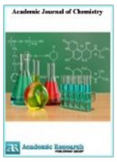
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Cadmium Removal from Synthetic Aqueous Solutions by Adsorption on Peanut Shells: Kinetic and Thermodynamic Studies

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Abstract

Peanut shells were used in this study as an adsorbent carrier in the treatment of wastewater containing a trace metal element (TME), namely cadmium, a pollutant with adverse effects on health and the environment. The influences of contact time between adsorbent and adsorbate, initial cadmium concentration, adsorbent mass, reaction medium temperature and solution pH were evaluated. To determine the optimum parameters, pseudo-first-order and pseudo-second-order kinetic models were applied to the experimental results of the contact time study. Similarly, experimental results relating to the influence of concentration and temperature were modeled. Modeling of the data from the adsorbate-adsorbent contact time study showed that the kinetics of the cadmium adsorption reaction are well described by the pseudo-second-order kinetic model. The isotherm of the adsorption mechanism of cadmium ions (Cd^{2+}) by peanut shells is perfectly described by the Freundlich equation. Thermodynamic studies using temperature variation have shown that adsorption is spontaneous and endothermic. This study led to a low entropy during cadmium adsorption. The best adsorption capacity was obtained at low pH = 5, i.e. favorable adsorption in acidic media.

Keywords: Cadmium; Metallic trace element; Peanut shell; Pollutants.

1. Introduction

The increased establishment and intensification of industrial activities (Alban Tano, 2009) in Côte d'Ivoire has led to a strong demand for labor. As most industries are located in urban areas, the urban population is growing. This growth is the main cause of the exponential expansion of automobile transport. This sector generates numerous organic and inorganic substances, such as trace metals (TMEs), which are released directly into the environment.

The presence of trace metals (TMEs) in these discharges generally results from the waste streams of various activities such as mining, metal plating, the electroplating industry, the paper industry and leather manufacture, as well as from the use of pesticides [1]. Although some TMEs, such as zinc (Zn), copper (Cu) and nickel (Ni), are necessary for the human organism, they are toxic above a certain concentration limit [2]. However, other TMEs such as cadmium (Cd), lead (Pb) and chromium (Cr) are very harmful to humans, whatever their concentration [3-5]. Depending on their concentration, TMEs are likely to cause harm to humans, ranging from simple discomfort to serious illnesses such as cardiovascular complications and even death [6]. In ecosystems, they can lead to trophic chain dysfunction [7]. Cadmium, the target TME in this study, is non-biodegradable and passes easily through the food chain. In humans, cadmium ingestion has no adverse effects at 0.05 mg/L, but may cause nausea and vomiting at concentrations of 15 mg/L. Above 15 mg/L, nausea and vomiting may occur. Above 15 mg/L, symptoms of renal dysfunction, hypertension and anemia have been observed [7]. It is therefore essential to remove cadmium from effluents before releasing them into the environment.

Generally, there is a number of different technologies for eliminating TMEs or reducing their content in contaminated water. These include chemical precipitation, electrocoagulation, ion exchange, electrolytic reduction, solvent extraction, biological treatment and evaporative recovery [8-10]. However, the high cost of implementation equipment, difficulties in separating by-products and the low efficiency of these technologies limit their fields of application. The adsorption removal method represents a cost-effective alternative for the treatment of environmental TMEs. Adsorption is a fluid treatment process in which molecules or ions, called adsorbates, attach themselves to the surface of a solid, called an adsorbent. This process is easy to implement, less costly and requires little equipment. Depending on the type of pollutant, the quality of the adsorbent is decisive for efficient elimination.

Several adsorbents have been used for TMEs removal. In recent studies, scientists are increasingly turning to the use of local agricultural waste as an adsorbent medium. Adsorbents such as activated carbon, banana peels, date pits, cashew shells and coconut husks [11-13] have been used.

The aim of the present study is to evaluate the adsorption capacity of cadmium on peanut shells. The effects of physico-chemical parameters such as contact time between adsorbent and adsorbate, initial cadmium concentration, adsorbent mass, reaction medium temperature and solution pH will be evaluated.

