Sorption Isotherm Models on Methylene Blue-Baking Filter Dust Activated Carbon

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Abstract. The aim of this study was to investigate the removal characteristics of methylene blue on baking filter dust carbon using three models of isotherm, Langmuir, Freundlich and Temkin. Baking filter dust from aluminum industrial waste was prepared as an adsorbent to adsorb methylene blue under room temperature. The BFD was prepared using KO+KI as impregnation agent. Isotherm characteristics were analyzed using Langmuir, Freundlich, and Temkin models. BFD-MB was contacted and stirred in a batch process, sampling was done in a specific interval of time in order to acquire the concentration adsorbed onto the BFD. Spectrophotometer UV-VIS was used to analyze the absorbance of the MB samples and the concentrations were determined using the calibration method. According to the result, Freundlich model best fit adsorption system data sets studied in this work with the $R^2$ 0.949.

1. Introduction
Activated carbon adsorption was the cheapest and the widest used method in recovering the contaminant in water, waste water, and any other application in industrial processes. Many researches have been conducted in the past and continue in current works in order to find out cheaper and compatible adsorbent to specific adsorbate. Many

Various types of materials that can be used as adsorbents include activated carbon [1]; [2]; [3], bentonite [4] and many other adsorbents. Research on continuous adsorbents is carried out to continuously increase the capacity and quality of the adsorbent. In this study, waste from the INALUM industry in the form of Baking Filter Dust (BFD) was used to adsorb methylene blue (MB) in batches.

In other cases, some industrial wastes were studied to get the beneficial uses of them. Due to regulatory, industries are not allowed to sell nor spent their wastes directly to the environment without any appropriate treatments, they should, in some extent, do researches to find out the best way to resolve to their waste. As an example, INALUM industry in Indonesia produces about 115 ton per day of carbon dust from the aluminum melting process which commonly called baking filter dust. The carbon has comparatively high carbon, up to 85 %, which is potential to convert into activated carbon. Nevertheless, limiting data was available in using this waste as some useful product. Therefore, continuous research should be conducted to cope the accumulate waste in the site.

[2] has performed the textile sludge impregnated KOH, KI, and hybrid in certain composition to be activated carbons. It indicated that the use of hybrid KOH+KI gives better surface area than the others.
In the malachite green adsorption, it yielded the adsorption capacity up to 146 mg/g. Hence, this study focused on the hybrid KOH-KI as impregnation agent to activate the BFD carbon.

Some of the possible concern to be done is using the BFD in removing aqueous MB solution. In this work, MB in wide range initial concentration was contacted with a certain weight of BFD in order to find out the removal characteristics. The isotherm characteristic was fitted to Langmuir, Freundlich and Temkin.

2. Material and Methods

2.1. Materials

Baking filter dust (BFD) was obtained from PT. INALUM (Indonesia Asahan Aluminum), Medan. Chemicals used in this research were KOH pellets KI powder from Merck, Germany, and methylene blue powder from Sigma Aldrich, Germany. The chemicals were all analytical grades.

2.2. Impregnation of BFD

BFD carbon was mixed with (KOH+KI) in 0.5 ratio (w/w) of precursor and the activating agent. The composite was heated for 1.5 h 90°C and then dried at 110°C overnight for impregnation. Furthermore, it was heated using furnace by 500°C for 1 h. A certain volume of purified water was added to the sample until neutral pH was obtained to remove nonorganic materials. The BFD activated carbon was 24 h oven dried at 110°C.

2.3. Adsorption of MB onto BFD carbon

Methylene blue solution was prepared from it powdered form to 1000 ppm stock solution. The various concentrations MB 1-8 ppm then made from this by dilution in distilled water. The volume of 250 ml of a certain concentration was poured in a flask, a 0.5 g BFD was added into the flask and the stopwatch was on. Sampling was performed every 5 minutes intervals until the constant concentration of remain solution obtained.

The remaining solution concentrations were determined using Spectrophotometer UV-VIS at 661 nm wavelength. Calibration concentrations were 2, 4, 6, 8, and 10 ppm. Calibration curves were performed to get the concentrations.

