



# Equilibrium and Kinetic Study of the Removal of Cadmium from the Human Blood Plasma Using Dried *Allium cepa* Biomass

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**Abstract:** The therapeutic importance of *Allium cepa* has been reported in previous studies. However, little has been known of its efficiency to remove heavy metal from the human blood plasma. Thus this study seeks to investigate the efficiency of *Allium cepa* biomass in detoxification of Cadmium ions from aqueous solution of human blood plasma in-vitro. The biosorbent was prepared and identified using FTIR which revealed the presence of carbonyl group, amide group, sulphoxide group, ether and aromatic groups. The optimum sorption was investigated and the experimental data revealed an equilibrium pH 4, optimum Cd<sup>2+</sup> concentration 70 mgL<sup>-1</sup> and optimum biosorbent dose 0.80 g were obtained and used to study the equilibrium sorption rate which occurred at 80 mins with 99.98% removal at ambient temperature. The experimental data fitted Pseudo Second order kinetic as indicated by the correlation coefficient value (R<sup>2</sup>)=0.9961 with a rate constant K<sub>2</sub>=0.3730 g.mg<sup>-1</sup>.min<sup>-1</sup>. The experimental data conforms to Freundlich isotherm and Jovanovic isotherm, however Freundlich isotherm showed best fit with correlation coefficient (R<sup>2</sup>)=0.632, sorption capacity (KF)=3.3113 and sorption intensive (n)=1.870. The separation factor of the Langmuir isotherm (RL)=0.0141, which suggests that the overall adsorption process was favourable.

**Keywords:** *Allium cepa*, Biosorption, Flory-Huggins Isotherms, Freundlich Isotherms, Jovanovic Isotherms, Langmuir Isotherms, Kinetic Models, Toxic Metal

## 1. Introduction

Health challenges often arise due to the presence of toxic metals present in industrial chemicals which adversely affect animals and human lives. Toxic metals (Cadmium) are harmful because of their ability to bio-accumulate in various plants and body parts due to their non-biodegradable nature [1]. These metals have a wide application in agriculture as fungicides, pesticide and herbicide to control pest and improve agricultural yield. Exposure to these metals even at low concentration often causes a wide range of illness such as nausea, vomiting, destruction of the blood cell, damage liver and kidney failure to mention but a few. The metals enter the human body through inhalation, drinking of

contaminated water and consumption of contaminated plant and animal. These metals are often transported to various body parts through the human blood plasma and tend to bind to proteins by displacing the essential metals from their natural binding sites in the body thereby causing health challenges [2]. Techniques such as adsorption, coagulation, biosorption, solvent extraction, oxidation, reverse osmosis and phytoremediation have been employed in the removal of these metals. However, increase in the amount of hazardous chemicals makes these conventional techniques insufficient.

Biosorption technology exploits biomaterials in the elimination of toxic metals from aqueous solution via physicochemical and/or metabolically mediated pathways. This practice has been employed for a while as it provides an

eco-friendly, effective and low cost material option. Amongst these biomaterials are *Allium cepa*, which is vegetable plant widely consumed for its therapeutic importance. They have been reported to contain health promoting phytochemicals (quercetin, disulphides, trisulphides and vinylthiols), acrid and strong antioxidants [3]. They are important in disease prevention, tracking free radicals in the body and preventing blood platelets from sticking together and forming clots and reduce blood pressure [4].

The human blood plasma is a component of the human blood. It makes up 55 per cent of total blood volume. It helps in suspension of the blood cells and also in the transportation of essential nutrients, antibodies, unwanted materials and toxic substances around the body. Several studies have been carried out utilizing plants and animal remaining in heavy metal removal. For instance, Chromium (IV) adsorption from aqueous solution by *Leucaenaleucocephala* seed shell [5]; Biosorption of heavy metals from aqueous solution using *Streptomyces rimosus* biomass [1]; Lead biosorption from aqueous solution by waste biosorbent from derived from biotrickling filter: Kinetics and, isotherm and thermodynamics [2]; Multicomponent isotherm for biosorption of Zn (II), Co (II) and Cd(II) from tarmy mixture onto pretreated dried *Aspergillus niger* biomass [6]; Cadmium biosorption from aqueous solution using *Aspergillus cristatus* [7]; Evaluation of *Ulva Fasciata* and *Sargassum* sp for the biosorption of  $\text{Cu}^{2+}$  from aqueous solution [8]; amongst others. This study seeks to investigate the kinetics and equilibrium of the decontamination of cadmium from human blood fluid utilizing dehydrated *Allium cepa* biomass. The equilibrium pH, initial  $\text{Cd}^{2+}$  concentration and biosorbent dose will be established and employed to study the rate of  $\text{Cd}^{2+}$  removal from human blood fluid. The experimental data obtained will be fitted into Freundlich Isotherm, Freundlich isotherm, Jovanovic Isotherm and Langmuir Isotherm. The kinetic study was investigated using pseudo first and second order kinetic model.

