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Biopolymer-based cellulose-zeolite composite membranes for cationic and anionic dye adsorption in wastewater systems

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ABSTRACT

Textile wastewater contaminated with both cationic and anionic dyes remains a major environmental challenge. This study aims to evaluate the adsorption performance of a biopolymer-based cellulose–zeolite composite membrane as an environmentally friendly adsorbent for methylene blue (MB) and remazol red (RR). The membrane was fabricated by incorporating cellulose acetate with natural zeolite and subsequently applied in batch adsorption experiments. Membrane stability under different pH conditions and surface functional groups were evaluated using FTIR analysis. The adsorption behavior was investigated by examining the effects of solution pH, contact time, and initial dye concentration, followed by analysis using Langmuir and Freundlich isotherm models. The results demonstrated excellent agreement between experimental data and both isotherm models. The maximum adsorption capacities calculated using the Langmuir model were 0.94 mg g^{-1} for MB and 0.29 mg g^{-1} for RR, demonstrating preferential adsorption toward dyes with different charge characteristics. The adsorption of MB was governed by electrostatic interactions, whereas RR adsorption was governed by hydrogen bonding and physical interactions. These results demonstrate that the cellulose-zeolite composite membrane exhibits selective adsorption behavior and confirm its effectiveness as a sustainable adsorptive material for the treatment of complex textile wastewater.

Keywords: cellulose-zeolite, composite membrane, adsorption, methylene blue, remazol red.

INTRODUCTION

Water pollution caused by synthetic dyes discharged from textile industries remains a critical global environmental issue (Periyasamy, 2025; Sukarta, Suyasa, Mahardika, Suprihatin, & Sastrawidana, 2025). Cationic and anionic dyes, such as methylene blue and remazol red, are widely recognized for their toxicity, chemical stability, and resistance to biological degradation, leading to their persistence in aquatic environments and posing serious ecological and human health risks (Sudiana et al., 2022; Sukarta et al., 2021). In addition to deteriorating water aesthetics, dye contamination reduces light penetration, inhibits photosynthetic activity, and disrupts aquatic ecosystems (Adenan & Zaini, 2024; Aziz et al., 2024; Shukor et al., 2022).

Numerous treatment technologies have been developed for dye removal, including use of activated carbon kaolin filter using response surface methodology (Belay et al., 2025), flocculation, electrocoagulation, advanced oxidation processes (Lucas et al., 2025), photodegradation (Sukarta et al., 2025; Yerima et al., 2024), and ion exchange (Naim et al., 2022).

Nevertheless, many of these approaches are limited by high energy consumption, elevated operational costs, secondary waste generation, and insufficient efficiency toward structurally complex dyes (Fadhil & Abbas, 2025; Ledakowicz & Pa, 2021; Singh & Ransingh, 2020). Consequently, adsorption remains one of the most extensively applied techniques due to its operational simplicity, high removal efficiency, and economic feasibility (Ahmaruzzaman et al., 2025; Choudhary & Maurya, 2024; Dawood & Sen, 2014).

In recent years, adsorptive membrane technology has emerged as a promising hybrid approach that integrates filtration and adsorption mechanisms within a single system. This strategy enables the simultaneous physical retention and chemical interaction of contaminants with the membrane surface. Compared to conventional powdered adsorbents, adsorptive membranes offer significant advantages, including easier solid–liquid separation, continuous operation potential, and improved regeneration capability, while operating under relatively low energy requirements (Akinyemi et al., 2024; Valiollah & Mohadeseh, 2025).

Biopolymer-based membranes, particularly those derived from cellulose and its derivatives, have attracted increasing attention owing to their biodegradability, hydrophilicity, and abundant availability. Cellulose acetate is widely used as a membrane material due to its excellent film-forming properties and chemical stability. However, pristine cellulose-based membranes often exhibit limited adsorption capacity and poor selectivity toward contaminants with different charge characteristics. To overcome these limitations, the incorporation of porous inorganic fillers such as zeolite has been proposed as an effective modification strategy.

Zeolite is a crystalline aluminosilicate characterized by a high specific surface area, a well-defined pore structure, and strong ion-exchange capacity. Incorporation of zeolite into a cellulose matrix produces composite membranes that combine the mechanical flexibility and hydrophilicity of cellulose with the adsorption capacity and selectivity of zeolite (Chen et al., 2021).

Previous studies have reported the use of cellulose–zeolite composite membranes for the removal of heavy metals and individual dyes. However, available investigations predominantly examine either a single adsorbate or dyes of the same charge type, providing limited comparative analysis of the adsorption behavior of cationic and anionic dyes within a single membrane system (Hierarchical et al., 2022; Ibrahim et al., 2024; Radoor et al., 2022).

