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Efficient adsorptive removal of Remazol Black dye from textile wastewater using quaternary ammonium polymers: Mechanistic insights, isotherm modeling, and thermodynamic evaluation

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ABSTRACT

This study evaluates the efficiency and adsorption mechanism of Remazol Black dye removal from textile wastewater using quaternary ammonium polymers (QAPs) as adsorbents. The adsorption behavior was systematically investigated through isotherm, kinetic, and thermodynamic analyses to elucidate the interaction mechanisms between the dye molecules and the adsorbent. Batch adsorption experiments were conducted to examine the effects of adsorbent dosage, contact time, temperature, and initial dye concentration. Adsorption data were analyzed using Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich isotherm models, while kinetic behavior was evaluated using pseudo first order and pseudo-second-order models. Surface and structural characterizations before and after adsorption were performed using SEM, FTIR, and BET analyses to identify morphological changes and functional group interactions. The optimum adsorption conditions were achieved at an adsorbent dosage of 0.10 g, a temperature range of 25–30 °C, and a contact time of 20–30 minutes, resulting in removal efficiencies of 97.65% for synthetic dye solutions and 95.64% for textile wastewater. The Langmuir model provided the best fit, indicating monolayer adsorption, while the pseudo-second-order kinetic model suggested chemisorption as the dominant rate-controlling mechanism. Thermodynamic parameters ($\Delta H° = 65.37$ kJ/mol, $\Delta S° = 0.21$ kJ/mol.K, and $\Delta G°$ values ranging from +1.54 to -8.10 kJ/mol) indicated that the adsorption process is endothermic and becomes spontaneous at elevated temperatures. A decrease in removal efficiency in textile wastewater compared to synthetic solutions was attributed to matrix effects, including the presence of competing ions and organic substances. The findings provide mechanistic insights into the adsorption performance of QAPs for dye removal from complex aqueous systems.

Keywords: azo dyes, Remazol Black, quaternary ammonium polymer, adsorption mechanism, environmental remediation, wastewater.

INTRODUCTION

The textile sector is recognized as one of the major consumers of water, leading to the generation of significant amounts of wastewater, especially during dyeing and finishing stages. Textile effluents commonly contain a complex combination of dissolved substances, including organic compounds, salts, heavy metals, and synthetic dyes, many of which are highly resistant to natural degradation processes (Al-Tohamy et al., 2022). When released without adequate treatment, these effluents pose serious environmental risks, such as deterioration of water quality, harmful effects on aquatic life, and inhibition of photosynthetic activity in aquatic ecosystems. Furthermore, a large proportion of dyes and auxiliary chemicals used in textile processing exhibit poor biodegradability, limiting the effectiveness of conventional biological treatment methods (Jahan et al., 2022;

Jorge et al., 2025). Consequently, there is an increasing demand for advanced and efficient treatment approaches that can effectively eliminate persistent dye contaminants rather than relying solely on traditional biological processes.

Therefore, advancing efficient wastewater treatment technologies is essential to limit the discharge of toxic compounds into aquatic systems and to support sustainable water management within the textile industry (Panhwar et al., 2024). A wide range of treatment strategies has been explored for textile effluent remediation, including physical, chemical, and biological methods such as coagulation–flocculation, membrane-based separation, chemical oxidation, biodegradation, and adsorption (Ayed et al., 2020; Wang et al., 2022). Among these approaches, adsorption has attracted considerable attention due to its high removal efficiency, operational flexibility, economic feasibility, and capability to eliminate pollutants even at trace concentrations (Tuciilo et al., 2023). Nonetheless, the overall performance of adsorption processes is largely governed by the physicochemical characteristics of the adsorbent material and the nature of the interactions occurring at the adsorbent–pollutant interface.

Remazol dyes constitute an important class of reactive dyes extensively employed in textile manufacturing owing to their high water solubility, strong fiber fixation, and vivid coloration (Suhartini et al., 2025; Khaliq et al., 2021). Remazol Black is one of the most commonly applied dyes in Indonesia's textile sector, particularly for coloring a wide range of fabric materials (Ayu and Kasiamdari, 2022). Despite its industrial advantages, Remazol Black is an azo dye with high persistence and potential toxicity, making its removal from wastewater particularly challenging. Its anionic character and molecular complexity often limit removal efficiency when non-selective or weakly interactive adsorbents are used. Polymeric materials have attracted increasing interest as adsorbents due to their chemical stability, mechanical durability, and tunable surface properties (Alkhaldi et al., 2024; Hernández-Cruz et al., 2025).

Polymers are high–molecular weight materials formed from repeating monomer units through polymerization processes (Speight, 2020). In recent years, polymer-based adsorbents have been increasingly reported as effective materials for dye removal from aqueous systems (Kolya and Kang, 2025; Mihai et al., 2025; Panić et al., 2013). Within this class of materials, quaternary ammonium polymers (QAPs) are particularly notable due to the presence of permanently charged cationic functional groups, which promote strong electrostatic attraction toward anionic dye species. Previous studies have documented the high removal performance of QAPs for various contaminants, including nitrate, phosphate, and several synthetic dyes (Rahayu et al., 2023b) (Musnamar et al., 2024; Rahayu et al., 2023). Moreover, these polymers have shown promising applicability in complex wastewater matrices, such as ethanol production vinasse, where phosphate removal efficiencies of up to 75.70% have been achieved (Veranica et al., 2024). Collectively, these results highlight the considerable potential of quaternary ammonium polymers for practical wastewater treatment under conditions involving multiple pollutants.

Motivated by these gaps, the present study focuses on a systematic evaluation of the adsorption behavior and underlying mechanisms involved in the removal of Remazol Black dye using quaternary ammonium-based adsorbents. The objectives are to (i) evaluate adsorption performance under varying operational parameters, (ii) elucidate adsorption mechanisms through isotherm, kinetic, and thermodynamic modeling, and (iii) correlate adsorption behavior with structural and surface characteristics before and after adsorption using SEM, FTIR, and BET analyses**.** The central hypothesis of this study is that the cationic functional groups of QAPs promote strong electrostatic and chemical interactions with Remazol Black dye, leading to monolayer-dominated adsorption governed by chemisorption mechanisms, even in complex wastewater matrices. By addressing these aspects, the study seeks to generate new mechanistic insights into polymer-based dye adsorption and to fill the existing knowledge gap regarding QAP performance in realistic textile wastewater systems.

MATERIALS AND METHODS

Materials

All chemicals employed in this study were of analytical grade and utilized directly without additional purification. The monomer 2-(methacryloyloxy)ethyl trimethylammonium chloride (META) was supplied by Sigma-Aldrich. 2,2′-azobisisobutyronitrile (AIBN, $[(CH_3)_2C(CN)]_2N_2$) served as the radical initiator, while ethylene dimethacrylate (EDMA, $C_{10}H_{14}O_4$), was used as the crosslinking agent; both reagents were obtained from TCI (China). Ethanol (C_2H_6O) and poly(ethylene glycol) (PEG, $H(OCH_2CH_2)_nOH$, M_n=400) were also obtained from TCI. Isopropyl alcohol (IPA, C_3H_8O; Wako 1st Grade, Japan) was employed as the porogenic solvent.