## 2. Materials and Methods

### 2.1. Collection and Preparation of Sample Materials and Chemicals

All chemicals used were of analytical grade. The stock solution of  $\text{Cd}^{2+}$  was prepared in  $1.0 \text{ gL}^{-1}$  concentration using  $3\text{CdSO}_4 \cdot 8\text{H}_2\text{O}$  then diluted to the required concentration prior to analysis (Abdel-Aty *et al.* 2013).  $0.1 \text{ molL}^{-1} \text{HNO}_3$  and  $0.1 \text{ molL}^{-1} \text{NaOH}$  were employed to adjust the pH value of the aqueous solution. The human blood sample from which the plasma was extracted and used for the study was collected from the Department of Hematology, Ahmadu Bello University, Zaria. *Allium cepa* sample was collected from samaru market and taken to the Department of Chemistry Multi User Research Science Laboratory (MURSL) prior to analysis. Preparation and characterization of samples (*Allium cepa* and Human Blood plasma) has been reported by previous study [9].

### 2.2. Biosorption Experiment

#### 2.2.1. Study of Equilibrium pH

The equilibrium pH was studied at varying pH range (0.5 – 10) by introducing 0.80 g of biosorbent into  $100 \text{ cm}^3$  conical flask containing  $50 \text{ cm}^3$  aqueous solution composed of human blood plasma and  $70 \text{ mgL}^{-1} \text{Cd}^{2+}$  concentration. The mixture was adjusted to the desired pH using  $0.1 \text{ HNO}_3$  and  $0.1 \text{ NaOH}$  then agitated for 120 mins at 150 rpm using a rotary shaker [10]. The filtrate separated from the mixture using Whatman No 11 filter paper, digested and analyzed for residual  $\text{Cd}^{2+}$  concentration using Atomic Adsorption Spectrophotometer (AAS).

#### 2.2.2. Study of Equilibrium Concentration

The equilibrium concentration was studied at varying metal ion concentration ( $20 - 100 \text{ mgL}^{-1}$ ) by introducing 0.80 g of biosorbent into  $100 \text{ cm}^3$  conical flask containing  $50 \text{ cm}^3$  aqueous solution composed of human blood plasma and  $20 \text{ mgL}^{-1} \text{Cd}^{2+}$  concentration. The mixture was adjusted to the equilibrium pH, then agitated for 120 mins at 150 rpm using a rotary shaker [9]. After the preset contact time, the filtrate separated from the mixture using Whatman No 11 filter paper, digested and analyzed for residual  $\text{Cd}^{2+}$  concentration using Atomic Adsorption Spectrophotometer (AAS).

#### 2.2.3. Study of Equilibrium Biosorbent Dosage

The equilibrium biosorbent dosage was investigated at varying biosorbent dosage ( $0.20 - 1.00 \text{ g}$ ) by introducing 0.20 g of biosorbent into  $100 \text{ cm}^3$  conical flask containing  $50 \text{ cm}^3$  aqueous solution composed of human blood plasma and equilibrium metal ion concentration. The mixture adjusted to equilibrium pH, then agitated for 120 mins at 150 rpm using a rotary shaker [11]. After the preset contact time, the filtrate separated from the mixture using Whatman No 11 filter paper, digested and analyzed for residual  $\text{Cd}^{2+}$  concentration using Atomic Adsorption Spectrophotometer (AAS).

