

Utilization Of Nata De Pina As Adsorbent For Adsorption Of Remazol Black B Textile Dyes

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Abstract: This research was conducted with the aim to determine the efficiency percent, the pattern of adsorption isotherm, and the maximum adsorption power of the adsorption of remazol black B textile dyes using cellulose nata de pina membrane. The cellulose nata de pina membrane in this study was made with the basic ingredients of pineapple skin waste. The result showed that the percentage of the efficiency value of remazol black B textile dyes using the cellulose nata de pina membrane obtained was 31.69119%. This value is obtained at the condition of the optimal remazol black B solution at pH 2, concentration 60 mg/L, and contact time for 60 minutes. Remazol black B isotherm adsorption pattern by cellulose nata de pina membrane fulfilled the Langmuir adsorption isotherm curve with $R^2 = 0.952$ and maximum membrane adsorption capacity of 12.5 mg per gram of adsorbent.

Keywords: adsorption, cellulose nata de pina membrane, remazol black B.

I. INTRODUCTION

The textile industry is one of the largest foreign exchange earners in Indonesian. The textile is large export commodity. This textile exports always increase every year. In 2017 textile exports reached US\$ 12.4 billion, up 4.4% from the previous year [Muslim, 2018].

In addition to contributing to positive impacts in the form of large foreign exchange for Indonesian, textile production also contributes to negative impacts in the form of sizable waste, which can pollute the environment. Environmental pollution arises in the form of water pollution due to liquid waste from washing and coloring textiles. In the coloring process hazardous textile dye waste is produced. One of the textile dyes that are widely used in industry is azo type remazol black B [Erkurt, 2010].

Remazol black B (RBB) is widely used because it is not easily degraded under ordinary aerobic conditions and is easily soluble in water. RBB is a dye having a molecular formula $C_{26}H_{21}N_5Na_4O_{19}S_6$ with a molecular weight of 991.8 g/mol. The RBB structure is presented in Figure 1.

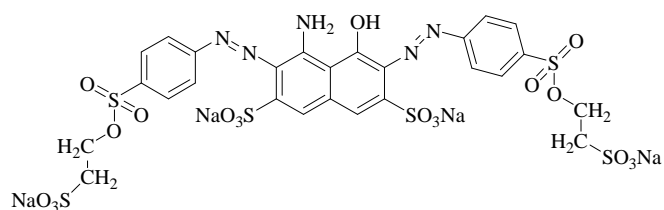


Figure 1: Structure of RBB

Reactive dyes containing the azo chromophore group are widely used as black dyes in textiles. RBB is synthesized so that it is not easily damaged by photolytic or chemical treatment. If this waste is not handled properly it can cause a foul odour, disturb the aesthetics and can poison the biota in the waters. The presence of this dye in water can also cause aesthetics problems and inhibit the penetration of sunlight into the water, thereby interfering with the photosynthetic activity of aquatic organisms and triggering anaerobic activity which produces unpleasant odour compounds [Sudiana, et.al., 2018]. Other impacts that can be caused by these dyes are health problems ranging from skin diseases to skin cancer [Sulistya, 2013]. Therefore, it is necessary to do further handling of this waste before it is discharged into the waters.

One method that can be used to treat RBB waste is the adsorption method. This adsorption can be done using membrane adsorbents. The membrane was chosen because the production process is very simple, economical, low energy consumption is needed, does not damage the material and is classified as clean technology [Lindu, et.al., 2009]. One membrane that can be used to adsorb RBB waste is the cellulose nata de pina membrane. Nata de pina is a pineapple liquid waste product that is fermented using fiber-rich *Acetobacter xylinum*. The cellulose content in nata de pina has strong adsorbing properties. Therefore, this nata is suitable for use as a main ingredient in the manufacture of cellulose membranes that can adsorb RBB dyes.

This research was conducted with the aim to determine the efficiency percent, the pattern of adsorption isotherm, and the maximum adsorption power of the adsorption of remazol black B textile dyes using cellulose nata de pina membrane. The success of this research can reduce the danger of RBB staining. In addition, this research can indirectly reduce environmental pollution by pineapple skin waste which is used as a basic ingredient in making nata de pina.

II. MATERIALS AND METHODS

MAKING OF NATA DE PINA

Clean pineapple skin is added to the water by a ratio (1:4) which is 200 grams of pineapple skin plus 800 mL of water. The two are then mixed and filtered to get the filtrate. The obtained filtrate was added with 7.5% (w/v) sucrose and 0.5% (w/v) ammonium sulfate. Next, the filtrate is boiled at 100°C for 15-20 minutes. The filtrate was collected in a tray the pH was adjusted to 4.5 with the addition of 2% (v/v) acetic acid. The tray is wrapped in sterile paper and tied with string and black isolation. After cooling, *Acetobacter xylinum* inoculum is added to the tray by 10%. The media was incubated for 7 days at room temperature until nata was formed.

PURIFICATION OF NATA DE PINA

Nata de pina is soaked in 1 liter of 1% (w/v) NaOH solution for 24 hours. Nata is then neutralized with 1 liter of 1% (v/v) acetic acid for 24 hours. After that, Nata is soaked with 1 liter water for 24 hours. Nata is also washed several times to a neutral pH.

