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Biosorption of fluoride from aqueous phase onto *Padina sanctae crucis* algae: evaluation of biosorption kinetics and isotherms

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ABSTRACT

Biosorption is considered as one of the favorable treatment technologies for fluoride removal from aqueous solutions. The present study is the first attempt to evaluate the biosorption capability of the *Padina sanctae crucis* algae in removal of fluoride from aqueous phase. Biosorption was studied in a series of batch experiments at room temperature (25 ± 1˚C). The effects of experimental parameters such as biosorbent dose (0.8–64 g/L), contact time (5–120 min), pH (3, 7, and 11), initial fluoride concentration (2–8 mg/L), and the presence of competing anions (bicarbonate, nitrate, carbonate, sulfate, and chloride) were investigated. The highest removal biosorption was observed at 48 g/L biosorbent dose, 5 min contact time, and initial fluoride concentration at 8 mg/L (97% fluoride removal). Competing anions had no significant effect on the fluoride biosorption by the biosorbent. Freundlich model was better fitted than Langmuir model that showed heterogeneous biosorption surface and the possibility of multilayer biosorption of fluoride by biosorbent. The biosorption kinetics was controlled by the pseudo-second-order and pore diffusion models. It was concluded that the *Padina sanctae crucis* algae can be used as an effective, low-cost, and environmental friendly biosorbent for fluoride removal from aqueous solutions.

Keywords: Biosorption; Fluoride; Freundlich model; *Padina sanctae crucis*; Persian Gulf; Pseudo-second-order kinetic

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1. Introduction

Earth's crust contains fluoride in the average concentration of 0.3 g/kg. Background concentration level of fluoride in the air throughout the world is also about 3 ng/m. Entrance of fluoride into the water bodies is principally occurred through natural resources. Moreover, industries that use inorganic fluoride-containing compounds, including phosphate fertilizer, aluminum, steel, brick, glass fiber, tile, and ceramic production industries are the main artificial resources of fluoride entrance into water bodies and play a significant role in fluoride water pollution [1,2].

Fluoride can cause an extensive range of adverse health effects such as teeth and bone damage, as well as adverse health consequence in soft tissues [3–7]. Due to possible adverse health effect of fluoride, the World Health Organization [8] has set a recommendation value for fluoride in drinking water at 1.5 mg/L. Also US Public Health Service has established a new guideline value of 0.7 mg/L for fluoride and changed the previous recommended range of 0.7–1.2 mg/L [9]. So with view to the special concerns of fluoride, various studies have been done in relation to fluoride concentration levels in drinking water, air, fish, and sea like Iran in concerned areas [10–16].

A number of processes including biosorption, ion-exchange, precipitation, electrodialysis and electrochemical, were developed to eliminate extra fluoride from aqueous solution [17]. Some of these systems are relatively expensive and/or possess certain limitations such as high volume of sludge formation, and therefore there is a need to improve cost-effective choices. Among these methods, biosorption is a commonly used one for fluoride removal [18–25].

At present, significant interests were observed on the application of biosorbent materials for removal of different impurities. Biosorption has a potential marketing interest over other common treatment methods as it is cheaper and environmental favorable, especially when natural biomass is used. Low-cost biosorbent could be generated from numerous raw materials such as industrial and agriculture waste. Many efforts have been described on pollutants elimination mainly heavy metals by biosorption [26–32]. Biomass of marine algae such as brown algae is available in large quantities and can form a good basis for the development of biosorbent materials [33]. Brown algae are generally used as nutritional supplements, animal feed, fertilizers, and as a source of thickeners such as alginate [34]. But only a limited number of studies have so far been focused on the usage of non-living biomass of marine algae for fluoride removal from aqueous solution.

To our best knowledge, the present study is the first attempt to evaluate the biosorption capability of the *Padina sanctae crucis* algae (Fig. 1) in removal of fluoride from aqueous solution. In this study, the effect of several important factors which can affect fluoride biosorption such as biosorbent dosage, initial fluoride concentration, pH, and contact time, was also discussed. Beside this, we determined the biosorption kinetics and modeling, sorption isotherms, and the effect of competing anions on fluoride biosorption by *Padina sanctae crucis* algae.

2. Materials and methods

2.1. Biosorbent preparation and activation procedure

The brown algae *Padina sanctae crucis* was obtained along the Persian Gulf in the Bushehr port coastline area (N 28˚58′26.35′′ and E 50˚49′33.25′′). After collection and transfer to laboratory, the algae were washed three times by tap water and then two times by distilled water to take away clay, sands, and other impurities. The washed algae were then dried in oven at 105˚C for 24 h and finally powdered and sieved through a 0.71 mm screen. For activation, sieved biomass was placed in a furnace (Carbolite, England) at three different temperatures of 200, 400, and 600˚C.

