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BIOSORPTION OF MN (II) FROM AQUEOUS SOLUTION BY *SARGASSUM HYSTRIX* ALGAE OBTAINED FROM THE PERSIAN GULF: BIOSORPTION ISOTHERM AND KINETIC

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Abstract

In the present study the biosorption of manganese (Mn (II)) onto *Sargassum hystrix* algae were assessed in a batch system by evaluation of different parameters such as biosorbent dosage (0.5-10 g/l), contact time (3-120 min) and initial manganese concentration level (0.5-100 mg/l). The maximum biosorption rate was obtained at initial Mn (II) concentration level of 10 mg/l and biosorbent dose of 10 g/l at contact time of 120 min with 85.6 percent removal efficiency. The Langmuir and Freundlich isotherms were used to describe the biosorption of Mn (II) from aqueous solution and Freundlich model was better fitted than Langmuir model. According to the results, pseudo first order model ($R^2 = 0.99$) compared to the pseudo second order and diffusion models was better for description of biosorption process. The results of this study suggested that *Sargassum hystrix* algae can be used as a low cost biosorbent for removal of Mn (II) from aqueous phase.

Keywords: Biosorption, Freundlich model, Manganese, *Sargassum hystrix* algae, Pseudo first order model, Persian Gulf.

Introduction

Organic and inorganic contaminants can be presence in the atmosphere, groundwater, sediment, and food [1-12]. From these contaminants, heavy metals are widely distributed in the environment and is hence of particular concern. Industrial

wastewater effluents contain some heavy metals such as cadmium, iron, copper, lead, zinc and manganese. These heavy metals are toxic for human and can make different diseases [13]. Manganese ions are discharged to aquatic environment from different industrial processes such as dry battery cells, glass and ceramics, electrical coils, ink and dyes [14, 15]. Mn (II) exposure is associated with damages to the nervous system function, muscle tightness, reduced learning ability and problems related to liver in children [13]. The U.S. Environmental Protection Agency (EPA) has set the permissible limit of 0.05 mg/l for Mn (II) [16] and World Health Organization (WHO) has set a maximum acceptable concentration level of 0.1 mg/l for Mn (II) [17]. Several methods such as chemical precipitation, oxidation/reduction, ion exchange, evaporate recovery, reverse osmosis, ultra filtration, electro dialysis, solvent extraction have been developed for removal of heavy metals from aqueous solution. However, these conventional methods have disadvantages in addition to their performance in the removal of heavy metals from aqueous solution such as sludge production, high cost of operation and need to large quantity of chemicals[18-21].

Another method that works well in removing heavy metals from aqueous solution is biosorption due to its high performance, low cost, reduced sludge production, biosorbent recovery, possibility of metal recovery and no need for chemicals [22]. Biosorbent are cheap and reproducible materials that are obtained naturally from different sources [23]. Yeast, inactive bacteria, sawdust, agricultural wastes and seafood processing wastes, fungus and algae are examples of biosorbents that have been used for inorganic contaminants removal from aqueous solution [24-36]. In recent years use of algae as biosorbents has increased and among different groups of algae, brown algae have been shown high performance in removal of metals[37].

The main objectives of the present study were to evaluate the effects of different parameters such as biosorbent dosage, initial Mn (II) concentration level ,and contact time in removal of Mn (II) from aqueous solution. Also in this study we investigated the biosorption isotherms as well as kinetics.

Materials and methods

Biosorbent preparation

The brown algae *Sargassum hystrix* was collected along the Persian Gulf in the Bushehr port beaches. After collection, the algae was washed three times by tap water and two times by distilled water to remove impurities, then dried in oven at 105 °C for 24h and finally powdered and sieved through a 0.71 mm screen.

