

Study of mango biomass (*Mangifera indica* L) as a cationic biosorbent

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ABSTRACT: Unfertilizable fruiting buds of mango plant *Mangifera Indica* L, an agrowaste, is used as a biomass in this study. The efficacy of the biosorbent was tested for the removal of lead, copper, zinc and nickel metal ions using batch experiments in single and binary metal solution under controlled experimental conditions. It is found that metal sorption increases when the equilibrium metal concentration rises. At highest experimental solution concentration used (150 mg/L), the removal of metal ions were 82.76 % for lead, 76.60 % for copper, 63.35 % for zinc and 59.35 % for nickel while at lowest experimental solution concentration (25 mg/L), the removal of metal ions were 92.00% for lead, 86.84 % for copper, 83.96 % for zinc and 82.29 % for nickel. Biosorption equilibrium isotherms were plotted for metal uptake capacity (q) against residual metal concentrations (C_r) in solution. The q versus C_r sorption isotherm relationship was mathematically expressed by Langmuir and Freundlich models. The values of separation factor were between zero and one indicating favourable sorption for four tested metals on the biosorbent. The surface coverage values were approaching unity with increasing solution concentration indicating effectiveness of biosorbent under investigation. The non-living biomass of *Mangifera indica* L present comparable biosorption capacity for lead, copper, zinc and nickel metal ions with other types of biosorbent materials found in literature and is effective to remove metal ions from single metal solutions as well as in the presence of other co-ions with the main metal of solution.

Keywords: Biomass; Efficacy; Isotherm; Multi metal; Single metal; Sorption

INTRODUCTION

Water is essential for life on earth. Ninety-seven percent of the water on the earth is salt water. Two percent of the water on earth is glacier ice at the north and south poles and only less than one percent of all the water on earth is fresh water that actually available for drinking, agriculture, domestic and industrial consumption. Moreover, the rest is locked up in oceans as salt water, polar ice caps, glaciers and underground reservoirs. Rapid industrialization and population growth have increased water demands but the supply is limited. This available limited quantity of water is also polluted by variety of pollutants. The major sources of water contamination are domestic, industrial and agricultural, as well as solid waste pollution, thermal pollution, shipping water pollution and radioactive wastes (Tyagi and Mehra, 1994). Heavy metals such as zinc, lead and chromium have a number of applications in basic engineering works, paper and pulp industries, leather tanning, organochemicals, petrochemicals,

fertilizers, etc. These industries are main source of heavy metals into the environment. Rapid industrialization and technological development enhance the concentration of heavy metals, posing a significant threat to the environment and public health because of their toxicity, accumulation in the food chain and persistence in nature (Ceribasi and Yetis, 2001; Samarghandi *et al.*, 2007).

Over a few decades several chemical methods have been devised for the treatment and removal of heavy metals. The commonly used procedures for removing metal ions from aqueous streams include phytoremediation, chemical precipitation, lime coagulation, ion exchange, reverse osmosis and solvent extraction (Priyantha *et al.*, 2008; Shah *et al.*, 2009). However the disadvantages like incomplete metal removal, high reagent and energy requirements, generation of toxic sludge or other waste products that require careful disposal has made these methods comparatively cost effective for removing heavy metals from aqueous effluents (Ahalya *et al.*, 2003; Abdel-

