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Research Paper

Biosorption Process Using by Sunflower Straw (*Helianthus annuus* L.) Biosorbent for Removal Malachite Green Dye from Aqueous Solutions

Ekrem Güllüce [1,*], Elif Aslan [2] , Yaren Nur Sulanç [2], Afranur Başak [2]

[1] *Faculty of Engineering, Department of Environmental Engineering, Atatürk University, Erzurum, Turkey*
[2] *Erzurum Anatolian High School, Erzurum, Turkey*

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Abstract

The aim of this study was to investigate the malachite green dye removal by using sunflower straw biosorbent, which is agricultural waste. Batch experimental of the initial dye concentration, biosorbent dosage and stirring speed parameters have been carried out. According to these findings, it was observed that 97% of the biosorbent of the sunflower straw was removed from malachite green dye. Additionally, kinetic and isotherm studies have been carried out. According to the results of these kinetic studies, the correlation coefficient (R^2=1) of the pseudo second order kinetic model was found to be the highest. As a result of the isotherm studies, the freundlich isotherm correlation coefficient (R^2=0.983) was found to be the highest. Therefore, Sunflower straw is an effective biosorbent that can be used in dye wastewater removal, which is harmful to the environment, due to its easy availability, cheapness and sustainability.

Keywords: Sunflower straw; Biosorption; Malachite Green; Isotherm; Kinetic.

1. Introduction

Industrial wastewater is a problem in most countries. Wastewater from factories that carry out industrial activities such as textiles, cosmetics, printing and paper making are contaminated as dye [1]. This the dye residues both prevent sunlight from passing into the water and cause toxic, mutagenic, carcinogenic diseases [2].

Malachite green (MG) dye used in industries such as textile, leather and yarn have the feature of cationic *N*-methylated diaminotri-phenylmethane dye. MG dye is dangerous to human and animal health, and environmental concerns are raised if this dye is discharged without treatment [3]. Considering these situations, the MG dye should be discharged after pre-treatment is brought to the effluent limit.

Processes such as coagulation, flocculation and membrane filtration are technological methods used for dye removal. However, these processes have many disadvantages such as high costs, using high reagent of amounts and energy requirement [4].

Biosorption process is an effective method used in dye removal. The biosorption process attracts more attention due to its low cost, environmental friendliness and sustainability. In recent years, different biosorbents have been used to remove dyes in the biosorption process. It is increasing day by day to use cheap and effective materials that require less processing, such as the by-products of agriculture and food industries, which are abundant in nature, for dye removal [5].

Many of biosorbents such as mango stone biocomposite [6], almond shell residues [7], garlic straw [8], calcined mussel shells [9], cucumis sativus peel [10], sunflower seed hull [11], haloxylon recurvum plant stems [12], modified spent tea leaves [13], wheat bran [14] and *Syringa vulgaris* L. Hull [2] have been used the dye removal in the literature.

Sunflower production is abundant in Turkey. The sunflower straw formed after the production of sunflower has a large amount of environmental pollution. Burning process is generally applied to eliminate the sunflower straw. However, with the implementation of this burning process, it brings many problems. It creates serious problems such as the burning of organic substances, which have the greatest effect on the growth of the plant, the soil being suitable for erosion, and the occurrence of diseases in the roots of the plants [15].

[*] *Correspondance: Ekrem Güllüce, Faculty of Engineering, Department of Environmental Engineering, Atatürk University, Erzurum, Turkey*
E-mail: gulluceekrem@hotmail.com

The aim of this study was to make a study to remove MG dye using sunflower straw biosorbent. In this study, the effects of parameters such as initial dye concentration, adsorbent amount and stirring speed on biosorption were investigated. In order to evaluate the biosorption process, isotherm and kinetics of the biosorption process were also studied.

2. Materials and Methods

2.1. Preparation of malachite green

100 mg/L MG dye stock solution was prepared by dissolving 100 mg malachite green in 1 L distilled water. In our study, experimental studies were carried out in the range of 10-50 mg L^{-1} of MG dye from aqueous solutions. MG dye using in the study was supplied by Sigma-Aldrich. The properties of the MG dye are shown in Table 1.

