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Biosorption of As(III) and As(V) from aqueous solutions by brown macroalga *Colpomenia sinuosa* biomass: kinetic and equilibrium studies

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ABSTRACT

In this research, capability of brown macroalga *Colpomenia sinuosa* for biosorption of arsenic [As(III) and As(V)] from aqueous solutions was investigated in a single-component batch system. Kinetic experiments indicated that As(III) and As(V) biosorption by *C. sinuosa* was rapid, and that 90–98% of equilibrium capacity of biosorption in 30 min was reached. Biosorption kinetics of As(III) and As(V) by the biomass at different concentrations were well described in terms of pseudo-second-order rate model ($R^2 > 0.999$ and $\varepsilon\% < 12.1\%$). In the pH range of 2–9, the optimum pH values for As(III) and As(V) biosorption were determined to be 6 and 2, respectively. The optimal pH of As(V) biosorption is highly acidic and its provision in operating the process would be difficult. Isotherm experiments were conducted in initial ion concentration range of 2–100 mg/L. The isotherm data were fitted to the Langmuir, Freundlich, Langmuir–Freundlich, and Redlich–Peterson models. Isotherm data of As(III) biosorption were found to be in the best fitness with the Freundlich–Langmuir model ($R^2 > 0.996$ and $\varepsilon\% < 4.2\%$) while the Langmuir model ($R^2 > 0.995$ and $\varepsilon\% < 5.6\%$) described the isotherm data of As(V) biosorption better than the other isotherm models. According to the Langmuir model, the maximum biosorption capacities (q_m) of As(III) and As(V) were obtained to be 95.6 and 59.9 mg/g, respectively. This study indicated that *C. sinuosa* biomass could be used as an efficient biosorbent for removal of As(III) and As(V) from aqueous environments.

Keywords: As(III); As(V); Biosorption; *Colpomenia sinuosa*; Kinetic; Isotherm

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1. Introduction

Nowadays, contamination of the aqueous environments by arsenic (As) is a worldwide environmental problem. Although this element exists in oxidation states of −3, 0, +3, and +5 in the environment, but arsenate (As^{5+} or As(V)) is the most abundant in water and only under anaerobic conditions, it might be reduced to arsenite (As^{3+} or As(III)) [1–5]. Acute exposure to high doses of arsenic is manifested in different ways including gastrointestinal symptoms, disturbance of cardiovascular, nervous systems functions, and death. Chronic exposure symptoms through arsenic-contaminated drinking water could be certain dermal lesions (somewhere it shows blackfoot

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disease), which may bring about skin cancer. Other symptoms related to arsenic-contaminated water ingestion reported are bladder and lung cancers and peripheral neuropathy disease [1,6–8]. As a consequence of these effects, World Health Organization (WHO) has assigned a provisional guideline value of 0.01 mg/L for arsenic in drinking water [1].

The conventional technologies for the removal of arsenic from water mainly include chemical precipitation, ion exchange, adsorption, and membrane processes that require high capital investment and running costs. Therefore, there is an urgent need for development of innovative but low-cost processes, where arsenic can be removed economically. The intensive research activities indicate that biosorption is one of the most promising processes for arsenic removal. This novel approach is competitive, effective, and economical [9–14].

Biosorption or bioadsorption is a process in which pollutant can uptake into dead or inactive biomass or materials extracted from biological sources in passive manner. Mechanisms of biosorption involve physical and/or chemical interaction between pollutant and functional groups of the cell wall which mainly include ion exchange, complexation, coordination, chelation, physical adsorption, and microprecipitation [12–15]. The most important advantages of biosorption are high removal efficiency, low capital and operation costs, regenerability, rapid kinetic, and low sludge generation. Biosorption has also some disadvantages including leaching dissolved organic matter, problems in regeneration, and need to granulate biomass that should be taken into more consideration [14–17].

