



Modeling of adsorption isotherm, kinetics for Cd (II) removal using brown marine algae *Sargassum myriocystum*

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Abstract

Adsorption of heavy metal Cd (II) by brown algae *Sargassum myriocystum* was examined to appraise the usefulness of biomass to eliminate heavy metals from environment. Different optimizing conditions such as contact time, biomass dosage, solution pH, temperature, and initial Cd (II) concentration was examined by batch studies. The most favorable environment was (1 h, 100, 150 and 200 rpm shaking speed, 25 °C, pH = 5 with heavy metal cadmium 50 mg/L, 100 mg/L, 150 mg/L and 200 mg/L). Several kinetic and isotherm models are used to distinguish the Cd (II) sorption method against biomass. Kinetic studies used in this biosorption method, are Pseudo first and Second order, Elovich model as well as Intra-particle diffusion models. Langmuir and Freundlich isotherms models were tested for modeling the adsorption isotherms. The separation factor R_L and surface coverage θ is both the essential parameters. The present kinetic records fit best to the Second order when compared to other models. Freundlich isotherm fit well to the experimental data, and gave the highest correlation coefficient values ($R^2 > 0.90$). This study influenced that the natural biomass *Sargassum myriocystum* showed as an effective choice along with eco-friendly adsorbent for Cd (II) ion elimination as of environment.

Keywords: Brown algae, *Sargassum myriocystum*, heavy metal, cadmium, low cost adsorbent, adsorption isotherms, kinetic studies

1. Introduction

In developing countries, due to industrial activities, we faced a set of troubles owing to offensive clearance of hazardous wastes. This resulted in rising contaminants in water bodies causing rigorous health problems even at low concentrations. Effluents discharged from various industries such as tanning, metal complex dyes, pesticides, pigments, mining, textile, fixing agents mordants, fertilizer, bleaching agents, and pharmaceuticals are mainly contributed to enhancing contaminated wastewaters all over the world. Metals are non-degradable as well as have the tendency to gather in living organisms, cause various diseases [1]. Therefore, it is important to diminish the levels of toxic metals from wastewaters before discharged into the surroundings [2]. Field crops when irrigated with wastewater contaminated by heavy metal ions are one of the main significant as well as most dangerous sources for the entry of non-biodegradable heavy metals into the human body [3]. Adsorptions of heavy metals are taking place by two different methods from the environment, chemical along with physical. On the other hand, these methods are not economically reasonable. For that reason, it is compulsory to look into low-cost successful alternatives. Therefore, a researcher shows an increased interest in the low-cost, effective alternatives named adsorbents. Numerous natural materials used for the sorption of heavy metals ions from the environment, such as plant parts, algae, fungi, bacteria, fruit peels, fruits seeds, activated charcoal, rice husk, rice straw [4, 5, 6]. Kinetics and isotherms models for the removal

of cadmium metal on seaweed were studied and described by several models. Industrial effluent discharge from various industries contains a range of organic as well as inorganic pollutants. Among these heavy metals which are toxic, carcinogenic, and harmful to humans and other living organisms [7]. Legislation in urbanized countries increasingly releases a large amount of metals in water. Maximum pollutant level in India for heavy metals be 0.05 for chromium, 0.01 for cadmium, 0.25 for copper, 0.20 for nickel, 0.80 for zinc, 0.006 for lead, 0.00003 for mercury, 0.050 for arsenic, respectively [8]. Among the different techniques, adsorption is more powerful compared to various methods and has many disadvantages such as to generate a large quantity of sludge, low competence with high cost. This method is comparatively new as well as rising as a potential choice for heavy metal removal since it gives suppleness in design, best-quality sewage treatment with reusable [9]. The USEPA classified cadmium metal as a human carcinogen and it causes toxic effects to bone and kidney [7]. Adsorption kinetics learns is important as it gives expensive insights and the process consists of four steps: Transport of metals from solution to the adsorbent, Transport of metals from the boundary to the surface of adsorbent (surface diffusion), Transfer of the metals from the surface to active sites (pore diffusion) and Adsorption by the active sites. As a result, water quality is checked, particularly at what time their metal content was high when compared to the permissible limits to make them suitable for human use. In this study, the brown seaweed *S. myriocystum*