Polymer preparation

A schematic illustration outlining the sequential steps of the polymer synthesis is shown in Figure 1. The polymer was prepared via a one-pot polymerization route using ethylene dimethacrylate (EDMA, 0.375 mL) as the crosslinking agent and 2-(methacryloyloxy)ethyl trimethylammonium chloride (META, 1.25 mL) as the functional monomer. Polymerization was initiated by adding 0.005 g of 2,2′-azobisisobutyronitrile (AIBN) as a free-radical initiator. Poly(ethylene glycol) (PEG, M_n = 400; 1.4 mL) was introduced as a co-solvent, while ethanol (1.25 mL) and 75 mL of isopropyl alcohol (IPA) were employed as porogenic solvents. The reaction mixture was first homogenized and subsequently subjected to thermal polymerization in a water bath maintained at 70 °C for 24 h (Rahayu et al., 2023a). Upon completion of the reaction, the formed polymer monolith was removed from the mold and mechanically fragmented into smaller pieces. To remove unreacted monomers and residual porogens, the polymer was repeatedly rinsed with ethanol until the washing solution became transparent. The cleaned polymer was then dried in an oven at 50 °C until a constant mass was achieved. A schematic representation of the proposed polymerization mechanism using the one-pot synthesis strategy is provided in in Figure 2.

of META and EDMA adsorption of Remazol Black dye

The adsorption behavior of the synthesized quaternary ammonium polymer was examined using Remazol Black as a representative dye contaminant commonly found in textile effluents. Batch adsorption experiments were conducted by bringing the polymer adsorbent into contact with dye solutions under a range of operating conditions in order to identify optimal adsorption parameters. The investigated variables included adsorbent dosage (0.10, 0.15, 0.20, 0.25, and 0.30 g), contact duration (10, 20, 30, 40, 50, and 60 min), reaction temperature (25, 30, 40, 50, 60, and 70 °C), initial dye concentration (10, 20, 30, 40, and 50 ppm), and wastewater composition (10, 20, 40, 50, and 100%). Following the adsorption experiments, the remaining concentration of Remazol Black in the aqueous phase was quantified using UV–visible spectrophotometry. Adsorption performance was assessed in terms of dye removal percentage and adsorption capacity of the polymer. The reliability of the adsorption process was further confirmed through run-to-run and day-to-day reproducibility evaluations. To investigate changes in the physicochemical characteristics of the polymer resulting from dye uptake, the adsorbent was analyzed both prior to and after adsorption using scanning electron microscopy (SEM), Brunauer-Emmett-Teller (BET) surface

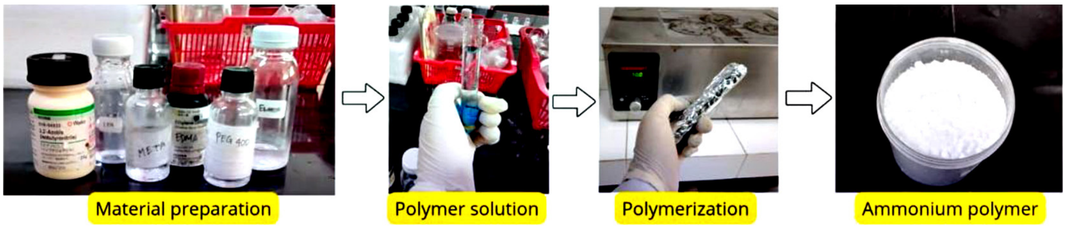


Figure 1. Step-by-step process of polymer preparation

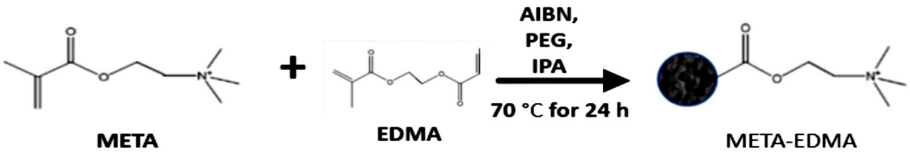


Figure 2. Schematic representation of the proposed polymerization mechanism

area measurements, and Fourier-transform infrared (FTIR) spectroscopy. SEM analysis was employed to examine surface morphology, while BET analysis provided insights into variations in surface area and pore structure. FTIR spectroscopy was used to identify possible interactions between functional groups on the polymer surface and dye molecules. A schematic illustration of the adsorption procedure using the quaternary ammonium polymer is presented in Figure 3.

The removal of Remazol Black (RB) dye from both synthetic aqueous media and textile wastewater was investigated using quaternary ammonium polymers (QAPs) as adsorbent materials. Batch adsorption tests were performed by contacting 0.10 g of the QAP adsorbent with 25 mL of RB dye solution. Adsorption isotherm analysis was applied to evaluate the maximum adsorption capacity (q_{max}) of the polymer toward the dye. The adsorption capacity at equilibrium (q_e) and efficiency *(RE*) at equilibrium were calculated according to the following expressions:

$$q_e = \frac{(C_0 - C_e) \times V}{W} \tag{1}$$

$$RE = \frac{(C_0 - C_e) \times 100\%}{C_0} \tag{2}$$

where: C_0 and C_e denote the initial and equilibrium concentrations of Remazol Black in solution (mg/L), W represents the mass of the adsorbent (g), and V is the volume of the solution (L).

Adsorption experiments were conducted at varying times (10, 20, 30, 40, 50, and 60 minutes) To assess the influence of contact duration, adsorption experiments were carried out over a range of interaction times (10, 20, 30, 40, 50, and 60 min). The concentration of RB remaining in solution before and after adsorption was determined using UV-visible spectrophotometry, and the corresponding removal efficiencies were subsequently calculated based on the measured concentration changes.

Adsorption isotherm estimation

Freundlich

The Freundlich isotherm model is commonly employed to assess adsorption behavior under equilibrium conditions. This empirical model is particularly suitable for systems involving heterogeneous adsorbent surfaces, where adsorption sites exhibit different affinities toward the adsorbate and multilayer adsorption may occur. In contrast to the Langmuir model, which assumes uniform surface sites and monolayer coverage, the Freundlich isotherm accommodates variations in adsorption energy and a non-homogeneous distribution of binding sites across the adsorbent surface. The correlation between the equilibrium concentration of the adsorbate and the quantity adsorbed is described by the Freundlich isotherm Equation 3.

$$\log qe = \log k + \frac{1}{n} \log Ce \tag{3}$$

where: q_e (mg g^{-1}) represents the amount of adsorbate adsorbed per unit mass of adsorbent, and C_e (mg L^{-1}) denotes the equilibrium concentration of the adsorbate in solution. K_F ((mg g^{-1})(L mg^{-1})$^{1/n}$) is the Freundlich constant indicative of adsorption capacity, while n represents the adsorption intensity reflecting the heterogeneity of the surface.

