

# Biosorption of $Pb^{2+}$ and $Cd^{2+}$ from Simulated Wastewater Using Melina (*Gmelina arborea*) Tree Leaves Powder

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## Authors' contributions

This work was carried out in collaboration between both authors. Author NOI designed the study, reviewed the literature searches and managed the experimental process and performed statistical analyses of the experiment. Author AAO performed the experimental process and analysis, wrote the first draft of manuscript. Both authors read and approved the final manuscript.

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## ABSTRACT

Increased anthropogenic activities have led to serious environmental problems due to pollution caused by toxic materials such as heavy metals whose levels are rising in the environment. The inefficiency and high cost of conventional methods of waste treatment have prompted the investigation of environmentally friendly and cheaper methods of treatment using natural products. In this study, *G. arborea* leaves powder was investigated with a view of using it as cheap material for the biosorption of  $Pb^{2+}$  and  $Cd^{2+}$  from wastewater. The effects of operational parameters like pH, biosorbent dose [g/L], initial metal ions concentration [mg/L], contact time [minutes] and stirring speed [rpm] on the biosorption efficiency [%] were determined. The optimum solution pH for  $Pb^{2+}$  and  $Cd^{2+}$  adsorption was 5.0 and peak adsorption of 91.33% and 82.53% for  $Pb^{2+}$  and  $Cd^{2+}$ , respectively. 5 g/L Melina leaves were enough to achieve peak removal of both metal ions. The removal of the metal ions was comparatively quick, and stability was achieved after 30 minutes. The optimum stirring speed was 250 rpm for both metal ions. The uptake efficiency of the biosorbent was determined by Langmuir and Freundlich isotherm models. The value of Langmuir isotherm

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separation parameter [RL] of  $Pb^{2+}$  ion [0.0446 - 0.78125] and  $Cd^{2+}$  [0.1005- 0.9482] were within range of 0 -1 indicating favorable biosorption for both metal ions. The degree of non-linearity [n] values between  $Pb^{2+}$  [12.79] and  $Cd^{2+}$  [11.79] solution concentration and biosorption in Freundlich equation were greater than 1, indicating physical biosorption of  $Pb^{2+}$  and  $Cd^{2+}$  on to *G. arborea* leaves. *G. arborea* can serve as efficient biosorbent not only for  $Pb^{2+}$  and  $Cd^{2+}$  ions but also for other heavy metal ions in a wastewater stream

**Keywords:** Biosorption; lead; cadmium; Melina (*Gmelina arborea*); Langmuir isotherm; Freundlich isotherm.

## 1. INTRODUCTION

Elements whose atomic density are above 6 g/cm<sup>3</sup> are heavy metals and example are lead, mercury, arsenic, copper, zinc, cadmium, etc. They are major pollutants of wastewater and freshwater reserves because of their toxic, non-biodegradable and persistent nature, which when consumed can bioaccumulate causing cancer, brain damage, and so forth.

Lead, a metabolic poison, if present in drinking water, can cause mental retardation, anaemia, [1,2], hepatitis and nephropathy [3] damages to different organs of the body [4] The permissible limit of lead in wastewater as set by the Environment Protection Agency is 0.1 mg/L, whereas in drinking water it is 0.05 mg/L [5].

Wastewater discharged into the environment by industries are the principal sources of poisonous heavy metals in the environment. Industries of concern include the paper, pesticides, batteries, electronics and non-ferrous metal works, to mention a few [6,7,8]. Reducing the concentration of heavy metals in industrial effluents to within acceptable limits before discharge into the environment is therefore very necessary. Various methods have been proposed over the years, all aimed at meeting the aforementioned need. Among them are precipitation, electro dialysis, coagulation, floatation, ultra-filtration, etc. These methods, however, are characterised by varying disadvantages such as the production of toxic sludge, technical and economic constraints, low selectivity, etc [9,10,11,12].

The problems of these usual physical and/or chemical methods have necessitated the search for cheap, more eco-friendly, and readily available substitutes like biosorption. Agricultural wastes are abundant and cheap biomaterials (sorbents) of concern with the potential of heavy metal removal from industrial waste effluents. Various authors have reported on the use and

effectiveness of biosorption as a possible substitute for conventional remediation techniques [13,14,15].