be experienced to assess the prospective removal of metals from aqueous solutions. The rate of adsorption with an adsorption capacity of seaweed *S. myriocystum* might be the important parameters in the wastewater treatment process. Consequently, the parameters, as well as the adsorption mechanisms, were identified before adsorption. The main objective of this work is to explore the possibility to utilize *S. myriocystum* biomass for the sorption of metal cadmium.

2. Materials and Methods

2.1 Adsorption Studies

Kinetic adsorption studies with 1000 ml solutions of Cd (II) concentrations (i.e., 50, 100, 150, and 200 mg/L) were prepared as of the stock with seaweeds containing (1, 2, and 3 g). The agitation speed of the metal solution was adjusted to the desired value (150, 200, and 250 rpm) using a shaker bath. After attaining equilibrium, the seaweed samples were drawn and filtered for cadmium analysis. The percentage of cadmium removal taking place was calculated by AAS. Values are noted in duplicates, average values are noted.

2.2 Kinetics of biosorption

There is a different kinetic mechanism that controls the biosorption method, pseudo-first-order, pseudo-second-order, Elovich model, and intra-particle diffusion models.

2.2.1 Pseudo-first order

The rate constant for sorption of metals such as Cd (II) from aqueous solution onto adsorbent *S. myriocystum* was determined using the Lagergren equation [10].

$$\frac{dq_t}{dt} = k_1(q_e - q_t)$$

The above equation becomes

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303}$$

Whereas both q_e and q_t is the amount of adsorption (mg/g) at a particular time and, k_1 is the rate constant. Value k_1 and q_e can be determined as of the slope and intercept of plot $\log(q_e - q_t)$ vs t .

2.2.2 Pseudo-second order

The pseudo-second order kinetic model was described by [11]. The equation is written as

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

Where, q_e (mg/g) and q_t (mg/g) denotes the amount of metal sorbed at equilibrium and t (min) at the time, correspondingly and k_2 rate constant ($\text{g mg}^{-1}\text{min}^{-1}$). The parameters k_2 and q_e can be obtained from the curve.

2.2.3 Elovich Equation

Elovich is most appropriate for heterogeneous surfaces [12]. The model is commonly uttered as:

$$\frac{dq_t}{dt} = \alpha \exp(-\beta q_t)$$

Where, q_t amount of metal sorbed at time 't', α sorption rate ($\text{mg g}^{-1}\text{min}^{-1}$), and β desorption constant (g mg^{-1}).

2.2.4 Intra-particle diffusion rate equation

Transfer of heavy metals from the solution into adsorbent [13].

$$q = K_p t^{1/2}$$

Where K_p is diffusion rate constant and 't' time in minutes.

2.3 Sorption isotherms

In this work, Langmuir and Freundlich's isotherms were studied for the sorption of metal ions.

2.3.1 Langmuir isotherm

Langmuir isotherm was proposed originally by [14]. Langmuir isotherm equation is given as:

$$\frac{C_e}{Q_e} = \frac{1}{Q_0 b} + \frac{C_e}{Q_0}$$

Where, Q_0 and b are constants associated with monolayer sorption (mg/g) and the speed of adsorption (L/mg), respectively. The Langmuir plots obtained by plotting $1/Q_e$ vs $1/C_e$ are linear showing the applicability of Langmuir adsorption isotherm for metal adsorption using the adsorbents selected for this study. This is also evidenced by the best fit of the linear equation as seen from the correlation coefficient values (R^2).

Separation Factor (RL)

The separation factor RL and surface coverage are both the essential parameters in the Langmuir isotherm model. The separation factor (SF) is the equilibrium parameter that specifies the nature of adsorption and the shape of the isotherm. SF can be defined by the following relationship

$$\text{Separation factor } R_L = \frac{1}{1 + bC_0}$$

Where, b represents the constant and C_0 the initial cadmium concentration [15].

