

## THE BIOSORPTION OF CADMIUM AND LEAD IONS FROM AQUEOUS SOLUTION USING *MUSA PARADISIACA* STALK

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### ABSTRACT

The biosorption potentiality of *Musa paradisiaca* stalk at removing cadmium and lead ions from aqueous solution was investigated. The biosorption experiment was carried out as a function of contact time, initial pH, initial metal ion concentration and biosorbent dose. Adsorption equilibria were obtained from batch experiment and described using the Langmuir and Freundlich isotherm models. Kinetics of adsorption was fitted into the pseudo - first and second - order kinetic equations to quantitatively describe cadmium and lead uptake from aqueous solution. The results showed that the maximum biosorption capacity was 7.73 mg/g and 6.78 mg/g for cadmium and lead ion respectively at 300 K and a pH of 6. At a constant concentration of adsorbate, cadmium ion reached saturation faster than the lead ion. The biosorption kinetics followed the pseudo second model for the two ions.

**Keywords:** *Musa paradisiaca*, Biosorption, Cadmium, Lead, Isotherm

### INTRODUCTION

Human activities, development in science and technology and the exploitation of various world natural resources over many decades have culminated into the generation of toxic wastes and harmful substances into the environment. Unfortunately, majority of these wastes especially heavy metals are persistence and not subjected to deterioration or biochemical decomposition (Ruello *et al.*, 2011). Therefore, they find their way into the environment and get accumulated into the human body through water, air and along the food chains.

Heavy metals such as cadmium, lead and mercury have been classified as the three most destructive pollutants in the environment, highly detrimental to health even at low concentrations (Jalali *et al.*, 2002). These metals are released into the environment through industrial activities such as ore refining, sludge disposal, processing of radioactive materials, metal plating or the manufacture of electrical equipment, paints, alloys, batteries, pesticides or preservatives (Ahalya *et al.*, 2003). Lead has been reported to cause certain health problems ranging from behavioural problems and learning disabilities to seizures and death (Deng *et al.*, 2007). In individual of all ages, Pb may cause anaemia, kidney disease,

brain damage, impaired function of the peripheral nervous system, high blood pressure, reproductive abnormalities, developmental defects, abnormal vitamin D metabolism, and in some situations death (U.S.EPA, 1992; Hrudey, 1995). Similarly, the presence of cadmium in the human body could result into kidney malfunction (Jarup, 1998). The kidneys lose their function to remove acids from the blood in *proximal renal tubular dysfunction*. The kidney damage inflicted by cadmium poisoning is irreversible. The *proximal renal tubular dysfunction* creates low phosphate levels in the blood (hypophosphatemia), causing muscle weakness and sometimes coma. The dysfunction also causes gout, a form of arthritis due to the accumulation of uric acid crystals in the joints because of high acidity of the blood (hyperuricemia). Another side effect is increased levels of chloride in the blood (hyperchloremia). The kidneys can also shrink up to 30% (Jarup, 1998).

Various methods have been used for the treatment and removal of metals from the environment which include chemical precipitation, ultra-filtration, electro-dialysis, ion exchange, reverse osmosis, phytoremediation, precipitation, evaporation, electroplating, membrane processes, solvent extraction and ultra filtration (Deng *et al.*,

2007). Apart from biosorption, most of these methods are inefficient especially at low concentration of metals. Other problems associated with these conventional methods range from high cost and energy requirement, generation of large volume of toxic sludge which is difficult to handle and dispose, to incomplete removal of certain ions (Cordero, 2004; Hubbe *et al.*, 2011). On the other hand, biosorption is relatively effective, cheaper and easy to set up. Besides, the biosorbents are renewable, recyclable and biodegradable. Large volume of agricultural by products and residues abound and can be readily utilized as adsorbents for most of these metals. A typical example is *Musa paradisiaca* (plantain) stalk which is relatively abundant in major Nigerian markets with no specific usage but rather constitute pollution and waste disposal problems.

The present work was aimed at investigating the feasibility of using *Musa paradisiaca* stalk for the removal of cadmium and lead metals from aqueous solution. Kinetic experiments were performed to study the effect of the solution pH value, initial metal ion concentration, biosorbent dose and contact time on biosorption. A range of equilibrium adsorption isotherms (Langmuir and Freundlich) and kinetic models (pseudo-first-order and pseudo-second-order) were used to quantitatively describe cadmium and lead uptake from aqueous solution.

## MATERIALS AND METHODS

### Preparation of Biomass

Samples of *Musa paradisiaca* stalks were collected from Oje market in Ibadan, Oyo State, Nigeria. The stocks were washed twice with running water and once with deionized water. After washing, it was oven-dried at 60°C for 24 hr, crushed with an analytical mill and sieved to a size fraction of 0.5 - 1.0 mm before being stored in paper envelopes. All chemicals used in the study were of analytical grade and solutions were prepared using deionized water. Experimental stock solutions of 1000 mg/L of Cd (II) and Pb (II) metal ions were prepared by dissolving 2.751 g Cd(NO<sub>3</sub>)<sub>2</sub> · 4H<sub>2</sub>O and 1.599 g Pb(NO<sub>3</sub>)<sub>2</sub> separately in deionized water and making it up to 1000ml in standard flasks. Working solutions were prepared from the stock solution by serial dilution to different initial

concentrations for the sorption experiment.

### Biosorption Experiment

The biosorption experiment was conducted at 27°C to study the effect of initial solution pH, ion concentration, contact time and adsorbent dosage on the adsorption of lead II and cadmium II ions (Babarinde *et al.* 2009). Each experiment was conducted in a thermostated water bath (Haske Wia model) and the residual metal ions were analyzed using Atomic Absorption Spectrophotometer (AAS).

