution with a concentration of 1000 mg/l, solutions with a concentration of 20, 90, 180, 300 mg/l were prepared. The pH of all solutions (solutions with a concentration of 20, 90, 180, 300 mg / l) was adjusted to the numbers 4, 6 and 8 with the help of 0.1 normal nitric acid (HNO3) and 0.1 normal NaOH. Then 40 ml of all solutions was removed and 0.4g of adsorbent was added to it and it was stirred for 15, 30, 60 and 120 minutes at 200 rpm with a magnetic stirrer at 25 °C. The solutions were smoothed with Whatman brand No. 41 filter paper, then added to each of the filtered solutions of Dithizone reagent, and the adsorption rate was measured for lead at a wavelength of 520 nm with the help of UV-VISIBLE spectrophotometer (CamSpec model M550). Finally, equilibrium isotherms were used to describe the data. Equilibrium isotherms are equations that show the distribution of the absorbed material between the soluble phase and the absorbed in equilibrium state [14]. The Langmuir adsorption isotherm shows the single-layer adsorption on the homogeneous surface without the reaction between the adsorbed molecules and the uniform energies of adsorption on the surface by Equation (1) [15].

$$q_e = \frac{q_e b\, c_e}{k_l + k_l c_e} \quad (1)$$

In this Equation:

C_e: Concentration of the absorbed substance in equilibrium state in the liquid phase (milligrams per liter).

q_e: The amount of absorbed ion per unit mass of adsorbent in equilibrium state (mg/g).

b: Adsorption capacity in solid phase (mg/g).

k_l: Constant of adsorption.

The Freundlich adsorption isotherm is an experimental model for explaining multi-layered adsorption with heterogeneous energy distribution with the reaction between the adsorbed molecules that shown by Equation (2) [16].

$$q_e = k_f c_e^{\frac{1}{n}} \quad (2)$$

In this Equation:

k_f: Constant of isotherm in connection with the amount of adsorption (liters per gram).

1/n: This is the amount of adsorption intensity that changes with the non-uniformity of the material.

One of the important studies in the adsorption process is the study of the effect of contact time with the adsorption value, which is known as adsorption kinetics studies. In order to study the control mechanisms of the adsorption process, the kinetics models of the Lagergren Pseudo-First-Order and Ho Pseudo-Second-Order will be used [14]. Equation (3) shows the Lagergren model [14].

$$\log(q_e-q_t)=\log q_e-(k_1 t/2.303) \quad (3)$$

In this Equation:

q_e: The amount of ions absorbed in the equilibrium time (mg/g).

q_t: The amount of ions absorbed in the t time (mg/g).

k_1: Constant of adsorption in the Lagergren model.

Equation (4) shows the Ho model [14].

$$\frac{t}{q_t}=\frac{1}{k_2 q_e^2}+\frac{t}{q_e} \quad (4)$$

In this Equation:

q_e: The amount of ions absorbed in the equilibrium time (mg/g).

q_t: The amount of ions absorbed in the t time (mg/g).

k_2: Constant of adsorption in the Ho model (g/mg min).

Results and Discussion

The effect of pH on lead adsorption rate

The initial pH of the solution is an important parameter in the process of adsorption and removal of heavy metal ions by the adsorbent, because the hydrogen ions (active functional group) inside the solution compete severely with the heavy metal ions to be located on active sites in the cell wall. The rate of adsorption efficiency is largely dependent on the concentration of hydrogen ions in the solution [17]. Figures 1 to 3 show that by increasing the pH from 4 to 6, the adsorption rate of lead by the absorbent is increased. At low pH, the adsorption rate of lead ions was low, because the concentration of hydrogen ions in the solution was high, and the hydrogen ions competes with the lead ions for active sites on the adsorbent cell wall and occupies the adsorbent active sites. As the pH increases, the amount of negative charges on the adsorbent surface and inside the solution increases, and these ions increase the adsorption efficiency of lead ions, and the maximum adsorption efficiency at pH is 6. By increasing the pH from 6 to 8, the adsorption rate of lead by adsorbent decreased, because in this case the concentration of hydroxide ions in the solution increased and formed a complex with lead metal ions and competed with lead ions in order to be located on adsorbent active sites, as a result, the efficiency of process adsorption decreases again [18].

The effect of the initial concentration of lead ions on the adsorption rate of lead

In the process of discontinuous adsorption, the initial concentration of metal ions in the solution provides the main and key role in creating the force required to transfer the mass between the solid and liquid phases [19]. According to Figures 1 to 3, the adsorption rate of lead increases, by increasing the concentration of lead ions from 20 to 300 mg/l, respectively. The reason for this can be explained by the fact that in general, at low concentrations such as concentrations of 20, 90, 180, 300 mg / l, certain sites are responsible for metal adsorption, which these sites at high concentrations are saturated and the metal adsorption is reduced. At low concentrations, ion adsorption sites are occupied, but with increasing concentrations, ion exchange sites are also filled.

