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ISOTHERM, KINETICS, AND THERMODYNAMIC STUDIES FOR ADSORPTION METHYLENE BLUE SOLUTION USING SHRIMP SHELL

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A B S T R A C T

This research was carried out adsorption of methylene blue dye using shrimp shell (SS) biosorbent. Methylene blue dye is commonly used in the textile and paper industries. Previously, research had been carried out on the adsorption of anionic dyes by shrimp shells, then in this study a batch adsorption was carried out to absorb methylene blue cationic dyes.It is carried out in batches with several parameters to obtain the optimum conditions. Several studies were carried out such as isotherm studies, kinetics and thermodynamics to analyze how the adsorption process occurs. The adsorption process followed the Langmuir isotherm model for the two biosorbents which showed the formation of monolayers, the kinetic model followed the second-order pseudo. The thermodynamic study of shrimp shells showed that the adsorption process was non-spontaneous, endothermic, and increased randomness. FTIR analysis shows that there are electrostatic interactions, cation exchange, and hydrogen bonding. SEM-EDX analysis shows that the pore filling of the biosorbent surface by methylene blue molecules occurs.

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INTRODUCTION

There are many environmental problems that are quite serious. One of the environmental problems that is of concern to people all over the world is water pollution and inadequate clean water. One of the most dangerous pollutants in water are synthetic dyes which contain several components or parts that can be toxic, carcinogenic, teratogenic or mutagenic to aquatic life and humans. Synthetic dyes are widely used in the textile, paper, food, rubber and printing industries. Worldwide, about 10-15% of the total dyes used in various textile and other industrial processes are then discharged into wastewater causing widespread pollution(Konicki et al., 2017).

Textile dyes in wastewater cause several chemical reactions and pose a major threat to aquatic systems even at very low concentrations. Textile dyes can cause kidney damage and reproductive disorders, even affecting the central nervous system. Therefore removal of this dye is very important. One of the most commonly used toxic dyes includes Methylene Blue. Methylene blue (methylthioninium chloride, 1876) is a dye widely used in the textile industry and this industrial waste consists of unused dyes. Even small amounts (under 1 ppm) of cationic thiazine methylene blue affect the environmental balance(Ravi & Pandey, 2019).

Various ways to overcome the problem of dye waste both physically and chemically have been widely reported. Of the several methods, adsorption is superior to other techniques for wastewater treatment in terms of initial cost, simplicity of design and ease of operation (Eletta et al., 2020). A popular technique is adsorption by natural materials which is also known as biosorption. Biosorption provides several advantages such as being environmentally friendly, highly efficient, easy to prepare, reducing organic solid waste, and does not require complicated and expensive equipment (Eletta et al., 2020).

According to the Center for Statistical Data and Information of the Ministry of Maritime Affairs and Fisheries (KKP) RI 2020, shrimp production in Indonesia reached 434,872.72 and was exported without shell and head as much as 239,282 tons. Shrimp shell waste contributes 40–66% of the weight of the shrimp, and most of it contains chitin, protein, and calcium carbonate which will interact with dye molecules(Ramadhani et al., 2020). In this research studied the isotherm, kinetics and thermodynamics of adsorption to predict the reaction mechanism that occurs during the adsorption process and the thermal stability of shrimp shells.

MATERIAL AND METHOD

Materials and Instruments

The material used in this study was shrimp shell waste (Metapenaeusmonoceros) obtained from a traditional market in Padang, West Sumatra. Dyes methylene blue (Merck), HNO_3p.a (Merck), technical NaOH (Merck), and Potassium chloride (KCl). (all chemicals used are analytical grade). FTIR (IRTracer-100-Shimadzu), SEM-EDX (Hitachi FLEXSEM 1000), UV-Vis Spectrophotometer (Genesys 20 Thermo Scientific).

Sample preparation

Shrimp shell cleaned with water, then air-dried at room temperature. The shrimp shells were then crushed and uniform in size using a 100 mesh sieve to form a powder. Shrimp shell powder was weighed ± 50 grams and then soaked for 3 hours in 0.01 M HNO_3, where the ratio of powder to HNO_3 was 1: 3. After that, it was washed with distilled water until the pH was neutral, filtered, and air-dried (Ramadhani et al., 2020).

