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Biochemical Engineering Journal 23 (2005) 185–192

Biochemical Engineering Journal

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Study on biosorption of Cr(VI) by *Mucor hiemalis*

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Received 9 December 2003; received in revised form 14 September 2004; accepted 5 January 2005

Abstract

Many investigations have been carried on metal binding capacity of different groups of microorganisms. However, the reports on the kinetic, thermodynamic and desorption study of biosorption process are quite limited. The present study was carried out in a batch system using *Mucor hiemalis* for its sorption and desorption study of Cr(VI). *M. hiemalis* exhibited the highest Cr(VI) uptake of 53.5 mg/g at an initial pH of 2.0. Equilibrium data fitted well to Langmuir isotherm model. Biosorption showed pseudo-second order rate kinetics at different initial concentration of Cr(VI) and different dose of *M. hiemalis*. The activation energy of the biosorption (E_a) was estimated as 4.0 kJ/mol using Arrhenius equation. Using the equilibrium constant value obtained at different temperature, the thermodynamics properties of the biosorption (ΔG°, ΔH° and ΔS°) were also determined. The biosorption of Cr(VI) onto *M. hiemalis* was found to be endothermic. Desorption data showed that nearly 99% of the Cr(VI) adsorbed on *M. hiemalis* could be desorbed using 0.1N NaOH. Study with the cyclic use of a batch of *M. hiemalis* repeatedly after desorption, showed that it retain its activity up to five sorption and desorption cycles.

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Keywords: Adsorption; Biosorption; Bioremediation; *Mucor hiemalis*; Wastewater treatment; Chromium; Desorption

1. Introduction

Rapid industrialization and increase in population has led many-fold increase in the utilization and release of chemicals including heavy metals. As consequences, these metals are found in toxic concentrations in aqueous systems. Heavy metals like mercury, cadmium, lead, nickel, and chromium are toxic, even in extremely minute quantities. Anthropogenic sources of chromium are industries viz., electroplating, leather tanning, metal finishing, chemical industries and many others. Chromium exists in several oxidation states out of which Cr(III) and Cr(VI) are most stable form. Because of its high toxicity and potential carcinogenicity Cr(VI) is of especial concern. In humans, Cr(VI) causes severe diarrhea, ulcers, eye and skin irritation, kidney dysfunction and probably lung carcinoma [1]. Various methods used for removal of Cr ions include chemical reduction and precipitation, reverse osmosis, ion exchange and adsorption on activated carbon or similar material. But all these methods suffer from severe constraints, such as incomplete metal removal, high reagent or energy requirements, generation of toxic sludge or other waste products that require safe disposal. Some of the treatment methods involve high operating and maintenance cost. There is, therefore, a need for some alternative technique, which is efficient and cost effective. Biosorption could be such an alternative method of treatment. It employs wide variety of biomass for removal of metal ions such as algae [2], fungi [3] and bacteria [4]. Biosorption has distinct advantages over conventional methods of treatments: the process does not produce chemical sludge, hence non polluting, it is more efficient, easy to operate. It is very efficient for removal of pollutants from very dilute solutions also. Since biosorption often employs dead biomass this eliminates the need of nutrient requirement and can be exposed to environments of high toxicity [5]. A major advantage of biosorption is that it can be used in situ, and with proper design may not need any industrial process operations and can be integrated with many systems in the most eco-friendly manner. In this study, *Mucor*

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doi:10.1016/j.bej.2005.01.011

hiemalis was used as biosorbents for the removal of Cr(VI). The objective of the present investigation was to study the biosorption equilibrium, kinetic, thermodynamics and desorption.

1.1. Equilibrium study of biosorption

The equilibrium established between adsorbed phase on the biosorbents and that in solution can be represented by adsorption isotherms. The most widely used isotherms for modeling equilibrium are Freundlich and Langmuir isotherm equations. The Freundlich isotherm equation is an empirical equation based on the sorption on a heterogeneous surface suggesting that binding sites are not equivalent and/or independent. The monocomponent Freundlich isotherm equation [6] is given below:

$$q_{\mathrm{eq}} = K_{\mathrm{f}} C_{\mathrm{eq}}^{1/n} \tag{1}$$

where K_{f} and n are Freundlich constants related to sorption capacity and sorption intensity, respectively. From the linearized plot of $\log q_{\mathrm{eq}}$ versus $\log C_{\mathrm{eq}}$, these constants can be determined.

