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Biosorption of Cd (II) from Solution using Cashew (Anacardium occidentale) Leaf: Kinetics, Equilibrium, and Thermodynamics

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ABSTRACT

The presence of metal ions in aqueous solutions represents a major environmental problem. These inorganic species are persistent and non-biodegradable pollutants that should be eliminated from water. It occurs due to the direct and indirect discharge of diverse chemicals into the water bodies without sufficient treatment to reduce and diminish the harmful compounds. The biomass materials utilized in this research were cashew (*Anacardium occidentale*) leaves. This was discussed along with the principal factors affecting the biosorption process, such as solution pH, biomass dose, metal ion concentration, and contact time. The potential health and environmental hazards of metal ions, in addition to the kinetic and isothermal models assessed to fit the biosorption experimental data, were also considered. The kinetic and isothermal models follow pseudo second order and Freundlich, respectively. The maximum biosorption was obtained at 180 minutes at pH 5 and 323 K for an initial Cd (II) concentration of 90 mg/L. This indicates that the cashew (*Anacardium occidentale*) leaf is a good biosorbent for the treatment of Cd (II)-bearing wastewater.

Keywords: Biosorption, Biomass dosage, pollutant, wastewater

INTRODUCTION

Industrial revolution has led to the increased use of heavy metal like Cr, Cu, Cd, Pd, Zn etc. Heavy metals such as Cr, Cu, Cd, Pd, Zn are essential for plants and microbes as they form many enzymes and proteins[1].

The presence of inorganic pollutants such as metal ions in the ecosystem causes a major environmental problem. Toxic metal compounds coming to the earth's surface not only contaminate earth's water (seas, lakes, ponds and reservoirs), but can also contaminate underground water in trace amounts by leaking from the soil after rain and snow[2]. The numerous metals which are significantly toxic to human beings and ecological environments, include chromium, copper, lead, cadmium, mercury, zinc, manganese and nickel, etc[3].

It is necessary to remove metal ions from wastewater before it can be discharged. In this respect, many physicochemical methods have been developed for the removal of metal ions from aqueous solutions including precipitation, evaporation, electro deposition, ion exchange, membrane separation, coagulation etc[4].

However, these methods have disadvantages such as secondary pollution, high cost, high energy input, large quantities of chemical

reagents or poor treatment efficiency at low metal concentration[5]. It can be said that the conventional methods for metal ions removal from wastewater are limited by technical and economic barriers, especially when concentration of metals in the wastewater is low (<100 ppm)[6]. Therefore, the search and development of efficient and low-cost metal removal processes is of utmost importance. The effective and economical technologies involving the removal of toxic metals from wastewater have directed attention to biosorption based on metal binding capacities of various biological materials at little or no cost[7-17].
In this endeavor, biosorption has emerged as an alternative and sustainable strategy for cleaning up water[15].

MATERIALS & METHODS

Biomass preparation

Cashew (*Anacardium occidentale*) leaves were harvested from a compound along Ijesa road, opposite fountain hospital, Ago Iwoye, Ogun State, Nigeria. The leaves were properly rinsed with water, sun dried immediately and later cut into pieces of approximately 0.5 cm. The leaf sample was kept dry till the time of usage.

Batch Biosorption Study

The biosorption study was carried out by contacting 0.5 g of cashew (*Anacardium occidentale*) leaf with 25 mL of the metal ion solution under different conditions for a period in a boiling tube. The biosorption studies were conducted at 27 ^oC using thermostatic water bath to determine the effect of pH, contact time and initial metal ion concentration on the biosorption. The residual metal ion was analyzed using an Atomic Absorption Spectrophotometer. The amount of metal ion biosorbed was determined by difference and the mean value calculated.

