



## Biosorption of Heavy Metals from Aqueous solution using Mangrove fern *Acrostichum aureum* L. leaf Biomass as a Sorbent

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### Abstract

Investigation was carried out to analyze the biosorption behavior of the Mangrove fern (*Acrostichum aureum* L.) leaf biomass, for the removal of heavy metals from the aqueous solution. The uptake for Cu (II), Pb (II) and Zn (II) were  $46.86 \pm 0.54$ ,  $47.21 \pm 0.65$  and  $47.81 \pm 0.43$  mg g<sup>-1</sup> respectively when 500 mg L<sup>-1</sup> of the metal solution was used. The Langmuir and Freundlich equilibrium adsorption isotherms were studied and data obtained from equilibrium experiment are found well fitting with the Freundlich model than the Langmuir model with correlation coefficient of 0.99. Kinetic studies indicated that biosorption of heavy metal by the mangrove fern followed well the pseudo second order. The presence of hydroxyl, carboxyl, and amino groups in the biomass and their role in metal ion adsorption process was confirmed by FTIR studies. By this study plentifully accessible a new biosorbent for the exclusion of heavy metals from the polluted water was identified.

**Keywords:** *Acrostichum aureum*, biosorption, heavy metal.

### Introduction

Heavy metal pollution is endangering the aquatic systems and affecting the water quality. Studies on the sorption of metal ions from aqueous solution are vital in water pollution control. Increase in heavy metal pollution is associated with population growth and industrial progress. Heavy metal contamination is caused by industrial emissions, mining, smelting and application of fertilizers and sewage sludge contaminated with heavy metals<sup>1</sup>. Heavy metals including copper (Cu), and zinc (Zn) at high concentrations induce phytotoxicities in plants<sup>2,3</sup>. Heavy metals may cause haemolysis, gastrointestinal, skin, liver and kidney disorders<sup>4</sup>. The elimination of pollutant metal ions from the wastewaters is therefore important.

Ionic exchange, oxidation–reduction, membrane filtration, adsorption chemical precipitation and reverse osmosis, are cost-demanding methods used in pollution treatments<sup>5,6</sup>. Biosorption, is the capacity of some biomaterials to remove heavy metals from the aqueous solutions. It is an alternative to the conventional technologies for heavy metal removal from the polluted effluents<sup>7</sup>. In the recent years various plant materials were tried for biosorption. Leaves of poplar, neem, peepal, maize, Ulmus, *Araucaria heterophylla* have been studied to find out metal removal potential<sup>8-13</sup>.

Biosorption studies on Tree fern for the removal of lead ions from aqueous solution bracken fern in the elimination of mercury and Lady fern in the removal of Zn and Cu from aqueous solution were carried out to investigate the potential of pteridophytes in bioadsorption<sup>14-16</sup>. *Acrostichum aureum* L. is a mangrove fern growing luxuriantly in coastal regions of Karnataka, India. This fern shows clumped distribution and

produces golden yellow leaves; rhizome paste is used in wound treatment<sup>17</sup>. There are no reports on the major applications of leaves mainly in bioremediation. The present study investigates whether the mature leaf biomass of mangrove fern *Acrostichum aureum* can be used remove Lead, Copper and Zinc ions from aqueous solutions as a sorbent

### Material and Methods

**Effect of pH, Temperature, Contact Time and Initial Concentration on Metal Biosorption:** Leaves of *A. aureum* was oven dried at 60°C, powdered, sieved and preserved in sealed polythene bags. Stock (1,000 mg L<sup>-1</sup>) of Cu (II) from CuSO<sub>4</sub>, Pb (II) using Pb(NO<sub>3</sub>)<sub>2</sub> and Zn (II) from ZnSO<sub>4</sub>.6H<sub>2</sub>O solutions in deionized water were prepared. The adsorption study was performed with 50 ml of metal solution (300 mg L<sup>-1</sup>) in 100-ml conical flasks. Glassware's used for experimental purposes were washed in 20% nitric acid and rinsed with distilled water to eliminate any interference by other metals. The pH of solutions was adjusted from 2 to 6. The desired pH was adjusted with 1N NaOH and 1N HCl. Biomass of 0.5 g weight was taken in the each conical flask. The temperature effect of on biosorption was studied at 10°C, 20°C, 30°C, 40°C and 50°C. The metal ion concentration was determined after 100 minutes of incubation. To examine the effect of contact time on biosorption samples were withdrawn at different time intervals (20, 40, 60, 80 and 100 minutes) and the metal ion in the supernatant was determined using a flame atomized atomic absorption spectrophotometer (GBC 932 plus AVANTA spectrophotometer). Initial concentration was varied from 50 to 500 mg L<sup>-1</sup> of the metal ion to study the effect of on biosorption. Experiments were conducted in triplicates.