2.3.2 Freundlich isotherm

This equation is used for shaping the heterogeneous nature of the sorption process. The Freundlich model is given by [16]

$$Q_e = K_f C_e^{1/n}$$

Where, K_f constant (mg/g), n intensity, C_e the equilibrium amount of metal (mg/L), Q_e cadmium sorbed per gram of biomass at equilibrium (mg/g).

3. Results and Discussion

Biosorption has been reported as one of the many alternative methods that can be categorized as a green technology for heavy metal removal from industrial effluents [17]. The present study was examined and the results of the investigation are discussed below.

3.1 Kinetics of biosorption

A range of models are available to study the kinetics of biosorption and it may also help to understand the efficiency of biosorption as well as it was a great application in effluent treatment for the design and optimization of a system. The kinetics of sorption of selected heavy metals on *S. myriocystum* under a different set of experimental conditions namely range of time of contact of biosorbent and metal ions and different initial concentrations of metals was modeled using first-order kinetics, second-order kinetics, Intraparticle diffusion rate equation, and Elovich rate equation. The results obtained are discussed as follows:

3.1.1 Pseudo-first-order kinetics

The values of k_1 and q_e for the sorption of Cd (II) onto *S. myriocystum* were designed from the curves of $\log(q_e - qt)$ against the t plots (Table 1). The first-order plots of Cd (II) biosorption on to *S. myriocystum* are shown in (Fig. 1). As presented in (Table 1) and (Fig. 1) the regression coefficient ($R^2 > 0.9$) of the first-order model suggested that the q_e support the first-order model to explain the sorption kinetics of the metal ions. But it can be inferred from the tables that the calculated q_e values were very much less than the experimental q_e values for all the selected heavy metals under the selected experimental conditions. Hence it could be suggested that the adsorbent and the heavy metals are not strongly support the adsorption process. Sahnurova *et al.* [18] also indicated that first-order model was not fit well to explain the biosorption of Cd (II) onto *Enteromorpha compressa* macroalgae as a function of initial concentration and agitation rate.

3.1.2 Pseudo-second-order kinetics

The second-order curves for the sorption of Cd (II) on *S. myriocystum* are shown in (Fig. 2). For Cd (II) the curve for t/q_t versus t provides a straight line and so this kinetic is useful. The linear form of pseudo-second-order kinetics at different initial metal concentrations with the biomass at various contact time as presented in (Fig. 2) indicated the applicability of this model to explain the biosorption kinetics. As demonstrated from (Table 2) the calculated q_e in mg/g values for the biosorption of Cd (II) at initial metal ion concentration namely 50 mg, 100 mg, 150 mg, and 200 mg were found to be 41.67, 41.67, 40.0, and 40.0 respectively. The values obtained are in good concord with the corresponding q_e data namely 39.6 mg, 38.4 mg, 37.3 mg, and 36.8 mg respectively (Table 1). The correlation coefficients (R^2) were above 0.99 for the biosorption of Cd (II) heavy metal on *S. myriocystum*. The best fit of this model for the sorption of metals onto different biosorbents was reported in the earlier literature. To list a few, second-order model was most excellent for the sorption of Cd (II) with *Borassus aethiopicum* and *Cocos nucifera* [19], Cd (II) on *Cystoseira baccata* [20].

3.1.3 Elovich equation

The Elovich data obtained for the biosorption of Cd (II) from aqueous solution onto *S. myriocystum* are summarised in (Table 3) and it is graphically shown in (Fig. 3). The values of correlation coefficients (R^2) for the elovich kinetic model ranged between 0.848 and 0.974 as evident from (Table 3). From (Fig. 3) it could be suggested that the kinetic data for the elovich equation for the biosorption of Cd (II) heavy metal on *S. myriocystum* did not support the

elovich model as the R^2 values were found to be less than those obtained for the pseudo-second-order model. The Elovich model did not support the kinetic data for the biosorption of Cd (II) onto different biosorbents as reported in the earlier literature. To list a few, Elovich model was not fitted for the biosorption kinetics of Cd (II), Cr (III) and Pb (II) onto the olive stone [21].

