

## Evaluation of Aluminum and Copper Biosorption in Two-Metal System using Algal Biosorbent

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Biomass of non-living brown seaweed *Sargassum fluitans* pretreated with NaOH is capable of taking up more than 10% ( $q_{\max}$  : 3.85 mmol/g for Al and 1.48 mmol/g for Cu) of its dry weight in the Al and Cu at pH of 4.5. However, the maximum Al and Cu uptakes calculated from Langmuir isotherm were 1.58 mmol/g for Al and 1.35 mmol/g for Cu at pH 3.5. Equilibrium batch sorption study was performed using two-metal system containing Al and Cu. The mathematical model of the two-metal sorption system enabled quantitative estimation of one-metal biosorption inhibition due to the influence of a second metal.

NaOH-treated *S. fluitans* contained 2.19 mmol (43 wt.%) carboxyl groups per gram of biomass. A modified form of Langmuir, which assumes binding of Cu as  $\text{Cu}^{2+}$  and Al as  $\text{Al}(\text{OH})_2^+$ , was used to model the experimental data. This result agrees with the one of mono-valent sorption for Al in single-metal system. The modified Langmuir model gives the following affinity correlated coefficients: 0.196 for Cu and 6.820 for Al at pH 4.5, and 2.904 for Cu and 3.131 for Al at pH 3.5. The interference of Al in Cu biosorptive uptake was assessed by 'cutting' the three dimensional uptake isotherm surfaces at constant second-metal final concentrations. Equimolar final equilibrium concentrations of Cu and Al of 1 mM at pH 4.5 give Cu and Al uptakes reduced by 82.5% and 5.4%, respectively. However, these values at pH 3.5 were 55% (Cu) and 31% (Al).

Key words : aluminum, copper, biosorption, *Sargassum fluitans* biomass,  $\text{Al}(\text{OH})_2^+$ , alginate, two-metal system

### 1. Introduction

Passive metal uptake, observed with a broad range of microbial biomass types, has been investigated with the aim of using it to remove residual toxic or valuable heavy metals from industrial effluents. This biosorption technology and its potential for the treatment of wastewater and environmental pollution has been outlined recently (Volesky, 1990). Different dead biomass types, such as bacteria, fungi and algae have been screened for their capability of adsorbing heavy

metals from solutions (Volesky, 1995). Considerable work carried out with seaweeds, specially from genera *Sargassum* and *Ascophyllum* (Leusch, 1995; Holan, 1993; Kuyucak, 1989), showed high and even commercially interesting sorbent potential of such organisms for metal removal/recovery from dilute solutions. The presence of a large number of metals in industrial metal-bearing solutions makes it necessary to investigate their effect on the final metal uptake by individual biosorbent materials.

Considering the number of metals and different

biosorbent materials of interest, in the absence of a theoretical apparatus, experimental testing of the metals present on the metal uptake by biosorbent materials would require a large volume of laboratory work. Although aluminum is not considered as a major environmental problem, its ubiquitous presence in solutions makes it an obvious investigation target as to its effect on the biosorbent uptake of many other metals of interest. Among them, copper is of a primary interest for its extensive uses and increasing levels in the environment although it is not acutely toxic to humans.

Simple sorption isotherm curves are usually constructed as a result of studying equilibrium batch sorption behavior of different biosorbent materials. They enable quantitative evaluation of sorption performance of these materials in conjunction conventionally only with one metal (de Carvalho, 1994). However, when more than one metal is present in the sorption system, the evaluation of biosorption results, their interpretation and representation become much more complicated. With two metals in the solution, instead of a 2-dimensional biosorption isotherm curve, the system evaluation results in a series of 3-dimensional sorption isotherm surfaces (de Carvalho, 1995). This approach to representing the cadmium biosorption by *Ascophyllum nodosum* biomass in a two-metal system revealed the competitive and inhibitive behavior of the metals present. At the same time, in order to facilitate such a study, mathematical models to represent the experimental data and sorption surfaces had to be proposed and examined for their suitability (Chong, 1995). However, the sorption isotherm surfaces have not been smoothed and showed irregularities which may or may not truly reflect the behavior of the two-metal sorption system. Moreover, the sorption performance of the virgin biosorbent studied then was affected by the

alginate leached from it (de Carvalho, 1994) making it difficult to extrapolate the sorption uptake values over the range of the biosorption studies conducted.

