



Batch Sorption of Copper (II) Ions from Simulated Aqueous Solution by Banana Peel

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Abstract

This research presents the possibility of using banana peel (arising from agricultural production waste) as biosorbent for removal of copper from simulated aqueous solution. Batch sorption experiments were performed as a function of pH, sorbent dose, and contact time. The optimal pH value of Copper (II) removal by banana peel was 6. The amount of sorbed metal ions was calculated as 52.632 mg/g. Sorption kinetic data were tested using pseudo-first order, and pseudo-second order models. Kinetic studies showed that the sorption followed a pseudo second order reaction due to the high correlation coefficient and the agreement between the experimental and calculated values of q_e . Thermodynamic parameters such as enthalpy change (ΔH°), entropy change (ΔS°) and Gibbs free energy change (ΔG°) were also investigated. Free energy change showed that biosorption of Cu (II) was spontaneous and nature endothermic at all studied temperatures (25–45 °C).

Keywords: Sorption capacity, biosorption kinetic, Copper removal, Banana peel sorbent.

1. Introduction

Heavy metals located in the wastewater originated mainly from electroplating, metal finishing, metallurgical, chemical manufacturing, mining and battery manufacturing processes in large amounts. Heavy metals are known toxic elements and their discharge into streams of water reason for harmful effects on human health and the environment [1]. As they are not biodegradable, they begin to accumulate in living organisms. Thereby they cause many diseases and disturbance. Addition, because of their toxicity, the existence of metals in unreasonable quantities interferes with profitable use of water [1]. Thus, it is imperative to find a way to get rid of heavy metals in wastewaters before being discharged into the environment.

Copper is one of the metallic elements that exist in soil at a concentration of approximately 50 parts per million (ppm). It is existent in all animals and plants and is a necessary nutrient for

humans and animals in small amounts. The main sources of copper in the environmental involve the mining, smelting and refining of copper, manufacture producing products from copper such as wire, pipes and sheet metal, and fossil fuel combustion. Water pipes are many times made of copper and bath equipment may be made from brass and bronze alloys that include copper. The major source of copper in drinking water occurs as a result of the leaching of copper from pipes and bath fixtures because of acidic water. Blue-green stains remaining on bath fixtures are a sign of the existence of copper in water [2]. Other sources of copper in environment involve agricultural use against plant diseases and treatments applied to water bodies to get rid of algae [3]. From all above, the concentration of copper should be reduced to reasonable standard via treating the effluent from industrial wastewater before throwing to the river. It is possible to remove these metals by using various techniques, such as precipitation, membrane

filtration, ion exchange, sorptive flotation and adsorption.

Adsorption technology has the potentiality to remove, recover and recycle of metals from wastewater. Activated carbon is an adsorbent which is used extensively to remove heavy metals because it has a high specific surface area and attractiveness of metals. Despite this, due to it being high cost and technical preparation lead to a development of new adsorbents with similar characteristics there is a study to find an alternative adsorbent which have a low-cost and high capacity [4]. Many researchers have examined low-cost adsorbents produced from agro-wastes for removal of heavy metals (Kumar [1]; Hossain [4]; Husoon [5]; Aloma [6]; Torab et al., 2013 [7]; Nour [8], and others).

In this paper an agricultural waste such as banana peel has been utilized as bio-sorbents for the removal of copper ion by adsorption process. Banana plants affiliated to the family Musaceae. They are ploughed mostly for their fruit and to a lesser extent for the produce of fiber also as decorative plants. These plants are usually tall and strong, it is one of the most consumed fruits in the world and useless peels, due to this reason, makes one of the main agriculture-waste problems [9]. Previous studies have shown that many tons of banana peels are generated daily in market places and domestic waste that cause environmental inconvenience. For this, banana peels have been selected as adsorbents for copper ions from industrial wastewaters. Adsorption isotherms and kinetics were investigated and various adsorption models were examined to estimate the experimental data and to clarify the possibility of adsorption mechanism.

