Exploring Isotherm Models for the Efficient Adsorption of Remazol Yellow on Polymer-Based Adsorbents

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ABSTRACT

Remazol Yellow is an azo dye widely used in the textile industry due to its vibrant color and ease of application. However, it poses serious environmental and health concerns. This dye contains benzene groups that are known to be toxic and carcinogenic. Additionally, its discharge into aquatic ecosystems can cause water turbidity and inhibit photosynthesis by blocking sunlight penetration, thereby disrupting the balance of aquatic life. Therefore, the effective treatment of dye-contaminated wastewater before it is released into the environment is essential. This study investigates the adsorption behavior of Remazol Yellow using quaternary ammonium polymers as adsorbents. The research focuses on determining the most suitable isothermal adsorption model by varying operational parameters, including contact time (10–60 minutes), temperature (25–70°C), and initial dye concentration (10–50 ppm). Dye concentrations in the filtrate were measured using a UV-Vis spectrophotometer, while surface morphology and pore structure of the adsorbent residue were analyzed using Scanning Electron Microscopy (SEM). The experimental data were fitted to four isothermal models: Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich. Among these, the Langmuir model showed the best fit with a linear correlation coefficient (R^2) of 1, indicating monolayer adsorption on a homogeneous surface. SEM analysis revealed that the quaternary ammonium polymers possess macropores, with pore sizes ranging from 0.34 to 1.35 µm at the optimum time and from 0.33 to 1.43 µm at the optimum temperature. Additionally, union sizes (aggregate clusters) ranged from 0.48 to 1.14 µm and 0.45 to 1.18 µm under the same respective conditions. These results confirm that quaternary ammonium polymers are effective for removing Remazol Yellow from wastewater, with the Langmuir isotherm providing the best description of the adsorption mechanism.

1. Introduction

Synthetic dyes are widely used in the textile industry. Its low price, wide range of colors, brighter colors, and extended durability are some of the reasons why it is popular in the textile industry [1], [2]. The use of synthetic dyes in the textile industry contributes significantly to environmental pollution, particularly in water sources. Therefore, it is necessary to treat textile industry liquid waste before it is discharged into the environment.

The most widely used dyes in industry are azo. Azo compounds and their derivatives, which often take the form of benzene groups, are one of the primary sources of natural contamination caused by colors. Azo substances can cause turbidity in water, be toxic and carcinogenic, and can inhibit photosynthesis in water. Some types of azo dyes, such as remazol, methylene, congo red, and so on [2]–[6]. One of the azo dyes widely used in Indonesia is Remazol Yellow. This dye is widely used because of its bright and long-lasting color. Remazol yellow is commonly used in the textile, batik, and weaving industries [7]–[10].

Various types of liquid waste treatment have been applied, such as photocatalytic, precipitation, adsorption, oxidation process, membrane filtration, flocculation, coagulation, and biological treatment [11]–[13]. The most widely applied method for treating liquid waste is the adsorption method. This method has a simple design, is cheap both in initial installation and operation, can adsorb pollutants in small concentrations, can adsorb various types of contaminants (heavy metals, organics, dyes, pathogens, and nutrients), and is easy to operate [14]–[16].

Polymers have been widely applied in various fields, including waste processing. Several studies have shown that polymers can adsorb pollutants efficiently [17]–[19]. Quaternary ammonium polymers are polymers made using the one-step method. The one approach method is an environmentally friendly, inexpensive, and efficient method of making polymers. Quaternary ammonium polymers have successfully adsorbed various pollutants as adsorption media through several studies. Research has been conducted on synthetic nitrate, synthetic nitrite, synthetic phosphate, phosphate applications on vinasse waste, and synthetic yellow remazol dye with adsorption percentages of 94.58%, 87.44%, 95.11%, 75.70% and 98.64%, respectively [2], [20], [21].