In addition, systematic evaluations of the combined influence of key operational parameters-such as solution pH, contact time, and initial dye concentration–on adsorption mechanisms and isotherm characteristics in cellulose-zeolite composite membranes remain limited. These parameters directly regulate electrostatic interactions, hydrogen bonding, and surface heterogeneity, which determine adsorption capacity, selectivity, and overall performance.

MATERIALS AND METHODS

Materials

Cellulose acetate was used as the polymer matrix, while natural zeolite served as the inorganic filler for composite membrane preparation. Methylene blue (MB) and remazol red (RR) were selected as representative cationic and anionic dyes, respectively. Analytical-grade reagents including acetic acid (CH_3COOH, 0.1 M), sodium hydroxide (NaOH, 0.1 M), and hydrochloric acid (HCl, 0.1 M) were used for pH adjustment. Distilled water was used throughout all experiments.

Experimental methods

Preparation of cellulose-zeolite composite membrane

Natural zeolite was washed with distilled water and dried in an oven at 110 °C for 4 h, followed by grinding and sieving to obtain a particle size of 230 mesh. The zeolite was then calcined at 400 °C for 2 h and stored in a desiccator prior to use. Cellulose acetate solution (20% w/w) was prepared by dissolving the polymer in acetone under continuous stirring for 24 h. The solution was subsequently degassed by refrigeration for 24 h to remove entrapped air bubbles. For composite membrane fabrication, zeolite was incorporated into the cellulose acetate solution at varying loadings (5–20% w/w relative to polymer weight) and stirred for an additional 24 h to ensure homogeneous dispersion. The resulting solution was cast onto a glass plate and solvent evaporation was carried out in a desiccator for seven days to obtain free-standing membranes (Ernawati, 2014). The composition for making cellulose-zeolite composite membrane is presented in Table 1.

Membrane characterization

The pH stability of the membranes was assessed by immersing pre-weighed samples in 30 mL of methylene blue solution (30 mg L^{-1}) adjusted to pH 3, 5, 7, 9, and 11 for 2 h. After immersion, the membranes were removed, dried to constant

Table 1. The composition for making cellulose–zeolite composite membrane

Code	Cellulose acetate (g)	Zeolite (g)	Acetone (mL)	DMSO (mL)
Saz I	1	0.05	47.5	2.5
Saz II	1	0.15	42.5	7.5

weight, and reweighed to determine mass changes associated with swelling or structural degradation.

The functional groups of the membranes were characterized using fourier transform infrared (FTIR) spectroscopy. Approximately 2 mg of membrane sample was thoroughly mixed with 200 mg of potassium bromide (KBr), compressed into a pellet, and analyzed over the wavenumber range of 400–4000 cm^{-1}.

The adsorption experiment studies

Stock solutions (1000 mg L^{-1}) of methylene blue and remazol red were prepared by dissolving 1.0 g of each dye in 1000 mL of distilled water under continuous stirring to ensure complete dissolution. Working solutions of the desired concentrations were subsequently obtained by serial dilution with distilled water. The maximum absorption wavelength (λ_{max}) of each dye was determined using UV–Vis spectrophotometry by scanning in the wavelength range of 400–650 nm, and the identified λmax values were used for subsequent concentration measurements.

Effect of pH

Five Erlenmeyer flasks were prepared, each containing 30 mL of Remazol Red solution at an initial concentration of 30 mg/L. The pH of each solution was adjusted to 3, 5, 7, 9, and 11 by adding 0.05 M HCl or 0.05 M NaOH, and the pH values were verified using a calibrated pH meter. An identical procedure was applied to the methylene blue solution. Subsequently, 0.5 g of the cellulose-zeolite composite membrane was introduced into each flask, and the mixtures were agitated in a shaker for 60 minutes to ensure homogeneous contact. After adsorption, the suspensions were filtered and further centrifuged at 5000 rpm for 15 minutes to obtain a clear supernatant. The residual dye concentration was then determined using a UV-visible spectrophotometer at the respective maximum absorption wavelength (Setiawan et al., 2019).

Contact time

The dye solution was prepared at an approximate concentration of 30 mg/L, and its actual concentration was verified using a UV-visible spectrophotometer. Subsequently, 25 mL of the dye solution was adjusted to the optimum pH determined from the pH variation study. A mass of 0.5 g of the membrane was then added to the solution, and the mixture was agitated using a shaker for different contact times of 10, 20, 40, 60, 80, and 100 minutes (Elystia et al., 2018). After the adsorption process, the suspension was filtered and centrifuged at 5000 rpm for 15 minutes to obtain a clear solution. The absorbance of the supernatant was then measured at the maximum absorption wavelength using a UV-visible spectrophotometer.