### Effect of pH

Experiments were conducted at 27°C to study the effect of initial solution pH on the adsorption of the metal ions by contacting 0.5 g of the plantain stalk with 25ml of 25 mg/L solutions of each of lead and cadmium salt in a 250 ml bottle. The pH of each of the metal solution was adjusted to the desired value with either 0.1 M sodium hydroxide or 0.1 M nitric acid. Experiment was conducted at pH values of 2, 3, 4, 5, 6 and 7. The reaction mixture was agitated on a shaker at 150 rpm for 30 minutes after which it was filtered to separate the supernatant, and residual pH measured. About 5 ml of the supernatant was then taken for the analyses of the residual metal content by AAS. Each experiment was carried out in duplicate

### Effect of Biosorbent Dose

Powdered plantain stalk (0.25 g - 2.0 g) were added to 25 ml of 25 ppm solutions of each of the metals at the neutral pH for both Cd<sup>2+</sup> and Pb<sup>2+</sup>. The solutions of the mixtures were agitated for 30 mins on a shaker at 150 rpm and then filtered. The residual metals in each of the supernatant were then determined by Atomic Absorption Spectroscopy (AAS). Each experiment was carried out in duplicate. A biomass dose of 0.5g was chosen for all the other experiments.

### Effect of Contact Time

The adsorption of the metal ions by plantain stalk was studied at various time intervals ranging from 0 - 120 minutes. A constant concentration of 25 mg/L each of Cd and Pb solutions was used. The agitated solution mixtures were filtered. 5 ml of each of the filtrate was determined for residual metal ion concentration by the Atomic Absorption Spectroscopy (AAS).

### Effect of Initial Concentration on Biosorption

Sample of powdered plantain stalk powdered (0.15g) was added to 10–100 mg/L of the metal solutions (25 ml) at a temperature of 27°C and at the experimental pH and equilibration time of 1 hour. After the mixtures were filtered, 5ml of each filtrate was determined for residual metal ion concentration. The uptake of metal ion was calculated by concentration difference method as follows:

$$q_e = (C_i - C_e)V/1000w \quad (1)$$

where  $q_e$  is the metal uptake or adsorptive capacity (mg metal adsorbed/g adsorbent),  $C_i$  and  $C_e$  are the initial and equilibrium or final concentration of metal ion at various time intervals (mg/L),  $V$  is the volume of the solution in  $\text{dm}^3$  and  $w$  the mass of the sorbent in g.

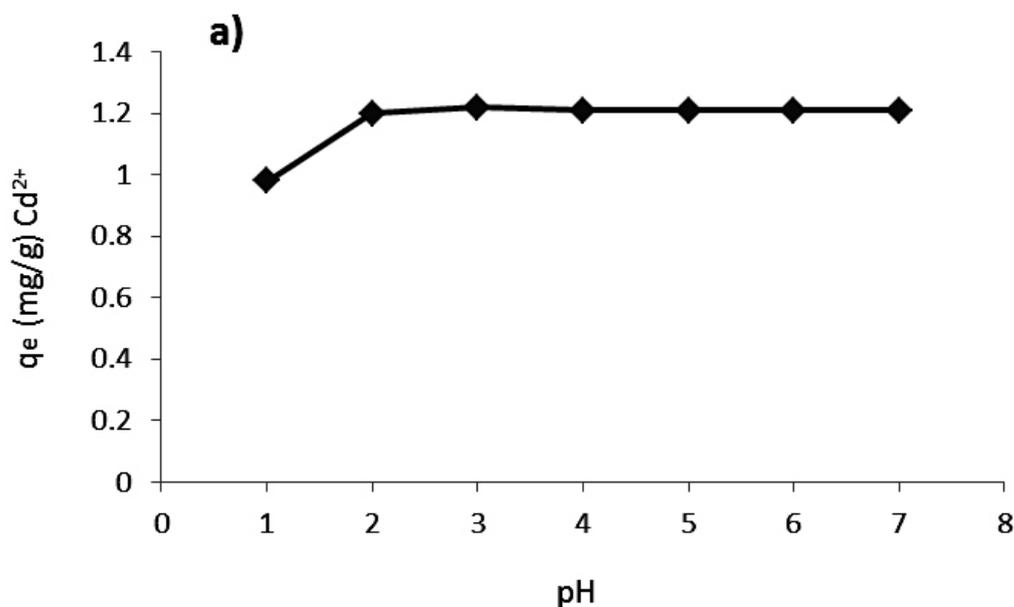
Kinetic data were generated by contacting 0.5g of

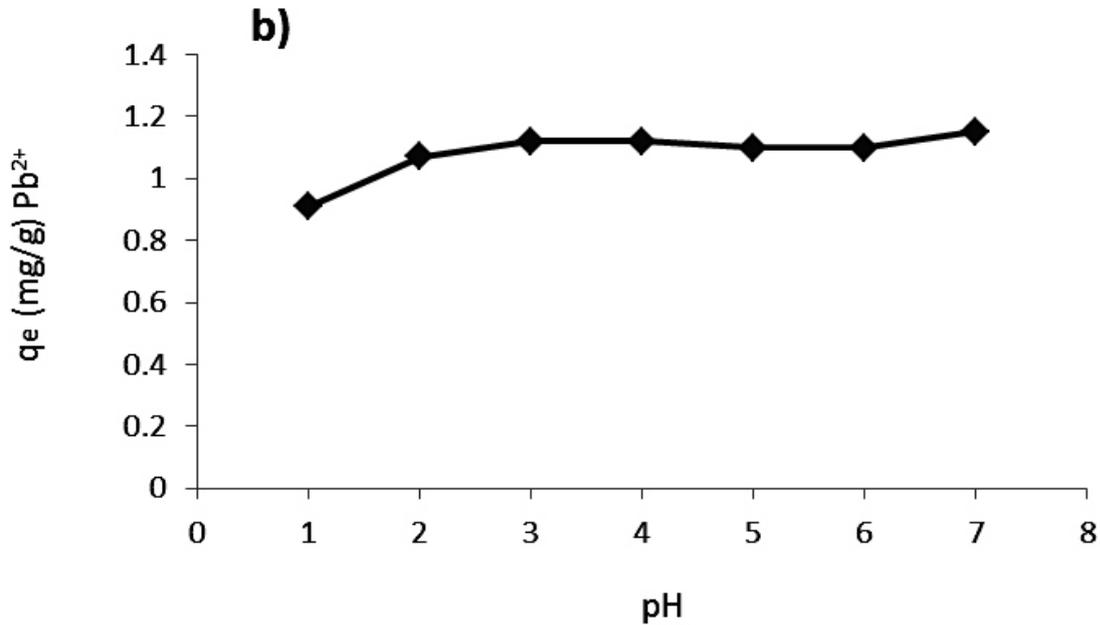
the adsorbent with 25 mg/L of each metal solution in plastic bottles. The bottles were shaken at 150 rpm for different time intervals (ranging from 2–120 minutes). At different interval of time, the reaction mixture was filtered and 5 ml of the supernatant was taken for AAS analysis.