Previous research has been conducted on the efficiency and influence of variations in contact time, temperature, and concentration on the adsorption process of remazol yellow pollutants to quaternary ammonium polymers. This study aims to determine the adsorption interactions in Remazol Yellow with quaternary ammonium polymers by analyzing the isothermal approach that occurs.

2. Research Methodology

2.1. Materials

Materials come from different sources. The principal component was monomer 2-(methacryloyloxy) ethyl] trimethyl ammonium chloride solution $H_2C=C(CH_3)CO_2CH_2CH_2N(CH_3)_3Cl$ or META derived from Sigma Aldrich, crosslinker: ethylene dimethacrylate or EDMA ($C_{10}H_{14}O_4$) and AIBN or 2,2'-azobisisobutyronitrile ($[(CH_3)_2C(CN)]_2N_2$) were supplied by TCI (China), porogen: Isopropyl alcohol or IPA (C3H8O), poly (ethylene glycol) or PEG ($H(OCH_2CH_2)nOH$) with Mn = 400, ethanol (C_2H_6O), sourced from Wako 1st Grade, Japan. While the materials for making synthetic dye solutions were distilled water supplied from Yogyakarta, and Remazol Yellow was purchased from Jombor.

2.2. Synthesis of Ammonium Kuarterner Polymer

The polymer was made from 1.4 mL of PEG Mn=400 (polyethylene glycol 400), 0.375 mL of ethylene dimeth acrylate (EDMA), 1.75 mL of isopropyl alcohol (IPA), 1.25 mL of 2-[(methacryloyloxy)ethyl] trimethylammonium (META), 2,2-azobisisobutyronitrile (AIBN) 0.005 g, and 1.25 mL of homogenized ethanol. Then polymerization was carried out in a waterbath for 24 hours at a temperature of 70°C [20].

2.3. Isothermal Studies

Isothermal studies were conducted with time variations of 10-69 minutes, temperature variations of 25-70°C, and concentration variations of 10-50 ppm, and the amount of adsorbent used was 0.15 g [2]. After stirring for 20 minutes, the remaining concentration in the solution was determined by a spectrophotometer with a wavelength of 363 nm. Various isothermal models were applied to calculate the different values of adsorption parameters.

3. Results and Discussion

3.1. Isothermal Approach

Adsorption isotherm calculations are performed to comprehend and define the adsorption mechanism on a surface. Furthermore, insights derived from the adsorption isotherm methodology can assist in the development of an effective adsorption system by identifying the optimum operational conditions [22]. The calculated adsorption isotherm equation approaches are the Freundlich, Langmuir, Temkin, and Dubinin-Radushkevich. The Freundlich isotherm illustrates that adsorption occurs physically, so that the bonds are not strong and occur in several layers [23]. The Langmuir isotherm demonstrates that there is just one layer (monolayer) of substances sticking to the surface of the adsorbent, leading to the highest capacity of the adsorbent and no interaction among the molecules that are adsorbed and the nearby elements [24]. The Temkin isotherm indicates that as coverage increases, the heat of adsorption for all molecules in the layer decreases linearly with coverage. This change is due to the interactions between the adsorbent and the adsorbate, along with the characterization of adsorption based on the even distribution of binding energies that extends to the maximum binding energy [25]. The Dubinin-Radushkevich isotherm is rarely applied to liquid phase adsorption because it has a complex nature. This approach accepts heterogeneous surfaces with Gaussian energy distributions [26]. The linear equation of the Freundlich isotherm can be seen in equation 1 [27].