3. Results and Discussion

3.1. Isotherm Models

There are three models studied in this experiment, Freundlich, Langmuir and Temkin. The models were applied to fit the equilibrium data on the MB-BFD sorption. The data were tabulated in Table 1. Isotherm models were shown in Fig. 1-Fig 3. The Freundlich model shows in eq. 1.

\[ Qe = Kf \cdot Ce^{1/n} \]  

(1)

Where \( Qe \) is the equilibrium concentration of MB onto BFD activated carbon. \( Kf \) and \( n \) are the Freundlich constant. \( Kf \) value indicates the sorbat capacity where the \( 1/n \) value is an empirical parameter related to adsorption intensity.
According to the linear form of Freundlich model, the linear equation is $y = 1.0914x - 0.4089$, which $R^2$ of 0.949 (See Fig.1). This equation is fit enough to describe the adsorption model of MB onto BFD impregnated KOH-KI, rather than the two other models. It describes the adsorption characteristics for lower adsorbate concentration and the heterogenous surface of the adsorbent.

In this work, the $n$ value obtained 0.916, where $1/n$ is more than 1. It indicated the cooperative adsorption occurred in the sorbate-sorbent. The adsorption concentration has a significant influence to make the solute (MB) go through the adsorbent surface area. Conversely, when the value of $1/n$ is less than 1, the adsorption follows Langmuir model [5]. Furthermore, [6] stated that the $1/n > 1$ means the physical sorption which van der Waals bond was obtained between the adsorbate and adsorbent. When the value is less than 1, the adsorption occurred through chemical bonds. Freundlich isotherm model signifies that the adsorption takes place on multilayer of the adsorbent. Langmuir isotherm model is given as:

$$Q_e = \frac{Q_o \cdot b \cdot C_e}{(1+b \cdot C_e)}$$

The Langmuir constant of $b$ and $Q_o$ were identified by using the linearization equation from Fig. 2. Unfortunately, the correlation of the experimental equilibrium data poorly correlated to the Langmuir isotherm model as indicate in extremely low of $R^2$ value of 0.008. The Langmuir isotherm expresses the sorption of sorbate-solute only in monolayer system. It represents the solute – sorbate interaction in this experiment do not in the monolayer surface, but according to the Freundlich model, where is in multilayer as indicated above.

Temkin isotherm model represents the interface interactions among adsorbates that influence the adsorption characteristics. There is no correlation of the concentration whether it is low or high enough towards the heat of adsorption. The Temkin model shows as:

$$Q_e = B \ln A \cdot C_e$$

Where $B$ corresponds to the thermal energy of the adsorption, which is $RT/b$. A value shows the Temkin constant related to the highest joint energy of the adsorbates. The plot of $Q_e$ vs $\ln C_e$ give the linear quadratic value, $R^2$ of 0.8979, and the linear equation of $y = 0.7766x + 0.4823$ (Fig. 3).
This work ensured the adsorption thermal energy, which state as B constant of 0.7766 J/mol (Table 1). The B value < 12 J/mol denotes the domination of physic sorption of MB onto the BFD activated carbon besides chemical sorption and ion exchange. On the other hand, when the value is more than 12 J/mol, the adsorption approach could be on the opposite. In this case, there is likelihood of both of the sorption method took place simultaneously. Additionally, the A value informs the maximum energy among the MB solutes which are 1,861 L/g in this experiment.

| Isotherm | Freundlich | Langmuir | Temkin |
|----------|------------|----------|--------|
|          | n | Kf | R² | Qo | b | R² | B | A | R² |
| Temkin   | 0.916 | 0.390 | 0.949 | -32,258 | -0.012 | 0.0088 | 0.7766 | 1,861 | 0.8979 |

4. Conclusion

The sorption efficiency is affected by the initial concentration of MB. The longer time of adsorption more MB adsorbed until the equilibrium is obtained. The MB sorption onto BFD fit to Freundlich isotherm with R² 0.949.

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