2. Materials and Methods

2.1. Chemicals and Materials

Synthetic wastewater which used in this experiment containing the required concentration of copper was prepared by dissolving the calculated amount of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ in distilled water. The mass of these metals required to achieve the required concentration was calculated according to equation (1) assuming complete dissociation:

$$W = V \times C_i \times \frac{M.wt}{At.wt} \quad \dots (1)$$

Where:

W= weight of heavy metal salt (mg).

V= volume of solution (L).

C_i = initial concentration of metal ion in solution (mg/L).

M.wt= molecular weight of metal salt (g/mole).

At.wt= atomic weight of metal ion (g/mole).

2.2. Apparatus

Atomic Adsorption Spectrometer (AAS) (GBC 933 plus, Australia) was used to measure the concentrations of soluble copper. Batch experiments for optimization of process parameters were carried out in 250 ml round bottom flasks with working volume of 100 ml at 200 rpm in an incubator cum orbital shaker (Heidolph, No.549-59000-0-0, Germany).

2.3. Preparation of Sorbent

Banana peel was used as a biosorbent for the removal of copper ions. It was collected from local juice market and washed with distilled water to remove surface impurities and dried at 100°C for 24 hours in the oven to remove the moisture content. The dried peel was milled and sieved, the particles sizes ≤ 0.5 mm were chosen for this study.

2.4. Sorption Experiments

Batch sorption experiments were carried out to detect the optimum pH, amount of dosages of adsorbent, contact time and equilibrium and kinetics isotherms. Effects of all factors were identified with keeping other variables constant. In the experiments 100 ml of synthetic solutions containing 50 mg/L of copper ions have been added into flask with various amounts of banana peels varying between 0.05 and 0.5g. pH adjustments were made by using 0.1N hydrochloric acid and 0.1N sodium hydroxide. Solution was stirred at 200 rpm at a predetermined period of time. At the end of agitation time sorbents were filtered (Whatman 70mm filter paper) and metal contents of solution were analyzed by AAS.

3. Results and Discussion

3.1. FTIR Analysis of Banana Peel

The banana peel has been characterized by FTIR spectral analysis; it is requisite to determine the functional groups of the adsorbent, which are responsible for the adsorption of metal ions [10]. The functional groups were identified in Fig.1.

From these figures the spectrum pattern of loaded banana peel appeared changes in the peak absorption compared with unloaded banana peel resulting from the adsorption process. The role of each functional group in this process is summarized in Table 1.

3.2. Effect of pH

process. So as to study the effect of the pH The pH is an important parameter that affects the sorption on removal of copper by banana peels, 100 ml of 50 mg/l metal solutions were used. Experiments were carried out in the pH range 2-8. Add a constant amount of peels to all flasks; solutions were agitated for 2h at 200 rpm speed. Effect of pH on removal efficiency is shown in Fig. 2. The percentage sorption increases with increase in pH. The minimum sorption was observed at low pH. This behavior may be

referred to the existence of higher concentration and higher mobility of H^+ ions favored sorption compared to $M(II)$ while the acidic medium due to high solubility and ionization of metal ions. The surface of the adsorbent becomes more positively charged at high H^+ concentration such that the attraction between adsorbents and metal cations is lowered. Into reverse with increase in pH the negatively charged surface area becomes more thus facilitate greater metal removal and then at very high pH also the percentage removal decreases [11]. The maximum sorption was observed at pH 6 which might be because of partial hydrolysis of metal ions. More increase in pH i.e., above 8 of the solution causes precipitation of metal ions on the surface of the adsorbent by nucleation [10]. To obtain high extraction efficiency without metal hydroxide precipitation, pH of 6 for Cu (II) was selected for subsequent experiments.