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Ghani and Elchaghaby, 2007; Chien and Shih, 2007). The search for new technologies involving the toxic metals from waste waters has directed attention to biosorption, based on metal binding capacities of various biological materials. Biosorption is considered to be a fast physico-chemical process. The biosorption process involves a solid phase (sorbent or biosorbent; biological material) and a liquid phase (solvent, normally water) containing dissolved species to be sorbed (sorbate, metal ions) due to higher affinity of biosorbent for the sorbate species, the later is attracted and bound there by different mechanisms (Kratochvil and Volesky, 1998; Abdel-Ghani et al., 2009). Different parameters may affect the biosorption mechanism to various extents (Soltanali and Shams Hagani, 2008; Gadd, 2009; Zvinowanda et al., 2009). pH is the most important parameter that can affect the solution chemistry of the metals, the activity of the functional groups in the biomass and the competition of metallic ions (Galun et al., 1983; Friis, 1986). Temperature normally not effects the biosorption performances in the range of 20-35 °C (Asku, 1992). Biomass concentration in solution seems to influence the specific uptake. For lower values of biomass concentrations there is an increase in the specific uptake (Fourest and Roux, 1992; Gadd and De Rome, 1988). Biosorption is used to treat waste water where more then one metal ions are present; the removal of one metal ion may be influenced by the presence of other metal ions. For example: Uranium uptake by biomass of bacteria, fungi and yeasts was not affected by the presence of manganese, cobalt, copper, cadmium, mercury and lead in solution (Sakaguchi and Nakajima, 1991). In contrast, the presence of Fe²⁺ and Zn²⁺ was found to influence uranium uptake by *Rhizopus arrhizus* (Tsezos and Volesky, 1982) and cobalt uptake by different microorganisms seemed to be completely inhibited by the presence of uranium, lead, mercury and copper (Sakaguchi and Nakajima, 1991; Nwuche and Ugoji, 2008). Metal affinity to the biomass can be manipulated by pretreating the biomass with alkalis, acids, detergents and heat, which may enhance the amount of the metal sorbed. When non-viable biomass is used in the removal of heavy metals, alkali pre-treatment is an effective method to improve the biosorption capacity for metal ions (Yan and Viraraghavan, 2000). Hence, it is necessary to carry out more detailed studies to understand why enhancement or reduction in absorption capacity occurs under specific pre-treatment conditions. Biomass which is available in large quantities as a waste product is of

high interest as a potential metal biosorbent. Previous studies on metal biosorption have been restricted to simple solution of only one metal. Masud and Anantharaman (2005) found biosorption activity of copper (II) on negative bacteria *Thiobacillus ferrooxidans* while Nuhoglu et al. (2002) used *Ulothrix zonata* for removal of Cu (II) from aqueous system. King et al. (2007) used *Azadirachta indica* bark while Mamoon et al. (2008) used *Neem* biomass for the removal of zinc from aqueous solution. Pb (II) removal by citrus pectin and *Pseudomonas aeruginosa* were studied by Ankit and Silke (2005) and Yong Kang et al. (2008), respectively. Biosorption of Fe (II) from aqueous solution on tamarad bark (*Tamarindus indica*) and potato peel waste (*Solanum tuberosum*) have been studied by Devi and Abdulsalam (2009) while Ahalya et al. (2005) used Bengal gram husk (husk of channa dal, *Cicer arietinum*) in the biosorption of Fe (III). Biosorption of Ni (II) ions have been investigated by Vijayaraghavan et al. (2006) and Mogollon et al. (1998) onto *Sargussum wightii* and filamentous fungi strains *Rhizopus sp.* respectively. Hashim and Chu (2004) studied the binding ability of Cd (II) on seven different species of brown, red and green seaweeds while Aravindhyan et al. (2009) studied the biosorption of cadmium metal ion from waste water using *Hypnea valentiae* biomass.

The objective of this study is to develop inexpensive and effective biosorbent that is easily available in large quantities and feasible economically for multiple metal ions in solution. Biosorbent, unfruiting buds (battoor) of mango *Mangifera Indica L* is tested for biosorption of multiple metal ions in single metal system (SMS) and binary metal system (BMS).

MATERIAL AND METHODS

Preparation of biosorbent

Mangifera Indica L biomass was collected from the botanical gardens of Borneo. The biomass was dried in sun for fifteen days. The buds were removed and further dried in sun for another fifteen days. This biomass was washed with tap water to remove any dust or foreign particles attached to biomass and thoroughly rinsed with distilled water. The washed biomass was dried at 50 °C and ground to powder with kitchen grinder. Grinded biomass was further thoroughly washed with distilled water till the color of washing water was cleared. The powdered biomass was dried in oven at 50 °C to a



constant weight. The biosorbent was again ground to powder and was sieved with three different sized meshes. Four different grade particle sizes were obtained. First particle size was less than 255 µm, second 255-355 µm, third 355-500 µm and fourth was 500-710 µm. The aforesaid particle size no. 2 was selected for further study because the amount of no. 1 size was too less and no. 3 and no. 4 sizes will show less efficiency because of less surface area as compared to no. 2 size.

