



Competitive biosorption of cadmium(II) and zinc(II) ions from binary systems by *Sargassum filipendula*

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ABSTRACT

The present work describes our study on the competitive biosorption of Cd(II) and Zn(II) ions onto *Sargassum filipendula* from single component and binary systems. Results showed that *S. filipendula* was an efficient biosorbent for the removal of Cd(II) and Zn(II) ions from an aqueous solution. The equilibrium experimental data for the single component system for the two metallic species fitted well to Langmuir and Langmuir–Freundlich isotherm models. Seven isotherm models were tested with the equilibrium data for modeling of the binary system. Based on the sum of squares error, the Langmuir–Freundlich isotherm model showed the best fit to the binary adsorption data. The influence of the additional cation on the behavior of the biosorption of Cd²⁺ and Zn²⁺ was analyzed comparing single component and binary isotherms. It was observed that Zn²⁺ had a relevant effect on the Cd²⁺ biosorption, but the interference of Cd²⁺ on the sorption of Zn²⁺ was considerably less intense.

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1. Introduction

Industrial activities have contributed to the increased contents of metallic ions in natural waters, due to the great volumes of effluent contaminated with toxic metals such as cadmium, copper, lead, chromium, and zinc.

Cadmium has attracted wide attention of environmentalists as one of the most toxic metals. The major sources of cadmium release into the environment by waste streams are electroplating, smelting, alloy manufacturing, pigments, plastic, battery, mining and refining processes (Tsezos, 2001). Cadmium has been recognized for its negative effects on the environment where it readily accumulates in living systems (Sharma, 1995).

Zinc is an essential element for the human organism, since it participates in protein and nucleic acid metabolism, stimulates the activity of more than a hundred enzymes and plays an important role in effectively operating the immunologic system. However, it is toxic for humans at levels of 100–500 mg/day (Volesky and Holan, 1995). This metal is very extensively used by many industries, such as in galvanizing and in manufacturing brass and other alloys, batteries and pigments.

A number of methods exist for the removal of heavy metal pollutants from liquid wastes. These include physico-chemical methods, such as chemical precipitation, chemical oxidation or reduction, electrochemical treatment, application of membrane

technology, evaporation recovery, solvent extraction and ion-exchange processes. However, these techniques may be ineffective or extremely expensive, especially when the metals are dissolved in large volumes of solution at relatively low concentrations (around 1–100 mg/L) (Volesky, 2003).

In this context, biosorption can be considered as an alternative technology for industrial wastewater treatment when the concentration of transition metals is low. Biosorption is based on the ability of biological materials to accumulate heavy metals from wastewater by physico-chemical pathways of uptake. This innovative depurative process uses biomaterials, such as marine algae or wastes coming from industrial and biological processes, which are either low-cost and abundant in nature (Benguella and Benaissa, 2002; Holan et al., 1993; Volesky and Holan, 1995; Volesky and May-Phillips, 1995).

The biomass of brown algae of the *Sargassum* genus is reported to possess a metal binding capacity superior to other organic and inorganic sorbents. Although actual wastewater treatment systems often have to deal with a mixture of heavy metals, most biosorption studies involving *Sargassum* and other biomasses only focus on the removal of heavy metal cations from single metal solutions (Aksu, 2001; Antunes et al., 2003; Brierley et al., 1986; Britto et al., 2007; Cruz et al., 2004; Davis et al., 2004; Luna et al., 2007; Martins et al., 2006; Matuschka et al., 1993; Pagnanelli et al., 2003; Vieira et al., 2007). On the other hand, studies concerning the adsorption in multi-metal systems found in the literature are limited (Aksu et al., 2002; Apiratikul and Pavasant, 2006; Carvalho et al., 1995; Chong and Volesky, 1995; Fagundes-Klen et al., 2007; Kumar

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et al., 2008a; Sanchez et al., 1999; Srivastasa et al., 2006; Sun et al., 2008). In multi-metal systems, systematic studies on the biosorption equilibrium of transition metals are very important since it is influenced not only by the biomass surface properties and the operational conditions (pH, temperature, metallic ion concentration) but also by the competition among the different cations for the active sites of biosorbent and/or the screening effect of the cations, which interfere with the capacity of biosorption of the metallic species of interest.

Thus, since a multi-metal biosorption system would more closely represent the composition of industrial effluents, in the present work the competitive biosorption of Cd^{2+} and Zn^{2+} by *Sargassum filipendula* was investigated. A quantitative approach based on the use of different isotherm models was used to evaluate the biosorption capacity of the biomass in the binary metal species system. These results were also compared with previous experimental data obtained for single-metal systems (Britto et al., 2007; Cruz et al., 2004) in equivalent conditions.