2.2. Preparation of biosorbent

Sunflower straw was collected from a sunflower field in Erzurum. It was washed with plenty of tap water to remove contaminants on the surface of the sunflower straw, then rinsed with distilled water. Subsequently, the biosorbent was dried at 50 ºC for 96 hours. After drying, the biosorbent was pulverized. The powdered biosorbent was then stored in an airtight container.

2.3. Biosorption experiment studies

The initial dye concentration (10, 20, 30, 40 and 50 mg L^{-1}) and the biosorbent dosage (0.5, 1, 1.5 and 2 g) were used in the biosorption experimental studies. The effect of stirring speed (75, 100, 125 and 150 rpm) and equilibrium time was also investigated. When the experiments reached equilibrium, the samples were centrifuged. Then, the supernatant part was taken and the remaining dye concentration was measured with the help of UV spectrophotometry device. The biosorption capacity of the dye was calculated with the help of Eq. (1):

$$q_e = \frac{(C_0 - C_e)}{m} \times V \quad \text{(Eq. 1)}$$

where q_e value is the biosorption capacity at equilibrium (mg g^{-1}). C_o and C_e values are the initial concentration of MG dye (mg L^{-1}) and the concentration of dye at equilibrium (mg L^{-1}), respectively. V and m values are the solution volume (V) and the amount of Sunflower straw biosorbent (g), respectively.

2.4. Biosorption kinetic studies

In this study, the kinetics of the biosorption process was investigated in order to determine the rate of biosorption. The biosorption kinetics study was carried out in 100 ml Erlen bottles of MG dye solutions (10-50 mg L^{-1}). The bottles were periodically mixed at stirring speeds of 75, 100, 125 and 150 rpm. Samples were taken at certain time intervals and centrifuged. Subsequently, analysis of residual MG dye was performed. Kinetic studies of MG dye were performed using pseudo first order, pseudo second order and elovich kinetic models. The kinetic models used in our study (pseudo first order, pseudo second order and elovich) were calculated with the help of Eq. (2)-(4):

$$\text{In}(q_e - q_t) = \text{In}(q_e) - k_1 t \quad \text{(Eq. 2)}$$

where qt value is the biosorption capacity at time t (mg g^{-1}). k_1 is constant value of the pseudo first order (min^{-1}). In the linear graph of In(q_e-q_t) versus t, In(q_e) and k_1 are calculated from the cut-off point and from the slope point, respectively.

$$t/(q_t) = 1/(k_2 q^2_e) + t/q_e \quad \text{(Eq. 3)}$$

Linear graph of t versus (t/q_t) was drawn and values of $1/(k_2 q^2_e)$ and 1/qe was calculated from cut-off point and slope point, respectively.

$$q_t = 1/\beta \, \text{In}(\alpha\beta) + 1/\beta \, \text{Int} \quad \text{(Eq. 4)}$$

Values of α is initial desorption rate (mg (g min)$^{-1}$), value of β is desorption coefficient (g mg^{-1}). Linear graph of q_t versus In(t) was drawn. Values of ((1/β)In(αβ)) and (1/β) are calculated from cut point and slope point, respectively.

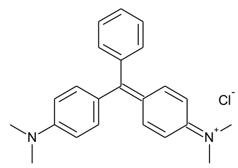
2.5. Biosorption isotherm studies

Isotherm study has been carried out to examine the interaction between adsorbate and biosorbent. In this study, the isotherm models of Langmuir, Freundlich, Elovich, Dubinin-Radushkevich (D-R) and Harkins-Jura (H-R) were used.

The linear equation of the Langmuir isotherm is given in Eq. (5). The separation factor (R_L) is given in Eq. (6).

$$\frac{1}{q_e} = \frac{1}{q_n a} * \frac{1}{C_e} + \frac{1}{q_n} \quad \text{(Eq. 5)}$$

Table 1. Properties of MG dye

Name	Molecular Weight (g/mol)	Chemical Formula	Class	Molecular Structure
Malachite green	329.46	$C_{23}H_{25}ClN_2$	Cationic *N*-methylated diamino triphenyl methane dye	

Linear graphs of (1/qe) versus (1/Ce) was drawn. Values of ($1/q_n$) and ($1/q_n a$) are calculated from cut point and slope point, respectively. Ce is initial concentration of MG dye at equilibrium (mg L^{-1}). qn is monolayer biosorption capacity (mg g^{-1}). a is the constant of the Langmuir isotherm (L mg^{-1}).