Several studies have been reported on toxic elements removal from aqueous solution using different biosorbent materials (algae, fungi, bacteria, wheat shell, leaves and bark of trees, etc.). Among biosorbent materials, algae have received a great attention because of their cheapness and easy availability. Hansen et al. [18] used alga *Lessonia nigrescens* as biosorbent for As(V) removal from wastewater. They observed that the highest adsorption capacities in pH values of 2.5, 4.5, and 6.5 were 45.2, 33.3, and 28.2 mg/g, respectively. Naddafi and Saeedi [19] investigated biosorption of copper(II) from aqueous solutions using brown alga *Cystoseira myrica* biomass. In this research, the maximum uptake capacities of the original and protonated *C. myrica* for copper(II) were determined 97.9 and 76.9 mg/g, respectively.

The brown macroalga *Colpomenia sinuosa* is one of the abundant species in Persian Gulf that can be harvested economically, but no study has been conducted on its arsenic biosorption potentiality until now. The aim of the present study was to examine the feasibility of As(III) and As(V) removal from aqueous solutions using *C. sinuosa*. Kinetic and isotherm experiments of As(III) and As(V) biosorption were conducted and the effect of pH on uptake capacities of As(III) and As(V) was studied in a batch system.

2. Materials and methods

2.1. Biosorbent preparation

The brown macroalga *C. sinuosa* was collected from Persian Gulf on the coast of Bandar Lengeh, Iran. At first the biomass was washed with tap water and deionized water to remove sand and other impurities and then was sun dried and dried in an oven at 70°C. The biomass dried was ground by a blender. Later, the biomass was sieved in order to segregate particles with suitable size between 0.2 and 0.3 mm. Finally, the biomass again was dried at 70°C for 24 h and then used as biosorbent.

2.2. Solution preparation

All of the chemicals used in the experiments were of analytical reagent grade. As(III) and As(V) standard solutions were prepared from As_2O_3 and As_2O_5, respectively. Concentration of As(III) and As(V) in the standard solutions was adjusted to 1,000 mg/L. Initial pH of solutions was adjusted using a pH meter (Model 827 pH lab, Metrohm, Switzerland) to the desired values by using 0.1–1 M HCl and/or 0.1–1 M NaOH. Deionized double-distilled water was used for the preparation of standard and wash solutions.

2.3. Biosorption experiments

All of the biosorption experiments were conducted using both As(III) and As(V) as adsorbates inside a single-adsorbate batch system. Solution volume was 250 mL and the mixture of solution and biosorbent was agitated at 200 rpm using a rotary shaker at room temperature (20 ± 2°C). An amount of 125 mg of the biosorbent was added to each Erlenmeyer flask as experiment vessel. In this study, each experiment was duplicated and the average value was presented.

Kinetic experiments were performed in three initial concentrations of metalloid ion (As(III) or As(V)); 20, 50, and 100 mg/L. Initial pH of the solutions was adjusted to 5.0. Kinetic experiments were continued for 6 h and samples were drawn from the vessels at predetermined time intervals (5, 10, 15, 20, 30, 60, 120, 180, 240, and 360 min) for analysis. The end point of kinetic experiments was selected to be 6 h, because the results of these experiments showed that all of the experiment vessels reached to equilibrium at 4 h. The biosorption equilibrium studies were also conducted at equilibrium time determined through kinetic experiments to be 4 h. In isotherm experiments, the initial ion concentrations were in the range of 2–100 mg/L with initial pH of 5.0. Moreover, the effect of pH on the equilibrium biosorption capacities of As(III) and As(V) was investigated in the range of 2.0–9.0.

2.4. Analytical methods

After completion of each test, the mixture was filtered through 0.45 μm membrane filters (mixed cellulose ester) and the filtrate was kept for analysis of effluent arsenic level. The arsenic concentration was determined by an inductively coupled plasma/optical emission (ICP/OE) (Spectro Arcos, Germany) according to the instruction of Standard Methods [20]. To check analytical quality, standard solutions of arsenic were measured between experiment samples occasionally. In addition, reagent blanks were used for detection of any lost arsenic in samples with mixing and without biosorption process.

2.5. Calculation

The results of kinetic experiments of As(III) and As(V) biosorption were analyzed using the pseudo-first-order and pseudo-second-order rate equations given below as Eqs. (1) and (2), respectively:

$$q_t = q_e(1 - \exp^{(-k_1 t)}) \tag{1}$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \tag{2}$$

where t is time (min), q_e and q_t are the amounts of ion sorbed (mg/g) at equilibrium and at any time, respectively; k_1 is the pseudo-first-order rate constant (1/min) and k_2 is the pseudo-second-order rate constant of adsorption (g/mg min) [19,21].