**Adsorption Isotherms:** The biosorption potential ( $q_{eq}$ ) at equilibrium was calculated as follows:

$$q_{eq} = \frac{C_0 - C_{eq}}{W} \times V \quad (1)$$

Where:  $C_0$  was the initial metal ion concentration ( $\text{mg L}^{-1}$ );  $C_{eq}$  the metal ion concentration at equilibrium ( $\text{mg L}^{-1}$ );  $V$  the volume of solution used (L); and  $W$  was weight of the dry leaf powder (g).

To investigate the sorption isotherm, two equilibrium models, the Langmuir and the Freundlich isotherm equations, were analysed. The Langmuir sorption isotherm is useful in describing sorption of a solute from a liquid solution<sup>18</sup>.

$$\frac{C_e}{q_e} = \frac{1}{K_L q_{max}} + \frac{1}{q_{max}} C \quad (2)$$

Where  $K_L$  is the Langmuir constant linked to the adsorption energy and  $q_{max}$  is the maximum adsorption capacity ( $\text{mg g}^{-1}$ ),  $q_e$  is the equilibrium sorption capacity ( $\text{mg g}^{-1}$ ),  $C_e$  the equilibrium liquid phase concentration ( $\text{mg L}^{-1}$ ). Values of Langmuir parameters  $q_{max}$  (monolayer sorption saturation capacity) and absorption constant  $K_L$  were obtained from the slope and intercept of the linear plot of  $C_e/q_e$  versus  $C_e$ .

The Freundlich isotherm<sup>19</sup>, which is the first known sorption isotherm, is expressed by the following equation

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (3)$$

Where:  $q_e$  is the equilibrium sorption capacity ( $\text{mg g}^{-1}$ ),  $C_e$  the equilibrium liquid phase concentration ( $\text{mg L}^{-1}$ ).  $K_F$  and  $n$  are isotherm constants.

**Kinetic studies:** The pseudo-first-order, pseudo-second-order and intraparticle diffusion equations were applied to the experimental data<sup>20-22</sup>.

Linearized form of pseudo first order rate equation as described here is used.

$$\text{Log}(q_{eq} - q_t) = \ln q_{eq} - K_1 t \quad (4)$$

Where  $q_{eq}$  and  $q_t$  are the metal ions adsorbed at equilibrium ( $\text{mg g}^{-1}$ ) and at time  $t$  (min), respectively, and  $K_1$  is the rate constant of the first order adsorption equation ( $\text{min}^{-1}$ ). The sorption rate constants ( $K_1$ ) can be determined by plotting  $\ln(q_{eq} - q_t)$  versus  $t$ .

Experimental data was further analyzed using pseudo second order kinetic equation.

$$\frac{t}{qt} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \quad (5)$$

Where:  $K_2$  is the rate constant of pseudo-second-order adsorption,  $t$  is the contact time (min),  $q_e$  ( $\text{mg/g}$ ) and  $q_t$  ( $\text{mg/g}$ ) are the amount of the solute adsorbed at equilibrium and at time  $t$ .

Weber and Morris<sup>22</sup> proposed a model to examine the intraparticle diffusion.

$$q_t = K_{id} t^{(1/2)} \quad (6)$$

Where:  $q_t$  is the amount of adsorbed metal ions on the biomass at time  $t$  ( $\text{mg g}^{-1}$ ). The intraparticle rate constant,  $K_{id}$  was obtained from the slope of the plot.