$$R_L = \frac{1}{1+aC_o} \quad \text{(Eq. 6)}$$

where C_o is initial concentration of MG dye (mg L^{-1}), a is constant of the Langmuir isotherm. Linear equation of the Freundlich isotherm is given in Eq. (7).

$$Inq_e = \frac{1}{n_f} InC_e + InK_f \quad \text{(Eq. 7)}$$

Linear graph of Inq_e versus InC_e was drawn. Values of InK_f and $1/n_f$ are calculated from cut point and slope point, respectively. K_f is constant of the Freundlich isotherm. n_f is the adsorption intensity on heterogeneous surfaces. If $n_f > 1$ indicates that adsorption is suitable. Linear equation of the Elovich isotherm is given in Eq. (8).

$$In\left(\frac{q_e}{C_e}\right) = In(K_e q_m) - \left(\frac{q_e}{q_m}\right) \quad \text{(Eq. 8)}$$

Linear graph of $In(q_e/C_e)$ versus q_e was drawn. Values of $In(K_e q_m)$ and $1/q_m$ are calculated from cut point and slope point, respectively. K_e is constant of the Elovich isotherm. q_m is maximum biosorption capacity of the Elovich isotherm (mg g^{-1}). Linear equation of Dubinin-Radushkevich (D-R) isotherm is given in Eq. (9).

$$Inq_e = Inq_d - \beta\varepsilon^2 \quad \text{(Eq. 9)}$$

Linear graph of $In(q_e)$ versus ε^2 was drawn. Values of $In(q_h)$ and β are calculated from cut point and slope point, respectively. q_d is theoretically the monolayer sorption capacity (mg g^{-1}). β is the biosorption energy coefficient ($mol^2\ j^{-2}$). Polanyi adsorption potential (ε) is calculated with the help of Eq. (10).

$$\varepsilon = RT * In\left(1 + \frac{1}{C_e}\right) \quad \text{(Eq. 10)}$$

The value of R given in the formula is the universal gas constant (8.314 J $K^{-1}mol^{-1}$). T is the absolute temperature (K). Biosorption average energy (E) is calculated with the help of Eq. (11).

$$E = \frac{1}{\sqrt{2\beta}} \quad \text{(Eq. 11)}$$

Linear equation of the Harkins-Jura (H-R) isotherm is given in Eq. (12).

$$\frac{1}{q_e{}^2} = \frac{B}{A} - \frac{LogC_e}{A} \quad \text{(Eq. 12)}$$

Linear graph of ($1/q_e^2$) versus $LogC_e$ was drawn. Values of (B/A) and (1/A) are calculated from cut point and slope point, respectively. Values of A and B are constant of Harkins-Jura (H-R) isotherm.

3. Results and Discussion

3.1. Effect of biosorbent dosage

The effect of biosorbent dosage on the biosorption capacity and biosorption removal efficiency were investigated of the initial dye concentration at 10 mg L^{-1}, 30 °C temperature and 150 rpm stirring speed during 30 minutes equilibrium time. As seen in Figure 1, the biosorption removal efficiency was 93% in the biosorbent dosage of 0.5 g, while the biosorption removal efficiency in highest biosorbent dosage of 2 g was 97%. This increase can be explained by the excessive of adsorption dosage, by increasing the surface area and keeping the dye molecules on the surface too much. While the biosorption capacity was 0.93 mg g^{-1} in the biosorbent dosage of 0.5 g, it was observed that the biosorption capacity was 0.24 mg g^{-1} in the biosorbent dosage of 2 g. The biosorption capacity decreased as the biosorbent dosage increased. This may be due to consisting in a lower surface area the partial aggregation of the biosorbent [16-18].

3.2. Effect of initial dye concentration

In this parameter, the effect of initial dye concentration on the biosorption capacity and biosorption removal efficiency was investigated by using different initial dye concentrations (10, 20, 30, 40 and 50 mg L^{-1}). As the initial dye concentration increases, the biosorption removal efficiency decreases, while the biosorption capacity increases with the increase of the initial dye concentration (Figure 2). This may be due to an increase in the initial dye concentration by creating a driving force to reduce the mass transfer resistance of MG dye between the aqueous and solid phase [19]. The highest biosorption removal efficiency of 97% was obtained at an initial dye concentration of 10 mg L^{-1}. It reached a maximum biosorption capacity of 1.15 mg g^{-1} at an initial dye concentration of 50 mg L^{-1}.