Preparation of the dye solution

The main methylene blue solution is made by weighing 0.1 g of methylene blue with distilled water to a volume of 100 mL in a volumetric flask. The solution is diluted into several concentration variations, namely 0, 1, 2, 3, 4, and 5 mg/L for series solutions standards with a volume of 100 mL each. Furthermore, the maximum wavelength of methylene blue is obtained by scanning the wavelength from the range of 600-700 nm using a UV-Vis spectrophotometer, selecting the wavelength that shows the maximum absorbance value(Hevira et al., 2019)

Determination of methylene blue adsorption isotherm model

The adsorption isotherm models studied in this study are the Freundlich isotherm model and the Langmuir isotherm model. The adsorption isotherm parameters were obtained by calculation using data on the effect of the initial concentration of methylene blue dye on the absorption capacity. The Freundlich isotherm equation is obtained by plotting log Ce versus log qe data, while the Langmuir isotherm equation is obtained by plotting Ce versus Ce/qe data(Ramadhani et al., 2020). The Langmuir isotherm can be represented by the following equation.

$$\frac{1}{q_e} = \frac{1}{k_l q_m C_e} + \frac{1}{q_m}$$

where qm is the maximum monolayer adsorption capacity of the adsorbent (mg g^{-1}), K_L is the Langmuir adsorption constant (L mg^{-1}).

For the Freundlich model equation is expressed by the following equation.

$$\log q_e = \log K_f + \frac{1}{n} \log C_e$$

K_f and n are Freundlich constants. The Freundlich adsorption isotherm model describes a reversible and non-ideal adsorption process. In contrast to the Langmuir isotherm model, the Freundlich model is not limited to monolayer formations where its application to multilayer adsorption is possible(Zein & Suciandica, 2022).

Determination of methylene blue adsorption kinetics model

Kinetic studies were evaluated using pseudo-first-order and pseudo-second-order kinetic models. The adsorption kinetic parameters were obtained by calculating using data on the effect of contact time on the absorption capacity of methylene blue in shrimp shells. The first-order pseudo-kinetic equation is obtained by plotting data t (minutes) versus ln (qe-qt), while the second-order pseudo-kinetic equation is obtained by plotting data t (minutes) versus

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t/qt(Zein, Purnomo, et al., 2022). Adsorption kinetics was studied using pseudo first order and pseudo second order, using the following equations

Pseudo first order : $\ln (q_e - q_t) = \ln q_e - k_1 t$

Pseudo second order : $\frac{1}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$

The kinetic model and isotherm adsorption that matches the adsorption process is chosen on the model that has an R^2 value that is close to one

Adsorption thermodynamic studies

In 15 Erlenmeyer pieces 25 mL were added 0.1 g of biosorbent and 10 mL of dye solution with various concentrations of 10-50 mg/L. Adjusted the optimum pH and contact time that has been obtained previously. The temperature was varied respectively 25, 35 and 45 °C then stirred at a stirring speed of 150 rpm. The mixture was filtered and the filtrate was analyzed using a UV-Vis spectrophotometer at λmax. = 664 nm(Zein & Suciandica, 2022).The thermodynamic constant can be estimated from the Langmuir isotherm model using the following equation:

$$\Delta G^0 = \Delta H - T\Delta S$$

$$\ln K_c = \frac{\Delta H}{RT} + \frac{\Delta S}{R}$$

$$\Delta G^0 = -RT\ln K_c$$

where R is the natural gas constant and KC is the equilibrium constant and is calculated as

