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Biosorption Technique for Naphthalene Removal from Aqueous Solution By Chara sp., Algae

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Keywords-Naphthalene, Chara sp., Biosorption, Algae, PAHs, Langmuir isotherm, Freundlich isotherm, Intraparticle diffusion

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1. Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) consist of two or more fused aromatic rings, and some of them are found as animal carcinogens and suspected human carcinogens. PAHs represent type of compounds obtained by means of the environment in air, soil and in water. They exist physically in coal, coal tar, crude oil, and creosote. More than one hundred PAH were known in different environment [1]. In general, PAHs have no color. Since PAHs do not dissolve easily in water and usually do not burn, they remain in the environment from 100 days to a couple of years [1]. Numerous various strains of microorganisms are able to reduce the grading of PAHs. Naphthalene, the simplest member of the PAHs, consists of two fused rings, and is a colorless compound, crystalline solid powder, balls, scales, and cakes. It possesses a vigorous aromatic odour connected with mothballs. In order to preserve the waters at acceptable levels of quality, various purification ways were made for wastewaters treatment [2]. Adsorption is widely used for removing poly aromatic hydrocarbon PAHs from wastewater, but the higher price of adsorbent promoted to search the utilization of less expensive substances such as; Bentonite [3], dry activated sludge fly ash [4], paper mill sludge [5], and chitosan [6] are some

of the sorbents investigated in previous works. Adsorption is central to a number of biological, chemical and physical processes and operation in environmental studies. The dissolved impurities adsorption from the solution has been extensively applied to purification of water, and is presently seen as a highly ranked procedure for treating the wastewater and reclaiming the water [7]. Sorption methods have manifested to be influential for removing the dissolved organic compounds, including polycyclic aromatic hydrocarbons (PAHs) [8]. Algae are an extremely big and different group of eukaryotic organisms, extending from unicellular genera like *Chlorella* to multi cellular shapes like the giant kelp, a big brown alga may increase in length up to 50 m. They are one of most adaptive and diverse group of organisms on earth. They develop themselves to adapt in polluted environment and not only that, they eventually flourish well. *Chara sp.*, it is really a multi-cellular macro-alga as shown in Figure 1 [9].

Chara sp., exists at the lowest parts of ponds, lakes, rivers, and ditches, and can make submersed beds of vegetation. The size of the individual Algae ranges from a few inches to several feet in length. *Chara sp.*, having a convolution of 6-8 branches lets that occurs from the nodes along the stem. *Chara sp.*, algae have been satisfactorily utilized as a biosorbent for

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highly dense metals [10], and their high performance for removing fabric dyes has been lately revealed [11]. The ion exchange has a major role in the mechanism of sorption in the adsorption process of highly dense metals, and the hydrophobic interactions are responsible for the alga-dye binding. In case of biosorption of phenol compounds, hydrophobic and donor acceptor interactions have been proposed to propel the sorption processes of phenol [4-5]. Therefore the present study aimed to evaluate the efficiencies of *Chara sp.*, for the removal of naphthalene from aqueous solutions. For this purpose, kinetic, isotherm and intraparticle diffusion models studies have been done.



Figure 1: *Chara sp.*, Algae

2. Biosorption Studies in Batch Systems

I. Adsorption Isotherms

The experimental adsorption isotherm of naphthalene was performed at room temperature ($25 \pm 2^\circ\text{C}$) on *Chara sp.*, alga. Adsorption isotherms were obtained at pH 7 by mixing 50 mL of naphthalene solutions of different concentrations (10–50 mg/l) with amount of the *Chara sp.*, alga (0.15 g) in conical flasks. The flasks were stirred by electrical shaker [Type: BS-21, Origin: Germany] at 175 r.p.m. The adsorbent was separated by a syringe filter 0.2 μm for the short-time experiments. The data of adsorption for naphthalene were fit into Temkin, Freundlich and Langmuir isotherms.

II. Adsorption Kinetics

The adsorption kinetics experiments by using *Chara sp.*, algae were performed by 50 ml of naphthalene solution of 30 mg/l concentration and pH 7. A 0.15 g *Chara sp.*, alga was added to a flask at temperature ($25 \pm 2^\circ\text{C}$). An electrical shaker was used to stir the flask at 175 r.p.m. An equilibrium investigation was conducted by removing the sample from the shaker at certain periods of time (30, 90, 150, 210, 240, 300, 360 and 420 minute). For every sample, a syringe filter 0.2 μm was used to separate the adsorbent for the short-time experiments to avoid potential interference from suspended scattering particles

in the UV–vis analysis. The rate of sorption was analyzed by three kinetic models, namely pseudo-first-order, pseudo-second-order and Intraparticle diffusion.