This work illustrates a quantitative approach to evaluating the sorption capacity of a well-stabilized NaOH-treated *S. fluitans* biosorbent, prepared from the biomass of the brown marine alga, in conjunction with solutions containing two metals of interest: Al and Cu. The objective of the present work was to evaluate a two-metal biosorption system behavior involving copper, aluminum and a generally high metal-sorbing biosorbent material of *Sargassum* seaweed. This is considered a first step in studying multimetal biosorption systems which would more closely represent composition of industrial effluents.

## 2. Materials and Methods

### 2.1. Biomass preparation

Raw *S. fluitans* biomass was collected sun-dried on the beach near Naples, Florida. Dry raw biomass was treated by soaking in 0.1 N NaOH solutions in flasks shaken gently on a gyrotory shaker. That is, 1.2 g of raw biomass was added to 200 mL of 0.1 N NaOH (100 rpm shaking overnight at room temperature). Biomass was filtered and washed with the same volume of distilled water and then dried overnight at 60°C. The weight loss of biomass was approximately 37%.

### 2.2. Sorption experiments

The metal solutions were prepared by dissolving  $\text{Al}(\text{NO}_3)_3$  and  $\text{Cu}(\text{NO}_3)_2$  in distilled deionized water to desired initial concentrations.

All sorption experiments were performed by suspending 100 mg of biomass in 100 mL of the metal-bearing solution and shaking on a gyrotory shaker for 30 hours. 0.1 N HCl or 0.1 N NaOH were used for pH adjustments. At the end of each experiment, the samples were filtered (Millipore membrane, 0.45  $\mu\text{m}$ ) and the filtrate was analyzed by AAS (Thermo Jarell Ash, model Smith-HieftjeII, Waltham, MA) for the equilibrium metal content. The filtered biomass was washed with distilled water and dried overnight at 60°C and then weighed for desorption experiments. All desorption experiments were performed by suspending 100 mg of metal-loaded biomass in 100 mL of 0.1 N HCl (pH 1.1) and shaking on a gyrotory shaker for 8 hours. At the end of each desorption experiment, the samples were filtered (Whatman No. 1) and the filtrate was analyzed by AAS. The filtered biomass was washed with distilled water, dried overnight at 60°C and then weighed. The metal uptake was calculated from the results of desorption :

$$\text{experiments as } q \text{ (mmol/g)} = V \cdot C_f / M$$

where:  $C_f$  is the final eluted metal concentration in the solution (mM),  $V$  is the solution volume (L), and  $M$  is the initial mass of the biosorbent used (g).

Alginate was extracted from the dry biomass using 3% solution of  $\text{Na}_2\text{CO}_3$ , according to the method of Percival and McDowell [Percival and McDowell, 1967]. The concentration of alginate in the crude extracts of biomass was determined according to the method of Kennedy and Bradshaw [Kennedy and Bradshaw, 1987], using poly(hexamethylene-biguanidinium chloride)[PHMBH<sup>+</sup>Cl<sup>-</sup>] and UV spectrophotometry.

### 2.3. Three-dimensional sorption isotherm surfaces

The procedure for the equilibrium batch

sorption experiments was the same as described above. The 3-D sorption surfaces were obtained by plotting the experimentally determined final (equilibrium) metal concentrations of both metals respectively on the x and y coordinates against the Al, Cu or total metal uptakes, respectively, on the z-coordinate. Computer program MATLAB 4.0 was used for this purpose. The computer program, MATLAB(Version 4.0), a high performance interactive software package for scientific and engineering numeric computation, is capable of plotting a 3-D diagram based on randomly generated experimental data. It can either simply connect the experimental data points using a 3-D interpolating mesh or it can fit a smoothed surface to data. The latter approach, which is more desirable, requires the MATLAB user to input an appropriate equation representing the surface.