3.3. Effect of stirring speed

The effect of stirring speed on the biosorption removal efficiency of Sunflower straw biosorbent for MG dye removal was investigated using stirring speeds 75-150 rpm interval. It was observed that the biosorption removal efficiency increased with the increase of stirring speed from 75 rpm to 150 rpm (Figure 3). While adsorption removal efficiency 94% was obtained at 75 rpm stirring speed, adsorption removal efficiency 97% was obtained at 150 rpm stirring speed. By increasing the stirring speed, the Sunflower straw of the dye solution passes through the biosorbent and ensures that it clings to the surface. It may be caused by excessive fragmentation of sunflower straw biosorbent particles with increased stirring speed [2].

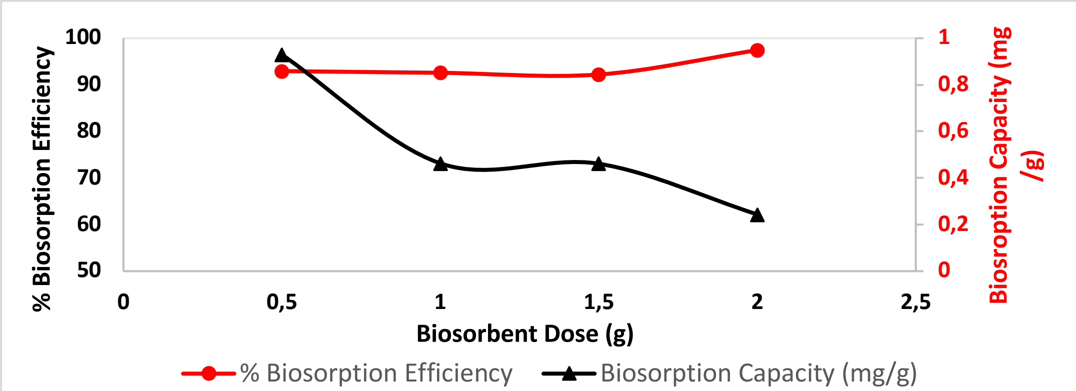


Figure 1. Effect of biosorbent dosage on biosorption capacity (mg g^{-1}) and biosorption efficiency (%) using Sunflower straw biosorbent for removal MG dye.

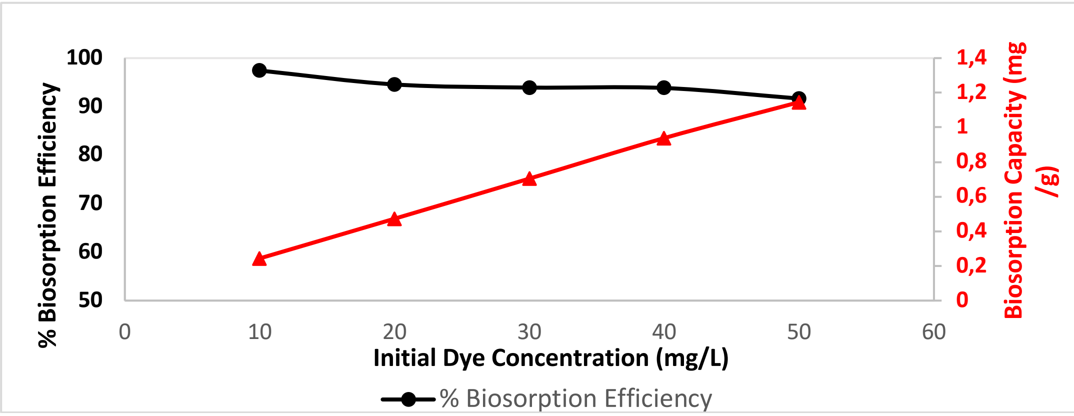


Figure 2. Effect of initial dye concentration on biosorption capacity (mg g^{-1}) and biosorption efficiency (%) using Sunflower straw biosorbent for removal MG dye.