3. Adsorption Isotherm

The most widely used isotherms for water and wastewater treatment applications are the Freundlich, Langmuir and Temkin equations.

I. Isotherm Model of Langmuir

It is a simple and more widely utilized model, and considered a semi-empirical isotherm derived from the suggested kinetic mechanism [12]. This model depends upon the assumption that the adsorption occurs at certain homogeneous positions in the adsorbent, and no significant interaction exists among the adsorbed species. The adsorbent becomes saturated after one layer of the adsorbed molecules is made up on the surface of adsorbent. This model has the form:

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

Where, q_e is mg of adsorbate /g of sorbent, q_{max} (in mg/g) is capacity of adsorption, C_e (in mg/L) is concentration of equilibrium solution phase, and b (in L/mg) is coefficient of affinity, which is related to the energy of adsorption. Equation (1) can be used as a linear form as follows:

$$\frac{C_e}{q_e} = \frac{1}{q_{max}} C_e + \frac{1}{q_{max} b} \quad (2)$$

A dimensionless constant, called separation factor or equilibrium parameter (R_L) was defined by Webber and Chakkravorti [13] can be described as:

$$R_L = \frac{1}{1 + b C_0} \quad (3)$$

Where, C_0 (mg/L) is the adsorbate initial concentration.

The value of R_L indicates how favorable the isotherm is, as shown in Table (1) [14].

Table 1: Type of isotherm according to value of R_L

R_L	Isotherm Type
$0 < R_L < 1$	Favorable
$R_L > 1$	Unfavorable
$R_L = 1$	Linear
$R_L = 0$	Irreversible

II. Model of Freundlich Isotherm

This model is non-linear and based on the assumption that the system of sorption concerns with multilayer and non-homogenous surfaces, bounded sorption positions, and the interactions of potential energy [15]. Freundlich model is expressed mathematically, as shown in equation (4).

Where, K_F and n are constants

Data are normally fit to a logarithmic form of the equation, which provides a straight line with a slope of $1/n$ and an intercept equals to the value of $(\ln K_F)$. The intercept indicates approximately the capacity of sorption and the adsorption intensity slope, $1/n$. Commonly, the $(1/n)$ values between 0.1 and 1 show a promising adsorption [16]. Linearized Freundlich isotherm equation can

$$\ln q_e = \ln K_f + (1/n) \ln C_e \quad (5)$$

III. Temkin Isotherm

The Temkin isotherm can be stated by equation (6):-

$$q_e = \frac{RT}{b_T} \ln K_T C_e \quad (6)$$

Where, q_e is the amount of adsorbed naphthalene per adsorbent unit weight (mg/g), K_T is the constant of equilibrium binding (L/g) relevant to the peak binding energy, b_T is the constant of Temkin isotherm and is connected to the heat adsorption heat (kJ/mol), R is the constant (8.314 J/mol.K) of universal gas, T is the absolute temperature (K), and C_e is concentration of adsorbate in solution at equilibrium (mg/L). Equation (6) can be written in the linear form as follows:

where, constant $B = RT/b_T$.

This isotherm is based on two assumptions

1. The adsorption heat of whole number of molecules in the layer reduces linearly with coverage owing to adsorbent-adsorbate interactions.
2. The adsorption is described by a consistency spread of binding energies, reaching to almost peak binding energy [17-18].

4. Adsorption Kinetic Study

Pseudo-first-order, second-order and intraparticle diffusion models are utilized for kinetic study.

I. Pseudo first order

Pseudo-first-order equation was described by Lagergren for the oxalic acid sorption and malonic acid sorption on charcoal [19]. The model is generally

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (8)$$

Where, q_e equilibrium sorption capacity (mg/g); q_t is the sorption capacity at time t (mg/g) and k_1 is the rate constant (min^{-1}). After integration, equation (10) yields:

$$\log(q_t - q_e) = \log q_e - \frac{k_1}{2.303} t \quad (9)$$

The adsorption rate constant, k_1 can be experimentally determined by the slope of linear plots $\log(q_t - q_e)$ vs. t . Ho and McKay [20] observed through the literature that, in many situations, the pseudo-first-order equation of Lagergren [19] does not fit properly the complete range of contact time, and is usually usable during the first 20 to 30 min of sorption process.