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

Where:
q = amount of adsorbate adsorbed on the surface (mg/g)
Ce = final concentration of remazol yellow after adsorption (mg/L)
n = empirical constant
k = freundlich constant

Based on Equation 1, a linear equation will be obtained with log q as the y-value and log C as the x-axis, with a slope value of 1/n and an intercept of log k (Sawyer et al., 1994). The Langmuir isothermal linear equation can be seen in equation 2 [28].

$$\frac{Ce}{qe} = \frac{1}{Kl \,.\, qm} + \left(\frac{1}{qm}\right).Ce \tag{2}$$

Where:
Kl = Langmuir constant
Ce = final concentration of remazol yellow after adsorption (mg/L)
qm = maximum adsorption capacity (mg/g)
qe = amount of adsorbate adsorbed on the surface (mg/g)

Based on equation 2, a linear equation will be obtained with Ce/qe as the y value and Ce as x, with a slope value of 1/qm and an intercept of 1/(Kl.qm). Temkin's isothermal linear equation can be seen in equation 3 [26].

$$qe = \frac{RT}{B_T} \ln K_T + \frac{RT}{B_T} \ln C_e \tag{3}$$

Where:
qe = amount of adsorbate adsorbed on the surface (mg/g)
Ce = final concentration of remazol yellow after adsorption (mg/L)
BT = constant related to heat of adsorption (J/mol)
R = gas constant (8.314 J/mol K)
T = temperature (K)
KT = Temkin equilibrium constant (L/g)

Based on equation 3, a linear equation will be obtained with qe as the y value and ln Ce as the x value, with slope RT/BT and intercept $(RT/BT)\ln K_T$. The Dubinin Radushkevich isothermal linear equation can be seen in equation 4 [26].

$$ln\ qe = ln\ qm - K_{DR}\ \varepsilon^2 \tag{4}$$

Where:
qe = amount of adsorbate adsorbed on the surface (mg/g)
qm = maximum adsorption capacity (mol/L)
K_{DR} = adsorption energy constant (mol^2/kJ^2)
ε = Polaniyi potential

Based on Equation 4, a linear equation will be obtained with ln qe as the y-value and ε2 as the x-value, having a slope of KDR and an intercept of ln qm.