**FTIR analysis:** For FTIR analysis was carried out for control biomass sample and metal-loaded samples which was dried and blended with KBr. The FTIR spectra were collected at transmission mode ( $4,000-400 \text{ cm}^{-1}$ ) using a (IR Prestige-21 Shimadzu) FTIR spectrophotometer.

## Results and Discussion

**Effect of pH:** The pH of the solution is the important parameters which influence the adsorption of metal ions<sup>23</sup>. Uptake of Cu (II), Pb (II) and Zn (II) by biomass increased with an increase in pH of the solution (figure-2a). Studies on biosorption using neem and *F. religiosa* leaves also reported enhanced biosorption with increased pH<sup>8,9</sup>. Biomass was most competent at pH 6.0 in removing Cu (II) and Zn (II) ions. Above this pH Cu (II) ions will precipitate<sup>24</sup>. In case of Pb (II) ions, the highest adsorption was noted at pH 5.0. With increasing pH, formation of lead hydroxide results in decreased adsorption. Functional groups get protonated at lower pH values which will avoid the positively charged metal ions to their surface<sup>9</sup>. Functional groups achieve negative charge at higher pH values and have a high attraction for the metal ions with positive charge, which may results in improved biosorption of the metal ions.

**Effect of Temperature:** Absorption rate increased from 7.45 to 20.34, 8.84 to 24.45, 7.19 to 19.5  $\text{mg/g}$  for Cu, Pb and Zn respectively with temperature (figure-2b). Increased biosorption with temperature were also reported in tree fern biomass by Ho and Wang<sup>14</sup>. The temperature generally activates the process by increasing the heat or mass transport processes<sup>13</sup>. Temperature increase causes activation of adsorption sites<sup>25</sup>. The biomass of *A. aureum* would include many types of binding sites and the effect of temperature on each binding site would differ and it will change the overall metal adsorption process.

**Effect of contact time:** From the experiment it was prominent that Cu (II) ions uptake was most quick compared to Pb (II) or Zn(II) ions. Smaller ion radius of the Copper ions may be responsible for quick adsorption<sup>26</sup>. The equilibrium was attained within 1 h. Repulsive forces between the solute molecules on the adsorbent and aqueous solution may also contribute to the slowing down of uptake of metal at equilibrium<sup>13</sup>.

**Effect of Initial Concentration of Metal Ions:** The study has shown that biosorption capacities ( $q_{eq}$ ) are influenced by initial concentration of the metal ions. The uptake was enhanced from  $3.56 \pm 0.43$ , to  $46.32 \pm 1.02$ ,  $4.37 \pm 0.68$  to  $47.34 \pm 1.43$ ,  $3.83 \pm 0.54$  to  $47.67 \pm 1.23 \text{ mg g}^{-1}$  for, Pb (II), Zn (II) and Cu (II), respectively when initial concentration was varied from 50 to 500  $\text{mg L}^{-1}$ . According to Lata et al.<sup>27</sup> initial metal

concentrations increase will acts as a positive driving force in overcoming mass transfer resistance of metal ions between the aqueous and solid phases. This would, in turn, enhance the uptake of metal ions. Similar observations were also made by earlier investigators in their studies on adsorption of metals on plant biomass<sup>14,13</sup>.