## RESULTS AND DISCUSSION

### Effect of pH on Biosorption

The biosorption capability of *Musa paradisiaca* stalk for cadmium and lead, at different pHs (1–7), were illustrated in Figures 1(a) and (b) respectively. The adsorption of  $\text{Cd}^{2+}$  ion remained constant with increase in pH from 3 to 7. Similar observations were made by Parvathi *et al.* (2007) and Mohammadi *et al.*, (2012). The same trend was also observed for  $\text{Pb}^{2+}$  ion. Metal uptake increased from pH 1 to 3 and then remains constant up to pH of 6. At pH of 7, the solution became slightly cloudy, getting ready to precipitate. Any further increase in pH will cause precipitation.





**Figure 1:** Effect of pH on (a) Cd<sup>2+</sup> and (b) Pb<sup>2+</sup> Uptake by the *Musa paradisiaca* stalk (Temperature = 27°C; time: 30 min; agitation rate 150 rpm)

#### Effect of Biosorbent Dose

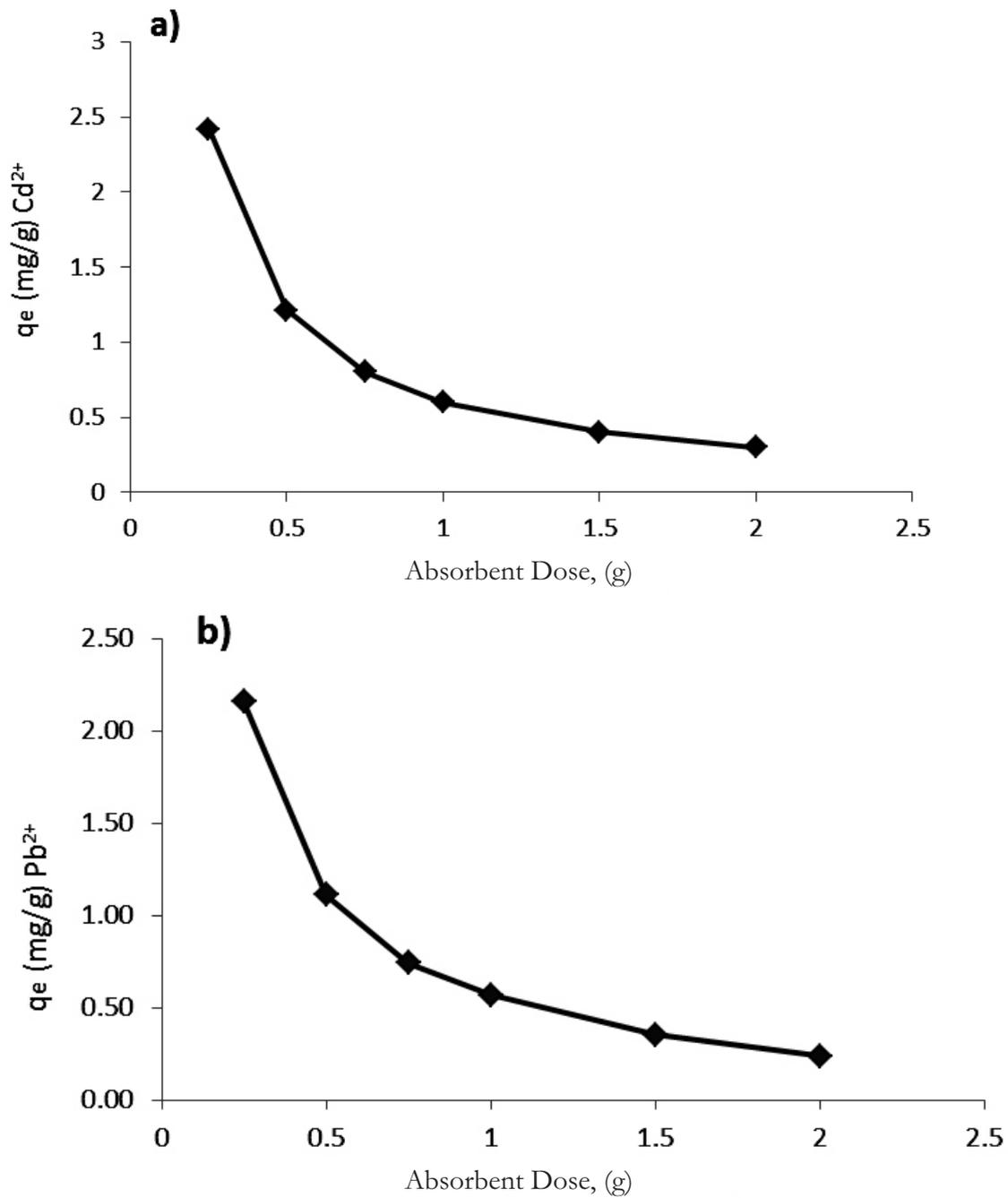
Biosorbent dose seemed to have a great influence in biosorption process. Dose of biomass added into the solution determine the number of binding sites available for adsorption (Zafar *et al.*, 2006). The results and plots of the effect of *Musa paradisiaca* stalk dose (Figures 2(a) and (b)) showed that metal uptake values increased with a decrease in biomass dose. This could be attributed to interference between binding sites at higher concentrations (De rome and Gadd, 1987). Similar observations were reported by other researchers (Sampedro *et al.*, 1995). Reduction in metal uptake with increasing biomass concentration was attributed to insufficiency of metal ions in solution with respect to available binding sites (Fourest and Volesky, 1992). Higher specific uptake at lower dry mass concentrations could be due to an increased metal-to-biosorbent ratio, which decreases upon an increase in dry mass concentration.