**Table 1. Result of Fourier transformed infrared and ultraviolet spectra of chloroform – methanol extract**

Wave number (cm <sup>-1</sup> )	Suspected chromophores
3458.66	O-H stretch for alcohol, phenol and carboxylic acid.
3436.30	O-H stretch for alcohol
3421.83	O-H stretch for alcohol
2855.71	C-H stretch for alkanes
2106.36	C=N stretch for nitriles
1645.33	C=O stretch for ketones, acid amides and esters.
1458.23	C=C stretch for alkenes and aromatics
1105.25	C-O stretch for alcohol, carboxylic acids and esters
1016.52	C-H deformation bonds for alkyl groups.
UV max 277	Indicating highly conjugated trisubstituted
366 and 669	Aromatic compound

Source: Udeozo et al. [18]

*G. arborea*, an important tropical flowering plant that belongs to the Verbenaceae family. It is a hardwood with *gmelina* as its common name. In Nigeria, its Igbo name is *gmelina*, *igi-Melina* in Yoruba and *kalankuwa* in Hausa [16,17]. It is a white teak veware and a fast-growing deciduous tree, occurring naturally throughout the greater part of Asian and African countries and has been widely cultivated in Nigeria especially in Ogun State. Melina sheds its leaves every day and if unutilised becomes an environmental nuisance. The results of the FTIR and UV analysis (Table 1) on Melina leaves carried out by Udeozo et al [18] showed its chemical structure contains anionic functional groups such as carbonyl, hydroxyl, and amino groups that required to react with cationic pollutants in solution.

The ready availability of *G. arborea* leaves and the dearth of information on its use as biosorbent necessitated this work. The objectives of this work were to use powdered *G. arborea* leaves for sorption of  $Pb^{2+}$  and  $Cd^{2+}$  ions from simulated wastewater taking into consideration, the effects of pH, initial metal ion concentration, contact time and agitation speed on biosorption efficiency.

## 2. METHODOLOGY

### 2.1 Sample Collection

Dried matured *G. arborea* leaves were obtained from the Federal Polytechnic, Ilaro, Nigeria. The leaves were rinsed to remove adhering dirt, mopped dry and oven-dried at 80°C for 72 h. Dried leaves were ground and sieved to pass through a 200-300 µm mesh, and stored in an air tight container until required.

### 2.2 Preparation of Solution

Analytical grade reagents that required no further purification were used to prepare standard stock solutions of  $Pb^{2+}$  and  $Cd^{2+}$ . Working solutions ranging from 10 mg/L to 100 mg/L metal ions were prepared by serial dilutions of the stock solutions using deionised water and maintaining the initial pH of the solution.

### 2.3 Batch Biosorption Study

The biosorption study was carried out by the procedure similar to those earlier reported by Babarinde et al. [19]. The experiment was performed in triplicates by stirring 0.5 g of the dried Melina leaf powder with 30 ml of the metal ion solution on a thermostated hot plate magnetic stirrer (Corning™ PC-420D) at constant temperature of 30°C varying the experimental conditions (pH, contact time, initial metal ion concentration, adsorbent dose, and agitation speed) for a period of time in a boiling tube. The remaining metal ion in the solution was determined using Atomic Absorption Spectrophotometer (Bulk Scientific model 210). The amount of metal ion removed from the solution was the difference between the concentration pre and post adsorption of each metal ion. The equilibrium biosorption capacity  $q_e$  (mg/g) as the difference between the initial and final equilibrium concentration of metal.

$$q_e = (C_o - C_e)V/m \quad (1)$$

Also, the % Removal at equilibrium was determined from this equation:

$$\% \text{ Removal} = \frac{[C_o - C_e]}{C_o} \times 100 \quad (2)$$

Where,  $C_o$  (mg/L) is the initial metal ions concentration,  $C_e$  (mg/L) is the equilibrium concentration of metal ions,  $m$  is the mass of Melina leaves used (g) and  $V$  is the volume of the solution (L).

### 2.4 Effect of pH on Biosorption

The effect of pH on the biosorption of the metal ion was carried out within the range that would not be influenced by the metal precipitated. The method used is comparable to Babarinde et al. [19]. 0.5 g of Melina leaves were contacted with 30 ml of 10 mg/l metal ion solution in a boiling tube. 0.01 M Hydrochloric acid (HCl) and/or 0.01 M Sodium Hydroxide (NaOH) added in dropwise to bring the pH of each solution to the desired value. The studies were conducted at pH 2, 3, 4, 5, 6 and 7. The conical flask containing the mixture was left on a thermostated hot plate magnetic stirrer (Corning™ PC-420D) at the speed of 250 rpm for 30 minutes. The residue was obtained by decanting the supernatant and the remaining metal ions concentration in the solution were determined.

### 2.5 Effect of Contact Time on Biosorption

The adsorption of the metal ions by Melina leaves was studied at various time intervals (5-60 mins) using a method described by Babarinde et al. [19]. This was done by weighing 0.5 g of Melina leaves into each conical flask and 30 ml of 10 mg/L of metal ion solution at optimal pH was introduced into it. The biomass solution in the conical flask was decanted at different time intervals from the first to the last flask. The aliquot was then taken for analysis using an Atomic Absorption Spectrophotometer (Bulk Scientific model 210). The amount of metal ions biosorbed was then calculated for each sample.