Concentration effect

Remazol red and methylene blue dye solutions were prepared under optimum pH conditions at initial concentrations of approximately 10, 30, 50, 70, and 100 mg/L. The actual concentrations were verified using a UV-visible spectrophotometer. Subsequently, 0.5 g of the cellulose-zeolite composite membrane was introduced into 30 mL of each dye solution, and the mixtures were agitated at the predetermined optimum contact time. After the adsorption process, the suspensions were filtered using filter paper and centrifuged at 5000 rpm for 15 minutes to obtain a clear supernatant. The absorbance was then measured at the respective maximum wavelength using a UV-visible spectrophotometer. The adsorption efficiency of the cellulose–zeolite composite membrane under variations of pH, contact time, and initial dye concentration was calculated using the following equation.

$$\%E = \frac{C_0 - C_e}{C_0} \times 100\% \quad (1)$$

where: E represents the adsorption efficiency (%), C_0 is the initial dye concentration before adsorption (mg/L), and C_e is the equilibrium dye concentration after adsorption (mg/L).

Meanwhile, for the adsorption capacity, the following equation is used.

$$\textit{Capacity of adsorption}\ (\text{mg/g}) = = \frac{C_0 - C_e}{w} \times V \quad (2)$$

The adsorption isotherms studies

A total of 30 mL of remazol red solution was prepared at pH 6 with an approximate concentration of 30 mg/L. The actual initial concentration was determined using a UV-visible spectrophotometer at its maximum absorption wavelength. Subsequently, 0.5 g of the cellulose-zeolite membrane was added to the solution, and the mixture was agitated for 200 minutes. To evaluate the adsorption isotherm behavior and determine the maximum adsorption capacity, the procedure was repeated at initial dye concentrations of 10, 30, 50, 70, and 100 mg/L. After equilibrium was reached, the residual dye concentration was measured and recorded as the equilibrium concentration (C_e), and the adsorption capacity (q_e) was calculated accordingly. The same experimental procedure was applied to methylene blue dye.

Under the Langmuir adsorption isotherm framework, *Ce*/*Qe* is linearly proportional to *Ce*, as represented by the equation below:

$$\frac{Ce}{Qe} = \left(\frac{1}{Qmax}\right)\left(\frac{1}{b}\right) + \left(\frac{1}{Qmax}\right)Ce \qquad (3)$$

Alternatively, under the Freundlich isotherm model, log *Qe* is linearly correlated with log *Ce*, as expressed in the corresponding equation.

$$\log Qe = \log kF + \frac{1}{n}\log Ce \qquad (4)$$

where: *kF* is the Freundlich constant, which reflects the adsorption capacity and intensity of the adsorbent.

Data analysis

The adsorption capacity (q_e), removal efficiency (%R), and equilibrium adsorption behavior were quantitatively determined using standard adsorption equations based on mass balance relationships. The equilibrium adsorption capacity was calculated from the difference between initial and residual dye concentrations, taking into account the solution volume and membrane mass.

To elucidate the adsorption mechanism and surface characteristics of the composite membrane, the experimental equilibrium data were fitted to the Langmuir and Freundlich isotherm models. The Langmuir model was employed to assess monolayer adsorption capacity and surface homogeneity, whereas the Freundlich model was applied to describe heterogeneous surface interactions and adsorption intensity. Model parameters were obtained by nonlinear regression analysis, and the goodness of fit was evaluated using correlation coefficients (R^2).

Adsorption capacity, removal efficiency, and equilibrium behavior were calculated using standard adsorption equations. Langmuir and Freundlich isotherm models were applied to evaluate adsorption mechanisms and maximum adsorption capacity.

RESULTS AND DISCUSSION

The results of the synthesis of cellulose-zeolite composite membranes are presented in Figure 1. The cellulose-zeolite composite membranes were then tested for their pH resistance.

Characterization of cellulose–zeolite composite membrane

pH resistance test

The cellulose-zeolite membrane did not experience significant physical or chemical changes across all pH variations tested. After immersion in acidic pH (pH 3–5), neutral pH (pH 7), and basic pH (pH 9–11), the membrane retained its color, shape, flexibility, and mechanical strength as before testing. No indication of swelling, cracking, or degradation of the membrane surface was found. This resistance indicates that the interaction between the cellulose hydroxyl groups and the aluminosilicate framework in zeolite forms a strong and stable bond, thus being able to withstand attacks by H^+ ions in acidic conditions and OH^- ions in basic conditions. Zeolite also acts as a structural reinforcement that increases the thermal and chemical stability of the cellulose matrix. It can be concluded that the cellulose-zeolite membrane has excellent stability against pH changes. This membrane is resistant to acidic, basic, and neutral conditions without experiencing degradation or changes in physical properties, so it has the potential to be applied in various filtration or separation processes involving environments with wide pH variations.