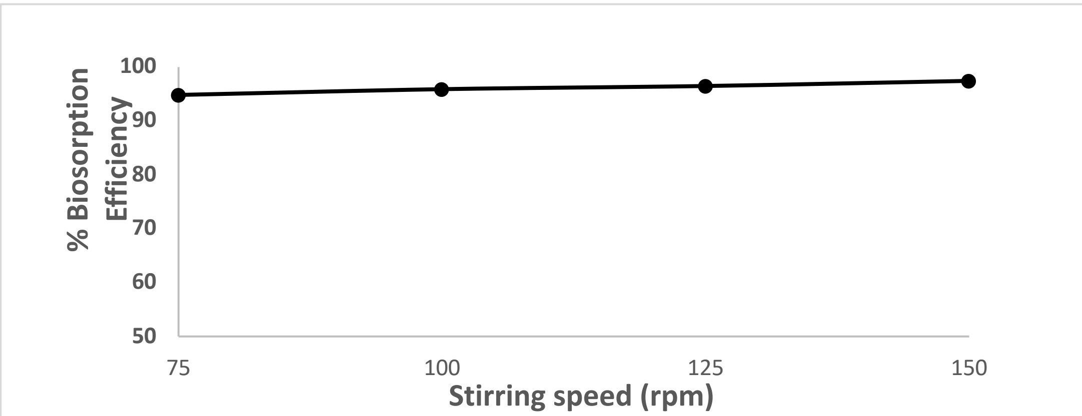


Figure 3. Effect of stirring speed on biosorption efficiency (%) using sunflower straw biosorbent for removal MG dye.

3.4. Effect of contact time

As shown in Figure 4, the effect of contact time on the biosorption removal efficiency of Sunflower straw biosorbent for MG dye removal was studied. The removal of MG dye increased within 22.5 minutes, while no significant increase was observed between 22.5-30 minutes. This can be explained by the fact that MG dye molecules surround the outer layer of the Sunflower straw biosorbent [20].

3.5. Batch kinetic studies

The purpose of the kinetic studies is to express how the adsorption mechanism occurs. In this study, we investigated the Pseudo First Order, Pseudo Second Order and Elovich Kinetics models. The values calculated using these kinetic models the equations 2-4 are shown in Table 2. As a result of these calculated values, the correlation coefficient (R^2=1) of the Pseudo Second Order kinetic model was found to be the highest. These findings showed that the q_{cal} value calcu-

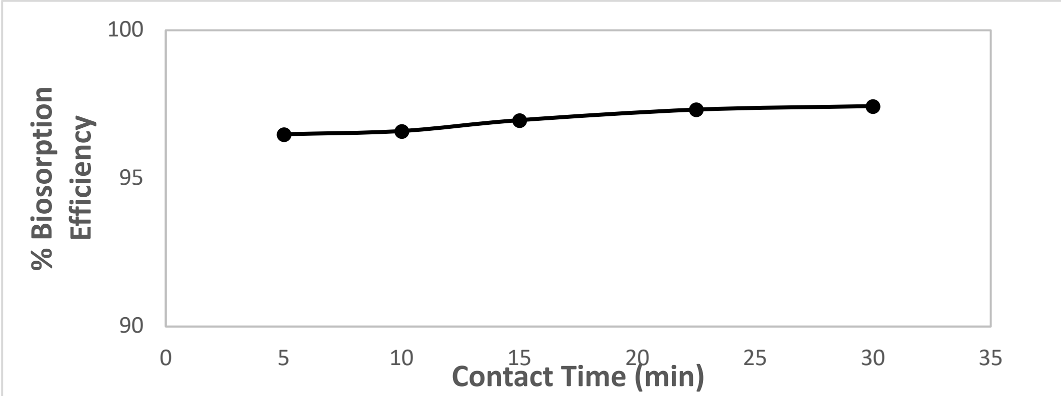


Figure 4. Effect of contact time on biosorption efficiency (%) using sunflower straw biosorbent for removal MG dye

Table 2. Kinetic parameters for Pseudo First Order, Pseudo Second Order and Elovich Kinetics models

Kinetic Models	Kinetic Parameters	Value
Pseudo First Order Kinetic	k_1	0.122
	q_{cal}	0.006
	q_{exp}	0.244
	R^2	0.916
Pseudo Second Order Kinetic	k_2	44.02
	q_{cal}	0.244
	q_{exp}	0.244
	R^2	1
Elovich Kinetic	α	1.81E+66
	β	666.67
	R^2	0.929

lated from the Pseudo Second Order kinetic model is appropriate for the q_{exp} value calculated in sunflower straw biosorbent for removal MG dye.