### 2.6 Effect of Metal Ion Concentration on Biosorption

Batch adsorption study of metal ion was carried out using a concentration range of 10 -100 mg/L, 0.5 g of the dried Melina leaf powder was introduced into each of the conical flask employed and 30 ml of 10 mg/L of metal ion solution at optimal pH was added to the flask [19]

The flasks were left on thermostated hot plate magnetic stirrer (Corning™ PC-420D) maintained at 30°C. The Melina leaves were removed from the solution and the concentration of the residual metal ion in each solution was determined

### 2.7 Effect of Stirring Speed on Biosorption

The experiment was carried out to study the effect of stirring speed on Pb<sup>2+</sup> and Cd<sup>2+</sup> removal efficiency using method similar to the procedure reported by Opeolu et al. [20]. The study was done varying the speed from 100 to 600 rpm, at initial pH 5, initial metal concentration 10 mg/L and 5.0 g Melina leaf powder. The flask was left on thermostated hot plate magnetic stirrer (Corning™ PC-420D) maintained at 30°C for 30 minutes. The residual metal concentration in the filtrate was determined after each agitation speed.

### 2.8 Adsorption Isotherms

The Langmuir and Freundlich isotherms were used to explain the experimental data in the study. The Langmuir model [21] was found on the assumption that sorption occurs on an even surface by monolayer sorption without the adsorbed molecules interacting with each other. It is expressed as:

$$q_e = \frac{q_{max} b C_e}{1 + b C_e} \quad (3)$$

$$C_e/q_e = C_e/q_{max} + 1/bq_{max} \quad (4)$$

Where,  $q_{max}$  represents the maximum biosorption capacity and  $b$  is an affinity parameter, related to the energy of biosorption [22]. The Langmuir isotherm separation factor (RL) can be used to forecast attraction between the adsorbate and adsorbent, and it is expressed as

$$R_L = 1/1 + b C_i \quad (5)$$

The Freundlich model [23] proposes an even layer sorption with variously dispersal active sites and the adsorbed molecules interact with each other. It is expressed mathematically as:

$$q_e = K_F (C_e)^{1/n} \quad (6)$$

Equation [6] can be written in linear form as:

$$\log q_e = 1/n \log C_e + \log K_F \quad (7)$$

Where;  $K_F$  and  $n$  are the Freundlich coefficients [23].

## 3. RESULT AND DISCUSSION

### 3.1 Effect of pH

Biosorption of the two metal ions by *Gmelina arborea* Leaves powder was strongly affected by pH (Fig. 1). A trend of increasing metal ion binding with increasing pH was observed for both metals between pH 2.5 to 5. There was a reduction in the uptake of both metals at pH values of 5.5 to 7 the result agreed with Venkateswara et al. [24] reports on different sorbents such as Coir, Barley straw, and *Ficus benghalensis* L.

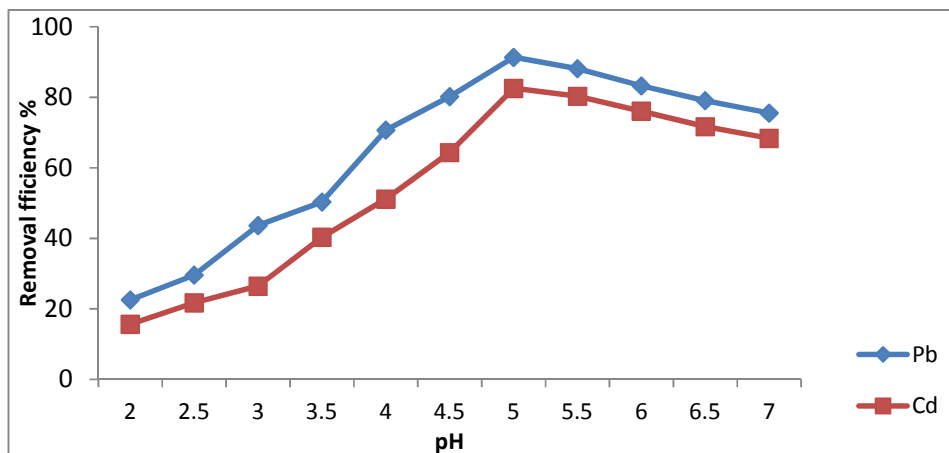


Fig. 1. Effect of pH on metal biosorption using *Gmelina arborea* leaves powder

There seems to be a dominance of hydrogen ions over metal cations on the sorption sites in the solution, when the pH is 2.0, hence hindering the approach of metal cations, causing a reduction in metal ion removal efficiency. According to Sa'g et al. [25] rise in pH reduces the dominance proton over metal cations, leading to decrease in the surface charge of sorption sites, and this explained the increase in metal removal as pH increases. As the pH increased, the ligands such as carboxylate groups in *Gmelina* would be exposed increasing the negative charge density of the biomass surface, which in turn increased the attraction of metallic ions with the positive charge, and allowing the biosorption onto the cell surface.