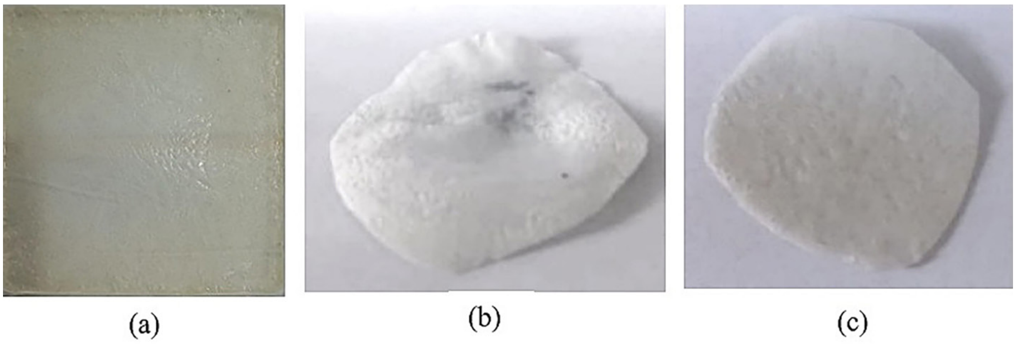


Figure 1. (a) Membrane cellulose acetate, (b) Saz I, (c) Saz II

Functional group analysis

The FTIR spectra of the cellulose-zeolite composite membrane before and after dye adsorption are presented in Figure 2.

The results of the FTIR spectrum analysis in Figure 2 show differences in the characteristics of functional groups between cellulose acetate, zeolite, and Saz I and Saz II composites. The cellulose acetate spectrum displays a characteristic absorption band at a wave number of around 3400 cm^{-1} indicating O–H stretching, a band at 2900 cm^{-1} for C–H stretching, and a strong band at around 1735 cm^{-1} indicating the presence of carbonyl groups (C=O) from the acetate group. Zeolite shows a broad absorption band at around 3450 cm^{-1} originating from the O–H stretching of adsorbed water and a band at 1630 cm^{-1} related to the H–O–H bending vibration, while the strong bands in the 1000–450 cm^{-1} region indicate Si–O–Si and Al–O–Si stretching which are characteristic of the aluminosilicate framework. In the SAz I and SAz II composites, a combination of the characteristics of both materials is seen, but with changes in intensity and a slight shift in the absorption bands. The decrease in the intensity of the O–H and C=O bands and a small shift in the Si–O–Si region indicates an interaction between the active groups of cellulose acetate and the zeolite surface, mainly through the formation of hydrogen bonds. The more pronounced changes in the SAz II spectrum compared to SAz I indicate that the modification or composite formation process in stage II results in a stronger interaction and a more homogeneous composite structure between cellulose acetate and zeolite.