#### 2.2.4. Study of Equilibrium Contact Time

The rate of biosorption was studied at varying contact time ( $10 - 120$ ) mins, by the introduction of equilibrium biosorbent dose into  $100 \text{ cm}^3$  conical flask, containing a solution composed of  $50 \text{ cm}^3$  human blood plasma solution and equilibrium metal ion concentration. The mixture adjusted to equilibrium pH using  $0.1 \text{ HNO}_3$  and  $0.1 \text{ NaOH}$ , then agitated for various contact time ( $10 - 120$ ) mins respectively [12]. After the preset contact time, the filtrate separated from the mixture using Whatman No 11 filter paper, digested and analyzed for residual  $\text{Cd}^{2+}$  concentration using Atomic Adsorption Spectrophotometer (AAS).

### 2.3. Data Analysis

The percentage removal of  $\text{Cd}^{2+}$  was calculated using equation 1

$$\%R = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

Where  $C_0$  and  $C_e$  represent the initial and final metal ion

concentration ( $\text{mg/L}^{-1}$ ), present in the human blood plasma solution respectively [10].

The amount of metal ion absorbed per unit mass ( $q_e$ ) is calculated using equation 2.

$$q_e = \frac{(C_0 - C_e)V}{M} \quad (2)$$

where  $C_i$  and  $C_e$  represents the initial and final (Equilibrium) metal ion concentration after time  $t$  min.,  $V$  represents the volume of human blood plasma in  $\text{cm}^3$  and  $M$  is the mass of adsorbent in gram [10].

#### 2.4. Adsorption Kinetics

The adsorption kinetics was investigated by employing the pseudo first and second order kinetic models to analyze metal ion uptake from the aqueous solution at different time intervals. When plotted, the linearity indicates if the models suitably describe the adsorption process [13].

The pseudo first order kinetic model is expressed in equation 3.

$$\text{Log}(q_e - q_t) = \text{Log}(q_e - K_1/2.303) \quad (3)$$

Where,  $q_e$  and  $q_t$  represents the sorption capacity at equilibrium and at time  $t$ , respectively ( $\text{mgm}^{-1}$ ) and  $K_1$  is the rate constant of pseudo first order sorption ( $\text{L.min}^{-1}$ ) [14]. A plot of  $\text{Log}(q_e - q_t)$  against  $t$  give a straight line with  $\text{log}(q_e)$  as slope and  $K_1$  as intercept.

The Pseudo second order kinetic is expressed in equation 4.

$$\frac{t}{qt} = \left(\frac{1}{K_2 q_e^2}\right) + \left(\frac{t}{q_e}\right) \quad (4)$$

Where  $q_e$  and  $qt$  represents the sorption capacity at equilibrium and at time  $t$ , ( $\text{mgg}^{-1}$ ) respectively, while  $K_2$  is the pseudo second order kinetic rate constant ( $\text{g.mg}^{-1}.\text{min}^{-1}$ ) [15].

#### 2.5. Adsorption Isotherms

The adsorption isotherm describes the interaction between the adsorbate in the solution and the adsorbent to establish equilibrium between the metal ions absorbed and the residual metal ions [16]. However, the potential of two parameter isotherms (Flory-Huggins Model, Freundlich model, Jovanovic model and Langmuir model) were tested to identify the model that best describe the interaction between the metal ions in the solution and the adsorbent.

The linear form of Flory-Huggins is expressed in equation 5.

$$\text{Log}\frac{\theta}{C_0} = \text{Log} K_{FG} + n \text{Log}(1 - \theta) \quad (5)$$

Where  $K_{FG}$  is Flory-Huggins equilibrium constant ( $\text{Lmg}^{-1}$ ),  $\theta = (1 - C_e/C_0)$  represents fractional coverage, and  $n$  is the interaction energy between the absorbed molecules [17]. A plot of  $\text{Log}\frac{\theta}{C_0}$  against  $\text{Log}(1 - \theta)$  is used to ascertain the value of  $n$  and  $K_{FG}$  as the slope and intercept respectively.

The Gibbs free energy is calculated using equation 6.

$$\Delta G = -RT \ln K_{FH} \quad (6)$$

The linear form of Freundlich isotherm is expressed in

equation 7.

$$\text{Log } q_e = \text{Log } K_F + \frac{1}{n} \text{Log } C_e \quad (7)$$

where  $K_F$  is the Freundlich adsorption capacity ( $\text{L/mg}$ ) and  $1/n$  represent the adsorption intensity, which also is an indication of the relative distribution of the energy level of heterogeneity of the active site on the adsorbent. A plot of  $\text{Log } q_e$  against  $\text{Log } C_e$  will produce a straight line with slope  $1/n$  and intercept  $K_F$  [18].