### 3.2 Effect of Contact Time

Contact time is an important factor that determines the efficiency of biosorption and is the period required for the reaction to attain equilibrium when the metal ions removal is constant based on the differences in the characteristics properties of the biosorbents.

Fig. 2, shows that both metal ions adsorbed in two phases with variation in the contact time of biosorbent and solution. The first phase was extremely rapid in the case of both investigated metals, regardless of the influence of metal nature [26,27,28]. This study also showed that *G. arborea* leaf powder was effective for the removal of  $Pb^{2+}$  ions than  $Cd^{2+}$  ions, the differential sorption of the ions may be ascribed to the differences in their ionic radii which

confirmed the claim of Wulfsberg [29] that ions with smaller radius have high tendency to hydrolyse, hence causing reduction in sorption.

### 3.3 Effect of Biosorbent

The effect of biosorbent dosage for the removal of lead and cadmium is depicted in Fig. 3 with dosages varying from 1 – 10 g/L. As the biosorbent dose increased, the percentage removal efficiency also increased from 71% – 83% for lead and 58% – 72% cadmium. This is because of the active sites available at the higher dosage of the biosorbent, thus making easier penetration of the metal ions to the adsorption sites [20]. Thus, maximum uptake capacity was at 5.0 g/L, above which the removal rate of metal ions remarkably decreased because of the partial clump of adsorbent which reduces the effective sorption site  $Pb^{2+}$  for metal uptake. Similar results were reported for the removal of  $Pb^{2+}$  by El-Said [5] using groundnut hull and rice husk and its ash, and *Azadirachta indica* [30].

### 3.4 Effect of Stirring Speed

*G. arborea* leaves powder floats on the surface of the solution due to its lightweight, therefore, necessitating stirring of the solution, in order to achieve contact between sorbent and water phase. Fig. 4 shows that the removal efficiency increased with an increase of stirring speed because increasing the agitation rate led to decrease in the film resistance to mass transfer surrounding the adsorbent particle [31].