2. Materials and Methods

2.1. Description of Peanut Shells

The peanut is a plant with a height between 20 and 90 cm. The leaves are composed of 2 or 3 pairs of membranous, oval leaflets. They have sheathing stipules at their base. Flowers are almost stalkless and appear in the leaf axils, singly or in small groups. The papilionaceous corolla is orange-yellow. The nine stamens are fused into a tube by their threads. The fruit ripens at a depth of 3 to 5 cm. For this reason, the plant requires light, well-drained soil. The fruit is a 3 to 4 cm-long pod, known commercially as a hull, containing seeds, which are externally reticulated and strangulated between the seeds but not partitioned.

Peanut shells are mainly composed of fiber, with a crude fiber content often exceeding 60% dry matter and a lignin content in the order of 6 to 45% dry matter. Peanut shells contain small, variable amounts of protein (on average 7% dry matter) and oil (2% dry matter) [14].

The peanut shells were harvested in the town of Katiola, around 394 km from Abidjan (Côte d'Ivoire). This is a town with no industrial zones and therefore no potential for contamination.

Samples were taken at a peanut shelling site. Harvested peanut shells are transported to the laboratory.

Figure-1. Peanut shells

2.2. Sorting, Grinding, Drying and Sieving

In the laboratory, peanut shells are sorted by hand to remove soil, leaf, dust and insect debris. They are then washed with distilled water. An appropriate quantity of peanut shells is placed in an oven at 60°C for 48 hours to remove all traces of water. The peanut shells are ground and sieved using a sieve shaker (Saulas, Paris, France). The peanut shell grain sizes obtained are arranged in the following four ranges: [125-250 μm], [250-500 μm] and [500-800 μm]. The particle size of ground peanut shells is used [250-500 μm] [15]. Figure 2 below shows dried and ground peanut shells.

Figure-2. Dried, ground peanut shells

2.3. Preparation of Stock Solutions

A mass of 1 g of cadmium sulfate is dissolved in a 1 L flask with distilled water, to form a stock of 1g/L stock solution in a 2 L volumetric flask.

2.4. Influences of Physico-Chemical Parameters

2.4.1. Influence of Contact Time between Adsorbent and Adsorbate

In 250 mL beakers, 50 mL of a 10 mg/L solution, obtained by diluting the cadmium stock solution, is brought into contact with 1g of peanut shell powder. The mixture is stirred at 450 rpm for periods ranging from 10 to 200 min.

2.4.2. Influence of Initial Cadmium Concentration

Solutions with concentrations ranging from 10, 20, 30, 40 to 50 mg/L are obtained by dilution of the cadmium stock solution. To 50 mL of each solution, 1 g of peanut shells are added in 250 mL beakers. The beakers are stirred at 450 rpm for 120 min.

2.4.3. Influence of Peanut Shell Mass

The adsorbent support's mass effect is studied to determine the quantity of cadmium adsorbed as a function of the adsorption sites available. Different quantities of peanut shells (0.5-4g) in diameter are brought into contact with 50 mL of 10 mg/L cadmium solution in 250 mL beakers. The mixture is stirred at 450 rpm for 120 min.

2.4.4. Influence of Temperature

The study of the temperature effect allows the determination of thermodynamic quantities and the influence of temperature on the adsorption process. 1 g of peanut shells are taken and brought into contact with 50 mL of 10 mg/L cadmium solution in 250 mL beakers. The mixture is stirred at various temperatures ranging from 0 to 50°C for 120 min at 450 rpm.

2.4.5. Influence of pH of Cadmium Solutions

Daughter solutions of 10 mg/L cadmium are obtained by dilution of the stock solution. The pH of the solutions is adjusted by adding hydrochloric acid (HCL: 0.1 M) or sodium hydroxide (NaOH: 0.1 M). The pH values of the solutions are set from 3 to 10.