#### Effect of Contact Time

The kinetic study of *Musa paradisiaca* stalk with Cd<sup>2+</sup> and Pb<sup>2+</sup> ions in aqueous solution is presented in Figures 3 (a) and (b). Optimum adsorption was observed for Cd<sup>2+</sup> ion at 4 min of agitation, after which equilibrium was reached and the adsorption remains constant with further increase in time. For Pb<sup>2+</sup> ions, adsorption reaches saturation in the first few minutes and then remains constant with increase in time.

#### Effect of Initial Metal Concentration

Experimental data points obtained for the biosorption of cadmium and lead by *Musa paradisiaca* stalk at several concentrations are shown in Figures 4 (a) and (b). The result clarifies the relation between capacities and the metal ion concentrations, which shows that as the metal ion concentration increased, metal uptake also increased. In general, the data indicated that



**Figure 2:** Effect of Biosorbent Dose on (a) Cd<sup>2+</sup> and (b) Pb<sup>2+</sup> Uptake by the *Musa paradisiaca* stalk (Temperature = 27°C; time = 30 min; agitation rate = 150 rpm; pH = 6)

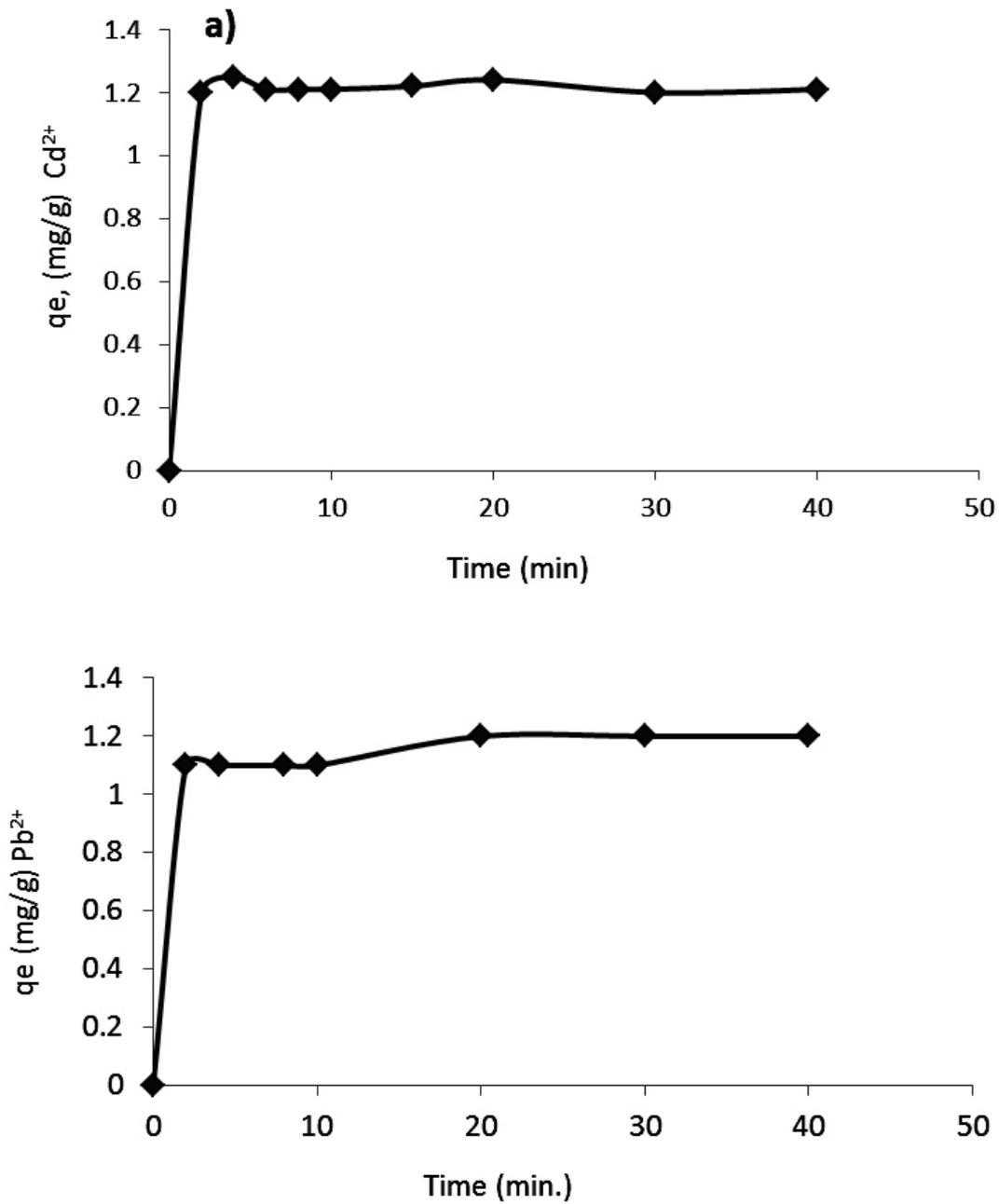
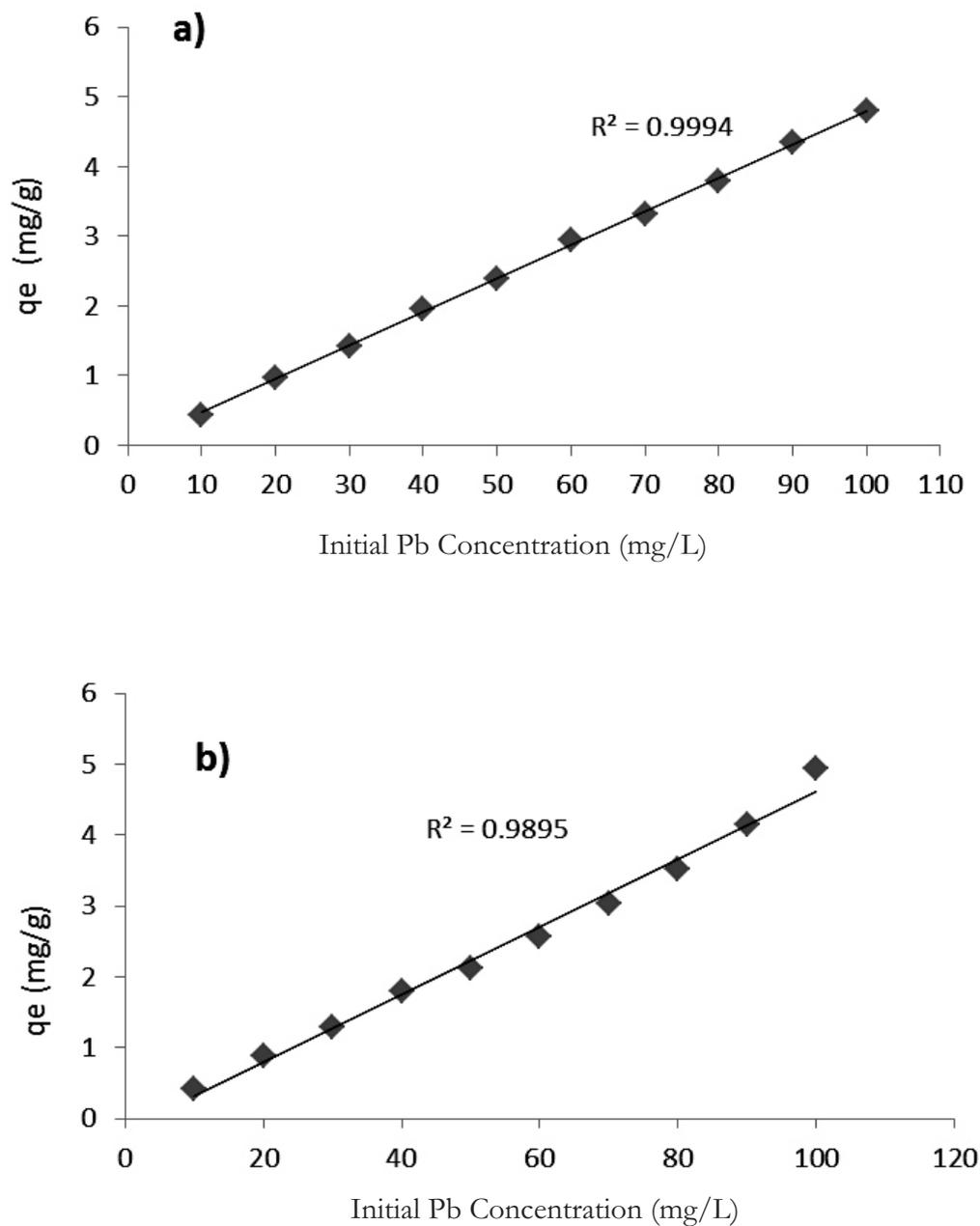