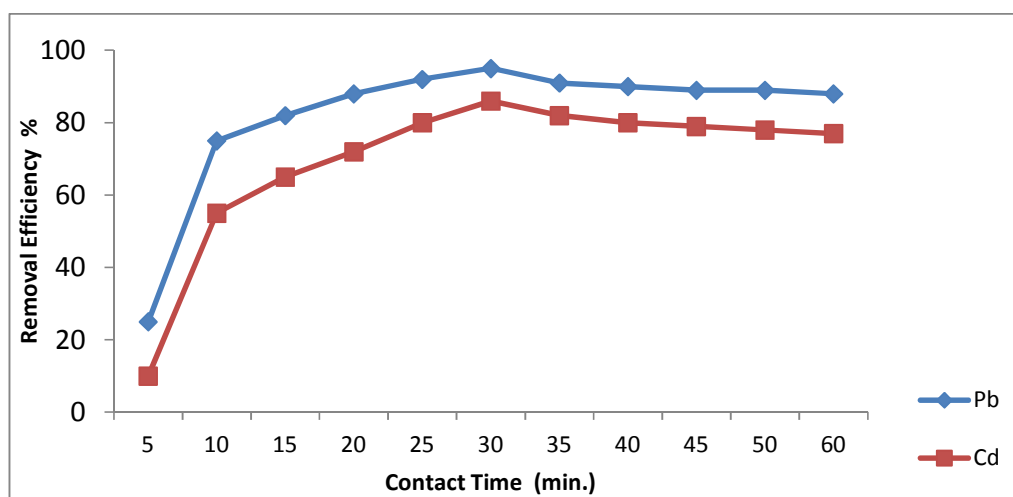


Fig. 2. Effect of contact time

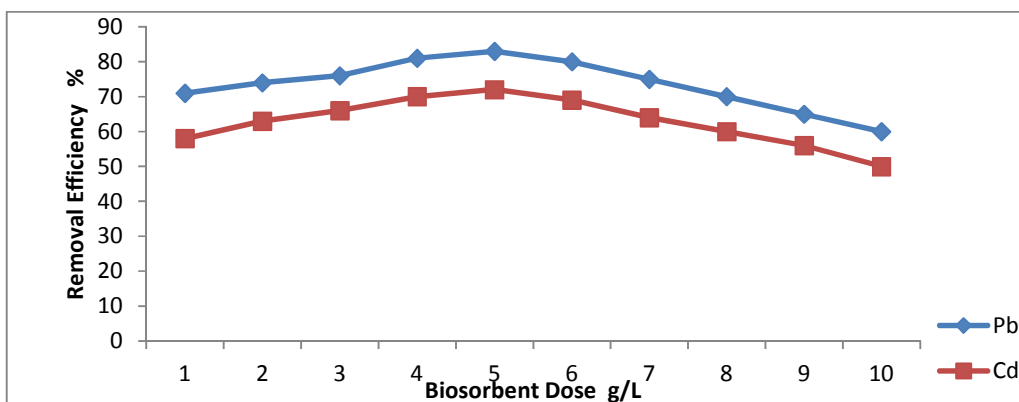


Fig. 3. Effect of biosorbent dose

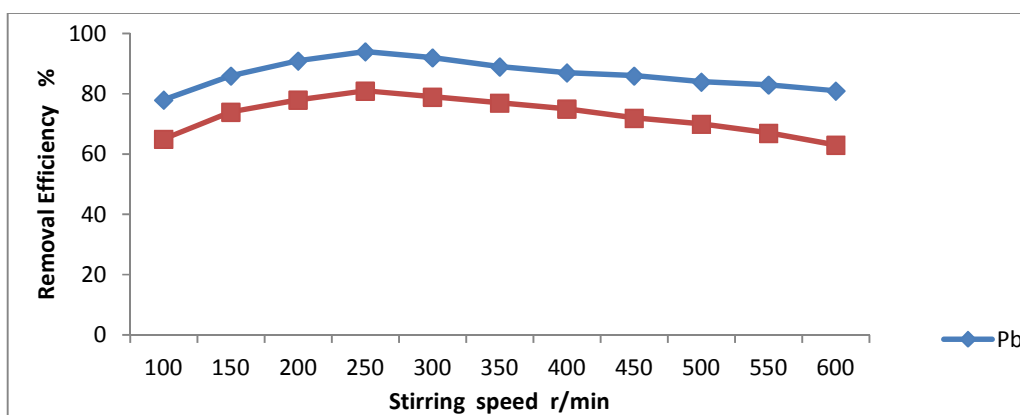


Fig. 4. Effect of stirring speed

Maximum removal efficiency was achieved at 250 rpm. and the quantity of metal ion sorbed from the solution reduced when stirring speed got beyond 300 r/min, because of vortex phenomena, which reduce the contact surface between phases.

### 3.5 Effect of Initial Metal Concentration on Biosorption

Figs. 5 and 6 showed the effect of initial concentration on the adsorption of lead and cadmium by *Gmelina arborea* leaves. The equilibrium uptake (q) of metal increased and the percentage removal efficiency of lead and cadmium ions reduced as their quantity in the solution increase. This increase in uptake of lead (4.2–22 mg/g) and cadmium (2.4 – 14 mg/g) was as a result of the increase in the driving force that is a concentration gradient [24].

However, the percentage removal efficiency of metal ions by *G. arborea* leaves has decreased

from 88 to 60% and 79 to 56%, respectively, which may be credited to the insufficient surface area to contain the initial high quantity of the ions. and thus the removal efficiency was higher at lower concentrations because there was more interaction between the sorption sites and the ions. A similar phenomenon was observed by Han, et al. [32] and Oboh, et al. [33], who had independently reported the adsorption of  $Pb^{2+}$  and  $Cu^{2+}$  on chaff and Neem leaves from aqueous solution, respectively.

### 3.6 Isotherm Models of the Biosorption

The isotherm models are mathematical models describing the distribution of cations between two phases that are in contact. The extent of the attraction of the biosorbent for the metal ions determines their distribution between the solid and liquid phases. The Langmuir and Freundlich isotherms used in explaining the experimental data in this study were the most commonly used [13,15,19,20].