An aliquot of 1 g peanut shell powder is contacted with 50 ml of each cadmium solution in 250 ml beakers. The mixture is stirred at 450 rpm for 120 min.

The cadmium content of each filtrate obtained during the various adsorption tests is measured using iCE 3400 AAS Atomic Absorption Spectrometer.

2.5. Quantity of Adsorbed Cadmium

The quantity of cadmium adsorbed at equilibrium in each influence is determined by the following expression:

$$Q_e = \frac{(C_0 - C_e)V}{m} \tag{1}$$

Where :

Qe (mg/g): is the quantity of adsorbed solute at chemical equilibrium;

V (L): is the volume of synthetic cadmium solution;

m (g): is the mass of peanut shell powder;

C_0 (mg/L): is the initial concentration of cadmium;

C_e (mg/L).: is the concentration of cadmium at equilibrium

2.6. Modeling of Experimental Data

2.6.1. Adsorption Kinetics

The characteristic quantities of the pseudo-order one and pseudo-order two kinetic models are determined using equations 1 and 2 respectively :

$$\ln(Q_e - Q_t) = \ln(Q_e) - k_1 t \tag{2}$$

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e} \tag{3}$$

With :

k_1(min^{-1}): rate constant for pseudo-first-order kinetics;

k_2(($g.mg^{-1}min^{-1}$).): rate constant for pseudo-second-order kinetics

Q_t(mg/g): adsorption capacity at time t;

Q_e(mg/g): adsorption capacity at chemical equilibrium;

t: Time of adsorption process.

2.6.2. Adsorption Isotherm

Langmuir and Freundlich isotherms represented by equations 4 and 5 respectively were used.

$$\frac{C_e}{Q_e} = \frac{1}{Q_{max} K_L} + \frac{C_e}{q_{max}} \tag{4}$$

$$\ln Q_e = \ln K_F + \frac{1}{n} \ln C_e \tag{5}$$

With :

Q_e (mg/g) : amount of solute adsorbed per gram of adsorbent at chemical equilibrium,

C_e (mg/L) : concentration of solute in the solution at chemical equilibrium;

K_F (L/g) : Freundlich constant relating to the sorption capacity of the adsorbent ;
n : Empirical parameter for adsorption intensity.

2.6.3. Thermodynamic Quantities

In adsorption studies, the thermodynamic component is based on the estimation of variations in standard Gibbs free energy (ΔG^o), standard enthalpy (ΔH^o) and standard entropy (ΔS^o).

The relationship between (ΔG^o), (ΔH^o) and (ΔS^o) is given according to the Ellingham approximation equation (Eq. 6):

$$\Delta G^o = \Delta H^o - T\Delta S^o \qquad (6)$$

The standard Gibbs free energy (ΔG^o) having as expression the following relation:

$$\Delta G^o = -RT\ln(K_e) \qquad (7)$$

By combining these two equations, we obtain the following relationship:

$$\ln(K_e) = -\frac{\Delta H^o}{RT} + \frac{\Delta S^o}{R} \qquad (8)$$

The thermodynamic equilibrium constant K_e is equal to the distribution coefficient K_D at adsorption equilibrium (Elmoubarki *et al.*, 2015).

This equality is translated by the following equation:

$$K_e = K_D = \frac{Qe}{Ce} \qquad (9)$$

This gives the following final expression :

$$\ln\left(\frac{Qe}{Ce}\right) = -\frac{\Delta H^o}{RT} + \frac{\Delta S^o}{R} \qquad (10)$$

Where:
R: is the universal perfect gas constant,
T: adsorption temperature,
Q_e (mg/g): quantity of adsorbed solute at chemical equilibrium
C_e (mg/L): solute concentration at chemical equilibrium.