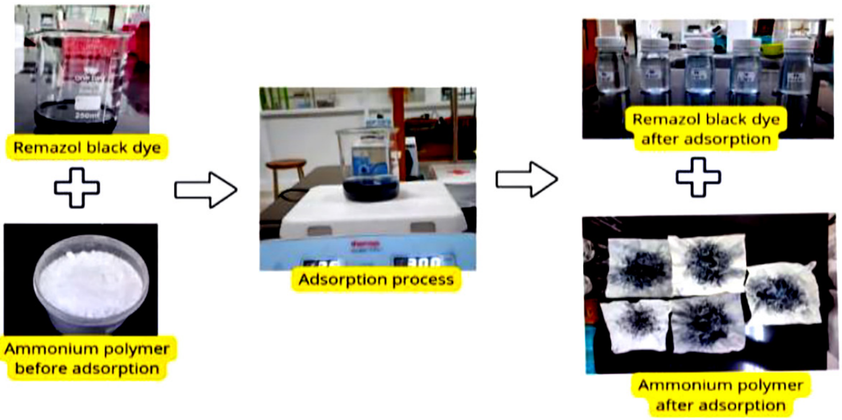


Figure 3. Dye adsorption using the quaternary ammonium polymer adsorbent

When $1/n = 0$, the adsorption process is irreversible; values of $1/n$ between 0 and 1 indicate favorable adsorption, whereas $1 > n$ suggests an unfavorable adsorption process (Alam et al., 2021; Murtihapsari, 2012; Musah et al., 2022).

Langmuir isotherm

The Langmuir isotherm describes an adsorption process in which adsorbate molecules form a single molecular layer on a uniform adsorbent surface composed of identical and energetically equivalent active sites. Under this assumption, each adsorption site can accommodate only one adsorbate molecule, and once occupied, no additional adsorption can occur at that location. This model is therefore well suited for adsorption systems that exhibit a finite adsorption capacity associated with complete surface saturation. The linearized Langmuir equation is given in Equation 4.

$$\frac{Ce}{qe} = \frac{1}{Kl.qm} + \left(\frac{1}{qm}\right).Ce \tag{4}$$

where: q_e (mg g^{-1}) is the amount of adsorbate adsorbed per unit mass of adsorbent at equilibrium, C_e (mg L^{-1}) is the equilibrium concentration of the adsorbate, K_L (L mg^{-1}) is the Langmuir adsorption constant related to the rate of adsorption, and q_m (mg g^{-1}) represents the theoretical maximum adsorption capacity (Harahap et al., 2018; Ho, 2004).

A higher q_m value indicates a greater adsorption capacity, while K_L reflects the affinity between the adsorbate and the adsorbent surface.

Temkin isotherm

The Temkin isotherm model assumes that the heat of adsorption of all molecules in the layer decreases linearly with coverage due to adsorbate–adsorbent interactions. This model considers a uniform distribution of binding energies up to a certain maximum energy. The linear form of the Temkin equation is given in Equation 5.

$$qe = \frac{RT}{B_T} \ln K_T + \frac{RT}{B_T} \ln C_e \tag{5}$$

where: q_e (mg g^{-1}) is the amount of adsorbate adsorbed at equilibrium, C_e (mg L^{-1}) is the equilibrium concentration of adsorbate, R is the universal gas constant (8.314 J mol^{-1} K^{-1}), T (K) is the absolute temperature, B_T (J mol^{-1}) is the Temkin constant related to the heat of adsorption, and K_T (L g^{-1}) is the Temkin equilibrium binding constant (Temkin, 1941; Benmessaoud, 2020).

This model helps to identify the influence of adsorbate–adsorbent interactions on adsorption capacity and energy distribution (Temkin, 1941; Benmessaoud, 2020).

Dubinin-Radushkevich (D-R) isotherm

The Dubinin-Radushkevich (D–R) isotherm model is frequently applied to describe adsorption mechanisms involving a Gaussian energy distribution over heterogeneous surfaces. It is particularly useful for distinguishing between physical and chemical adsorption processes. The linearized D–R isotherm equation is represented in Equation 6.

$$ln\ qe = ln\ qm - K_{DR}\ \varepsilon^2 \tag{6}$$

where: q_e (mg g^{-1}) is the amount of adsorbate adsorbed at equilibrium, q_m (mg g^{-1}) is the theoretical maximum adsorption capacity, K_{DR} (mol^2 kJ^{-2}) is the D–R constant related to the adsorption energy, and ε represents the Polanyi potential, calculated as $\varepsilon = RT\ln(1 + 1/C_e)$.

The mean adsorption energy (E) can be estimated from K_{DR} using the relation $E = (2K_{DR})^{-0.5}$, where $E < 8$ kJ mol^{-1} indicates physical adsorption, and $E > 8$ kJ mol^{-1} suggests chemical adsorption (Benmessaoud et al., 2020; Dada et al., 2012).

Adsorption kinetic estimation

Pseudo-first-order model

Kinetic analysis provides valuable insight into the adsorption mechanism and rate-determining steps of sorption processes. The reaction rate constant is a key parameter in understanding the efficiency and dynamics of adsorption. The pseudo-first-order kinetic model, proposed by Lagergren, is one of the most widely used equations for describing adsorption in liquid–solid systems. The model assumes that the rate of occupation of adsorption sites is proportional to the number of unoccupied sites. The linearized form of the pseudo-first-order equation is expressed in Equation 7.

$$\ln(q_e - q_t) = \ln q_e - k_1.t \quad (7)$$

where: q_e (mg g^{-1}) represents the amount of adsorbate adsorbed at equilibrium, q_t (mg g^{-1}) denotes the amount of adsorbate adsorbed at time t (min), and k_1 (min^{-1}) is the pseudo-first-order rate constant.

The values of q_e and k_1 can be determined from the slope and intercept of the plot of ln (q_e – q_t) versus t. The correlation coefficient (R^2) is used to evaluate the model's ability to describe the experimental kinetic data.

Pseudo-second-order model

The pseudo-second-order kinetic model, often referred to as the Blanchard model, assumes that adsorption involves chemisorption as the rate-limiting step, which may occur through electron-sharing or electron-exchange between the adsorbate and the adsorbent (Benjelloun et al., 2021). The model is expressed by the following linearized Equation 9.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (8)$$

where: q_e (mg g^{-1}) and q_t (mg g^{-1}) are the amounts of adsorbate adsorbed at equilibrium and at time t (min), respectively, and k_2 (g mg^{-1} min^{-1}) is the pseudo-second-order rate constant (Benjelloun et al., 2021).

The parameters q_e and k_2 can be determined from the slope and intercept of the plot of t/qt versus t. A higher correlation coefficient (R^2) indicates a better fit of the experimental data to the pseudo-second-order model, suggesting that the adsorption process may be controlled by chemisorption involving valence forces, such as electron sharing or exchange between the adsorbent and the adsorbate.

Thermodynamic approach

The thermodynamic parameters of the adsorption process were determined by studying the effect of temperature on the equilibrium distribution of Remazol Black dye between the solution and the quaternary ammonium polymer adsorbent. Batch adsorption experiments were conducted at different temperatures (25, 30, 40, 50, 60, and 70 °C) under constant initial dye concentration, adsorbent dosage, and pH. After reaching equilibrium, the supernatant was separated by filtration, and the residual dye concentration was measured spectrophotometrically at the maximum wavelength (λ_max) of Remazol Black. The adsorption distribution coefficient (K_d), which was calculated using Equation 9.

$$K_d = \frac{C_a}{C_e} \quad (9)$$

where: C_a (mg/L) is the amount of dye adsorbed per liter of solution and C_e (mg/L) is the equilibrium dye concentration.

