Adsorption Capacity and Isotherm of Methylene Blue Removal in Aqueous Solution onto Regenerated Activated Carbon

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Abstract. Adsorption is one of the potential ways to removal of methylene blue in aqueous solution. The adsorbent used in the adsorption process is the activated carbon of betel nuts that produced from regeneration. The purpose of this study was to determine the effect of regeneration method to capacity and isotherm adsorption. The research occurs in a batch with three various regeneration conditions that is chemical, physical, and chemical-physical. In chemical, activated carbon is regenerated with HCl with various concentrations of 5 N, 6 N, and 7N. In physical, activated carbon is regenerated by heating it at temperatures of 160°C, 175°C, and 190°C. In chemical-physical regeneration, the condition is combination of the two. The concentration of methylene blue was analyzed using ultraviolet-visible spectrophotometer (UV-visible). The result showed the best regeneration conditions occur in chemical-physical regeneration with HCl concentration of 7 N, temperature of 190°C, and contact time of 25 minutes with an adsorption capacity on the surface is 3.57 mg/g and efficiency of 98.98%. Adsorption isotherm of removal methylene blue onto regenerated of betel nuts activated carbon follows the Langmuir model with regression value 1 ($R^2 = 1$).

1. Introduction
Dyestuffs and pigments are very widely used in several industries such as textile, paper, plastic, leather, food, and cosmetics. Industrial wastewater that contains dyes or pigments will pollute the aquatic environment [1]. One of the dyes found in aquatic is methylene blue. Methylene blue causes irritation to the digestive tract by ingested, cyanosis by inhalation, and irritation to the skin [2]. Methylene blue is an aromatic hydrocarbon compound that is toxic and includes cationic dyes with very strong adsorption power. In general, methylene blue is used as silk dyes, wool, textiles, papers, equipments, and cosmetics. Methylene blue is one type of base color that is widely used in the textile industry [3]. This compound is a dark green crystal. Dissolved methylene blue in water or alcohol will produce a blue solution.

The textile industry is one of the fastest growing industries in Indonesia. According to the regulation of the Minister of Environment No.51/MENLH/10/1995 concerning the standard quality for liquid waste, the maximum concentration of permissible methylene blue is 5-10 mg/L [4]. The method that can be used to reduce methylene blue as an aquatic environmental pollutant is adsorption. Adsorption is the best alternative that is done to overcome dye pollution [1]. One of the adsorbents that can be used in the adsorption process is activated carbon. Activated carbon is an amorphous compound that is produced from materials containing carbon or charcoal that are

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specifically needed to obtain high adsorption power. Activated carbon adsorb gas and certain chemical compounds or selective adsorption properties, depending on the size or volume of pores and surface area [5]. Raw material of the activated carbon that developed is betel nuts [6-7]. Betel nuts contain 32.56% water, 3.35% protein, 2.17% fat, 1.06% ash, and 60.86% carbohydrate [7]. Betel nuts also have a cellulose content of 70.2% which shows more carbon content in betel nuts [8]. The removal direct dye onto betel nuts activated carbon was 76.4% adsorption [7]. The activated carbon to be used is the activated carbon of betel nuts produced from regeneration. Regeneration is usually cheaper than replacing adsorbents [9].

Sanada et al [10] showed that removal of methylene blue with KATK (Coconut Shell Activated Carbon) and KABB (Coal Activated Carbon) were respectively 48.54% and 66.79%. Kow et al [11] used chemical regeneration to the activated carbon. The results showed that optimal regeneration conditions were produced at a volume of 15 ml of 6 N NaOH solution per gram of carbon with a contact time of 30 minutes, with a regeneration efficiency of 98% in the adsorption of methylene blue [11]. Bello et al [12] conducted research on the adsorption of methylene blue using treated sawdust by physical regeneration method. The results showed that the regeneration efficiency was 90% for 6 hours of contact time.

Hassan et al [13] conducted a study on adsorption capacity and isotherm in batches using commercial activated carbon from regeneration in the removal of synthetic dyes. The results showed that the adsorption capacity using physical methods was 61.73 mg/g and the chemical method was 53.72 mg/g. Physical methods have a higher regeneration efficiency value than chemical methods, 96.30% and 83.80% respectively.