3.6. Batch isotherm studies

Adsorption isotherms explain the relationship between adsorbate and adsorbent at equilibrium time (Değermenci et al. 2019). In this study, Langmuir, Freundlich, Elovich, Dubinin-Radushkevich (D-R) and Harkins-Jura (H-R) isotherms were used. The results of the isotherm models are shown in Table 3. The linear graph of the isotherms Langmuir, Freundlich, Elovich, Dubinin-Radushkevich (D-R) and Harkins-Jura (H-R) is shown in Figures 5, 6, 7, 8 and 9. According to these results, the correlation coefficient (R^2=0.983) of the Freundlich isotherm was found to be the highest.

Freundlich and Harkins-Jurassic (H-R) isotherms occur on homogeneous surfaces [21]. According to these findings, it is thought to occur on the heterogeneous surface of the sunflower straw. Freundlich isotherm constant, n_f, was found to be 1.75. Since this value is n_f>1, it is seen that adsorption is suitable.

The separation factor (R_L) value was calculated using Equation 6. According to the calculated values, it was found in the range 0 <R_L<1. If the separation factor value is in this range, it means that the adsorption is suitable. The initial dye concentration is an effective parameter for finding the R_L value. It can be seen that as the initial dye concentration increases, the R_L value approaches zero (Figure 10).

4. Conclusions

In the present study, MG dye was removed from aqueous solutions using sunflower straw of biosorbent. Maximum removal efficiency was found to be 97% in MG dye removal at 10 mg / L initial dye concentration, 2 g biosorbent dosage and 150 rpm stirring speed. In order to investigate the relationship between adsorbent and adsorbate, isotherm studies were conducted using the isotherm models of Langmuir, Freundlich, Elovich, Dubinin-Radushkevich (D-R) and Harkins-Jura (H-R). According to the results of the isotherm study, Freundlich isotherm of correlation coefficient (R^2=0.983) was found to be the highest. Kinetic studies have been carried out to determine how the biosorption mechanism takes place. Pseudo First Order, Pseudo Second Order and Elovich kinetic models were used in kinetic studies. The correlation coefficient (R^2=1) of the Pseudo Second Order kinetic model was found to be the highest. As a result, it is seen that sunflower straw is a cheap and effective biosorbent for MG dye removal.

Table 3. Kinetic parameters for Langmuir, Freundlich, Elovich, Dubinin-Radushkevich (D-R) and Harkins-Jura (H-R) isotherms

Isotherm Models	Kinetic Parameters	Value
Langmuir Isotherm	q_n	1.105
	b	1.08
	R^2	0.958
Freundlich Isotherm	n_f	1.747
	K_f	0,508
	R^2	0.983
Elovich Isotherm	K_e	2.84E-3
	q_m	111.11
	R^2	0.8023
Dubinin-Radushkevich (D-R) Isotherm	β	2.05E-3
	q_d	0.94
	E	0.156
	R^2	0.871
Harkins-Jura (H-R) Isotherm	A	0.07
	B	0.5
	R^2	0.921

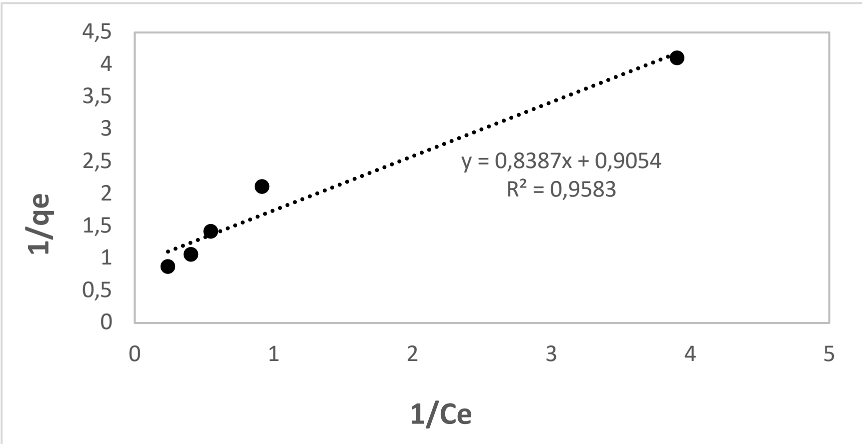


Figure 5. Linear graph of Langmuir isotherm for MG biosorption dye onto sunflower straw