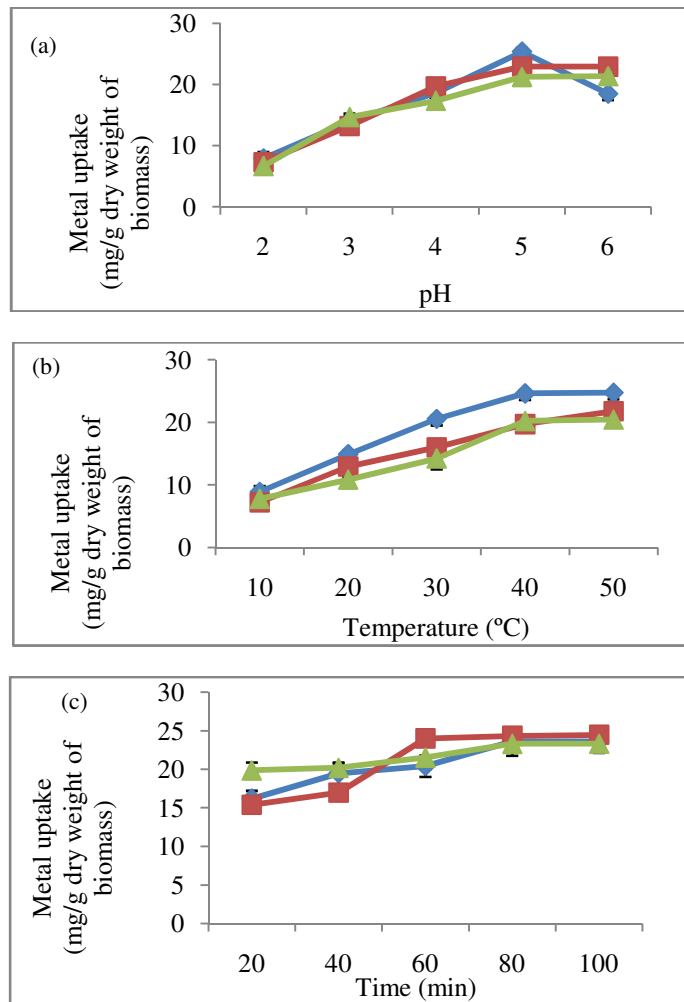


Figure-1

(a) Effect of initial pH on metal biosorption by *A. aureum* [Ci=300 mg L<sup>-1</sup>, leaf powder =0.5g, time (t) = 80 min] (b) Effect of temperature on metal biosorption by *A. aureum* [Ci=300 mg L<sup>-1</sup>, leaf powder =0.5g, t=80 min. and T=10°C, 20°C, 30°C, 40°C and 50°C] (c) Effect of contact time on metal biosorption [Ci=300 mg L<sup>-1</sup>, leaf powder =0.5g, t=20, 40, 60, 80 and 100 min], [Cu (II) (square), Pb (II) (diamond) and Zn (II) (triangle)]

**Equilibrium isotherm:** According to Langmuir isotherm finite numbers of binding sites are available on the solid surface of the adsorbent with uniform energies for the adsorption. In a monolayer adsorption there will be no interaction between the metal ions adsorbed on the adsorbent surface. The correlation coefficients for Langmuir isotherm were 0.96 for Pb (II) and Zn

(II), 0.98 for Cu (II) respectively (table-1). Metal sorbing capacity of sorbent varies with various sorbents and sources. For example q<sub>max</sub> of Pb determined using biomass of tree fern is 40mg/g and for Zn and Cu using lady fern was 0.073, 0.051 mg/g respectively<sup>14,16</sup>. However q<sub>max</sub> reported in the present study is lower than the tree fern and significantly higher than lady fern.

The Freundlich isotherm suggests that on the heterogeneous surfaces of the adsorbent binding sites are not equivalent. It also indicates that sorption energy exponentially reduces on completion of the sorptional centers of an adsorbent. For a linear adsorption n = 1, for a chemical process in absorption n < 1 and n > 1 if it is a physical process. In this study Freundlich constant n was greater than 1, indicating that adsorption process here is a physical process and sorbate intensity on the surface of the sorbent was high, revealing favourable sorption even at high metal concentration. The R<sup>2</sup> values show fitness of the experimental data to the isotherm models. This also indicates high adsorption capacity of the sorbent and metal ions could be separated from its aqueous solution efficiently by the sorbent. The correlation coefficients value for Freundlich isotherm were 0.99 this indicating that the heavy metal sorption data fitted well to the Freundlich model.