Figure 3: Effect of Contact Time on (a) Cd<sup>2+</sup> and (b) Pb<sup>2+</sup> Uptake by the *Musa paradisiaca* stalk (Temperature = 27°C; agitation rate = 150 rpm; pH = 6)



**Figure 4:** Effect of Concentration on (a)  $Cd^{2+}$  and (b)  $Pb^{2+}$  Uptake by the *Musa paradisiaca* stalk ( $C_o = 10-100$  mg/L; Temperature =  $27^\circ C$ ; time 60 min; agitation rate 150 rpm; pH = 6)

sorption capacity increased with increase in initial metal ion concentration for the two metals on the biomass. Thus, sorption characteristic indicated that surface saturation was dependent on the initial metal ion concentrations. The equation obtained from the plot can be used to predict the sorption capacity of the stalk at a given concentration of metal ions.

### Isotherm Model

The Langmuir isotherm (Langmuir, 1916) model assumes in its formulation a monolayer adsorption at maximum capacity occurring through a uniform or equivalent surface of adsorbent without any interaction between the adsorbate molecules. The equation is expressed as follows:

$$q_e = q_m b C_e / (1 + b C_e) \quad (2)$$

where  $q_e$  (mg/g) and  $C_e$  (mg/L) are the concentration of the adsorbate solution in the adsorbent and in solution at equilibrium respectively,  $q_m$  is the maximum sorption capacity (mg/g) and  $b$  is the Langmuir constant related to the energy of the adsorbate for the adsorbent (L/g).

The linearised form of the equation is given as:

$$C_e / q_e = 1 / b q_m + C_e / q_m \quad (3)$$

The Freundlich isotherm (1906) relates the solute concentration on the adsorbent surface to the solute concentration in the liquid phase. The isotherm assumes that adsorption occur on a heterogeneous adsorbent surface, i.e. multilayer adsorption.

Accordingly, Freundlich proposed the equation:

$$q_e = K_f C_e^{1/n} \quad (4)$$

which on linearisation becomes

$$\log q_e = \log K_f + 1/n \log C_e \quad (5)$$

where  $K_f$  and  $1/n$  are the Freundlich coefficients related to biosorption capacity and biosorption

intensity respectively.