$$K_c = \frac{Ce}{1-Ce}$$

RESULT AND DISCUSSION

Biosorbent Characterization

FTIR

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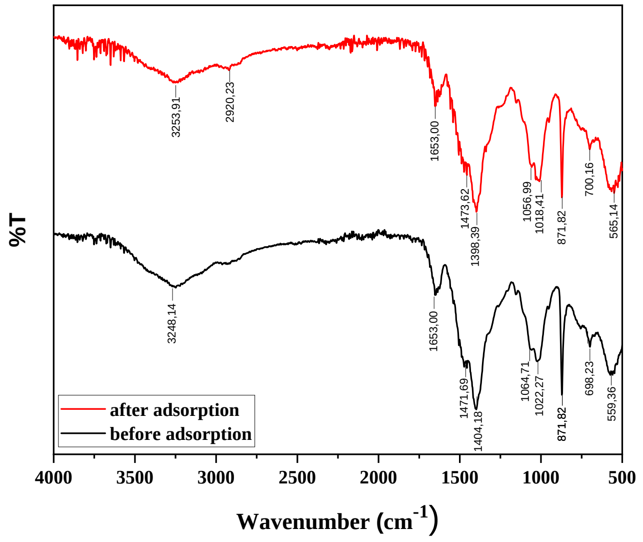

Figure1. The FTIR spectrum of SS before and after methylene blue dye adsorption.

Fig. 1. The FTIR spectrum of SS before and after methylene blue dye adsorption.The characterization of the biosorbent using FTIR was carried out over a wavenumber range of 4,000–500 cm^{-1}, as can be seen in Figure 1 the wavenumbers of several SS functional groups shifted due to chemical interactions between the biosorbent and methylene blue (Makeswari et al., 2016). In SS, there is a distinctive peak representing CO_3^{2-} from the aragonite compound ($CaCO_3$) appearing at wave numbers 1,404.18 and 871.82 cm^{-1}. After adsorption of methylene blue, the peaks of each functional group shifted. The broad and strong peak at 3,248.13 cm^{-1} which represents the –OH strain on the biosorbent. The peak at 1022.27 cm^{-1} indicates a stretch of the C-N group, and the N-H bond at the peak of 698.23 cm^{-1} which indicates that the SS contains proteins bound to chitin and SS minerals (Ramadhani et al., 2021). The appearance of a peak at wave number 1348.39 cm^{-1} indicates that the amine group in methylene blue has been adsorbed on the surface of the two biosorbents. Methylene blue is a dye with an amine group attached to an aromatic ring and a heterocyclic amine (Zein, Purnomo, et al., 2022).

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SEM-EDX

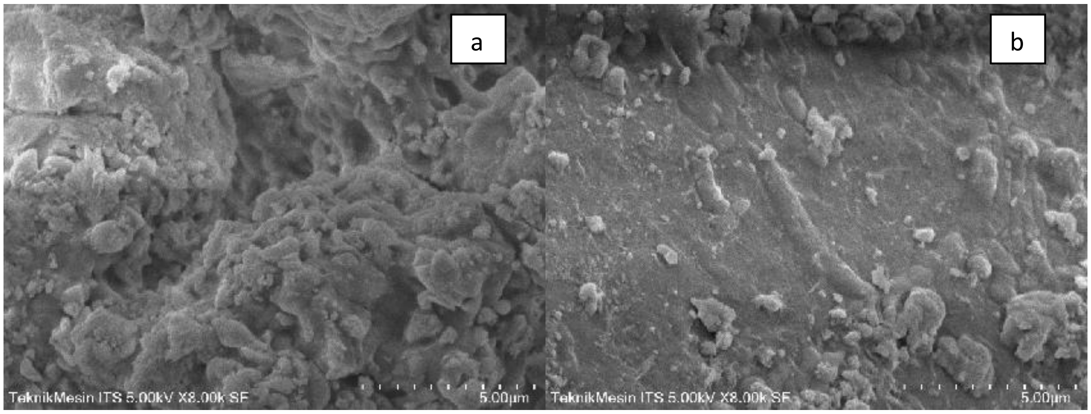

Figure2. SEM images of shrimp shell before adsorption (a), after adsorption (b)

Identification using SEM-EDX aims to see a surface directly on the biosorbent by scanning the surface using an electron beam with magnification up to a specific scale. Changes in the surface morphology of the biosorbent can be caused by the reorganization of the surface functional groups that bind to the methylene blue molecule. Alghouti (2020) explained that the smooth surface of the biosorbent after absorption showed the formation of a monolayer where all sites had the same affinity for the adsorbate, and there was no transmigration of the adsorbate in the surface plane (Al-Ghouti & Da'ana, 2020).

Table 1. The relative abundance of elements on the surface of the biosorbent before and after methylene blue absorption.

