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IRON BIOSORPTION FROM AQUEOUS SOLUTION BY PADINA SANCTAE CRUCIS ALGAE: ISOTHERM, KINETIC AND MODELING

Mozhgan Keshtkar[1], Sina Dobaradaran[1,2,3*], Samad Akbarzadeh[4], Masoud Bahreini[5], Dariush Ranjbar Vakil Abadi[1], Sara Ghaderi Nasab[5], Farshid Soleimani[6], Nahid Khajeahmadi[6], Masoud Mohamadi Baghmolaei[1]

[1]Department of Environmental Health Engineering, Faculty of Health, Bushehr University of Medical Sciences, Bushehr, Iran.
[2]The Persian Gulf Marine Biotechnology Research Centre, The Persian Gulf Biomedical Sciences Research Institute, Bushehr University of Medical Sciences, Bushehr, Iran.
[3]Systems Environmental Health, Oil, Gas and Energy Research Center, The Persian Gulf Biomedical Sciences Research Institute, Bushehr University of Medical Sciences, Bushehr, Iran.
[4]Faculty of Medicine, Biochemistry Department, University of Medical Science, Bushehr, Iran.
[5]Faculty of Nursing, Bushehr University of Medical Sciences, Bushehr, Iran.
[6]Student Research Committee, Bushehr University of Medical Sciences, Bushehr, Iran.
Email: S.dobaradaran@bpums.ac.ir

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Abstract:

The aim of this study was to determine Padina sanctae crucis algae efficiency in iron (Fe) biosorption from aqueous solutions. In a series of batch experiments, the experimental parameters were studied using the dried algae: initial Fe concentration (5-100 mg/l), biosorbent dose (0.1-10 g/l), pH (3-11), and contact time (5-120 min). The highest capacity biosorption was observed at 0.1 g/l of biosorbent at 120 min contact time, and initial Fe concentration level of 100 mg/l (34.65 mg/g). The biosorption was found to be better fitted to the Langmuir model and follow a pseudo-second-order kinetic model. Finally, it was concluded that the Padina sanctae crucis algae can be used as an environmental friendly, cheap and effective biosorbent in removal of Fe from aqueous solutions.

Keywords: Biosorption, Iron, Kinetic and isotherm models, Padina sanctae crucis algae, Persian Gulf.

1. Introduction

The pollution of water resources especially groundwater due to the indiscriminate disposal of pollutants has been causing worldwide concern over the last few decades. It is well known that contamination of water by some metals can have toxic or harmful effects on many forms of life. Metals, which are highly toxic particularly in high concentration level to human beings and ecological environments include: Cr, Cu, Pb, Hg, Mn, Cd, Ni, Zn and Fe [1, 2]. Iron (Fe) is one of the most common metals in the earth's crust [3]. It is used in paints and pigments, blue-print paper, laundry bluing, and in disposal of sludge from water treatment plants [4]. Although Fe is a vital element for humans and other forms of life but in high concentrations can cause tissue damage to organs by catalyzing the conversion of H_2O_2 to free radical ions that attack cell membranes and cause hemorrhagic necrosis, sloughing of mucosal areas in the stomach, breakage of DNA strands, and oncogene activation [4, 5]. Moreover, Fe toxicity can cause diabetes mellitus, hormonal abnormalities, atherosclerosis and related cardiovascular diseases, and a dysfunctional immune system. Oxidative stress induced by excess Fe can also cause brain damage [5]. In this regard, USA has stipulated a permissible limit of 1.6 mg/l for inland surface waters under the National Pollution Discharge Elimination System [6]. The World Health Organization (WHO) and European Commission have also recommended a guideline values of 0.3 mg/l and 0.2 mg/l for Fe in drinking water respectively [7, 8]. So Fe removal from the aquatic environment and water supply is, thus, essential. Various technologies have been employed to enhance water quality by removing inorganic contaminants [9]. The technologies available for Fe removal are either physicochemical or biological. Physicochemical iron oxidation can be performed by ion exchange, microfiltration, reverse osmosis, air stripping, or the use of strong oxidizing agents [10, 11]. Aeration-filtration requires a long detention time and the removal percentage is not as good as that obtained by an increase in pH alone; hence, a strong oxidant is required. Chlorine is the least expensive oxidant used to remove Fe, but can react with natural organic matter and may produce disinfection by-products (DBPs). Ozone is a very strong oxidizing agent and requires a very low oxidation time; however, it is expensive and has higher technological requirements than other oxidants [12-14]. A simple and widely-applicable treatment method is needed to remove Fe from aqueous solutions. Natural based methods can offer viable alternatives to chemical methods for removal of contaminants from aqueous solutions [15-19]. The advantages of natural based methods over conventional physicochemical treatment are that they do not require the use of chemicals, are environmentally friendly, and require lower operation and maintenance costs [20-24].