II. The Pseudo-Second-Order

The pseudo-second-order equation based on the sorption equilibrium capacity may be expressed as [21]:

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (10)$$

Integrating equation (10) gives:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (11)$$

Where, K_2 is the pseudo second-order rate constant for adsorption (g/mg.min) which can be determined by plotting t/q_t versus t .

5. Materials and Methods

A 5.0 mg of naphthalene was dissolved in a 5 mL methanol the resulting was 1000 mg/l standard solution. From the standard solution, different naphthalene concentrations (1- 50 mg/L) were made by the dilution with distilled water. The concentration of naphthalene solution was estimated by measuring absorbance using UV spectrophotometer [Type: U.V-1100, Origin: China] at maximum wavelengths of the naphthalene ($\lambda_{\text{max}} = 275.5$ nm). The removal efficiency (R %) was calculated by Eq. (12), and the adsorption amount q_t was computed by Eq. (13).

$$R\% = \frac{C_0 - C_t}{C_0} \times 100\% \quad (12)$$

$$q_t = (C_0 - C_t)V/W \quad (13)$$

Where, q_t is amount of adsorption at time t , C_0 is primary naphthalene solution concentration, C_t is

naphthalene solution concentration at time t , and V is naphthalene solution volume, and W is *Chara sp.*, algae mass.

I. Preparation of the biosorbent (*Chara sp.*)

Chara sp., alga was collected from the Tiger's River at Baghdad University bank. Then, it was rinsed several times with deionized water, and then dried in the ambient air and kept in an oven at 60°C for 6 hrs. The dried *Chara sp.* Algae was ground in the laboratory blender and sorted by using the standard sieves range of (0.5-1 mm).

II. Characterization of *Chara sp.*

Surface morphology of the *Chara sp.*, powder was investigated using scanning electron microscopy (SEM) [Type: VEGA 3 LM, Origin: Germany] it is available in University of Technology/ Center of Nano Technology. The specific surface area of *Chara sp.*, powder was measured using instrument [Type: Q-surf 9600, Origin: USA] the instrument is available in Petroleum Research and Development Centre in Baghdad. FT-IR spectroscopy is provides bonding and the grafted functional groups for material. The analysis was carried out using FT-IR [Type: Bruker –Tensor 27, Origin: Germany] the instrument is available in the College of education for pure science Ibn Al-haitham.

6. Results and Discussion

I. Characterization of the *Chara sp.*

Surface morphological images obtained by SEM for *Chara sp.*, powder, which was used in this study, as shown in Figure 2.

The SEM images were obtained for *Chara Sp.*, powder with different magnifications consisted of a brick type, plate-like structure and how the presence of agglomerated and irregular surface particles with this result *Chara Sp.*, has porosity making it used for the adsorption of naphthalene. BET surface area and pore volume of *Chara sp.*, were measured at 1.067 (m²/g) and 0.003 (cm³/g) respectively. Figure 3 shows the FTIR spectrum of *Chara Sp.*, The existence of –OH, –COOH, C=O and C-H groups are around 3402, 2926, 1635 and 1035 cm⁻¹, respectively. Absorptions between 1585-1560 cm⁻¹ characteristics of amide bonds NH bending and CN stretching absorption bands around 2920 and 1660 cm⁻¹ correspond to characteristics of reflects amide bonds, which is the result of the influence of hydrogen bonding (C=O...H-N).

