Arabian Journal of Chemistry (2015) 8, 506–511

King Saud University

Arabian Journal of Chemistry

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ORIGINAL ARTICLE

Removal of mercury(II) from aqueous solutions by biosorption on the biomass of *Sargassum glaucescens* and *Gracilaria corticata*

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Received 12 August 2011; accepted 31 January 2012
Available online 8 February 2012

KEYWORDS

Mercury(II);
Removal;
Adsorption;
Gracilaria corticata;
Sargassum glaucescens

Abstract Previous studies have shown the batch removal of Pb^{2+} ions from wastewater and aqueous solution using two different algae; *Gracilaria corticata* (red algae) and *Sargassum glaucescens* (brown algae). The objective of this research is to study the batch removal of Hg^{2+} from aqueous solution, and wastewater using marine dried red algae *G. corticata* (red algae) and *S. glaucescens* (brown algae). Marine alga was used as a low-cost absorbent. The effects of pH, biosorption time and the concentration of the Hg^{2+} solution were examined in the adsorption process with *S. glaucescens* and *G. corticata*. Results showed that when we used *S. glaucescens* as the biosorbent, optimum conditions of pH, Hg^{2+} concentration and equilibrium time were at 5, 200 ppb and 90 min, and when *G. corticata* was used, these conditions were at 7, 1000 ppb and 30 min. The equilibrium adsorption data are fitted to the Frundlich isotherm model, by both *S. glaucescens* and *G. corticata*. The Hg^{2+} uptake by *S. glaucescens* was best described by the first-order rate model and *G. corticata* was best described by the second-order model.

1. Introduction

The presence of toxic heavy metals in water resulting from rapid industrialization and technological advances is a worldwide environmental problem. Removal of these pollutants from aqueous effluents has conventionally been accomplished through biotic processes (Padilla and Tavani, 1995; Yabe and De Oliveira, 2003; Oubagaranadin et al., 2007). However, these processes can be expensive and not fully effective. Recently, an increasing interest in the application of materials of biological origin in heavy metals' removal from diluted, large volume solutions has been observed (Regime et al., 2000; Mehta and Gaur, 2005). Biosorption, which uses the ability of biological materials to remove and accumulate heavy metals from aqueous solutions, has received considerable attention in recent years because of a few advantages compared with traditional methods (Karthikeyan et al., 2007).

Surface adsorption is found to be an important basis for the treatment of toxic Ni^{2+} contaminated water (Goswami and

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Peer review under responsibility of King Saud University.

Production and hosting by Elsevier

http://dx.doi.org/10.1016/j.arabjc.2012.01.008

Ghosh, 2005). The presence of carboxylic and sulfate groups in algae cell wall polysaccharides can act as binding sites for metals. Alginate, which is composed of mannuronic and guluronic acids, is a major polysaccharide in brown algae and acts as the source of carboxylic groups (Davis et al., 2003). The cost of decontamination of toxic metals from industrial effluents and wastewaters using ion exchange resins is exorbitant. Hence, the usage of indigenous biodegradable resources for treating hazardous waste being less expensive is relevant to regional emerging environmental biotechnology (Babel and Kurmiawan, 2003). Activated carbon has unquestionably been the most popular and widely used absorbent in wastewater treatment employments throughout the world. However, activated carbon remains a costly material since the higher the quality of activated carbon, the greater its cost. Therefore, searching for low-cost activated carbon and other absorbent materials for wastewater treatment is of great importance (Fourest and Roux, 1992; Abdelwahab et al., 2006; Kapoor et al., 1999).