The values of ln K_d were plotted against 1/T (K^{-1}) according to the Van't Hoff Equation 10.

$$In\, K_d = -\frac{\Delta H^o}{R}\left(\frac{1}{T}\right) + \frac{\Delta S^o}{R} \quad (10)$$

The enthalpy (ΔH°) and entropy (ΔS°) changes were obtained from the slope and intercept of the linear plot, respectively, while the Gibbs free energy change (ΔG°) was calculated using Equation 11.

$$\Delta G^o = \Delta H^o - T\Delta S^o \quad (11)$$

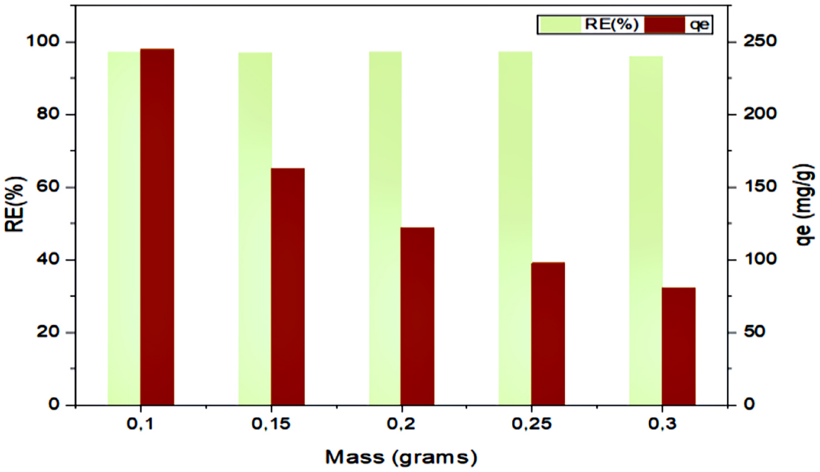
This approach allowed determination of the spontaneity, endothermic or exothermic nature, and entropy change associated with the adsorption of Remazol Black onto the polymeric adsorbent.

RESULT AND DISCUSSION

Influence of adsorbent dosage on Remazol Black removal

The quantity of adsorbent is a key parameter affecting adsorption performance, as it directly influences the availability of active sites for dye uptake. Accordingly, the effect of quaternary ammonium polymer dosage was investigated by varying the adsorbent mass from 0.10 to 0.30 g, while maintaining a constant stirring speed of 300 rpm and a contact time of 20 min at ambient temperature. The impact of adsorbent mass on dye removal efficiency (RE, %) and equilibrium adsorption capacity (q_e) for Remazol Black is illustrated in Figure 4. The corresponding values were calculated using Equations 1 and 2, as described in the Methods section.

As depicted in Figure 4, changes in adsorbent dosage significantly affected the adsorption behavior of Remazol Black. In aqueous model solutions, the maximum removal efficiency of 97.27% was achieved at an adsorbent mass of 0.10 g. In contrast, experiments conducted using textile



Mass (grams)	0.1	0.15	0.2	0.25	0.3
RE (%)	97.27%	97.03%	97.20%	97.15%	95.97%
Qe (mg/g)	244.60	162.80	122.24	97.76	80.81

Figure 4. Effect of increasing adsorbent mass on Remazol black adsorption

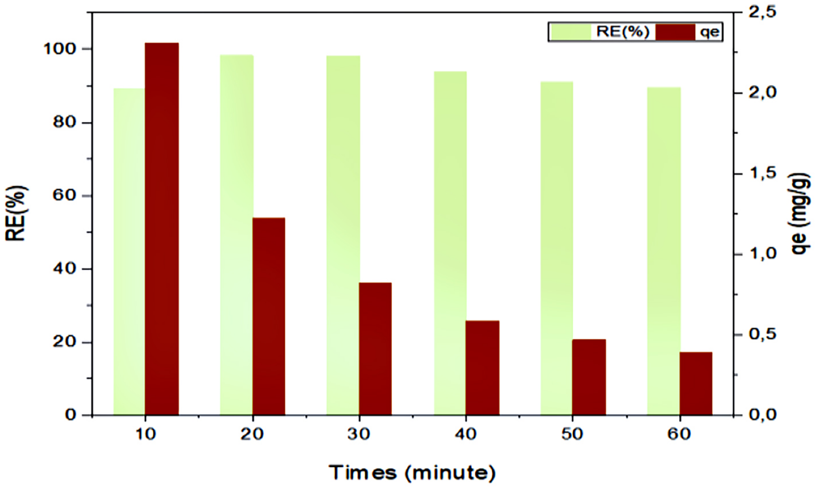
wastewater showed that a higher adsorbent dosage of 0.25 g was required to attain the optimal removal efficiency, reaching of 95.28%. This difference suggests that the presence of competing constituents and matrix complexity in real wastewater systems influences the adsorption process, necessitating a greater amount of adsorbent to achieve comparable dye removal. As illustrated in Figure 4, the amount of adsorbent exerted a pronounced effect on the adsorption performance of Remazol Black. Increasing the adsorbent dosage led to an initial rise in dye removal efficiency, which reached an optimum at a certain mass. Beyond this point, further addition of adsorbent did not produce a commensurate improvement in removal efficiency. This trend is clearly reflected in the graphical data, where the equilibrium adsorption capacity (q_e) tends to level off with increasing adsorbent mass, suggesting that the available adsorption sites gradually become saturated. Overall, the results demonstrate a clear correlation between adsorbent dosage and dye removal behavior.

The reduction in adsorption capacity (q_e) observed at higher adsorbent dosages can be attributed to particle aggregation phenomena. At elevated concentrations, adsorbent particles are more likely to agglomerate, leading to a decrease in the effective surface area and reduced accessibility of active adsorption sites. Such aggregation can also hinder the diffusion of dye molecules into internal pores, thereby diminishing adsorption efficiency despite the presence of a larger adsorbent mass (Istghfarini et al., 2017; Reyra et al., 2003; Ristianingsih and Lestari, 2025). Similar behavior was reported by (Samarghandy et al., 2011) who observed a substantial decline in the adsorption capacity of a potato peel–based bio adsorbent for Remazol Black, from 205.25 mg g^{-1} to 47.53 mg g^{-1} as the adsorbent mass increased from 0.2 to 1.0 g. In contrast, adsorption capacities obtained from textile wastewater samples were comparatively lower and showed minimal variation with changes in adsorbent mass. This behavior is likely associated with the complex composition of textile wastewater, which contains a mixture of dyes along with various organic and inorganic constituents. These components may compete for adsorption sites or interfere with electrostatic interactions between the quaternary ammonium polymer and dye molecules, ultimately limiting adsorption performance (Herawati et al., 2018).

Effect of contact time on Remazol Black adsorption

The contact time between the adsorbent and the adsorbate is a key parameter that influences adsorption efficiency. Figure 5 illustrates the effect of contact time on the removal efficiency (RE, %) and adsorption capacity (q_e) of Remazol Black by the quaternary ammonium polymer. The experiments were conducted using a polymer mass of 0.10 g at a stirring speed of 300 rpm under ambient conditions.