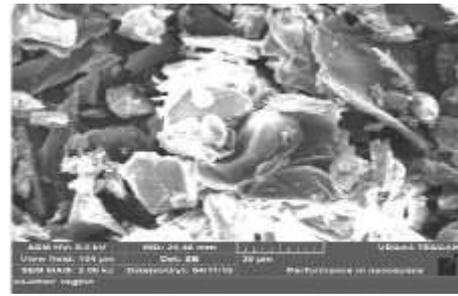


Figure 2: Scanning electron micrographs of the *Chara sp.*

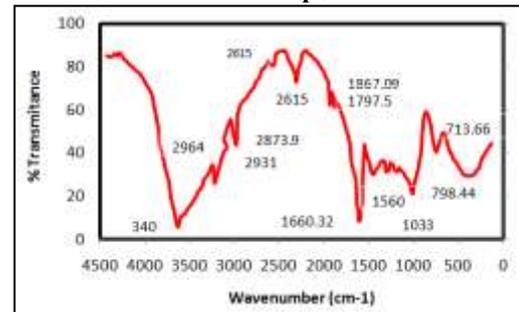


Figure 3: FT-IR Spectra of *Chara sp.*, powder

II. Effect of pH on the Biosorption Process

The results of the naphthalene removal by algae at various pH is presented in Figure 4. The highest percentage of adsorption of naphthalene by *Chara* powder was recorded at pH= 7 (45% at 50 mL of C₀=30 mg/L, dose= 0.15g, 298K⁰ and r.p.m=175). With increasing pH from 7 to 10, the percentage of naphthalene adsorption decreased from 45% to 25%, indicating that naphthalene was more effectively removed at pH = 7 by *Chara sp.*, algae powder; however, the removal efficiency decreased on either side of these pH values Figure 4. The same result has been reported by [22-23].

III. Effect of Adsorbent Dose

Experiments were done to establish an appropriate adsorbent dose for naphthalene removal Figure 5. The first concentration adsorbate imparts a significant driving force to eliminate the resistances of whole mass transfer of molecules ions between the solid phases and the aqueous [24].

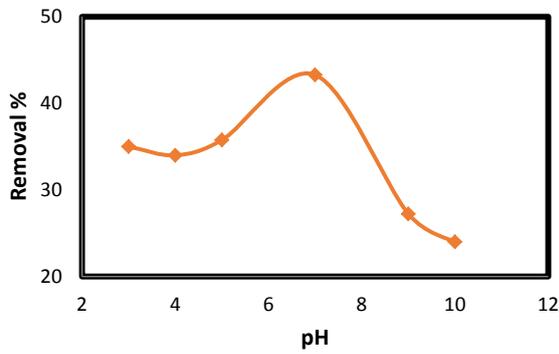


Figure 4: Effect of pH on the removal efficiency of naphthalene.

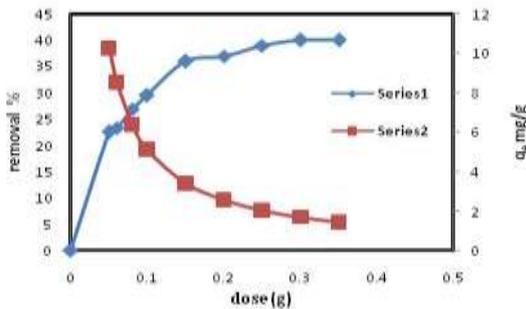


Figure 5: Effect of Chara sp., dose on adsorption of naphthalene.

The percentage of naphthalene removed increased from 25 to 40 % with increasing the amount of dose from 0.05 to 0.35 gram of *Chara sp.*, (50 mL of $C_0=30$ mg/L, 298^0 K and r.p.m=175).The adsorption increase with the amount of dose can be ascribed to the existence of larger surface area and many adsorption positions [25].

IV. Effect of Contact Time

The effect of contact time of removal of naphthalene by *Chara sp.*, at $C_0= 30$ mg/L and dose = 0.15 g is shown in Figure 6. The rate of the naphthalene is rapidly adsorption in the first 60 min and, thereafter, the adsorption rate increased gradually and the adsorption reached equilibrium at 240 min . Increasing the time of contact to 475 min revealed that the removal of naphthalene is nearly 7% beyond that found for 240 min. Since the difference in the adsorption values at 240 min and at 475 min is small, 240 min is assumed in equilibrium situation. All batch experiments were conducted at a contact time of 240 min under shaking speed of 175 rpm and pH = 7. Many of destitute surface sites are available for adsorption through the first stage, and after a certain time, the destitute surface sites that left are not easily filled because of the repulsive forces between the solute molecules on the solid and

bulk phases [26-27].This result is in agreement with [28].

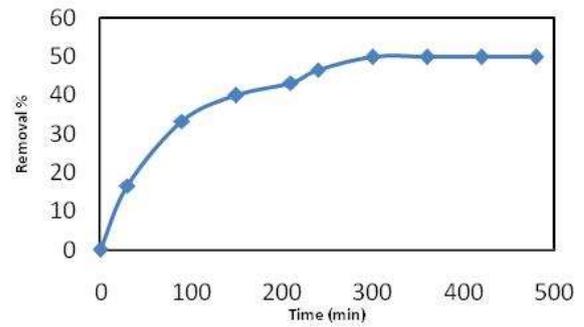


Figure 6: Effect of contact time on the removal of naphthalene by Chara Sp.,.