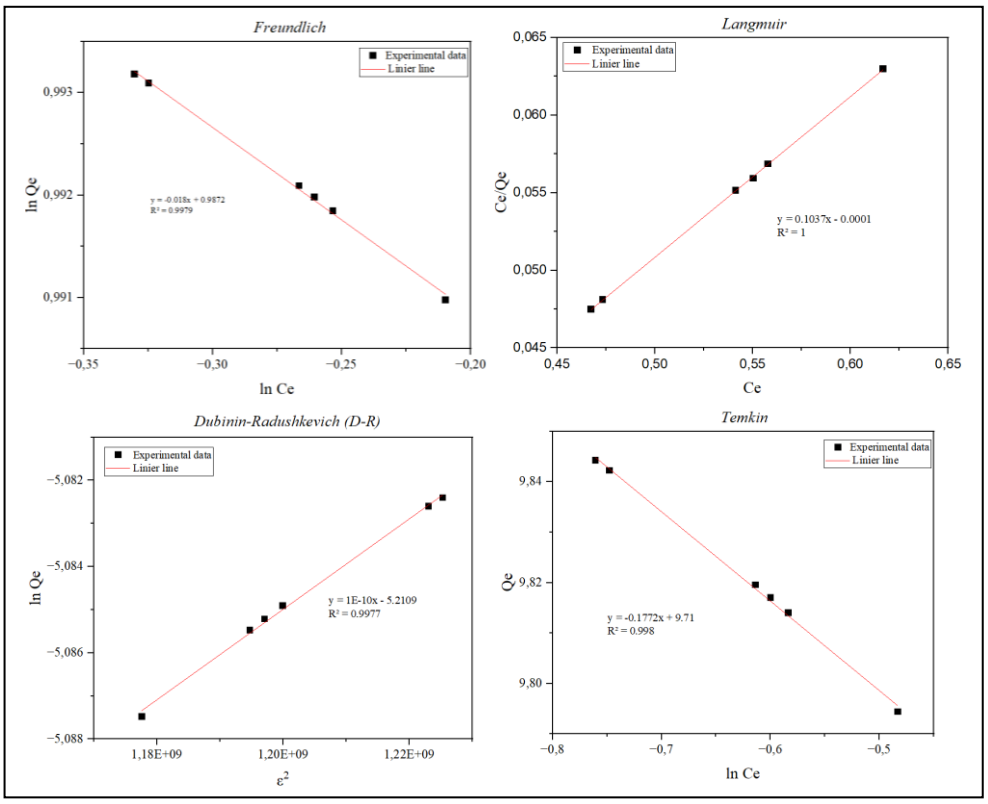

Fig. 1. Adsorption isothermal approach curves in variation of contact time

According to Fig. 1, the isothermal curves for the Freundlich, Langmuir, Temkin, and Dubinin-Radushkevich are depicted. In the variation of contact time, each isothermal approach obtained R^2 values of 0.9979, 1, 0.998, and 0.9977. The best isothermal approach to time variation is in the Langmuir approach. Because it has the highest linearity value, which is 1, furthermore, the isothermal approach to temperature variation can be seen in Fig. 2.

The variation of temperature, based on Fig. 2, yields isothermal curves for Freundlich, Langmuir, Temkin, and Dubinin-Radushkevich, each with a linear value of 0.9991, 1, 0.9991, and 0.999, respectively. The linearity value in Langmuir is highest at temperature variations. In this variation, the Langmuir approach is more suitable.

The change in concentration is illustrated in Fig. 3, which shows that the isothermal curves for Freundlich, Langmuir, Temkin, and Dubinin-Radushkevich yielded linear values of 0.397, 0.664, 0.4449, and 0.4054, respectively. Among these, the Langmuir model exhibits the highest linearity value across varying temperatures. Therefore, the Langmuir model is the most suitable for this temperature variation.

Based on these values, it can be seen that for the adsorption of remazol yellow solution dye with quaternary ammonium polymer as an adsorbent, the suitable isothermal illustration is Langmuir. This is because the R^2 value in the Langmuir isothermal approach is the largest of the others. The Langmuir isothermal model posits that adsorption takes place at designated binding sites situated on the adsorbent surface, asserting that every adsorption site on the adsorbent surface is uniform, the adsorbent surface is provided with a monolayer of adsorbed molecules, and there is an absence of interactions among the molecules adhered to the adsorbent surface [29]. The isothermal approach in

this study is in line with previous studies of quaternary ammonium polymers with Langmuir adsorption processes [21], [22], [30].