3.1.4 Intra-particle diffusion model

The rate constant K_p and regression coefficients (R^2) for various contact time of metals biosorption using an aqueous solution to the biosorbents were obtained from the slope of time (\sqrt{t}) with quantity of biosorbent sorbed (q). The values of K_p obtained in this study for the biosorption of Cd (II) are given in (Table 4). It is evident from table that R^2 ranged between 0.787 and 0.960 for intraparticle diffusion model. The regression standards for Cd (II) were found to be low. The linear plots against Cd (II) as shown in (Fig. 4) did not pass through the origin. The variation in curve is the differences in the stages of the biosorption [22]. The intra-particle diffusion not fitted to the data onto biosorption of cadmium onto biosorbents was reported on the earlier literature. To list a few, intra-particle diffusion was not fitted for the biosorption kinetics of Cd using water lilly (*Nymphaea ampla*) root biomass [23], Cd(II) by *Chitosanabrus precatorius* blended beads [24]. The present information in shape best to the pseudo-second-order models on sorption of Cd (II) when compared to all other selected models.

3.2 Sorption Isotherms

The purpose of sorption isotherm is extremely helpful to explain the relations to the metals and the sorbent. The constants obtain by the adsorption isotherms give main information such as surface character and affinity of sorbent [25]. The arithmetical model that illustrates the distribution of the adsorbate species among adsorbent and liquid is known as the adsorption isotherm [26]. Three Isotherm equations namely Langmuir and Freundlich Isotherm were tested in the present study.

3.2.1 Langmuir Isotherm

In order to see the validity of the Langmuir adsorption isotherm data obtained from uptake of metals from metal solutions to *S. myriocystum* the isotherm parameters are summarised in (Table 5). The standards of parameters Q_0 and K_L were considered from the plot of $1/q_e$ vs $1/C_e$ for Cd (II). The linear plot of $1/q_e$ vs $1/C_e$ obtains shows the monolayer exposure of metals to the surface of biosorbent *S. myriocystum*. The resulting sorption isotherm curves are illustrated with (Fig. 5a to Fig5 c). The sorption capacities (Q_0) increased to increase in sorbent dosages for Cd (II) as observed from (Table 5). The correlation coefficients in case sorption of Cd (II) were in the range of 0.466-0.994.

Separation Factor (R_L)

The separation factor, R_L and surface coverage, θ are both the essential parameters in Langmuir isotherm model. Separation factor (SF) is the equilibrium parameter that specifies the nature and shape. The shape can reveal by R_L . From (Table 6) it can be revealed that the R_L values ranged between 0 and 1 for all the heavy metals selected at different sorbent dosage and agitation speed. This indicated the favourable adsorption of Cd (II) by *S. myriocystum*.

3.2.2 Freundlich isotherm

The plots obtained were linear which showed the applicability of Freundlich adsorption isotherm meant for the uptake of metals from solutions and using *S. myriocystum* as an adsorbent. The isotherms drawn for the given experimental data are given in (Fig 6a-c). The slope values (1/n) indicate the adsorption intensity and the intercept values (Kf) give an idea about the adsorption capacity. The 'n' is an observed factor so as to differ the scale and is interrelated with the sharing of metal species on algal exteriors. In common, higher 'n' values, gives stronger adsorption intensity [27]. The Freundlich constant, Kf represents the degree of metal sorption and higher values of Kf confirm an easy uptake of adsorbate from the solution. The correlation coefficient (R2) confirms the validity of Freundlich adsorption isotherm with the experimental data [28].