Fig. 1. *Padina sanctae crucis* algae along the Persian Gulf in the Bushehr port coastal area (photo credit: image courtesy of Reshad Balef).

2.2. Fluoride sorption by batch study

A stock solution of 100 mg/L fluoride was prepared by dissolving sodium fluoride (Merck, Germany) in ultrapure water. Fluoride solutions were prepared at 2, 3, 5, and 8 mg/L concentration levels. At each run, 100 mL of fluoride solution with particular initial fluoride concentration was agitated at 120 rpm. The effect of five different contact times (5–120 min), four initial fluoride concentrations (2–8 mg/L), three different pH values (3, 7, and 11), and various mass ratios of biosorbent to the initial fluoride concentration (five ratios within the range of 400–8,000) were studied in the batch systems at room temperature (25 ± 1˚C). The standard SPADNS method [1] was used by using a spectrophotometer (HACH, USA, model CAM Spec M501) for analysis of the remaining fluoride level in the aqueous solution after each experiment. The removal efficiency was calculated using the following Eq. (1) [35]:

$$\text{Biosorption yield} = \frac{(C_\mathrm{i} - C)}{C_\mathrm{i}} \times 100 \qquad (1)$$

where C_i and C are the concentrations of fluoride before and after the experiment in any time (mg/L).

The equilibrium biosorption capacity of *Padina sanctae crucis* algae at different fluoride concentration levels was calculated by using the following Eq. (2):

$$q_\mathrm{e} = \frac{(C_\mathrm{i} - C_\mathrm{e})V}{m} \qquad (2)$$

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

2.3. Characterization of biosorbent

FTIR spectra of *Padina sanctae crucis* algae samples were recorded by using a FTIR spectrometer (PerkinElmer FT-IR, model Spectrum RXI) over the wave number range from 4,000 to 400 cm^{-1}.

3. Results and discussion

3.1. Characterization of the biosorbent

FTIR spectra of *Padina sanctae crucis* algae before and after fluoride biosorption were characterized and show in Fig. 2(a) and (b). The band at wavenumbers of 3,416.3, 2,922.88, 2,852.11, 2,143.56, 1,054.54, 1,627.07, 670.22 cm^{-1}, are indicative of the appearance of the stretching of –OH, $-\mathrm{CH}_2$, –C≡C–, C–O–, >C=C<, C–X. Comparison of FTIR spectra of *Padina sanctae crucis* algae (Fig. 2) before (a) and after fluoride biosorption (b) showed that the –OH band shifted from 3,416.3 to 3,449.32 cm^{-1}, the >C=C< band shifted from 1,627.07 to 1,626.36 cm^{-1}, and $-\mathrm{CH}_2$ shifted from 2,924.44 to 2,922.88 after fluoride biosorption. The peak at 1,054.54 cm^{-1}, due to stretching vibration of C–OH group of carboxylic acid also shifted to 1,032.2 cm^{-1} due to interaction with fluoride [36].

3.2. Effect of pH

The effect of pH on fluoride removal was studied in the pH range of 3 to 11 and results are explained in Fig. 3(a). pH played a key role in fluoride biosorption. Biosorption process is dependent on the pH value of aqueous phase, the functional groups on the biosorbent, and their ionic states at specific pH value [37,38].

It has been reported that biosorbent biomass contains a large quantity of polysaccharides and some of them are associated with proteins and other components [39,40]. These biomacromolecules on the biosorbent surfaces have various functional groups (such as, alcohol, amino, phenol, carboxyl, thiol, sulfydryl, and phosphate groups) and biosorption phenomena is influenced by the protonation or deprotonation of these functional groups [39]. The ionic form of fluoride in aqueous solution and the electrical charge of the biosorbent surface influence by the pH value of solution. The fluoride biosorption from aqueous solution was highly dependent on the pH value of solution. The maximum removal efficiency was observed at a pH value of 7. It was observed that by increasing the pH 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. Decrease in fluoride removal at lower pH value in present study (pH 3) may be due to the formation of the weakly ionized HF ($\mathrm{p}K_\mathrm{a} = 3.2$) and at higher pH value (pH 11) may be because of the competitiveness of the OH^- and F^- ions [17].