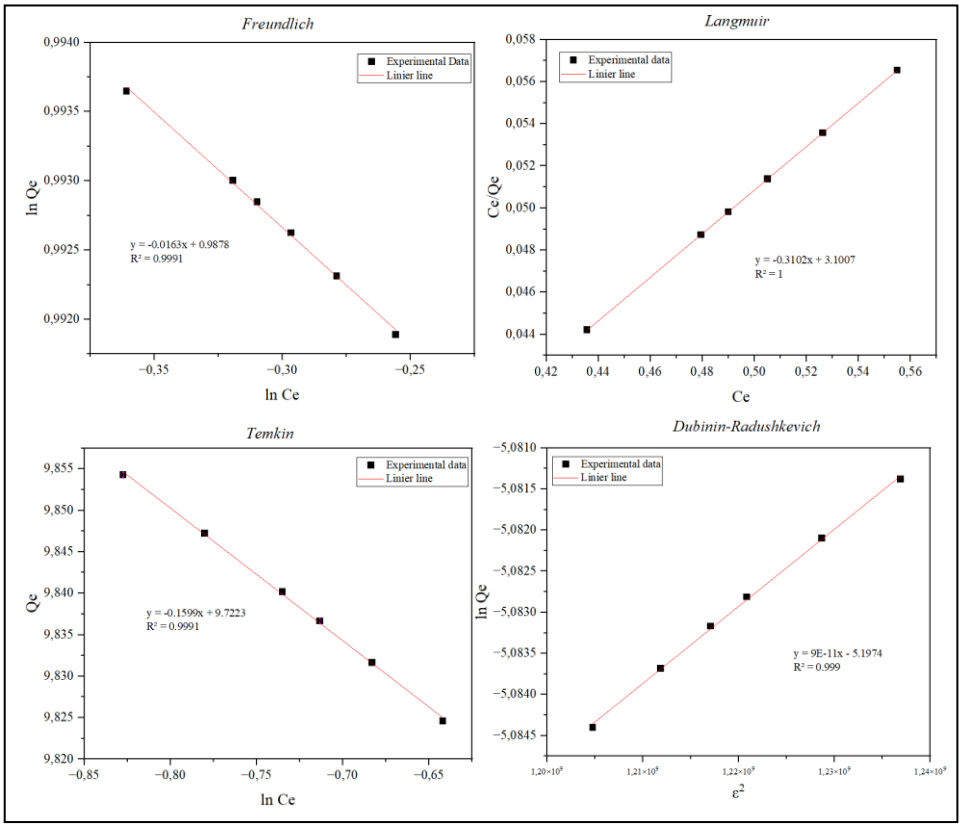

Fig. 2. Adsorption isotherm approach curves at varying temperatures

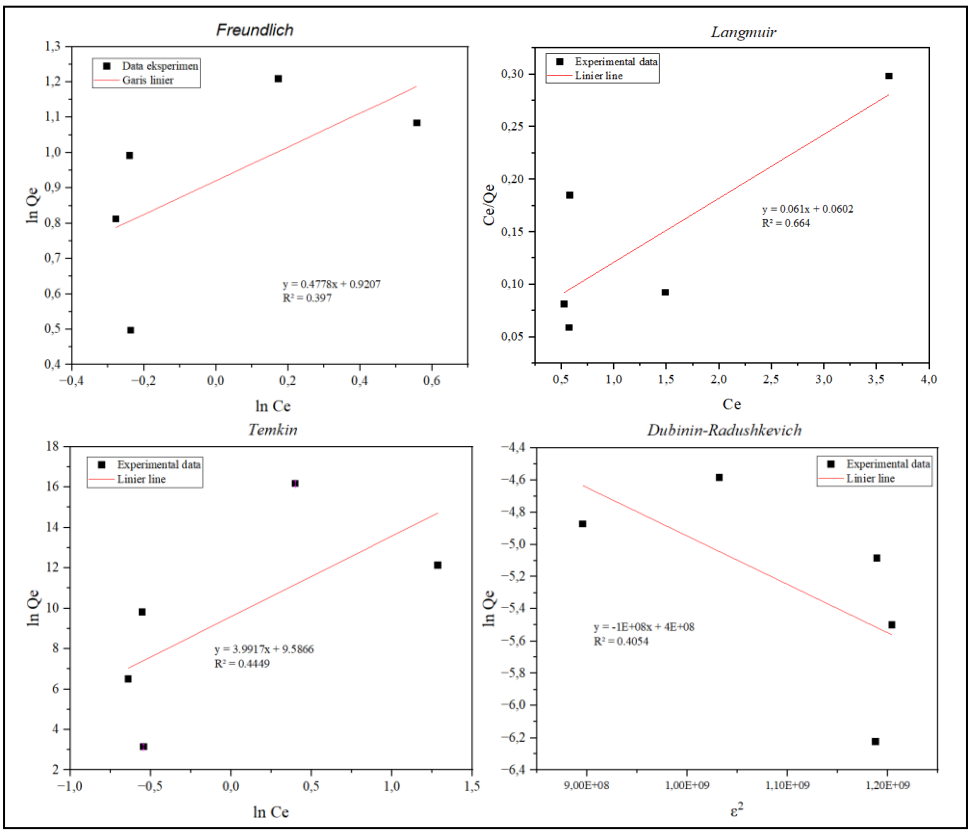

Fig. 3. Adsorption isotherm approach curves at varying temperatures

3.2. Adsorption Kinetics

1) Pseudo-First-Order

To obtain a clearer understanding of the mechanism of remazol yellow dye absorption, pseudo-first-order and pseudo-second-order models were used [31], Showed that the pseudo-first-order model is an initial model that describes the absorption rate in terms of absorption capacity. This model is usually represented by Equation 5.