V. Analysis According to Isotherms

Figures 7, 8 and 9 show the analysis of the experimental data according to Langmuir, Freundlich, and Temkin isotherms model, respectively. It is obvious that the experimental data fit very well to models. Freundlich model is perhaps very convenient to demonstrate the binding of equilibrium, because the saturation point is not attained but Langmuir model yields the peak capacity of adsorption, which is a beneficial factor to compare the efficiency with that of other adsorbents. The Langmuir, Freundlich and Temkin models constants are given in Table 2.

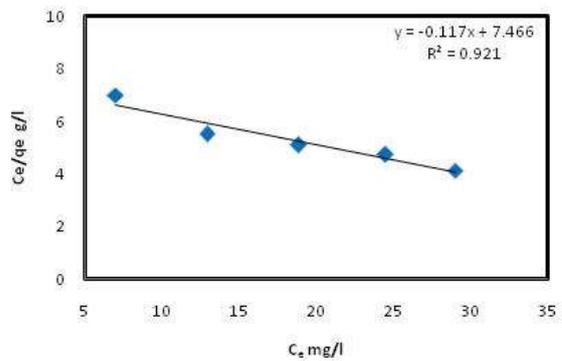


Figure 7: Langmuir Isotherm model for Chara Sp.,.

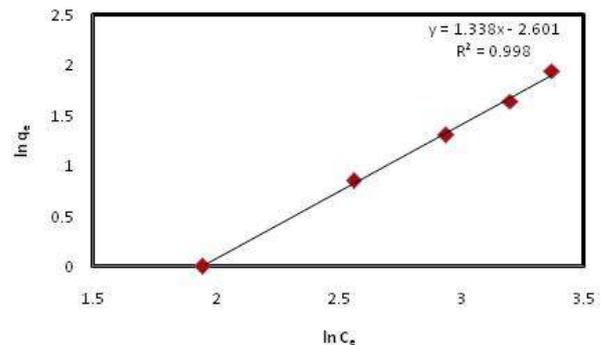


Figure 8: Freundlich Isotherm model for Chara sp., algae.

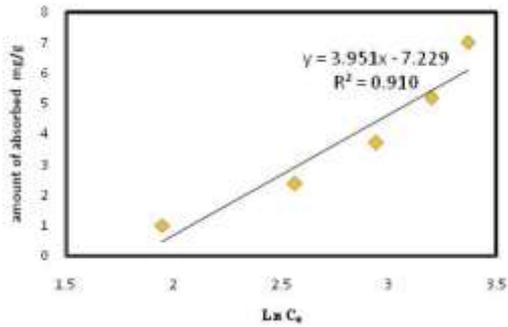


Figure 9: Temkin Isotherm model for Chara sp., algae.

VI. Kinetics

The kinetics of naphthalene adsorption on Chara algae was shown in Figures 10, 11 and 12. It can be noted from Table 4 that the values of q_e , computed from the kinetics of pseudo-second order, are almost in agreement with the experimental ones. The resulted data exhibited that the naphthalene adsorption by the *Chara sp.*, algae follows the kinetics of pseudo-second order. The obtained results from the linearized of both models for adsorption of naphthalene by *Chara sp.*, algae are listed in Table 3.

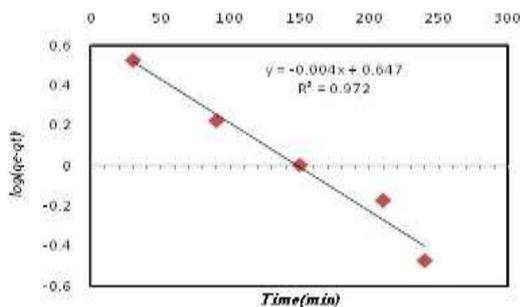


Figure 10: Pseudo-first order kinetics model for Chara sp., algae.

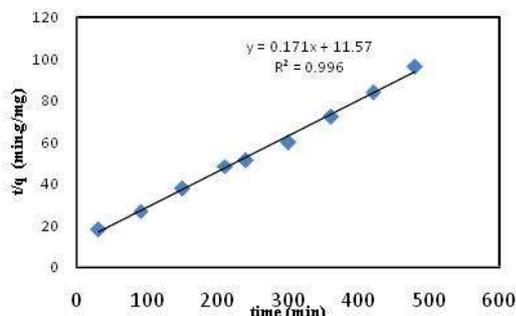


Figure 11: Pseudo-second order kinetics model for Chara sp., algae.