Elements	Before adsorption		After adsorption	
	Wt (%)	At (%)	Wt (%)	At (%)
C	0,62	4,22	2,36	1,07
O	53,04	71,3	60,41	78,36
Si	0,76	0,58	0,52	0,38
S	2,11	1,37	2,75	1,84
Ca	41,10	22,06	36,34	18,82

The percentage of dominant elements on the surface of shrimp shells before and after adsorption is shown in Table 1. The results of EDX analysis showed that the dominant elements on the surface of shrimp shells were C, O, Si, S, and Ca. These elements are the main components in shrimp shells that play a role in the adsorption of the removal of methylene blue dye. The EDX results showed that the C content after adsorption was higher

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than before. Considering that element C is present in the methylene blue molecular framework, it can be concluded that a large amount of methylene blue is adsorbed by both biosorbents(Ramadhani et al., 2020). The percentage of elemental O also increased after adsorption; this proves the strong affinity of the adsorbent for methylene blue dye. Then, the percentage of Ca and Si elements decreased after adsorption. The Ca element percentage indicated a cation exchange between Ca ions and methylene blue molecules.

Adsorption Model

Adsorption isotherm

This adsorption isotherm study aims to predict the adsorption mechanism between adsorbate and adsorbent. The adsorption isotherm explains how the distribution of adsorbate molecules on the adsorbent is at equilibrium(Zein, Chaidir, et al., 2022). In this study, two isotherm models were used to analyze equilibrium data, namely Freundlich and Langmuir. Freundlich's isotherm model is an empirical equation that describes adsorption on heterogeneous surfaces that form a multilayer layer, with groups having different adsorption energies. Then the Langmuir isotherm model relates to adsorption on a homogeneous surface that createscreates a monolayer layer of the adsorbent, and all groups on the adsorbent have the same energy(Eljiedi & Kamari, 2017). The linear equation of the Freundlich isotherm model is obtained by connecting the log qe value to the log Ce, and for the Langmuir isotherm, it is accepted by connecting the Ce value with Ce/qe(Hevira et al., 2021). The graph of the adsorption isotherm study can be seen in Figure 3a-b

Table 2. Parameters of methylene blue adsorption isotherm by shrimp shell

Isoterm Model	Parameters	Value
Langmuir	q_m (mg/g)	38,61
	K_L (L/mg)	0,00547
	R^2	0,9795
	R_L	0,1770-0,9481
Freundlich	K_F	1,8967
	1/n	0,278
	R^2	0,3797

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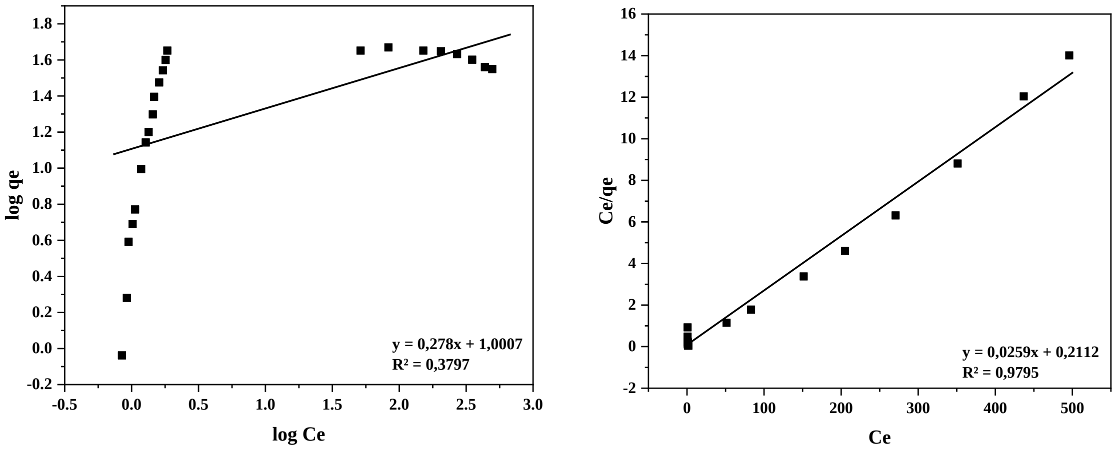