Mn (II) biosorption by batch study

MnSO₄(Merck, Germany) was used as the source of metal in solutions. Mn (II) solutions were prepared at 0.5, 1, 5, 10, 20, 50, and 100 mg/l concentration levels by prepared stock solution (1000 mg/l). At each run, 100 ml of Mn (II) solution with specific initial concentration level of Mn (II) was agitated at 120 rpm. The effects of six contact times (3, 10, 20, 40, 60, and 120 min), seven initial Mn (II) concentration levels (0.5, 1, 5, 10, 20, 50, and 100 mg/l) and different doses of biosorbent (0.5-10 g/l) were investigated in the batch experiments. Atomic absorption spectrophotometer (Varian 240, Australia) was used to analyze the remaining Mn (II) concentration level in the aqueous solution after each experiment. Following equation was used to calculate the removal efficiency of biosorbent during experiments.

Eq. 1

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

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

The equilibrium biosorption capacity of *Sargassum hystrix* algae at different Mn (II) 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 Mn (II) concentration levels at initial time (mg/l); C_e is the concentration levels of Mn (II) in solution at equilibrium time (mg/l); V is the solution volume (L); and m is the biosorbent dose (g).

Results and discussion**Effect of biosorbent dose**

The biosorption studies of Mn (II) ions by *Sargassum hystrix* algae were done at room temperature, pH value of 6.5, initial concentration level of 10 mg/l Mn (II), different dose ranges of biosorbent from 0.5 to 10 g/l and different contact times (3-120 min). The effect of biosorbent dosage in the removal of Mn (II) is shown in Fig 1. As seen, in the first 3 min, biosorption of Mn (II) quickly increased and then after 60 minutes biosorption rate was almost constant. At the first 3 min, biosorption efficiency increased from 2.8% with a biosorbent dose of 0.5 mg/l to 73.64% with a biosorbent dose of 5 mg/l, this showed the high effect of biosorbent dose on biosorption efficiency. This trend is predictable because with increasing biosorbent dosage additional binding sites and surface area are available for the ions [38]. The highest removal

rate was obtained at initial concentration level of 10 mg/l Mn (II), biosorbent dose of 10 g/l and contact time of 120 min with 85.6 percent efficiency. Similar results were obtained by other studies. Khalilnezhad et al. examined removal of Mn (II) from aquatic solution by using *Penicillium camemberti* biomass at various dose in batch and fix bed reactors. They obtained maximum biosorption rate at biomass dosage under 1.5 g/l. By increasing the dosages of nano-biomass and biomass from 1 to 1.5 g/l, the removal efficiency of Mn (II) increased from 98.4% to 99.88% and 39.5% to 97.85% respectively[39]. In another study, Akpomie et al. examined adsorption of nickel(II) and manganese(II) ions from solution onto an alkaline-modified montmorillonite and observed that the removal efficiency of Ni (II) and Mn (II) ions from aqueous solution increased with increasing of adsorbent dosage[40]. Also Adeogun et al. studied comparative biosorption of Mn(II) and Pb(II) ions by raw and oxalic acid modified maize husk and found that the removal rate increased with increasing biosorbent dosage. The highest percentage of removal obtained at biosorbent dosage of 0.8 g and then biosorption efficiency decreased with increasing biosorbent dosage[41]. In another study, Dawodu et al. reported simultaneous adsorption of Ni(II) and Mn(II) ions from aqueous solution onto a Nigerian kaolinite clay. They found by increasing the adsorbent dose from 0.1 to 0.5 g, adsorption efficiency of Ni (II) and Mn (II) increased from 61-76% and 51-65% respectively[42].