The use of Freundlich and Langmuir isotherms is carried out with the aim of obtaining an equilibrium equation that can be used to determine the mass of the adsorbate absorbed by the adsorbent (adsorption capacity). Liquid-solid phase adsorption usually adheres to the Freundlich and Langmuir isotherm types [14]. The Freundlich isotherm is used with the assumption that multilayer bonds between the adsorbent and the adsorbate occur due to Van der Walls style, so the bond is not too strong. The Langmuir isotherm assumes that the formed layer is a monolayer which has a strong adsorbate because of chemical bonds [15]. Based on the previous studies, the development study of the effect of regeneration method of betel nuts activated carbon to the capacity and isotherm adsorption in removal of methylene blue was obtained.

2. Research methodology

In this study, the activated carbon made from betel nuts is a waste produced from the adsorption process of Jumputan wastewater. The activated carbon was saturated. This adsorbent obtained in three regeneration method, namely chemical, physical, and chemical-physical regeneration. Chemically method used 5 N, 6 N, and 7 N HCl for 30 minutes immersion. Physically method occurs at a temperature of 160°C, 175°C, and 190°C for 1 hours heating. Chemically-physically regenerated carried out by using 5 N, 6 N, and 7 N HCl for 30 minutes immersion, then heating at 160°C, 175°C, and 190°C for 1 hour. The regenerated activated carbon is dried in an oven 105°C to remove the water content. The quality of regenerated activated carbon analyzed by the water content.

The process of making a standard curve is done by making a standard solution of methylene blue with concentrations of 20, 40, 60, 80, and 100 mg/L. Each standard of methylene blue solution is measured for absorbance using UV-Visible Spectrophotometer at a wavelength of 560.5 nm. The experiment carried out by mixing 6 grams of regenerated activated carbon into 200 ml of methylene blue solution with a stirring speed of 150 rpm. Samples were filtered and analyzed using a UV-visible spectrophotometer.

The adsorption efficiency is determined using the equation in (1). Adsorption efficiency is a comparison between the initial concentration (Co) (mg/L) and the final concentration (Ce) with the initial concentration.

$$\text{Adsorption efficiency} = \frac{C_0 - C_e}{C_0} \times 100\% \quad (1)$$
Langmuir adsorption isotherm can be based on several assumptions, adsorption only occurs in a single layer (monolayer), adsorption heat does not depend on surface closure, and all sites and surfaces. The Langmuir equation assumes that the surface of the adsorbent is homogeneous, and a site can only be regenerated once the activated carbon

\[ \log q_e = \log k_F + \frac{1}{n} \log C_e \]  

From this concept, the equation can be derived as follows (equation 2):

\[ \frac{C_e}{q_e} = \frac{1}{q_mK} + \frac{C_e}{q_m} \]

The approach to the adsorption isotherm can be explained by the Freundlich equation. If the concentration of the solution in equilibrium is plotted as ordinate and the concentration of adsorbate in the adsorbent as abscissa in logarithmic coordinates, gradients n and intercept will be obtained [16]. The Freundlich isotherm model shows that the adsorbate layer formed on the surface of the adsorbent is multilayer [16]. \( q_e \) is the number of adsorbates adsorbed (mg/g), \( K_F \) is the Freundlich constant (L/mg), and \( n \) is the adsorption constant. These model constants are calculated by the slope and intersection of the plot obtained from \( \log q_e \) versus \( \log C_e \). Where the Freundlich equation is written as follows (equation 3):

\[ \log q_e = \log k_F + \frac{1}{n} \log C_e \]

Based on the calculation of concentration dyestuff, it can be found that the adsorption capacity \( q_e \) using the equation (4), where \( C_0 \) and \( C_e \) are initial and final concentration (mg/L), \( V \) is volume of solution treated (L), and \( m \) is the dosage of the adsorbent used (grams).

\[ q_e = \frac{(C_0-C_e) \times V}{m} \]

3. Result and discussion

The quality of the regenerated activated carbon tested based on the water content. The fresh activated carbon contains 0.05% water and waste activated carbon (before regeneration) of 0.059%. The best water content results are obtained on chemical-physical regeneration with 7 N and a temperature of 190°C, which is equal to 0.02%. The regenerated one has lower water content because the HCl concentration used greater that HCl used in the activation process, and the addition of heating process. The low water content is because the surface of the activated carbon contains fewer polar functional groups [17]. Water content analysis results obtained from this study meet the activated carbon quality standards in accordance with SNI 06-3730-1995 [18]. The result showed that regenerated activated carbon has a good performance as an adsorbent and can be used to adsorption process.