Similar results have been reported in the removal of fluoride from aqueous solution using calcined Zn/Al hydrotalcite-like compound (HTlc) [41]. In other studies, Viswanathan et al. examined fluoride removal by using Fe(III) loaded carboxylated chitosan beads and La(III) incorporated carboxylated chitosan beads, respectively [42,43]. Initial pH values of solution in their study were in the range of 3–11 (3, 5, 7, 9, and 11). In both studies, the maximum biosorption was

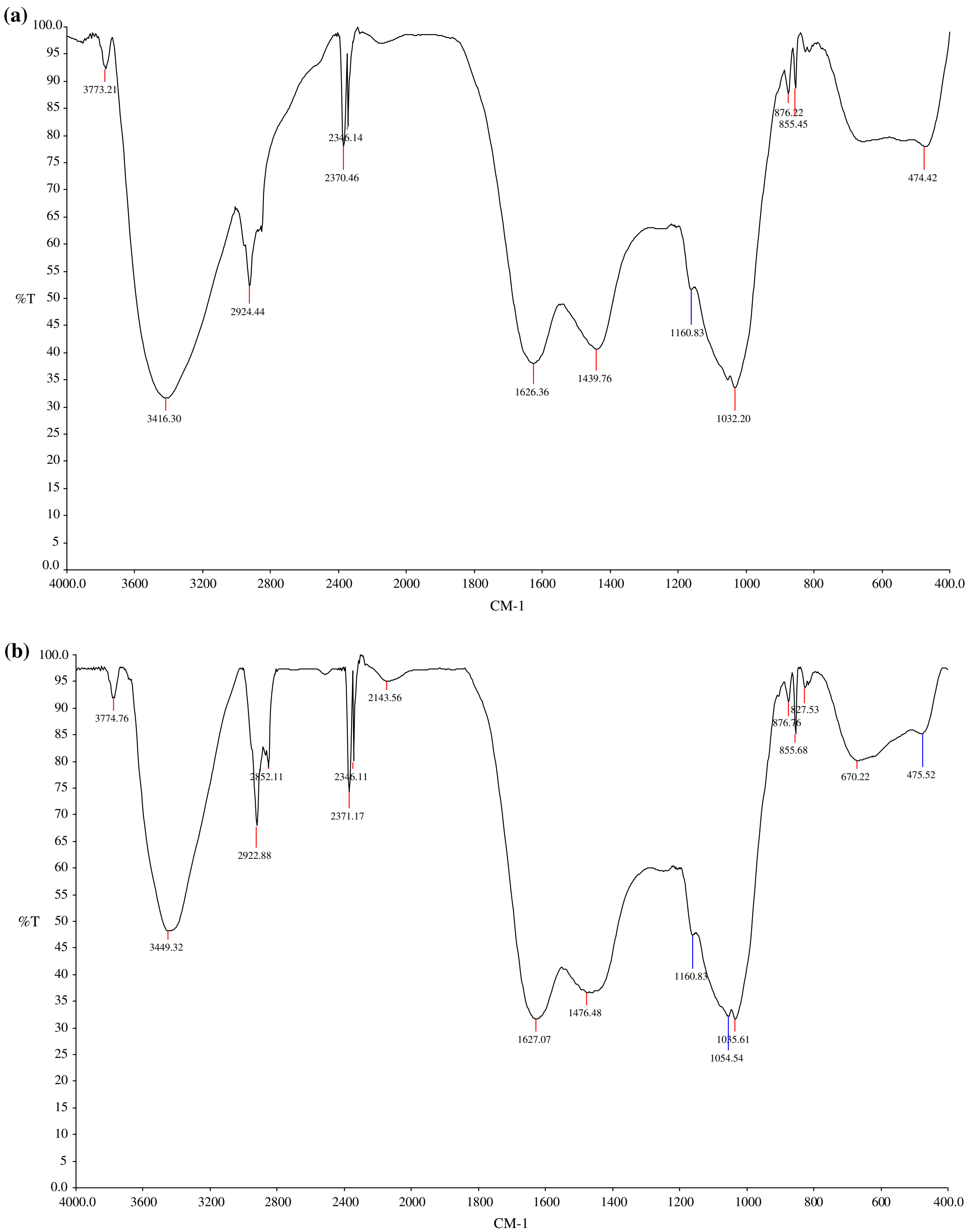

Fig. 2. FTIR spectra of *Padina sanctae crucis* algae before (a) and after (b) fluoride biosorption.

