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Sorption of chromium from aqueous solutions using *Fucus vesiculosus* algae biosorbent

Amany A. Asaad^{1*}

Abstract

The presence of heavy metals in wastewater is an environmental concern and the current treatment procedures are very expensive so it is necessary to find effective and inexpensive biosorbents. In this study, *Fucus vesiculosus* was used as a biosorbent for the biosorption of Cr(III) ions from the aqueous solutions. Biosorption parameters, such as pH, adsorbent dose, contact time, and initial concentrations of Cr(III) had the most impact on the sorption process. The required pH value for sorption was 5, the biosorbent dose was 4.0 g/L, the contact time was seen to occur after 90 min, and the Cr(III) removal decreased from 98.9 to 92%. The maximum biosorption capacity of chromium was 14.12 mg/g. FTIR analysis of *Fucus vesiculosus* biomass before the sorption process, it was found that Cr(III) metal ions were incorporated within the sorbent during the interaction with (=C–H) active functional groups. The biosorp-tion data were found to be perfectly suited by Langmuir equilibrium isotherm model. According to the results of this study, *Fucus vesiculosus* is an effective biosorbent for the removal of Cr(III) from aqueous solutions.

Keywords Fucus vesiclosus, Biosorption, Heavy metals, Chromium, Water treatment

Introduction

Heavy metals are toxic and carcinogenic, and cannot be biodegraded. Heavy metals such as zinc, copper, nickel, mercury, cadmium, lead, chromium, and arsenic have a tendency to build up in living organisms and cause a decrease in species diversity [1-3]. Metal contamination is a global environmental problem that persists and should be addressed with sufficient measures to prevent its exposure to the public. Heavy metals deposited because of industrial processes should be removed before they are received into the water since they are particularly baleful to aquatic habitats. Among these metals,

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¹ Central Laboratory for Environmental Quality Monitoring, National Water Research Center, El-Qanater-Qalubeya 13621, Egypt chromium is one of the worthy environmental troubles that continue to cause contamination of aqueous systems and it is present in various oxidation states such as Cr(III) and Cr(VI). Chromium is used in several industries such as iron, steel, leather, metal coating, textile industry, electric power plants, coil coating, electroplating, film, photography, galvanometer, and automotive battery manufacturing industries [4-7]. The disposal of this commonly used metal in the environment causes critical pollution [8]. Moreover, searching for an important approach to remove such contaminants is an indispensable task for researchers. In this regard, various biological, physical, and chemical methods have been adopted for eliminating such heavy metals from industrial effluents such as chemical precipitation, ion exchange, and membrane separation techniques. It is preferable to use biological materials (sorbents) as an alternative method for removing chromium from aqueous solutions because the



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commonly used procedures for removing Cr(III) from effluents include chemical precipitation, lime coagulation, ion exchange, reverse osmosis and solvent extraction were apart from being economically expensive have disadvantages like incomplete metal removal, high reagent and energy requirements, and generation of toxic sludge or other waste products that require disposal. Efficient and environment friendly methods are thus needed to be developed to reduce heavy metal content. In this context, considerable attention has been focused in recent years upon the field of biosorption for the removal of heavy metal ions from aqueous effluents. Biosorption is a property of certain types of inactive, non-living microbial biomass to bind and concentrate heavy metals from even very dilute aqueous solution. Biomass exhibits this property, acting just as chemical substance, as an ion exchanger of biological origin [9]. Due to some benefits over conventional methods, the use of sorption materials in the removal and accumulation of heavy metals from aqueous solutions has recently received a lot of attention.

Several adsorbents have been used to remove Cr(VI) from water over the last few years, including commercial inorganic materials such as clay, silica gel, zeolite, alumina, and activated carbon, as well as bio products [10, 11] which may be alive or dead, and their effectiveness is determined by their loading capacity, selectivity, affinity, and rate of ion adsorption [12]. Lignocelluloses materials, in general, are piquing researchers' interest due to their simple design, ease of handling, cheap operating costs, ease of availability, eco-friendliness, efficiency, and production of minimal toxic chemicals and biological sludge [13]. Furthermore, these materials are abundant in poly functional groups, which can contribute significantly to the selective adsorption of Cr(VI) from aqueous solutions [14]. Another notable property of biosorbents is their ability to convert Cr(VI) to Cr(III) at lower pH values and to totally remove Cr(VI) at moderate concentrations [15]. The removal ability of the biosorbents is affected by parameters such as pH, adsorbent dose, size, concentration, and contact time during the process.

