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Research Article

BIOLOGICAL UPTAKE OF METALS LEAD AND CADMIUM BY MEANS OF ACACIA AND FRAXINUS EXCELSIOR USING CONTINUOUS COLUMN METHOD

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ABSTRACT
In this paper, acacia trees and ash plant have been used as absorbents for the removal of metal ions of lead and cadmium from aqueous in continuous systems. The effects of some factors such as PH and particle size have been studied. PH 5 is optimal for absorption. Thomas model and Yoon and Nelson model are used for comparison with experimental data of break curve. For all adsorbents, both models showed good agreement with the experimental data. The optimum conditions for both the absorbers and for both metals the PH 5, particle size is 0/1 to 30/0 mm. In the optimum conditions for maximum adsorption capacity of acacia and ash respectively is 26/36 and 65/50 mg/g for lead. Similarly, cadmium uptake capacity of the ash in the optimal conditions is 74/18 mg/grams. Lead removal efficiency for locust and ash respectively is 25/46 and 25/50%. Cadmium removal efficiency for Ash is 66/47 %. The results of absorbs with fixed platform show that with increasing the adsorbent weight and PH solution , break time and saturation time increases and with increasing adsorbent particle size, flow rate and initial concentration of the solution, these times are reduced. Both Thomas and Yoon and Nelson model showed good agreement with experimental data, and the correlation coefficient in all phases of the experiment was 90/0> R □ respectively.

Keywords: *Metals, Lead, Cadmium, Biological Absorption*

INTRODUCTION
Water is a condition of life and the vast majority of chemical reactions taking place in aqueous environments. Water changes to wastewater after various uses (domestic, agriculture, industry, etc.). To prevent contamination of water and the environment by these wastewaters, we must adopt some strategies for treatment and reusing it (Panswad *et al.*, 1988).
Pollution is the increase in the amount of each reagent, whether chemical, physical or biological properties that contributes to change in its fundamental role and its properties in its specific applications. Generally, environmental pollution can be divided into several categories: 1- Water resource pollution, 2- pollution of soil, 3- air pollution, 4-light and sound pollution, (in large cities). One of the most important types of pollution is water pollution.
Today, pollution from heavy metals threatens the Earth. Wastewater from industries plays a significant role in polluting the environment as one of the sources of these metals. Heavy metals are elements with atomic weight 54/63 to 59/200 and higher densities of 5 grams per cubic centimeter. Among the conventional methods for the removal of heavy metals from wastewater, chemical deposition as hydroxide or sulfide and ion exchange is are so common. These methods, in addition to the high costs that result in avoiding industry men not to use of such methods, sludge production problems from the chemical sediments have followed as well. Because this problem in the aqueous environment is changed into a new problem in effluent which is not compatible with the environment. Biological uptake involves the use of biomass, which the ligands or functional groups of these materials can have physical or chemical interaction with heavy metals (Veglio and Beolchini, 1997; Volesky and Holan, 1995). In this process, dead and alive adsorbent can be used at two discontinuous (discrete or modular) and continuous (column with a fixed-bed).

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Previous work

In 2005, Vijayaraghavan *et al.*, studied the copper removal from aqueous solution by brown seaweed called Behnam Turbinaria ornata in both discrete and continuous systems. Done tests at different pH in discrete systems showed that the maximum adsorption capacity for copper is 06/47 mg per g at pH 6 based on Langmuir model (Vijayaraghavan *et al.*, 2005). In 2010, Muhamad *et al.*, studied the absorption of cadmium and copper ions by wheat crust. In this study, packed bed column has been used. In continuous system, the effects of air pressure such as flow rate, bed height were measured. Thomas model, Yun and Nelson and Adams Buhart has been used for continuous system in order to explain the adsorption process. According to the results, Thomas model was more consistent with the experimental data (Muhamad *et al.*, 2010).

MATERIALS AND METHODS

The leaves used in the study include locust and ash. Heavy metal cations in our study are lead (Pb^+) and cadmium (Cd^+). Solutions used in this study are prepared by nitrate salts to prepare the synthetic wastewater of these metals (all with reagent grade, made by Merk). In this study, the packed bed column was used.

Experimental study and biological absorption theory in column

Evaluation of the failure curve

Packed bed column performance usually is expressed by the failure curve. Other name for the failure curve is the concentration-time profile. Failure curve is a curve that ratio of C_{ad} (absorbed concentration) which is equal to(C, Concentrations output _C_0(input concentration) or $\left(\frac{C}{C_0}\right)$in terms of time (min) or the output volume (mL). Concentrations are in terms of milligrams per liter (Guibal *et al.*, 1995).