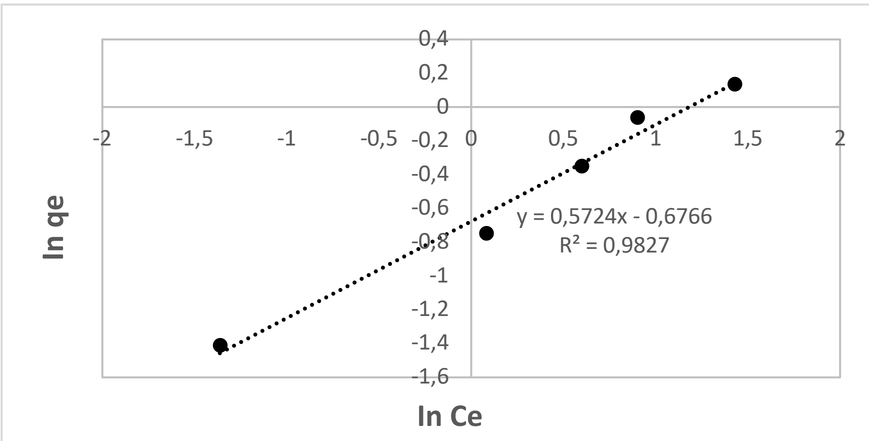


Figure 6. Linear graph of Freundlich isotherm for MG biosorption dye onto sunflower straw

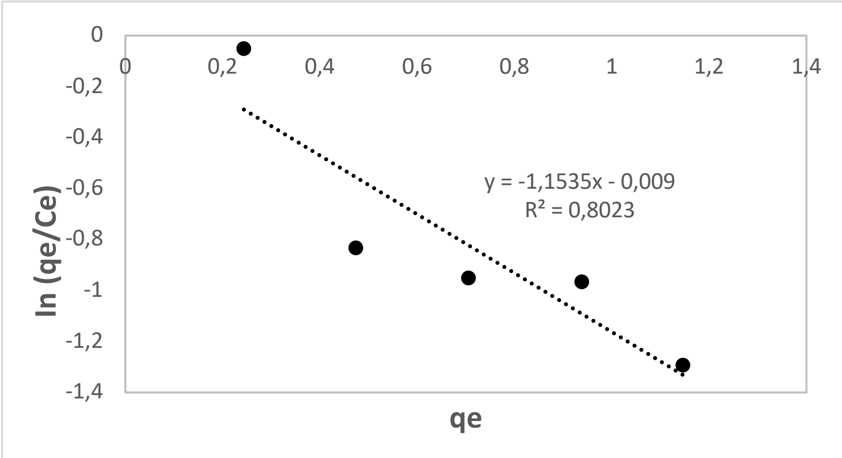


Figure 7. Linear graph of Elovich isotherm for MG biosorption dye onto sunflower straw