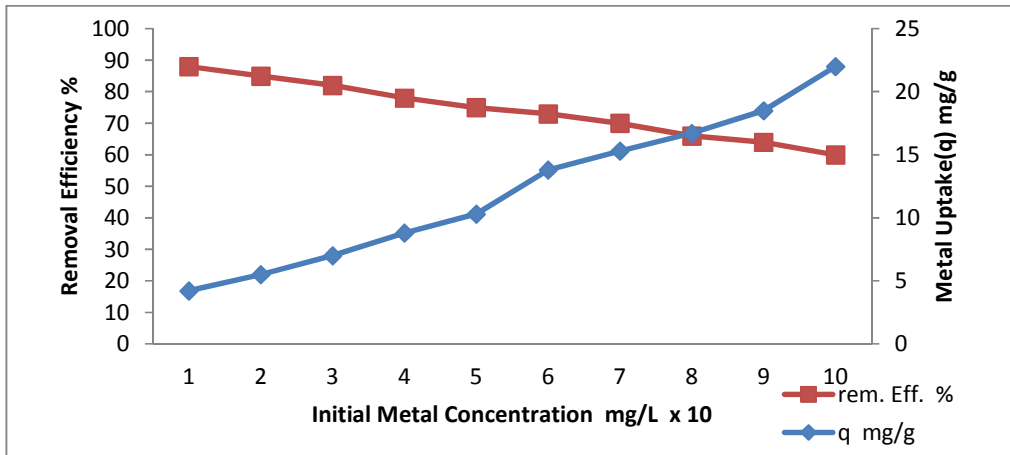


Fig. 5. Effect of initial metal ion (Pb) concentration

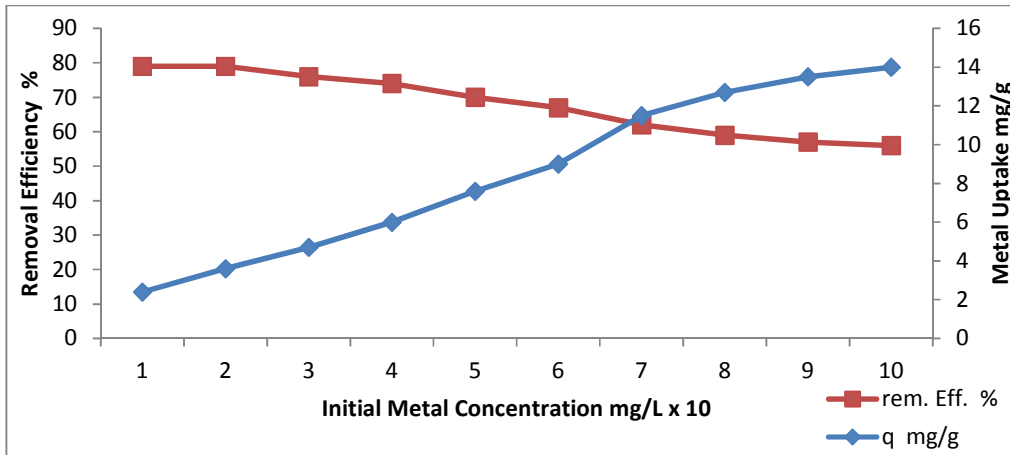


Fig. 6. Effect of initial metal ion (Cd) concentration

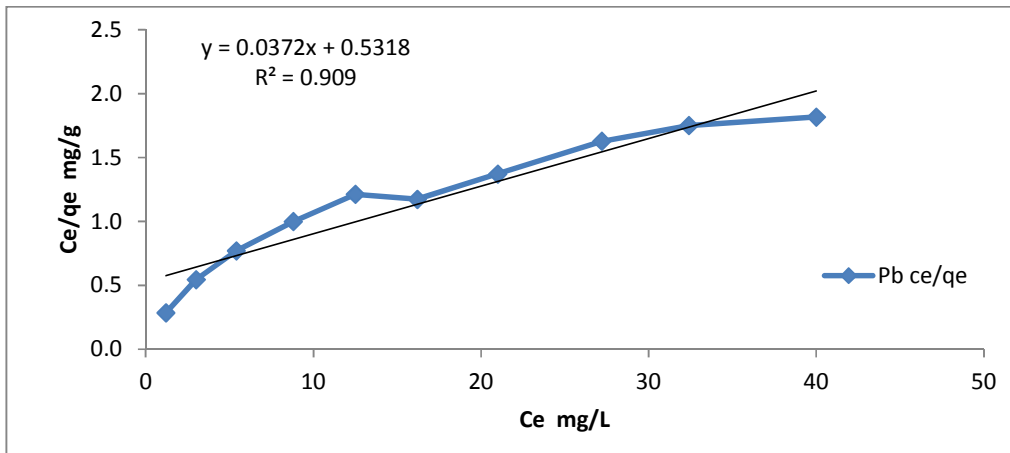
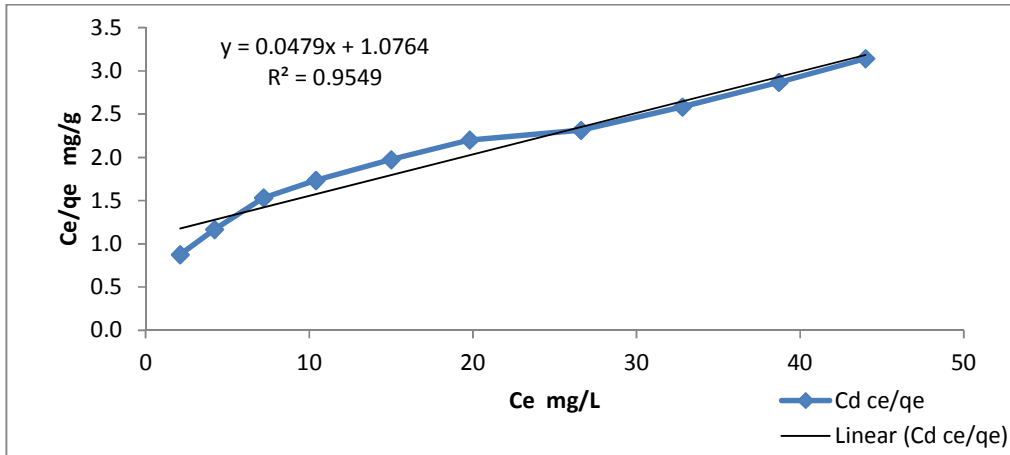
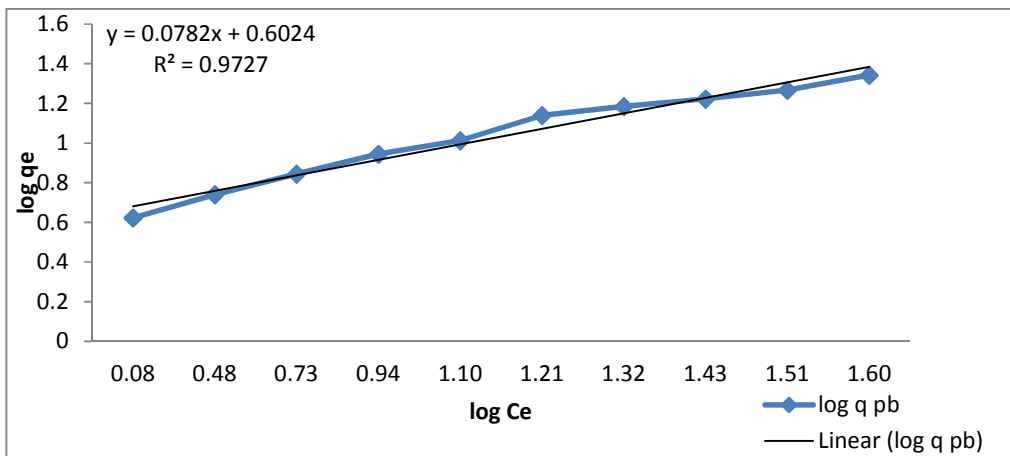


Fig. 7. Langmuir biosorption isotherm for lead at 5 g in 30 mL of biosorbent concentration and solution pH of 5.0



**Fig. 8. Langmuir biosorption isotherm for cadmium at 5 g in 30 ml of biosorbent concentration and solution pH of 5.0**



**Fig. 9. Freundlich biosorption isotherm for cadmium at 5 g in 30 mL of biosorbent concentration and solution pH of .50**

The Langmuir model Langmuir [21] is based on the hypothesis that uptake occurs on a homogenous surface by monolayer sorption without interaction between adsorbed molecules. Its equations are expressed as equations 3 and 4. A graph of  $C_e$  against  $C_e/q_e$  with straight lines as shown in Figs. 7 and 8 were fitted by regression for lead ion and cadmium ion respectively. as shown in Table 2.

Maximum biosorption capacity of biosorbent ( $q_m$ ) is found to be 26.88 mg/g (lead) and 20.88 mg/g (cadmium) of biosorbent which as shown in Table 3 is higher than biosorbents used by some authors [34, 35,36,37,38,39,40,41,42,] and lower than biosorbents used other authors [43,44,45, 46,47].

The attraction between the adsorbate and adsorbent can be forecasted using the Langmuir isotherm separation factor (RL), as expressed in the equation 5. The value of separation parameters RL gives valuable data about the nature of adsorption. The value of RL signified the kind of Langmuir isotherm to be permanent (RL = 0), good (0 < RL < 1), linear (RL = 1) or poor (RL > 1). The RL was found to be 0.0446 - 0.78125 for concentration of 10 mg/L– 70 mg/L of  $Pb^{2+}$  and 0.1005- 0.9482 for concentration of 10 mg/L –60 mg/L of  $Cd^{2+}$ . They are in the range of 0 -1 which indicates favourable biosorption for both metal ions.