**Adsorption kinetics:** For the pseudo first-order model ln (q<sub>eq</sub> - q<sub>t</sub>) versus t was plotted. The correlation coefficients (R<sup>2</sup>) for Cu (II), Pb (II), and Zn (II) adsorption onto biomass were 0.93, 0.90, and 0.83, respectively. The linear plots of t/q<sub>t</sub> versus t (figure-5) of the pseudo second-order for the adsorption of Cu (II), Pb (II), and Zn (II) ions onto biomass was plotted. The rate constants (K<sub>2</sub>), the q<sub>eq</sub> and the R<sup>2</sup> values are given in the table-2. The correlation coefficients obtained was 0.99 for Cu (II), Pb (II), and Zn (II) adsorption. The R<sup>2</sup> values suggest that the adsorption fits well to the pseudo-second-order kinetic equation.

The results of intraparticle diffusion can be represented by a linear relationship, but the linear plot did not pass through the origin and the curve is dual natured due to variation in initial and final adsorption experiment.

**FTIR analysis:** Functional groups of the biomass were studied through FTIR spectra, which is a valuable tool for functional group analysis. Figure-2 to 5 shows the functional groups. IR peak observed at 3,421 cm<sup>-1</sup> due to stretching vibration of OH group indicated the involvement of hydroxyl group in metal binding<sup>26</sup>. Shifting of 2,372 cm<sup>-1</sup> to 2,374 cm<sup>-1</sup> for Pb, 2,376 cm<sup>-1</sup> for Zn is due to asymmetric stretching of -N=C=O group. Shift in the wave number from 2,339 to 2,341 cm<sup>-1</sup> for Cu, Pb and Zn is due to the stretching vibration of N-H or C=O group. This indicates that oxygen and nitrogen are involved in metal adsorption. C-H stretching of methyl, methylene and methyne group was observed when band in region of 2,926 cm<sup>-1</sup> shifted to 2922 cm<sup>-1</sup> in Cu and Pb, 2,924 cm<sup>-1</sup> in Zn treated leaf powder. Broadening and stretching of the bond characteristic of C=O groups of amide linkage at 1,654 to 1,651 cm<sup>-1</sup> indicates the

interaction of metals with C=O groups of the biomass. IR peak in the region of 1,440 shifted to 1,444  $\text{cm}^{-1}$  for Pb and Cu loaded leaf powder showed the participation of aliphatic or aromatic (C-H) group in the plane deformation vibration of methyl, methylene and methoxy groups. Peak shift in the region 1,321 to 1,317  $\text{cm}^{-1}$  indicate the aromatic amines involvement in

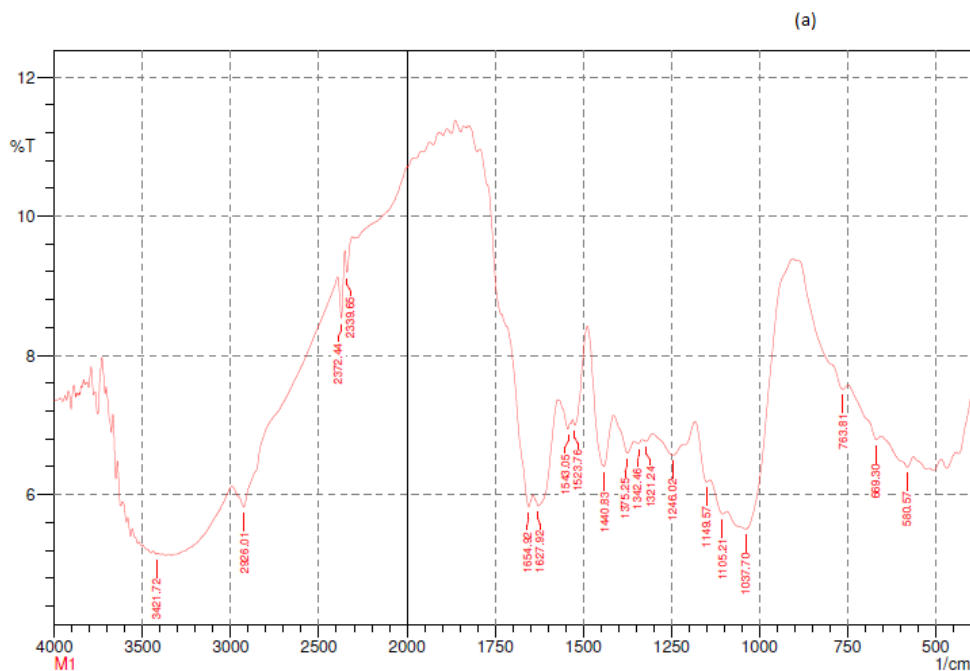
adsorption. Band in the region 669  $\text{cm}^{-1}$  shifted to 665  $\text{cm}^{-1}$  indicating the involvement of amide nitrogen in biosorption. After the binding of metal ion on the biomass, changes in intensity and shift in the position of peaks occur indicating the removal of metal ions by leaf biomass.