$$\ln(q_e - q_t) = \ln q_e - k_1 \, . \, t \tag{5}$$

where k_1 (min^{-1}) is the pseudo-first-order rate constant, and qe and qt denote the amount of remazol yellow adsorbed (mg/g) at equilibrium and at time t (min), respectively. The results of data processing are shown in Fig. 4.

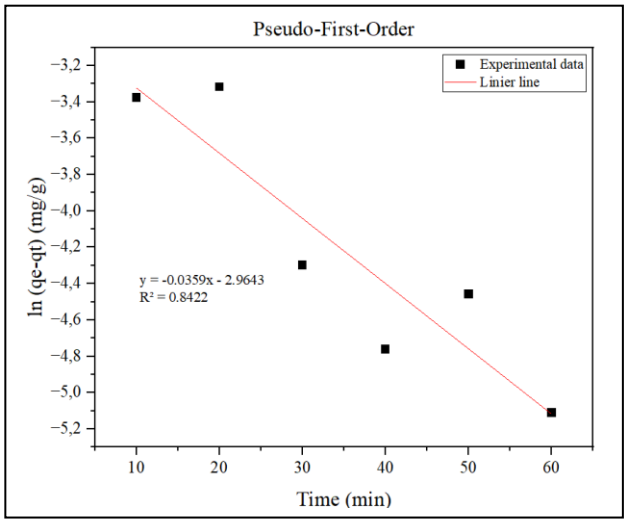

Fig. 4. Example of a figure caption. *(figure caption)*

2) Pseudo-Second-Order

The pseudo-second-order kinetic model based on [31] It is written in linear format through Equation 6.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{6}$$

where k_2 (g/[mg.min]) denotes the pseudo-second-order rate constant, and qe and qt maintain the same definitions as previously mentioned. The results of data processing are shown in Fig. 5.

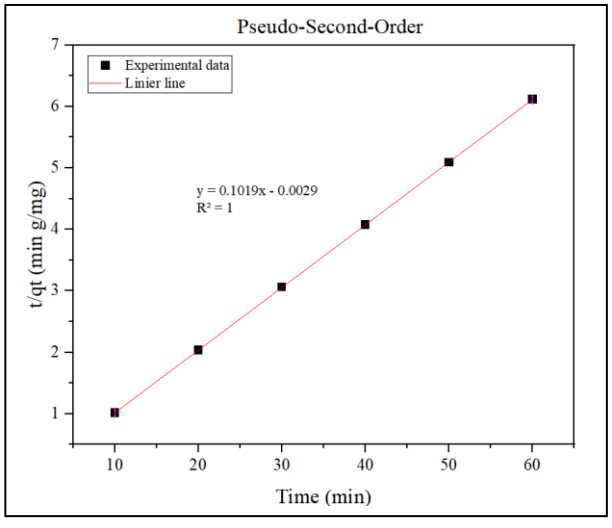

Fig. 5. Second-order-kinetic model of remazol yellow adsorption on quaternary ammonium polymer surface

Table 1. Parameters of pseudo-first-order and pseudo-second-order kinetic models

Kinetic models	Parameter	Value
Pseudo-first-order	qe (mg/g)	0.96
	K_1 (min^{-1})	2.96
	R^2	0.84
Pseudo-second-order	qe (mg/g)	9.81
	K_2 (g/mg min)	3.58
	R^2	1.00