Effect of pH on biosorption

The effect of pH on the biosorpton of the metal ion was carried out within the range that would not be influenced by the metal precipitated. This was done by contacting 0.5 g of Cashew leaf (*Anacardium occidentale*) with 25 mL of 100 mgL^{-1} metal ion solution in a boiling tube within the range pH 1-7. The pH of each solution was adjusted to the desired value by dropping 0.1 M HNO_3 and/or 0.1 M NaOH. The boiling tubes containing the mixture were left in a water bath for 3 hours. The biomass was removed from the solution by decantation. The residual metal ion concentration in the solution was analyzed. The optimum pH was determined as the pH with the highest biosorption of each metal ion. All experiments were conducted in duplicates and the mean value was determined for each.

Effect of contact time on biosorption

The biosorption of the metal ion by Cashew (*Anacardium occidentale*) leaf was studied at various time interval (0-300 min) and at the concentration of 100 mg/L. This was done by weighing 0.5 g of Cashew leaf into each boiling tube and 25 mL of 100 mg/L of metal ion solution at optimum pH was introduced into it. The leaf was left in solution for varying periods. The solution in the boiling tubes was decanted at different time intervals from the first to the last tube. The aliquot was then taken for analysis using an Atomic Absorption Spectrophotometer. The amount of metal ions biosorbed was calculated for each sample.

Effect of initial metal ion concentration on biosorption

Batch biosorption study of metal ions were carried out using a concentration range of 10-100 mg/L. This was done by introducing 0.5 g of Cashew (*Anacardium occidentale*) leaf into each of the boiling tubes employed and 25 mL of 100 mg/L of metal ion solution at optimal pH was added to the tube. A boiling tube was used for each concentration. The tubes were left in a thermostatic water bath maintained at 27 ^oC. The metal bound cashew leaf was removed from the solution and the concentration of residual metal ion in each solution was determined.

Effect of temperature on biosorption

The batch biosorption process was studied at different temperatures of 20-50 ^oC in order to investigate the effect of temperature on the biosorption process. This was done by contracting 0.5 g of cashew leaf with 25 mL of 100 mg/L of metal ion solution at the optimal pH. The biosorption of metal ion may involve chemical bond formation and ion exchange since the temperature is a main parameter affecting them.

Effect of biomass dosage

The effect of biomass dosage on biosorption efficiency is the general trend of increase in metal ion biosorbed with increase in biomass dosage, this indicates an increase in uptake due to more binding sites on the biomass available for biosorption.

RESULT AND DISCUSSION

FT-IR studies of the free and metal-bound cashew *(Anacardium occidentale)* leaf

The FT-IR spectra of dried unloaded and Cd- loaded cashew *(Anacardium occidentale)* leaf was taken to obtain information on the nature of possible interaction between the functional groups of cashews *(Anacardium occidentale)* leaf biomass and the metal ion as presented in figure 1. The FT-IR spectrum was measured within the range of 500-4000 cm^{-1} wavenumber. As shown in figure 1, the FT-IR spectrum displays several absorption peaks indicating the complex nature of the biosorbent. The presence of the carboxylic, hydroxyl and amines groups on the biosorbent makes biosorption possible. These bands are due to the functional groups of the cashew leaves that participate in the biosorption of Cd (II). The spectra shows that there are clear band shifts and decrease in intensity of bands as reported in Table 1. Analysis of the FT-IR spectra showed the presence of ionisable functional groups (C-N, C=O, C-O and O –H) which are able to interact with cations. This implies that these functional groups would serve in the removal of positively charged ions from solution.