Marine algae are biological resources which are available throughout the world. The use of algae, the batch removal of nickel(II) ions from aqueous solution under different experimental conditions using activated carbon prepared from red alga *Gracilaria* (Esmaeili and Ghasemi, 2009), biosorption of copper from wastewater by activated carbon preparation from alga *Sargassum* sp. (Esmaeili et al., 2010a), and the batch removal of toxic hexavelant chromium ions from wastewater and aqueous solution using two different activated carbon marine algae *Gracilaria* (red algae) and *Sargassum* sp. (brown algae) were all examined. Activated carbon was prepared from *Gracilaria* and *Sargassum* sp. (Esmaeili et al., 2010b). The use of algae, *Durvillae pototatorum* (Matheickla et al., 1997) *Ecklonia radiate* (Matheickla and Yu, 1996) *Ascophyllum nodosum* (Volesky, 1990) and *Saccharomyces crevisiae* (Ghorbani and Younesi, 2008) *Spirogyra* (Gupta and Shirivastava, 2001) for heavy metal removal has been reported. In this research, removal of mercury(II) from aqueous solutions by biosorption on the biomass of *Sargassum glaucescens* (brown algae) and *Gracilaria corticata* (red algae) was examined. The effects of contact time, initial concentration, pH and equilibrium isotherms on the process were studied.

2. Experimental

2.1. Preparation of biomass

G. corticata (red algae) and *S. glaucescens* (brown algae) were collected from the Persian Gulf on Queshm Island. Before use, these were washed several times with tap water to remove sand particles and salts. They were then sun dried for 6 days. Dry biomass was chopped, milled (size fraction of 0.5–1 mm) and then used for biosorption experiments (Esmaeili et al., 2008a, 2011).

2.2. Preparation of Hg^{2+} solution

Stock Hg^{2+} solution (800 mg/L) was prepared by dissolving 0.37 g of $Hg(CH_3COO)_2\ 3H_2O$ (Merck) in 250 mL of deionized water. Different concentrations of Hg^{2+} solution were prepared by dilution of deionized water. The initial pH of the sample solution was adjusted with 1 M HCl/1 M NaOH using a digital pH meter. All the adsorption experiments were carried out at room temperature (23 ± 2 °C).

2.3. Preparation of biomass

Red marine macroalgae *G. corticata* and brown algae *S. glaucescens* were used for the removal of Hg^{2+} from aqueous solution. They were collected from the Persian Gulf on Queshm Island, Iran and washed several times with tap water to remove the sand particles and dirt. Dry biomass (after 5 days) was ground to particle size.

2.4. Determination of the Hg^{2+} contents

Concentrations of Hg^{2+} in the solution before and after the equilibrium were determined using a Perkin Elmer Analyst 300 atomic absorption spectrometer equipped with a deuterium lamp as the background corrector and an air-acetylene burner, and controlled by an IBM personal computer. The hollow cathode lamp was operated at 15 mA, and the analytical wavelength was set at 324.8 nm (Welz and Sperling, 2007).

2.5. Adsorption experiments

A series of flasks containing Hg^{2+} solutions varying in concentration from 200, 400, 600, 800, 1000 ppb of biomass for *G. corticata* and *S. glaucescens* were prepared from the stock solution. Adjustment of pH was carried out using 1 N NaOH and 1 N H_2SO_4. Batch equilibrium sorption experiments were carried out in Erlenmeyer flasks for *G. corticata* and *S. glaucescens* for 90 (200 ppb of biomass of metal solution) and 30 min (200 ppb of biomass, of metal solution) respectively, in a rotary shaker. These experiments were done at pH 1.0–13.0. After the sorption equilibrium was reached (90 and 30 min), the solution was separated from the biomass by membrane filtration. The initial and equilibrium Hg^{2+} concentrations in each flask were determined by atomic absorption spectrometry (AAS). To maximize Hg^{2+} removal by the absorbent, batch experiments were conducted at a constant temperature using the optimum conditions of all pertinent factors, such as dose, pH, initial concentration, and contact time. Subsequent adsorption experiments were carried out using only the optimized parameters.

2.6. Equilibrium isotherm

2.6.1. Lead uptake capacities and sorption isotherm

The amount of metal adsorbed by activated carbon was calculated from the difference between the metal quantity added to the biomass and the metal content of the supernatant using the equation:

$$q_e = (C_0 - C_e)V/M \tag{1}$$

where q_e is the metal uptake (mg metal adsorbed per g absorbent), C_0 and C_e are the initial and equilibrium metal concentrations in solution (mg/L), V is the volume of the solution (mL), and M is the weight of algae marine (g). To examine adsorbed and aqueous concentrations at equilibrium, sorption isotherm models and Langmuir and Freundlich adsorption models were used for the data. These isotherm constants for Hg^{2+} are presented in Table 1.