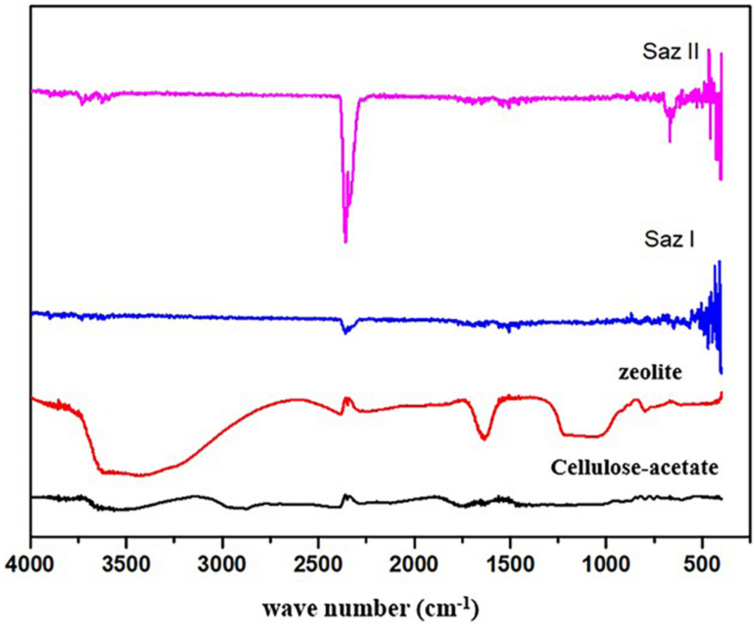


Figure 2. FTIR spectrum

Adsorption of cationic and anionic dyes

The effect of pH variations on adsorbate concentration can be seen in Figure 3. Based on it, the effect of pH variation on adsorbate concentration, it can be seen that at all pH variations, the final concentration of remazol red (sample II) is higher than methylene blue (sample I). This indicates that the adsorption efficiency of methylene blue tends to be greater than remazol red. Remazol red shows higher adsorption results than methylene blue at all pH variations due to several factors related to the chemical properties of the dye and the surface characteristics of the adsorbent (cellulose acetate-zeolite membrane).

Remazol red is an anionic dye containing a highly polar sulfonate ($–SO_3^–$) group that interacts strongly with the hydroxyl (–OH) and acetyl ($–OCOCH_3$) groups in cellulose acetate, as well as with the silanol (–Si–OH) groups in zeolite through hydrogen bonding and electrostatic forces. Zeolite surfaces tend to have a weak positive charge in the low-neutral pH range, thus attracting negative ions from remazol red more effectively (Hierarchical et al., 2022). Furthermore, the larger molecular size of remazol red and the numerous active groups increase the likelihood of multipoint interactions with the adsorbent surface. In contrast, methylene blue is a cationic (positively charged) dye, which tends to experience electrostatic repulsion on the cellulose acetate-zeolite adsorbent surface, particularly at neutral to alkaline pH when the adsorbent surface becomes negatively charged (Shukor et al., 2022). This limits the ability of methylene blue to adhere strongly to the adsorbent surface. Thus, the adsorption of remazol red is higher than that of methylene blue because remazol red has stronger electrostatic interactions and hydrogen bonds with the cellulose acetate-zeolite surface, as well as the possibility of forming multipoint bonds that increase the adsorption stability, while methylene blue only interacts through the van der Waals force mechanism and a small part of electrostatic (Gürses et al., 2023).

The results of the adsorption of methylene blue and remazol red with time variations are presented in Figure 4.

Figure 4 shows the variation of adsorption percentage (%Ads) over time, showing that both methylene blue (I) and remazol red (II) experienced an increase in adsorption percentage as the contact time increased until they reached a nearly constant state after 90 minutes. At the beginning of the time (30 minutes), the adsorption of methylene blue was still low compared to remazol red because the diffusion process of methylene blue molecules to the surface and pores of the membrane had not yet taken place optimally (Bai et al., 2025). However, after 60 minutes, there was a sharp increase in the adsorption percentage of methylene blue which even exceeded that of remazol red, indicating that at that time an adsorption equilibrium had been reached between the number of dye molecules bound to the adsorbent surface and those still in solution. Remazol red showed a high adsorption value from the beginning (30 minutes), because the sulfonate group ($–SO^{3-}$) in this dye had a fast interaction with the hydroxyl and acetyl groups in cellulose acetate and the silanol groups in zeolite. However, the subsequent increase tended to stabilize after 60 minutes, indicating that the adsorbent surface had begun to saturate. Overall, increasing the contact

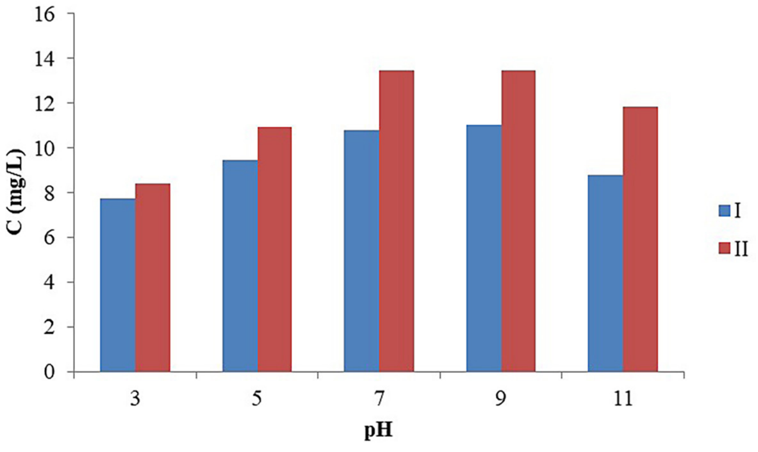


Figure 3. Adsorption results of methylene blue and remazol red with pH variations