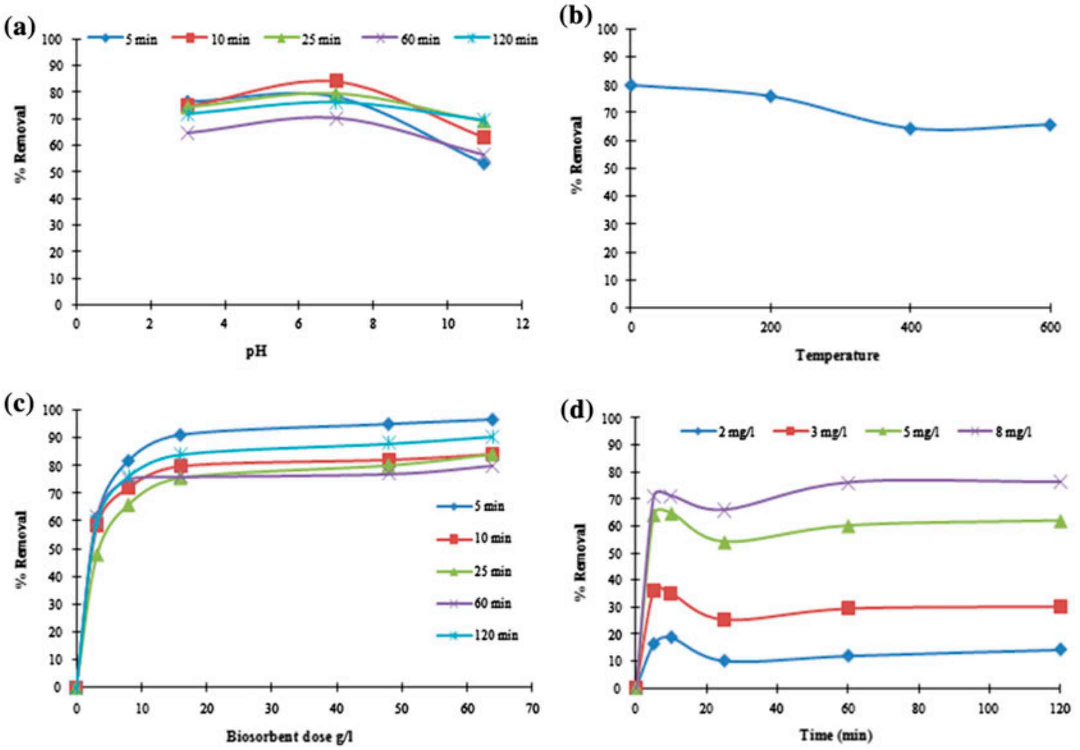

Fig. 3. Fluoride biosorption as a function of (a) pH (biosorbent dose: 8 g/L; initial fluoride concentration: 8 mg/L), (b) activation temperature (biosorbent dose: 3.2 g/L; fluoride concentration: 8 mg/L; contact time: 25 min; pH 7), (c) biosorbent dose (initial fluoride concentration: 8 mg/L; pH 7), and (d) initial fluoride concentration (g/L biomass to mg/L fluoride: 6,000; pH 7).

observed at neutral pH values. Whereas, it has been reported that the removal efficiency for fluoride biosorption by shrimp shell waste increased by increasing pH value from 3 to 11 [44]. However, Venkata Mohan et al. showed that removal of fluoride by using algal biosorbent, *Spirogyra* sp.-IO2 decrease with increasing the pH value from 2.0 to 10.5 [45]. In contrast, Kemer et al. and Nigussie et al. found that pH had no effect on fluoride removal by waste residues from waste mud and alum manufacturing process, respectively [46,47]. According to results of this study, the pH value of 7 was the optimum pH. This can be an advantage of using *Padina sanctae crucis* algae as biosorbent because of cost saving and lower requirement of biosorbent in the rural areas, where pH adjustment may not be a preferable solution.

3.3. Effect of algae activation temperature

In order to determine the optimal activation temperature, different temperatures (0 (without activation), 200, 400, and 600˚C) were used for activation of biosorbent. As shown in Fig. 3(b), by increasing temperature from 200 to 600˚C the biosorption efficiency decreased. The possible reason for the decrease in biosorption efficiency was probably due to the loss of functional groups upon thermal activation [48].

Wu et al. reported that fluoride removal by using a novel Fe-Al-Ce trimetal oxide biosorbent decreased significantly from 83.4 to 18.2% by increasing temperature from 700 to 850˚C [49]. In another study, Hauge et al. investigated fluoride binding capacity of clay and clayware at various firing temperature range from 500 to 1,000˚C. They reported that the effect of firing temperature on fluoride biosorption by pottery declined by increasing firing temperature from 600 to 900˚C [50]. According to results of this study, the biosorption efficiency of *Padina sanctae crucis* algae in removal of fluoride after drying process (at 105˚C for 24 h) was higher in comparison to use activation process at higher temperatures (200, 400, and 600˚C) plus drying process. This can be another advantage of this biosorbent as a fluoride biosorbent, because it doesn't need high energy for activation process.