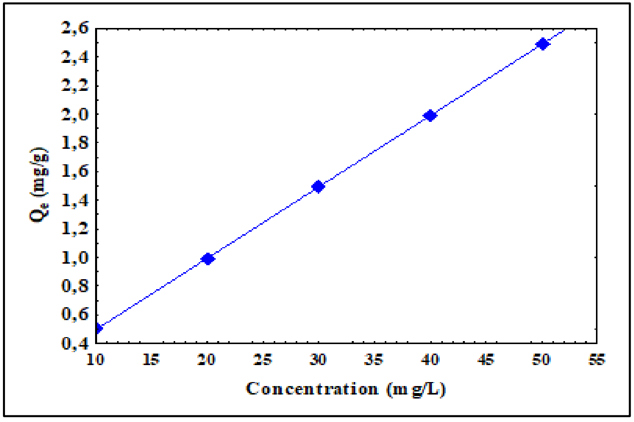
3. Results

3.1. Effect of Cadmium Concentration

3.1.1. Concentration Variation

Figure 3 shows the results of concentration variation in the synthetic cadmium solution.

Figure-3. Effect of initial cadmium concentration



It can be seen that an increase in the initial cadmium concentration leads to an increase in the adsorption capacity of peanut shells. This phenomenon could be explained by the massive and accelerated diffusion of cadmium ions towards peanut shell adsorption sites. This acceleration is due to a driving force generated by the growing concentration gradient. Some authors observed such a phenomenon in their study of nitrate adsorption on activated banana skins [16].

3.1.2. Adsorption Isotherms

In order to determine the nature and mechanism of cadmium adsorption by peanut shells, Freundlich and Langmuir isotherms were applied to the experimental data. The linear regression lines for these different isotherms are shown in Figures 4 and 5.

Figure-4. Freundlich isotherm for cadmium adsorption

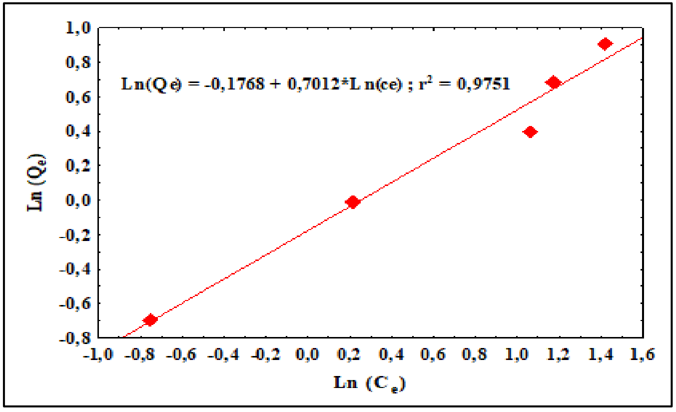
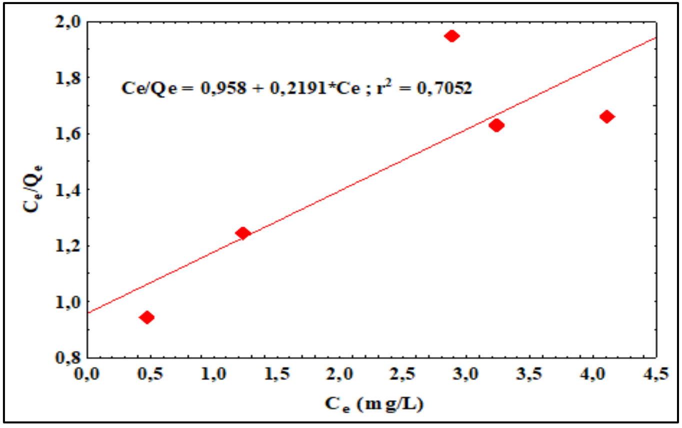


Figure-5. Langmuir isotherm for cadmium adsorption



The values of the various parameters obtained from these isotherms are given in Table 1.

Table-1. Cadmium adsorption parameters according to Langmuir models

Langmuir			**Freunlich**		
Q_{max} (mg/g)	K_L (L/mg)	r^2	1/n	K_F (L/g)	r^2
4,5641	0,2287	0,7052	0,7012	0,3083	0,9751

The adsorption intensity value (1/n) is 0.7012 for the Freundlich isotherm. As this value is less than 1 (1/n = 0.7012 < 1), this indicates that adsorption of cadmium ions (Cd^{2+}) by the adsorbing biomass (peanut shells) is favorable [17].