Stock solutions of metal ions

Stock solution were prepared in deionized water from the respective salts of four metals (Pb^{2+} , Cu^{2+} , Zn^{2+} , Ni^{2+}) under investigation. The resulting stock solutions were stored in the air tight plastic bottle.

Batch studies of SMS and BMS

For SMS, solutions of fixed volume (100 mL) with varying concentrations in conical flasks were thoroughly mixed with 0.5 g of biosorbent dose, size of 255 to 355 µm at 30 °C and 100 revolutions per minute (rpm) shaking speed for 12 h. Twelve hours of equilibrium period for sorption experiment were used to ensure equilibrium after conducting equilibrium studies of the biosorbent. The pH range was adjusted from 4-6 by using 0.1M HNO_3 and 0.1M HCl solutions. The flasks were kept on rotating shaker with constant shaking. At the end of experiment the flasks were removed and the solution was separated from the biomass by filtration through filter paper. For BMS 100 mL volume of solutions with varying concentrations (25, 50, 75, 100, 125 and 150 mg/L) of main metal and 25mg/L of affecting metal concentration in the same solution in conical flasks were thoroughly mixed under same conditions stated for SMS solutions. At the end of experiment, the solution was separated from the biomass by filtration through filter paper. Filtrates of SMS and BMS were diluted to 10 mg/L or 20 mg/L with deionized water and analyzed for metal concentration using flame atomic absorption spectrometry (Perkin Elmer AA300). After metal concentration analysis, the final concentration was subtracted from the initial concentration in order to find the metal to be sorbed.

Calculation of metal uptake

The quality of biosorbent is judged by the metal uptake (biosorption capacity), q .

Amount of metal bound by the biosorbent which disappeared from the solution was calculated based on the mass balance for the biosorbent in the system.

$$q = \frac{V(C_i - C_f)}{S}$$

q = Metal ion uptake capacity (mg/g)

C_i = Initial concentration of metal in solution, before the sorption analysis (mg/L).

C_f = Final concentration of metal in solution, after the sorption analysis (mg/L).

S = Dry weight of biosorbent (g)

V = Solution volume (L)

The difference between the initial metal ion concentration and final metal ion concentration was assumed to be bound to the biosorbent.

Freundlich and Langmuir model

To characterize the biosorption for SMS, Langmuir and Freundlich models are used. The Langmuir model makes assumptions such as monolayer adsorption and constant adsorption energy while the Freundlich model deals with heterogeneous adsorption

Langmuir equation of adsorption isotherm is $q = \frac{q_{max} C_i}{K + C_i}$, Where q_{max} and K are the Langmuir constants. The Freundlich equation of adsorption isotherm is $\log q = \log K + (1/n) \log C_i$, Where q is the amount adsorbed per unit mass of adsorbent and C_i is equilibrium concentration. The plot of $\log q$ vs $\log C_i$ is linear and constants K and n is evaluated from slopes and intercepts.

Separation factor

The shape of the isotherm can be used to predict whether adsorption system is favorable or unfavorable in a batch adsorption system. Accordingly, the essential feature of Langmuir isotherm was expressed in term of dimensionless constant called the separation factor. It is defined by the following relationship.

$$Sf = 1/(1+bC_i)$$

Surface coverage (θ)

To account for adsorption behavior of the metal ions on the biomass the Langmuir type equation related to surface coverage is used. The equation is expressed as follow.

$$bC_i = \theta/(1-\theta) \text{ and } \theta = bC_i/(1+bC_i)$$

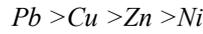
RESULTS AND DISCUSSION

Efficiency of the non conventional biosorbent of plant origin is tested for removal of metal ions Pb^{2+} , Cu^{2+} , Ni^{2+} and Zn^{2+} from synthetic solutions in SMS



and BMS solutions. The rate of absorption is a function of the initial concentration of metal ions, which makes it an important factor to be considered for effective biosorption (Ahalya *et al.*, 2005). Fig.1 reveals that capacity of biosorbent increases with increase in initial concentration of metal ions. This characteristic represents that surface saturation was dependent on the initial metal ion concentrations. At low concentrations biosorbent sites take up the available metal more quickly. However, at higher concentrations, metal ions need to diffuse to the biomass surface by intraparticle diffusion and greatly hydrolyzed ions will diffuse at a slower rate (Horsefall and Spiff, 2005). It is found that as metal ions concentration is reduced, biosorption rate increased and when the metal ions concentration is high the metal removal rate decreased. Such decline in % removal rate is probably caused by the saturation of some adsorption sites. The results are in agreement to Chen and Wang (2007).