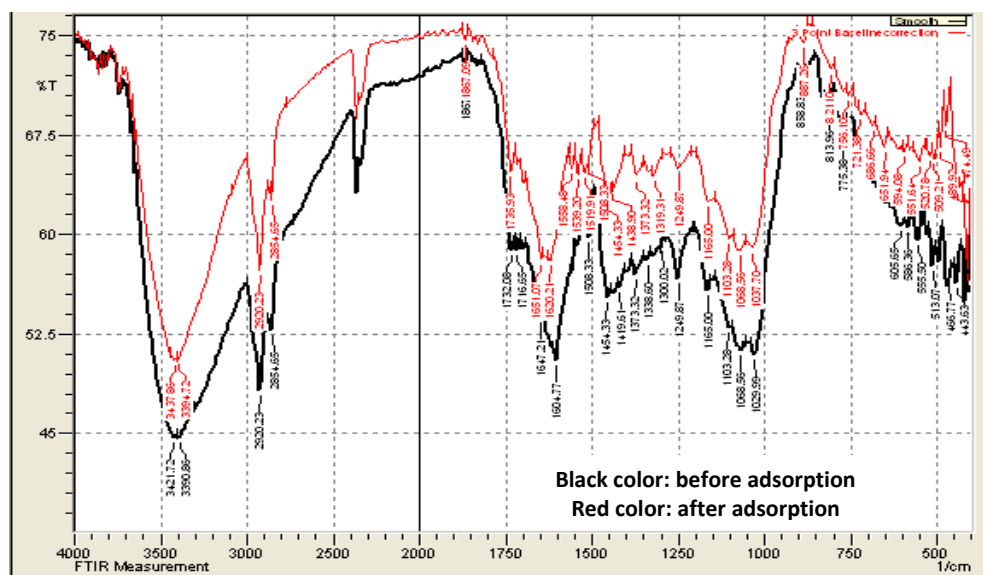


Fig. 1. FTIR before and after biosorption of Cu (II) onto banana peels.

Table.1,
FTIR functional groups responsible for copper adsorption.

Wave number (cm ⁻¹)	Assignment Groups	Wave number (cm ⁻¹) after adsorption
3421.72	Carboxylic acid, Amide, Amine	3437.86
1604.77	Alkane, Amine	1620.21
1508.33	Carboxylic acid	1558.43
1419.61	Carboxylic acid	1454.33
1373.32	Carboxylic acid	1438.90
1338.60	Carboxylic acid	1373.32
1300.02	Carboxylic acid	1319.31
858.83	Alkane	887.26
775.38	Aromatic	796.10
605.65	alkynes	651.94
586.36	alkyl halides	551.64
555.50	alkyl halides	520.78

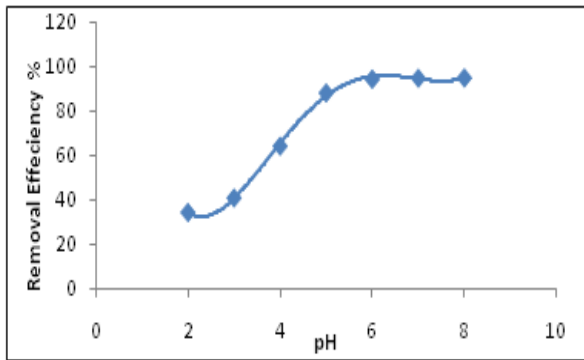


Fig. 2. Effect of pH on of Cu (II) uptake using banana peel.

3.3. Effect of Banana Peels Dose

The sorbent amount is also one of the important parameters to obtain quantitative uptake of metal ion. The effect of dose was studied by changing the amount of banana peels (0.05-0.5g), whereas the other parameters such as pH 6, initial metal concentration 50 mg/l, agitating time 2h and stirring speed 200 rpm stayed constant. Sorption of metal ions was increased as the sorbent amount increased. The results were expected because for a fixed initial metal concentration, increasing adsorbent amount provides greater surface area or sorption site [12] [13]. The higher removal efficiency was achieved by using 0.45 g/100 ml sorbent dosages (Figure 3).