## 3. Results and Discussion

### 3.1. Single-metal sorption

The sorption performance of *S. fluitans* biomass was evaluated through determining the sorption isotherms for the material with Al and Cu, respectively, before examining it in solutions containing both metals together. The single-metal sorption behavior(Fig. 1) of the material followed the Langmuirian pattern:

$$C_f / q = C_f / q_{\text{max}} + 1 / (K \cdot q_{\text{max}}) \quad (1)$$

Equation (1) allowed the determination of the maximum metal uptake ( $q_{\text{max}}$ ) and the Langmuir coefficient  $K(k_{\text{adsorption}}/k_{\text{desorption}})$ , relating to the affinity of the biosorbent for the given metal, both calculated from fitting the Langmuir sorption model to the experimental data.

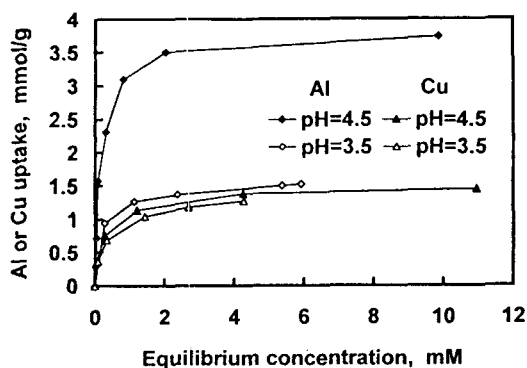


Fig. 1. Effect of pH on the single-metal sorption for the NaOH-treated *S. fluitans* biomass.

Results calculated by Equation (1) for the modified *S. fluitans* are given in Table 1.

The maximum Al uptake value obtained at pH 4.5 is much higher than that for the other metals.

The weight gain of biomass during the Al sorption process was up to 23% with 101 mg/g of Al uptake at pH 4.5. That is, the weight gain for high Al uptake by biomass was 2.3 times higher than the Al uptake. This may be an indication that the biomass hydroxyl groups were involved in sequestering the aluminum in the form of  $Al(OH)_2^+$ . As shown in Fig. 1, the biosorbent uptake of aluminum was strongly affected by the pH of the solution, while the uptake of copper was only slightly affected by the pH. This is also the reason why the concentration of hydroxyl ions have an important effect upon Al uptake.

On the other hand, the alginate content of NaOH-treated *S. fluitans* was 2.19 mmole(43 wt.%) carboxyl groups per gram of biomass, indicating the lower value than the binding sites of Al uptake at pH 4.5. Fourest et al(1996) reported that cadmium binding by *S. fluitans* biomass was based on bridging or bidentate complex formation with the carboxyl groups of

the alginate and that this biomass contained sulfonate groups( $0.27 \text{ meq/g} \pm 0.03$ ) and alginate (45% of the dry weight) corresponding to 2.25 meq. of carboxyl groups per gram of biomass. This indicated that other binding sites, likely the polyphenol groups, in addition to alginate and sulfonate groups, contribute to aluminum binding.

Table 1. Single-metal sorption constants for NaOH-treated *S. fluitans* biomass.

metal	equilibrium pH	$q_{\max}$ (mmol/g)	$K$ (mM) <sup>-1</sup>
Al	4.5	3.85	6.487
Al	3.5	1.58	3.923
Al	2.5	1.05	2.592
Cu	4.5	1.48	3.820
Cu	3.5	1.35	3.210
Cu	2.5	0.55	2.104

### 3.2. Biosorption in a two-metal system

Creating an interpolated sorption isotherm surface without smoothing the experimental data randomly generated results in surface irregularities. Smoothing of the sorption isotherm surface is possible through assigning a mathematical model to them which can represent the randomly distributed experimental data(Volesky, 1995; Chong, 1995). This approach makes it possible to eventually derive two-dimensional sorption isotherm curves from the 3-D sorption isotherm surface image by cutting with parallel iso-concentration planes for one or the other metal selected concentration values, respectively. The resulting set of sorption isotherm curves depicts either the effect of the second metal on the biosorption of the first one or vice versa in an easily understandable manner. These curves reflect then correctly the actual equilibrium biosorption conditions as appropriate (de Carvalho, 1995).