As shown in Figure 5, the adsorption of Remazol Black increased rapidly during the initial stages of contact due to the high availability of active adsorption sites on the polymer surface. The



Times (minute)	10	20	30	40	50	60
RE (%)	89.14%	98.29%	98.12%	93.88%	91.01%	89.51%
qe (mg/g)	2.31	1.22	0.82	0.58	0.47	0.39

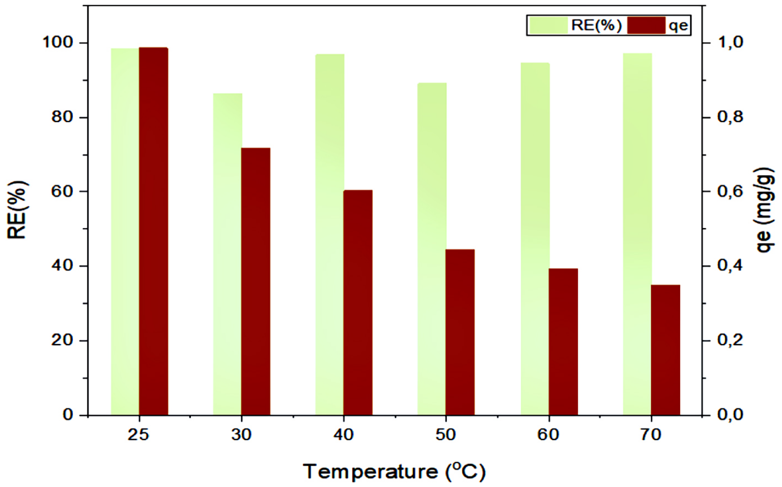
Figure 5. Effect of increasing contact time on Remazol Black adsorption

maximum removal efficiency in aqueous solution was achieved at 20 minutes, with an adsorption percentage of 98.29%. For textile wastewater, the optimal contact time was 30 minutes, yielding a removal efficiency of 95.64%. Beyond these times, the removal efficiency and adsorption capacity gradually decreased, suggesting that equilibrium had been reached and the active sites were saturated. At prolonged contact durations, desorption phenomena may occur as dye molecules detach from the polymer surface and re-enter the solution phase (Irwandi et al., 2015; Patel, 2021). Additionally, extended contact time can lead to partial degradation or structural alteration of the quaternary ammonium polymer due to prolonged exposure to aqueous conditions, thereby reducing the overall adsorption capacity (Satyam and Patra, 2024). The optimum contact time thus represents a balance between the adsorption rate and the onset of desorption or material degradation. Determining this balance is crucial for achieving high pollutant removal efficiency while maintaining the structural stability and reusability of the adsorbent in subsequent adsorption cycles.

Effect of temperature on Remazol Black adsorption

Temperature is a critical factor influencing the adsorption efficiency of Remazol Black onto quaternary ammonium polymers. The optimization of adsorption temperature was performed to evaluate its impact on dye removal performance. Experiments were conducted at a stirring speed of 300 rpm for 20 minutes using 0.10 g of adsorbent at various temperatures (25, 30, 40, 50, 60, and 70 °C). The effect of temperature on the removal efficiency (RE, %) and adsorption capacity (q_e) is presented in Figure 6.

As shown in Figure 6, an increase in temperature led to a noticeable decline in both the removal efficiency and adsorption capacity of the quaternary ammonium polymer. The highest adsorption efficiency (98.46%) was achieved at 25 °C for the aqueous solution, whereas in textile wastewater, the optimum temperature was 30 °C, with a removal efficiency of 95.64%. Beyond these temperatures, a decrease in adsorption efficiency was observed, indicating that the adsorption process is exothermic. The decline in adsorption performance with increasing temperature can be attributed to the increased kinetic energy of the dye molecules, which weakens the adsorptive interactions between the polymer surface and the dye, promoting desorption (Wijayanti and Kurniawati, 2019). Elevated temperatures also increase molecular mobility, thereby reducing the residence time of dye molecules at active sites. This behavior confirms that the adsorption of Remazol Black onto the quaternary ammonium polymer occurs via an exothermic mechanism, in which higher temperatures diminish the binding strength between the dye and the adsorbent surface (Muther and Dahaghi, 2025). These results are consistent with previous studies reporting the exothermic adsorption of anionic dyes (Alghamdi



Temperature (°C)	25	30	40	50	60	70
RE (%)	98.46%	86.47%	96.83%	89.15%	94.57%	97.14%
qe (mg/g)	0.99	0.72	0.60	0.44	0.39	0.35

Figure 6. Effect of increasing temperature on Remazol Black adsorption

and Mannoubi, 2021; Khalaf et al., 2021) where anionic dyes typically act exothermically. Similarly, (Qatta and Mohammad, 2021) observed that increasing temperature reduced the adsorption efficiency of methylene orange and methylene blue onto sand-based adsorbents, further supporting the exothermic nature of the adsorption process.

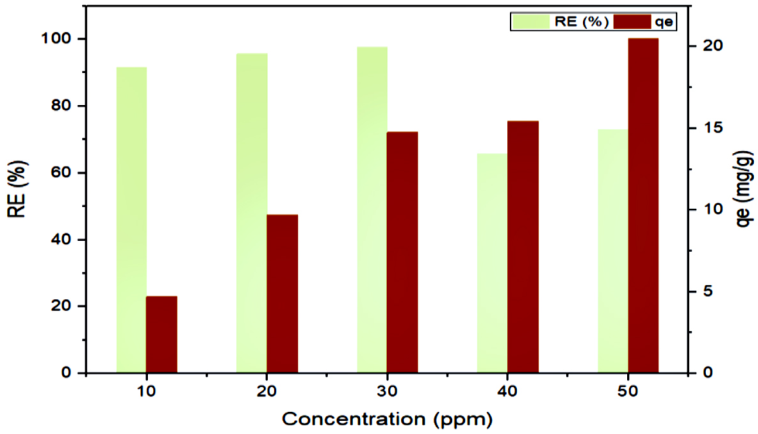
Effect of initial concentration on Remazol Black adsorption

The initial dye concentration is an essential factor influencing the adsorption performance of the quaternary ammonium polymer. In this study, the adsorption behavior was evaluated at different initial concentrations of Remazol Black (10, 20, 30, 40, and 50 ppm) under constant operating conditions. Figure 7 presents the relationship between the initial dye concentration and both the removal efficiency (RE, %) and adsorption capacity (q_e).

As shown in Figure 7, the initial concentration significantly affected the adsorption efficiency of Remazol Black onto the quaternary ammonium polymer. The highest removal efficiency of 97.65% was obtained at an initial concentration of 30 ppm. In comparison, the optimum adsorption in textile wastewater (10% dilution) was achieved at an initial concentration of 14.70 ppm, corresponding to a removal efficiency of 94.46%. At lower dye concentrations, a high proportion of the polymer surface's active sites was accessible, resulting in efficient adsorption. However, as the initial dye concentration increased, competition among dye molecules for active sites became more pronounced, resulting in a decline in removal percentage once surface saturation occurred (Njeri et al., 2023). This behavior can be explained by the increased number of dye molecules in the system, which enhances the likelihood of collisions between dye molecules and the polymer surface. Nevertheless, after the saturation of adsorption sites, additional dye molecules remain unadsorbed, decreasing the overall removal efficiency. In contrast, the adsorption capacity (q_e), which increased with higher initial concentrations, indicating that more dye molecules were adsorbed per unit mass of polymer until the available sites were fully occupied. Similar trends have been reported for the adsorption of Reactive Red 198 using $ZnCl_2$-activated pistachio hull waste and Acid Orange 7 using Kenya tea pulp ash (Bazrafshan et al., 2013; Saeidi et al., 2017). The concurrent increase in adsorption capacity and decline in removal efficiency with increasing concentration confirms the existence of competitive adsorption among dye molecules for the limited active sites, a behavior commonly observed in heterogeneous adsorption systems operating under fixed adsorbent dosage.