**Table-1**  
**Adsorption isotherm parameters for Pb (II), Cu (II), and Zn(II) ions on *A. aureum* leaf powder**

Metal	Langmuir isotherm			Freundlich isotherm		
	q m (mg g <sup>-1</sup> )	K L(L mg g <sup>-1</sup> )	R <sup>2</sup>	KF(mg g <sup>-1</sup> )	n	R <sup>2</sup>
Pb(II)	28.571	0.39	0.966	2.12	3.69	0.994
Cu(II)	27.77	0.51	0.983	2.15	3.703	0.993
Zn(II)	37.03	0.211	0.966	2.37	3.816	0.993

**Table-2**  
**Pseudo-second-order and intraparticle diffusion parameters for the biosorption of metal ions onto *A. aureum* leaf powder**

Metal	Pseudo-second-order			Intraparticle diffusion		
	K <sub>2</sub>	q <sub>eq</sub>	R <sup>2</sup>	K <sub>i</sub> (mg/g min <sup>-1/2</sup> )	Intercept (mg g <sup>-1</sup> )	R <sup>2</sup>
Pb(II)	2	27.77	0.999	0.19	14.98	0.926
Cu(II)	3	28.57	0.955	0.1	18.63	0.925
Zn(II)	3	27.77	0.999	0.25	13.39	0.815