The linear form of Jovanovic isotherm is expressed in equation 8

$$\text{In } q_e = \text{In } q_{max} - K_f C_e \quad (8)$$

where  $q_e$  ( $\text{mgg}^{-1}$ ) represent the amount of  $\text{Cd}^{2+}$  on the biosorbent at equilibrium,  $q_{max}$  represents the maximum metal ions absorbed obtained from a plot of  $\text{In } q_e$  against  $C_e$ ,  $K_f$  represents the Jovanovic isotherm constant [19].

The linear form of Langmuir isotherm is expressed in equation 9.

$$\frac{C_e}{q_e} = \frac{1}{Q_m} C_e + \frac{1}{K_L Q_m} \quad (9)$$

Where,  $q_e$  represents equilibrium conc. of metal  $\text{Cd}^{2+}$  in solid phase,  $C_e$  represents equilibrium conc. of metal  $\text{Cd}^{2+}$  in liquid,  $Q_m$  represents Langmuir constant while  $K_L$  represents Langmuir sorption equilibrium constant [20]. A plot of  $C_e/Q_e$  against  $C_e$  gives straight line from which  $Q_m$  and  $K_L$  can be calculated from the slope and intercept respectively. The isotherm further predicts the affinity between the metal ions in aqueous solution and the biosorbent as expressed in equation 10.

$$R_L = \frac{1}{1 + K_L C_0} \quad (10)$$

Where  $R_L$  represents the separation factor and  $C_0$  represents the initial metal ion concentration in the aqueous solution. A favorable adsorption is recorded when value of  $R_L$  is in between 0 and 1. Thus  $R_L > 1$  is an unfavourable adsorption. However were  $R_L = 1$  suggests linear adsorption and  $R_L = 0$  suggests irreversible adsorption [21].

#### 2.6. FTIR Spectroscopy

FTIR spectra of adsorbent before and after  $\text{Cd}^{2+}$  binding were recorded to identify the active site believed to be responsible for the metal ion uptake as reported in previous study [22].

### 3. Result and Discussion

#### 3.1. FTIR Spectroscopy

The FTIR spectra of the adsorbent before and after biosorption are presented in Figure 1 and Figure 2 respectively. The result revealed a general shift in the peaks from  $3272.6 \text{ cm}^{-1} - 3268.9 \text{ cm}^{-1}$ ,  $1636.3 - 1625.1 \text{ cm}^{-1}$ ,  $1364.2 \text{ cm}^{-1} - 1319.5 \text{ cm}^{-1}$ ,  $1252.4 \text{ cm}^{-1} - 1237.5 \text{ cm}^{-1}$ ,  $920.7 \text{ cm}^{-1} - 890.8 \text{ cm}^{-1}$ . The general shift in the peak revealed presence of O-H stretch of carbonyl group, C=O stretch of amide group, S=O of

sulphoxide group,  $=C-O-O$  asymmetric stretch of ether group and  $=C-H$  bend of aromatic group were involved in the overall adsorption process. These functional groups are believed to have participated in the process of adsorption [23].

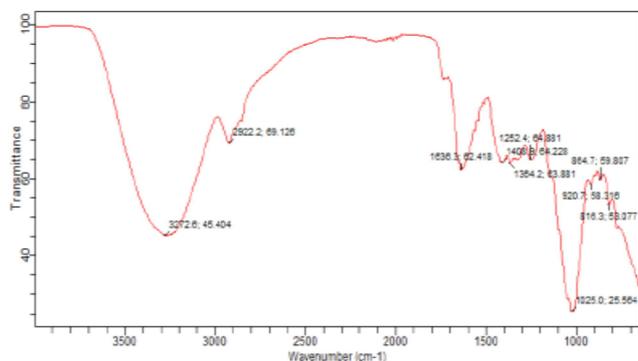


Figure 1. FTIR of *Allium Cepa* biomass.