When both ions were present in the solution

together(2-metal experiments), some reduction of the Cu uptake could be observed with increasing Al concentrations. This is well indicated in the series of simple isotherm curves generated as iso-concentration cuts of the 3-dimensional plots whereby the two(equilibrium) metal concentrations are plotted against the Cu(Fig. 2), Al(Fig. 3) and total metal uptakes(Fig. 4), respectively.

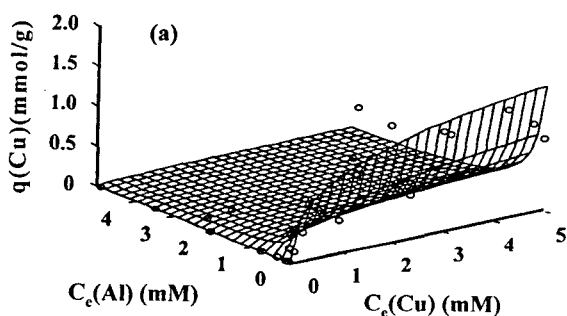


Fig. 2(a). A 3-dimensional sorption surface for the Cu-Al biosorption system: Copper uptake at pH 4.5.

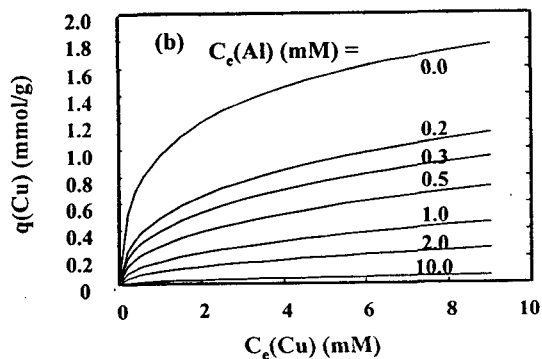


Fig. 2(b). Effect of Al on the equilibrium uptake of Cu by NaOH-treated *S. fluitans* biomass at pH 4.5.

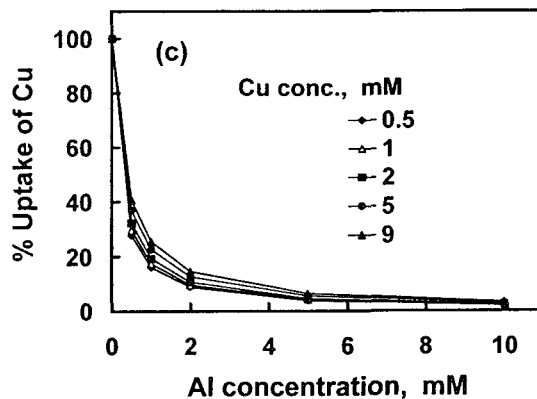


Fig. 2(c). The summary of the effect of Al presence on the Cu uptake NaOH-treated *S. fluitans* biomass at pH 4.5. Equilibrium Cu concentrations arbitrarily selected.

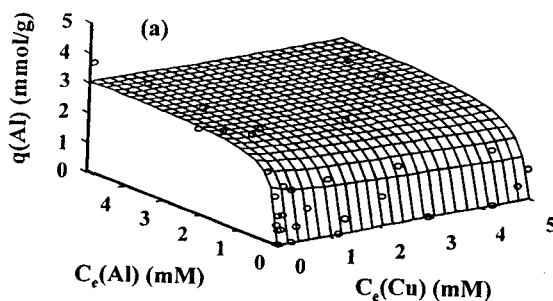


Fig. 3(a). A 3-dimensional sorption surface for the Cu-Al biosorption system: Aluminum uptake at pH 4.5.

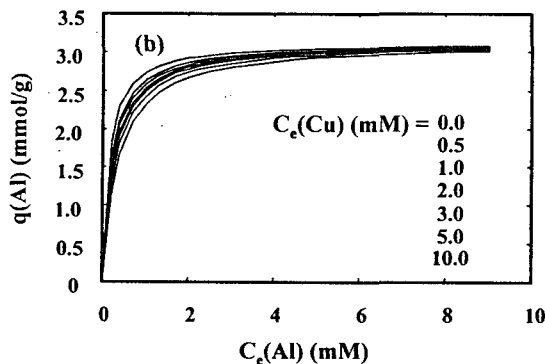


Fig. 3(b). Effect of Cu on the equilibrium uptake of Al by NaOH-treated *S. fluitans* biomass at pH 4.5.