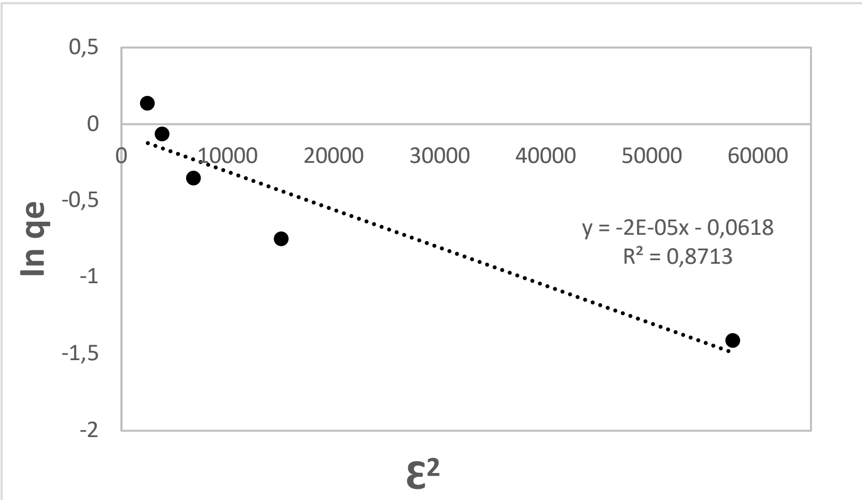


Figure 8. Linear graph of Dubinin-Radushkevich (D-R) isotherm for MG biosorption dye onto sunflower straw

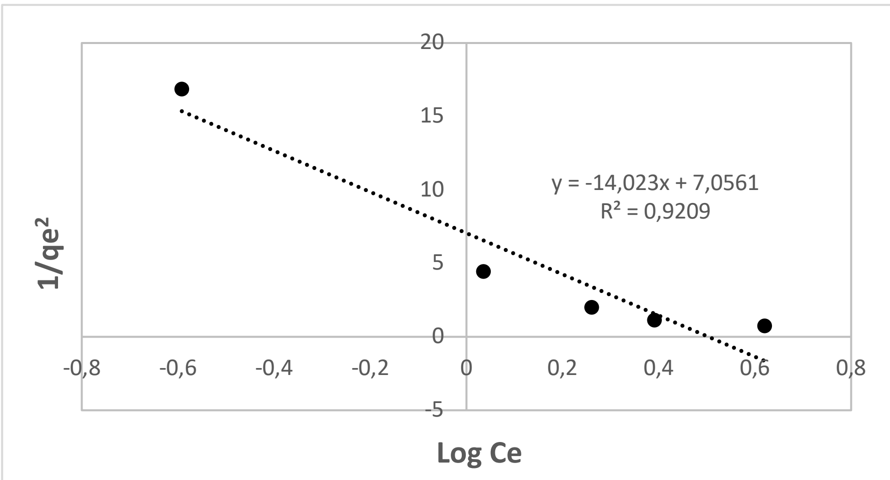


Figure 9. Linear graph of Harkins-Jura (H-R) isotherm for MG biosorption dye onto sunflower straw

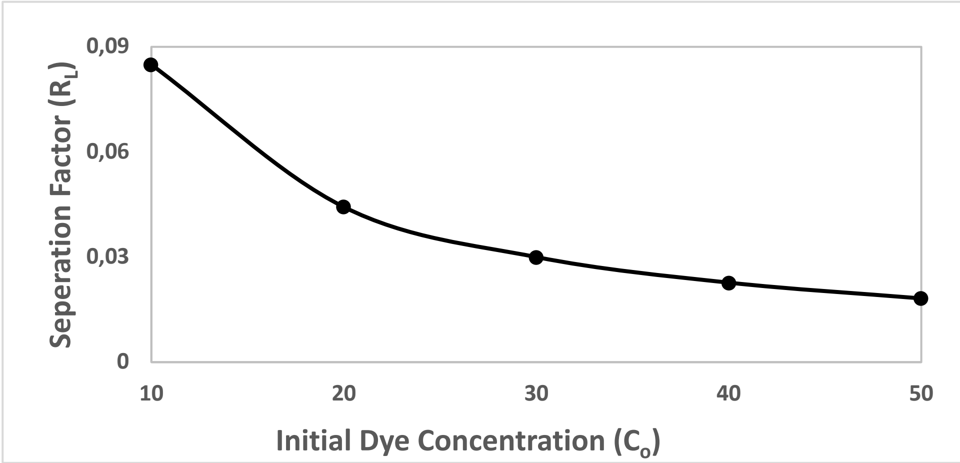


Figure 10. Variation of separation factor (RL) as a function of initial dye concentration

The correlation coefficient (R^2) of the D-R isotherm was found to be 0.8713. Biosorption average energy (E) was found to be less than 8 kj/mol. If the biosorption average energy (E) is <8 kJ/mol, the biosorption process is considered to be physical biosorption. According to these findings, it is thought to be physical biosorption.

Conflict of Interest

The authors declare no conflict of interest.

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