The results of the kinetic study are shown in Table 1. The illustration images can be seen in Fig. 4. and Fig. 5. Based on the comparison of the correlation coefficient of the kinetic model (R2), it can be said that the kinetics of remazol yellow adsorption through quaternary ammonium polymer adsorbents have the best agreement with the second-order pseudo model. In addition, the K_1 value in the first-order pseudo model is smaller than the second-order pseudo model K_2. Therefore, the adsorption of remazol yellow through quaternary ammonium polymer adsorbents follows the second-order kinetic model. These kinetic results show that the adsorption process of quaternary ammonium polymers on remazol yellow is chemical adsorption, with the dye forming a coordination covalent bond with the adsorbent [32].

3.3. Illustration of the absorption of remazol yellow dye on quaternary ammonium polymer

The illustration of the absorption of Remazol yellow dye on quaternary ammonium polymer based on the results of the Langmuir isotherm approach and kinetic model. The illustration is shown in Fig. 6.

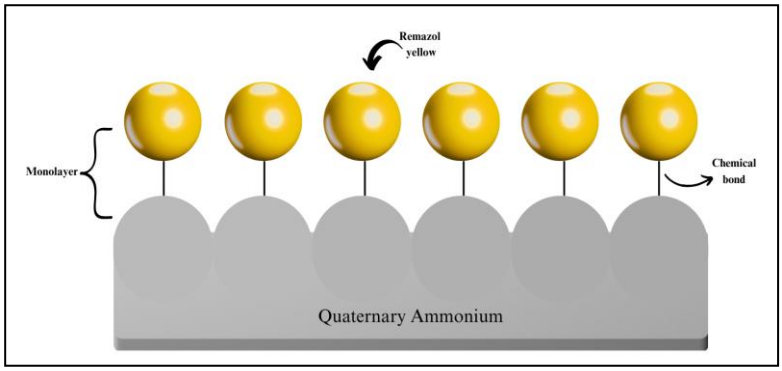

Fig. 6. Illustration of quaternary ammonium polymer adsorption on remazol yellow

An illustration of quaternary ammonium polymer absorption with Remazol Yellow is shown in Fig. 4. A chemical bond forms between the adsorbent and the adsorbate. This makes the ammonium polymer difficult to degrade, rendering it irreversible. Remazol yellow forms a layer on the surface of the quaternary ammonium polymer [33], [34].

3.4. Scanning Electron Microscope (SEM) Characterization

Morphological characterization was performed using a Scanning Electron Microscope (SEM) instrument. Morphological analysis was performed with a magnification of 10004 times at the optimum time and 10000 times at the optimum temperature. The morphology of the quaternary ammonium polymer after undergoing yellow Remazol adsorption is shown in Fig. 7.

In Fig. 7, the morphology of the quaternary ammonium polymer is shown after adsorbing yellow Remazol at both the optimum time and temperature, in the form of solid granules, bright in color, with various macropore and micropore sizes. The distribution of macropore and union sizes is presented in Table 2.