3.4. Effect of biosorbent dose

The effect of biosorbent dosage in the range from 3.2 to 64 g/L on fluoride biosorption at pH value of 7, initial concentration of fluoride at 8 mg/L, and different contact time were studied. As shown in Fig. 3(c), the biosorption rate rapidly increased in the first 5 min and then biosorption rate was almost constant (with the exception of biosorbent dosage of 64 g/L at contact time of 120 min). The biosorption efficiency was depended on the increasing biomass dosage in the solution. This can be attributed to the additional number of biosorption sites, which are resulted from the increase in the biosorbent dosage. In the first 5 min, fluoride removal increased from 61 to 97% with increasing biosorbent dosage from 3.2 to 64 g/L. It should be noted with the exception of biosorbent dosage of 64 g/L at contact time of 120 min there was no significant difference between fluoride removal at biosorbent dosage from 16 to 64 g/L. This trend could be explained as a consequence of a partial compression of biomass at higher biomass concentration level, which results in a reduction in effective surface area for the biosorption [51]. Similar results have been reported by Dobaradaran et al. in defluoridation of aqueous solutions by using shrimp shell waste and *Moringa oleifera* seed ash [44,52]. In aqueous solution, defluoridation by using *M. oleifera* seed ash, the fluoride removal percentage increased with increasing biomass dosage from 2 to 40 g/L but there were no significant differences in the fluoride removal percentage between biosorbent dosage of 30 and 40 g/L. In another study, Jagtap et al. reported that by increasing the biomass doses of modified chitosan-based, biosorption rate increased from 19 to 81.98%. The maximum fluoride biosorption was obtained at biomass dosage of 1 g/L and it was almost fixed at higher dosages of biosorbent [53]. Sivasankar et al. also examined removal of fluoride by using MnO_2-coated Tamarind Fruit (*Tamarindus indica*) shell at different doses and observed that the percentage sorption of fluoride increased with increasing of biomass doses from 4 to 16 g/L. However, there were no significant differences between fluoride removal at adsorbent dosage higher than 16 g/L [17].

In contrast, Thakre et al. used lanthanum incorporated chitosan beads (LCB) for removal of fluoride from drinking water and reported that LCB at dosage range from 0.2 to 2 g/L has no significant fluoride removal capacity [54].

3.5. Effect of contact time and initial fluoride concentration

The effect of initial fluoride concentration level on the fluoride removal is illustrated in Fig. 3(d). It was observed that by increasing substrate concentration from 2 to 8 mg/L, at a fixed mass ratio of biosorbent to initial fluoride concentration, the removal efficiency increased. This might be due to high biosorption capacity of *Padina sanctae crucis* algae. Fluoride removal reached a maximum after 10 min and then decreased with increasing contact time which can be due to desorption and a reduction of the active surface area. In a similar trend, Dobaradaran et al. examined the efficiency of seed *M. oleifera* ash and shrimp shell waste in removal of fluoride from aqueous solutions. They reported that by increasing the fluoride concentration from 2 to 8 mg/L, the removal efficiency increased from 33.14 to 80.84 and 33 to 81%, respectively [44,52]. Mahramanlioglu et al. and Viswanathan et al. reported the same results in the fluoride removal from aqueous solutions by using acid treated spent bleaching earth and protonated chitosan beads, respectively [55,56]. In contrast, Ramanaiah et al. performed adsorptive experiments on fluoride biosorption by using waste fungal biomass (*Pleurotus ostreatus* 1804) derived from laccase fermentation process and found that the biosorption efficiency of fluoride by biosorbent decreases with increasing initial fluoride concentration [57]. The biosorption of fluoride in this study increased up to initial concentration level of 8 mg/L and then decreased with increasing initial concentration of fluoride. The reason was that the capacity of the biosorbent substances gets exhausted with increase in the initial concentration level of fluoride. This was because of the point that for a fixed biosorbent dosage, the whole existing biosorption sites were limited, which became saturated at a higher concentration levels of fluoride. Similar trend has been reported for removal of fluoride by using Neem charcoal by Chakrabarty and Sarma [58].