RBB ADSORPTION

The RBB parent solution was made at a concentration of 100 mg/L. The maximum wavelength of the RBB parent solution to obtain a concentration of 50 mg/L at a wavelength of 400-800 nm. The 4x4 cm nata de pina membrane sheet was put into 6 Erlenmeyers, each filled with a RBB solution concentration of 60 mg/L pH 1, 2, 4, 7, 9, and 11. The solution was shaken for 120 minutes. At optimal pH, variations in concentrations of 10, 30, 60, 80, and 100 mg/L are performed. Under optimal pH and concentration conditions, variations in contact time are 30, 60, 120, 150, and 180 minutes.

DATA ANALYSIS

Optimal pH, concentration, and contact time data can be obtained from the RBB correlation curve adsorbed with pH, concentration, or contact time. The value of x/m can be calculated by the following equation.

$$\frac{x}{m} = \frac{(C_0 - C_{st}) \times V}{m} \dots\dots\dots(1)$$

With,

$\frac{x}{m}$ = the mass of the RBB adsorption by the nata de pina membrane (mg/g)

C_0 = the concentration of RBB before adsorption (mg/L)

C_{st} = the concentration of RBB after adsorption (mg/L)

m = the mass of nata de pina membrane adsorbent

Meanwhile, the efficiency of RBB adsorption by the nata de pina membrane can be calculated by the following equation.

$$\%E = \frac{C_0 - C_{st}}{C_0} \times 100\% \dots\dots\dots(2)$$

With,

x/m : The mass of RBB dyes adsorbed by each gram mass of the nata de pina membrane

$\%E$: Percent efficiency

C_0 = the concentration of RBB before adsorbed (mg/L)

C_{st} = the concentration of RBB after adsorbed (mg/L)

m = the mass of nata de pina membrane adsorbent

The RBB dyestuff adsorption isotherm pattern is obtained by substituting the adsorption data obtained into the linear equation of the linear equation of the Langmuir and Freundlich. The Langmuir equation is:

$$\frac{C}{x/m} = \frac{1}{x/m_{mak}k} + \frac{1}{x/m_{mak}} \times C \dots\dots\dots(3)$$

Meanwhile, Freundlich equation is:

$$\text{Log}(x/m) = \text{Log} k + \frac{1}{n} \text{Log} C \dots\dots\dots(4)$$

III. RESULT AND DISCUSSION

Based on the adsorption carried out at the pH variation, the adsorption results are presented in the form of a curve in Figure 2. The pH variation curve shows that the optimal adsorption occurs at pH 2. The adsorbed RBB at pH 2 is 11.36562 mg per gram of adsorbent. This is in line with research conducted by [Ayuni, et. al., 2016]. Ayuni, et. al.2016, also succeeded in adsorbing RBB using a polyvinyl alcohol chitosan-pectin binding membrane which binds cross with the optimum pH at pH 2. Thus, this show that indeed it is true that adsorption of RBB dyes can occur optimally at pH 2.

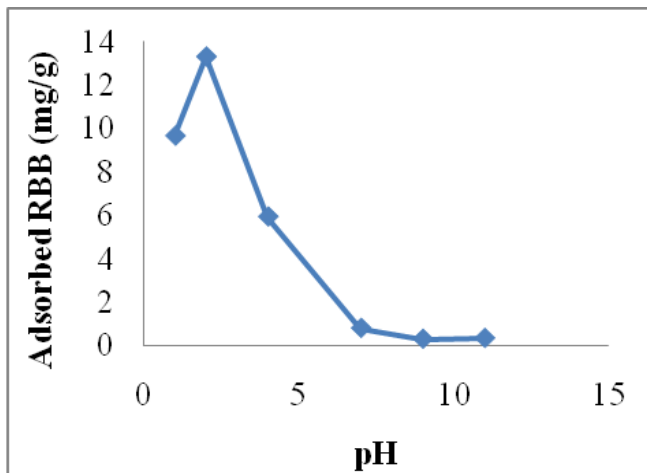


Figure 2: RBB adsorbed pH variations

Increased membrane adsorption power at pH 2 is caused by the presence of H^+ ions contained in RBB solution. The amount of H^+ ions will increase when the pH decreases (the pH becomes more acidic). But at pH 1 the adsorbed RBB content is less than pH 2, because the H^+ ions in the RBB solution pH 1 is very much. This excess H^+ ions prevents interaction between the RBB dye and the active group on the surface of the adsorbent. Whereas in RBB solutions above pH 2 there is a decrease in the adsorbed RBB level, because the number of H^+ ions decreases and there are OH^- ions. This OH^- ions are found in basic pH which has a relatively small number of protons. This causes the dye ions to form hydroxide deposits, thereby reducing the rate of adsorbed RBB [Maghfiroh, 2016]. The efficiency obtained in this condition is 22.11108%.