As for the Langmuir model, the value of the maximum adsorption capacity (Q_{max}) is 4.5641 mg/g. This value indicates the maximum amount of metal adsorbed per unit mass of peanut shell, to form a complete monolayer on the surface at a given concentration of cadmium ions (Cd^{2+}) at equilibrium [18].

The correlation coefficient for cadmium adsorption is 0.7052 for the Langmuir model and 0.9751 for the Freundlich model. A comparison of these coefficients shows that the Freundlich model best describes the adsorption of cadmium ions (Cd^{2+}). The authors [19] have also shown that the Freundlich isotherm best describes the adsorption of Cadmium and lead.

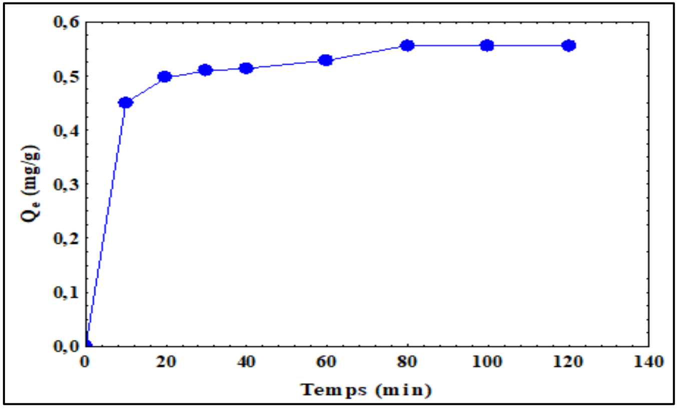
On the other hand, work carried out by a number of authors on the removal of cadmium from wastewater using waste resources has shown that the Langmuir model best describes the adsorption process [20].

3.2. Study of Adsorption Kinetics

3.2.1. Adsorption Equilibrium Time

The results of the study of contact time in solution are shown in Figure 6. Thus, the amount of adsorbed metal increases following the time until a plateau is reached, reflecting the chemical equilibrium of adsorption. The chemical equilibrium of adsorption corresponds to the occupation of most sites by the metal ion and is reached after 80 minutes.

Figure-6. Cadmium adsorption versus time curve



The maximum amount of cadmium adsorbed at chemical equilibrium is 0.5565 mg/g. After chemical equilibrium, the active sites of the peanut shells are saturated and there is no significant binding of metal ions. This result is a constant adsorption capacity of peanut shells over time (Figure 6).

3.2.2. Adsorption Kinetic Models

Experimental kinetic data were applied to the pseudo-first-order and pseudo-second-order kinetic model to characterize metal adsorption kinetics on peanut shells.

The pseudo-first-order and pseudo-second-order kinetic parameters were determined by plotting the respective graphs ln $(Q_e - Q_t) = f(t)$ (Figure 7) and $t/Q_t = f(t)$ (Figure 8).