The selectivity order for metal ions towards the studied biomass is:



This order is same as searched by Zhang and Banks (2005) and comparable to Saeed *et al.* (2005). This differential absorption of metal ions may be ascribed to the difference in their ionic radii. The smaller the ionic radius, the greater its tendency to hydrolyzed leading to reduced sorption. The ionic radii of Pb^{2+} , Cu^{2+} , Zn^{2+} and Ni^{2+} are 1.19\AA , 0.73\AA , 0.74\AA and 0.67\AA , respectively. The result is in similar to previous study by Horsefall and Spiff (2005) and Hanif *et al.* (2007). Table 1 illustrates the relationship between absorbed and aqueous concentration at equilibrium. The Langmiur and Freundlich adsorption constants are evaluated from the isotherms with correlation coefficients. Both the models represent better absorption process due to high value of correlation coefficients (R^2).

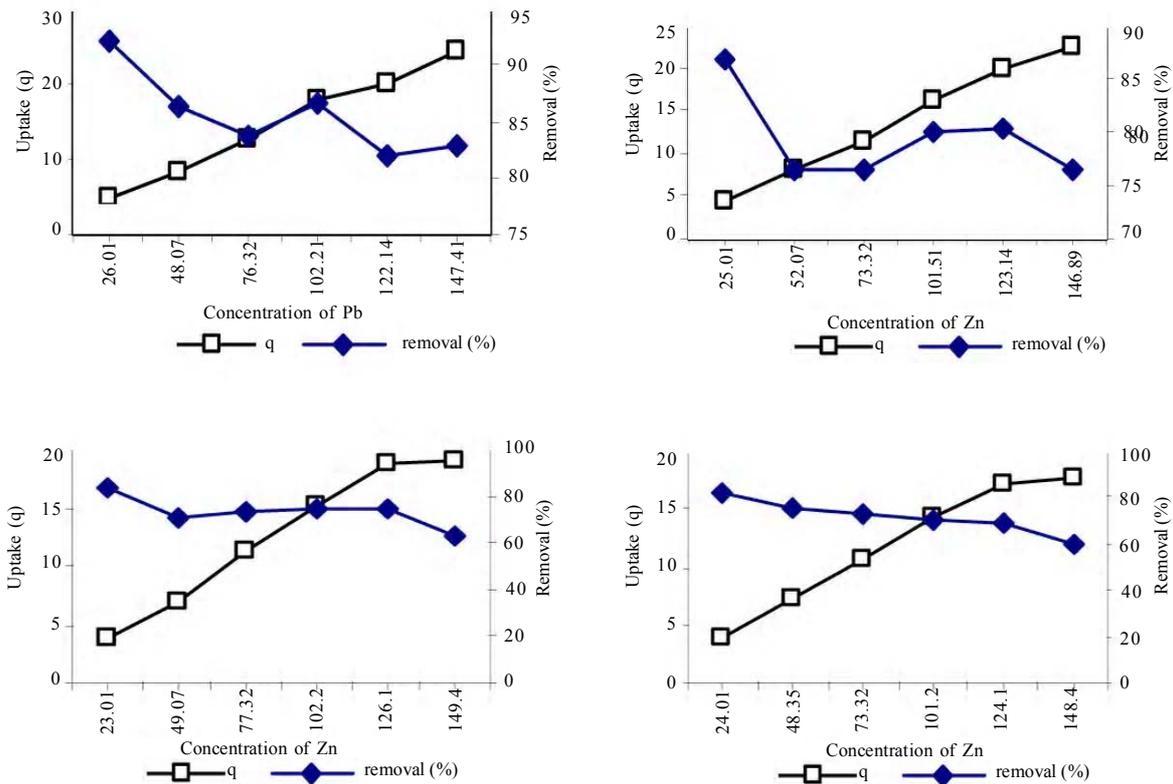


Fig. 1: Comparison of uptake capacity (mg/g) and percentage removal as a function of metal ions concentration by *Mangifera Indica L* biosorbent



Table 1: Percentage removal of metal ions on biomass *Mangifera indica* L in single metal system