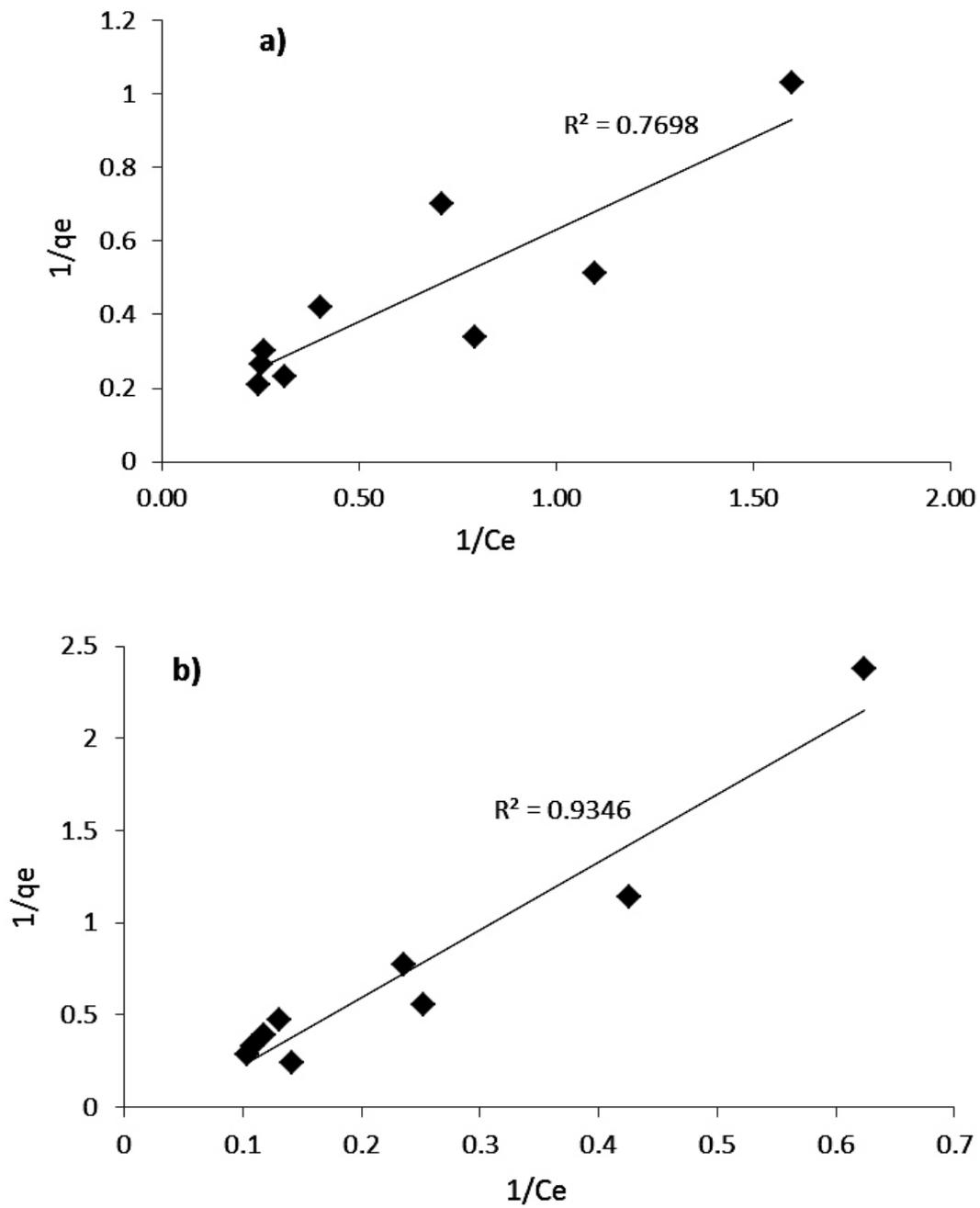
The Langmuir and Freundlich isotherms for the adsorption of Cd(II) and Pb(II) are shown in Figures 5 and 6 respectively. The parameters for these isotherms are summarized in Table 1. The results showed that the biosorption data for the lead ion gave better fit for both Langmuir and Freundlich isotherm with correlation coefficients of 0.94 and 0.88 respectively than the cadmium ion. As indicated in Table 1, the monolayer adsorption capacity  $q_m$  (mg/g) of Cd and the Langmuir constant  $b$  (L/g) related to the energy of adsorption are higher than for Pb. The  $q_m$  values for both ions were quite lower than those reported for *Cassia fistula* (Golden Shower) biomass (Hanif *et al.*, 2007). The  $K_f$  value of the metal ions also indicated that cadmium ion had greater capacity towards the adsorbent than the lead ion. This may be attributed to the differences in hydrated ionic sizes of the metals in solution. Cadmium ion being smaller than lead ion, not only migrate faster but occupied more space than lead ion (Babarinde *et al.*, 2009). The intensity of the sorption of cadmium ion unto plantain stalk implied that it is a chemisorption process ( $1/n < 1$ ) while that of lead ion suggested cooperative adsorption ( $1/n > 1$ ) (Haghseresht and Lu, 1998)