3.1. Effect of chemical regeneration method on methylene blue adsorbed

The adsorption capacity in various HCl concentration was in range 2.37 to 3.28 mg/g. At HCl concentration 5 N, \( q_e \) increases through the first 15 minutes rapidly, then continue to decline slowly. The 6 N HCl concentration has the same trend, \( q_e \) increases rapidly to the first 20 minutes adsorption. Regeneration using HCL 7 N shows slowly increases by the time, with initial adsorption capacity lower than 5 N and 6 N. Up to the 35th minute, the adsorption capacity has increased over the 5 N and 6 N.

Figure 1 shows that the adsorption of methylene blue on regenerated activated carbon has increased with increasing HCl concentration. At a concentration of 7 N and a contact time of 35 minutes, the ability adsorbent to adsorb the methylene blue was 3.28 mg/g. The higher concentration will produce greater the surface area available, more pores that can be passed, so contact of dyes with activated carbon wider [19]. Figure 1 is also shown that at a concentration of 7 N HCl the amount of mass of methylene blue adsorbed increases with increasing contact time. Kow [11] reported that at a concentration HCl of 8 N the adsorbed mass decreases. This shows that if the concentration of regenerator used more than 7 N will damage the activated carbon structure that effect its ability to adsorb the dye. In this study, longer contact time is needed to see a decrease in the mass of adsorbed, because longer contact makes the
methylene blue pass through the pores longer. Regeneration efficiency of 98% in the adsorption of methylene blue onto activated carbon achieved at 30 minutes contact time [11]. The highest adsorption efficiency of methylene blue achieved by regenerated of betel nuts activated carbon was 89.87%. This efficiency is greater than activated carbon based on coconut and coal [10].

3.2. Effect of physical regeneration methods on methylene blue adsorbed
Physical regeneration method occurs by heating the waste activated carbon at 160°C, 175°C, and 190°C. The adsorption capacity is 3.13 – 3.51 mg/g. Overall data shows that qₑ go up rapidly to first 10 minutes adsorption then fluctuate up and down slowly. The adsorption process occurs in two roles, the initial adsorption occurs fast and then the releases of substances slowly because the amount of the adsorbed substance exceeds the maximum adsorption capacity. Langmuir explained that on the surface of the adsorbent there are active sites whose numbers are proportional to the surface area of the adsorbent [20]. If the active site has been saturated, the addition of the contact time will not affect the increasing of adsorption ability, the qₑ tends to decrease.

Figure 1. Effect of Chemical Regeneration Method on Methylene Blue Adsorbed

Figure 2 describes the largest mass of methylene blue was obtained at a temperature of 190°C for 25 minutes of contact time, which is 3.51 mg/g. At a temperature of 160°C and 175°C, the water and organic matter content evaporated are less than at 190°C. The higher the temperature and the length of contact
time, the more water and organic matter evaporates and makes the pore surface area even bigger. Increasing the pore surface area will affect the adsorption capacity of an adsorbent [21]. The amount of $q_e$ in the three various temperature does not show a significant difference.

The highest adsorption efficiency of methylene blue with the physical regeneration method is 96.16%, that achieved at 190°C and 25 minutes contact time. This amount is greater than efficiency methylene blue removal onto treated sawdust [12]. The efficiency of adsorption of physical regeneration is higher than chemical, same report as Hassan et al [13].

3.3. Effect of chemical-physical regeneration method on methylene blue adsorbed

Chemical-physical regeneration method obtains 2.01-3.62 mg/g of adsorption capacity. The combination of chemical-physical regeneration will make the pore surface of activated carbon greater than just chemical or physical regeneration. Larger pores will certainly facilitate the process of adsorption of methylene blue substances to be removed.

Most of the $q_e$ on the chemical-physical regeneration with a concentration HCl of 5 N for various temperature is below 3 mg/g. Variation of 5 N HCl and 190°C show the smallest $q_e$ all the time. This shows that active site, water and volatile matter on pores of waste adsorbent has not opened completely. At 6 N and 7 N HCl concentration at various temperature, the amount of $q_e$ is almost the same, most of in range 3.60-3.62 mg/g. This shows that with increasing HCl concentration, there is an increase in the ability to clean the pore surface of waste activated carbon by dissolving into cations and heating. The ion H⁺ in this activator has a higher electronegativity, with the addition of high temperature and contact time, so the ability of the regenerated adsorbent to adsorb of methylene blue will increase.