Various studies indicate that non-living sorbents are more effective for binding metals than biological sorbents [7]. Marine algae are known to possess excellent mineral binding capacity in different bio-selective procedures. The cell membranes of brown algae are usually composed of cellulose and algal acid, which is a straight-chain polysaccharide with a carboxyl group (–COOH) primarily responsible for binding to minerals, while sulfated polysaccharide algae bind to ion salts.. Because of these properties, algae (sorbents) are a good choice for adsorbing metal ions from the aqueous solutions in a short period and reducing heavy metal concentricity to the ppb range [16]. The sorption mechanism of heavy metals on biosorbents is thought to involve one or more of these: ion exchange, biosorption, complexation, partial precipitation formation, chelation, and electrostatic interaction. However, the most significant way that heavy metal ions are adsorbed by algae is through the ion exchange process [17]. The presence of sulfate groups, as well as a large number of carboxylic groups in brown marine algae, has been attributed to the biosorption of trivalent metal cations [18]. The characterization of the biosorbent structure and exploration of the reaction mechanism of sorbate ions and biosorbents can be identified with the help of FTIR (Fourier transform infrared) spectroscopy and SEM (A scanning electron microscope coupled with an energy dispersive spectrometer) is important to determine the structure of the *Fucus vesiculosus* [19, 20]. The reports on this kind of biosorbents and their utilization in eliminating heavy metals are few [21].

This paper investigates the biosorption capacity of *Fucus vesiculosus* (algae known by different common names such as bladder wrack, black tang, rockweed, sea grapes, sea oak, cut weed, and rock wrack) and the chromium affinity toward it. To determine the best conditions for biosorption, the effects of pH, equilibrium time, and initial concentrations were examined. FTIR spectroscopy is used to determine the specific functions involved in the association of chromium with this type of algae.

Experimental methods

Preparation and analysis of biomass

Sun-dried *Fucus vesiculosus* brown algae were brought from a local market in Cairo and then dried and grounded by an electrical grinder to conduct the biosorption procedures. FTIR spectroscopy was used to determine the effective groups that chromium can occupy before and after the biosorption. The spectra ranges were $600-4000 \text{ cm}^{-1}$ using "Thermo Fisher Nicolet 50 spectroscopy" [22]. A scanning electron microscope (SEM) is one of the common methods for imaging the microstructure and morphology of the materials [23].

Preparation of metal solution

The preparation of 20 mg/L Cr(III) solution was carried out by diluting the working standard solution 1000 mg/L (CrCl₂ Merck) to different concentrations from 10 to 50 mg/L. The pH values of those prepared solutions were adjusted using 1N NaOH and/or 1N HCl.

Biosorption experiment

Biosorption of chromium by (*Fucus*) was performed by contacting 4g/L of *Fucus* with chromium concentration(20 mg/L) in a 1000 cm³ Pyrex conical flask intermittently for 90 min on the stirrer at 300 rpm. The mixture was filtered, and the residual concentration of the filtrate was analyzed

using Inductively coupled plasma optical emission spectrometry (ICP-OES) which is well suited for such analysis because it is precise for lower concentrations [24]. The adsorbed amount of chromium (mg/g) was calculated using the following formula:

$$q_e = \frac{(C_o - C_e)V}{M},\tag{1}$$

where the equilibrium biosorption capacity of *Fucus* for chromium is denoted by q_e (mg/g), the weight of the biosorbent is symbolized by M(g), and the sorbate volume is symbolized by V(L). C_o and C_e represent the metal concentration before and after sorption (mg/L). Hence the chromium uptake ratio can be evaluated by the next equation:

$$R\% = \frac{\mathbf{Co} - \mathbf{Ce}}{\mathbf{Co}} \times 100. \tag{2}$$

Effects of operational parameters

The determination of the optimal adsorption parameters such as pH of the biosorption solution, dose of biosorbent, biosorption time, and concentration of the adsorbate solution were essential in knowing the biosorption efficiency of the biosorbent under equilibrium condition [25]. The optimal effective adsorption parameters determined when equilibrium occurs can be achieved by preparing a series of a series of chromium solutions with pH values from 2 to 7 at a concentration of 20 mg/L, a shaking speed of 300 rpm, and a doses of (0.5, 0.1, 0.2 and 0.3) g of the adsorbent (Fucus vesiculosus) at 25 °C. The pH adjustment was performed using 1 N NaOH and/or 1 N HCl solutions. To establish the optimal contact time for biosorption studies, 0.2 g of Fucus vesiculosus powder was added to 50 mL of chromium solution at a concentration of 20 mg/L for 10-120 min at 25 °C. After the sorption process, the samples were filtered off through 0.45 µm membrane filter paper. Also the effect of biosorbent dose was investigated in the 0.05–0.3 g range. This was performed by adding a specific dose of 50 mL of chromium solution (20 mg/L) and shaking it for 90 min. After that, the sorption capacity of Fucus vesiculosus was calculated using the aforementioned equations. The impact of chromium initial concentration on the biosorption capacity of the biosorbent was studied by utilizing 0.2 g of Fucus powder and various concentrations of chromium solution (10, 20, 30, 40, and 50 mg/L) for 90 min at 25 °C and pH 5.

Biosorption isotherm models

Biosorption isotherms were employed to determine the biosorption behavior of the biosorbent and to provide a connection between the sorbate concentration C_e and the biosorption capacity q_e per mass unit of the biosorbent at equilibrium. Langmuir isotherm shows that the biosorption occurs on a homogeneous monolayer containing large biosorption sites [26]. The linear form of the Langmuir equation is presented as the following:

$$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m},\tag{3}$$

where q_e is the adsorption capacity at equilibrium, C_e represents the equilibrium concentrations of chromium, q_m is the maximum adsorption capacity at equilibrium and K_L is Langmuir constant which indicates the adsorption energy. The basic characteristics of the Langmuir isotherm can be described in terms of dimensionless factor R_L , which is assumed by:

$$R_L = \frac{1}{(1+bC_o)} \tag{4}$$

 C_o is the initial concentration of adsorbate and R_L explains the adsorption preference of this isotherm and indicates whether the adsorption is irreversible if $R_L = 0$, linear if $R_L = 1$, or unfavorable if $R_L > 1$.

Freundlich isotherm postulates that biosorption occurs at the available locations on heterogeneous surfaces [27]. The correlation factor R^2 is used to evaluate the applicability of an isothermal model. The known logarithmic form of the Freundlich model is presented in Eq. (5).

$$\ln q_e = \ln q K_F + \frac{1}{n} \ln C_e, \tag{5}$$

where q_e and C_e are the capacity of biosorption (mg/g) and the concentration of sorbate (mg/L) at equilibrium, 1/n is related to the intensity of biosorption, K_F , and n are constants.

Temkin model postulates the interactions of adsorbent-sorbate. It exhibits that the heat of an adsorbed substance is reduced linearly than logarithmically [28]. This model is characterized by a uniform binding energy distribution up to maximum binding energy and it is implemented by plotting q_e against lnC_e and then the constants can be calculated from their slope and intercept.

$$q_e = \beta_{\mathsf{T}} \ln \mathsf{K}_{\mathsf{T}} + \beta_{\mathsf{T}} \ln \mathsf{C}_{\mathsf{e}},\tag{6}$$

where q_e denotes the quantity of adsorbed molecules that reach a state of equilibrium (mg/g); C_o is related to the concentration of the metal (mg/L). β constant is linked to the heat of biosorption, while R is the gas constant (8.314 J/mol K), and K represents the Temkin isotherm constant (L/g) [29].