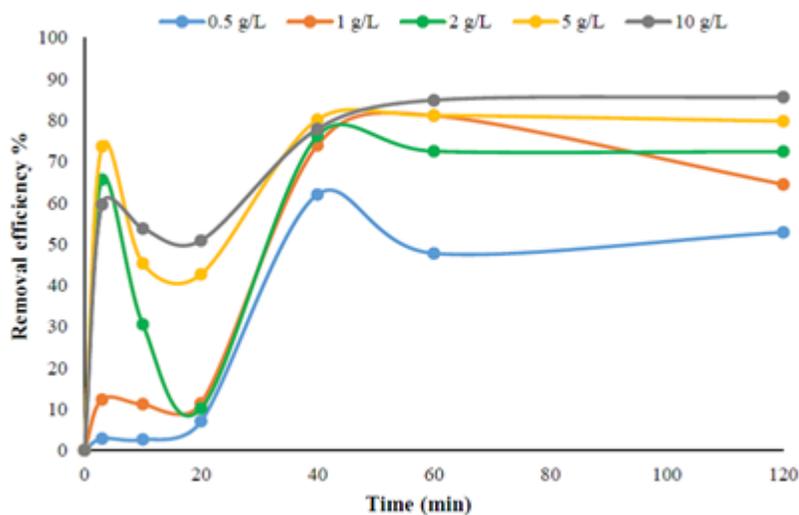


Fig 1. Mn (II) biosorption as a function of biosorbent dose (initial Mn (II) concentration: 10 mg/l, pH: 6.5).

Effect of initial concentration level of Mn (II)

The initial concentration level of metal ions plays a significant role in performance of biosorption process. The effect of initial metal concentration levels on Mn (II) biosorption from aqueous solution is shown in Fig 2. As shown, by increasing Mn (II) concentration level from 0.5 to 100 mg/l at a constant amount of biosorbent (10 g/l) biosorption

efficiency decreased. This can be due to certain and limited number of active sites of biosorbent as well as saturation of active binding sites [43, 44]. In agreement with our study, Khalilnezhad et al. reported that biosorption efficiency of Mn (II) by *Penicillium camemberti* biomass decreased with increasing concentration level of metal [39]. Also Dawodu et al. found that by increasing Mn (II) concentration levels from 100 mg/l to 300 mg/l biosorption efficiency by using an unmodified Nigerian kaolinite clay (UAK) reduced from 50 to 30% [42].

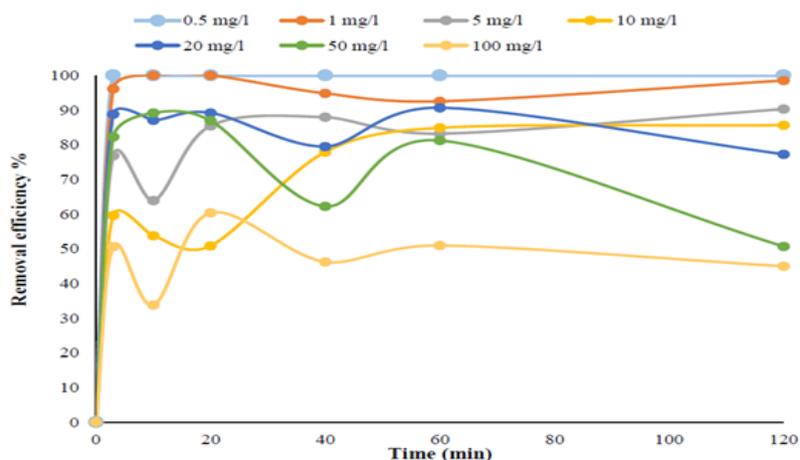


Fig 2. Mn (II) biosorption as a function of initial Mn (II) concentration (biosorbent dose: 10 g/l, pH: 6.5).

Biosorption isotherms

Two different biosorption isotherms, the Langmuir and the Freundlich isotherms, were used to determine the biosorption capacity of *Sargassum hystrix* algae for Mn (II) the removal from aqueous solutions.

Freundlich equation [45] can be represented as Eq. 3:

$$\log(q_e) = \log(K_f) + \frac{1}{n} \log C_e \quad (3)$$

Where q_e (mg/g) is the amount of adsorbed Mn (II) ions per mass of biosorbent biomass, K_f and n are Freundlich equation constants that K_f is the biosorption capacity and $1/n$ (mg/l) represents the equilibrium concentration level of Mn (II) ions.