Concentration	Removal (%)			
	Pb	Cu	Zn	Ni
25	92.00	86.84	83.96	82.29
50	86.16	76.37	70.85	74.93
75	83.58	76.52	73.31	72.43
100	86.58	80.00	73.73	70.21
125	81.85	80.26	74.22	69.19
150	82.76	76.60	63.35	59.35

The Langmuir isotherm considers absorption as a chemical phenomenon. It was first theoretically examined in the adsorption of gases on solid surfaces. The linearized Langmuir equation is

$$1/q = 1/q_{max} + 1/(b \cdot q_{max})(C_f)$$

Constant b which is related to the energy of absorption through the Arrhenius equation. The higher b, the higher is the affinity of the biosorbent for the metal ions. q_{max} can also be interpreted as the total number of binding sites that are available for biosorption and q as the number of binding sites that are in fact occupied by the metal ions at the concentration C_f . (Volesky, 2003).

According to the above data the affinity order of *Mangifera Indica* L biosorbent is

$$Pb > Cu > Zn > Ni$$

Langmuir isotherm, which represents that monolayer of metal ions (sorbate), is formed on biosorbent.

Adsorption-partition constants are determined for metals using the following log form of the Freundlich isotherm

$$\log q = \log K + (1/n) \log C_f$$

Where q is the metal ion sorbed (mg/g), C_f the equilibrium concentration of metal ion solution in mg/L, K and n are Freundlich constants. The constants K and 1/n were determined by linear regression from the plot of log q against log C_f . K is a measure of the degree or strength of adsorption. Small value of K indicate the minimal absorption and large value indicates the more absorption (Horsefall and Spiff, 2005) while 1/n is used as an indication of whether absorption remains constant (at 1/n = 1) or decreases with increasing metal ions concentrations (with 1/n = 1). The q_{max} value is the maximum value of q, is important to identify the biosorbent highest metal uptake capacity and as such useful in scale-up considerations (Hanif et al., 2007). The magnitude of the experimental q_{max} for *Mangifera Indica* L biomass is found to 24.40, 22.51, 18.93 and 17.62

mg/g for all the four i.e., lead copper zinc and nickel metal ions are comparable with theoretically calculated q_{max} values from Langmuir and Freundlich isotherm models.

The maximum absorption capacity is observed of lead on *Mangifera Indica* L suggesting that it is a potential biosorbent for removal of lead, as well as the other three tested metals.

The shape of the Langmuir isotherm can be used to predict whether a biosorption system is favorable or unfavorable in a batch adsorption process. Accordingly, the essential features of the Langmuir isotherm was expressed in terms of a dimensionless constant called the equilibrium parameter, SF, which is defined by the following relationship

$$SF = 1/(1 + bCi)$$

Where, SF is the dimensionless equilibrium parameter or separation factor, b, the constant from Langmuir equation and C_i the initial metal ion concentration of 100 mg/L. The parameter, SF, indicates the shape of the isotherm and nature of the sorption process. SF value between 0 and 1 represents favorable isotherm. The SF values of Pb^{2+} Cu^{2+} Zn^{2+} Ni^{2+} for *Mangifera Indica* L biomass is calculated from above equation and plotted against initial metal ion concentration (Hanif et al., 2007).

The data in Table 1 shows that, the sorption of metals on *Mangifera Indica* L biomass increase as the initial metal ion concentration increase from 25 to 150 mg/L, indicating that biosorption is even favorable for the higher initial metal ion concentrations (Fig. 1). The biosorption process is favorable for metal removal at all concentrations investigated. According to this classification, removal ability tends to be in the order:

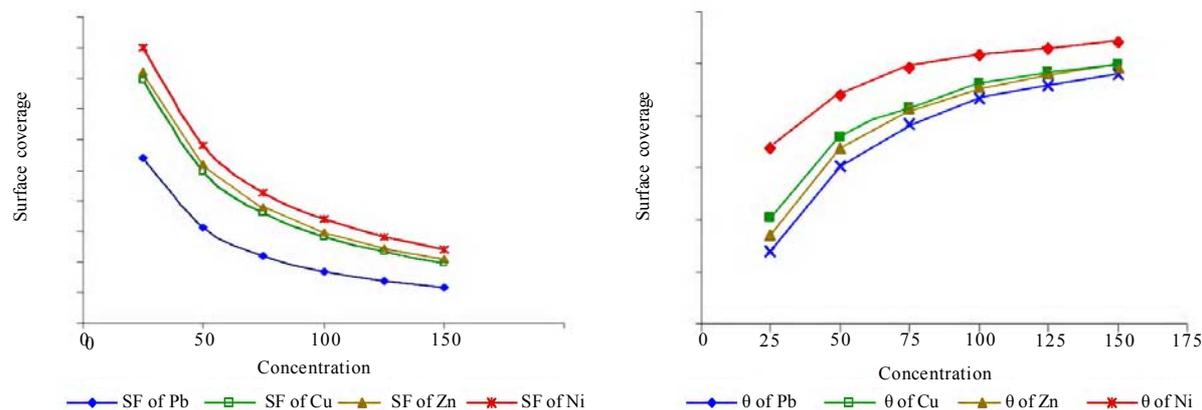
$$Pb > Cu > Zn > Ni$$

Above given order illustrates that initially equilibrium for metals uptake is more favorable for *Mangifera Indica* L, its sorption capacity and selectivity is the same as presented in previous Table 2. The trend presented by SF in Fig. 2 is also providing information that the



Table 2: Surface coverage (θ) of lead, copper, zinc and nickel for *Mangifera Indica L* biomass

C_i (mgL ⁻¹)	θ of Pb	θ of Cu	θ of Zn	θ of Ni
25	0.737	0.603	0.568	0.540
50	0.838	0.760	0.737	0.703
75	0.891	0.814	0.815	0.782
100	0.916	0.860	0.854	0.832
125	0.929	0.882	0.878	0.858
150	0.940	0.899	0.895	0.879

Fig. 2: A plot of SF and surface coverage (θ) against concentration of lead, copper, zinc and nickel for *Mangifera Indica L* biomass

Mangifera Indica L biomass is more effective and excellent adsorbent for metal at lower metal concentrations.

To account for absorption behavior of the metal ions on the biomass the Langmuir type equation related to surface coverage is used. The equation is expressed as follow:

$$bC_i = \theta/(1-\theta) \text{ and } \theta = bC_i/(1+bC_i)$$

Where, b is the absorption coefficient, C_i the initial concentration and θ the surface coverage.

The fraction of biomass surface covered by metal ion was studied by plotting the surface coverage values (θ) against metal ions concentration. The data is presented in Fig. 2. The figure shows that, increase in initial metal ion concentration for *Mangifera Indica L* biomass increases the surface coverage on the biomass until the surface is nearly fully covered with a monomolecular layer. Further examination of Fig. 2 reveals that the surface coverage ceases to vary significantly with concentration of metal ions at higher levels. Surface coverage value for metal ions on adsorbents of *Mangifera Indica L* is in following order:

$$Pb > Cu > Zn > Ni$$

Comparing the intercationic effect in BMS, biosorption of Pb^{2+} by *Mangifera Indica L* biomass in Table 3 and Fig. 3 it is observed the inhibition of Pb^{2+} sorption in the presence of Cu^{2+} , Ni^{2+} and Zn^{2+} co-ions. The sorption of Pb reduces in the presence of Cu^{2+} and Ni^{2+} from 24.4 to 24 and 23.95mg/g, respectively compared to its sorption from SMS. It is not much significant change. In case of Zn^{2+} co-ion, the uptake capacity of biosorbent reduces to 19.29 mg/g. This reduction in sorption or uptake capacity is observed at highest concentration of lead in solution. The % removal of lead is observed in all binary metal system of lead as compared to % removal in SMS. Interestingly Ni^{2+} sorption increases in the presence of co-ions Cu^{2+} , Pb^{2+} and Zn^{2+} as compared to its sorption in SMS. Uptake capacity of *Mangifera Indica L* biosorbent for Ni^{2+} increases from 17.61 in SMS to 20.44 and 20.17mg/g in the presence of Zn^{2+} and Pb^{2+} co-ions and a slight increase in the presence of Cu^{2+} that is 18.07 mg/g.