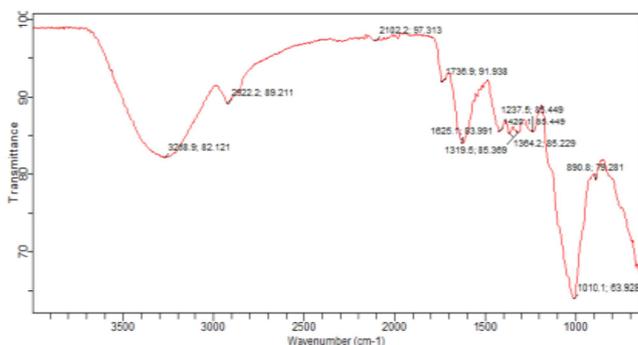


Figure 2. FTIR of  $Cd^{2+}$  loaded *Allium cepa* biomass.

### 3.2. Equilibrium pH

The pH of the adsorption process is significant as it directly affects the chemistry of the active sites on the adsorbent with an attendant effect on the adsorption capacity [24]. A plot of percentage removal against varying pH is presented in Figure 3. The result showed that there was an increase in percentage removal with increase in the pH of the solution. Equilibrium sorption was attained at pH 6 with 98.446% removal. Further increase in the pH of the solution resulted to a decrease in percentage removal. This suggests a low adsorption capacity of the *Allium cepa* biomass in alkaline medium which is an indication that the solubility of the metal ions in the aqueous solution is affected by the availability of hydrogen ions however, at high pH, these ions are precipitated to hydroxides [25].

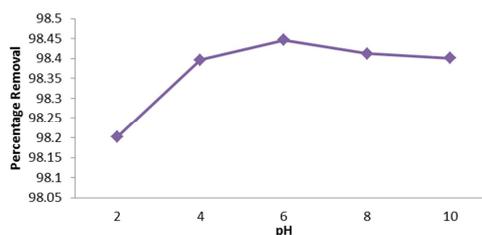


Figure 3. Percentage removal of  $Cd^{2+}$  as a function of pH.

### 3.3. Equilibrium Metal Ion Concentration

Figure 4 represents the plot of percentage removal against varying metal ion concentration. The result revealed a steady increase in percentage removal with an increase in the metal ion concentration. Equilibrium concentration was attained at  $70 \text{ mgL}^{-1}$  with 98.89% removal. Further increase in the concentration of metal ions in the solution, resulted to a reduction in the percentage removal. This suggests unavailability of active sites for metal ion uptake due to high saturation of the binding sites by metal ions [26].

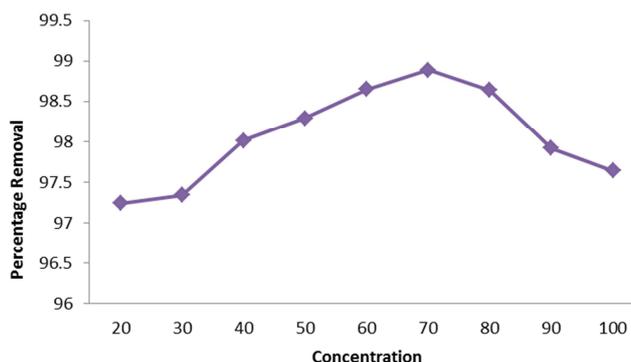


Figure 4. Percentage removal as a function of metal ion concentration.

### 3.4. Equilibrium biosorbent Dosage

Figure 5 represents a plot of percentage removal against biosorbent dosage. The result revealed an increase in percentage removal with an increase in biosorbent dosage. Maximum sorption was attained at  $0.80 \text{ g}$  with 99.92% removal. Further increase in biosorbent dosage yields a decrease in percentage removal. This is attributed to high competition for the available metal ions in the solution by the active sites.

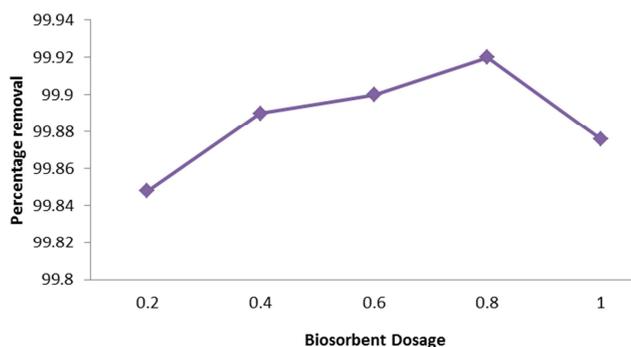


Figure 5. Percentage removal as a function of Biosorbent dosage.