2. Isotherms

Biosorption is a phenomenon based on ion exchange, but also potentially involving additional physicochemical effects, such as, adsorption (chemisorption and physisorption) and complexation. Despite the complexity of this process, the basic approach is to describe the equilibrium of metal ions between the solution and the biosorbent by sorption isotherms. Although traditional isotherms have been obtained in a completely different context (e.g., gas adsorption on activated carbon and/or metal surfaces), their application for biosorption description shows good results (Maurya and Mittal, 2006).

2.1. Single component isotherms

2.1.1. Langmuir isotherm

The Langmuir isotherm assumes the form:

$$q_e = \frac{Q_0 K_L C_e}{1 + K_L C_e} \quad (1)$$

where Q_0 is the maximum amount of metal ion per unit weight of sorbent to form a complete monolayer on the surface, and K_L is the equilibrium adsorption constant which is related to the affinity of the binding sites. The parameter Q_0 represents a practical limiting adsorption capacity when the surface is fully covered with metal ions and allows the comparison of adsorption performance, particularly in the cases where the sorbent does not reach its full saturation in experiments (Aksu, 2001).

2.1.2. Freundlich isotherm

This isotherm is an empirical equation which considers the heterogeneity of the adsorbent surface:

$$q_e = K_F C_e^{1/n} \quad (2)$$

where K_F and n are the Freundlich parameters. Differing from the Langmuir isotherm, Freundlich isotherm does not predict the saturation of adsorbent sites.

2.1.3. Langmuir–Freundlich isotherm

Langmuir–Freundlich isotherm is expressed by the following equation:

$$q_e = \frac{Q_0 K_{LF} C_e^{1/n}}{1 + K_{LF} C_e^{1/n}} \quad (3)$$

where Q_0 , K_{LF} and n are the isotherm parameters. Depending on their values, this isotherm may result in the following: if $n = 1$ then

it becomes the Langmuir isotherm, if K_{LF} is very small then it becomes the Freundlich isotherm.

2.2. Binary isotherms

2.2.1. Non-modified Langmuir isotherm

This isotherm is based on the utilization of single component sorption data to describe the behavior of sorption of mixtures using the Langmuir isotherm (Aksu et al., 2002):

$$q_{e,i} = \frac{Q_{0,i} K_{L,i} C_{e,i}}{1 + \sum_{j=1}^2 K_{L,j} C_{e,j}} \quad \text{for } i = 1, 2 \quad (4)$$

where the parameters in this equation are obtained from the single-component Langmuir isotherm.

2.2.2. Modified Langmuir isotherm

In this alternative, parameters η_i estimated from binary data are inserted in the previous isotherm (Aksu et al., 2002):

$$q_{e,i} = \frac{Q_{0,i} K_{L,i} (C_{e,i}/\eta_i)}{1 + \sum_{j=1}^2 K_{L,j} (C_{e,j}/\eta_j)} \quad \text{for } i = 1, 2 \quad (5)$$

2.2.3. Competitive Langmuir isotherm

A competitive model based on the Langmuir hypotheses implies the following equilibrium equations between the cations and the adsorbent sites (Apiratikul and Pavasant, 2006):



Thus, the resultant isotherm can be represented by the expression:

$$q_{e,i} = \frac{Q_0 K_{L,i} C_{e,i}}{1 + \sum_{j=1}^2 K_{L,j} C_{e,j}} \quad \text{for } i = 1, 2 \quad (8)$$

where all parameters are obtained from multicomponent sorption data.

2.2.4. Uncompetitive Langmuir isotherm

Another variant of the Langmuir equation can be established based on the assumption that the two cations can also be attached to a single site simultaneously, i.e., together with the equilibrium equations expressed by Eqs. (6) and (7), there is the following relation:



These three equilibrium equations determine the following isotherm expression (Apiratikul and Pavasant, 2006):

$$q_{e,i} = Q_0 \left(\frac{K_{L,i} C_{e,i} + K_{L,ij} C_{e,i} C_{e,j}}{1 + K_{L,i} C_{e,i} + K_{L,j} C_{e,j} + K_{L,ij} C_{e,i} C_{e,j}} \right) \quad \text{for } i, j = 1, 2 \text{ with } i \neq j \quad (10)$$

where all parameters are obtained from multicomponent sorption data.

2.2.5. Partial competitive Langmuir isotherm

This Langmuir derived isotherm considers that a cation can be attached to a free binding site or to a site already occupied by a different cation. In this case, the equilibrium equations represented by Eqs. (6) and (7) are complemented by:



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