### Kinetic Model

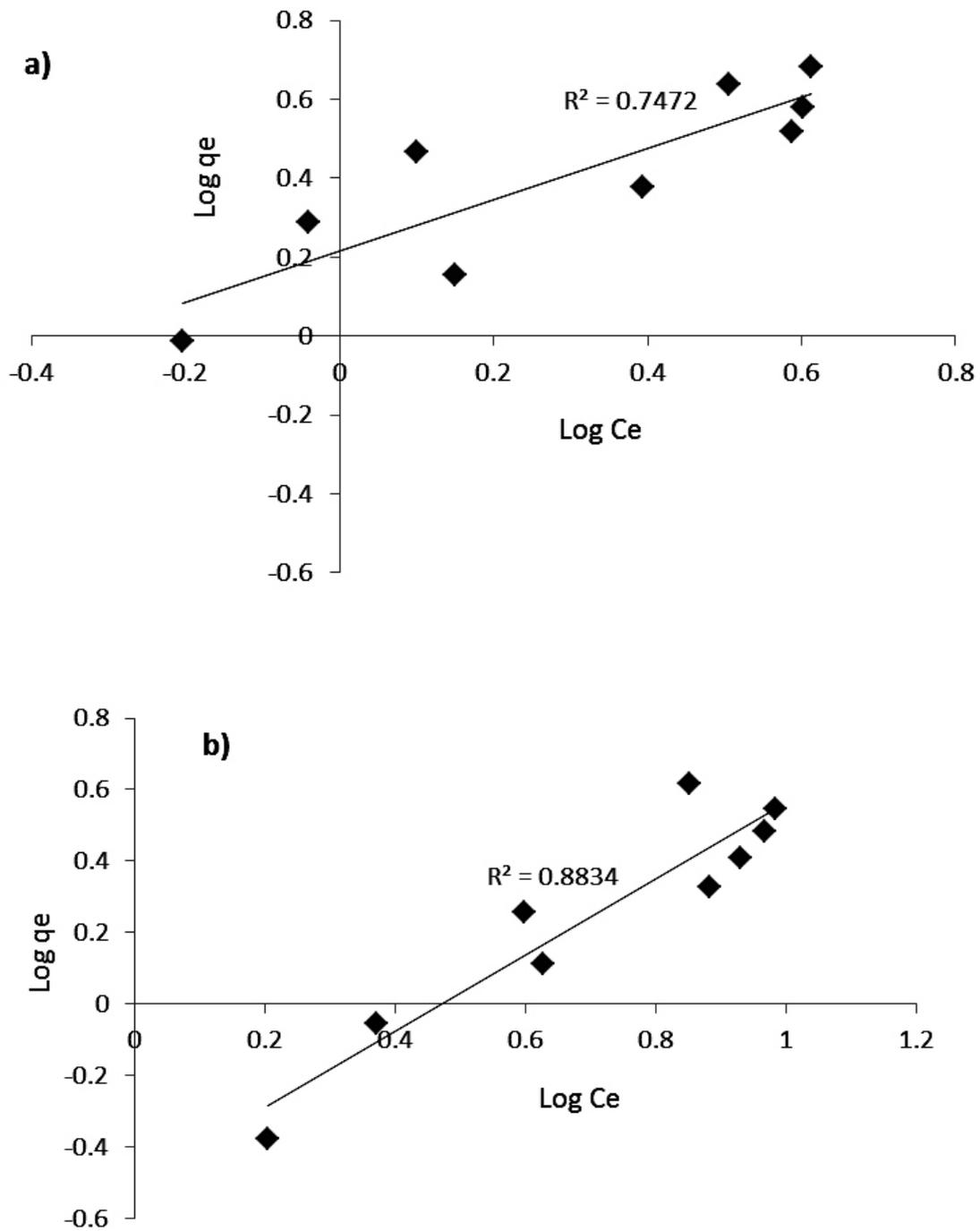
Biosorption kinetic is an important indicator of the rate and mechanism of the biosorption process (Augustine and Yuh-Shan, 2007). The linearised form of the pseudo-first order kinetic of Lagergren (1898) is given in the form:

$$\log (q_e - q_t) = \log q_e - (k_t / 2.303) t \quad (6)$$

where  $q_e$  and  $q_t$  are the amount of dye sorbed per unit weight of sorbent at equilibrium and at time  $t$  (min) respectively,  $k_t$  is the rate constant of the pseudo-first order sorption ( $\text{min}^{-1}$ ). The value of the rate constant ( $k_t$ ) is obtained from the linear plot of  $\log (q_e - q_t)$  versus  $t$ .



**Figure 5:** Langmuir Isotherm for Biosorption of (a) Cd<sup>2+</sup> and (b) Pb<sup>2+</sup> by *Musa paradisiaca* stalk (C<sub>0</sub> = 10–100mg/L; dosage = 0.5 g; pH = 6)



**Figure 6:** Freundlich Isotherm for Biosorption of (a)  $\text{Cd}^{2+}$  and (b)  $\text{Pb}^{2+}$  by *Musa paradisiaca* stalk ( $C_o = 10-100\text{mg/L}$ ; dosage = 0.5 g; pH = 6)

**Table 1:** Isotherm Parameters for the Sorption of Cd and Pb ions using *Musa paradisiaca* stalk

Isotherms	Parameters	Values for Cd	Values for Pb
Langmuir	$q_m$ (mg/g)	7.73	6.78
	$b$ (L/mg)	0.26	0.04
	$R^2$	0.77	0.94
Freundlich	$1/n$	0.65	1.07
	$K_f$	1.64	0.31
	$R^2$	0.75	0.88

The pseudo-second order rate equation (Ho *et al.*, 2006) can be expressed as:

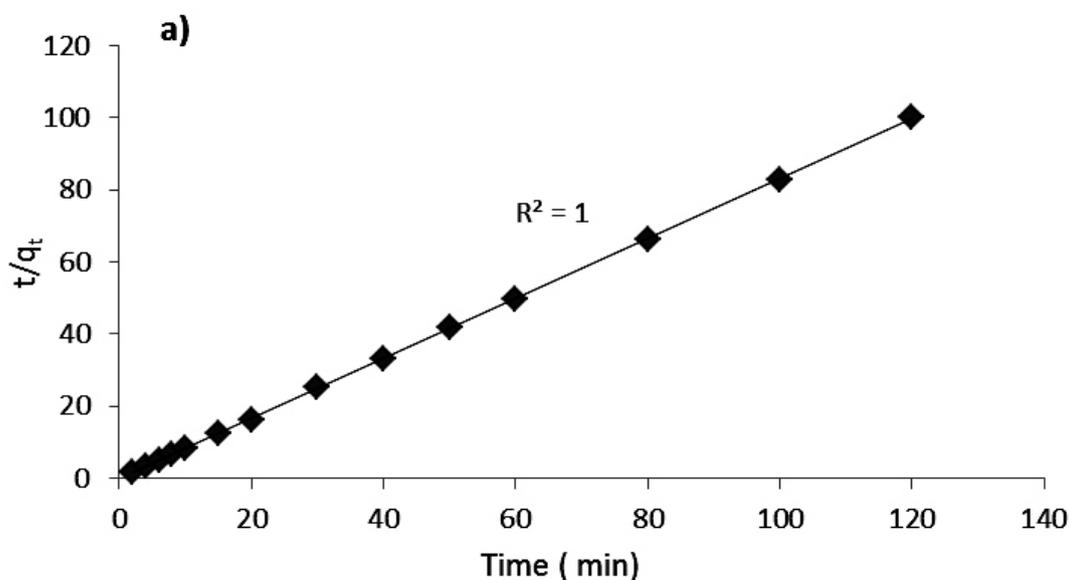
$$d_q/d_t = k_2(q_e - q_t)^2 \quad (7)$$