### 3.5. Equilibrium Contact Time

Figure 6 represents a plot of % removal against contact time. The curve revealed an increase in % removal with contact time. This increase in % removal is attributed to the availability of free binding sites on the adsorbent. Equilibrium sorption was attained at 80 mins with 98.98% removal. Thereafter, increases in the contact time yield a decrease in the percentage removal. This is an indication of

unavailability of free binding sites on the adsorbent [9].

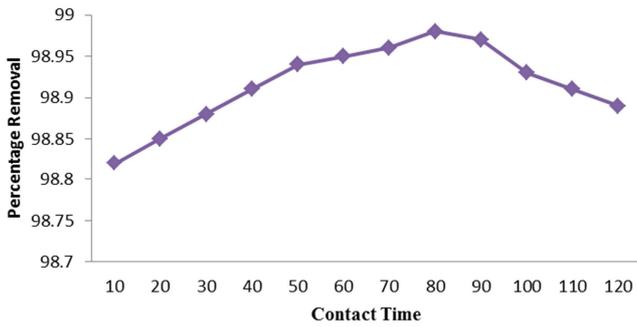


Figure 6. Rate of removal of Cadmium.

3.6. Adsorption Kinetics

3.6.1. Pseudo First Order Kinetic Model

Figure 7 represents the pseudo first order kinetic model for Cd<sup>2+</sup> sorption. The correlation parameters are presented in Table 1. The q<sub>e</sub> value (4.37 mgg<sup>-1</sup>) is an indication that significant amount of Cd<sup>2+</sup> was adsorbed at equilibrium [14]. The correlation coefficient (R<sup>2</sup>)=0.869 suggest that the experimental data fits pseudo first order kinetic model.

3.6.2. Pseudo Second Order Kinetic

Figure 8 represents the pseudo second order kinetic for Cd<sup>2+</sup> sorption, while the correlation parameters are presented in Table 1. The q<sub>e</sub> value (3.48 mgg<sup>-1</sup>) indicates that significant amount of Cd<sup>2+</sup> was absorbed by the biosorbent.

The correlation coefficient R<sup>2</sup>=1 indicates that the experimental data fits pseudo second order kinetic model.

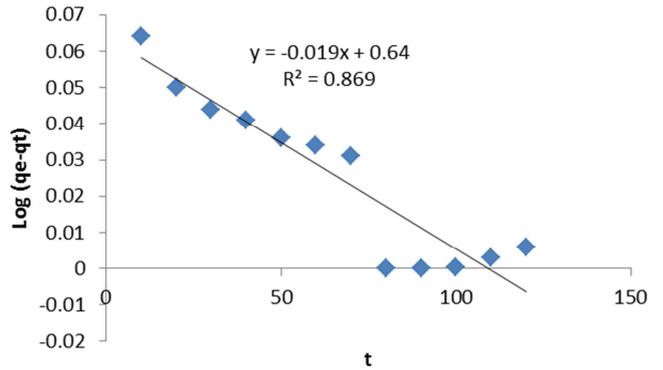


Figure 7. The pseudo first order kinetic model for Cd<sup>2+</sup> sorption.

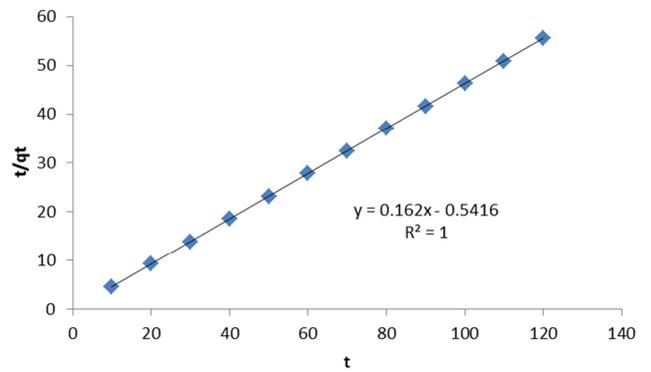


Figure 8. The pseudo second order kinetic for Cd<sup>2+</sup> sorption.