\[ q_e \text{ (mg/g)} \]
\[ \text{Time (minutes)} \]

\[ 5 N \text{ 160 C} \quad 5 N \text{ 175 C} \quad 5 N \text{ 190 C} \quad 6 N \text{ 160 C} \quad 6 N \text{ 175 C} \]
\[ 6 N \text{ 190 C} \quad 7 N \text{ 160 C} \quad 7 N \text{ 175 C} \quad 7 N \text{ 190 C} \]

**Figure 3.** Effect of Chemical-Physical Regeneration Method on Methylene Blue Adsorbed

Compare with the previous studies, the $q_e$ of methylene blue onto fresh coconut and coal activated carbon was 1.18 mg/g and 1.81 mg/g respectively [10]. This shows that the use of waste activated carbon made from betel nuts that regenerated is superior compared to coconut shell and coal activated carbon with maximum mass absorbed of 3.62 mg/g.

On the chemical-physical regeneration method, the highest adsorption efficiency of methylene blue is achieved under conditions of 7 N HCl concentration, temperature 190°C and contact time 25 minutes, which is equal to 98.98%. Larger pore surface area is due to chemical reactivation and followed by physical so that the activator solution can easily enter the pores and can release the adsorbate or impurities that trapped inside. The presence of high concentrations and temperatures will increase the pore size and push the formation of new pores. The removal of methylene blue onto regenerated
activated carbon made from betel nuts is more effective and efficient than coconut, coal, commercial activated carbon and sawdust [10-13].

3.4. Adsorption isotherm of activated carbon with chemical regeneration method

Based on Figures 4 and 5, the isotherm adsorption which has a value of R close to 1 is the model that best matches the adsorption process on regenerated carbon. The comparison of R² values from the Langmuir and Freundlich isotherm adsorption shows that the adsorption process to remove methylene blue in aqueous solution was fitted to the Langmuir isotherm.

Figure 4. Freundlich Isotherm of Methylene Blue in Chemical Regeneration Method

Figure 5. Langmuir Isotherm of Methylene Blue in Chemical Regeneration Method

Table 1 describes the function values of the Freundlich and Langmuir isotherm on the adsorption of methylene blue onto chemically regenerated carbon. Adsorption capacity in Freundlich was adapted from Kₓ, the greater is the best [23]. The slope value of 1/n with a range of 0-1 is the adsorption intensity or the heterogeneity of the adsorbent surface, increasingly heterogeneous if 1/n approaches to 0 [12, 22]. The cooperative and weak adsorption bond indicates with n value less than 1 [23-24]. As shown in Table 1, the n value of the process is 3.82-5.26, it means favorable sorption process occurred, where n refers to 1<n<10 [24].
Table 1. Adsorption Isotherms of Methylene Blue in Chemical Regeneration Method

| Isotherm Model | Chemical Regeneration | R²    | Constant |
|----------------|-----------------------|-------|----------|
| Freundlich     | 5 N HCl               | 0.981 | K_F     | 5.24 L/mg | n        | 5.24    |
|                | 6 N HCl               | 0.980 | K_F     | 5.25 L/mg | n        | 5.26    |
|                | 7 N HCl               | 0.966 | K_F     | 6.32 L/mg | n        | 3.82    |
| Langmuir       | 5 N HCl               | 0.998 | q_m     | 2.52 mg/g | K        | 2.87 L/mg |
|                | 6 N HCl               | 0.998 | q_m     | 2.52 mg/g | K        | 2.89 L/mg |
|                | 7 N HCl               | 0.994 | q_m     | 2.13 mg/g | K        | 4.73 L/mg |

Theoretical adsorption capacity (q_m) of the regenerated adsorbent that produces from chemical method is 2.13-2.52 mg/g. Value q_m at a concentration of 5 N and 6 N HCl is greater than 7 N HCl. It indicates that the higher concentration of activator affected to less methylene blue that adsorbed (q_m) [25]. The data obtained is in accordance with the theory that the specific surface area becomes smaller, because with the increasing pH (low activator concentration) more active sides will be available while at low pH (high activator concentration) will cause a decrease in the active site.