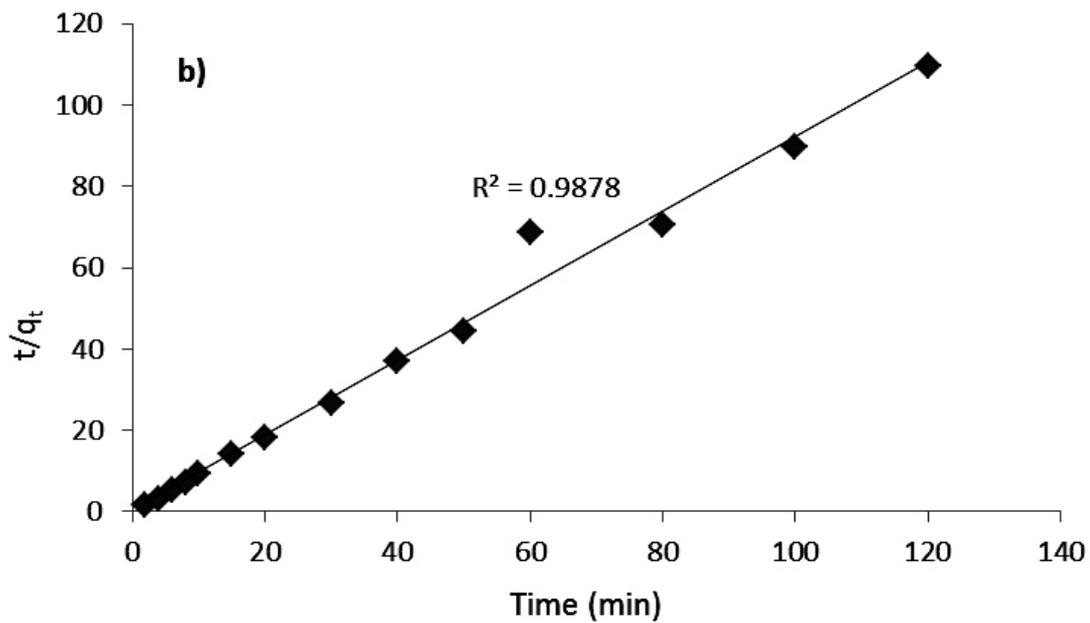
where  $k_2$  is the equilibrium rate constant of the pseudo-second order sorption in g/mg min. Upon rearrangement and integration, the equation becomes

$$t/q_t = 1/(k_2 q_e^2) + (1/q_e) t \quad (8)$$

The plot of  $t/q_t$  versus  $t$  should give a linear plot of slope  $1/q_e$  and intercept  $1/(k_2 q_e^2)$ .

In order to investigate the mechanism of biosorption of lead and cadmium by *Musa paradisiaca* stalk the kinetic models were used to test experimental data. The pseudo first order model did not give a good fit to the experimental data. The best fits, in the data range, were obtained using the pseudo second order model (Figures 7(a) and (b)), for sorption of both Cd and Pb ions. This suggests that the rate-limiting step is a chemical biosorption process between the metals and the biosorbent. The pseudo second-order model is based on the assumption that biosorption follows a second-order mechanism. So, the rate of occupation of adsorption sites is proportional





**Figure 7:** Pseudo-second-order Plot for the Biosorption of (a)  $\text{Cd}^{2+}$  and (b)  $\text{Pb}^{2+}$  by *Musa paradisiaca* stalk ( $C_0 = 10-100$  mg/L; dosage = 0.5 g; agitation rate = 150 rpm; pH = 6)

to the square of the number of unoccupied sites (Zafar *et al.*, 2006). Similar conclusions were found by Ho (2006), McKay (2006), Donmez and Aksu (2002) and Babarinde *et al.* (2009).

The values of the parameters,  $k_2$  and  $q_e$ , and of correlation coefficients,  $R^2$ , are presented in Table 2 for the two metals studied. It was observed that the rate of biosorption of cadmium is thrice that

of lead from the  $k_2$  values. The likely reason for this may be due to the differences in the hydrated ionic sizes of the two ions in solution. The correlation coefficients obtained in both cases are approximately one ( $R^2 \approx 1$ ), and the adequate fitting of theoretical and experimental  $q_e$  values for all the two metals suggest the applicability of second-order kinetic model.

**Table 2:** Pseudo-second-order Kinetic Model Parameters for Biosorption of  $\text{Cd}^{2+}$  and  $\text{Pb}^{2+}$  using *Musa paradisiaca* Stalk

Metal ion	$q_e$ (mg/g)	$k_2$ (g/mg/min)	$R^2$
Cadmium	1.204	6.154	1
Lead	1.089	1.93	0.9878

## CONCLUSION

The biosorption pattern of *Musa paradisiaca* stalk under the batch experimental conditions indicated that the rate of biosorption of cadmium and lead increases with a respective increase in the initial metal concentration. At a constant value of adsorbate concentration, maximum adsorption was observed at 4.0 minutes of agitation for Cd

ion and 20 minutes for Pb ion. The kinetic model obtained showed negative correlation coefficients with the pseudo first order model while good fitting was obtained using the pseudo second order kinetic model. The equilibrium biosorption studies showed that the biosorption data for the lead ion gave better fit for both Langmuir and Freundlich isotherm with correlation coefficients

of 0.94 and 0.88 respectively than the cadmium ion. The study indicated that *Musa paradisiaca* stalk would be a good biosorbent for the removal of Cd and Pb from industrial effluent.

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