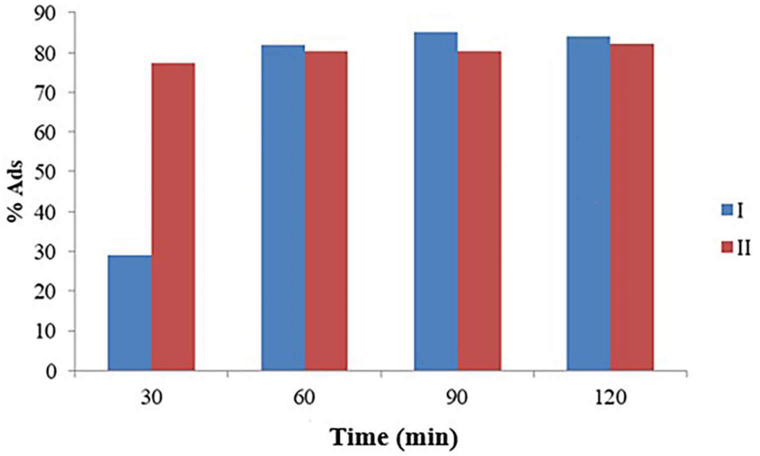


Figure 4. Results of adsorption of methylene blue and remazol red with time variation

time provides a greater opportunity for the dye molecules to diffuse and interact with the active sites on the cellulose acetate-zeolite membrane. The optimum adsorption time was reached at approximately 90 minutes, where both dyes showed the highest % adsorption values and approached equilibrium conditions.

Meanwhile, the results of the adsorption of methylene blue and remazol red with varying concentrations are presented in Figure 5.

Based on Figure 5 at low concentrations (10 mg/L), the number of active sites on the surface of the cellulose acetate–zeolite membrane is still large and able to interact with most of the dye molecules present, resulting in a high % adsorption. This can be seen in methylene blue which reaches around 90% and remazol red around 65%. However, when the dye concentration increases (≥ 7 mg/L), the number of dye molecules available in the solution becomes much greater than the number of active sites of the adsorbent. As a result, most of the dye molecules remain in the solution because the adsorption sites have begun to saturate, so that the % adsorption decreases. Methylene blue shows a higher % adsorption value than remazol red at most concentrations, possibly because the positive charge on methylene blue interacts more easily with the –OH and –Si–O$^-$ groups on the zeolite and the acetyl groups on cellulose acetate through electrostatic forces and dipole interactions. Meanwhile, remazol red, which is anionic, experiences repulsive forces on the negatively charged adsorbent surface, especially at neutral-alkaline pH, so its adsorption efficiency is slightly lower. Thus, it can be concluded that the higher the initial dye concentration, the lower the adsorption efficiency due to the limited number of active sites on the membrane, and differences in the chemical properties of the dyes also affect their respective adsorption capabilities.

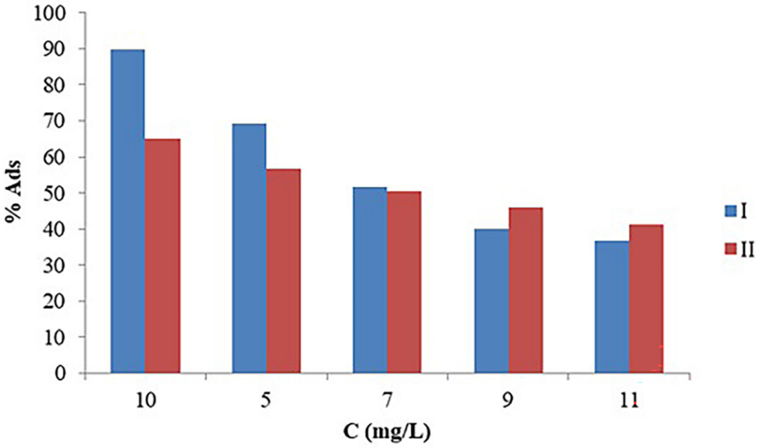


Figure 5. The adsorption of methylene blue and remazol red with varying concentrations

Adsorption isotherm

The adsorption isotherm patterns used are the Langmuir and Freundlich isotherm patterns. The determination of the adsorption isotherm is carried out by channeling the Langmuir and Freundlich adsorption isotherm equations into a straight-line equilibrium curve where the equilibrium model depends on the highest coefficient of determination (*R*). The determination of the adsorption isotherm aims to determine the number of methylene blue ions adsorbed by the adsorbent. The type of adsorption isotherm can be used to determine the adsorption mechanism of the adsorbent towards the adsorbate. Solid-liquid phase adsorption usually follows the Langmuir and Freundlich adsorption isotherm types, with bonds between the adsorbate molecules and the adsorbent surface can occur by physisorption and chemisorption. The Langmuir isotherm pattern curve is presented in Figure 6, while the Freundlich isotherm pattern is presented in Figure 7.