Langmuir isotherm equation is based on monolayer sorption onto a surface with finite number of identical sites, which are homogeneously distributed over the sorbent surface and is given by the Eq. (2) [7]

$$q_{\mathrm{eq}} = \frac{Q_{\max} b C_{\mathrm{eq}}}{1 + b C_{\mathrm{eq}}} \tag{2}$$

where $Q_{\max}$ and b are Langmuir constants denoting maximum adsorption capacity and the affinity of the binding sites, respectively. These constants can be determined from the $1/C_{\mathrm{eq}}$ versus $1/q_{\mathrm{eq}}$. This also represents the physical bondage between the component and adsorbent.

1.2. Kinetic model

Pseudo-first order and pseudo-second order rate equation have been used for modeling the kinetics of Cr(VI) biosorption. Pseudo-first order rate equation is expressed as follows [8]

$$\frac{\mathrm{d}q_t}{\mathrm{d}t} = k_1(q_{\mathrm{eq}} - q_t) \tag{3}$$

where q_t and q_{eq} is sorption capacity at time t and at equilibrium, respectively and k_1 is pseudo-first order rate constant. After integration and applying boundary conditions $t = 0$ and $q_t = 0$ to $t = t$ and $q_t = q_{\mathrm{eq}}$ at equilibrium, Eq. (3) becomes

$$\log(q_{\mathrm{eq}} - q_t) = \log q_{\mathrm{eq}} - \frac{k_1 t}{2.303} \tag{4}$$

where value of k_1 can be determined from the slope of the plot of the $\log(q_{\mathrm{eq}} - q_t)$ versus t.

Similarly, pseudo-second order rate equation is expressed as

$$\frac{\mathrm{d}q}{\mathrm{d}t} = k_2(q_{\mathrm{eq}} - q_t)^2 \tag{5}$$

where k_2 is pseudo-second order rate constant. After integration and applying the same boundary conditions $t = 0$ and $q_t = 0$ to $t = t$ and $q_t = q_{\mathrm{eq}}$ at equilibrium, Eq. (5) becomes

$$\frac{t}{q_t} = \frac{1}{k_2(q_{\mathrm{eq}})^2} + \frac{t}{q_{\mathrm{eq}}} \tag{6}$$

where value k_2 can be determined from the intercept of linearized plot of t/q_t versus t.

1.3. Thermodynamics of biosorption

1.3.1. Activation energy

Activation energy is determined according to the Arrhenius equation

$$\ln k = \frac{-E_{\mathrm{a}}}{RT} + \ln A_0 \tag{7}$$

where E_{a} is activation energy and A_0 is constant called the frequency factor. Value of E_{a} can be determined from the slope of $\ln k$ versus $1/T$ plot.

1.3.2. Gibbs free energy change (ΔG°)

ΔG° is the fundamental criterion of spontaneity. Reaction occurs spontaneously at given temperature if the value of ΔG° is negative. Value of ΔG° can be determined from the following equation

$$\Delta G^\circ = -RT \ln b \tag{8}$$

where R is gas constant (8.314 J/mol K), b is Langmuir constant and T is absolute temperature.

1.3.3. Enthalpy (ΔH°) and entropy (ΔS°) change

Relation between ΔG°, ΔH° and ΔS° can be expressed by the following equations

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \tag{9}$$

Eq. (9) can be written as

$$-RT \ln b = \Delta H^\circ - T\Delta S^\circ \tag{10}$$

or

$$\ln b = -\frac{\Delta H^\circ}{RT} + \frac{\Delta S^\circ}{R} \tag{11}$$

where values of ΔH° and ΔS° can be determined from the slope and the intercept of the plot between $\ln b$ versus $1/T$.

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