3.6. Effect of competing anions

A mixture of anions normally exists in fluoride-containing water that competes with fluoride for biosorption. In the present study, the removal of fluoride in the presence of competing anions such as bicarbonate (HCO_3^-), nitrate (NO_3^-), chloride (Cl^-), carbonate (CO_3^{2-}), and sulfate (SO_4^{2-}) was investigated with the concentration levels of 200 and 400 mg/L. The initial concentrations of fluoride and biosorbent

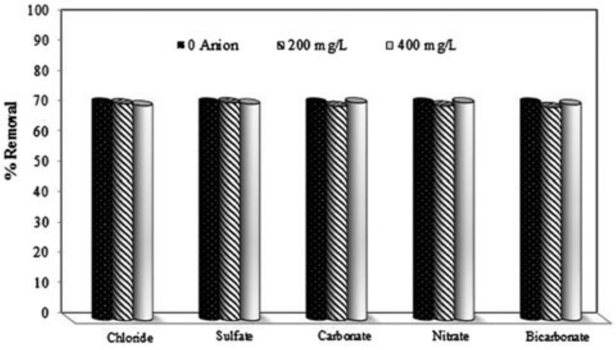

Fig. 4. Effect of competing anions concentration (200 and 400 mg/L) on fluoride biosorption (initial fluoride concentration: 8 mg/L; biosorbent dose: 8 g/L; contact time: 5 min and pH 7).

dosage were 8 mg/L and 8 g/L, respectively. As seen in Fig. 4, the results showed that the presence of competing anions did not have a significant effect on the biosorption rate of fluoride by *Padina sanctae crucis* algae.

In a similar trend, it has been reported that the presence of competing anions, including HCO_3^-, NO_3^-, Cl^-, CO_3^{2-}, and SO_4^{2-} at two concentration levels of 200 and 400 mg/L did not have a significant effect on fluoride biosorption by the shrimp shell waste [44]. Karthikeyan and Elango also found out that co-ions including Cl^-, NO_3^-, SO_4^{2-}, and HCO_3^- did not have any marked effect on the fluoride removal by aluminum-containing compounds (aluminum titanate) [59]. In another report, Maliyekkal et al. found that except SO_4^{2-} and HCO_3^- all other co-existing ions (Cl^-, NO_3^-, SiO_3^-, PO_4^{3-}, humic acid, and Ca^{2+}) have shown negligible effect on fluoride sorption by magnesia-amended activated alumina [60]. In contrast, Kumar et al. reported that the fluoride biosorption by using granular ferric hydroxide (GFH) was decreased in the presence of phosphate followed by carbonate and sulfate [61]. It has been also reported that the impact of major anions on fluoride biosorption by GFH was in order: $H_2PO_4^- > HCO_3^- > SO_4^{2-} > Cl^-$ [62]. In another study, Viswanathan et al. observed a reduction in defluoridation capacity of protonated chitosan beads by increasing the anions concentration of Cl^-, $NO_3{}^-$, SO_4^{2-}, and HCO_3^- [56].

3.7. Sorption isotherms

To quantify the sorption capacity of *Padina sanctae crucis* algae for the removal of fluoride from aqueous solutions, two frequently used isotherms namely Freundlich and Langmuir have been adopted.

The linear form of Freundlich [63] isotherm can be written as Eq. (3):

$$\log(q_e) = \log(K_f) + 1/n \log C_e \tag{3}$$

where q_e is the mass of fluoride adsorbed per unit weight of the sorbent (mg/g), K_f is the Freundlich capacity factor and a measure of biosorption capacity, $1/n$ is the equilibrium concentration of fluoride in solution (mg/L) after biosorption.

The values of $1/n$ and K_f for the sorbent were calculated from the slope and the intercept of the linear plot of $\log q_e$ vs. $\log C_e$ (Fig. 5(a)). Biosorption Freundlich isotherm parameters of fluoride onto *Padina sanctae crucis* algae are shown in Table 1.

The Langmuir biosorption isotherm [64] model is described as Eq. (4):

$$q_e = \frac{abC_e}{1 + bC_e} \tag{4}$$

And can be rewritten as Eq. (5):

$$\frac{C_e}{q_e} = \frac{1}{bq_{\max}} + \frac{1}{q_{\max}} C_e \tag{5}$$

where q_e is the mass of fluoride per unit mass of sorbent (mg/g), $q_{\max}$ is the monolayer sorption capacity, b is the Langmuir constant related to the free energy of sorption equilibrium concentration of fluoride in solution (mg/L) after biosorption.

The Langmuir constant can be specified by plotting C_e/q_e vs. C_e (Fig. 5(b)).

Biosorption Langmuir isotherm parameters of fluoride onto *Padina sanctae crucis* algae are shown in Table 1. As shown in Fig. 5(a) and (b), Freundlich model is better fitted than Langmuir model. Freundlich isotherm considers the heterogeneous biosorption surface and the possibility of multilayer biosorption.