**Figure-2**  
**FTIR spectra of *A. aureum* leaf powder before the biosorption of heavy metals**

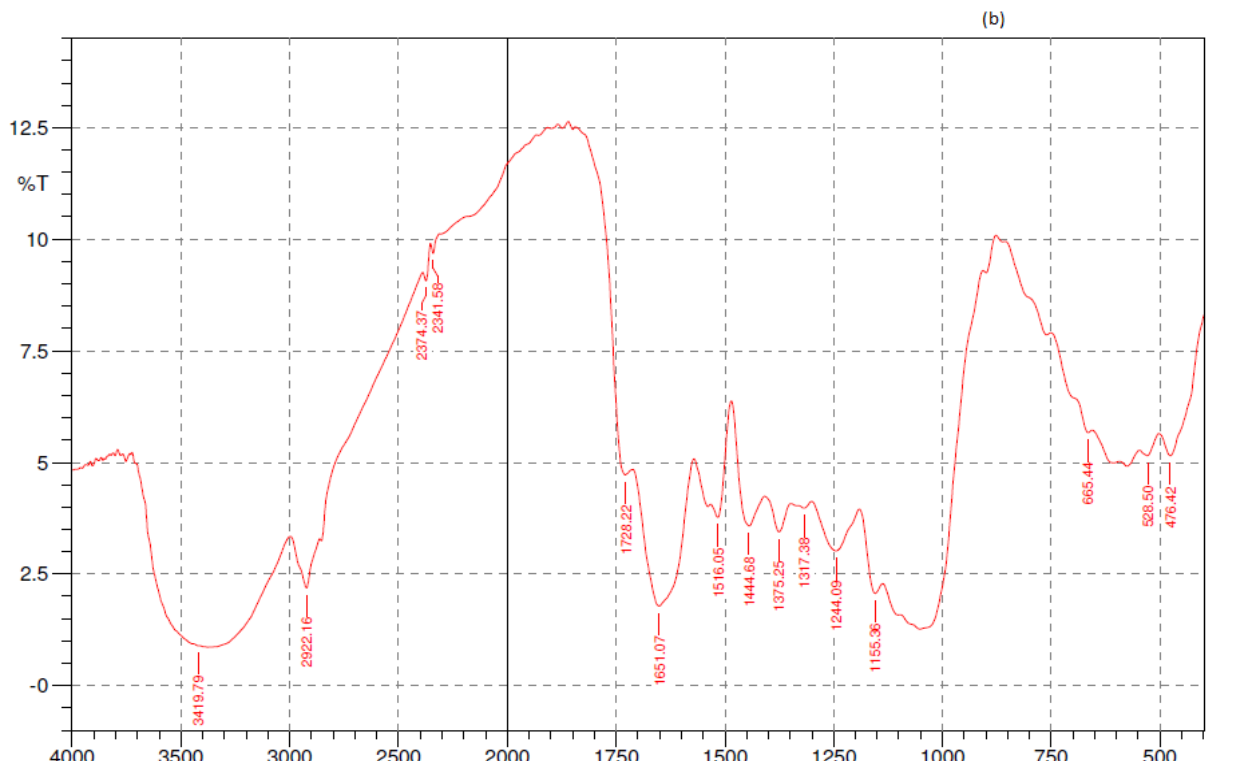


Figure-3  
FTIR spectra of *A. aureum* leaf powder after the biosorption of Pb (II)

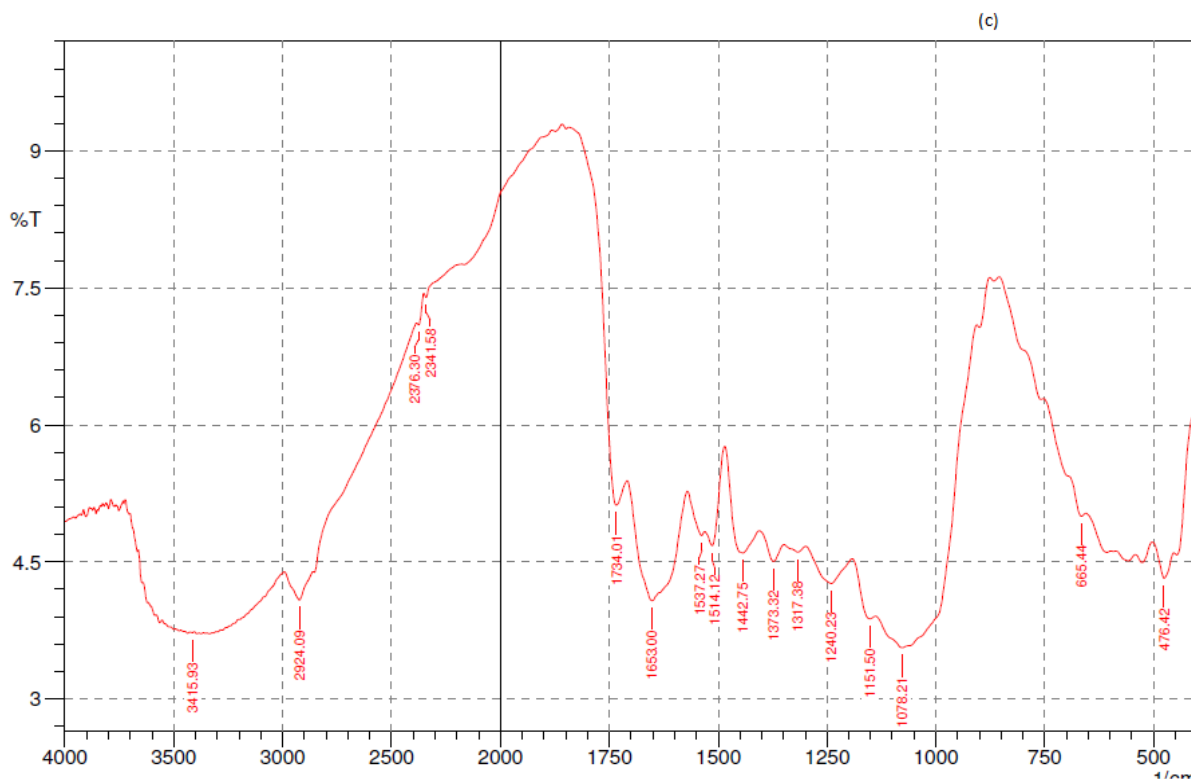


Figure-4  
FTIR spectra of *A. aureum* leaf powder after the biosorption of Zn (II)