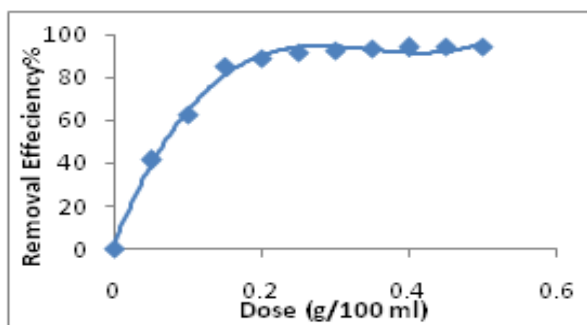


Fig. 3. Effect of sorbent dose on of Cu (II) uptake using banana peel.

3.4. Effect of Contact Time

The removal of copper ions increases with time and reaches saturation during 120 min. the effect of contact time was studied by taking 0.45g sorbent with 100 ml of a metal solution in different flasks. The flasks were vibrating at

various time intervals. Figure 4 shows the effect of contact time on the removal efficiency of copper ions, the removal of sorbate is rapid, but it gradually reduces as time goes until it reaches equilibrium. Rate of percent metal removal is higher at the beginning because of a larger surface area of the adsorbent being available for the adsorption of the metals. The binding site was shortly become limited and the remaining vacant surface sites are hard to be taken by copper ions due to the forming of repulsive forces between the copper on the solid surface and the liquid phase [4] [14].

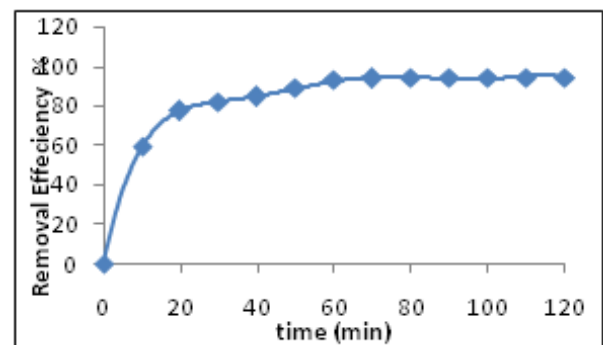


Fig. 4. Effect of time on of Cu (II) uptake using banana peel.

3.5. Sorption Isotherms

Sorption isotherms are very significant tools for analyzing the sorption process. Langmuir and Freundlich isotherms models are generally used to study the sorption process. The model parameters can be more explanation, preparation understanding on the mechanism of sorption, surface properties and an affinity of the adsorbent [15].

3.5.1. Langmuir Isotherm Model

Langmuir sorption isotherm models the monolayer coverage of sorption surfaces and supposed that sorption happens on a structurally homogenous adsorbent and all the sorption sites are strongly comparable. The mono-saturated layer curve can be represented by the expression [16]:

$$q_e = \frac{q_m b C_e}{(1 + b C_e)} \quad (\text{non linear form}) \quad \dots (2)$$

$$\frac{C_e}{q_e} = \frac{1}{b q_m} + \frac{C_e}{q_m} \quad (\text{linear form}) \quad \dots (3)$$

Where: q_e is the sorbed metal ions on the biomass (mg/g), q_m is the maximum sorption capacity for monolayer coverage (mg/g), b is the constant related to the affinity of the binding site (L/mg), and C_e is metal ions concentration in the solution at equilibrium (mg /L).The Langmuir isotherm is used mostly to describe the adsorption isotherm which is limited by the supposition of uniform energies of adsorption on the surface of adsorbent. Constant for the adsorbate-adsorbent equilibrium and the capacity of monolayer b and q_e were identified from the slope and intercept of the Langmuir plot (Figure 5).