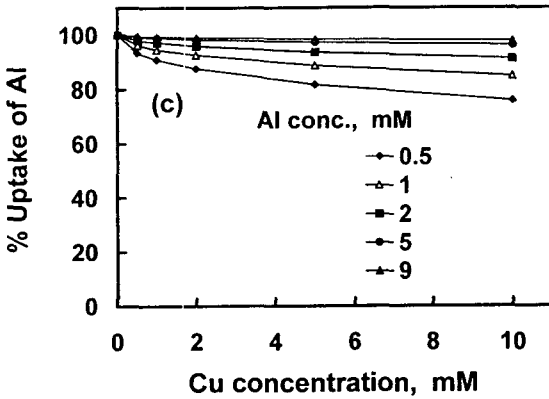


Fig. 3(c). The summary of the effect of Cu presence on the Al uptake NaOH-treated *S. fluitans* biomass at pH 4.5. Equilibrium Al concentrations arbitrarily selected.

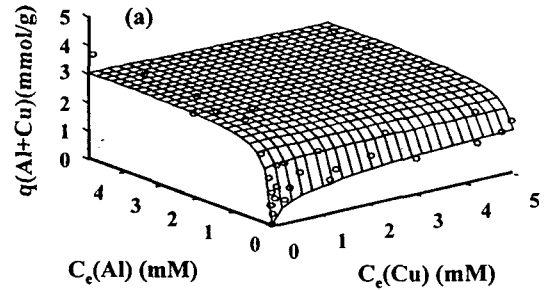


Fig. 4(a). A 3-dimensional sorption surface for the Cu-Al biosorption system: Total metal uptake at pH 4.5.

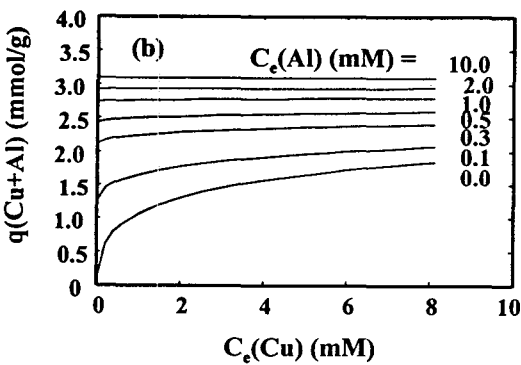


Fig. 4(b). Effect of Al on the equilibrium total metal uptake by NaOH-treated *S. fluitans* biomass at pH 4.5.

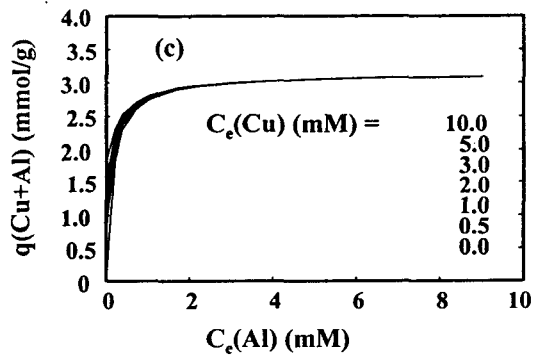
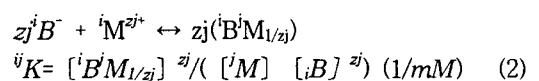


Fig. 4(c). Effect of Cu on the equilibrium total metal uptake by NaOH-treated *S. fluitans* biomass at pH 4.5.

### 3.3. Modeling in two-metal system at fixed pH

Multicomponent Langmuir isotherms for one binding site and formation of 1:1 sorbate/sorbent complexes have been described by Hill(1977). A case including one gaseous species that occupies two sites after dissociation was also considered by that author. These multicomponent isotherms were adapted for the case of multi-site and multi-ion biosorption for different ion valences.