The effect of Ni^{2+} , Pb^{2+} and Zn^{2+} on the uptake of copper by *Mangifera Indica L* biosorbent is observed as: Cu^{2+} sorption is inhibited by the presence of co-ion compared to its sorption from SMS and the amount of metal ions sorbed is less as compared to sorbed from Cu^{2+} solution of SMS. The data in Table 4 reveal



Table 3: Uptake capacity of *Mangifera Indica L* and percentage removal of lead in SMS and BMS

Ci	q	Removal SMS solution
26.01	4.78	92.00
48.07	8.28	86.16
76.32	12.75	83.58
102.21	17.70	86.58
122.14	19.99	81.85
147.41	24.40	82.76

Ci	(Pb and Cu) Binary solution		(Pb and Ni) Binary solution		(Pb and Zn) Binary solution	
	q	Removal (%)	q	Removal (%)	q	Removal (%)
26.01	4.38	84.31	4.62	90.31	3.83	75.92
48.07	7.88	82.00	8.27	84.38	7.76	78.46
76.32	12.35	80.96	12.46	82.16	11.65	77.90
102.21	17.30	84.62	17.27	85.45	15.53	78.15
122.14	19.59	80.21	19.99	81.19	19.06	76.57
147.41	24.00	81.40	23.95	80.28	19.29	64.79

Table 4: Uptake capacity of *Mangifera Indica L* and percentage removal of copper in SMS and BMS

Ci	q	Removal (%)
25.01	5.00	86.84
52.07	10.41	76.37
73.32	14.66	76.52
101.51	20.30	80.00
123.14	24.62	80.26
146.89	29.37	76.60

Ci	(Cu and Zn) Binary solution		(Cu and Pb) Binary solution		(Cu and Ni) Binary solution	
	q	Removal (%)	q	Removal (%)	q	Removal (%)
24.65	4.48	87.32	4.20	85.19	3.23	66.21
51.87	8.514	83.68	8.60	82.93	6.47	63.63
73.90	12.08	80.96	11.91	80.58	9.87	65.84
102.05	16.35	80.90	16.44	80.57	14.18	70.02
124.14	19.82	80.21	19.64	79.13	17.75	70.66
147.00	23.66	79.74	23.19	78.88	19.20	64.66

Table 5: Uptake capacity of *Mangifera Indica L* and percentage removal of zinc in SMS and BMS

Ci	q	Removal (%)
23.01	3.86	83.96
49.07	6.95	70.85
77.32	11.33	73.31
102.21	15.07	73.73
126.14	18.72	74.22
149.41	18.93	63.35

Ci	(Zn and Cu) Binary solution		(Zn and Ni) Binary solution		(Zn and Pb) Binary solution	
	q	Removal (%)	q	Removal (%)	q	Removal (%)
24.30	4.11	84.78	4.47	84.85	2.72	56.00
48.75	7.03	71.08	7.41	71.94	5.29	54.25
76.00	11.05	72.81	11.23	72.73	7.73	50.84
99.08	15.10	73.76	14.66	72.68	10.64	53.67
124.09	18.74	74.24	18.19	72.30	12.49	50.31
148.45	19.30	63.80	19.22	63.49	15.09	50.08



