



Removal of Cr(VI) from synthetic aqueous solutions by filamentous green algae *Spirogyra porticalis*

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Received 11 March 2015; Accepted 24 August 2015

ABSTRACT

Application of various biomaterials to toxic compounds and heavy metals removal from aqueous solutions and industrial wastewaters is an efficient and low-cost method, in this regard macro and microalgae have revealed high potential for the biosorption of such metals. In this work, removal of Cr(VI) from synthetic aqueous solutions by powdered filamentous green algae *Spirogyra porticalis* was studied as an effect of various parameters including pH, biomass dosage, initial Cr(VI) concentration, and contact time in batch experiments. In order to do the statistical analysis, repeated measure test using SPSS 16.0 software was applied and p -value < 0.05 was considered as significant level. The results showed that the removal of Cr(VI) increased with the increase in the algal dosage, metal concentration, contact time, and decreasing pH. The optimum pH, biomass dose, initial concentration of Cr(VI), and contact time were found to be 3.0, 1 g/L, 40 mg/L, and 60 min, respectively. Under these conditions, the removal percentage of chromium(VI) and the specific removal capacity were found to be 70% and 27.48 mg/g, respectively. The findings of the present study indicated that *S. porticalis* can be used as an available, affordable, and efficient biosorbent to Cr(VI) removal from aqueous solutions and industrial acidic wastewaters containing high concentrations of chromium.

Keywords: Biosorption; Chromium(VI); *Spirogyra porticalis*; Heavy metals

1. Introduction

Human industrial activities lead to production of hazardous environmental contaminants such as toxic organic compounds, heavy metals, and carcinogenic substances. Heavy metals are major pollutants in

nature due to their toxicity, non-degradability, and thus persistence in the environment. These metals may also be a threat to human health in high concentrations [1]. Chromium is widely used in chromate manufacturing, textile industries, electroplating, and leather tanning [2]. In aqueous solutions, chromium mostly exists in two oxidation states, the trivalent

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chromium and the hexavalent chromium [3]. Cr(VI) is unstable in the liquid phase due to high solubility and mobility as well as easy reduction [4]. Toxicity of hexavalent chromium is more than trivalent chromium; the Cr(VI) is carcinogenic and mutagenic to human as well as animals. Researchers found that chronic exposure to hexavalent chromium may cause an increased risk of cancer of the digestive tract and lungs. [2]. Cr(VI) can easily pass through membranes of cells and is easily absorbed, producing various toxic effects within cells due to formation of free radicals. It causes strong allergic reactions even in low concentrations (e.g. asthmatic bronchitis). Cr(VI) may also cause DNA damage [5,6]. Therefore, the World Health Organization (WHO) recommends that the permissible level of Cr(VI) in drinking water and total chromium discharged to wastewater should be 0.05 mg/L and below 2 mg/L, respectively [7]. Various methods of water and wastewater treatment may be used to remove the amount of Cr(VI) ions from aqueous solutions and industrial wastewaters. These methods include chemical precipitation, ion exchange, reverse osmosis, and other membrane processes [8–10]. Some disadvantages of aforementioned methods include: high energy requirement, incomplete metal removal, expensive, and generation of toxic sludge. Thus, it is necessary to find alternative techniques or adsorbents, which are efficient and affordable [11–13]. Biosorption is the process in which nonliving biomasses are applied to metal uptake from aqueous solutions by physico-chemical reactions such as electrostatic attraction [14,15]. In this process, metal ions form complexes with biomaterials using their ligands or functional groups and finally they are removed [16]. Biosorption has significant advantages over the conventional methods such as being used *in situ*, not producing chemical and toxic sludge, being more efficient, easy to operate, and biomass reusability [17,18]. Various types of biomass, including bacteria, fungi, yeasts, molds, viruses, and algae have been employed in the biosorption process [19,20]. Among them, algae have a higher adsorption capability due to the presence of polysaccharides, proteins, and lipids on the cell wall surface containing functional groups such as amino, hydroxyl, carboxyl, and sulfate, which can act as binding sites for metal ions [21]. Other features that make algae a desirable option for bioaccumulation and biosorption of heavy metals include availability in high quantities, high tolerance to toxicity of heavy metals, large surface area to volume ratio, and potential to genetic alterations [22,23]. Several researchers have reported the removal of heavy metals through macroalgae and microalgae from aqueous solutions. Kumar and Oommen [15] studied the removal of heavy metals by *Spirogyra*