The objective of this study was to determine the effectiveness of Padina sanctae crucis algae in Fe removal from aqueous solutions. In this study, the effect of several important factors which can affect Fe biosorption such as biosorbent dosage, initial Fe concentration level, pH and contact time, was also discussed. Beside this we determined the biosorption kinetics and models, as well as biosorption isotherms by using Padina sanctae crucis algae.

2. Materials and methods

2.1. Biosorbent preparation: The brown algae Padina sanctae crucis was obtained along the northern part along the Persian Gulf in Bushehr port coastline area (N 28˚58′21.6″ and E 50˚49′23.3″). After collection and transfer to laboratory, the algae was washed three times by tap water and then two times by distillated water to take away sands, clay, and other impurities. The washed algae were then dried in an oven (Memmert, Germany) at 105˚C for 24 h and finally powdered and sieved through a 0.71 mm screen.

2.2. Fe biosorption by batch study

A stock solution of 1000 mg/l Fe was prepared by dissolving $FeCl_3.6H_2O$ (Merck, Germany) in ultrapure water. Fe solutions were prepared at 5, 10, 20, 50 and 100 mg/l concentration levels. At each run, 100 mL of Fe solution with particular initial Fe concentration level was agitated at 120 rpm. The effect of seven different contact times (5, 10, 25, 45, 60, 90, and 120 min), five biosorbent doses (0.1, 0.5, 1, 5 and 10 g/l), five initial Fe concentration levels (5-100 mg/l) and five different pH values (3, 5, 7, 9, 11) were studied in the batch systems at room temperature (25 ± 1˚C). The remaining concentration level of Fe in solution after biosorption was determined by using an atomic absorption spectrometer (AAS) method (Varian, SpectrAA 240, Australia).

The removal efficiency was calculated by using the following Eq. 1 [25]:

$$\text{Biosorption yield} = \frac{(C_i - C_e)}{C_i} \times 100 \quad (1)$$

Where C_i and C_e are the concentration levels of Fe before and after the experiment in any time (mg/l).

The equilibrium biosorption capacity of Padina sanctae cruces algae at different Fe concentration levels was calculated by using the following Eq. 2:

$$q_e = \frac{(C_i - C_e)V}{m} \quad (2)$$

q_e is the equilibrium biosorption capacity (mg/g); C_i is the Fe concentration level at initial time (mg/l); C_e is the concentration level of Fe in solution at equilibrium time (mg/l); V is the solution volume (L); and m is the biosorbent dosage (g).

3. Results and discussion

3.1. Effect of pH: The effect of solution pH values on the Fe biosorption using the Padina sanctae crucis algae was investigated and the results are shown in Figure 1. Biosorption process is dependent on the pH value of aqueous phase, since it affects biosorbent surface charge, the degree of ionization, and the species of biosorbate [26]. The maximum removal efficiency was observed at a pH value of 7. It was observed that by increasing the pH value of solution from 3 to 7 the removal efficiency increased and then decreased with further increasing the pH value of solution from 7 to 11. At lower pH value, hydrogen ions are likely to compete with Fe and at pH values above 7, Fe might precipitate as hydroxides [1].