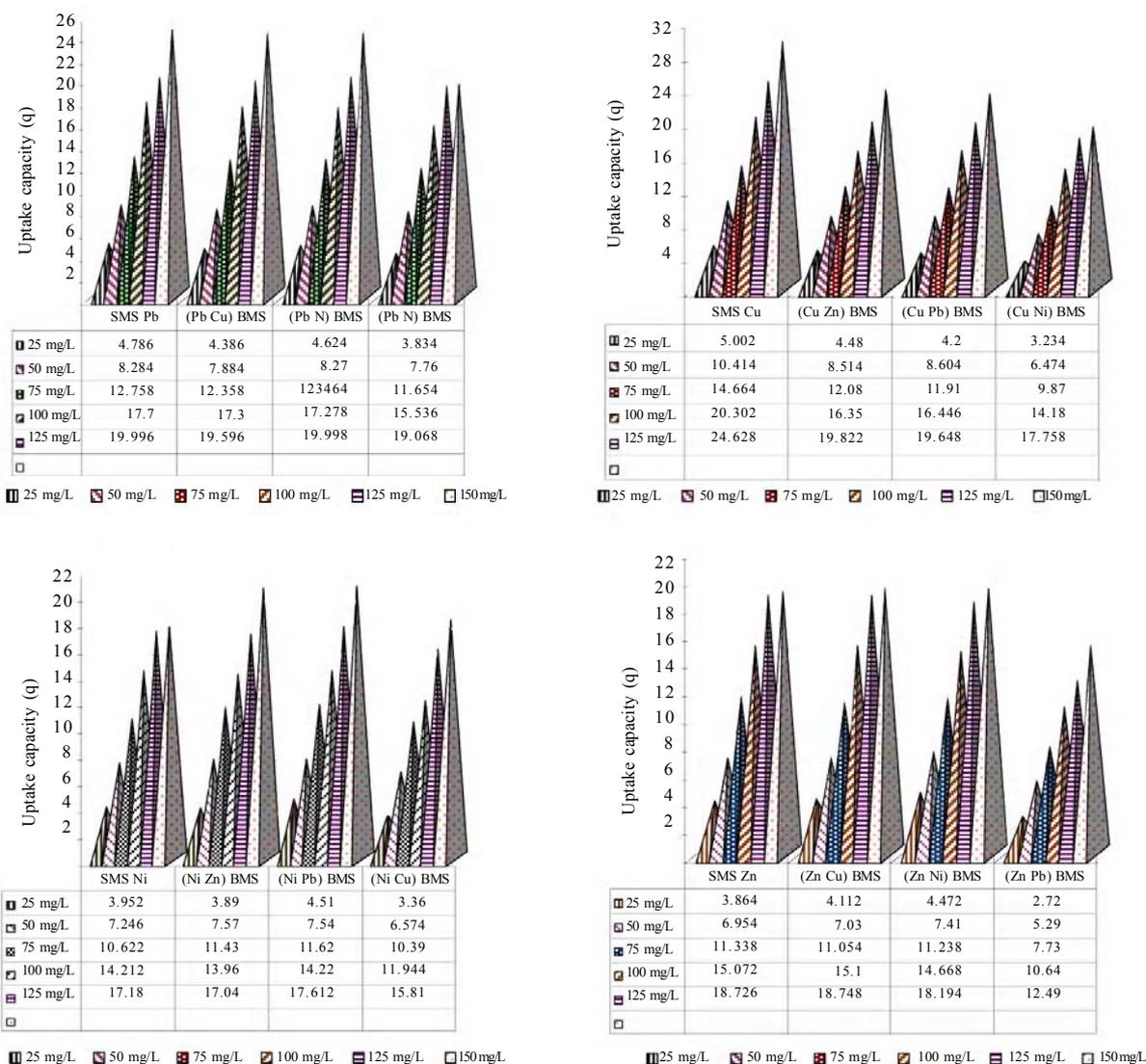


Fig. 3: Comparison of uptake capacity (q) of Pb²⁺, Ni²⁺, Cu²⁺, Zn²⁺ in SMS and BMS by *Mangifera Indica L* biomass

that uptake capacity of Cu²⁺ is 29.37mg/g in SMS but it reduces to 23.66, 23.19 and 19.20mg/g in the presence of Zn²⁺, Pb²⁺ and Ni²⁺, respectively. The reduction is significant in (Cu, Ni) binary solution.

Zn²⁺ uptake is not effected significantly in the presence of Cu²⁺ an Ni²⁺ co-ions in BMS and Pb²⁺ show significant effect on Zn²⁺ sorption. Zn²⁺ sorption reduces from 18.93 to 15.09mg/g in the presence of Pb²⁺ as compared to sorption by

Mangifera Indica L biomass in SMS. This is shown in Table 5 and Fig. 3.

CONCLUSION

The following conclusions can be with drawn from present study:

- 1) The harvesting of the *Mangifera Indica L* biomass is a relatively simple procedure and can be obtained without excessive cost.



- 2) The adsorption range of metal ions including Pb^{2+} , Cu^{2+} , Ni^{2+} and zinc $^{2+}$ ions from dilute acidic solutions at pH 4–6.
- 3) The non-living biomass of *Mangifera Indica L* present comparable biosorption capacity for Pb^{2+} , Cu^{2+} , Zn^{2+} and Ni^{2+} ions with other types of biosorbent materials found in literature.
- 4) Efficiency of the biomass indicates that it is effective to remove metal ions from binary metal solutions, as well as in single metal solution.

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