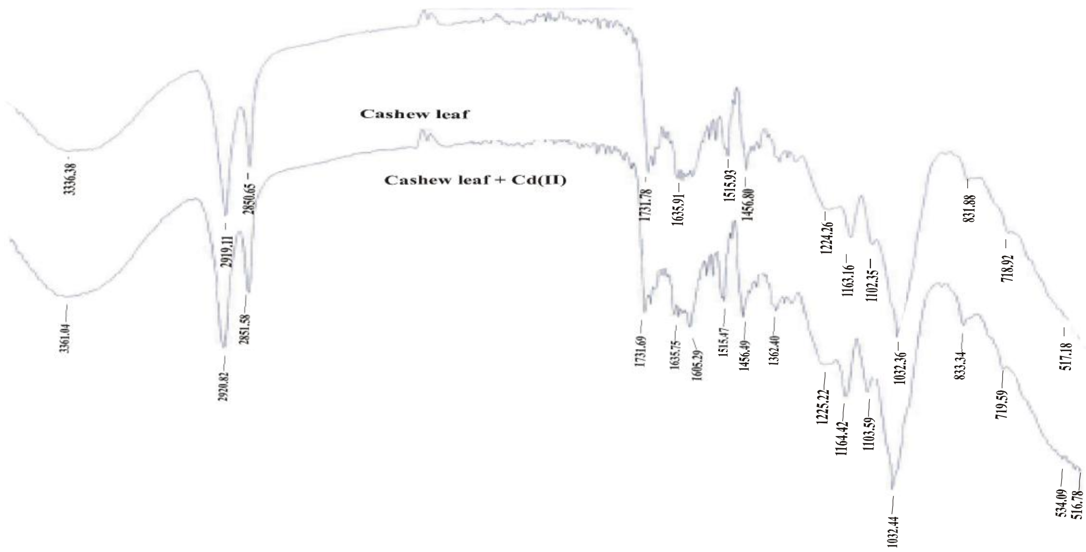

Figure 1. FT-IR spectra of the free and metal (Cd) bound cashew *(Anacardium occidentale)* leaf.

Table 1: FT-IR spectra characteristics of cashew (*Anacardium occidentale)* leaf before and after biosorption of Cd (II) for three hours

Metal ion	Absorption bands (cm^{-1})			Assignment
	Before	After	Difference	
Cd (II)	3336.38	3361.04	24.66	O-H stretch
	1731.78	1731.69	-0.09	C=O stretch

1635.91	1635.75	-0.16	N-H bend
1515.93	1515.47	-0.46	C=N, C-O stretch
1224.26	1225.22	0.96	C-N stretch
1163.16	1164.42	1.26	C-O (Ester)
1102.35	1103.59	1.24	C-O-C (Dialkyl Ester)
1032.36	1032.44	0.08	C-O stretch

Effect of solution pH on biosorption

It has been reported that the suitable pH ranges for the sorption of different metal ions were slightly different. Consequently, the suitable pH ranges for Cd (II) should be 1 – 7. The result of the pH study given in Figure 2 shows that maximum biosorption was obtained at pH 5. The percentage Cd (II) biosorbed is slightly lower at higher pH values. This result supports the earlier report that biosorption is pH dependent[18]. Surface adsorption is a physicochemical phenomenon. The cell walls of many plants consist of polysaccharides, proteins and lipids, hence, offer a host of functional groups capable of binding to heavy metals. These functional groups, such as amino, carboxylic, sulphydryl, phosphate and thiol groups, differ in their affinity as well as specificity for metal binding[19]. The charges of the adsorbate and that of the adsorbent often depend on the pH of the solution[20]. To understand the biosorption mechanism, the biosorption of Cd^{2+} as a function of pH was measured. It is observed from Figure 2 that there was an increase in the biosorption capacity of the biomass with an increase in pH from 2.0 to 3.0 but slightly lower biosorption was obtained at higher pH. As a result of net negative charge on the cell wall of the biosorbent at pH above the isoelectric point, the ionic state of the ligands such as carbonyl, phosphate and amino groups favors reaction with Cd^{2+}. On the other hand, on decreasing pH, the net charge on the cell wall is positive thereby inhibiting the approach of positively charge ions[21]. As the pH increased, the ligands in cashew leaf would be exposed, thereby increasing the attraction of metal ions with positive charge and allowing biosorption on the leaf surface. The result suggests that optimum biosorption is obtained at pH 5.0 and that initial pH would play a vital role in the removal of Cd^{2+} from aqueous solutions using cashew leaf.