From Table 7, it must clear that the Freundlich model fit well to the experimental data, and gave the highest correlation coefficient values ($R^2 > 0.90$) for the selected metals Cd (II). A linear regression of the experimental results from Cd (II) fitted better to Freundlich rather than Langmuir isotherms. An extremely high R2 value of Freundlich isotherms for all metal sorptions showed that sorption of metal ion cadmium had multilayer coverage on algae surface. The resulting sorption isotherm curves are illustrated with Fig 6a to Fig 6c. It indicated the multilayer biosorption nature of these metal ions on *S.myriocystum*. In the present study the intensity of adsorption (n) values were greater than unity which signifies that the forces between the Cd(II) and adsorbent surface are attractive which leads to the favourable adsorption and adsorption process was found to be physical in nature. The best fit of Freundlich isotherm for the biosorption of metals onto different biosorbents was reported on the earlier literature. Freundlich isotherm was best fitted for the biosorption of Cd (II) onto *Turbinaria conoides* [29].

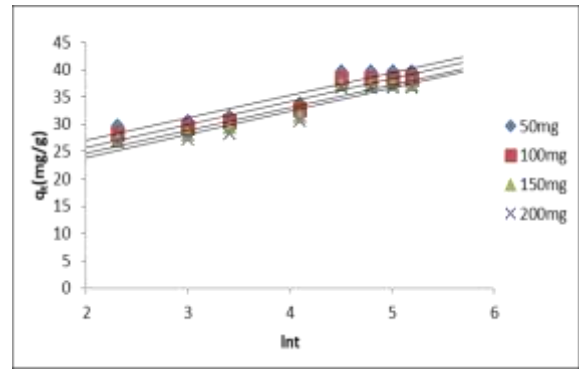


Fig 3: Elovich model for Cd²⁺ biosorption onto *S. myriocystum*

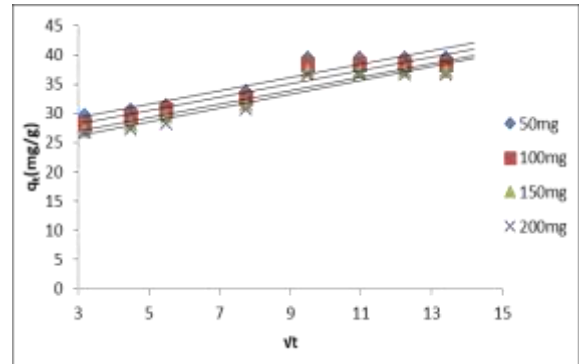


Fig 4: Intra-particle diffusion model for Cd²⁺ biosorption onto *S.myriocystum*

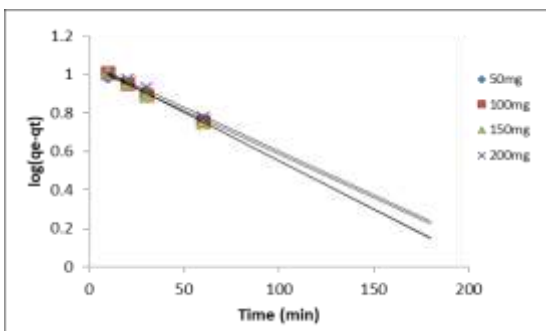


Fig 1: Pseudo-first order model for the biosorption of Cd²⁺ onto *S. myriocystum*

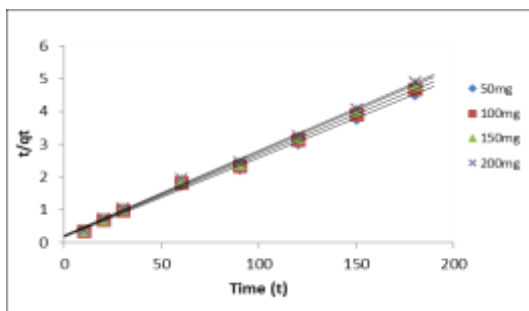


Fig 2: Pseudo-second order model for the biosorption of Cd²⁺ onto *S. myriocystum*