Based on the Langmuir isotherm analysis presented in Figure 6, the maximum adsorption capacities (q_{max}) of the cellulose–zeolite composite membrane toward methylene blue and remazol red were determined to be 0.94 mg g^{-1} and 0.29 mg g^{-1}, respectively. The notable difference in (q_{max}) values highlights the selective adsorption behavior of the composite membrane toward dyes with different charge characteristics.

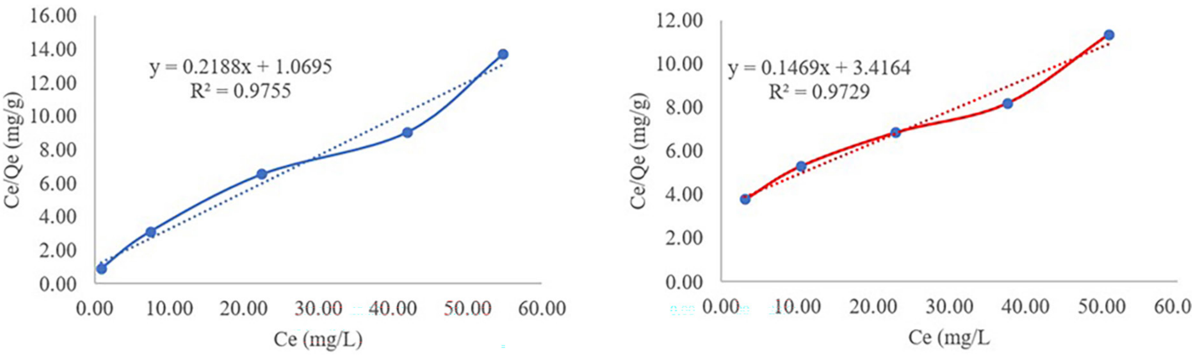
The higher (q_{max}) for MB indicates stronger adsorption affinity toward the cationic dye, which can be attributed to electrostatic attraction between positively charged MB molecules and the negatively charged membrane surface. This negative surface charge arises from deprotonated silanol groups of zeolite and hydroxyl groups of cellulose under neutral to alkaline conditions, promoting efficient monolayer adsorption.

In contrast, the lower maximum adsorption capacity observed for RR suggests that adsorption of anionic dyes is not primarily governed by electrostatic interactions. Instead, RR adsorption likely proceeds via hydrogen bonding and physical interactions between sulfonate groups and polar functional groups on the membrane surface, which generally result in lower adsorption capacities (Gürses et al., 2023; Tajat et al., 2022).

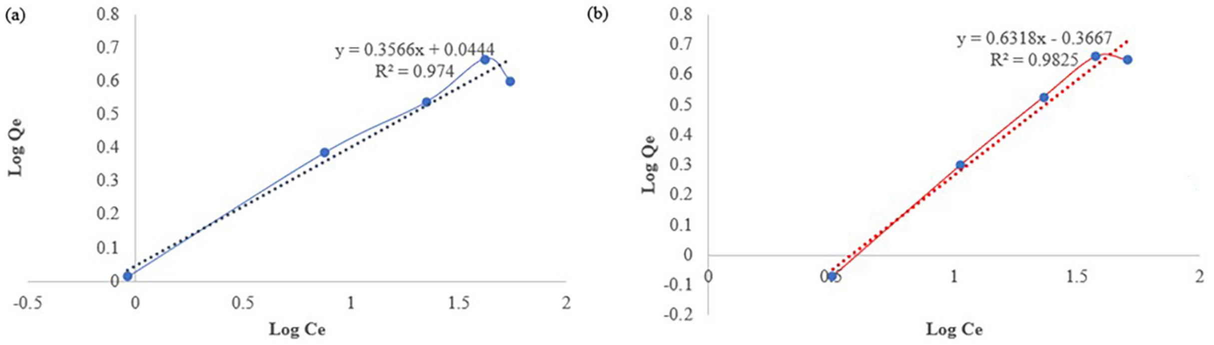
When compared with previous studies, the obtained (q_{max}) for MB falls within the expected range for cellulose-based adsorbents without extensive chemical modification. Busayo et al., (2025) reported that unmodified cellulose adsorbents typically exhibit relatively low MB adsorption capacities, while higher values are often achieved only after chemical activation. Chen et al. (2021) and Widiastuti et al. (2019) also noted that natural zeolites alone exhibit limited adsorption capacity due to restricted accessibility of active sites. The composite membrane developed in this study effectively integrates both materials to achieve stable and selective adsorption without complex surface modification.