(a) (b)

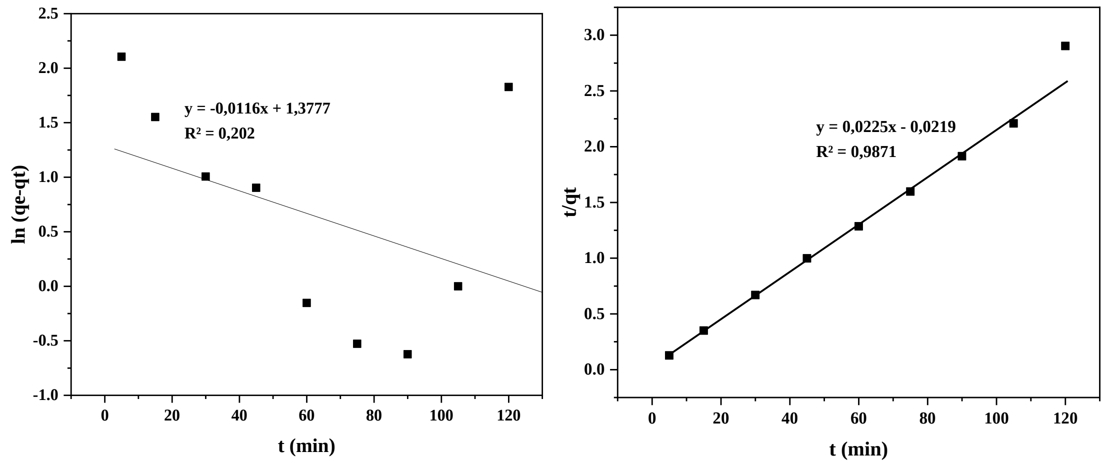
Figure3. (a) Freundlich isotherm model graph (b) Langmuir isotherm model

Table 2. q_m shows the maximum adsorption capacity obtained from the Langmuir model. K_L is the Langmuir equilibrium constant related to the interaction energy (Lmg^{-1}), K_f and n show the Freundlich affinity coefficient and the Freundlich linearity constant(Zein et al., 2019). It can be seen that the absorption of methylene blue by the adsorbent follows the Langmuir isotherm. Where the value of R^2 in the Langmuir isotherm model is 0.9795 for shrimp shells which shows the R^2 value is close to 1 and is greater than the R^2 of the Freundlich isotherm. The adsorption mechanism between methylene blue and the adsorbent shows the adsorption process on the surface of the adsorbent that occurs chemically by forming a monolayer layer on a homogeneous adsorbent.

An important characteristic of the Langmuir isotherm can be expressed by the separation factor (R_L). The R_L value indicates the nature of the adsorption process to be unfavorable ($R_L>1$), linear ($R_L = 1$), favorable ($0<R_L<1$), or irreversible ($R_L = 0$). The R_L values of 0.1770 to 0.9481 and 0.5460 to 0.9925 for methylene blue after adsorption by shrimp shells were in the range of $0 < R_L< 1$. This indicates that the adsorption of methylene blue is favorable (Eljiedi& Kamari, 2017). Thus, the shrimp shell is a suitable biosorbent for the absorption of methylene blue dye.Based on Table 2, it can be seen that according to the Langmuir isotherm, the value of qm is 38.61 mg L^{-1} for shrimp shells. This value is close to the experimental maximum adsorption capacity value. This indicates a good fit of the Langmuir model to the methylene blue equilibrium data.

Adsorption kinetics

The kinetic model can predict that the adsorption process occurs chemically or physically by constructing a curve by plotting the relationship between t vs -ln(qe-qt) for the first-order pseudo kinetic model and t vs t/qt for the second-order pseudo-kinetic model. The slope and intercept of the linear regression equation obtain the rate constant and adsorption capacity at equilibrium. The coefficient of determination (R^2) is considered a criterion of correspondence between the experimental data and the two proposed kinetic models(Naghizadeh & Ghafouri, 2017). The graph of the kinetic model can be seen in Fig. 4.