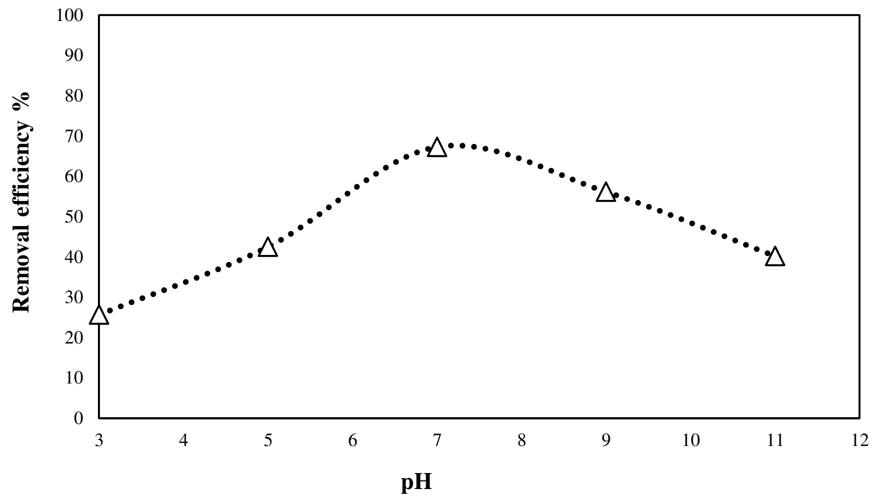

Figure-1: Fe biosorption as a function of pH (biosorbent dose: 1 g/l, contact time: 60 min, initial Fe concentration level: 50 mg/l).

In a similar study, Moreno-Pirajan et al. examined Fe removal using activated carbon prepared by coconut shells [1]. Initial pH values of solution in their study were in the range of 1 to 14. They reported the highest Fe adsorption at neutral pH values.

In another study, Kousalya et al. showed that Fe removal by using nano-hydroxyapatite chitin/chitosan hybrid biocomposites increased with increasing the pH values from 3 to 7 and then decreased with increasing the pH values from 7 to 11 [27].

3.2. Effect of biosorbent dosage

The effect of biomass dosage on the Fe removal by biosorbent was studied by using different biomass doses of Padina sanctae crucis algae in the range of 0.1– 10 g/l (Figure 2). The removal of Fe increased from 7.29, to 21.35, 41.71 and 95.08 % by increasing the biosorbent dosages from 0.1, to 0.5, 1 and 5 g/l, respectively. This can be explained by increasing of active site numbers with respect to increase of biosorbent dose. However, it was observed that after the biosorbent dose of 5 g/l, there was no significant change in the removal of Fe. This trend could be explained as a consequence of a partial compression of biomass at higher biomass dose level, which results in a reduction in effective surface area for the biosorption [28].

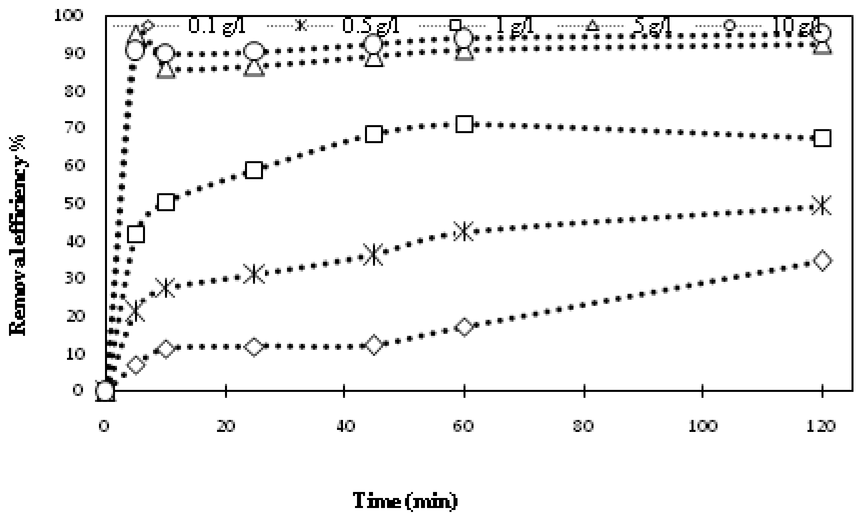

Figure-2: Fe biosorption as a function of biosorbent dose (initial Fe concentration level: 100 mg/l, pH=7).

Similar results have been reported by Bordoloi et al. in removal of Fe from aqueous solutions by using banana ash, carbonates and biocarbnates of Na, K and their mixtures [29]. In another study, Moreno- Pirajan et al. reported that by increasing the biomass doses of activated carbon from coconut shells, biosorption rate of Fe increased from 54 to 90 percent. The maximum Fe biosorption was obtained at biomass dosage of 0.3 g/l and it was almost fixed at higher dosages of biosorbent [1].