Equilibrium isotherms

Adsorption isotherms provide critical insights into the interaction between adsorbate molecules and the adsorbent surface at equilibrium. They describe the relationship between the amount of solute adsorbed per unit mass of adsorbent and its equilibrium concentration in the liquid phase



Concentrartion (ppm)	10	20	30	40	50
RE (%)	91.49%	95.69%	97.65%	65.72%	72.98%
qe (mg/g)	4.72	9.71	14.76	15.43	20.50

Figure 7. Effect of initial dye concentration on Remazol Black adsorption

at a constant temperature (Rahayu et al., 2025). The analysis of adsorption isotherms is essential for understanding the adsorption mechanism, optimizing system design, and accurately predicting adsorbent performance under varying conditions (Batool et al., 2018; Wang and Guo, 2020). In this study, four isotherm models (Freundlich, Langmuir, Temkin, and Dubinin-Radushkevich) were applied to evaluate the adsorption behavior of Remazol Black onto the quaternary ammonium polymer. The linearized plots for these models are shown in Figure 8.

As shown in Figure 8, the Langmuir model exhibited the best fit to the experimental data, with the highest correlation coefficient (R^2 = 0.9995) compared to the Freundlich (R^2 = 0.6964), Temkin (R^2 = 0.6898), and Dubinin-Radushkevich (R^2 = 0.8855) models. A similar trend was observed for the adsorption of Remazol Black from textile wastewater, where the Langmuir isotherm also showed the strongest linearity (R^2 = 1.0000). The strong agreement with the Langmuir model indicates that adsorption occurs via monolayer coverage on a homogeneous surface with identical adsorption sites, with negligible interaction between adsorbed molecules. This suggests that each binding site on the polymer surface can accommodate only one dye molecule, confirming uniform surface energetics and the formation of a single molecular layer. Similar findings have been reported for the adsorption of dyes such as Remazol yellow and other reactive dyes on chitosan-based and activated carbon adsorbents derived from orange peel (Annadurai et al., 2008; Bayrak and Uzgör, 2013; Musnamar et al., 2024). The Langmuir model's strong correlation supports the hypothesis that the quaternary ammonium polymer adsorbs Remazol Black primarily through specific active sites distributed uniformly on its surface, resulting in efficient monolayer adsorption.

Visual confirmation of the equilibrium stage was observed through the fading of the dye color in the solution, as shown in Figure 8. The image presents bottles containing RB dye solutions at different contact times (10, 20, 30, 40, 50, and 60 minutes). As the contact time increased, the dye concentration in the solution decreased, with a noticeable fading in color after 30 minutes, indicating the adsorption process was progressing. At 60 minutes, the solution became nearly clear, confirming that most of the dye had been adsorbed onto the polymer, and equilibrium was reached. These visual changes were further supported by SEM image (Figure 9).

Chemical component

The chemical structure and functional group interactions of the quaternary ammonium polymer before and after dye adsorption were analyzed using Fourier transform infrared spectroscopy (FTIR), as shown in Figure 10. The FTIR spectra provide insight into the surface chemistry and confirm the involvement of specific functional groups in the adsorption of RB dye. Before adsorption, the polymer spectrum exhibited characteristic peaks at 1094 cm^{-1} (C–O stretching), 1243 cm^{-1} (C–N stretching), 1647 cm^{-1} (N–H bending), 1719 cm^{-1} (C=O stretching), 2878 cm^{-1} (C–H stretching), and 3400 cm^{-1} (O–H stretching).

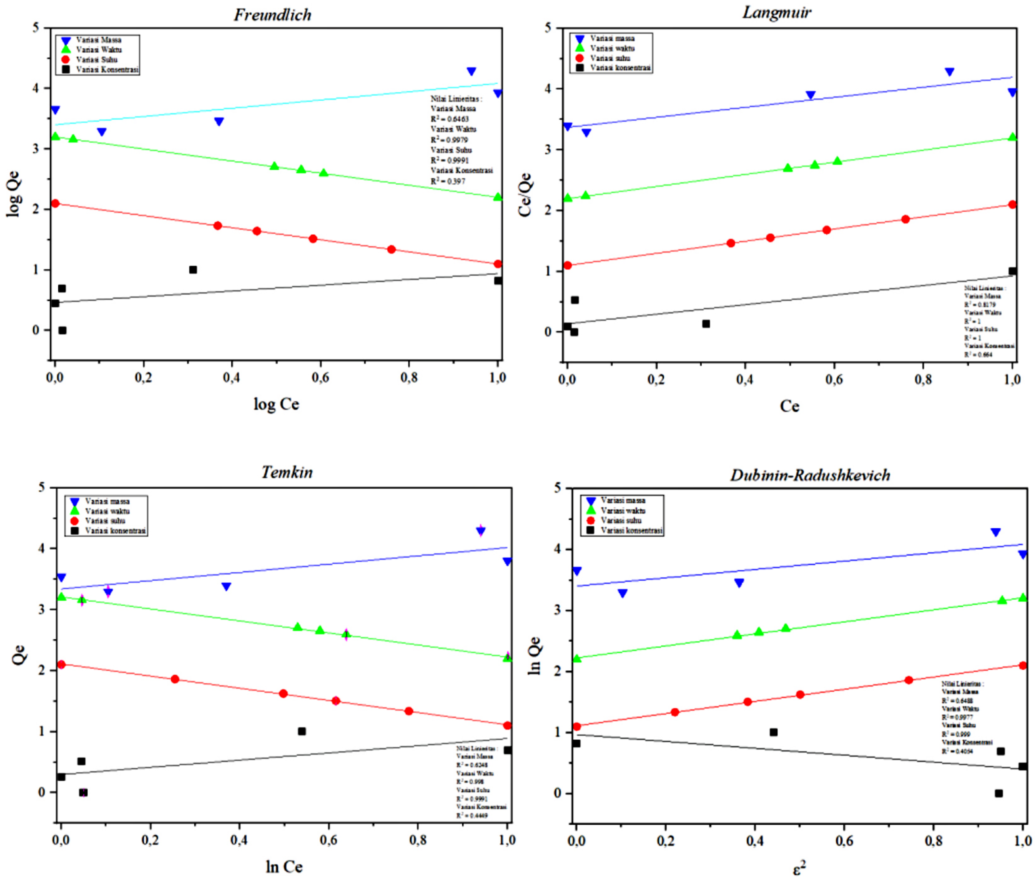


Figure 8. Comparative normalized representations of Remazol Black adsorption using (a) Freundlich, (b) Langmuir, (c) Temkin, and (d) Dubinin-Radushkevich