In this study to describe the equilibrium state for single-ion adsorption experiments, the Freundlich, Langmuir, Freundlich–Langmuir, and Redlich–Peterson models were used. The Freundlich model is given by Eq. (3):

$$q_e = K_F C_e^{1/n} \tag{3}$$

where C_e is equilibrium concentration of ion (mg/L) and K_F and n are indicators of biosorption capacity and intensity, respectively [22–25].

The Langmuir equation is presented below by Eq. (4):

$$q_e = \frac{b q_m C_e}{1 + b C_e} \tag{4}$$

where q_m is the maximum capacity of biosorption (mg/g) and b is a constant related to the affinity of the binding sites (L/mg) [26–28].

The three-parameter Freundlich–Langmuir model was developed to improve the fitness found by the Freundlich or Langmuir model. This model is given below by Eq. (5):

$$q_e = \frac{b q_m C_e^{1/n}}{1 + b C_e^{1/n}} \tag{5}$$

where b, q_m, and n are the constants of the Freundlich–Langmuir model [29,30].

The three-parameter Redlich–Peterson model is presented below by Eq. (6):

$$q_e = \frac{K_{RP} C_e}{1 + a_{RP} C_e^{\beta}} \tag{6}$$

where K_{RP} (L/g), a_{RP} $(\mathrm{L/mg})^{\beta}$, and β (dimensionless) are the Redlich–Peterson constants [31,32].

The linear regression coefficient (R^2) and average percentage errors ($\varepsilon\%$) were calculated for determination of fitness between the experimental data and the kinetic and isotherm models. The parameter $\varepsilon\%$ is calculated by use of the following equation:

$$\varepsilon\% = \frac{\sum_{i=1}^{N} \left| \frac{q_{\exp} - q_{\text{theo}}}{q_{\exp}} \right|}{N} \times 100 \tag{7}$$

where the subscripts "exp" and "theo" show the experimental and calculated values and N shows the number of measurements [19].

3. Results and discussion

3.1. Kinetic study

Batch kinetic experiments were carried out to determine the time required for the As(III) and As(V) binding process to reach equilibrium. Kinetic profiles of As(III) and As(V) biosorption using *C. sinuosa* and their fitness with the pseudo-first-order and pseudo-second-order rate equations are presented in Figs. 1 and 2. According to Fig. 1, As(III) biosorption in 30 min at initial concentrations of 20, 50, and 100 mg/L reached to 97.6, 98.5, and 93.9% of equilibrium uptake

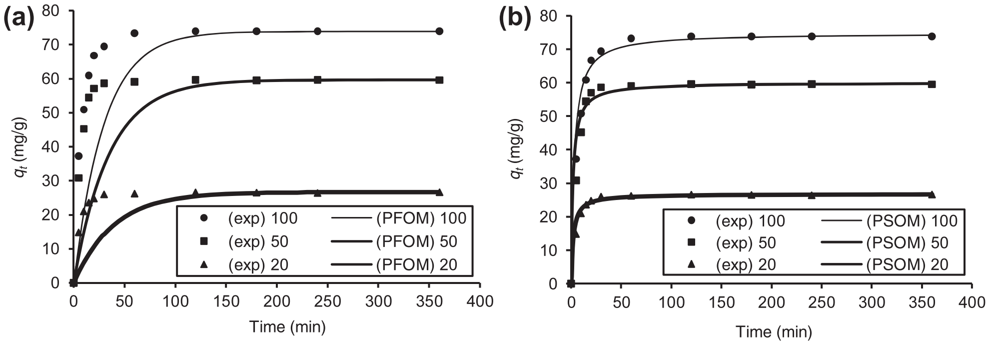

Fig. 1. Kinetic analysis of As(III) biosorption by *Colpomenia sinuosa* biomass using: (a) pseudo-first-order and (b) pseudo-second-order rate equations [experimental data (exp); pseudo-first-order model (PFOM); pseudo-second-order model (PSOM)].