(a) (b)

Figure4. (a) first-order pseudo kinetic model (b) second-order pseudo kinetic model

Table 3. Parameters of methylene blue adsorption kinetics by shrimp shell

Kinetics Model	Value
q_e(exp)	47,547
First-order pseudo	
k_1 (min^{-1})	0,0116
q_e (calc) (mg g^{-1})	3,9657
R^2	0,202
Second-order pseudo	
k_2 (min^{-1})	-0,0231
q_e (calc) (mg g^{-1})	44,4
R^2	0,9871

Based on the data in Table 4. The R^2 value of the pseudo-second-order kinetic model is more excellent for shrimp shell biosorbent 0.9871. It can be indicated that the adsorption process of methylene blue dye by shrimp shell biosorbent follows a pseudo-second order, kinetic model. Xu et al. (2016) explained that the adsorption process for methylene blue occurs by chemisorption or chemical interaction involving ion exchange between the adsorbent and the adsorbate(Xu et al., 2016)

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Thermodynamics Study

The thermodynamic parameters provide further information about the energy changes associated with the adsorption process. The most common equations used for thermodynamic parameters, such as changes in free energy (ΔG^o), enthalpy (ΔH^o), and entropy (ΔS^o), were tested at temperatures of 298, 308, and 318 K with concentrations of 10, 20, 30, 40, and 50 mg L^{-1} (Anastopoulos & Kyzas, 2016). A negative or positive value of ΔG confirms the spontaneity or non-spontaneity of the adsorption process. ΔH (enthalpy change) provides information about the energy release (exothermic process) or energy capture (endothermic process) during the adsorption process. Another thermodynamic parameter, ΔS (change in entropy), indicates whether randomness increases (positive value) or decreases (negative value) during the adsorption process (Taylor et al., 2015).The effect of methylene blue adsorption temperature on standard Gibbs free energy (ΔG^o) can be seen in Fig. 5.

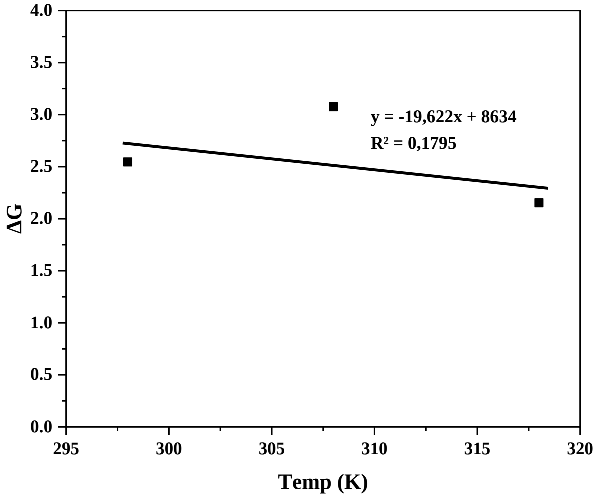

Figure5. Thermodynamic plots for methylene blue adsorption on shrimp shell

Table 1. Thermodynamic parameters of methylene blue adsorption by shrimp shell

Temperature (K)	ΔG (kJ mol^{-1})	ΔH (kJ mol^{-1})	ΔS (kJ mol^{-1} K^{-1})
298	2.55	8.6344	0.0196
308	3.08		
318	2.15		

Based on Table 5. For shrimp shells, ΔG is positive. This indicates that the adsorption process is not spontaneous. A positive ΔH value indicates that the adsorption process is endothermic. The non-spontaneous adsorption process requires external energy (endothermic), and a positive S value indicates a high level of disturbance on the surface of the adsorbent (Djelloul & Hamdaoui, 2014).

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CONCLUSION

Based on the research that has been done, the adsorption process followed the Langmuir isotherm model and pseudo-second-order kinetics model, which showed that the adsorption process occurred chemically through the interaction between methylene blue dye and biosorbent and formed a monolayer on the surface of the biosorbent. Thermodynamic studies for shrimp shells showed that the adsorption process was non-spontaneous, endothermic, and had high regularity. FTIR showed that there was a shift in the wavenumber indicating the involvement of functional groups between methylene blue and the biosorbent. SEM-EDX analysis showed the filling of pores on the surface of the biosorbent by methylene blue molecules.

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