3.8. Biosorption kinetics and modeling

Biosorption kinetics was examined in order to perceive the mechanism of fluoride biosorption on *Padina sanctae crucis* algae. For this, pseudo-first-order, pseudo-second-order, and intraparticle diffusion models were applied.

These kinetics can be represented in their nonlinear forms, as follows:

A simple pseudo-first-order kinetic model [65] can be represented as Eq. (6):

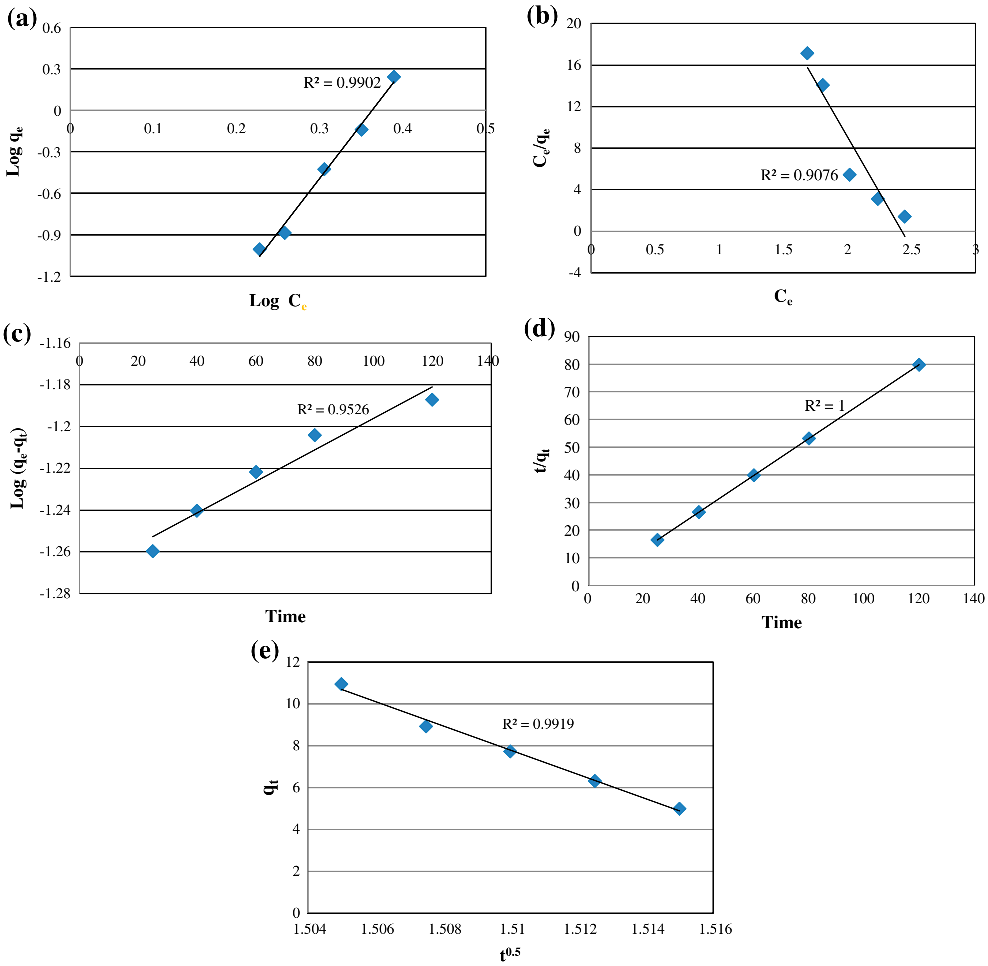

Fig. 5. (a) Freundlich, (b) Langmuir isotherms investigation of fluoride biosorption by *Padina sanctae crucis* algae, (c) Pseudo-first-order model, (d) Pseudo-second-order model of fluoride biosorption, and (e) intraparticle diffusion kinetic models by *Padina sanctae crucis* algae.

Table 1
Biosorption isotherm parameters for fluoride sorption onto *Padina sanctae crucis* algae

Isotherm	Parameter	Value
Freundlich	K_f (mg/g)	0.453
	$1/n$	7.831
	R^2	0.9902
Langmuir	b (L/mg)	0.4125
	R_L	0.2187
	Q_e (mg/g)	1.734
	R^2	0.9076

$$\log(q_e - q_t) = \log q_e - \frac{K_{1,\text{ads}}}{2.303} t \tag{6}$$

Pseudo-second-order model can be described as Eq. (7):

$$\frac{t}{q_t} = \frac{1}{q_e^2 K_{2,\text{ads}}} + \frac{1}{q_e} t \tag{7}$$

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

$$q_t = k_d t^{0.5} + C \tag{8}$$

where q_e (mg/g) and q_t (mg/g) are the amounts of sorbed fluoride on *Padina sanctae crucis* algae at equilibrium and time t (min), C is the intercept and K_1 (1/min), K_2 (g/mg min) and K_d (mg/g min$^{0.5}$) are the rate constants of pseudo-first-order, pseudo-second-order kinetic, and intraparticle diffusion models, respectively. A direct line of $\log(q_e - q_t)$ vs. t suggest the applicability of pseudo-first-order kinetic model. q_e and K_1 can be defined from the intercept and slope of