hyalina as function of initial concentration of the heavy metals and contact time of the biomass, and showed high capacity of *S. hyalina* in removing mercury and lead from aqueous solutions. Biosorption of mercury (II), cadmium(II), and lead(II) ions from aqueous system by microalgae *Chlamydomonas reinhardtii* was investigated in alginate beads by Bayramoglu et al. [23]. Roy et al. [24] studied the adsorption of heavy metals by green algae. The removal of metal ions by freshwater algae with different surface characteristics was investigated by Tien [25].

Spirogyra porticalis is filamentous algae that can often form flimsy green aggregates during the warm months in freshwater ponds, rivers, and streams. The present work, focused on the investigation of the capability of nonliving green algae *S. porticalis* to remove Cr(VI) from synthetic aqueous solutions as an effect of pH, initial metal concentration, algal dosage and, contact time.

2. Materials and methods

2.1. Preparation of biosorbent

The filamentous green algae *S. porticalis* was sampled along the Ghohrood River, Kashan, Iran (during the spring season). Firstly, they were washed with tap water and then with distilled water to remove foreign particles and dirt. The washed biomass was first air dried for 12 h and thereafter in an oven at 70°C for 12 h. Finally, the dried biomass was powdered using domestic mixer.

2.2. Preparation of chromium solution

The stock chromium solution (500 mg/L) was made by dissolving 1.417 g of 99% potassium dichromate ($K_2Cr_2O_7$) in 1 L distilled water. Synthetic solutions of different concentrations of Cr(VI) were obtained by diluting the stock solution. In such a way that for preparing the solutions containing 10, 25, and 40 mg/L of Cr(VI) respectively 2, 5, and 8 mL of the stock solution were added to 100 mL of distilled water.

2.3. Experimental procedure

This study was conducted experimentally in a laboratory scale batch. The synthetic solutions were examined at room temperature in 250-mL flasks containing 100 mL of metal solution with concentrations of 10, 25, and 40 mg/L of Cr(VI) and different pH values (3.0, 7.0, and 11.0). The biosorbent was used in three doses: 0.2, 0.5, and 1 g/L. Sulfuric acid (0.5 M) and caustic soda (1.0 M) were utilized to adjust the pH value. The samples were mixed at various contact times of 30, 45, and 60 min on a rotary shaker (agita-

tion rate, 150 rpm). After mixing time, the adsorbent was separated from the solution through a filter paper (Whatman). To measure the concentration of Cr(VI), the colorimetric method with reagent of 1,5-diphenylcarbazide using visible-spectrophotometer (Model DR/2010, HACH, USA) at wavelength of 540 nm was applied [26]. In an acidic solution, 0.5 mL of 1,5-diphenylcarbazide was added to 25 mL of each filtered sample containing various concentrations of Cr(VI). The color intensity created in the solution that follows the Beer-Lamberts law represents the remaining concentration of hexavalent chromium. Each experiment was repeated three times. The repeated measure test and one-way analysis of variance was used for statistical analysis (using SPSS 16.0 software) and p -value <0.05 was considered as significant level.

To estimate the removal percentage of chromium (VI) from aqueous solution, the following equation was used:

$$\text{Removal percentage} = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

where C_0 = initial concentration of Cr(VI), mg/L and C_e = remaining concentration of Cr(VI) in solution, mg/L.

The specific removal capacity is given by the following equation:

$$q_e = (C_0 - C_e)V/1000 W \quad (2)$$

where q_e = amount of Cr(VI) bound by per unit weight of biomass (mg/g), V = volume of the aqueous solution (mL), and W = biomass weight (g).