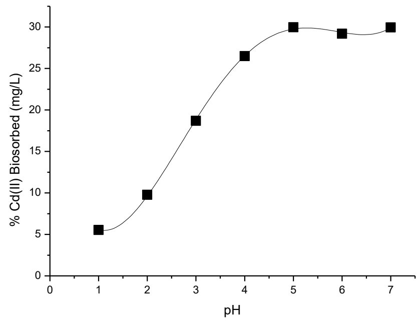

Figure 2. pH-dependent profile of the biosorption of Cd (II) by cashew (*Anacardium occidentale*) leaf.

Effect of contact time on the biosorption of Cd (II)

In batch biosorption experiments, the determination of the optimum contact time required to achieve the highest removal of metal ions is one of the key factors usually assessed. Equilibrium time is one of the important parameters for selecting a wastewater treatment system. The result of the effect of contact time on the biosorption of Cd (II) from aqueous solution is shown in Figure 3. It is observed that the adsorptive capacity of cashew leaf for Cd (II) increased with increase in contact time. The biosorption of Cd (II) by the biomass was rapid for the first 70 min as a result of available binding sites on the biomass. The biosorption approached equilibrium within 180 min as the binding sites were used up. The period of 180 min was therefore taken as the time required for the biosorption of Cd (II) by cashew leaf. Biosorption of metal ions has been reported to be biphasic[19]. The initial fast phase occurs due to surface adsorption on the biomass. The subsequent slow phase occurs due to diffusion of the metal ions into the inner part of the biomass.

It is observed in Figure 3 that the biosorption rate was high at the beginning, but equilibrium was approached within 180 min, similar to what was earlier reported[19].

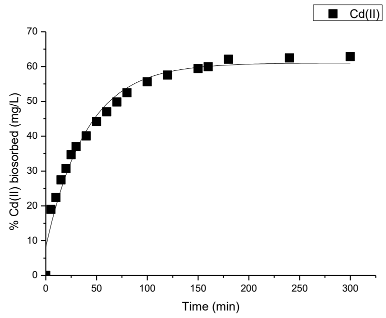

Figure 3: Time course of the biosorption of Cd(II) by cashew (*Anacardium occidentale*) leaf.

Biosorption kinetics

The prediction of the biosorption rate gives important information for designing batch biosorption systems. Information on the kinetics of pollutant uptake is required for selecting optimum operating conditions for full-scale batch process[22]. The sorption kinetics is significant in the treatment of wastewater, as it provides valuable insights into the reaction pathways and mechanisms of sorption reactions[23]. Several kinetic models have been applied to fit the biosorption data of different metal ions onto various biosorbents. These models include the pseudo-first order, pseudo-second order, Elovich, intraparticle diffusion, etc.

Pseudo-first order kinetics model

The pseudo-first order kinetic model (Lagergren model) assumes that metal ions bind only to one sorption site on the sorbent surface[24]. In Lagergren model, the rate of occupation of biosorption sites is proportional to the number of unoccupied sites[24]. The model is represented by:

$$rate = -\frac{d[A]}{dt} = k[A]^n \quad (1)$$

Which can also be writing as

$$\frac{d}{dt} q_t = k_1(q_e - q_t) \quad (2)$$

Integrating between the limit $q_t = 0$ at t=0 and $q_t = q_t$ at t =t, we obtain

$$\log[\frac{q_e}{(q_e - q_t)}] = \frac{k_1}{2.303} t \quad (3)$$

This can be re-arranged to obtain a linear form:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (4)$$

Where k_1 (min^{-1}) is the pseudo first order adsorption rate coefficient, q_e and q_t are the values of amount adsorbed per unit mass at equilibrium and at any time t. The values of k_1 and the calculated q_e can be obtained respectively from the slope and intercept of the linear plot of $\log(q_e - q_t)$ versus (t).