The amounts of Freundlich equation constants can be obtained by drawing $\log q_e$ vs $\log C_e$ (Fig 3.a). Table 1 shows the values of Freundlich equation parameters of Mn (II) ions onto *Sargassum hystrix* algae.

Langmuir isotherm [46] can be written as:

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

Where q_e is the mass of Mn (II) per unit mass of biosorbent (mg/g), q_{max} is the monolayer biosorption capacity, b is the Langmuir constant (mg/l). The most important parameter of Langmuir equation is R_L that is calculated by the following equation:

$$R_L = \frac{1}{1 + bC_0} \quad (5)$$

Since the value of calculated R_L is between zero and one ($0 < R_L < 1$) represents the optimal biosorption [47]. The amounts of biosorption Langmuir isotherm parameters are shown in Table 1. As shown in Fig 3.a,b, Freundlich model describe the biosorption process better in compare with Langmuir model.

Table 1: Freundlich and Langmuir equations parameter values of manganese onto *Sargassum hystrix* algae.

Isotherm	Parameter	Value
Freundlich	K_f (mg/g)	0.375
	$1/n$	2.185
	R^2	0.995
Langmuir	b (mg/l)	0.184
	R_L	0.351
	q_{max} (mg/g)	2.664
	R^2	0.864

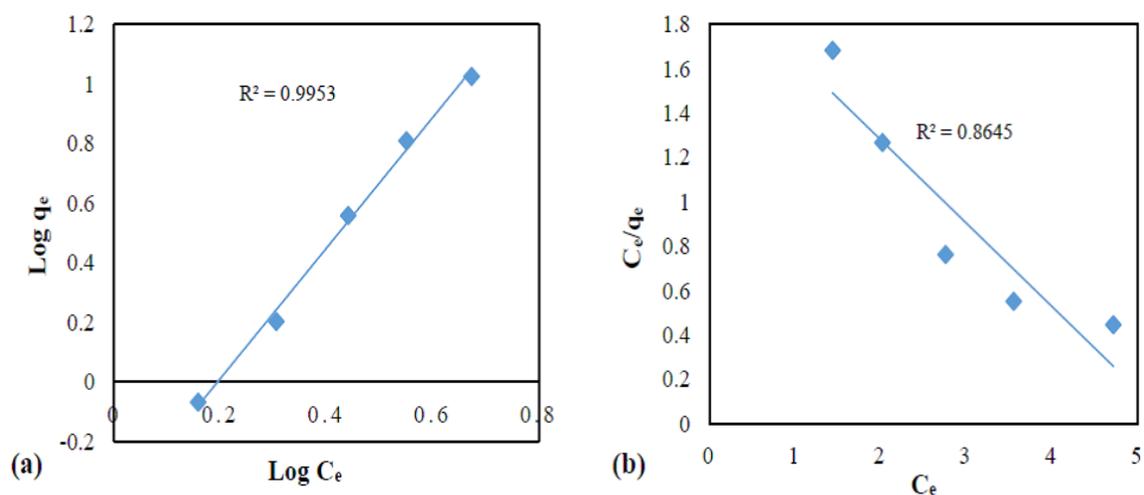


Fig 3. (a) Freundlich and (b) Langmuir isotherms investigation of Mn (II) biosorption by *Sargassum hystrix* algae.

Biosorption kinetics and modeling

Kinetic models of pseudo-first-order, pseudo-second-order and intraparticle diffusion models were used to analyze the mechanism of Mn (II) biosorption from aqueous solution.