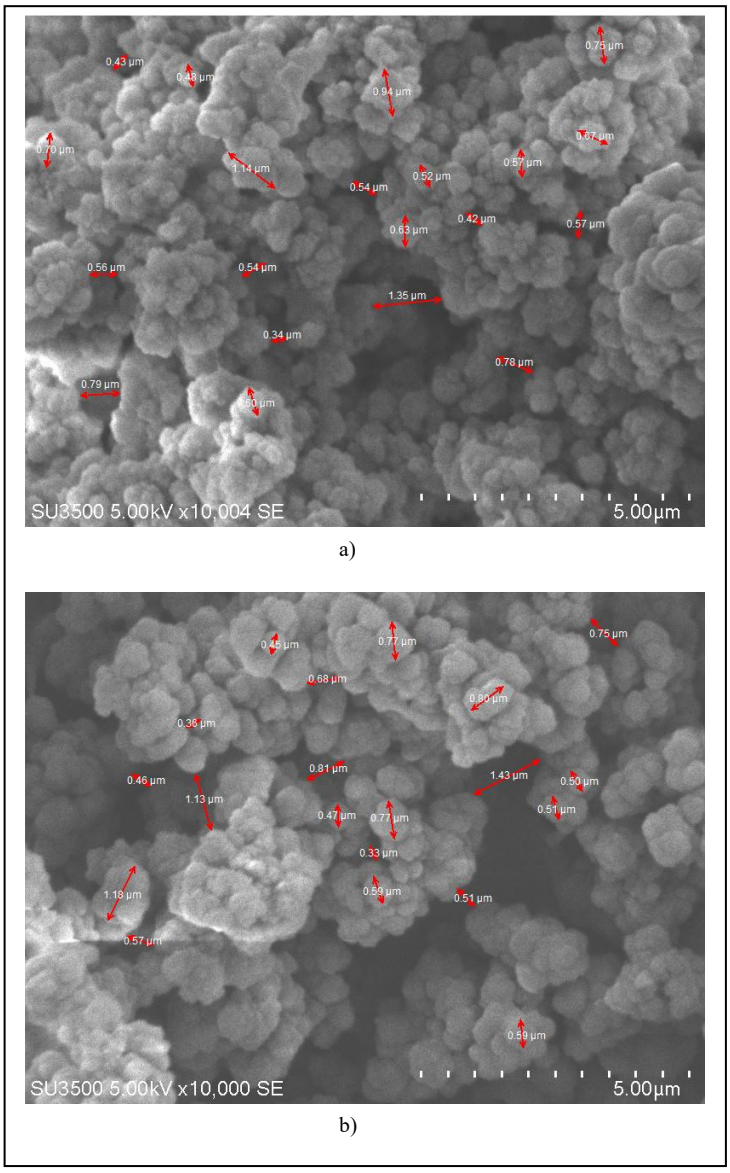

Fig. 7. Polymers ammonium kuarterner SEM illustration a) optimum time b) optimum temperature

Table 2. Macropore and union size of quaternary ammonium polymer after adsorbing Remazol Yellow

Sample	Length (µm)		
	Min	***Max***	***Average***
Macropore			
Time optimum	0.34	1.35	0.63
Temperature optimum	0.33	1.43	0.70
Union			
Time optimum	0.48	1.14	0.70
Temperature optimum	0.45	1.18	0.66

Based on Table 2, the macropore size at the optimum time and temperature is 0.34-1.35 µm and 0.33-1.43 µm, respectively. The union size at the optimum time and temperature is 0.48-1.14 µm and 0.45-1.18 µm, respectively. The macropore size at optimum temperature is larger than at optimum time, while the union size at optimum time is larger than at optimum temperature. Based on previous research [20], the morphology of quaternary ammonium polymers has a macropore size of 0.83-2.41 µm, and a union of 1.00-2.08 µm. After undergoing adsorption, the morphological size of the quaternary ammonium polymer, both macropore and union, changes. The size of the macropore and union after adsorption shrinks at both the optimum time and temperature. This

indicates that the yellow Remazol substance adheres to the quaternary ammonium polymer by filling the space within the polymer, and the size of the yellow Remazol granules is smaller than that of the quaternary ammonium polymer granules.

4. Conclusion

The most appropriate isothermal approach in this study is the Langmuir approach, as it yields the highest R^2 value, namely 1. This method outlines the adsorption mechanism that occurs at designated binding sites on the surface of the adsorbent. Every adsorption location on the adsorbent's surface is uniform, with the surface being filled by a single layer of adsorbed molecules, and there is no interaction between the adsorbed molecules on the adsorbent's surface. The macropore size at the optimum time and temperature is 0.34-1.35 µm and 0.33-1.43 µm, respectively. The union size at the optimum time and temperature is 0.48-1.14 µm and 0.45-1.18 µm, respectively.

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