Table 1 Isotherm parameters obtained for biosorption of Hg^{2+}.

Isotherm model	*S. glaucescens*	*G. corticata*
Langmuir		
Q_m (mg/g)	147.05	−4.71
b (L/mg)	4.4×10^{-3}	-9.12×10^{-3}
R^2	0.137	0.4386
Freundlich		
$1/n$	0.9111	3.5039
K_f (mg/g)(L/mg)$^{1/n}$	0.77	3.33×10^{-6}
R^2	0.935	0.7371

2.6.2. Freundlich adsorption model

The Freundlich (1906) model habitually gives a better fit for adsorption from liquids and can be expressed as

$$q_e = K_f C_e^{1/n} \tag{2}$$

In this model, the rate of adsorption is of the constants $1/n$ and K_f (L/g). For a good absorbent, $0.2 < 1/n < 0.8$, and a smaller value of $1/n$ shows better adsorption and formation of a rather strong bond between the adsorbate and the absorbent. Many researchers have used this model to interpret this sorption data for various systems (Padmesh et al., 2006; Kundu and Gupta, 2006; Golder et al., 2006; Ho and McKay, 1999).

2.6.3. Langmuir adsorption model

The most widely used isotherm equation for modeling equilibrium is the Langmuir equation. It is assumed that there is a finite number of binding sites that are homogeneously shared over the absorbent surface. These binding sites have the same absorption of a single molecular layer, and there is no interaction between absorbed molecules. The saturated monolayer isotherm can be represented as

$$q_e = bq_{\max} C_e/(1 + \mathrm{b}C_e) \tag{3}$$

where q_e is metal ions adsorbed (mg/g), C_e is the equilibrium concentration (mg/L), $q_{\max}$ is the maximum adsorption capacity, and b is an affinity constant; $q_{\max}$ represents a practical limiting adsorption capacity when the surface is fully covered with metal ions and assists in the comparison of adsorption performance, in cases where the sorbent did not reach its full saturation in experiments and b is constantly related to the affinity of the binding sites (Langmuir, 1916; Doenmez and Aksu, 2002).

2.7. Adsorption kinetic studies

The kinetics of adsorption describes the rate of lead ion uptake by prepared *G. corticata* and *S. glaucescens*, which control the equilibrium time. These kinetic models included the first-order and second-order equations.

2.7.1. Pseudo first-order model

The pseudo first-order rate expression is described by the equation:

$$dq_t/d_t = k_1(q_e - q_t) \tag{4}$$

where q_e is lead adsorbed at equilibrium per unit weight of sorbent (mg/g), q_t is Hg^{2+} adsorbed (mg/g), and k_1 is the rate constant (min^{-1}) (Doenmez and Aksu, 2002). The integrated form of Eq. (4) becomes

$$\log(q_e - q_t) = \log(q_e) - (k_{1/2.303})t \tag{5}$$

A plot of $\log(q_e - q_t)$ versus (t) indicates a straight line of slope ($k_{1/2.303}$) and an intercept of $\log(q_e)$.

2.7.2. Pseudo second-order model

The sorption data also analyzed a pseudo second order (Ag and Aktay, 2002), given by

$$dq_t/d_t = K_2(q_e - q_t)^2 \tag{6}$$

where K_2 is the equilibrium rate constant (g/mg min), and q_e and q_t are the sorption capacities at equilibrium and at time. The integrated form of Eq. (5) becomes

$$1/(q_e - q_t) = 1/(q_e) + K_2 t \tag{7}$$

This has the linear form:

$$t/q_t = 1/K_2 q_e^2 + (1/q_e)t \tag{8}$$

A plot t/q versus t shows a straight line of slope ($1/q_e$) and an intercept of ($1/K_2 q_e^2$) (Ag and Aktay, 2002). In this study, the batch method was used for the biosorption of Hg^{2+} by *G. corticata* and *S. glaucescens*.