3.5.2. Freundlich Isotherm Model

Freundlich equation is derived to model the multilayer sorption and for the sorption on heterogeneous surfaces. The Freundlich isotherm theory supposes that the percentage of the amount of solute adsorbed onto a given mass of sorbent to the concentration of the solute in the solution is not constant at different concentrations. The equation may be written as [16] [17]:

$$q_e = KC_e^{1/n} \quad \dots (4)$$

The linear form of Eq. (4) is:

$$\log q_e = \log K + \frac{1}{n} \log C_e \quad \dots (5)$$

Where: K is the constant indicative of the relative adsorption capacity of the adsorbent (mg/g). $1/n$ is the constant indicative of the intensity of the adsorption. Both K and n are constants, being an indication of the extent of adsorption and the degree of non-linearity between solution and concentration, respectively. The linear Freundlich plots are obtained by

plotting $\log q_e$ versus $\log C_e$ from which the adsorption coefficients. The linear Freundlich plots are acquired by plotting $\log q_e$ versus $\log C_e$ from which the adsorption coefficients could be estimated (Figure 6). All constants identified from Langmuir and Freundlich isotherms are given in Table 2.

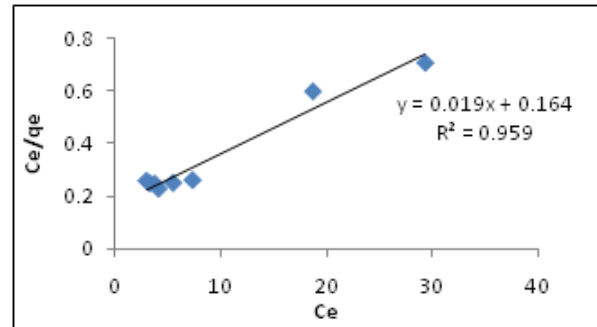


Fig. 5. Langmuir plot of Cu (II) sorption on banana peel (pH 6; rpm 200).

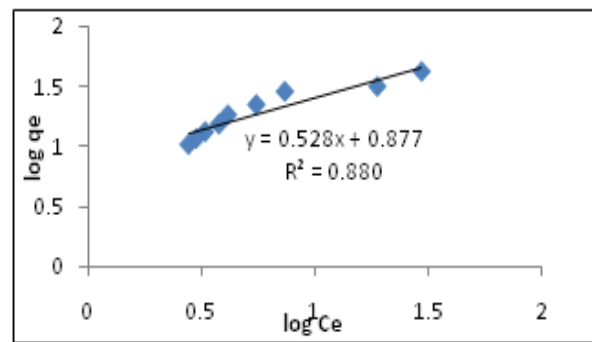


Fig. 6. Freundlich plot of Cu (II) on banana peel (pH 6; rpm 200).

Table.2,

Parameters of the Langmuir, Freundlich isotherms for the sorption of Cu (II) onto banana peel.

Langmuir model	Copper (II)	Freundlich model	Copper (II)
R^2	0.959	R^2	0.880
q_m (mg/g)	52.632	$1/n$	0.528
b (l/mg)	0.116	K (mg/g)(l/mg) ^{1/n}	7.534

The Langmuir model described the sorption data slightly better than Freundlich model according to the value of correlation coefficient (R^2).

3.6. Kinetics of Sorption

The kinetics clarifies the solute uptake rate which in turn controls the residence time of sorbate uptake at the solid–solution interface. Thus, it is essential to be able to predict the rate at

which sorbate is removed from aqueous solutions for design suitable sorption treatment processes [18]. The sorption kinetics, therefore, constitute a main principle in the definition of the interest of sorption processes. Two kinetic models which are the Lagergren-first order, pseudo-second-order used to describe the biosorption kinetics. The Lagergren- pseudo first order kinetic model equation, [19], is:

$$\ln(q_{eq} - q_t) = \ln q_e - k_1 t \quad \dots(6)$$

The Pseudo-second order reaction kinetic can be expressed as, [20]:

$$\frac{t}{q_t} = \left(\frac{1}{k_2 q_e^2} + \frac{t}{q_e} \right) \dots (7)$$

Where q_{eq} is the amount of metal sorbed at equilibrium (mg/g); q_t is the amount of metal sorbed at time t (mg/g); and k_1 is the rate constant of the first-order adsorption (1/min) and k_2 is the pseudo-second order rate constant (g/mg h). The slopes and intercept of $\ln (q_e - q_t)$ versus t plot (Fig.7a) were used to describe the pseudo first order rate constants (k_1) and q_e . A plot of t/q_t versus t (Fig.7b) was used to describe the pseudo-second-order rate. The values of correlation coefficient (R^2) indicate a better fit of pseudo-second- order model with the experimental data compared to pseudo- first- order model. The values of q_e calculated from the second order kinetic model Compatibility well with the experimental values. Results placed in Table 3 along with correlation coefficients (R^2) values.

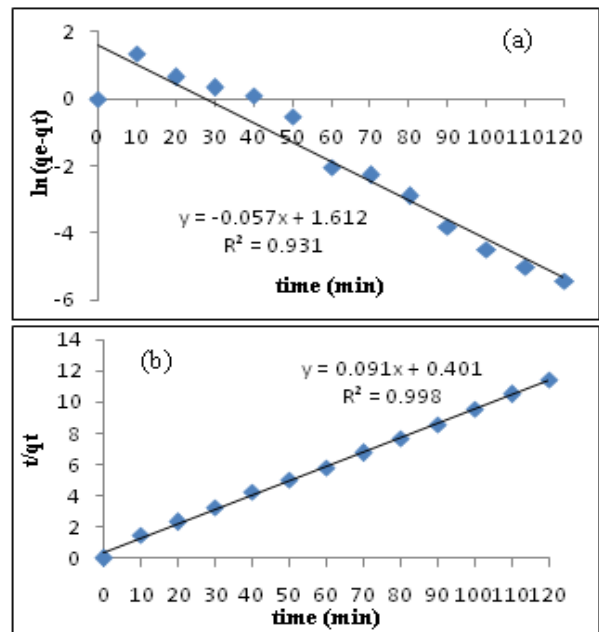


Fig. 7. (a) Pseudo-first-order (b) Pseudo-second-order plots of Cu (II) on banana peel.

Table 3,

Comparison of sorption rate constants, experimental and calculated q_e values for the pseudo-first- and -second-order reaction kinetics for component systems.

Metal	q_e experimental mg/g	Pseudo-first-order			Pseudo-second-order		
		k_1 1/min	q_e calculated mg/g	R^2	k_2 g/mg.min	q_e calculated mg/g	R^2
Copper	10.50	0.057	5.013	0.931	0.021	10.989	0.998

The values of correlation coefficient (R^2) suggest that a better fit of pseudo- second- order model with the experimental data compared to pseudo- first- order model. The values of q_e calculated from the second order kinetic model accepted very well with the experimental values, thus, the second-order model can be applied for Cu (II) ions sorption process.

3.7. Effect of Temperature and Thermodynamic Parameters

The effect of temperature on the adsorption of copper on banana peel was studied by carried out experiments for 50 mg/L of initial metals ion concentrations at 298, 308 and 318 K (25, 35, and 45 °C). By increasing the temperature percentage removal of heavy metals increased, which means that adsorption process was endothermic in nature. The results are plotted in Fig.8. The thermodynamic parameters were calculated by the following equations [21] [22]:

$$\ln K_d = \left(\frac{\Delta S^\circ}{R} \right) - \left(\frac{\Delta H^\circ}{RT} \right) \dots (8)$$

$$\Delta G^\circ = \Delta H^\circ - \Delta S^\circ T \dots (9)$$

Where K_d is the distribution coefficient; ΔH , ΔS , and T the enthalpy, entropy, and temperature in Kelvin, respectively; R is the gas constant (8.314 J/mol K) and Gibbs free energy change ΔG^0 . The values of distribution coefficient and thermodynamic parameters are presented in Tables 4 and 5, respectively.