3. Results

3.1. Effect of pH

The removal efficiency of Cr(VI) using *S. porticalis* was studied by pre-selected range of pH (Table 1). The results indicated that the Cr(VI) removal process was influenced significantly by variation of pH value

(Fig. 1). The maximum removal of Cr(VI) occurred at acidic pH. According to the obtained results, when the pH value reached 7.0, the removal percentage decreased gradually, while there was a small peak at pH 11.0. However, at pH 3.0 and 1 g/L of biomass, the removal percentage was found to be 70%. The findings showed that, when the pH value increased from 3.0 to 7.0, the removal percentage decreased by 26.48%. Besides, when it increased from 7.0 to 11.0, the removal percentage increased by 11.93%.

3.2. Effect of biomass dosage

The experiments were conducted with 40 mg/L synthetic solution of Cr(VI), 60 min contact time, pH 3.0, and algal dosages of 0.2, 0.5, and 1 g/L in 100 mL of metal solution at 25°C (Table 2). The results show that the removal percentage of Cr(VI) increased with increasing algal dosage, so that the optimum biosorbent dose was found to be 1 g/L. The specific removal capacity is a measure of the amount of Cr(VI) bound by per unit weight of biomass, as shown in Figs. 2 and 3. According to results, when the algal dosage increased from 0.2 to 1 g/L, the removal percentage also increased approximately by 15%.

3.3. Effect of contact time

The removal percentage of Cr(VI) under the influence of contact time has been depicted in Fig. 4. It was apparent that the removal percentage increased with increasing contact time (Table 3). The maximum removal was observed in 60 min of contact time. Under the optimum conditions, when the contact time increased from 30 to 60 min, the removal percentage increased by 18%.

3.4. Effect of initial metal concentration

The effect of initial metal concentration was studied in synthetic solutions containing 10, 25, and 40 mg/L initial Cr(VI) concentrations with various contact times but the same biomass dosage and pH value. The results showed that the removal efficiency increased with the increase in the initial Cr(VI) concentration (Fig. 4). According to findings, when the initial Cr(VI) concentration increased from 10 to 40 mg/L, the specific removal capacity increased from 4.78 to 27.48 mg/g at pH 3.0, from 2.51 to 20.92 mg/g at pH 7.0, and from 2.73 to 22.23 mg/g at pH 11.0. Moreover, when the initial metal concentration increased from 10 to 40 mg/L, the removal percentage increased by 22%.

Table 1

The mean and standard deviation of removal percentage as an effect of pH and initial Cr(VI) concentration in 60 min (regardless of the effect of biomass dosage)

C_0 (mg/L)	pH		
	3.0. $\bar{x} \pm \text{SD}$	7.0. $\bar{x} \pm \text{SD}$	11.0 $\bar{x} \pm \text{SD}$
10	37.93 \pm 7.63	21.61 \pm 3.07	24.80 \pm 2.4
25	48.09 \pm 3.67	35.73 \pm 0.97	42.42 \pm 1.22
40	60.87 \pm 5.91	50.86 \pm 1.15	54.65 \pm 0.971

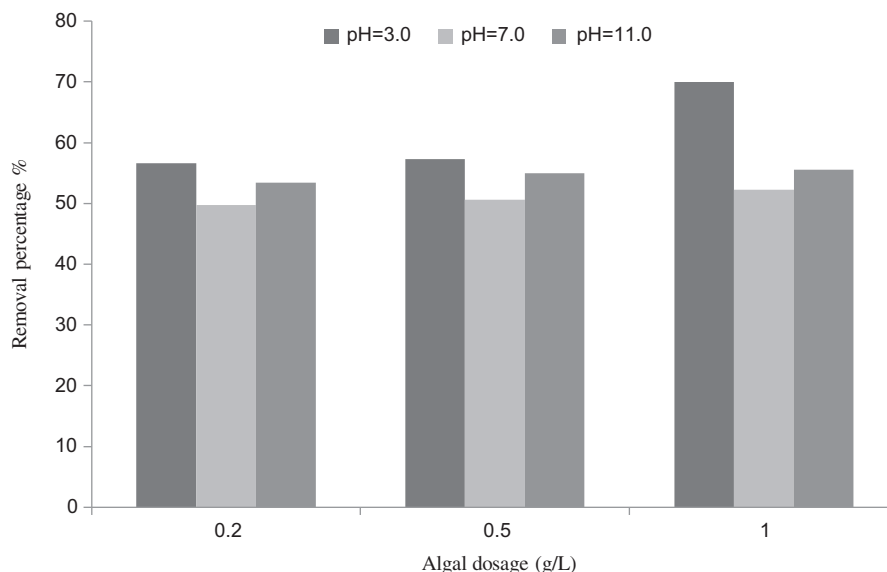


Fig. 1. Effect of algal dosage and pH on removal percentage of Cr(VI) by *S. porticalis*. ($C_0 = 40$ mg/L and contact time of 60 min).