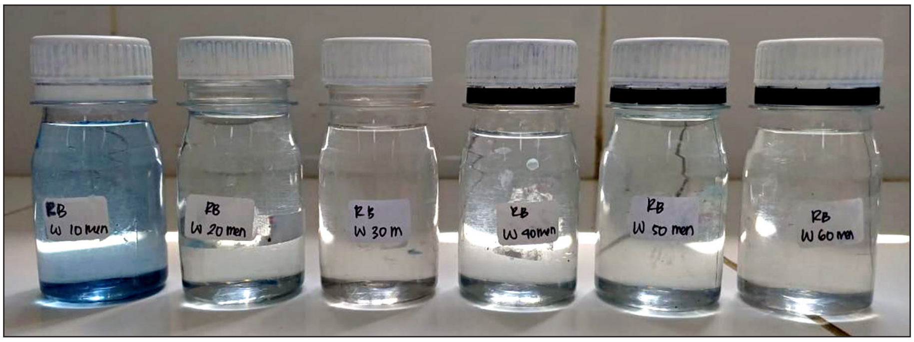


Figure 9. Photographic observation of Remazol Black solution appearance at varying contact durations

These bands are typical of quaternary ammonium-based polymers and confirm the presence of amine, hydroxyl, and carbonyl groups, which are responsible for electrostatic and hydrogen-bonding interactions. After the adsorption of Remazol Black dye, distinct spectral changes were observed. The appearance and enhancement of absorption bands at 1530–1543 cm^{-1}, corresponding to the azo group (–N=N–) of the dye, confirm successful attachment of the dye to the polymer surface. This shift indicates the formation of chemical interactions, likely through electrostatic attraction and hydrogen bonding between the dye molecules and cationic sites of the polymer.

Moreover, a decrease in the intensity or disappearance of the C–N stretching band (1243 cm^{-1}) was observed after adsorption, particularly in the polymer sample treated with textile wastewater. This suggests that the C–N functional sites actively participated in adsorption, possibly forming complexes with dye anions and other co-existing pollutants. The observed modifications

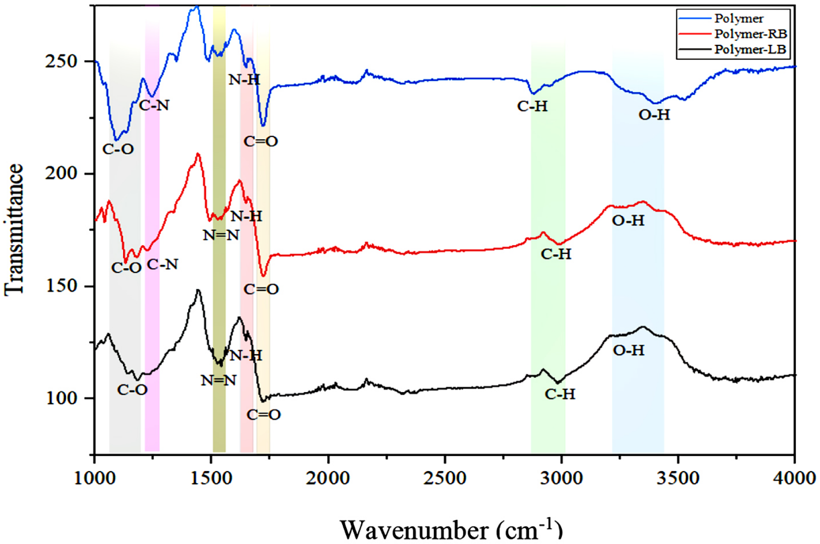


Figure 10. FTIR spectra of the quaternary ammonium polymer recorded before and after Remazol Black adsorption

imply that the adsorption process involves not only physical interactions but also chemical rearrangements at the polymer surface. These findings demonstrate that the quaternary ammonium polymer effectively interacts with the dye via electrostatic attraction, hydrogen bonding, and possible ion-exchange mechanisms, leading to significant alterations in its functional group environment. Similar FTIR spectral changes have been reported for other cationic polymer–azo dye systems (Silva et al., 2012; Kaur et al., 2024; Obulapuram et al., 2021), supporting the conclusion that adsorption occurs through multiple physicochemical interactions.

Morphological and surface characterization

Morphological analysis using scanning electron microscopy (SEM) was conducted to observe the surface characteristics of the quaternary ammonium polymer before and after dye adsorption. The SEM micrographs are presented in Figure 11.

As shown in Figure 11, the surface morphology of the polymer after adsorption exhibits aggregated and granular structures, indicating that dye molecules have attached to and partially filled the polymer pores. The observed macropore sizes of the polymer after adsorption were 0.42–1.42 µm in aqueous solution and 0.45–1.24 µm in textile wastewater. The union sizes were 0.55–1.47 µm and 0.55–1.38 µm, respectively. In comparison, the unmodified polymer exhibited larger macropores (0.83–2.41 µm) and union sizes (1.00–2.08 µm) before adsorption (Rahayu et al., 2023b). The reduction in both macropore and union sizes after adsorption suggests partial blockage of pore openings by dye molecules, confirming the successful adsorption of Remazol Black onto the polymer surface. The pore characteristics of the quaternary ammonium polymer before and after adsorption, determined by BET analysis, are summarized in Table 1.

BET analysis (Table 1) revealed significant changes in surface properties following dye adsorption. The pristine polymer exhibited a surface area of 3.32 m²/g, a total pore volume of 0.0006 mL/g, and an average pore size of 0.3495 nm, indicating a compact and microporous structure favorable for adsorption processes. After adsorption of Remazol Black from aqueous solution, the surface area decreased to 1.84 m²/g, while the total pore volume and average pore size increased to 0.0092 mL/g and 10.0388 nm, respectively. This suggests that dye molecules may have occupied the smaller pores, leading to surface restructuring and the apparent formation of larger pore openings. The reduction in surface area further confirms that dye molecules covered a portion of the active adsorption sites. Interestingly, after exposure to textile wastewater, the polymer showed increases in surface area (7.32 m²/g), total pore volume (0.0072 mL/g), and average pore size (1.9714 nm) compared to the pristine polymer. These changes may result from complex interactions between the polymer and wastewater constituents, including organic and inorganic

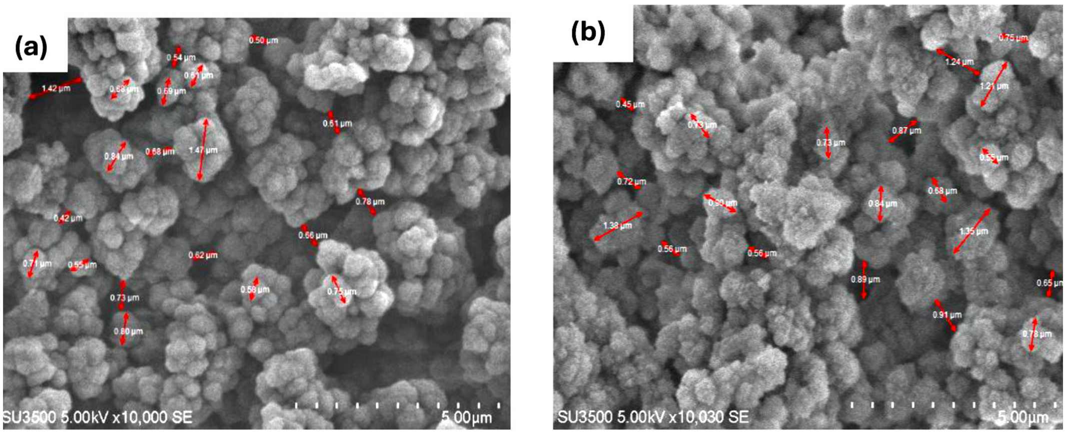


Figure 11. SEM images illustrating morphological changes of the quaternary ammonium polymer after adsorption of Remazol black from different solution matrices

Table 1. Surface structural properties of the quaternary ammonium polymer

Sample	Surface area (m²/g)	Total pore volume (mL/g)	Pore size (nm)
Polymer	3.32	0.0006	0.3495
Polymer-Remazol Black	1.84	0.0092	10.0388
Polymer-textile wastewater	7.32	0.0072	1.9714

compounds, which could induce surface etching or the formation of new pores, thereby increasing overall surface roughness.