3. Results and discussion

The present study shows the biosorption of Hg^{2+} ions onto two kinds of modified marine algae, *G. corticata* (red algae) and *S. glaucescens* (brown algae) biomass in aqueous solutions. The data obtained from this work supports the view that *S. glaucescens* is an effective and low-cost absorbent for the removal of Hg^{2+} from aqueous solutions. The adsorption of metal ions is dependent on *S. glaucescens*, concentration of metal ions, retention time, and the pH of the metal solution.

3.1. Effect of contact time

Fig. 1 indicates that the removal ability increased with increasing contact time and concentrated heavy metals before the equilibrium was reached. Other limits such as dose of absorbent, pH of solution and, initial concentration were kept opti-

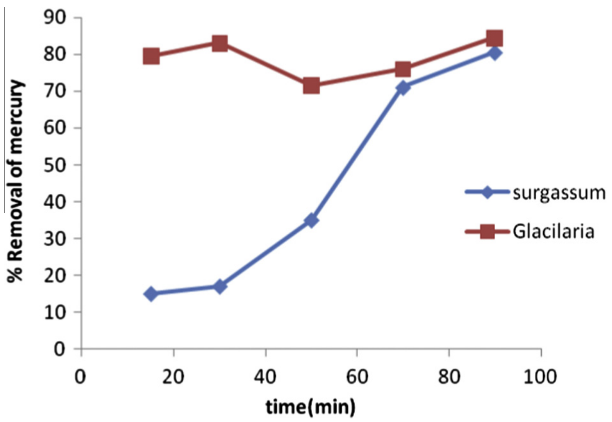

Figure 1 Biosorption of 100 ml Hg^{2+} solution, 100 and 25 mg/L, by *S. glaucescens* and *G. corticata*, 200 rpm shaking rate in different times.

mum. To find the suitable equilibrium time of the biosorption, the experiment was carried out at different time intervals (15, 30, 50, 70 and 90 min). As can be seen in Fig. 1 with the beginning of the process, the uptake of Hg^{2+} ions increased quickly and after 30 min for the 200 ppb and 90 min for the 200 ppb of Hg^{2+} solution, the exchange ion became slow, for *S. glaucescens* and *G. corticata*, respectively. It seems that *G. corticata* is more effective at removing high concentrations of lead solution.

The optimum contact time for both dried and activated carbon biosorbent was found to be 120 min. In previous literature, similar findings had been reported by the evaluation of the marine alga *G. corticata* for the adsorption of Cu(II) from wastewater in a packed column (Esmaeili et al., 2008b).

3.2. Effect of initial concentration

The initial concentration of metal ion provides an important driving force to overcome all mass transfer resistances of metal ions between the aqueous and solid phases (Malkoc, 2007). Fig. 2 represented the removal percent which was increased by using *G. corticata* and the maximum removal of Hg^{2+} ions, after 30 min was 1000 ppb in the range of 90.0%. As shown in Fig. 2, removal of Hg^{2+} ions by *S. glaucescens* was decreased by increasing the metal concentration and maximum removal after 90 min was 200 ppb in the range of 94.5%. Thus *S. glaucescens* is useful in removing high concentrations of Hg^{2+}. The initial concentration of metal ions provides an important driving force to overcome all mass transfer resistances of metal ions between the aqueous and solid phases (Esmaeili and Ghasemi, 2009). The influence of metal ion concentration on biosorption by *S. glaucescens* and *G. corticata* has been shown in Fig. 2.

Thus *S. glaucescens* is useful to remove high concentrations of Hg^{2+}. In recent studies, biosorption of Ni(II) with 30, 50 and 70 mg/L, by *G. corticata*, after 60 min was in the ranges of 99.03%, 97.99% and 95.12% (Esmaeili and Ghasemi, 2009). *G. corticata* was used for the biosorption of 180 mg/L Cu(II) solution in a packed column in the range of 80% (Esmaeili et al., 2008b).