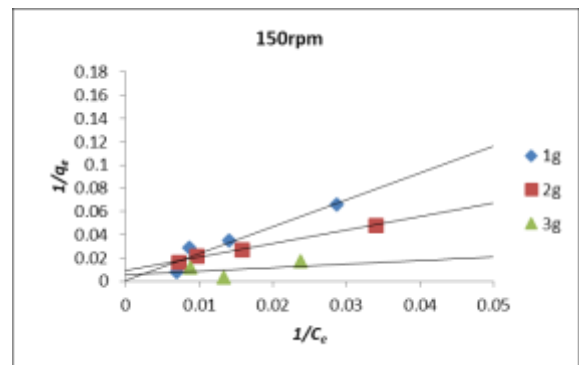


Fig 5a: Langmuir isotherms for Cd²⁺ biosorption at 150 rpm with varying initial Cd²⁺ concentration and adsorbent dose

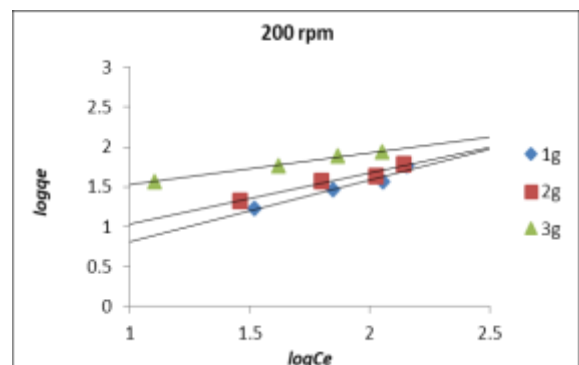


Fig 5b: Langmuir isotherms for Cd²⁺ biosorption at 200 rpm with varying initial Cd²⁺ concentration and adsorbent dose

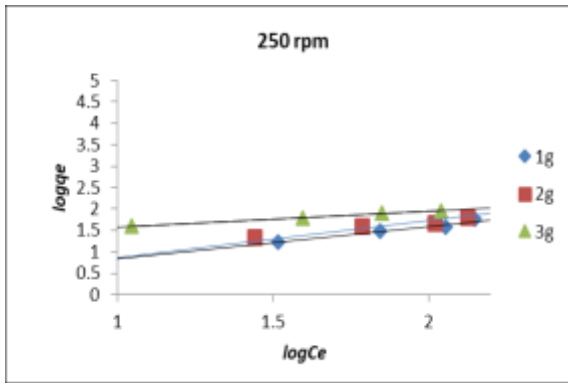


Fig 5c: Langmuir isotherms for Cd²⁺ biosorption at 250 rpm with varying initial Cd²⁺ concentration and adsorbent dose

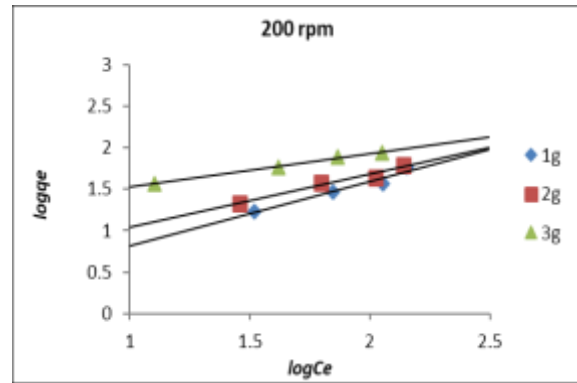


Fig 6b: Freundlich isotherms for Cd²⁺ at 200 rpm with varying initial Cd²⁺ concentration and adsorbent dose