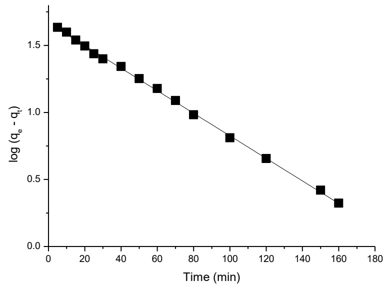

Figure 4. The pseudo first order kinetic plot for the biosorption of Cd (II) by cashew (*Anacardium occidentale*) leaf at 27 ºC.

Pseudo second order kinetics model

The second order kinetic model assumes that the rate limiting step is most likely to involve chemical interactions leading to binding of the ions to the surface by bonding as strong as covalent bonding[23]. The pseudo-second-order equation[25] based on equilibrium adsorption is expressed as:

$$\frac{d}{dt} q_t = k_1(q_e - q_t)^2 \quad (5)$$

Integrating between boundary conditions, we have

$$\frac{1}{(q_e - q_t)} = \frac{1}{q_e} + k_2 t \quad (6)$$

This can be re-arranged to obtain a linear form:

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

wherek$_2$ (g/mg min.) is the rate constant of second-order adsorption. Plots of (t/qt) versus (t) give the values of 1/qe as the slope and $\frac{1}{k_2 q_e^2}$ as the intercept.

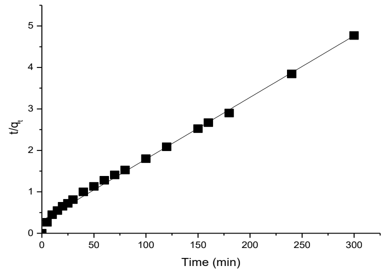

Figure 5. The pseudo second order kinetic plot for the biosorption of Cd (II) by cashew (*Anacardium occidentale*) leaf at 27 ºC.

Elovich kinetic model

The data were equally subjected to the Elovich kinetic model given by;

$$q_t = A + B \ln t \quad (8)$$

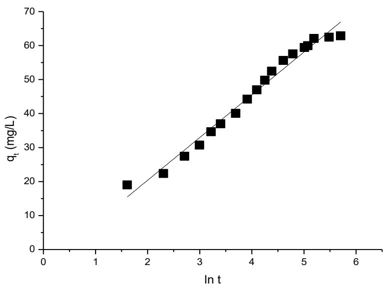

Figure 6. Graphical representation of Elovish kinetic model.

Intra paticle diffusion kinetic model

The intraparticle diffusion equation given as;

$$R = K_s t^b \quad (9)$$

has been used to indicate the behavior of intraparticle diffusion as the rate limiting step in the biosorption process. R is the percentage of metal biosorbed, K_s is the intraparticle diffusion constant, t is the constant time, while b is the gradient of the linear plot. In linear form the immediate equation above becomes.

$$\log R = b \log t + \log K_s \quad (10)$$

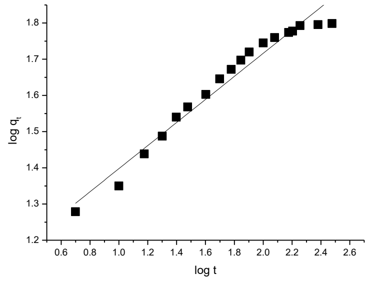

Figure 7. Intra particle diffusion kinetic model on biosorption of Cd (II) using cashew leaf.

Table 2: Kinetic parameters for the biosorption of Cd (II) onto cashew leaf a 100 mgL^{-1}

Kinetic models	Parameters	Cd (II)
First-order	q_e(mgg^{-1})	46.38
	k_1 (min^{-1})	1.94×10^{-2}
	R^2	0.9170
Second-order	$q_{e\ cal}$ (mgg^{-1})	67.20
	k_2(g mg^{-1} min^{-1})	7.34×10^{-4}
	R^2	0.9890
Elovich	A	-4.7662
	B	12.573
	R^2	0.9596
Intra-particle diffusion	K_d(mgg^{-1}min^{-1})	12.00
	B	0.3180
	R^2	0.9285