Biosorption kinetic models

Normally, the simulation of biosorption kinetics and evaluation of the reaction rates involve the utilization of the pseudo-first-order, pseudo-second-order, and Elovich kinetic models. The pseudo-first-order kinetic model clarifies the correlation between the adsorbent sorption sites that are occupied and the number of unoccupied sites but The relation between the adsorption capacity of the adsorbent and the time established by the pseudo-secondorder kinetic model [30]. Equations (7) and (8) provide the mathematical expressions for the pseudo-first-order and pseudo-second-order, respectively [31].

$$log(q_e - q_t) = logq_e - \left(\frac{K_1}{2.303}\right),$$
 (7)

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

where q_e is the amount of chromium adsorbed onto adsorbent at equilibrium in (mg/g), q_t is the amount chromium adsorbed onto adsorbent at any time in (mg/g), and K_1 is the kinetics rate constant of the pseudofirst-order model (min⁻¹). K_2 is the kinetics rate constant of the pseudo-second-order model (g mg⁻¹ min⁻¹).

Elovich model is utilized to explain the kinetics of chemical biosorption of gas onto solid adsorbents, but it has been proven to be effective in describing various types of biosorption [32]. The following equation illustrates the Elovich model:

$$q_t = \frac{1}{b}\ln(ab) + \frac{1}{b}\ln t,\tag{9}$$

where $q_t (mg/g)$ is the adsorbate quantity at time t, a is a chemisorption rate constant and b is a constant that represents the amplitude of surface coverage and they can be calculated from the relation between their slope and intercept by plotting q_t versus lnt. a (mg/g min⁻¹) represents the initial rate of sorption, and b (g/mg) represents the desorption constant.

Results and discussion

SEM analysis

The surface morphology and initial formation of this species of algae have been found to have rough surfaces with pores of various sizes and shapes, increasing the surface area for metal ions to interact as shown in Fig. 1.

FT-IR analysis

Fucus vesiculosus dried biomass before and after the sorption of chromium was analyzed using Fourier transform infrared (FTIR) spectroscopy to identify how metal ions and surface biomass interact. Fucus vesiculosus algae contain polysaccharides that include many negative charges and functional groups that can interact with chromium, and these functional groups include carboxylate, hydroxyl, amino, and nitro groups [33]. The spectra of adsorbents before and after chromium uptake were measured from 600 to 4000 cm⁻¹ wavenumber [34]. The spectra of *Fucus* vesiculosus before the biosorption process showed different absorption bands at 3280, 2922, 2318, 1259, and1080 cm⁻¹ were shifted to 3287, 2944, 1625, 1220, and 1029 cm⁻¹, respectively, after biosorption of chromium. This result indicated chemical bonding among binding sites on Fucus biomass and the chromium [35]. The sorption bands of the sorbent at 1535 and



Fig. 1 Scanning electron micrograph of dried Fucus vesiculosus brown algae

1416 cm⁻¹ remain unchanged after the sorption process while those at 1013 and 872 cm⁻¹ disappeared (Fig. 2a, b). The vibrational bands in the pure biomass of Fucus vesiculosus before chromium sorption at 3280 cm^{-1} and 2922 cm^{-1} are assigned to (C–H stretching) alkyne and alkene groups [36]. The vibrational band at 2318 cm⁻¹ is related to carbon dioxide (O=C=O). The band at 1535 cm⁻¹ is due to N-O functional group [37]. The sharp band at 1416 cm^{-1} is probably due to the bending vibration of the hydroxyl group (O-H) [38]. The vibrational band at 1259 cm^{-1} is restricted to (C–O) stretching [39]. The band at 1080 cm^{-1} relates to the (C-N) stretching mode [40]. The bands between the wavenumbers of $1800-750 \text{ cm}^{-1}$ (fingerprint regions) reflected the biochemical compositions, especially the moieties of carbohydrate, lipid, protein secondary and polyphenols [41] The band at 860 cm^{-1} is due to =C-H bending disappeared in the FTIR spectrum for the biomass sample of Fucus vesiculosus after the chromium sorption process [42]. Figure 2b demonstrated that the chromium was incorporated within the sorbent during the interaction with the active functional groups (=C-H) [43].