3.3. Effect of contact time and initial Fe concentration level

The effects of initial Fe concentration level and contact time on the Fe removal are shown in Figure 3. As seen in Figure 3, the Fe biosorption rate at different concentration levels of initial Fe (5, 10, 20, 50 and 100 mg/l) was rapid at the initial stage and then the biosorption rate progressively decreased with the progress of biosorption until the equilibrium reached in approximately 25 min after the run. With increasing the initial Fe concentration level, the process biosorption of Fe took a bit longer to reach equilibrium. This trend could be explained by the theory that many of metal cations self impact and contest for the active biosorption sites [30, 31]. The biosorption capacity of Padina sanctae cruces algae for Fe removal from aqueous solution was found to increase aggressively with an increase in the initial Fe concentration level. The biosorption capacity of Padina sanctae cruces algae at a constant biosorbent dosage level of 1 g/l increased from 0.5 mg/g to 8.45 mg/g when the initial Fe concentration level increased from 5 to 100 mg/l respectively. The enhancement in Fe biosorption could be due to an increase in electrostatic interactions, involving sites of progressively lower affinity for metal ions and high capacity of biosorbent in Fe biosorption [32]. When initial Fe concentration level was low, the availability of various active sites and the uncovered surface of Padina sanctae cruces algae were rather high. The higher initial Fe concentration level, the more ions binding with biosorption sites of Padina sanctae cruces algae, thus resulting in a further increase in biosorption capacity of Padina sanctae cruces algae. Victor-Ortega et al. and Hamdouni et al. found similar results in the removal of Fe from aqueous solutions by using strong-acid cation exchange resin and aqueous portlandite carbonation and calcite-solution, respectively [33, 34].

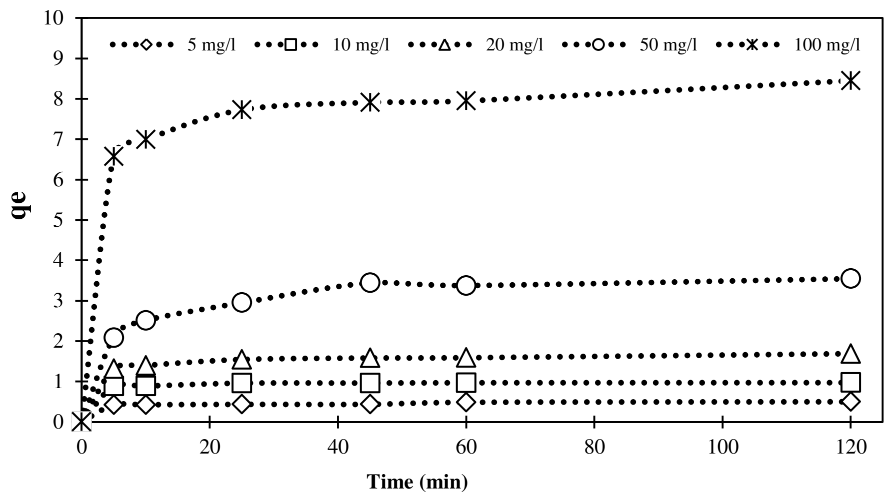

Figure-3: Fe biosorption as a function of initial Fe concentration level (biosorbent dose: 1 g/l, pH= 7).

3.4. Biosorption isotherms

To quantify the biosorption capacity of Padina sanctae cruces algae for Fe removal from aqueous solutions, two frequently used isotherms namely Freundlich and Langmuir have been adopted.

The linear form of Freundlich [35] isotherm can be written as Eq. 3:

$\text{Log}\,(q_e) = \text{Log}\,(K_f) + 1/n\ \text{Log}\,C_e$ …. (3)

Where q_e is the mass of Fe adsorbed per unit weight of the biosorbent (mg/g); K_f is the Freundlich capacity factor and a measure of biosorption capacity; 1/n is the equilibrium concentration of Fe in solution (mg/l) after biosorption, and C_e is equilibrium concentration level of Fe in solution.

The values of 1/n and K_f for the biosorbent were calculated from the slope and the intercept of the linear plot of log q_e vs log C_e (Figure 4a). Biosorption Freundlich isotherm parameters of Fe onto Padina sanctae cruces algae are shown in Table 1.

The Langmuir biosorption isotherm [36] model is describe as Eq. 4:

$$\frac{C_e}{q_e} = \frac{1}{bq_{max}} + \frac{1}{q_{max}} C_e \quad .... (4)$$

Where q_e is the mass of Fe per unit mass of biosorbent (mg/g), q_{max} is the maximum biosorption capacity of Fe (mg Fe/g biosorbent), b is the Langmuir constant related to the free energy of biosorption equilibrium concentration level of Fe in solution (mg/l) after biosorption. The Langmuir constant can be specified by plotting $\frac{C_e}{q_e}$ versus C_e (Figure 4b).