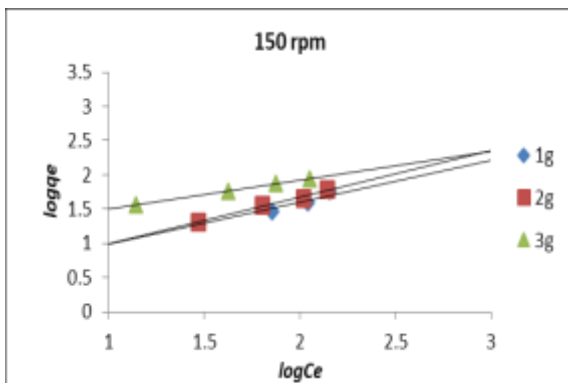


Fig 6a: Freundlich isotherms for Cd²⁺ at 150 rpm with varying initial Cd²⁺ concentration and adsorbent dose

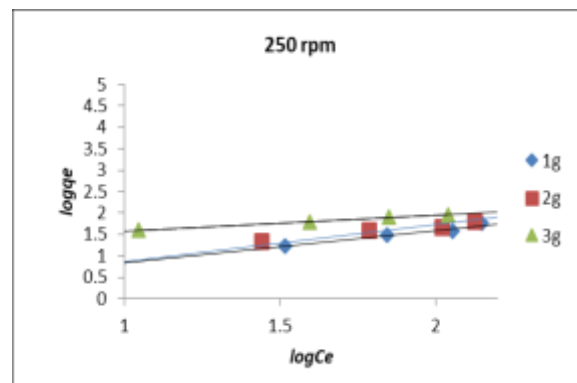


Fig 6c: Freundlich isotherms for Cd²⁺ at 250 rpm with varying initial Cd²⁺ concentration and adsorbent dose

Table 1: Pseudo-first order kinetic parameter for the biosorption of Cd (II) onto *Sargassum myriocystum*

Parameters	Initial Concentration of Cd(II)			
	50 mg/l	100 mg/l	150 mg/l	200 mg/l
Intercept log q _e	1.040	1.053	1.055	1.057
Slope (-K ₁ /2.303)	-0.004	-0.005	-0.005	-0.004
K ₁ in min ⁻¹ x10 ⁻²	9.212 x10 ⁻³	1.151 x10 ²	1.151 x10 ²	9.212 x10 ⁻³
q _e (cal)	2.829	2.866	2.872	2.877
Correlation Coefficient (R ²)	0.992	0.995	0.994	0.989

Table 2: Pseudo-second order kinetic parameters for the biosorption of Cd (II) onto *Sargassum myriocystum*

Parameters	Cd(II) (t/q _i)			
	50 mg/l	100 mg/l	150 mg/l	200 mg/l
q _e (mg/g)	41.67	41.67	40.0	40.0
K ₂ (g mg ⁻¹ min ⁻¹)	0.0032	0.003	0.0031	0.0028
h (mg g ⁻¹ min ⁻¹)	5.59	5.29	4.95	4.52
Correlation Coefficient (R ²)	0.997	0.997	0.997	0.996

Tables 3: Kinetic parameters of Elovich equation for the biosorption of Cd²⁺ onto *Sargassum myriocystum*

Parameters	Initial Concentration of Cd(II)			
	50 mg/l	100 mg/l	150 mg/l	200 mg/l
Intercept 1/β lnαβ	18.74	17.42	16.25	15.40
Slope 1/β	4.149	4.185	4.199	4.251
Initial adsorption rate (α) (mg g ⁻¹ min ⁻¹)	4.58967E+14	5.56443E+13	7.38151E+12	2.20115E+12
Desorption constant β (g mg ⁻¹)	0.241	0.2389	0.2381	0.2352
Correlation Coefficient (R ²)	0.900	0.915	0.915	0.893

Table 4: Intra-particle diffusion model for biosorption of Cd (II) onto *Sargassum myriocystum*

Time in minutes (t)	Initial concentration of Cd(II)			
	50 mg/l	100 mg/l	150 mg/l	200 mg/l
Intercept	25.89	24.69	23.54	22.71
Slope (K _p) x10 ⁻³	1.145	1.149	1.153	1.176
Correlation Coefficient (R ²)	0.903	0.907	0.907	0.900

Table 5: Langmuir isotherm parameters for Cd (II) at 150 rpm, 200 rpm and 250 rpm with varying initial metal ion concentration and adsorbent dose