Biosorption studies

The pH of the contact solution is an important parameter controlling the biosorption process. The variation of pH values changes the solution acidity or basicity and affects the *Fucus* surface charge. The pH of the initial chromium concentration (20 mg/L) varied from 2.0 to 9.0 at a constant dose (4 g/L) and the stirring rate at 300 rpm for 90 min at room temperature (25 °C), as shown in Fig. 3. The sorption of chromium was raised by increasing pH ranges from 2.0 to 5.0 where the capacity removal percentage of chromium reached 96.15%. It may be due to the active functional groups in the sorbent that facilitate biosorption by participating in metal ion binding [44]. This was followed by a gradual decrease in the chromium removal % at pH values greater than 5.0. The sorbent mass was then varied (0.05-0.3 g/50mL) for an initial Cr(III) concentration of 20 mg/L at 25 °C and pH 5.0 for 90 min as shown in Fig. 4. The sorption of Cr(III) increased with an increase in sorbent mass at an equilibrium time of 90 min from 46 to 96% and the equilibrium biosorption capacity reached 29 mg/g. This is because the higher the initial concentration of the metal ions, the higher the chance of collisions with adsorption sites on the surface of the adsorbent. Moreover, the driving force of mass



Fig. 2 a, b FT-IR spectrum of Fucus vesiculosus before and after Cr(III) sorption



Fig. 3 Effect of pH on Cr(III) sorption



Fig. 4 Effect of contact time on Cr(III) sorption

transfer is better, which is conducive to reduce the mass transfer resistance and increase the biosorption capacity [45]. Figure 5 shows the removal of Cr(III) which was accomplished within 90 min so there is no any additional sorption and an equilibrium state is reached. The rate of the biosorption process will increase significantly with increasing contact time so that it reaches the equilibrium point. Where, the longer the contact time, the greater the adsorption capacity. Figure 6 explains the effect of Cr(III) concentrations on the sorption process under study in which the higher uptake occurred at 90 min under equilibrium [46]. It may be imputed to the consumption of the available sites of the sorbent stable amount at equilibrium. Therefore, 90 min is the time required time for the sorption process. The sorption of Cr(III) declined from 10 mg/L to 50 mg/L and there was a significant rising in q_e of Cr(III) at the *Fucus* surface when the Cr(III) concentration ascents from 2.5 to 11.5 mg respectively.



Fig. 5 Effect of biosorbent mass on Cr(III) sorption



Fig. 6 Effect of initial concentration Cr(III) sorption

Hence the decrease in percentage removal from 98.9% to 92% may be attributed to the lack of other available sites of the sorbent and there is a repulsion force between the sorbate and bulk phase which reduces the uptake of the chromium [47, 48].

Biosorption isotherms

The biosorption isotherms are commonly used to reflect the performance of biosorbents in biosorption processes. Langmuir isotherm is useful for monolayer adsorption, the Freundlich isotherm shows adsorption on the heterogeneous surfaces of adsorbate-adsorbent systems and the Temkin isotherm model assumes that the adsorption energy of all molecules decreases linearly with increasing adsorbent surface occupancy. In this research, the biosorption isotherms were achieved for chromium solutions of different initial concentrations from 10 to 50 mg/L, an algae dose of 4 g/L at 300 rpm for 90 min, and

at pH 5 [49-51]. The concentration of adsorbed chromium was determined according to Eq. (3). Figure 7a-c represented the biosorption isotherms of Cr(III) by the Fucus surface at pH 5 using the Langmuir, Freundlich, and Temkin models, respectively. Isotherm parameters are reported in Table 1. The Langmuir constant values (K_1) show that strong interactions between metal ions and apparent functional groups are involved in the biosorption processes, regardless of the nature of metal ions or biosorbent. The separation factor in Langmuir isotherm (R_1) was less than one and the correlation factor (R^2) was 99%, which showed that the biosorption process was favorable [52]. The parameter 1/n in the Freundlich model is less than unity indicating that all biosorption processes are favourable. Moreover, the obtained values have better performance in biosorption of chromium(III) metal ions. These observations are also supported by the Temkin model parameters (Table 1), which show that in the biosorption process, the retention of metal ions is achieved through strong interactions, confirming the removal efficiency trend.