Table 1. Adsorption parameters for Pseudo First and Second order kinetic models.

| Pseudo First Order |                       |                                     |                | Pseudo Second Order   |                           |                |
|--------------------|-----------------------|-------------------------------------|----------------|-----------------------|---------------------------|----------------|
| Metal ion          | q <sub>e</sub> (mg/g) | K <sub>1</sub> (min <sup>-1</sup> ) | R <sup>2</sup> | q <sub>e</sub> (mg/g) | K <sub>2</sub> (g/mg/min) | R <sup>2</sup> |
| Cd <sup>2+</sup>   | 4.37                  | 0.0438                              | 0.869          | 3.48                  | 0.3730                    | 1              |

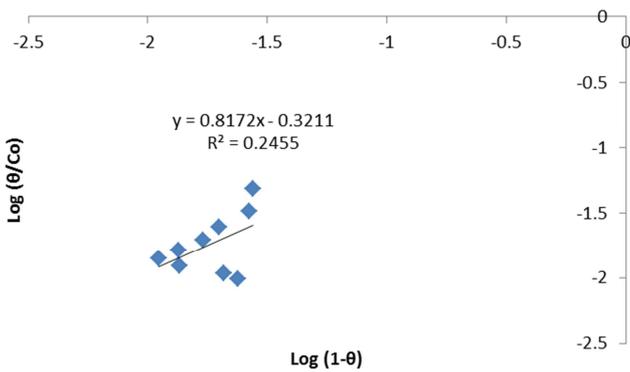


Figure 9. The Flory-Huggins adsorption isotherm for Cd<sup>2+</sup> sorption.

3.7. Adsorption Isotherm

3.7.1. Flory-Huggins Isotherm

The Flory-Huggins adsorption isotherm for Cd<sup>2+</sup> sorption is presented in Figure 9. The isotherm parameters are presented in Table 2. The result reveals that the experimental data obtained fits the Flory-Huggins adsorption isotherm as indicated by a correlation coefficient value R<sup>2</sup>=0.245 while the Flory-Huggins constant (K<sub>FG</sub>)=-2.0941 and the energy of

interaction between the absorbed molecules n=0.817. This indicates there is a low energy of interaction between the adsorbates [27]. The Gibbs free energy=55.05 KJmol<sup>-1</sup> suggests an enthalpy change in the overall physiochemical process.

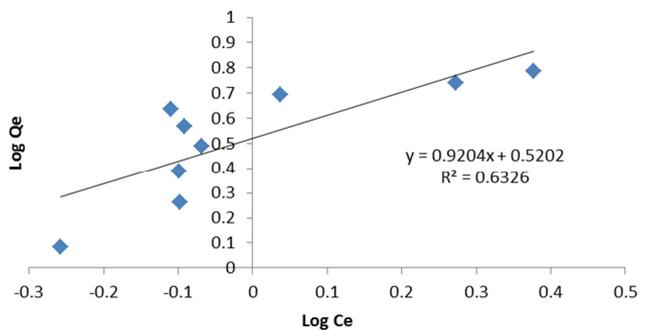


Figure 10. Freundlich Adsorption isotherm for Cd<sup>2+</sup> sorption.

3.7.2. The Freundlich Isotherm

The Freundlich isotherm for Cd<sup>2+</sup> sorption is presented in Figure10. The Freundlich isotherm parameters for Cd<sup>2+</sup> sorption are presented in Table 2. The result revealsthat the

experimental data fits Freundlich isotherm as indicated by the correlation coefficient value  $R^2=0.632$ . The Freundlich adsorption capacity  $K_F=3.3113$  indicates that a significant amount of  $Cd^{2+}$  was absorbed per site. The adsorption intensity ( $n$ )= $1.870 \text{ Lmg}^{-1}$  suggests a favorable adsorption of  $Cd^{2+}$  by *Allium cepa* biomass [28].