An experimental study of the failure curve

Area under the curve of failure (A) is calculated through the integration of absorbed concentration curve (C_{ad}) in terms of time. q_{total} Maximum capacity of the column (mg) in the concentration and flow rate was calculated from the equation (2-1) (Gulensoy *et al.*, 1984; Aksu and Gönen, 2004).

$$q_{total} = \frac{Q\,A}{1000} = \frac{Q}{1000}\int_{t=0}^{t=t_{total}} C_{ad}\,dt \qquad (1\text{-}2)$$

(W_{total}) the total amount of transferred metal to the column (mg) is calculated from following equation:

(2-2) $W_{total} = \frac{C_0 Q\, t_{total}}{1000}$

Output of metal ion removal is the relation of q_{total} to W_{total}:

(3-2) $Y = \left(\frac{q_{total}}{W_{total}}\right) \times 100$

By the use of mentioned relationships, we can determine the experimental parameters like (%Y, W_{total} ، $q_{eq(exp)}$، t_b و t_e)

Checking kinetic model of Thomas

In 1944 Thomas presented kinetic model for uptake of ions by zeolite (Thomas, 1944). Thomas model is one of the most reliable and authentic models to predict the adsorption process in the column. The absorptive capacity of the column and also Thomas rate constant (kinetic coefficient) can be calculated by this model. Thomas model is expressed as follows (Naddafi *et al.*, 2007).

$$\frac{C}{C_0} = \frac{1}{1 + \exp\left[{}^{K_{Th}}\!/_{Q}\left(q_{0(Th)}x - C_0 V_{eff}\right)\right]}$$

That K_{Th} is the rate constant of THOMAS in terms of (mL mg^{-1} min^{-1}), $q_{0(Th)}$ is the absorption capacity of columns per (mg g^{-1}), x is the amount of adsorbent in column (g), V_{eff} is the output volume (mL), C_0 is the initial concentration of metal ion solution (mg L^{-1}) and Q is the volumetric flow rate (mL min^{-1}). The linear form of Thomas equation is as follows.

$$\ln\left(\frac{C_0}{C}-1\right)=\frac{K_{Th}q_{0(Th)}x}{Q}-\frac{K_{Th}C_0}{Q}V_{eff}$$

From the slope and intercept of the diagram $\ln\left(\frac{C_0}{C}-1\right)$ against V_{eff}, we can calculate the K_{Th} and $q_{0\ (Th)}$ using linear regression analysis.

Study of kinetic model of Yoon and Nelson

This model is based on the assumption that the rate of decline in the probability of absorption per adsorbent molecule is proportional to adsorption probability of the adsorbent. This model has less complexity than other models. This model also requires no information about the properties of adsorbed material, type of adsorbent and physical properties of the bed. Yoon and Nelson equation for single-component is expressed as follows (Yoon and Nelson, 1988).

$$\frac{C}{C_0}=\frac{1}{1+\exp[K_{YN}(\tau-t)]} \qquad (7\text{-}2)$$

That (min^{-1}) K_{YN} is the constant rate of Yoon and Nelson,(min) τ is the time in which 50% of failure takes place $\left(\frac{C}{C_0}=0/5\right)$.t(min) is the time of sampling. The linear form of Yoon and Nelson equation is as follows:

$$\ln\frac{C}{C_0-C}=K_{YN}t-\tau K_{YN} \qquad (8\text{-}2)$$

From the slope and intercept of the diagram $\ln\frac{C}{C_0-C}$ versus t, we can calculate τ and K_{YN}. As noted above, t = τ is the time in which 50 percent of absorb takes place in the curve failure, so in time $t = 2\tau$, column bed is saturated. Using the following relations $q_{0(YN)}$ we can calculate the column adsorbent capacity (Ozturk and Kaval, 2005).

$$(9\text{-}2) \qquad q_{0(YN)}=\frac{q_{total}}{X}=\frac{(1/2)(Q/1000)(2\tau)C_0}{X}=\frac{C_0Q\tau}{1000\,X}$$

In both models it is reviewed in $\frac{C}{C_0}$ greater than 0/05 and lower than 0/9

Tests Results

Effect of adsorbent particle size of Zinc

Effect of adsorbent particle size of Zinc on the failure curve of Lead by acacia and ash

The diagram in Figure 2-1 shows the results of experiments on the effect of adsorbent particle size. Also according to the results obtained from Table 2-, the experimental parameters (t_b ‹t_e ‹q_{eq} and Y% were determined.