Biosorption Langmuir isotherm parameters of Fe onto Padina sanctae cruces algae are shown in Table 1. As shown in Figure. 5a, b, and Table 1, Langmuir model is better fitted than Freundlich model. Langmuir isotherm considers the homogeneous biosorption surface and the possibility of monolayer biosorption.

Table 1: Biosorption isotherm parameters for Fe biosorption onto Padina sanctae cruces algae

Langmuir parameter				Freundlich parameter		
b (L/mg)	q_m (mg/g)	R_L	R^2	K_F (L/g)	n	R^2
0.0091	21.186	0.5235	0.9853	0.124	0.79	0.9626

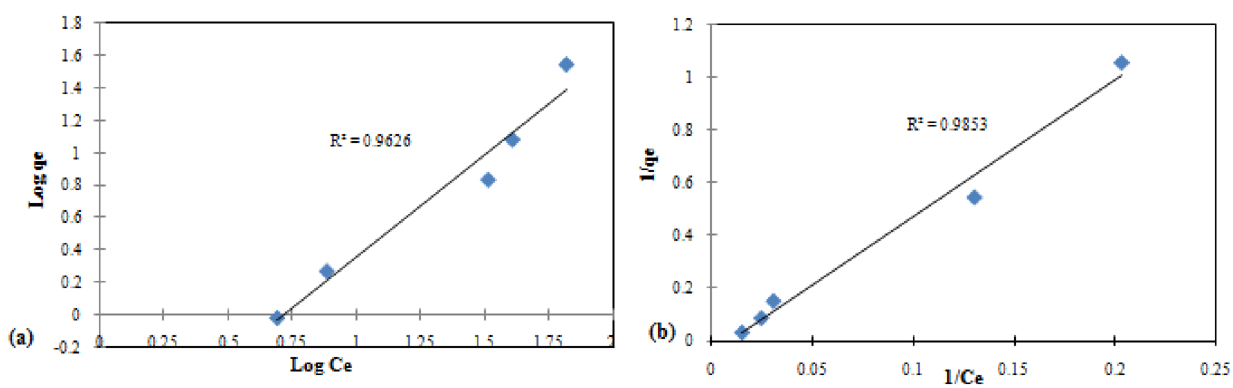

Figure-4: (a) Freundlich and (b) Langmuir isotherms investigation of Fe biosorption by Padina sanctae cruces algae.

3.5. Biosorption kinetics and modeling

The biosorption kinetics is important in the treatment of aqueous solution, as it presents significant insights into reaction and mechanisms of biosorption reactions. The experimental biosorption kinetics were defined by using pseudo- first and pseudo-second- order kinetics. This kinetics can be writtenas follows:

Pseudo-first- order model,

$\text{Log}\ (q_e - q_t) = \log q_e - \frac{K_{1,}}{2.303}\ t,$ ….(5)

Pseudo-second- order model,

$\frac{t}{q} = \frac{1}{{q_e}^2 K_2} + \frac{1}{q_e}\ t,$ …. (6)

The intraparticle diffusion model equation can be also described as Eq. 8:

$q_t = k_d\ t^{0.5} + C$ …..(7)

Where q_e is the mass of solute sorbed at equilibrium (mg/g), q_t is the mass of solute sorbed at time t (mg/g), K_1 is the first-order equilibrium rate constant (1/ min), and K_2 is the second-order equilibrium rate constant (1/ min).

The first-order equilibrium rate constant (K_1) for Fe biosorption was calculated from the slop of the linear plot of log $(q_e - q_t)$ versus time. In the case of the second-order equilibrium rate constant (K_2), kinetic data were plotted between t/q_t against time. The intraparticle diffusion model rate constant (K_d) and C can be measured from the slope and intercept of the linear plot of q_t vs $t^{0.5}$, respectively.

Biosorption kinetic parameters for Fe biosorption onto Padina sanctae cruces algae are shown in Table 2. As shown in Figure. 5 a, b, c, kinetics of Fe biosorption by Padina sanctae cruces algae followed the pseudo-second model, indicating that the biosorption limiting step may be chemisorption. This suggests that the biosorption of Fe maybe occurs via surface complexation reactions at specific biosorption sites of Padina sanctae cruces algae [37-39]. In addition, the plots showed that the intraparticle diffusion model was not the only rate controlling step because the line did not pass through the origin (C≠0). This showed that both intraparticle diffusion and boundary diffusion affected the Fe biosorption by Padina sanctae cruces algae.