The Freundlich model [23] is stated mathematically in equation 6 and 7. A graph of  $\log q_e$



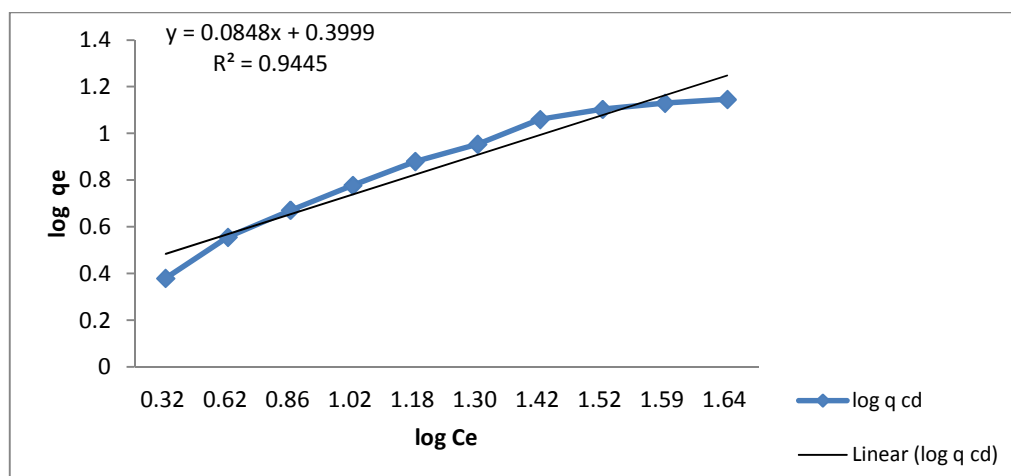
against  $\log C_e$  was plotted so as to appreciate the usefulness of Freundlich model, and straight lines were fitted to the plots by regression for lead ion and cadmium ion respectively (Figs. 9 and 10).

**Table 2. Adsorption isotherm constants for biosorption of lead (II) ions and cadmium (II) ion by *Gmelina arborea* leaves**

	Langmuir constants			Freundlich constants			
	$q_m$	b	$R^2$	1/n	n	$K_F$	$R^2$
Lead (II) ion	26.880	0.070	0.909	0.078	12.790	4.079	0.973
Cadmium (II) ion	20.880	0.045	0.955	0.085	11.790	2.302	0.945

**Table 3. Comparative analysis of bioabsorbents exhibited in different studies**

Metal	Qmax(mg/g)	Biosorbent	Reference
Pb	37.45	<i>Ficus religiosa</i> leaves	[43]
	26.88	<i>Gmelina arborea</i>	Present study
	18.9	Coir	[34]
	29.0	<i>Caulerpa lentillifera</i>	[44]
	10.1	<i>Aspergillus niger</i>	[35]
	31.54	Groundnut hull	[38]
	5.3	Barley straw	[36]
	14.7	<i>Fontinalis antipyretica</i>	[37]
	28.6	<i>Ficus benghalensis</i> L.	[46]
Cd	4.792	Bamboo Based Activated Charcoal	[38]
	20.88	<i>Gmelina arborea</i>	Present study
	6.81	<i>Saccharomyces cerevisiae</i>	[39]
	38.1	<i>R. rubra</i>	[47]
	8.82	Unmodified Rice husk	[40]
	11.00	African white star apple shell	[41]
	10.50	Ephagmum moss peat	[42]
4.594	Bamboo Based Activated Charcoal	[38]	



**Fig. 10. Freundlich biosorption isotherm for cadmium at 5 g in 30 mL of biosorbent concentration and solution pH of .50**

The higher value of  $K$  (4.0791) of  $Pb^{2+}$  indicates that the biosorbent has higher biosorption capacity for  $Pb^{2+}$  ion than  $Cd^{2+}$

#### 4. CONCLUSION

The study revealed that the *G. arborea* leaf could serve as a good biomass for the adsorption of lead and cadmium ions from wastewater. Also the abundance of *G. arborea* leaves biomass at little or no cost, makes it a good adsorbent for removing pollutants such as lead and cadmium ions from wastewaters. The optimal parameters such as solution pH, biosorbent dose, initial metal ions concentration, contact time and stirring speed determined in the experiment were effective in determining the adsorption efficiency of  $Pb^{2+}$  and  $Cd^{2+}$  onto *G. arborea* leaf powder. There was a rise in metal sorption by increasing the pH up to 5. The biosorption of all metals on the adsorbent was rapid as over 80 % of the metals were removed within the first 30 minutes of interaction. The equilibrium data of adsorption isotherm are in good agreement with the models of Langmuir and Freundlich. This adsorption can be a good choice for adsorption of not only  $Pb^{2+}$  and  $Cd^{2+}$  ions but also other heavy metal ions in the wastewater stream.

#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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