3.3. Effect of pH on adsorption

When *S. glaucescens* has been used for Hg^{2+} ion biosorption a significant difference was not observed in the removal percent, when changing the pH from 3 to 13. Fig. 3 shows that the biosorption of Hg^{2+} was increased for both *S. glaucescens* and *G. corticata*, when removal occurred in acidic conditions: pH 5 and 7 in the range of 92.5% and 97.3%, respectively. The lower uptake at higher pH value is probably due to the formation of anionic hydroxide complexes. Due to these lower uptakes at higher pH values the ligands such as carboxylate and sulfonate groups could uptake fewer metal ions (Kapoor et al., 1999). The initial pH of the metal solution is an important parameter affecting adsorption of metal ions (Matheickla and Yu, 1996).

Actually, in basic conditions, the groups of biosorbent could uptake fewer Hg^{2+} ions. Gupta and Shirivastava (2001) obtained maximum biosorption of chromium (VI) by green algae *Spirogyra* species in optimum pH of 2.

3.4. Effect of adsorbent dose on adsorption

The initial concentration of metal ion provides an important driving force to overcome all mass transfer resistances of metal ions between the aqueous and solid phases (Matheickla and Yu, 1996). The results show that the equilibrium concentration of Hg^{2+} increased with an increasing adsorbate concentration. It showed that the greatest adsorption was achieved using *G. corticata* and the maximum removal of Hg^{2+} ions, after 30 min was 1000 ppb in the range of 90.0%. As shown, removal of Hg^{2+} ions by *S. glaucescens* was decreased, by increasing the metal concentration and maximum removal after 90 min was 200 ppb in the range of 94.5%. This was due to the saturation of the sorption sites on absorbents (Nuhoglu and Oguz, 2003; Malkon and Nuhoglu, 2003). Equilibrium between absorbent and absorbate is described by adsorption isotherms; usually the ratio between the quantity adsorbed and that remaining in the solution at a fixed temperature (23 ± 2 °C), at equilibrium. To study the absorption isotherm, two models were analyzed.

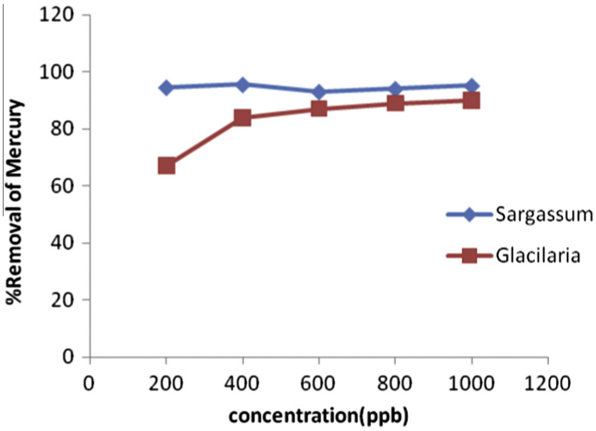

Figure 2 Biosorption of 100 ml Hg^{2+} solution at different metal concentrations, 200 rpm shaking rate by *S. glaucescens* during 90 min and *G. corticata* during 30 min.

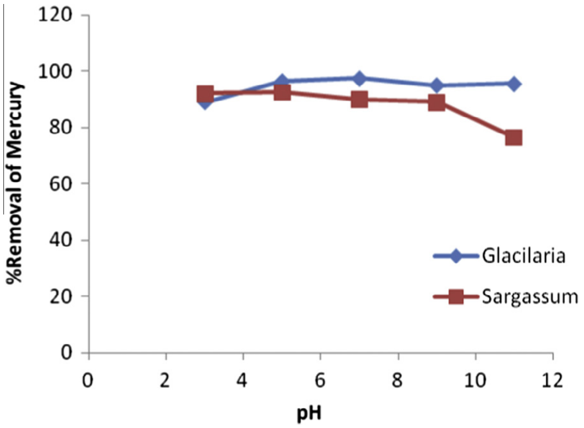

Figure 3 Biosorption of 100 ml Hg^{2+} solution at different initial pH, 200 rpm shaking rate Hg^{2+} solution by *S. glaucescens* in 90 min 1000 ppb by *G. corticata* in 50 min.