Since biosorption by seaweed biomass is largely an ion-exchange phenomenon, it was assumed that each cationic species sorbs to the number of monovalent ionized groups, which is equal to the valence of the cation, such that the overall charge is preserved. The equation for a cation  ${}^iM$  of the charge  $z_j$  sorbing to a binding site  ${}^iB$  is for ideal behavior(all activity coefficients equal to unity):



The first character of the upper left index of the equilibrium constant (or of the metal binding  $q$ ) refers to the binding site, the following to the sorbed ion. Protons are treated as one of the monovalent species with  $z_j=1$ . Divalent ions form  ${}^iB^jM_{0.5}$  complexes. Note that the term  $z_j^i B^j M_{1/z_j}$  has been chosen instead of  ${}^iB_{z_j}^j M$  because it implies that a number of bonds equal to the charge  $z_j$  of the ion have to be broken for the release of an  $z_j$ -valent ion, i.e., the dissociation reaction is of the order  $z_j$  and not of order one as this would be the case for the dissociation of a  ${}^iB_{z_j}^j M$  species.

The following equation (3) adapted, based on equation (2), for the modeling of multi-site and multi-component adsorption system was developed by many authors Schiewer and Volesky, 1996; Fritz and Schluender, 1974; Crittenden and Weber, 1978).

$${}^i q = \sum_{k=1}^m [{}^k B^j M_{1/z_j}] \\ = \sum_{k=1}^m {}^k B \frac{({}^k K [{}^i M])^{1/z_j}}{1 + \sum_{h=1}^m ({}^h K [{}^h M])^{1/z_h}} \quad (\text{meq/g}) \quad (3)$$

If only one site is present (i.e.,  $m=1$ ) eq. (3) can be regarded as a specific case of the Fritz-Schluender isotherm (Fritz and Schluender, 1974), which however needs  $n(2n+3)$  parameters in order to describe a  $n$  solute system (i.e., 14 for a two-solute system). The model presented here needs only  $(n+1)$  parameters (i.e., 3 for a two-solute system) for the one site case and  $(2n+2)$  for the two-site case. Since the exponents  $1/z_j$  are related to the ion valence, they do not constitute adjustable parameters. Since Fritz-Schluender isotherm needs such a high number of constants, it has frequently been used in different simplified forms (Crittenden, 1978; Liapis, 1977), which include the so called Langmuir-Freundlich isotherm (Ruthven, 1984).

There are the binding sites of carboxyl groups

of alginate as a major part (more than 90%) and the ones of sulfonate and polyphenol groups and etc. as a minor part in *Sargassum fluitans* biomass (Fourest, 1996). Therefore, the model case was considered as one site of carboxyl groups for the sorption system. In two-metal system of aluminum and copper, cations are protons and sodium ion added for pH adjustment in addition to aluminum and copper ions. Since previous work (Lee, 1997) noted that protons and sodium ion not sorbed at all in this system, eq. (3) is reduced to

$${}^1 q = \frac{{}^1 B ({}^1 K [{}^1 M])^{1/z_1}}{1 + ({}^1 K [{}^1 M])^{1/z_1} + ({}^2 K [{}^2 M])^{1/z_2}} \quad (4)$$

$${}^2 q = \frac{{}^2 B ({}^2 K [{}^2 M])^{1/z_2}}{1 + ({}^1 K [{}^1 M])^{1/z_1} + ({}^2 K [{}^2 M])^{1/z_2}} \quad (5)$$

The model produced an equation with three parameters ( ${}^1 B$ ,  ${}^1 K$ ,  ${}^2 K$ ). These parameters, related to each of the two-metal systems studied, were evaluated by minimizing the sum of squared residuals (residual refers to the difference between experimental metal uptake and that predicted from the model). Again, the MATLAB 4.0 program was used for this purpose.