VII. Intraparticle Diffusion

To get information about the mechanism and rate-controlling steps affecting the kinetics of naphthalene adsorption, intraparticle diffusion model was applied to the experimental data as shown in Figure 12. The figures exhibit multi-linear plots represented by two stages for *Chara sp.*, algae, as indicated by the dotted line representing the rate of change in naphthalene adsorbed. The first stage represents a rapid increase in adsorption rate; this is due to availability of free adsorption sites on the external surface of *Chara sp.*, algae and higher concentration gradient of naphthalene. In this stage the rate of adsorption depends on the external diffusion (surface diffusion), thus as illustrated in Figure 12 the larger slope of these dotted lines in this region means higher concentration gradient. After this stage and as time passed the quantity of naphthalene adsorbed onto *Chara sp.*, algae decreased significantly, this is due to the saturation of the external sites and the decrease in naphthalene concentration gradient. In this region, naphthalene will transport from the bulk of the solution to the adsorbent external surface and then diffusion within the pores will occur, this makes the rate of adsorption slower than the first stage. Since the second portion of the plots in this figure does not have zero intercept, the intraparticle diffusion is not the only rate that controls the adsorption process. The values of K_p that are obtained from this step are calculated from the slope of the second stage line and the results are shown in Table (4). This table shows that K_p increases for *Chara sp.*, algae from 0.0794 to 0.3041 and 0.2 mg/g.min^{0.5} as the initial concentration increases from 10 to 20 and 30 mg/L respectively this is mainly due to increase in driving force as a result of increasing the concentration gradient.

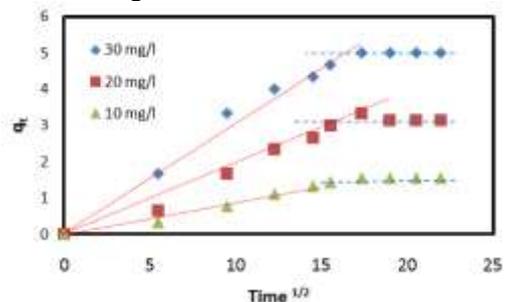


Figure 12: Weber-Morris model plots for biosorption of naphthalene.

Table 2: Isotherm parameters for naphthalene adsorbed onto Chara sp., algae.

Adsorbent	Langmuir	Freundlich	Temkin
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	q_{\max} mg/g	K_L (L/mg)	R^2	R_L	K_F $\text{mg}^{1-n} \text{g}^{-1} \text{L}^n$	1/n	R^2	K_t L/mg	B	R^2
<i>Chara sp.</i>	8.525	0.05707	0.921	0.25	13.48	1.338	0.9981	6.233	3.9511	0.9109

Table 3: Kinetic adsorption parameters obtained using pseudo-first-order and Pseudo- second order models.

Adsorbent	Pseudo first order				Pseudo second order		
	$q_{\text{exp}}^{\text{e}}$ mg/ g	$q_{\text{cal}}^{\text{e}}$ mg/ g	k_1 min^{-1}	R^2	$q_{\text{cal}}^{\text{e}}$ mg/ g	k_2 g /min mg	R^2
<i>Chara sp.</i>	5	4.4422	0.010133	0.9667	5.8	0.002529	0.9964

Table 4: Intraparticle diffusion model parameters

Adsorbent	C_0 (mg/L)	K_p (mg/g min ^{1/2})	I (mg/g)	R^2
<i>Chara sp.</i>	10	0.0794	0.0426	0.9432
	20	0.161	0.1194	0.9252
	30	0.2	0.6036	0.9206

7. Conclusion

Natural materials are one of the most promising adsorbents due to their availability and low costs. The present investigation reports batch type adsorption studies for the removal of naphthalene from aqueous solution using (*chara sp.*). Equilibrium adsorption data fitted very well to models of Langmuir, Freundlich, and Temkin isotherms. However, Freundlich isotherm model provided the best fit to the experimental data. The kinetic studies indicated that the adsorption process was extremely fast and follows the kinetics of pseudo-second order. The intraparticle adsorption model was also applied to the experimental data, and the results indicated that the intraparticle diffusion was not the only rate that controlled the adsorption process. It can be concluded that some Natural materials such as algae can be used as efficient and alternative adsorbents especially for the adsorption of organics due to easy handling readily available, and economic.

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