3.4.1. Langmuir and Freundlich isotherms

The Langmuir adsorption isotherm is the most widely applied absorption isotherm. A basic assumption of the Langmuir theory is that adsorption takes place at specific homogeneous sites within the absorbent. The applicability of the empirical Freundlich isotherm was also analyzed based on the adsorption on a heterogeneous surface, using the same set of experimental data of algae. The isotherm experimental results showed the data could be well modeled according to the Langmuir adsorption isotherm. The Langmuir constant ($q_{\max}$) is dependent on experimental conditions such as solution pH. Another importance in evaluating absorbent performance is the initial gradient of the absorption isotherm, since it indicates the absorbent affinity at low metal concentrations. In the Langmuir equation, this initial gradient corresponds to the affinity constant (*b*). As we can see from Figs. 4a and b data could be well modeled according to the Langmuir, absorption isotherm.

As we can see from Table 1 data could be well modeled according to either the Langmuir or Freundlich adsorption isotherm. In previous investigations, a similar method using the marine algae *Gracilaria* for the biosorption of Ni(II) had been reported (Esmaeili and Ghasemi, 2009).

3.5. Kinetic modeling

The second and first-order rate constants ($k_{1,\mathrm{ads}}$) and q_e determined from the model indicated that this model had failed to estimate q_e since the experimental values of q_e differed from those estimated in Table 2 and Fig. 5. Subsequently, the rate of uptake of Hg^{2+}onto the biomass increased quickly to 70 and 90 min, and no further adsorption was observed beyond this period. Using these equations by these two biosorbents in this study have been shown in Table 2 and Figs. 5a and b. As can be seen adsorption of Hg^{2+} by *G. corticata* (red algae) and *S. glaucescens* (brown algae) can be described by the first-order and second-order kinetic models, respectively.

In the previous literature for the evaluation of the activated carbon prepared from the algae *Gracilaria* for the adsorption of Cu(II), it had been observed that the adsorption follows

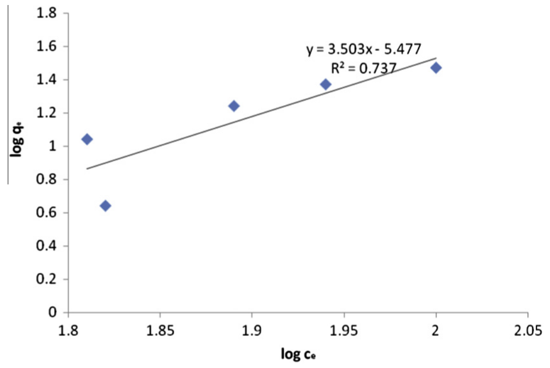

Figure 4a Isotherm models biosorption by *G. corticata*, Frundlich model.

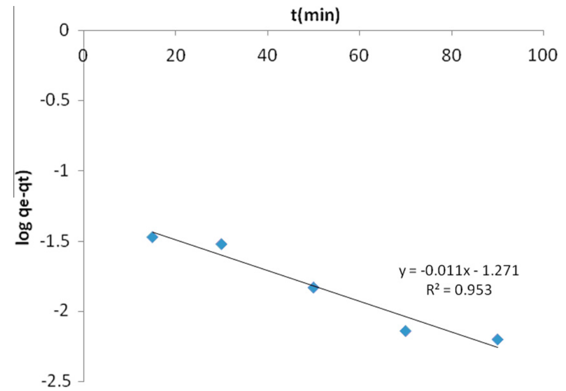

Figure 5a Kinetic modeling biosorption by *S. glaucescens*, first-order kinetic model.