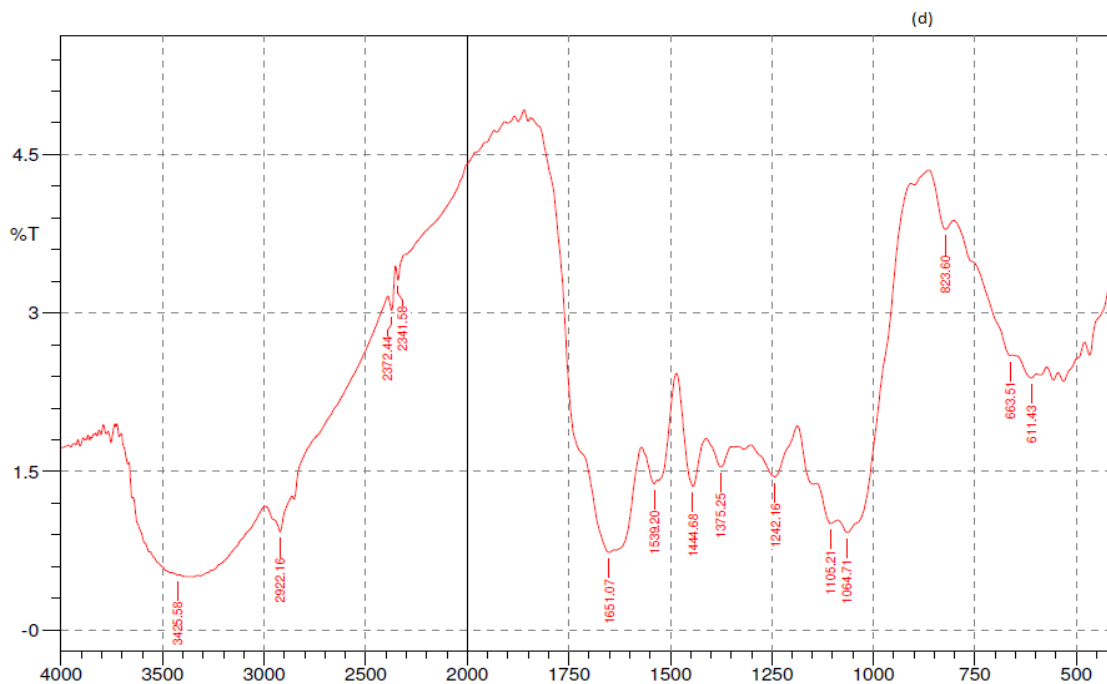


Figure-5  
FTIR spectra of *A. aureum* leaf powder after the biosorption of Cu (II)

## Conclusion

Biosorption of heavy metals was investigated. *A. aureum* is a suitable sorbent for the removal of Cu(II), Pb(II) and Zn(II) from the aqueous solution. The study indicated that the maximum uptake for Cu(II), Pb(II) and Zn(II) were  $46.86 \pm 0.54$ ,  $47.21 \pm 0.65$  and  $47.81 \pm 0.43$  mg g<sup>-1</sup> respectively at pH 5 when 500mg L<sup>-1</sup> of the metal solution were used at 40°C. Metal removal was a function of pH, temperature, time and initial concentration of metal ions in the solution. Freundlich isotherm fitted well to the adsorption data rather the Langmuir isotherm. FTIR studies revealed the presence of hydroxyl, carboxyl, and amino groups in the biomass and proved possible involvement of these groups in metal ion adsorption process. By this study the potential of a new cost-effective and plentifully available bioadsorbent for the removal of heavy metal ions from aqueous solutions has been identified.

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