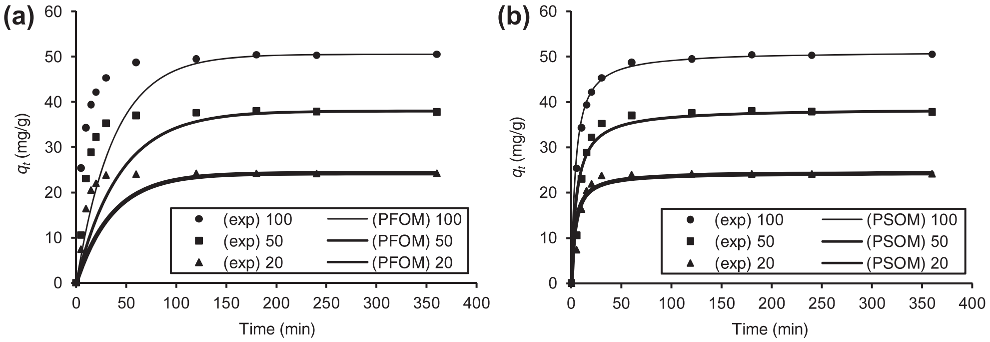

Fig. 2. Kinetic analysis of As(V) biosorption by *Colpomenia sinuosa* biomass using: (a) pseudo-first-order and (b) pseudo-second-order rate equations [experimental data (exp); pseudo-first-order model (PFOM); pseudo-second-order model (PSOM)].

capacities, respectively. The corresponding values for As(V) were obtained to be 98.4, 93.2, and 89.7% of equilibrium uptake capacities, respectively (Fig. 2). Therefore, removal of both As(III) and As(V) by the biosorbent was relatively fast and it is relatively faster for As(III).

This rapid kinetic has significant practical importance as it will reduce reactor volume ensuring efficiency and economy. Similar rapid uptake has been reported for the biosorption of As(III) and As(V) using rice polish (agricultural residue) wherein the system reached equilibrium time in 60 and 40 min for As(III) and As(V), respectively [16]. Hansen et al. [18] observed that As(V) biosorption using *L. nigrescens* reached to equilibrium after around 300 min of contact time under all of the experiment conditions. The biosorption of As(III) and As(V) by shelled *Moringa oleifera* seeds was rapid and the equilibrium approached at 60 min [33].

Kinetic parameters of the pseudo-first-order and pseudo-second-order rate equations for biosorption of As(III) and As(V) by *C. sinuosa* are given in Table 1. According to Table 1, the pseudo-second-order rate equation described kinetics of As(III) and As(V) biosorption ($R^2 > 0.999$ and $\varepsilon\% < 12.1\%$) better than the other equation. The rate constants of the pseudo-second-order rate equation for As(III) and As(V) biosorption were determined to be in the ranges of 0.004–0.018 and 0.005–0.010 g/mg min, respectively. The same results were obtained by Ranjan et al. [16], Sari and Tuzen [14], and Baig et al. [34] for biosorption of arsenic using other biomasses, so that they reported high correlation coefficients for pseudo-second-order equation in comparison to pseudo-first-order model. These observations indicate that biosorption of As(III) and As(V) can be approximated with the pseudo-second-order kinetic model.

3.2. Isotherm study

Isotherm data (statement of q_e as a function of C_e) is required for design of biosorption reactors at full scale; moreover analysis of biosorption isotherm is important to develop a model that accurately predicts

Table 1
Kinetic parameters of pseudo-first-order and pseudo-second-order rate equations for biosorption of As(III) and As(V) by *Colpomenia sinuosa* biomass

Type of ion	C_0 (mg/L)	Pseudo-first-order model				Pseudo-second-order model			
		q_e	k_1	R^2	ε%	q_e	k_2	R^2	ε%
As(III)	100	73.9	0.034	0.574	27.2	74.7	0.004	1.000	5.1
	50	59.6	0.029	0.110	31.9	60.0	0.009	1.000	6.5
	20	26.6	0.026	0.296	34.0	26.8	0.018	1.000	4.3
As(V)	100	50.5	0.027	0.797	30.7	51.2	0.005	1.000	2.1
	50	38.0	0.023	0.351	31.9	38.6	0.005	0.999	10.7
	20	24.2	0.028	0.407	29.9	24.6	0.010	0.999	12.1