Figure-7. Pseudo-first-order model of cadmium adsorption

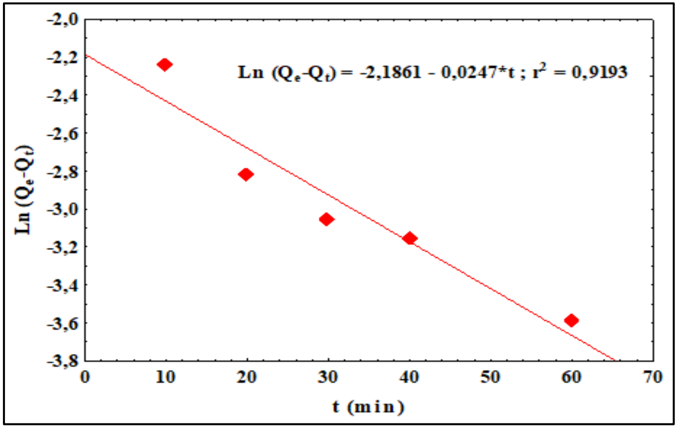
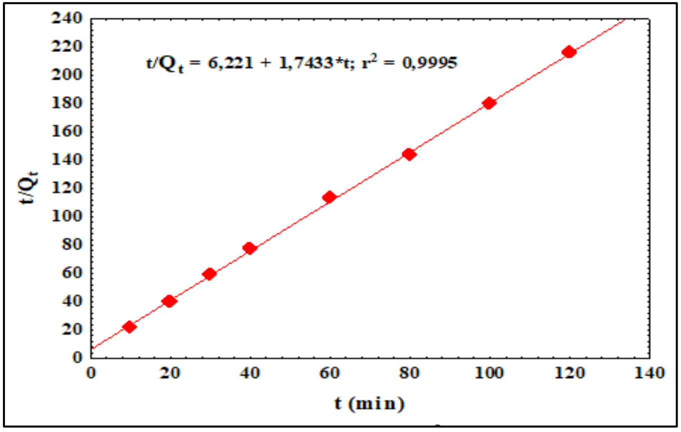


Figure-8. Cadmium adsorption pseudo-second-order model



The results of the parameters (k, Q_e, r^2) obtained are presented in Table 2.

Table-2. Kinetic parameters of Cadmium adsorption on peanut shells

Pseudo-first order				**Pseudo- second order**			
$Q_{e,th}$ (mg/g)	$Q_{e,exp}$ (mg/g)	k_1 (g/mg.min)	r^2	$Q_{e,the}$ (mg/g)	$Q_{e,exp}$ (mg/g)	k_2 (g/mg.min)	r^2
0, 1124	0,5565	0,0247	0,9193	0,5736	0,5565	0,4886	0,9995

Examination of Table 2 shows that the correlation coefficient r^2 (0.9995) of the pseudo-first-order kinetic model respectively is lower than that of the pseudo-second-order model r^2 (0.9726).

Furthermore, the theoretical Q_e value ($Q_{e,\ the}$) calculated from the pseudo-second-order model is close to that obtained experimentally (Q_e, exp) compared to that of the pseudo-first-order model. The latter result suggests that Cadmium adsorption on peanut shells is better described by the pseudo-second-order model. The authors [20] had also observed that the adsorption kinetics of Cadmium on their different supports were well compatible with the pseudo-second-order model.

3.3. Thermodynamic Study of Adsorption

3.3.1. Effect of Temperature

Temperature variation affects adsorption capacity and can help determine the mechanism of the adsorption process. The results of the study of the influence of temperature on Cadmium adsorption by peanut shells are shown in Figure 9.