3.5. Adsorption isotherm of activated carbon with physical regeneration method

Regression value of the Freundlich and Langmuir adsorption isotherm onto physically regenerated carbon shows that the Langmuir has a value of R² closes to 1. It can be concluded that the isotherm of adsorption is fitted to Langmuir. Freundlich constant in physical smaller than chemical, in range 3.97-4.04 L/mg, but the n value is greater with range 11.27-12.28 (over the favorable range). The q_m is in range 2.99-3.05 mg/g, greater than physical method, that shows more methylene blue can be adsorbed by the carbon.
3.6. Adsorption isotherm of activated carbon with chemical-physical regeneration method

Table 3 describes the function values of the Freundlich and Langmuir isotherm on the adsorption of methylene blue onto the chemical-physical regenerated adsorbent. From all the data, the regression value of the two approaches to 1, but Langmuir isotherm shows the best results. Based on the \( q_m \) and \( K_F \) values in Table 3, it can be seen that the greatest adsorption capacity occurs in waste activated carbon which regenerates chemical-physically with 5 N HCl concentrations and 190\(^\circ\)C, which is equal to 9.77 L/mg and 10.11 L/mg respectively. The Langmuir isotherm model has the highest adsorbent capacity is 3.57 mg/g.

From Figure 1-3 and Table 1-3, adsorption capacity experimental and Langmuir is adjacent, on average 3.09-3.25 mg/g. Determination of equilibrium models depends on the determinant coefficient (\( R^2 \)) at the highest value [26]. Based on the results of the calculation of \( R^2 \) from the three regeneration methods is closer to 1 by the Langmuir isotherm. The results obtained were also in accordance with the other results [27-30].
Table 3. Data of Adsorption Isotherm Methylene Blue in Chemical-Physical Regeneration Method

| Isotherm Model | Chemical-Physical Regeneration | R² | Constant |
|----------------|--------------------------------|----|----------|
| Freundlich     | 5 N 160°C                       | 0.9764 | 4.86 L/mg | n | 6.09 |
|                | 5 N 175°C                       | 0.9812 | 6.28 L/mg | n | 3.95 |
|                | 5 N 190°C                       | 0.9506 | 10.11 L/mg | n | 2.48 |
| Langmuir       | 6 N 160°C                       | 0.9994 | 3.62 L/mg | n | 82.64 |
|                | 6 N 175°C                       | 0.9999 | 3.62 L/mg | n | 83.33 |
|                | 6 N 190°C                       | 0.9994 | 3.62 L/mg | n | 89.28 |
|                | 7 N 160°C                       | 0.9974 | 3.62 L/mg | n | 50.50 |
|                | 7 N 175°C                       | 0.9986 | 3.62 L/mg | n | 70.92 |
|                | 7 N 190°C                       | 0.9992 | 3.62 L/mg | n | 88.49 |
|                | 5 N 160°C                       | 1     | qₘ₀ 3.56 mg/g | K | 0.01 L/mg |
|                | 5 N 175°C                       | 1     | qₘ₀ 3.56 mg/g | K | 0.01 L/mg |
|                | 5 N 190°C                       | 0.9812 | qₘ₀ 1.68 mg/g | K | 9.77 L/mg |
|                | 6 N 160°C                       | 1     | qₘ₀ 3.56 mg/g | K | 0.01 L/mg |
|                | 6 N 175°C                       | 1     | qₘ₀ 3.56 mg/g | K | 0.01 L/mg |
|                | 6 N 190°C                       | 1     | qₘ₀ 3.57 mg/g | K | 0.01 L/mg |
|                | 7 N 160°C                       | 1     | qₘ₀ 3.50 mg/g | K | 0.03 L/mg |
|                | 7 N 175°C                       | 1     | qₘ₀ 3.55 mg/g | K | 0.02 L/mg |
|                | 7 N 190°C                       | 1     | qₘ₀ 3.57 mg/g | K | 0.01 L/mg |

4. Conclusion
The best regeneration conditions of waste activated carbon made from betel nuts in the process of adsorption of methylene blue are by chemical-physical regeneration methods at HCl concentration of 7 N and temperature of 190°C. Adsorption capacity on the surface and the highest adsorption efficiency in the chemical-physical regeneration are 3.57 mg/g and 98.98%. Adsorption isotherm of removal methylene blue onto regenerated of betel nuts activated carbon follows the Langmuir model with regression value 1 (R² = 1).

Acknowledgments
This research was supported by the Separation and Purification Laboratory of Engineering Faculty of Universitas Sriwijaya. The authors thank Universitas Sriwijaya for funding the research under SATEKS Scheme.
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