The effect of contact time on lead adsorption rate

Another important parameter in the adsorption of heavy metal by the adsorbent is the contact time of adsorbent with heavy metal ions. According to Figures 1 to 3 and the results obtained, with increasing contact time between the adsorbent and the lead ion, the adsorption rate increases from 15 minutes to 60 minutes and then decreases in 120 minutes. The reason for this is related to the high number of adsorption active sites in the adsorbent surface in the early times. But over time, as the number of adsorption active sites in the adsorbent surface decreases and the competition for ions to occupy active sites increases, the adsorption upward trend is declining.

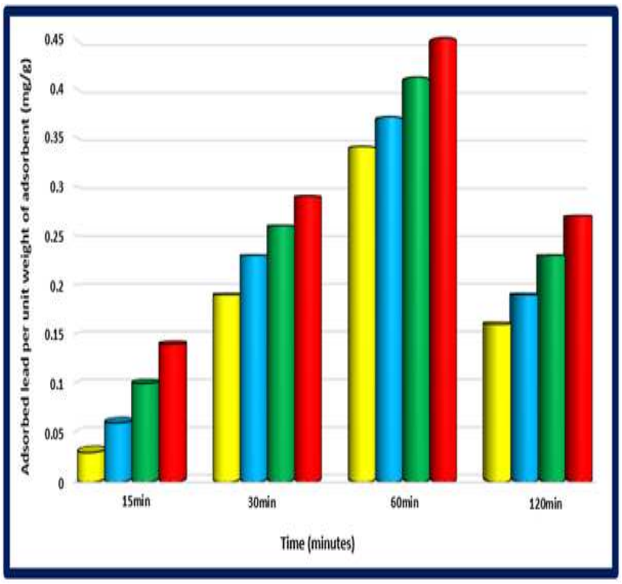

Figure 1-Investigating the cross impact of contact time and initial concentration in lead removal at pH=4

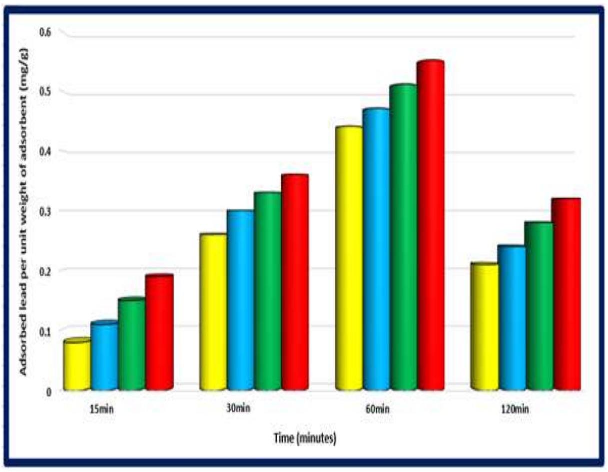

Figure 2-Investigating the cross impact of contact time and initial concentration in lead removal at pH=6

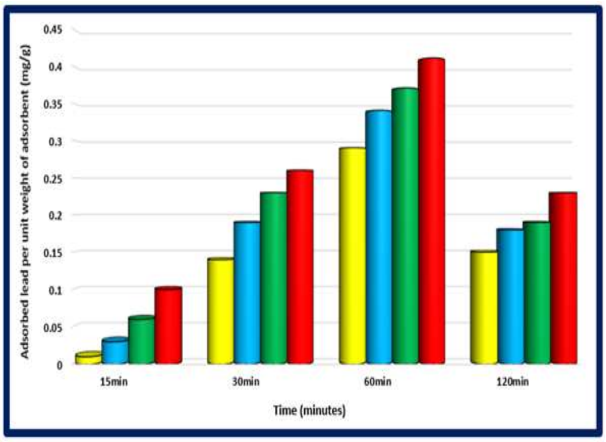

Figure 3-Investigating the cross impact of contact time and initial concentration in lead removal at pH=8

The results of the study of adsorption isotherms

Experimental data were studied in this study with two isotherms Langmuir and Freundlich. According to Figures 4 and 5, which show the equations of Langmuir and Freundlich, it is clear that the Langmuir model better describes the surface adsorption of lead on the palm tree leaves than the Freundlich model.