Table 2
Sorption kinetic parameters for fluoride biosorption onto *Padina sanctae crucis* algae

Model	Parameter	Value
First-order kinetic	q_e (mg/g)	0.104
	K_1 (1/min)	0.0016
	R^2	0.9697
Second-order kinetic	q_e (mg/g)	1.503
	K_2 (g/mg min)	2.647
	R^2	1
Intraparticle diffusion	K_d	0.0017
	C	1.523
	R^2	0.9919

the plot, respectively. The plot t/q_t vs. t should give a direct line if pseudo-second-order kinetics are applicable and q_e and K_2 can be defined from the slope and intercept of the plot, respectively.

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 [66].

Based on intraparticle diffusion model, the plot of q_t vs. $t^{0.5}$ should be linear ($C = 0$) if intraparticle diffusion is involved in the general mechanism of absorption and in this case the intraparticle diffusion is the only rate controlling step of the process. If this line did not through the principle ($C \neq 0$), there are intraparticle diffusion and boundary layer effects in sorption process. Whatever the intercept amount (C) increase, the effect of surface sorption in the amount controlling step increase [67].

The constants of pseudo-first-order, pseudo-second-order kinetic, intraparticle diffusion, and all other parameters of these kinetic models are shown in Table 2 and Fig. 5(c)–(e).

The plots were found linear with good correlation coefficients indicating the applicability of pseudo-second-order model in the present study. Based on the drawn plots, the experimental data were well fitted to the pseudo-second kinetic model with higher correlation coefficient (Table 2). Kinetics of fluoride biosorption by *Padina sanctae crucis* algae followed the pseudo-second model, indicating that the biosorption limiting step may be chemisorption. This suggests that the biosorption of fluoride maybe occurs via surface complexation reactions at specific biosorption sites of *Padina sanctae crucis* algae [68–70]. 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 ($C \neq 0$). This showed that both intraparticle diffusion and boundary diffusion affected the fluoride biosorption on *Padina sanctae crucis* algae.

Table 3
Comparison of fluoride sorption capacity by various biosorbent in some previous studies and present study

Biosorbent	Biosorption capacity	Refs.
Chitosan based mesoporous Ti-Al binary metal oxide supported beads	2.22	[71]
Modified chitosan-based (titanium)	7.21	[53]
Charcoal	7.88×10^{-5}	[72]
Red mud	6.28×10^{-3}	[73]
Activated carbon (*Morringa Indica*)	0.2314	[74]
Activated carbon (*Cynodon dactylon*)	4.755	[75]
Activated carbon (pecan nut shells)	2.3	[76]
Alginate (*Ulva japonica*)	39.9	[77]
Biomass carbon prepared at 300°C	0.52	[78]
Spirodela polyrrhiza	0.91	[79]
Used tea leaves	0.51	[80]
Moringa indica-based activated carbon	0.23	[74]
Powdered biomass (*Azadirachta indica* + *Ficus religiosa* + *Acacia catechu wild*)	0.04	[81,82]
Sawdust raw	1.73	[24]
Padina sanctae crucis	1.65	Present study

4. Conclusion

This study was the first attempt to evaluate the biosorption capability of the *Padina sanctae crucis* algae in removal of fluoride from aqueous solution. The results obtained in this study show that *Padina sanctae crucis* algae is an effective biosorbent for removal of fluoride from aqueous phase (Table 3). The biosorption of fluoride was influenced by the pH of the medium, initial fluoride concentration, biosorbent dose, and remained almost unchanged in the presence of other ions. The highest removal was reached only after 5 min contact time in optimum operation condition and activation temperature of biosorbent did not have a significant effect on biosorption rate. The biosorption followed the Freundlich model. The biosorption kinetics was controlled by the pseudo-second-order and pore diffusion models. Finally, it should be noted that *Padina sanctae crucis* algae can be used as an environmental friendly, effective, easy to source, and low-cost biosorbent for removal of fluoride from aqueous solution containing high level of fluoride especially in the rural and remote areas due to its easy operation.

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