Biosorption kinetics

The kinetics of the Cr(III) biosorption process was evaluated using different kinetic models. Pseudo-first, secondorder kinetics, and Elovich models were applied as shown in (Fig. 8a–c) and the estimated kinetic parameters have

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Models	Parameters	Values
	q _{max}	17.06 mg/g
	KL	0.52 L/mg
Langmuir	R _L	0.16
	R ²	0.99
	K _f	5.39 mg/g
Freundlich	n	1.7
	R ²	0.98
	β_T	0.27
Temkin	K _T	1.97 L/g
	R ²	0.97

 Table 1
 Fitting parameters of isotherm models for the biosorption of Cr(III) on the Fucus vesiculosus biosorbent

been illustrated in Table 2. The appropriate kinetic model of Cr(III) biosorption was governed by the linear correlation coefficient (\mathbb{R}^2) values taken from model plots. The value of \mathbb{R}^2 in the pseudo-second-order kinetic model (0.991) was higher than the value of \mathbb{R}^2 in the pseudo-first-order (0.933), hence it may be attributed to chemically induced biosorption kinetics including valence strength via ion exchange or through the electron interactions between adsorbed molecules on the *Fucus* surface and the adsorbent [53, 54]. It has been achieved that the high correlation coefficient indicate



Fig. 7 a-c Langmuir, Freundlich and Temkin Isotherms for the sorption of Cr(III) on the Fucus vesiculosus



Fig. 8 a-c Pseudo first order, pseudo second order and Elovich kinetic models for the sorption of Cr(III) on the Fucus vesiculosus biosorbent

Table 2 Kinetic parameters for the biosorption of Cr(III) on the

 Fucus vesiculosus biosorbent

Models	Parameters	Values
	q _e	20.30 mg/g
Pseudo-Frist-Order	K ₁	0.0449 min ⁻¹
	R ²	0.93
	q _e	23.62 mg/g
Pseudo-Second-Order	K ₂	0.062 g/mg. min
	R ²	0.99
	q _t	14.80 mg/g
Elovich	а	4.63 mg/g min ⁻¹
	b	0.23 g/mg
	R^2	0.99

a good fit of experimental data to the pseudo-secondorder model [55]. Also, this indicated that the rate constant (K_2) of Cr(III) was 0.062 g/mg min, which reveals that the pseudo-second-order kinetic model is based on the assumption that the rate-limiting step is chemical sorption or chemisorption and predicts the behavior over the whole range of adsorption. In this condition, the adsorption rate is dependent on adsorption capacity not on concentration of adsorbate [56]. Moreover, in order to comprehend the characteristics of chemisorption, the Elovich model has been utilized. The amounts of (1/b) and (1/b) ln (ab) have been evaluated by the slope and intercept of the linear correlation [57, 58]. The value of (1/b) represents the number of available sites required for biosorption, while the biosorption quantity is indicated by the value of $(1/b) \ln$ (ab). Elovich model data has been demonstrated in (Table 2).

Conclusions

This study presents a new approach using Fucus vesiculosus algae to remove chromium from aqueous solutions in a safe and environmental manner. The maximum chromium removal capacity was 96.15% at pH 5, and dose (4.0 g/L) in 90 min. The biosorption models described the biosorption equilibrium of chromium with Fucus vesiculosus, the maximum biosorption capacity of chromium was 14.12 mg/g and the isothermal constants were determined. The obtained results confirmed that the biosorption equilibrium data are excellently integrated into the Langmuir model and also the pseudo-secondorder equation gave an excellent correlation between the experimental and the calculated data in the biosorption of Cr(III). Finally, it was concluded that Fucus vesiculosus is an effective and environmentally friendly biosorbent and a suitable candidate for the removal of Cr(III) from aqueous solutions.

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Author contributions

A.A. wrote the main manuscript text and prepared all figures, data collection, analysis, and interpretation of results, and reviewed the manuscript.

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Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare no competing interests.

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