Table 2

The mean and standard deviation of chromium removal as an effect of biomass dosage and initial metal concentration in 60 min (regardless of the effect of pH)

C_0 (mg/L)	Biomass (g/L)		
	0.2 $\bar{x} \pm SD$	0.5 $\bar{x} \pm SD$	1 $\bar{x} \pm SD$
10	23.66 ± 5.83	27.28 ± 5.91	33.38 ± 10.88
25	40.23 ± 4.48	41.51 ± 4.75	44.50 ± 6.96
40	53.26 ± 3.008	54.26 ± 2.94	58.85 ± 7.54

4. Discussion

This study, removal of Cr(VI) from synthetic aqueous solutions by powdered filamentous green algae *S. porticalis* was investigated as a function of pH, initial Cr(VI) concentration, algal dosage, and contact time.

In aqueous solutions containing heavy metals, pH has an important role in chemical processes such as hydrolysis, formation of organic and inorganic substances, oxidation, and reduction. Hence, this parameter significantly affects the removal of heavy metals from water and wastewater (Table 4). In this study, results showed that at acidic pH, the removal percentage of Cr(VI) ions was higher, which is similar to other studies. For example, Valli et al. [4] found that maximum Cr(VI) removal by marine algae occurred at pH 2.0. This is because, the overall surface charge of Cr(VI) ions is negative but functional groups on the

surface of algal cell wall have positive charges. Therefore, in acidic pH, Cr(VI) ions bind onto the surface of biomass cell wall through electrostatic attraction. On the other hand, when the pH goes to the neutrality, removal efficiency decreases, probably because the surface charge on the algal cell wall becomes negative; it has been confirmed by Barrera et al. study [28]. Since with decreasing pH removal efficiency increased, industrial acidic wastewaters containing high concentrations of chromium can be directly treated without adding acid to remove Cr(VI) by *S. porticalis*. This can eventually lead to decrease in operating costs of industrial wastewater treatment plants [2,29].

In the biosorption process, more initial metal concentration in the solution leads to more biomass capability for metal adsorption. It is in accordance to previous studies. Ahuja et al. [30] in their study showed that any increase in the concentration of zinc can increase the metal uptake by *Oscillatoria angustissima*. It has been found that nickel and copper adsorption from aqueous solutions by green algae *Chlorella vulgaris* increased with increasing initial concentration of metal ions, Mehta and Gaur reported [27]. The present study also confirmed that increasing the initial Cr(VI) concentration can increase metal uptake at all pH values, used biomass dosages, and all contact times. This may be due to more accessibility of chromium ions in aqueous solutions. Furthermore, high concentrations of hexavalent chromium in solution lead to increase in the driving force to overcome mass transfer resistance of ions between the solid and liquid phases [31].