Figure-9. Cadmium adsorption versus temperature curve

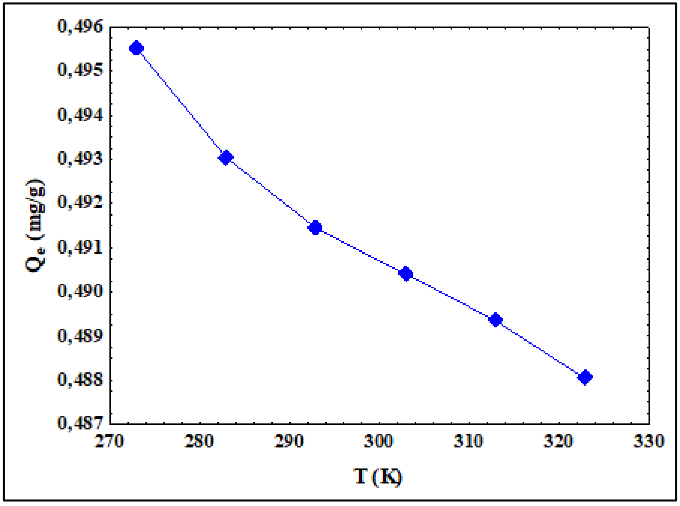
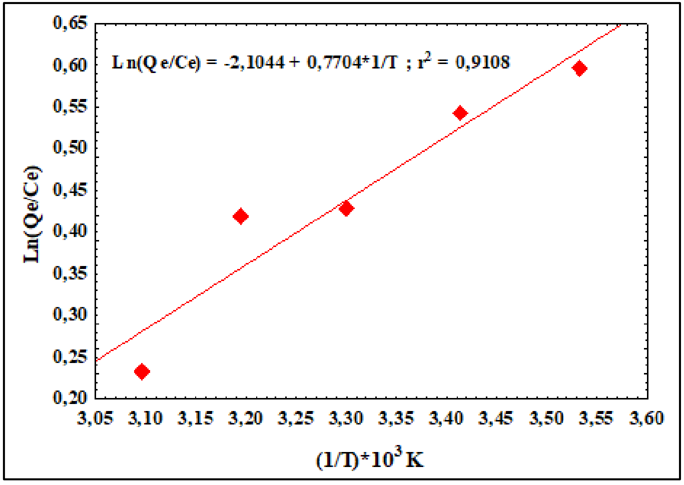


Figure 9 shows that the amount of cadmium removed decreases with increasing temperature. As the temperature of the medium rises, the diffusion rate of metal ions towards the internal adsorption sites of peanut shells decreases [21]. However, the work of Demirçivi [22], and Guyo, *et al.* [23] reported an improvement in the quantity of pollutants removed with increasing temperature.

3.3.2. Thermodynamic Quantities

To better understand the influence of temperature on cadmium adsorption, thermodynamic parameters such as the variation in enthalpy (ΔH°=, entropy (ΔS°) and free energy (ΔG°) were determined from experimental results (Figure 10).

Figure-10. Thermodynamic modeling of cadmium adsorption



The thermodynamic parameters obtained are shown in Table 3.

Table-3. Thermodynamic parameters

T(K)	ΔG°(kJ/mol)	ΔH°(kJ/mol)	ΔS°(J/mol)
283	-1402,02204	6,4051	-17,4959
293	-1322,09465		
303	-1078,63524		
313	-1089,88423		
323	-627,009958		

Examination of Table 3 shows that the Gibbs free energy (ΔG°) values are all negative. This means that the Cadmium adsorption process on peanut shells is spontaneous, feasible and favorable. These results confirm the affinity of the adsorbent for metal ions [24].

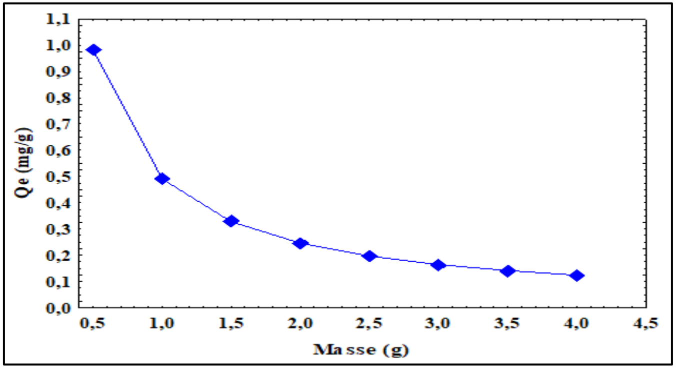
The enthalpy value ΔH^0 (kJ/mol) is equal to 6.4051. The positive value of ΔH^0 indicates the endothermic nature of the cadmium adsorption process. This value also shows that adsorption is favorable to a rise in temperature [25]. However, the authors Mingyue et al [19] showed that the adsorption process of cadmium ions (Cd^{2+}) ions from aqueous solutions by a biomass waste hydrogel was exothermic.

The low value of ΔH° (<40 $kJ.mol^{-1}$) obtained suggests that the cadmium adsorption process is physical in nature [26]. Furthermore, the negative value of ΔS^0 (-17.4959 kJ/mol/K) reflects a decrease in disorder at the cadmium/peanut shell interface. This decrease shows that the transition state is more ordered than the initial state and a well-organized distribution of metal ions at the adsorption sites [27].