Effect of initial metal ion concentration

The initial concentration of metal ions in the solution plays a key role as a driving force to overcome the mass transfer resistance between the aqueous and solid phases[26]. It is generally agreed that the biosorption capacity increases as the initial metal ion concentration in the solution increases, whereas the metal removal percentage (also called removal efficiency) decreases by increasing the metal ion initial concentration. As a rule, increasing the initial metal concentration results in an increase in the biosorption capacity because it provides a driving force to overcome mass transfer resistance between the biosorbent and biosorption medium[27]. The biosorption of cadmium was carried out at various concentration ranges from 10 to 100 mg/L and the most absorbed concentration is 90 mg/L.

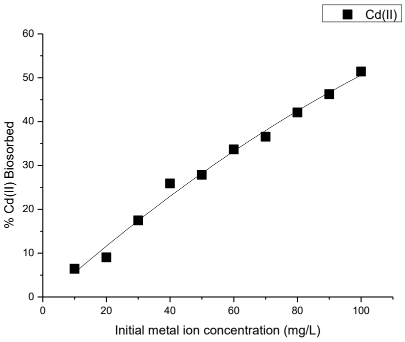

Figure 8. Effect of initial concentration of cadmium-on-cadmium biosorption by cashew leaf.

Biosorption isotherm

The Freundlich, Langmuir, Termkin, and D-R isotherms, were employed to calculate the biosorption capacity.

Freundlich isotherm

The Freundlich isotherm is an empirical equation describing adsorption onto a heterogeneous surface. The Freundlich isotherm model applies to adsorption on heterogeneous surfaces with the interaction between adsorbed molecules, and the application of the Freundlich equation also suggests that sorption energy exponentially decreases on completion of the sorption centers of an adsorbent[28]. The empirical Freundlich equation[29] based on sorption on a heterogeneous surface is given as:

$$\log Q = \frac{1}{n}\log C_e + \log K_f \qquad (11)$$

Where K_f and $\frac{1}{n}$ are the Freundlich constants characteristic of the system and are indicators of adsorption capacity and adsorption intensity, respectively[30]. The Freundlich equilibrium constants are usually determined from the plots of log Qeq versus log Ceq. The n value indicates the degree of nonlinearity between solution concentration and adsorption as follows: if n = 1, then adsorption is linear; if n < 1, then adsorption is a chemical process; if n >1, then adsorption is a physical process[31].

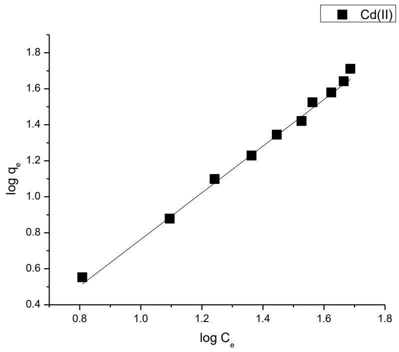

Figure 9. Freundlich isotherm plot on biosorption of Cd (II) using cashew leaf.

Langmuir isotherm

The Langmuir isotherm model assumes a surface with homogeneous binding sites, equivalent sorption energies, and no interaction between sorbed species[32]. In this model once a site is filled, no further sorption can take place at that site. As such, the surface will eventually reach a saturation point where the maximum adsorption of the surface will be achieved[28]. The Langmuir equation developed by Irving Langmuir in 1916[33] is represented by:

$$\frac{C_{eq}}{Q_{eq}} = \frac{1}{b}Q_{max} + \frac{C_{eq}}{A_S} \qquad (12)$$

where Qmax (mg/g) is the maximum amount of metal ion per unit weight of biosorbent to form a complete monolayer on the surface bound at high Ceq, and b (l/mg) is the Langmuir constant related to the energy of adsorption. The Qmax and b can be determined from the linear plot of Ceq/Qeq versus Ceq[30]. The essential characteristic of the Langmuir isotherm may be expressed in terms of dimensionless separation parameter,

$$R_L = \frac{1}{1+bC_O} \qquad (13)$$

which is indicative of the isotherm shape that predicts whether an adsorption system is favorable or unfavorable[30].