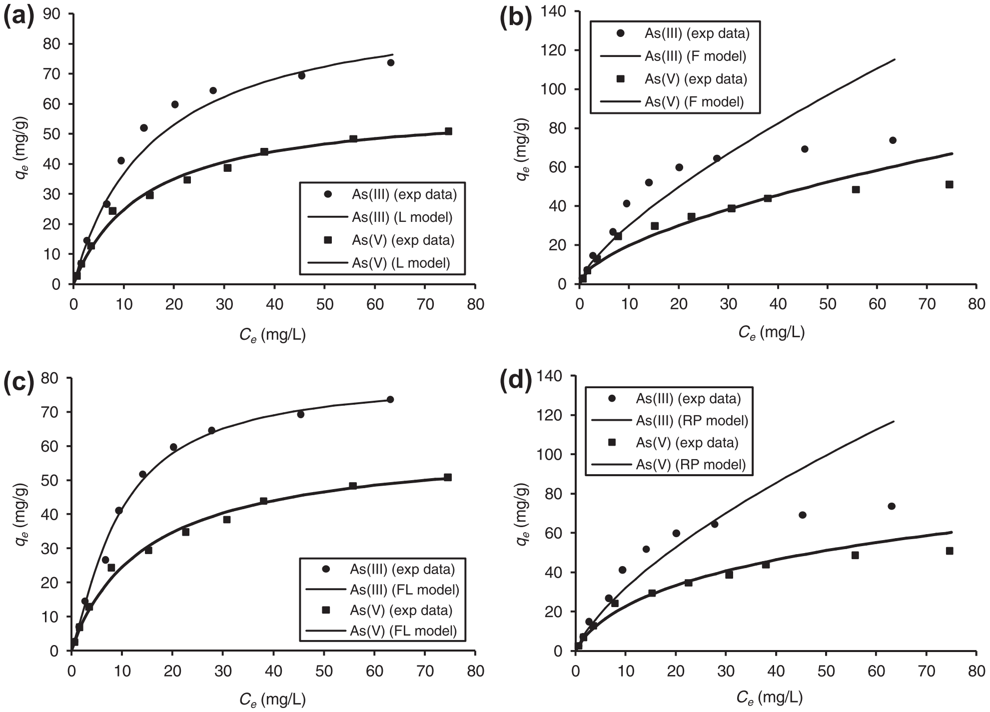

Fig. 3. Isotherm analysis of As(III) and As(V) biosorption by *Colpomenia sinuosa* biomass using: (a) Langmuir, (b) Freundlich, (c) Freundlich–Langmuir, and (d) Redlich–Peterson models [experimental data (exp data); Langmuir model (L model); Freundlich model (F model); Freundlich–Langmuir model (FL model); and Redlich–Peterson model (RP model)].

the isotherm data and that can be used for design purposes [30]. Fig. 3 shows isotherm data of As(III) and As(V) biosorption and their analysis by the Langmuir, Freundlich, Freundlich–Langmuir, and Redlich–Peterson models. Constants of these models and their correlation with biosorption of As(III) and As(V) by *C. sinuosa* biomass are given in Table 2. As indicated in Fig. 3 and Table 2, the Langmuir model described the isotherm of As(V) biosorption onto the biomass better than the other models, with high fitness ($R^2 > 0.995$ and $\varepsilon\% < 5.6\%$) while isotherm data of As(III) biosorption was determined to be in the best fitness with the Freundlich–Langmuir model ($R^2 > 0.996$ and $\varepsilon\% < 4.2\%$). Based on the linear regression coefficient and the average percentage errors, the second closest fitness with the isotherm data of As(III) and As(V) was provided by the Langmuir ($R^2 > 0.971$ and $\varepsilon\% < 12.0\%$) and Freundlich–Langmuir ($R^2 > 0.994$ and $\varepsilon\% < 5.7\%$) models, respectively. For both the As(III) and As(V), the worst fitness with the experimental data was produced by the Redlich–Peterson model ($R^2 > 0.672$ and $\varepsilon\% < 22.4\%$).