Similar trends have been reported in previous studies. For example, Magdy et al., (2025) observed a decrease in BET surface area and pore volume of poly (DVB) from 249.47 to 93.54 m²/g and from 0.877 to 0.106 cm³/g, respectively, after dye adsorption. Such decreases strongly support the effective adsorption of dye molecules within both external and internal pores. The combined SEM and BET results in this study confirm that Remazol Black molecules were successfully adsorbed onto and within the porous structure of the quaternary ammonium polymer. This finding corroborates the adsorption mechanism inferred from isotherm modeling, suggesting that both surface binding and pore-filling dynamics govern the process.

Thermodynamic approach

The thermodynamic parameters for the adsorption of Remazol Black dye onto quaternary ammonium polymers were determined from the Van't Hoff plot (ln Kd vs. 1/T), with R-squared equal to 0.9997, which confirms the validity of the thermodynamic model that can be seen in Figure 12. From the slope and intercept of the plot, the enthalpy change (ΔH°) and entropy change (ΔS°) were calculated to be 65.37 kJ/mol and 0.21 kJ/mol·K, respectively. The Gibbs free energy change (ΔG°) values were subsequently obtained using the Gibbs-Helmholtz equation as summarized in Table 2. The ΔG° values decreased from +1.54 kJ/mol at 25 °C to -8.10 kJ/mol at 70 °C, indicating that the adsorption process shifts from nonspontaneous at lower temperatures to spontaneous at higher temperatures.

The positive ΔH° value (65.37 kJ/mol) confirms that the adsorption of Remazol Black onto quaternary ammonium polymers is endothermic, indicating that heat input promotes dye–adsorbent interactions. The positive ΔS° (0.21 kJ/mol.K) reflects an increase in randomness at the solid–liquid interface, caused by the release of water molecules and structural rearrangement of dye ions on the adsorbent surface. The negative ΔG° values at higher temperatures indicate that the process becomes spontaneous with increasing temperature, consistent with the endothermic nature of adsorption. At low temperatures (25–30 °C) the process is nonspontaneous but becomes favorable above 40 °C due to improved dye mobility and stronger electrostatic attraction. The adsorption is endothermic, entropy-driven, and spontaneous at elevated temperatures, dominated by physical and electrostatic interactions between the anionic dye and cationic polymer sites, in agreement with

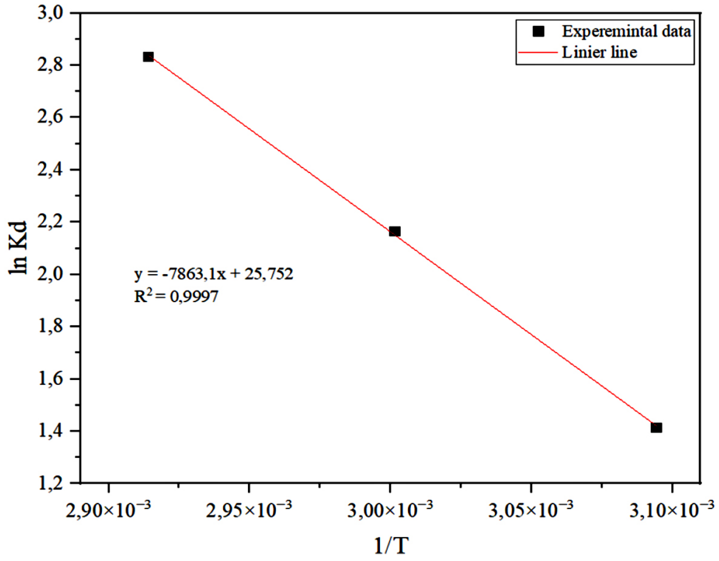


Figure 12. Van't Hoff analysis illustrating the thermodynamic behavior of Remazol Black adsorption on quaternary ammonium polymers

Table 2. Thermodynamic parameters obtained for Remazol Black adsorption on quaternary ammonium polymers

T (° C)	ΔH° kJ/mol	ΔS° kJ/mol.K	ΔG° kJ/mol
25	65.37	0.21	1.54
30			0.47
40			-1.67
50			-3.81
60			-5.95
70			-8.10

Table 3. Comparison of kinetic parameters obtained from pseudo-first-order and pseudo-second-order model fitting

Kinetic models	Parameter	Value
Pseudo-first order	q_e (mg/g)	0.18
	k_1 (min^{-1})	0.03
	R^2	0.13
Pseudo-second order	q_e (mg/g)	13.30
	k_2 (g/mg.min^{-1})	0.05
	R^2	1.00

previous reports on similar dye–polymer systems (Desnelli et al., 2022; Guerrero-fajardo et al., 2025; Obulapuram et al., 2021; Silva et al., 2012). The observed mechanism is consistent with the results reported by Kaur et al. (2024).

Kinetic analysis

The kinetics of dye adsorption are essential for understanding the rate of dye removal and the mechanisms governing the interaction between adsorbate and adsorbent at the solid–liquid interface (Markandeya et al., 2015). In this study, the adsorption kinetics of Remazol Black onto the quaternary ammonium polymer were analyzed using the pseudo-first-order and pseudo-second-order kinetic models, which describe the transfer of dye molecules from the liquid phase to the adsorbent surface. The kinetic parameters obtained from both models are summarized in Table 3.

The pseudo-second-order kinetic model shows a superior fit to the experimental data (R^2 = 1.00) compared to the pseudo-first-order model (R^2 = 0.13), indicating that the adsorption process follows pseudo-second-order kinetics. The higher rate constant (K_2) relative to (K_1) further supports this conclusion. These results suggest that Remazol Black adsorption onto the quaternary ammonium polymer is predominantly controlled by chemisorption, involving electron sharing or exchange between dye molecules and polymer functional groups (Ebelegi et al., 2020). Consequently, chemical interactions rather than mass transfer govern the rate-determining step.

CONCLUSIONS

Quaternary ammonium polymers are highly effective adsorbents for removing Remazol Black dye from both aqueous solutions and textile wastewater. The optimum adsorption conditions were determined to be a polymer mass of 0.10 g, a contact time of 20 minutes for synthetic solutions and 30 minutes for wastewater, an operating temperature of 25–30 °C, and an initial dye concentration of 30 ppm. Under these conditions, the adsorption efficiencies reached 97.65% and 95.64% for aqueous and wastewater samples, respectively. The adsorption behavior followed the Langmuir isotherm, indicating that the dye molecules formed a uniform monolayer on the polymer surface. FTIR characterization confirmed the presence of the azo (–N=N–) group after adsorption, verifying the dye's attachment to the polymer. SEM analysis revealed a reduction in pore size and surface roughness after adsorption, indicating that the dye molecules occupied the polymer's macropores. Furthermore, BET analysis showed that the polymer's surface area and pore structure changed significantly after adsorption, supporting the proposed surface interaction and pore-filling mechanism.

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