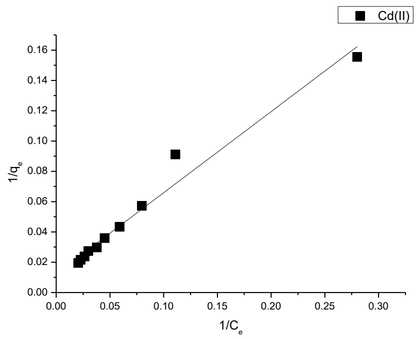

Figure 10. Langmuir Biosorption isotherm for cadmium using cashew leaf.

Table 3: Isothermal parameters for the biosorption of Cd (II) onto cashew leaf

Kinetic model	Parameters	
Freundlich	n	1.22
	$K_f(mgg^{-1})\ (Lmg^{-1})^{1/n}$	2.15
	R^2	0.9759
Langmuir	$q_{max}(mgg^{-1})$	80.78
	$K_L\ (L\ mg^{-1})$	0.0231
	R^2	0.9371

Biosorption efficiency

The result of the study on the effect of initial metal ion concentration on biosorption efficiency is shown on Fig. 13. The plots show that the biosorption efficiency of the biomass increased with an increase in the initial metal ion concentration for all the metal ions which might be due to an increase in effective collision between the metal ions and the active sites. The biosorption efficiency (E) for each metal ion was calculated as

$$E = 100\left(\frac{C_i - C_e}{C_i}\right) \qquad (17)$$

Where C_i and C_e are the initial and the equilibrium metal ion concentrations (mg L^{-1}), respectively.

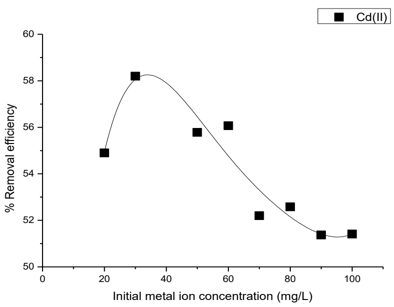

Figure 11. Percentage efficiency for the biosorption of Cd (II) by cashew leaf

Thermodynamics of cd(ii) biosorption by cashew leaf

The biosorption study can be regarded as a heterogenous and reversible process at equilibrium. The apparent equilibrium constant for the process has been shown[34-35] to be:

$$K_C = \frac{C_{ad}}{C_e} \qquad (18)$$

The Gibbs free energy of the biosorption process is thus given as:

$$\Delta G = -RT \ln K_C \qquad (19)$$

Where ΔG° is the standard Gibbs free energy change for the biosorption (kJmol-1), R the universal gas constant (8.314 $Jmol^{-1}k^{-1}$) and T the temperature (K). The free energy change obtained for the biosorption of Cd (II) at 323 K, initial Cd (II) concentration of 90 mgL^{-1} and pH 5 is –7.17 $kJmol^{-1}$. The large negative value of ΔG° obtained for the biosorption of Cd (II) shows the spontaneity of the biosorption process, thus signifying that cashew leaf could serve as biomass for biosorption of Cd (II).

The free energy change is related to other thermodynamic properties as;

$$\Delta G = \Delta H - T\Delta S \qquad (20)$$

$$\ln K_c = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \qquad (21)$$

Where t is the absolute temperature (K); R is the gas constant (8.314 $Jmol^{-1}K^{-1}$). ΔH ($Jmol^{-1}$) and ΔS (8.314 $Jmol^{-1}K^{-1}$) were calculated from the slope and the intercept of the linear plot of $\ln K_c$ versus $\frac{1}{T}$. The thermodynamic parameters obtained for this study are presented in Table 4

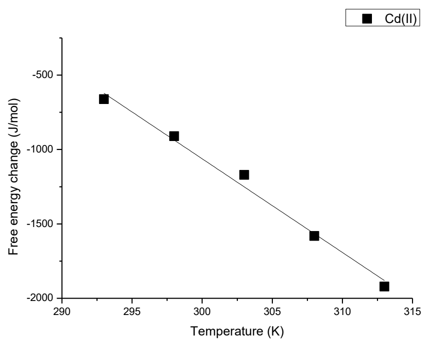

Figure 12. Thermodynamic plot of biosorption of Cd (II) with cashew leaf.