For anionic dyes, the (q_{max}) value for RR is consistent with reports by Foo and Hameed (2010), which indicate lower adsorption capacities of sulfonated dyes on negatively charged adsorbents. Despite the lower capacity, the ability of the composite membrane to adsorb both MB and RR demonstrates its practical relevance for treating textile wastewater containing mixed dye pollutants.

The Freundlich adsorption isotherm patterns of methylene blue and remazol red on the cellulose–zeolite composite membrane are presented in Figure 7a and Figure 7b. The Freundlich model describes adsorption on heterogeneous surfaces with non-uniform energy distribution, complementing



Figures 6. Langmuir adsorption isotherm pattern: (a) methylene blue dye and (b) remazol red dye



Figures 7. Freundlich adsorption isotherm pattern: (a) methylene blue dye and (b) remazol red dye

the Langmuir model. The linearized Freundlich equations obtained were:

For MB:

- log qe = 0.3566 log Ce + 0.0444 ($R^2 = 0.974$)

For RR:

- Log qe = 0.6318 log Ce − 0.3667 ($R^2 = 0.9825$)

The high R^2 values indicate that the Freundlich model adequately represents the adsorption behavior of both dyes, particularly at low to moderate concentrations. The Freundlich intensity parameter n, calculated from the reciprocal of the slope ($1/n$), was approximately 2.80 for MB and 1.58 for RR, indicating favorable adsorption ($n > 1$) for both dyes. The higher n value for MB suggests stronger adsorption intensity and greater affinity toward the composite membrane, consistent with the higher maximum adsorption capacity obtained from Langmuir analysis.

The Freundlich constant K_f, which reflects adsorption capacity at low concentrations, was higher for MB than for RR, indicating stronger initial interaction between MB molecules and the membrane surface. This behavior is primarily attributed to electrostatic attraction between cationic MB and negatively charged functional groups on the cellulose–zeolite matrix.

In contrast, the lower n value for RR implies adsorption on a more heterogeneous surface with a greater contribution of physical adsorption mechanisms. The sulfonate groups of RR interact mainly through hydrogen bonding and van der Waals forces, resulting in a broader energy distribution of adsorption sites.

Compared with previous studies, the obtained n values fall within the range commonly reported for cellulose- and zeolite-based adsorbents. According to Foo and Hameed (2010), n values between 1 and 10 indicate favorable adsorption, whereas higher n values are associated with stronger and more selective interactions. Therefore, the present results support the suitability of the cellulose–zeolite composite membrane for the adsorption of dyes with different charge characteristics.

Overall, the agreement of the experimental data with both the Langmuir and Freundlich isotherm models suggests that dye adsorption on the cellulose–zeolite composite membrane involves a combination of electrostatic interactions, hydrogen bonding, and surface heterogeneity rather than a single adsorption mechanism. The distinct adsorption behaviors observed for methylene blue and remazol red demonstrate the significant role of molecular charge in determining adsorption affinity and capacity. These findings provide mechanistic insight into the adsorption of dyes with different charge characteristics on biomaterial–mineral composite membranes and indicate that the cellulose–zeolite composite membrane exhibits selective and stable adsorption performance relevant to complex textile wastewater treatment.

CONCLUSIONS

This study demonstrates that biopolymer-based cellulose–zeolite composite membranes are effective, stable, and selective adsorbents for the removal of cationic and anionic dyes from aqueous solutions. pH stability tests and FTIR analysis confirmed that the integration of cellulose acetate and zeolite produced chemically robust membranes with strong interfacial interactions through hydrogen bonding and surface functional groups.

Adsorption experiments revealed that operational parameters such as solution pH, contact time, and initial dye concentration significantly influenced adsorption performance. Langmuir and Freundlich isotherm analyses showed excellent agreement with the experimental data, indicating the coexistence of monolayer adsorption and surface heterogeneity. The maximum adsorption capacities derived from the Langmuir model were 0.94 mg g^{-1} for methylene blue and 0.29 mg g^{-1} for remazol red, highlighting a higher adsorption affinity toward the cationic dye.

The distinct adsorption behaviors of the two dyes confirm that molecular charge and membrane surface characteristics play crucial roles in governing adsorption mechanisms and capacity. Methylene blue adsorption was primarily driven by electrostatic interactions, whereas remazol red adsorption was dominated by hydrogen bonding and physical interactions. Overall, this work provides fundamental insights into dye adsorption mechanisms on biomaterial–mineral composite membranes and underscores the potential of cellulose–zeolite composite membranes as environmentally friendly adsorptive materials for treating complex textile wastewater.

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