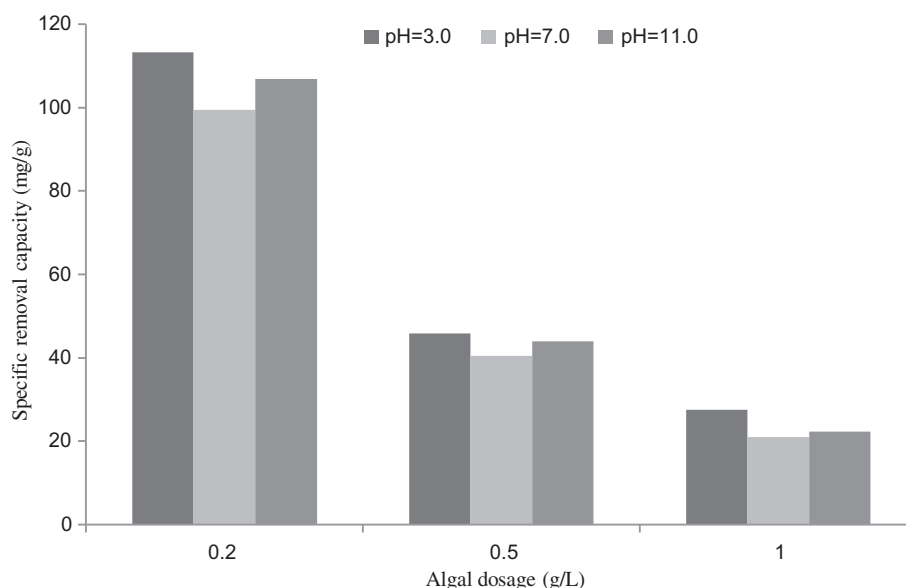


Fig. 2. The amount of Cr(VI) bound by per unit weight of biomass. ($C_0 = 40$ mg/L and contact time of 60 min).

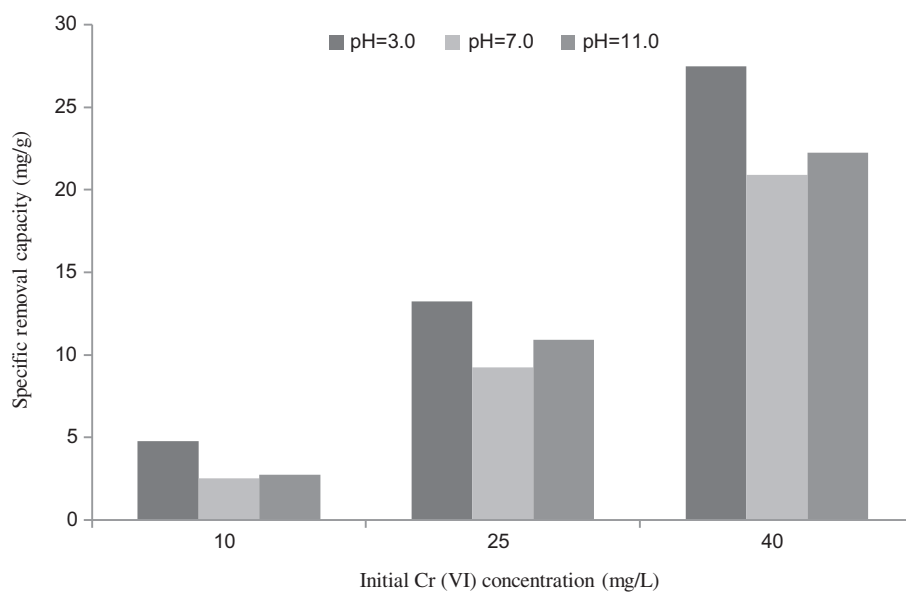


Fig. 3. The specific removal capacity as an effect of pH and initial metal concentration. (1 g/L biomass and contact time of 60 min).

Algae can be effectively used in the biosorption process through polysaccharides function, such as cellulose, chitin, glucans, and the presence of sulfate, carboxyl groups, and protein on the cell wall. Awasthi and Rai [32] found that more removal amount of nickel and cadmium was achieved by increasing algal dosage of *Anacystis nidulans*. Biosorption of Cr(VI) from aqueous solutions using dried macroalga *Clado-*

phora albida was studied by Deng et al. [2]. They recorded that removal efficiency by 1 and 10 g/L of biomass dosage was 45 and 95%, respectively, which is similar to present research findings and it is because more available adsorption surface was created by higher algal dosage. According to statistical analysis in this work, at constant pH, contact time and initial Cr(VI) concentration, the amount of algal dosage was