In the previous studies, several isotherm models were used for description of isotherm data of arsenic biosorption. According to Murugesan et al. [36], the isotherm data of As(III) and As(V) biosorption by pretreated waste tea fungal biomass were in good agreement with the Freundlich model. The Langmuir model described better isotherm data of As(III) and As(V) biosorption [5,14,33]. In addition, Ranjan et al.

Table 2
Isotherm parameters of Langmuir, Freundlich, Freundlich–Langmuir, and Redlich–Peterson models for biosorption of As (III) and As(V) by *Colpomenia sinuosa* biomass

Type of ion	Langmuir model				Freundlich model				Freundlich–Langmuir model					Redlich–Peterson model				
	q_m	b	R^2	$\varepsilon\%$	K_F	n	R^2	$\varepsilon\%$	q_m	n	b	R^2	$\varepsilon\%$	K_{RP}	a_{RP}	β	R^2	$\varepsilon\%$
As(III)	95.6	0.062	0.971	12.0	5.6	1.4	0.931	25.9	80.0	0.9	0.058	0.996	4.2	8.4	0.62	0.423	0.672	22.4
As(V)	59.9	0.070	0.995	5.6	4.8	1.6	0.948	17.1	61.6	1.0	0.072	0.994	5.7	5.2	0.25	0.714	0.916	9.8

Table 3
Comparison of maximum uptake capacities (q_m) of As(III) and As(V) by various biosorbent

Biosorbent	Type of biomass	Type of ion	C_0 range (mg/L)	q_m (mg/g)	Experimental conditions		Reference
					pH	T (℃)	
Sulfate-reducing bacteria	Bacterium	As(III)	–	0.2	6.5	25	Teclu et al. [35]
Waste tea fungal	Fungus	As(III)	–	1.109	7.2	30	Murugesan et al. [36]
Residue ‘rice polish’	Plant	As(III)	0.1–1.0	0.139	7.0	20	Ranjan et al. [16]
Garcinia cambogia	Plant	As(III)	50–2500	128.10	6	30	Kamala et al. [38]
Inonotus hispidus	Fungus	As(III)	10–500	51.9	6.0	20	Sari and Tuzen [14]
Moringa oleifera	Plant	As(III)	1–100	1.59	7.5	–	Kumari et al. [33]
Momordica charantia	Plant	As(III)	–	0.88	9	–	Pandey et al. [37]
Acidithiobacillus ferrooxidans BY-3	Bacterium	As(III)	0.5–3.0	0.293	4	30	Yan et al. [5]
Colpomenia sinuosa	Alga	As(III)	2–100	95.6	5	20 ± 2	This study
Sulfate-reducing bacteria	Bacterium	As(V)	–	1.76	6.5	25	Teclu et al. [35]
Waste tea fungal	Fungus	As(V)	–	4.951	7.2	30	Murugesan et al. [36]
Acid-washed crab shells	Animal	As(V)	–	8.25	2.51	–	Niu et al. [39]
Lessonia nigrescens	Alga	As(V)	50–600	45.2	2.5	20	Hansen et al. [18]
Residue ‘rice polish’	Plant	As(V)	0.1–1.0	0.147	4	20	Ranjan et al. [16]
Inonotus hispidus	Fungus	As(V)	10–500	59.6	2	20	Sari and Tuzen [14]
Moringa oleifera	Plant	As(V)	1–100	2.16	2.5	–	Kumari et al. [33]
Cod fish scale	Animal	As(V)	–	0.025	–	–	Rahaman et al. [26]
Chitosan	Biopolymer	As(V)	0.025–2	0.73	6	20	Gérente et al. [31]
Colpomenia sinuosa	Alga	As(V)	2–100	59.9	5	20 ± 2	This study

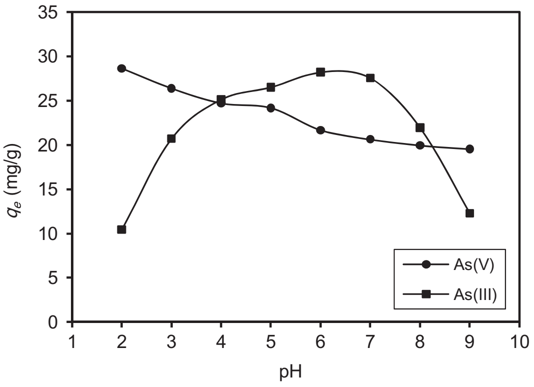

Fig. 4. Effect of pH on equilibrium capacity of As(III) and As(V) biosorption by *Colpomenia sinuosa* biomass.