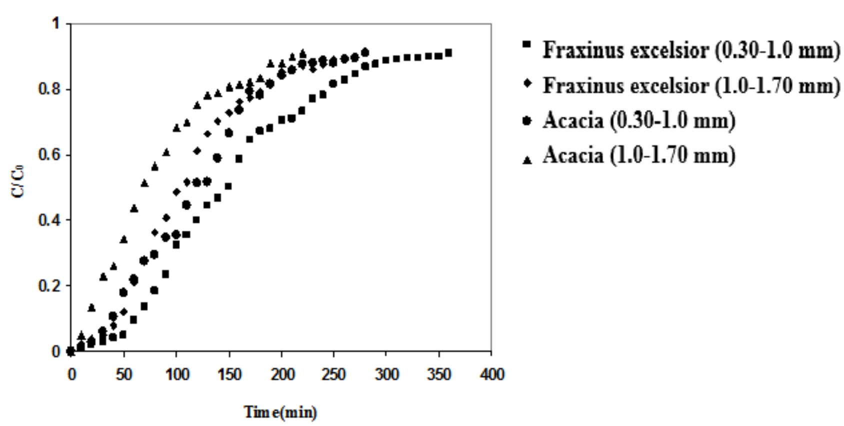

Figure 2-1 Effect of adsorbent particle size on the failure curve of Lead by acacia and ash

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(Concentration 50 milligrams per liter, Speed 5/6 ml/min, pH =5, adsorbent weight equal to 1g)

4.1.2 Effect of adsorbent particle size in parameters of theory of Thomas model and Yoon and Nelson lead by acacia and ash

For Thomas model parameters for the rate constant K_{Th} Thomas constant rate is calculated and$q_{0\ Th}$ the column adsorbent capacity and for model Yoon and Nelson K_{YN} the rate constant, τ the time in which 50% failure take place and $q_{0(Yn)}$adsorption capacity of the column. Figures 2-3 and Figure 2-4 shows the diagram related to test results. Theoretical parameters listed are given in Tables 2-2 and 2-3.

According to the results, particle size (0/1-30/0 mm) was selected as the optimal particle size for both adsorbents for further testing.

The effect of pH input solution on adsorption of lead

Effect of pH of solution in the input curve of lead failure by acacia and ash

In this section, the effect of pH on adsorption of lead intake by adsorbents was studied in the locust and ash. In order to determine the optimum pH amount for lead adsorbent by mentioned adsorbents, adsorbent particle size (0/1-30/0 mm), initial concentration of metal ion solution (50 mg), the weight of the adsorbent (1 g), and speed of flow (6/5 ml min) of the test prove that the only factor to be considered and taken into account the changing is the pH of the input solution (3, 4 and 5). Figure 2-10 shows a diagram related to the test results. According to the results, experimental parameters were defined in Table 2-7.

Table 2.2: Effect of adsorbent particle size in parameters of Thomas model in lead adsorbent for the locust and ash adsorbents

(Concentration 50 Mg/litre, Speed 6/5 ml/min, pH =5 and adsorbent Weight equal to 1 gram)

Adsorbent particle size (mm)	Acacia q_{eq} (Mg g^{-1})	$q_{0(Th)}$ (Mg g^{-1})	K_{Th} (M L mg^{-1} min^{-1})	R^2	Ash q_{eq} (Mg g^{-1})	$q_{0(Th)}$ (Mg g^{-1})	K_{Th} (ML mg^{-1} min^{-1})	R^2
(0/1- 30/0)	26/36	07/36	38/0	9 5/0	65/50	8 9/4 8	29/0	9 3/0
(70 / 1-0/1)	48/25	13/24	41/0	90/0	74/36	18/34	39/0	90/0

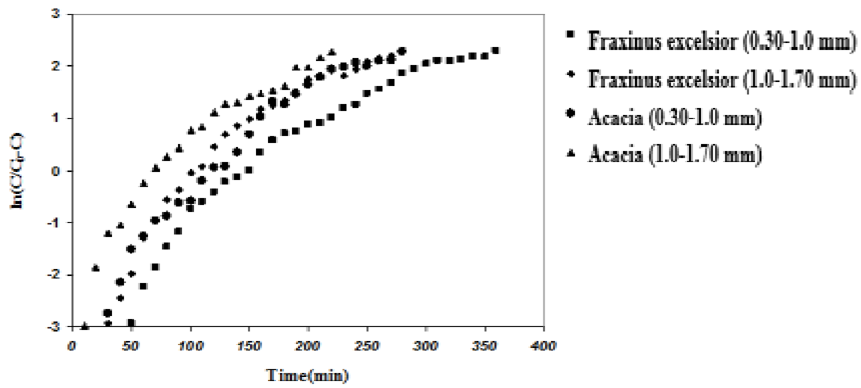

Figure 2-3: Graphs of the effect of sorbent particle size Yoon and Nelson model for lead capture by acacia and ash