The plot shown in Figure 13 is linear over the entire range of temperature investigated.

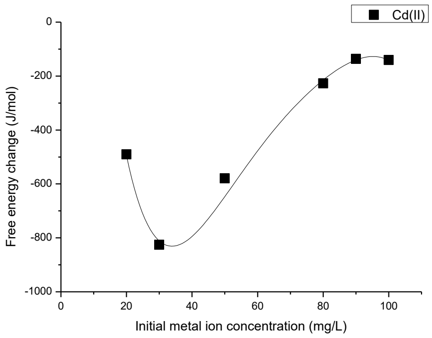

Figure 13. Thermodynamic plot with effect of initial metal ion concentration

Table 4: Thermodynamic parameters for the biosorption of Cd (II) by cashew (*Anacardium occidentale)* leaf.

Metal ion	ΔH° (kJ mol^{-1})	ΔS° (J $K^{-1}mol^{-1}$)	R^2	S. D	A (kJ mol^{-1}) @(303K)	A (kJ mol^{-1}) @(323K)
Cd (II)	17.82	+62.957	0.9519	9767.1211	20.34	20.51

Effect of biomass dosage

Biomass provides binding sites for the sorption of metal ions, and hence its concentration strongly affects the sorption of metal ions from the solution[36]. The amount of biosorbent used for the treatment studies is an important parameter, which determines the potential of biosorbent to remove metal ions at a given initial concentration.

For a fixed metal initial concentration, increasing the adsorbent dose provides greater surface area and availability of more active sites, thus leading to the enhancement of metal ion uptake[36]. At low biomass dosage, the number of ions adsorbed per unit adsorbent weight is high. The adsorption capacity is reduced when the biomass dosage increases as a result of lower adsorbate to binding site ratio where the ions are distributed onto larger amount of biomass binding sites.

However, at higher dosage, the ions adsorbed are higher due to the availability of more empty binding sites as compared to lower dosage which has fewer binding sites to adsorb the same amount of metal ions in the adsorbate solution[15].

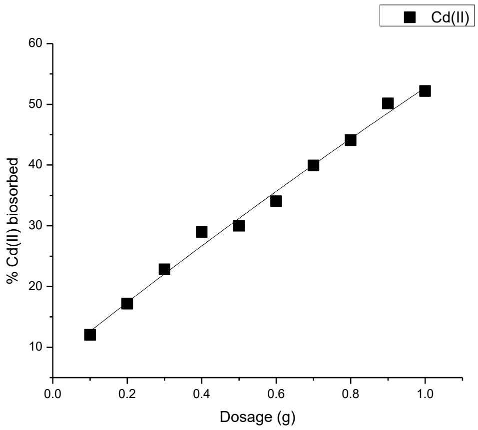

Figure14. Effect of biomass dosage plot on the biosorption of Cd (II) using cashew leaf.

CONCLUSION

This work confirms that the cashew (*Anacardium occidentale*) leaf could be used as an effective biosorbent for the treatment of Cd (II) bearing wastewater streams. The biosorption capacity was dependent on initial solution pH, contact time and the initial Cd (II) concentration. The maximum biosorption was obtained at 180 minutes at pH 5 and 323 K for initial Cd (II) concentration of 90 mgL^{-1}. The kinetic and isothermal models were assessed to fit the biosorption experimental data and these were found to follow the pseudo second order and Freundlich respectively.

The removal efficiency decreases with increase in initial Cd (II) concentration due to reduction in available binding site on the biosorbent for Cd (II).

Declaration by Authors

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