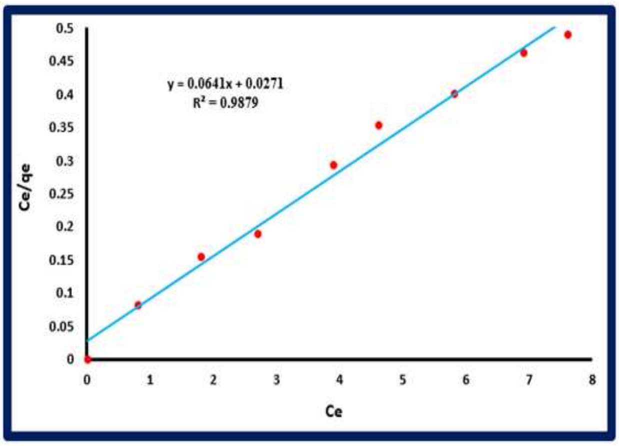

Figure 4-The results of the Langmuir isotherm related to adsorption of lead on adsorbent

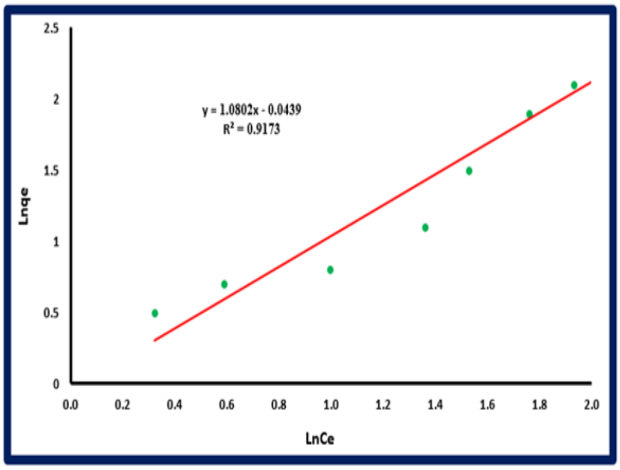

Figure 5-The results of the Freundlich isotherm related to adsorption of lead on adsorbent

The results of the study of adsorption kinetics models

Experimental data were studied in this study with Lagergren and Ho adsorption kinetics models. The results of the study of adsorption kinetics models for experimental data obtained from surface adsorption of lead on the studied adsorbent are presented in the Figure 6. According to Figure 6, it can be said that experimental data are more consistent with the Ho model.

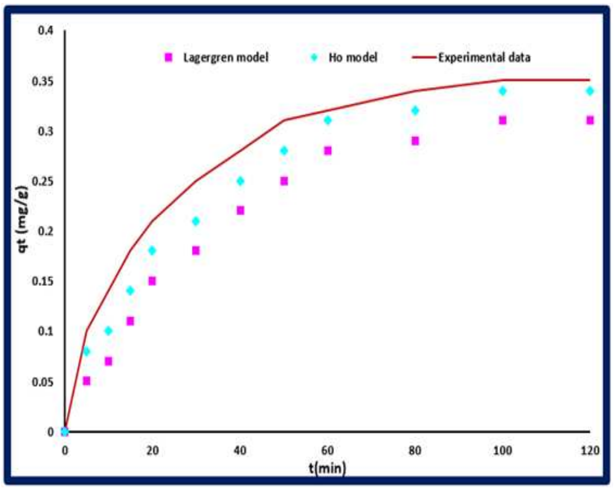

Figure 6-The fitting of kinetics models on experimental data of lead adsorption by leaves of palm trees

Conclusion

Surface adsorption is an effective and economical method compared to other methods of adsorption of heavy metals such as electrodialysis, chemical methods, extraction with solvent, membrane filtration and other methods that are expensive and complex and require expensive equipment. This method has a high flexibility in design and operation. Surface adsorption is a low-cost method, and the adsorbents used in this method are available and inexpensive. Surface adsorption method has a simple and convenient technology, the adsorption capacity of the adsorbents is high, and it performs separation operation in a wide range of pH. Agricultural, biological and livestock waste can be used as adsorbents. In Iran, date palm is one of the most important agricultural products. The palm is an evergreen tree, and after 3 to 7 years, its old leaves dry out and must be removed and pruned. Every year, a lot of waste is produced in the palm groves, which is the result of the operation of removing dry leaves and petioles, as well as separating tree fibers. In each palm tree, depending on the palm groves conditions and cultivar, about 15 to 25 leaves are dried during the year, each weighing an average of 1.5 to 2.5 kg. The generalization of this amount of plant residues to several million palm trees in Iran is a large figure. Therefore, in order to manage the productivity of plant residues, it is necessary to conduct research on the optimal use of this natural resource. In this study, the ability of palm leaves as a cheap and available adsorbent to remove lead (II) ions from aqueous solutions was investigated. The results showed that palm leaves were a good adsorbent for the removal of lead (II) ions from aqueous solutions.

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