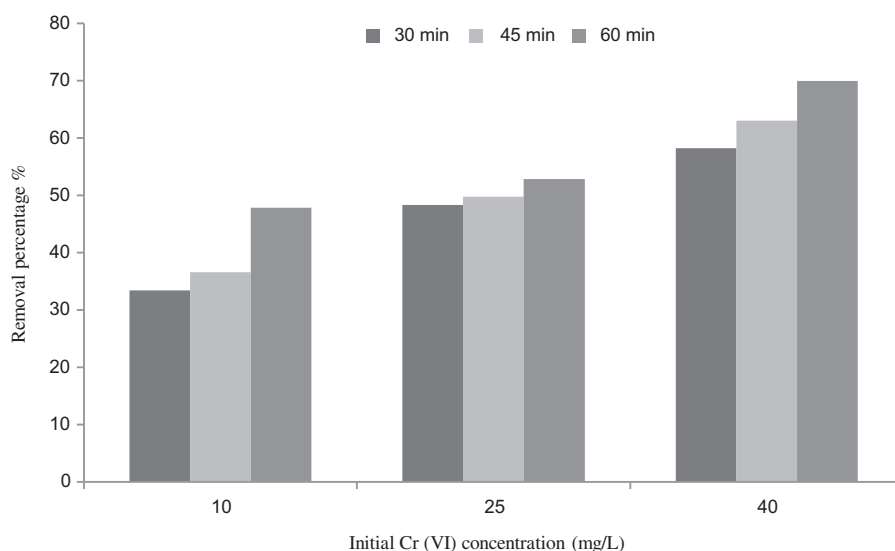


Fig. 4. Effect of contact time and initial metal concentration on Cr(VI) removal. (pH 3.0 and 1 g/L of biomass).

Table 3

The mean and standard deviation of removal efficiency as an effect of contact time and initial Cr(VI) concentration (regardless of the effect of pH and biomass dosage)

C_0 (mg/L)	Contact time (min)		
	30 $\bar{x} \pm SD$	45 $\bar{x} \pm SD$	60 $\bar{x} \pm SD$
10	21.73 \pm 6.63	24.82 \pm 6.60	28.11 \pm 8.62
25	40.11 \pm 4.60	40.90 \pm 4.83	42.08 \pm 5.60
40	52.83 \pm 3.40	54.002 \pm 4.19	55.46 \pm 5.39

significantly correlated with removal efficiency. It was noticed that the algal dosage is a key factor influencing the Cr(VI) removal process. Besides, the specific removal capacity decreased with the increase in the algal dosage. This was due to ions competition to locating on the available surface of biosorbent.

According to the results in the present study, there was a positive significant correlation between the removal efficiency of Cr(VI) and contact time at various ranges of pH, different biomass dosages, and all initial Cr(VI) concentrations. With passing of contact time, amount of collisions between Cr(VI) ions and

Table 4

Capability of various adsorbents to Cr(VI) removal

Adsorbent	Optimum pH	Maximum adsorption capacity (mg/g)	Literature
<i>S. porticalis</i>	3.0	27.48	This study
<i>Helianthus annuus</i>	2.0	7.2	[10]
<i>Dunaliella</i> spp.	2.0	45.5	[31]
Maize bran (agricultural waste)	2.0	312.52	[13]
Tamarind seeds (waste product)	1.12–1.46	29.7	[12]
<i>Cladophora albida</i> (macroalga)	2.0	24.8	[2]
<i>Rhizoclonium hookeri</i> (microalga)	2.0	67.3	[33]
Wild algal	2.0	7.007	[34]
<i>Limonia acidissima</i>	1.5	20.408	[8]

biomass increased and caused filling of empty spaces on the cell wall algae, so that removal efficiency enhanced.

5. Conclusions

In this work, the capability of *S. porticalis* under the influence of various factors including pH, biomass dosage, initial metal concentration, and contact time to remove chromium was studied. At the same contact times, the removal efficiency increased with increasing biomass dosage, metal concentration, and decreasing pH. Moreover, the amount of Cr(VI) removal increased as the contact time increased when pH, metal concentration, and biomass dosage were constant. Under the optimum conditions, the removal efficiency was found to be 70%. The results showed that *S. porticalis* which is broadly available at low cost, can be used as a suitable biosorbent to remove Cr(VI) from aqueous solutions and industrial acidic wastewaters containing high concentrations of chromium, even in low dosages. However, it is recommended that in future researches, the use of living green algae *S. porticalis* for bioaccumulation of toxic compounds and heavy metals from aqueous solutions and industrial wastewaters could be investigated.

Acknowledgments

The authors are grateful for the financial support of Vice Chancellor of Research in Kashan University of Medical Sciences, Iran.

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