The pseudo-first order model can be expressed as:

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

The Pseudo-second- order model can be expressed as:

$$\frac{t}{q_t} = \frac{1}{q_e^2 K_{2,ads}} + \frac{1}{q_e} t, \quad (7)$$

The intraparticle diffusion model can be represented as:

$$q_t = k_d t^{0.5} + C \quad (8)$$

where q_e is the mass of Mn (II) adsorbed per unit weight of the biosorbent (mg/g), q_{max} is the monolayer biosorption capacity, b is the Langmuir constant and K_f is the Freundlich capacity factor, $1/n$ is the equilibrium concentration level of Mn (II) in solution (mg/l) and q_t (mg/g) is the amount of biosorbed Mn (II) onto algae at 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 model.

Table 2: Biosorption kinetic parameters of manganese onto *Sargassum hystrix* algae.

Model	Parameter	Value
First-order kinetic	q_e (mg/g)	81.45
	K_1 (1/min)	4.697
	R^2	0.9992
Second-order kinetic	q_e (mg/g)	62.893
	K_2 (g/mg min)	0.00085
	R^2	0.6342
Intraparticle diffusion	K_d	0.0137
	C	0.3737
	R^2	0.2817

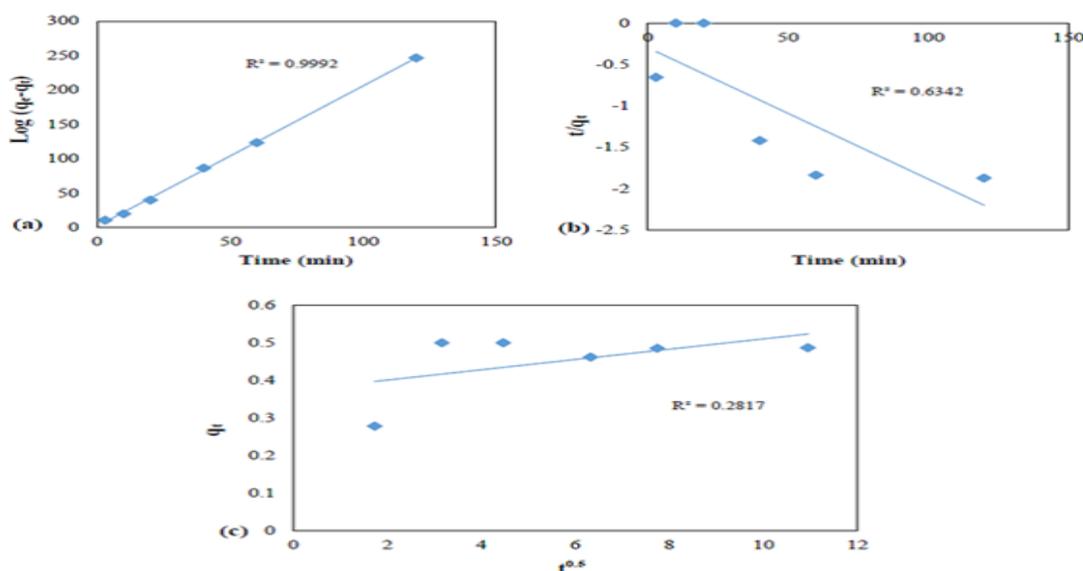


Fig 4. (a) Pseudo-first-order model (b) Pseudo-second-order model of Mn (II) biosorption and (c) intraparticle diffusion kinetic models by *Sargassum hystrix* algae.

Conclusion

The present study examined the efficiency of *Sargassum hystrix*algae in removal of Mn (II) from aqueous solutions. The biosorption studies were done in a batch system at room temperature and the effects of different variables such as initial Mn (II) concentration level, contact time and biosorbent dosage were investigated. The removal of Mn (II) by biosorbent at pH value of 6.5 increased by increasing biosorbent dose, decreasing initial concentration level of Mn (II). The biosorption followed the Freundlich isotherm. Among the biosorption kinetic models pseudo-first order kinetic model was described the biosorption process. At the end it is concluded that *Sargassum hystrix*algae can be used as a low cost and high efficient biosorbent for removal of Mn (II) from aqueous solution.

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