[16] found that As(III) and As(V) biosorption on the agricultural residue "rice polish" were in good fitness with all of the Langmuir, Freundlich, and Dubinin–Radushkevich (D–R) isotherm models.

According to the Langmuir equation, the maximum uptake capacities (q_m) of As(III) and As(V) by the biomass were obtained to be 95.6 and 59.9 mg/g, respectively. The Langmuir parameter q_m as maximum biosorption capacity is a suitable indicator for comparing different sorbents for the same sorbate. The parameter q_m for arsenic biosorption by different biomass obtained from this research and reported in the literature along with applied experimental conditions are summarized in Table 3. Although comparison of the parameter q_m obtained in different studies is difficult as a consequence of variable experimental conditions employed, but it can be seen from Table 3 that the q_m of *C. sinuosa* biomass for both As(III) and As(V) far exceeds those of most of the other biosorbent; therefore, brown macroalga *C. sinuosa* could be classified as an efficient biosorbent for As(III) and As (V).

At full scale, the most common reactors that are employed in the adsorption process are continuous flow columns [17]. The result of this study could be used for determination of operating pH, contact time, and life time of the columns, but further investigation is also required to develop suitable methods for regeneration and granulation of the biomass.

3.3. Effect of pH on biosorption

The pH of the aqueous solution is an important controlling parameter in the biosorption process [14,16,33,37–39]. The effect of pH on As(III) and As(V) biosorption was studied in the initial pH range of 2–9. Fig. 4 shows the effect of pH on equilibrium uptake capacities of As(III) and As(V) by the biomass. As shown in Fig. 4, the maximum As(III) biosorption capacity was obtained in pH value of 6. As(III) uptake had increased sharply between pH values of 2 and 3. Kamala et al. [38] found that uptake of As(III) by fresh and immobilized *Garcinia cambogia* was not influenced by pH solution extremely while optimum biosorption took place at around pH 6–8. Pandey et al. [37] reported that increase in pH up to 9.5 elevates As(III) adsorption capacity but it was reduced with a further increment of pH. The optimum sorption of As(III) was obtained at pH 9. But Ranjan et al. [16] reported that optimum adsorption was obtained at pH 7 and in the pH range of 2–5 almost no adsorption was observed.

However, As(V) biosorption showed different behavior, as As(V) uptake onto *C. sinuosa* decreased by pH increase and was the highest at pH 2. The optimal pH of As(V) biosorption is highly acidic and its provision in operating the process at full scale would be difficult and this topic needs further research. Boddu et al. [40] concluded that the sorption capacity of chitosan for As(V) depends on pH and it was raised with a decline in pH value. Under extremely acidic condition, the biosorbent surface becomes highly protonated and strong attraction between As (V) and positively charged surface of the biosorbent results in better adsorption [3,35].

4. Conclusions

Biosorption of As(III) and As(V) from aqueous solutions by brown macroalga *C. sinuosa* biomass was examined in a batch system. Kinetic analyses demonstrated that the biosorption of both As(III) and As(V) by biomass was relatively fast. The kinetic data signified that the pseudo-second-order kinetic model well described As(III) and As(V) adsorption onto *C. sinuosa* with high fitness ($R^2 > 0.999$ and $\varepsilon\% < 12.1\%$). The optimum pH for As(III) and As(V) biosorption was determined as 6 and 2, respectively. The isotherm data of As(III) and As(V) followed the Freundlich–Langmuir and Langmuir models, respectively. The maximum uptake capacities of As(III) and As(V) obtained from the Langmuir model were 95.6 and 59.9 mg/g, respectively. The results of the present investigation revealed that *C. sinuosa* biomass could be employed for arsenic eliminating from polluted water at full scale.

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