#### 3.4. Al-Cu system with NaOH-treated *S. fluitans*

With Al and Cu, there is the possibility of six pairs of binding cases:  $[Cu^{2+} - Al^{3+}]$ ,  $[Cu^{2+} - Al(OH)^{2+}]$ ,  $[Cu^{2+} - Al(OH)^{2+}]$ ,  $[Cu^+ - Al^{3+}]$ ,  $[Cu^+ - Al(OH)^{2+}]$ , and  $[Cu^+ - Al(OH)_2^+]$ . To quantify the agreement between the model predictions and experimental observations for the behavior of the two-metal sorption system, an objective function relating the experimental and the predicted metal uptakes may be defined for each component (Shallcross et al., 1988):

$$E_i = \sum \{ (q_i^{\text{exp}} - q_i^{\text{mod el}}) / q_i^{\text{exp}} \} \quad (6)$$

Among the six pairs of binding cases, the pair of binding case minimizing the value  $E_i$  of Equation (6) was  $[\text{Cu}^{2+} - \text{Al}(\text{OH})_2^+]$ . This result agrees with the cre for mono-valent Al-containing ionic species described in the single-metal sorption (Lee, 1998).

The modified Langmuir model equation can be represented by 3-D sorption isotherm surfaces as presented in Fig. 2(a), 3(a) and 4(a). While these 3-D isotherm surfaces represent the summary of the two-metal equilibrium results, the selected cuts through the 3-D diagrams presented in Fig. 2(b) and 3(b) better reveal the quantitative trends observed in the two-metal systems. Moreover, the effect of the secondary metal presence on the uptake of the primary metal can be summarized from these plots as seen in Fig. 2(c) and 3(c) for Cu and Al sorption, respectively.

The uptake of Cu was greatly reduced by the presence of Al, whereas the uptake of Al remained relatively constant when Cu was present. The Cu uptake reduction was much more pronounced at pH 4.5 (uptake  $q = 0.68$  mmol Al/g at  $C_{f, \text{Al}} = 0.04$  mM was taken as 100%; uptake  $q = 0.25$  mmol Cu/g was at  $C_{f, \text{Cu}} = 0.04$  mM was taken as 100%). Fig. 3(c) shows that at  $C_{f, \text{Cu}} = 2.0$  mM and  $C_{f, \text{Al}} = 2.0$  mM, Al uptake was reduced to 95.9%, whereas the uptake of Cu to 10.7% in the presence of the same 2 mM of Al and Cu (Fig. 2(c)). Another observation at pH 4.5 which can be made from Fig. 5 is that the maximum uptake of Al is much higher than that of Cu, 3.1 mmol/g versus 1.8 mmol/g. These values differ from the ones (3.85 mmol/g for Al and 1.48 mmol/g for Cu) obtained from the respective single-metal systems. The number of binding sites for Al was reduced only marginally in the presence of Cu, while those for Cu slightly increased in the presence of Al. On the other

hand, the maximum uptake of Al at pH 3.5 (1.5 mmol/g) was only slightly higher than that of Cu (1.3 mmol/g) in two-metal system. The Al uptake, was very sensitive to the pH of the solution as could be expected if Al was sorbed in the form of  $\text{Al}(\text{OH})_2^+$ .

Fig. 4(b) and (c) were obtained when the total metal uptake sorption surface is analyzed and viewed in two series of "iso-concentration cuts" by constant Al or Cu concentration planes, respectively. In summary, they reveal that when Sargassum biosorbent becomes saturated in the system containing high concentrations of both metals of interest in this study (Al and Cu), the common maximum total metal uptake approaches the value of 3.14 mmol/g. It should be noted that the lowest isotherm curves in Fig. 4(b) and 4(c) again represent the one-metal biosorption isotherm for Al and Cu, respectively.

The curves resulting from the cuts of the 3-D diagrams conform well to the modified Langmuir model. The Langmuir parameters,  ${}^1K$  for Cu and  ${}^2K$  for Al, the ratio of the adsorption rate constant to the desorption rate constant, are an indication of the "relative affinity" of the biosorbent toward a metal. Figure 5 shows that the  ${}^1K$  and  ${}^2K$  values from the modified Langmuir