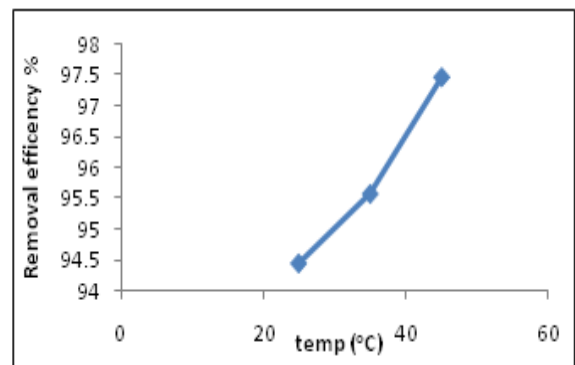


Fig. 8. Effect of temperature on the removal of Cu (II) using banana peel.

Table.4,
The distribution coefficients at different temperature.

Metal	k_d			R^2
	298k	308k	318k	
Copper(II)	17.05054	21.62443	38.37008	0.937

Table.5,
The thermodynamic parameters for the adsorption of Cu (II) ions on banana peel.

Metal	ΔH (J/mol)	ΔS (J/mol K)	ΔG (kJ/mol)		
			298K	308K	318K
Copper(II)	31.7927	-129.782	38.7067	40.00451	41.30232

The results indicated that the ΔG° values are positive and increased in their absolute values with temperature. This result suggested that a high temperature is favored for the adsorption of heavy metals on Banana peel, displayed a spontaneous adsorption process. The values of heat of adsorption, ΔH is positive for metals ion, showed that the adsorption process of heavy metals on banana peel was endothermic; negative ΔS suggested that the adsorption was enthalpy driven and spontaneous in nature [23].

4. Conclusion

A simple treatment procedure was suggested to remove copper ions by biosorption process using banana peels. Usage of agriculture (biological) waste (banana peels) for metal sorption has a lot of advantages; available and reasonable cost.

Experimental results were evaluated with Langmuir and Freundlich; the results show Langmuir isotherm fit the sorption equilibrium data better than the Freundlich. Pseudo-second-order reaction kinetic has provided a realistic description of removal of Cu (II) with closer experimental and calculated values of q_e . Also correlation coefficients are higher in pseudo-second-order kinetics.

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امتزاز ايونات النحاس بتجارب الدفعية من محلول المياه المحاكاة بواسطة قشر الموز

لهيب فيصل محيسن

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الخلاصة

يعرض هذا البحث إمكانية استخدام قشر الموز (الناجمة عن النفايات والمنتجات الزراعية) كوسط ماز لإزالة النحاس من محلول مائي المحاكاة . وأجريت التجارب الامتزاز الدفعة لأيجاد افضل قيمة لمعامل الدالة الحامضية، وكمية المواد المازة، ووقت التماس . وبلغت افضل قيمة الرقم الهيدروجيني لإزالة النحاس (II) بواسطة قشر الموز 6. تم حساب السعة القصوى للامتزازا 52,632 ملغم /غم. وتم اختبار البيانات الحركية من الدرجة الاولى و الثانية ، تبين انها ملائمة مع الدرجة الثانية اكثر من ملائمتها مع الدرجة الاولى حسب قيمة معامل الارتباط (R^2) و توافق القيم الناتجة من التجارب العملية مع القيم النظرية (q_e)، وقد تم اختبار المعلمات الحرارية مثل التغير في المحتوى الحراري (ΔH°)، تغير الانتروبي (ΔS) والتغير في الطاقة الحرة (ΔG). وكانت عمليات الامتزاز تلقائية و ذات طابع ماص للحرارة على وفق النتائج الحرارية.