### 3.7.3. The Jovanovic Isotherm

The Jovanovic isotherm for  $Cd^{2+}$  sorption is presented in Figure 11. The isotherm parameters are presented in Table 2. The result reveals that experimental data obtained fits Jovanovic adsorption isotherm as indicated by a correlation coefficient ( $R^2$ )= $0.517$ . The maximum uptake  $q_{max}=1.6242 \text{ mgg}^{-1}$  suggests that a significant amount of  $Cd^{2+}$  was absorbed by the biosorbent. The experimental data also suggests that the biosorbent has a poor retention capacity as indicated by Jovanovic constant ( $K_J$ )= $0.637$  [19].

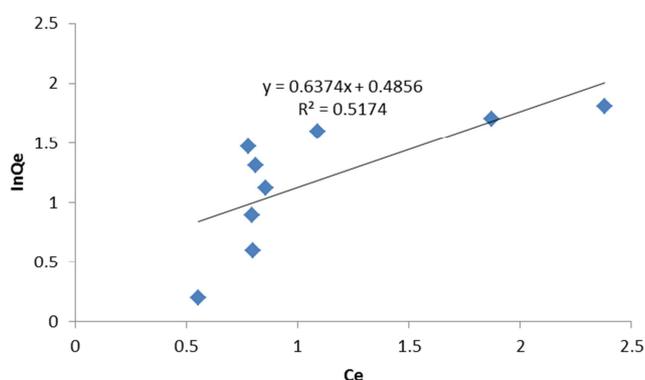


Figure 11. The Jovanovic isotherm for  $Cd^{2+}$  sorption.

### 3.7.4. The Langmuir Isotherm

The Langmuir isotherm for  $Cd^{2+}$  sorption is present in Figure 12. The isotherm's parameters as presented in Table 2. The result revealed that the experimental data did not conform to Langmuir isotherm as indicated by a correlation coefficient ( $R^2$ )= $0.063$ . The  $Q_{max}=2.0739 \text{ mgg}^{-1}$  suggests a significant amount of  $Cd^{2+}$  was absorbed by *Allium cepa* biomass, However  $K_L=0.1203$  suggest a low biosorption capacity of the *Allium cepa* biomass in removal of  $Cd^{2+}$  from the aqueous solution of human blood plasma. The value of  $R_L=0.0141$  suggests that adsorption process was favourable [29].

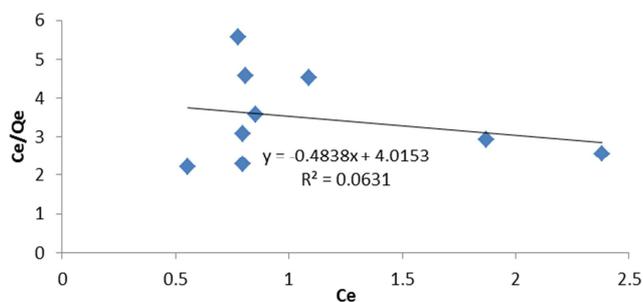


Figure 12. The Langmuir isotherm for  $Cd^{2+}$  sorption.

Table 2. Isotherm Parameters for  $Cd^{2+}$  sorption using *Allium cepa* biomass.

| Fowler Geggenheim | Freundlich   |
|-------------------|--------------|
| $K_{FG}=2.0941$   | $K_F=3.3113$ |
| $R^2=0.245$       | $R^2=0.632$  |
| $N=0.817$         | $n=1.870$    |

| Jovanovic        | Langmuir     |
|------------------|--------------|
| $K_J=0.637$      | $K_L=0.1203$ |
| $R^2=0.517$      | $Q_m=2.0739$ |
| $q_{max}=1.6242$ | $R^2=0.063$  |
|                  | $R_L=0.0141$ |

## 4. Conclusion

The study revealed that the efficacy of *Allium cepa* for the decontamination of aqueous solution of human blood plasma was dependant on pH, metal ion concentration and biosorbent dosage. The experimental data was well interpreted by the Freundlich isotherm. Though the experimental data did not conform to Langmuir isotherm, however the Langmuir separation factor suggests that the overall adsorption process was favourable. The study further revealed adsorption process followed a pseudo second order reaction path way. The Flory-Huggins isotherm revealed a low energy interaction between the adsorbates in the aqueous solution of human blood plasma. *Allium cepa* has proven to be an economically available and effective biosorbent for detoxification of

$Cd^{2+}$  from the aqueous solution of human blood plasma invitro. However further study can be done on chemically modified *Allium cepa* extract for removal of toxic metals from the human body in-vivo.

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