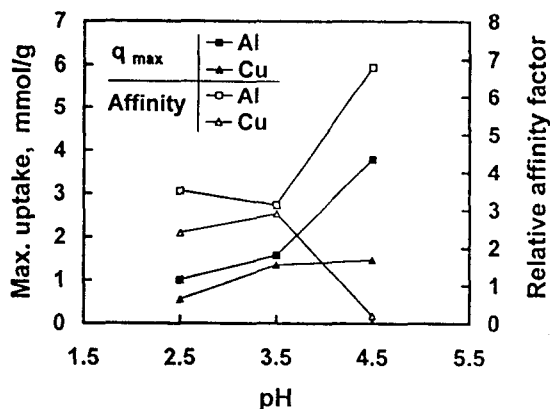


Fig. 5. Effect of pH on the maximum metal uptake and relative affinity factor in 2-metal biosorption system.



model are:  $0.196 \text{ (mM)}^{-1}$  and  $6.820 \text{ (mM)}^{-1}$  at pH 4.5,  $2.904 \text{ (mM)}^{-1}$  and  $3.131 \text{ (mM)}^{-1}$  at pH 3.5,  $2.393 \text{ (mM)}^{-1}$  and  $3.508 \text{ (mM)}^{-1}$  at pH 2.5. The maximum uptake of Al in the one-metal system relatively increased between pH 3.5 and pH 4.5. Within this pH range, the relative affinity of Al in the (Al+Cu) system greatly increased, but the one of Cu oppositely decreased.

Quantitative expressions of the preferential Al biosorption in the (Al+Cu) system are greatly dependent on the pH of solution and will be the subject of continuing studies which will yield further insight into the mechanism of the biosorbent metal uptake by *Sargassum* biomass of different metals. While ion exchange has been revealed as the predominant sequestering mechanism in this biosorption system for some metals (Schiewer, 1995), its presence in conjunction with other metals remains to be confirmed. Future work should include performing experiments to investigate these phenomena, which are apparently related to the mechanisms of metal biosorption. However, one has to realize the complexity of investigating the mechanisms of biosorption which could be any or a combination of these processes: Complexation, ion exchange, physical adsorption, chelation, or inorganic microprecipitation of metals. The sorption preferences observed with NaOH-treated *S. fluitans* biomass and the results of the competition aspects observed will likely have repercussions in the continuous-flow dynamic sorption column system. A net sorption preference for Al uptake over Cu in the relevant concentration range studied (0.0 to 5.0 mM) indicates the possibility of its use in a flow arrangement as a 'selective filter' for enrichment of given metallic species. Other authors reported that the use of microbial biosorbent for separation of Pd from Cu (Brierley, 1988) or resins for separation of Cu from Zn (Akita, 1990). It is important to stress that a change in the pH of the

sorption system could significantly alter the sorption capacities and selectivities in two-metal sorption system studied here.

#### 4. Conclusion

Equilibrium batch sorption study was performed using two-metal system containing Al and Cu. The mathematical model of the two-metal sorption system enabled quantitative estimation of one-metal biosorption inhibition due to the influence of a second metal.

The application of MATLAB 4.0 enabled the current work to produce results which can serve as a more reliable and accurate quantitative basis for judging the performance of new biosorbent material. At the same time, a suitable methodology is outlined here which represents step in standardizing the evaluation of biosorption performance.

The interference of Al in Cu biosorptive uptake was greatly affected at pH 4.5. Equimolar final equilibrium concentrations of Cu and Al of 1 mM at pH 4.5 give Cu and Al uptakes reduced by 82.5% and 5.4%, respectively. However, these values at pH 3.5 were 55% (Cu) and 31% (Al).

*Sargassum* biosorbent becomes saturated in the system containing high concentrations of both metals of interest in this study (Al and Cu), the common maximum total metal uptake approaches the value of 3.14 mmol/g.

#### Acknowledgement

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#### Nomenclature

$B_z^jM$ ,  $B_jM_{1/zj}$  : metal-biomass complexes

- $^tB$  : amount of binding site of biomass (meq/g)  
 $C_f$  : final equilibrium concentration (mM)  
 $^iK$  : equilibrium constant of complexation site i cation j (L/g mol)  
 $^iM^{zj}$  : ionic species j of valence zj  
 $^i q$  : uptake of metal i at equilibrium concentration  $[^iM]$  (meq/g)  
 $q_i^{exp}$  : experimental metal uptake for each component  
 $q_i^{model}$  : predicted metal uptakes  
 $zj$  : charge (valence) of ion j, stoichiometric factor

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