3.4. Adsorbent Mass Effect

The study of adsorbent mass enables us to determine the optimum mass required for a given initial concentration. Cadmium removal tests were carried out on different peanut shell masses. The results are shown in Figure 11.

Figure-11. Cadmium adsorption versus mass curve



Analysis of Figure 11 shows that the amount of metal adsorbed decreases with increasing peanut shell mass. When the mass varies from 0.5 to 4 g, there is a drop in the amount of metal removed. It therefore appears that when the mass of the adsorbent is low, the quantity of cadmium adsorbed is high. This is because at low masses, the active sites are easily accessible. In the opposite case, at high masses, these active sites become difficult to access by metal ions due to the formation of biomass aggregates [28]. These particulate aggregates of the adsorbent support form a screen which makes it difficult for cadmium ions (Cd^{2+}) to reach the adsorption sites on peanut shells [29].

3.5. pH Effect

In this study, the influence of pH was evaluated. Figure 12 shows the evolution of the amount of cadmium adsorbed as a function of pH.

Figure-12. Cadmium adsorption versus pH curve

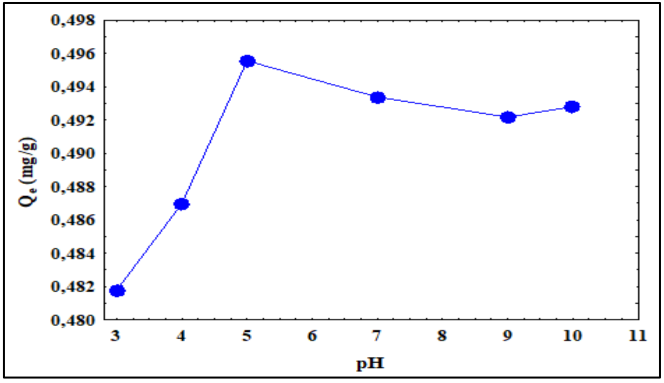


Figure 12 shows that the maximum amount of cadmium adsorbed increases with pH, reaching a maximum and then decreasing. This may be explained by the fact that, at low pH values, the support surface competes with cationic cadmium (Cd^{2+}) ions. This competition favors the adsorption of cadmium ions via the phenomenon of electrostatic interaction [16, 30] up to maximums (pH = 5). The results of our work on the influence of pH variation are in agreement with those of Victor et al [31] and Fumihiko et al [32]. Beyond this maximum, there is a drop in quantity. This could be explained by structure-related steric hindrance. This appears to be a factor limiting access to surface sites.

4. Conclusion

In this study, the adsorption capacity of peanut shells in the treatment of effluents loaded with pollutants such as cadmium was evaluated. In order to understand the removal of these pollutants, batch adsorption tests of cadmium on peanut shells were carried out. The effects of physico-chemical parameters such as the contact time between peanut shells and cadmium, the mass of the support, the initial concentration of cadmium in the solution, temperature and pH were evaluated. The study showed that contact time, carrier mass, initial concentration, temperature and pH influence the adsorption capacity of peanut shells for cadmium. Modelling of the data from the contact time study showed that the modelpseudo-second-order kinetic model perfectly describes the kinetics of the cadmium adsorption reaction. The Freundlich equation well describes the adsorption isotherm. Thermodynamic studies have shown that adsorption is spontaneous and endothermic. This study led to a low entropy during cadmium adsorption. The best adsorption capacity was observed in acidic media.

Further modification and characterization of the peanut shells surface would be necessary to better understand the cadmium ion adsorption mechanism.

This study is limited by the long-term biodegradation of peanut shells and the small quantity of solution treated. For this reason, it is necessary to envision a study of peanut shells conservation and an evolution towards continuous treatment mode of contaminated wastewater on a peanut shell's fixed-bed column.

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