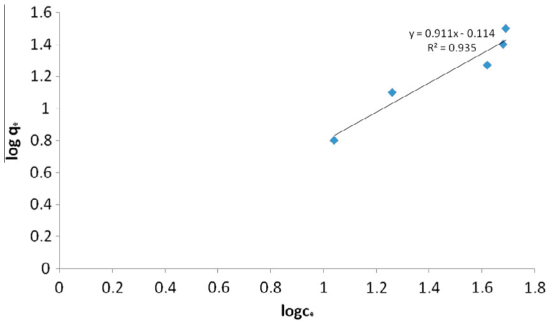

Figure 4b Isotherm models biosorption by *S. glaucescens*, Frundlich model.

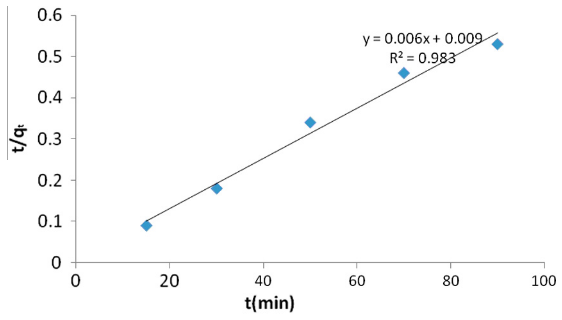

Figure 5b Kinetic modeling biosorption by *G. corticata*, second-order kinetic model.

Table 2 Parameters obtained for first- and second-order kinetic models by two biosorbents.

Algae	C_0, pb^{2+}	K_1	q_e (first order)	R^2	K_2	q_e (second order)	R^2
S. glaucescens	200	0.0253	0.535	0.9535	6.13×10^{-6}	−476.19	0.1172
G. corticata	200	0.07×10^{-3}	30.49	0.3701	3.95×10^{-3}	163.93	0.9832

the second-order kinetics reported (Malkon and Nuhoglu, 2003).

These isotherm constants for Hg^{2+} are presented in Table 2. The rate of uptake of Hg^{2+} for *G. corticata* and the maximum removal of Hg^{2+} ions, after 30 min was 1000 ppb. For *S. glaucescens* the rate was decreased, by increasing metal concentration and maximum removal was achieved after 90 min. In previous literature for the evaluation of the activated carbon prepared from the algae *Gracilaria* for the adsorption of Cu(II) the adsorption follows the second-order rate expression (Esmaeili et al., 2008b).

3.6. Adsorption equilibrium

The isotherm experimental results showed that the data could be well modeled according to the Freundlich adsorption isotherms (Figs. 4a and b). The Langmuir constant ($q_{\max}$) is dependent on experimental conditions such as solution pH. Another important factor in evaluating sorbent performance is the initial gradient of the absorption isotherm, since it indicates the sorbent affinity at low metal concentrations. In the Langmuir equation, this initial gradient corresponds to the affinity constant. The Freundlich and Langmuir isotherm constants were obtained and are presented in Table 1.

4. Conclusion

In this study, removal of mercury(II) from aqueous solutions by biosorption on the biomass of *S. glaucescens* (brown algae) and *G. corticata* (red algae) was examined. These treated biomass could be used as low-cost absorbents for the removal of Hg^{2+} from aqueous solutions. The adsorption of metal ions is dependent on the concentration of Hg^{2+}, pH and retention time. Absorption rate would increase by increasing the time to 90 and 30 min by *S. glaucescens* and *G. corticata*, respectively. Optimum Hg^{2+} uptake was obtained at pH = 5 and pH = 7 when we used *S. glaucescens* and *G. corticata*, respectively. Results showed that *S. glaucescens* is more effective for removing Hg^{2+}, in optimum conditions of the process in the range of 95.1%. *G. corticata* has adsorbed Hg^{2+}, in optimum conditions, in the range of 90.0%. The equilibrium adsorption data are correlated by Frundlich isotherm equation for both *S. glaucescens* and *G. corticata* and they can be described by first order for *S. glaucescens* and second order for *G. corticata*.

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