Metal ions	Agitation speed		
	150 rpm	200 rpm	250 rpm
Cd(II)	1g Q_o - 0.433 mg/g b - 0.023 L/mg R^2 - 0.906	1g Q_o - 1.519 mg/g b - 1.64 L/mg R^2 - 0.739	1g Q_o - 0.586 mg/g b - 2.86 L/mg R^2 - 0.990
	2g Q_o - 0.859 mg/g b - 1.29 L/mg R^2 - 0.994	2g Q_o - 1.769 mg/g b - 3.88 L/mg R^2 - 0.649	2g Q_o - 1.070 mg/g b - 0.72 L/mg R^2 - 0.847
	3g Q_o - 12.048 mg/g b - 0.17 L/mg R^2 - 0.777	3g Q_o - 4.255 mg/g b - 3.55 L/mg R^2 - 0.792	3g Q_o - 6.666 mg/g b - 0.11 L/mg R^2 - 0.466

Table 6: Separation factor parameter (R_L) for biosorption of Cd (II)

C_i mg/L	R_L for the removal of Cd(II) at 150 rpm			R_L for the removal of Cd(II) at 200 rpm			R_L for the removal of Cd(II) at 250 rpm		
	1g	2g	3g	1g	2g	3g	1g	2g	3g
50	0.4651	0.0153	0.1053	0.0120	0.0051	0.0056	0.0069	0.0270	0.1538
100	0.3030	0.0077	0.0555	0.0061	0.0026	0.0028	0.0035	0.0137	0.0833
150	0.2247	0.0051	0.0377	0.0040	0.0071	0.0019	0.0023	0.0092	0.0571
200	0.1786	0.0039	0.0286	0.0030	0.0013	0.0014	0.0017	0.0069	0.0435
150	0.0079	0.0039	0.0689	0.0009	0.0025	0.0689	0.0015	0.0069	0.1242
200	0.0059	0.0029	0.0526	0.0007	0.0019	0.0526	0.0011	0.0052	0.0961

Table 7: Freundlich isotherm parameters for Cd (II) at 150 rpm, 200 rpm and 250 rpm with varying initial metal ion concentration and adsorbent dose

Metal ions	Agitation speed		
	150 rpm	200 rpm	250 rpm
Cd(II)	1g $1/n$ - 0.607 K_f - 1.474 R^2 - 0.924	1g $1/n$ - 0.775 K_f - 1.036 R^2 - 0.954	1g $1/n$ - 0.754 K_f - 1.08 R^2 - 0.949
	2g $1/n$ - 0.680 K_f - 1.576 R^2 - 0.988	2g $1/n$ - 0.641 K_f - 1.483 R^2 - 0.965	2g $1/n$ - 0.620 K_f - 1.584 R^2 - 0.957
	3g $1/n$ - 0.419 K_f - 2.948 R^2 - 0.999	3g $1/n$ - 0.397 K_f - 3.099 R^2 - 0.996	3g $1/n$ - 0.374 K_f - 3.303 R^2 - 0.997

4. Conclusion

The kinetics used for this work are pseudo-first and second order, Elovich rate equation along with the intra-particle diffusion model were used to learn the kinetic data. Among the various kinetic models tested, correlation coefficients (R^2) with experimental data obtained for all the selected heavy metal ions supported the best fit of the pseudo second order model for adsorption. Biosorption isotherms are the mathematical models that explain the relationship between the adsorbent and adsorbate. The Freundlich model better represented the sorption process of Cd (II) in comparison of the model of Langmuir. The sorption capacities (K_f) increased from increasing adsorbent dosages under all set of experimental conditions. It indicated the multilayer biosorption nature of these metal ions on *S.myriocystum*. In the present study, the intensity of adsorption (n) values were greater than unity which signifies that the forces between the Cd(II) and adsorbent surface are attractive which leads to the favourable adsorption and the process of adsorption was found to be physical in nature.

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