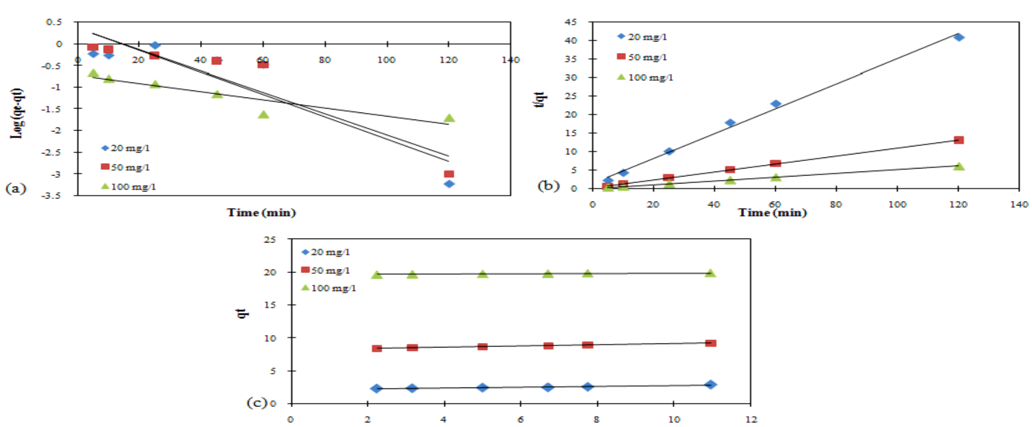

Figure-5: Biosorption kinetics of Fe biosorption by Padina sanctae cruces algae: (a) pseudo-first-order, (b) pseudo-second-order and (c) intraparticle diffusion kinetic.

Table-2: Biosorption kinetic parameters for Fe biosorption onto Padina sanctae cruces algae.

C_0 (mg/l)	q_{exp} (mg/g)	Pseudo-first- order			Pseudo-second-order			Intraparticle diffusion model		
		q (mg/g)	K_1	R^2	q (mg/g)	K_2	R^2	K_d	C	R^2
20	2.94	2.33	0.059	0.82	2.972	0.0781	0.9944	0.0634	2.1179	0.9285
50	9.216	2.29	0.056	0.866	9.259	0.0764	0.9996	0.0948	8.1741	0.997
100	19.82	0.18	0.021	0.842	19.802	0.5313	1	0.0271	19.563	0.959

4. Conclusion

In this study the efficiency of Padina sanctae crucis algae in iron (Fe) biosorption from aqueous solutions was evaluated. Our results revealed that Padina sanctae cruces algae is a suitable biosorbent for Fe removal from aqueous solutions. The operating parameters including pH, initial Fe concentration level, biosorbent dosage and contact time were effective in Fe removal from aqueous solutions. The removal efficiency increased by increasing contact time, biosorbent dose, and initial Fe concentration level at a fixed biosorbent dosage. Biosorption efficiency was highest at neutral pH value (pH=7) and the highest biosorption capacity (34.65 mg/g) was achieved at 0.1 g/l of biosorbent dosage, 120 min contact time, initial Fe concentration level of 100 mg/l. Langmuir model was slightly better fitted than Freundlich model and showed homogeneous biosorption surface and the possibility of monolayer biosorption of Fe by biosorbent. The biosorption process was observed to follow a pseudo-second-order kinetic, indicating that the biosorption limiting step may be chemisorptions. Finally, it should be noted that applied biosorbent in this study can be used as an effective, environmental friendly, and cost effective biosorbent for Fe removal from aqueous solutions. This process may sufficiently be used for Fe removal from industrial effluents containing high concentration level of Fe.

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Interest's statement

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

Authors' contributions

MK has done some of experiments and wrote the first draft of manuscript, SD has design the study as well as manuscript structure, writing and editing of manuscript. SA and DRV have guided the manuscript preparation. SGN, FS, NK, and MMB have collected and prepared the algae for experiments as well as helped MK in performing experiments.

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Corresponding Author:

Sina Dobaradaran*,

Email: *S.dobaradaran@bpums.ac.ir*