(Concentration of 50 Ml/G, Speed 6/5 M Liters per minute, pH =5, adsorbent Weight equal to 1 gram)

Table 2-3: Effect of particle size Yoon and Nelson model of parameters for the uptake of lead absorber in the locust and ash

(Concentration f 50 Mg/liter, Speed 6/5 ml/ min, pH =5, adsorbent Weight equal to 1 gram)

Adsorbent particle size (mm)	Acacia q_{eq} (Mg g^{-1})	(Min)	$q_{0(YN)}$ (Mg g^{-1})	K_{YN} (Min^{-1})	R^2	Ash q_{eq} (Mg g^{-1})	(Min)	$q_{0(YN)}$ (Mg g^{-1})	K_{YN} (Min^{-1})	R^2
(0/1- 30/0)	26/36	128	84/35	01 6/0	9 5/0	65/50	1 of 74	72/48	01 5/0	9 3/0
(70 / 1-0/1)	48/25	87	36/24	020/0	90/0	74/36	122	16/34	0 to 20/0	90/0

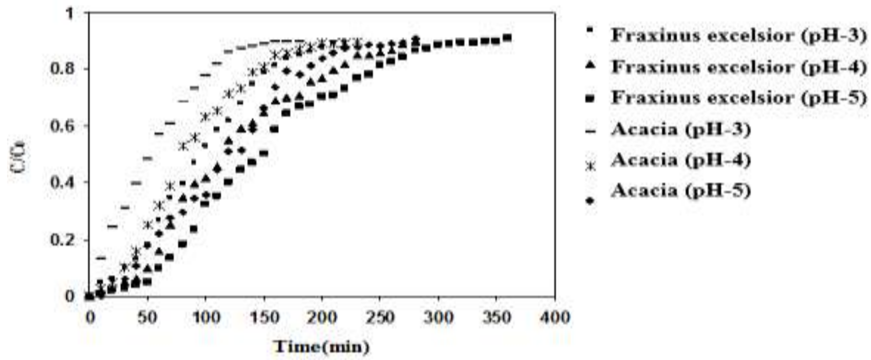

Figure 2-4: graph of the effect of pH on the failure curve Lead by acacia and ash
(Concentration 50 milligrams per liter, Speed 6/5 mL min, particle size (0/1-30/0 mm) and adsorbent weighs 1 gram)

Table 2-4: Effect of pH in the experimental parameters of lead failure curve by acacia and ash
(Concentration 50 mg/l, Speed 6/5 ml/min, particle size (0/1-30/0 mm) and adsorbent weighs 1 gram)

pH	**Acacia**					**Ash**				
	Failure time (t_b) (Min)	Saturation time (t_e) (Min)	W_{total} (Mg)	q_{eq} (Mg g^{-1})	Y (%)	Failure time (t_b) (Min)	Saturation time (t_e) (Min)	W_{total} (Mg)	q_{eq} (Mg g^{-1})	Y (%)
3	8	180	4/50	69/17	10/35	10	220	6/61	59/29	03/48
4	20	230	**4/64**	36/25	37/39	30	280	4/78	92/38	64/49
5	30	280	4/78	26/36	25/46	50	360	8/100	65/50	25/50

Conclusions

In previous studies, the method of biological absorption has been done by Batch that consist some limitations such as low volume of wastewater for treatment, low speed of work and lack of industrial application. In this study, the continuous method of (Column) was studied that in addition to removing the above restrictions has some benefits, such as simplicity, being automatic, achieving a high degree of purity, adsorption desorption cycles, the possibility to achieve high absorption capacity at very low concentrations of heavy metals and also the use of columns to use for practical purposes in industry. Studies have shown that these adsorbents have a suitable capacity in removal of lead and cadmium from wastewaters. By comparing the two metal adsorption capacity and efficiency, respectively Cd >Pb was obtained. Results showed that the ash adsorbent has a greater capacity for Lead than Acacia. Also, lead absorption capacity by ash is higher than metal cadmium. Reviews show that in the continuous system, parameters like flow rate, adsorbent particle size, the weight of adsorbent, initial concentration of dissolved metal ions and input solution of pH are the factors affecting the uptake of bio packed bed column. In the research, Thomas and Nelson model were used in linear mode for the interpretation of obtained experimental data. In most cases, both models have good agreement with experimental data. Biological absorption method is cheap and effective way to remove heavy metals.

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