In the variation of concentration, the optimum condition occurs at a concentration of 60 mg/L with RBB levels adsorbed at 17.49048 mg per gram of adsorbent as shown in Figure 3. At a concentration of 10 mg/L the RBB is adsorbed at 7.93033 mg per gram of adsorbent, increasing up to 17.40825 mg per gram of adsorbent at a concentration of 30 mg/L. The peak of the increase is at a concentration of 60 mg/L. After that the RBB dye adsorbed is reduced to 12.01627 mg per gram of adsorbent at a concentration of 80 mg/L and ends with 7.14084 mg per gram of adsorbent at a concentration of 100 mg/L.

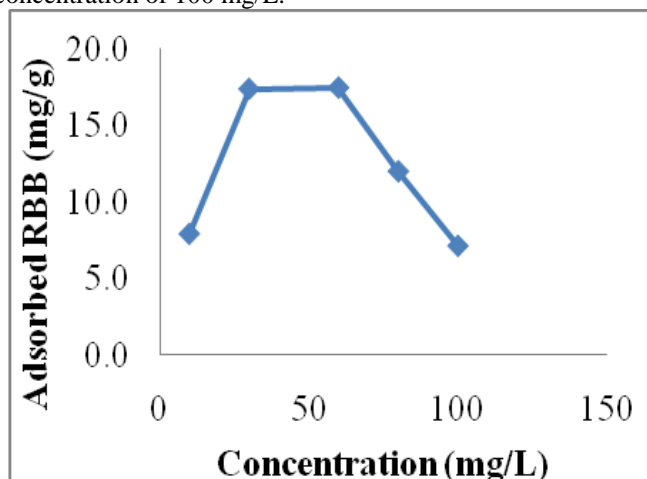


Figure 3: RBB adsorbed at concentration variations

This decrease occurs because the ability of the adsorption of the nata de pina membrane to the RBB dye has reached the

maximum limit, so that the membrane will reach the saturation point and the adsorbate that has not been adsorbed will diffuse out of the pore and return to fluid flow. This is consistent with Langmuir's theory which explains that on the surface of the adsorbent there are active sites whose amount is proportional to the surface area of the adsorbent, so that if the active site is saturated, then the addition of concentration can no longer increase the adsorbent adsorption capacity, resulting in a decrease in adsorption power by the adsorbent [Nurhasni, et. al., 2010]. The efficiency obtained at this optimal concentration is 29.56613%.

Based on the curve in Figure 4, RBB adsorption occurs maximally at 60 minutes contact time. In this condition, the adsorbed RBB is 17.01007 mg per gram of adsorbent. The adsorbed RBB level begins to decrease after this contact time. At 120 minutes of contact time, the adsorbed RBB dropped to 11.33626 mg per gram of adsorbent. Staining of adsorbed RBB continues to decrease until the contact time is 180 minutes. After contact, the adsorbed RBB dye decreased to 10.27311 mg per gram of adsorbent.

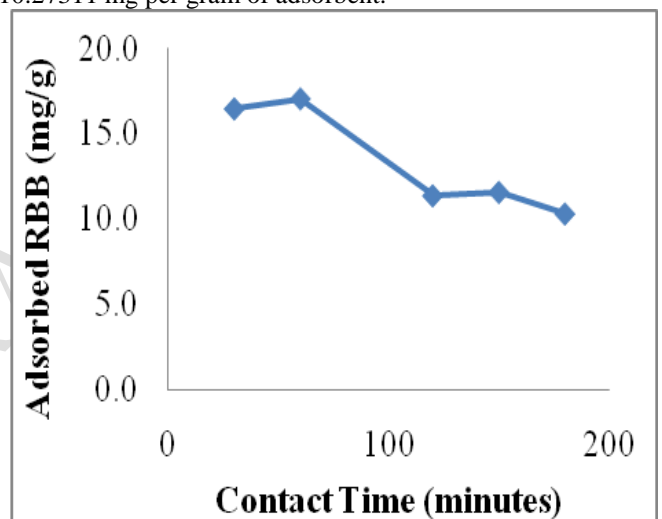


Figure 4: RBB adsorbed at contact time

In this condition, the active site of the membrane is fully filled so the RBB adsorbed level decreases with increasing contact time. This can also be caused by not all adsorbates which are bound to the membrane which is an electrostatic bond, resulting in desorption on long contact time on saturated adsorbents. Desorption occurs due to adsorption of RBB dyes by the nata de pina membrane due to ionic interactions. This interaction make the amine group (NH_2) in the protonated RBB to become amine ion (NH_3^+) which is alkaline. Thus, increasing contact time during adsorption will cause the adsorption process environment to become more alkaline. An alkaline environment will tend to cause desorption events, like is the release of dye back to the membrane [Arifin, et. al., 2012].

At the optimum conditions namely pH 2, concentration of 60 mg/L, and contact time for 60 minutes, the adsorption efficiency of RBB solution using nata de pina membrane was obtained, amounting to 31.69119%.

From the analysis of the adsorption isotherm pattern, the RBB coloring adsorption follows the Langmuir adsorption isotherm pattern with R^2 values of 0.952 and does not meet the

Freundlich adsorption isotherm pattern because the value of the R^2 is 0.466 less than 95% or 0.95 in Figures 5 and 6.

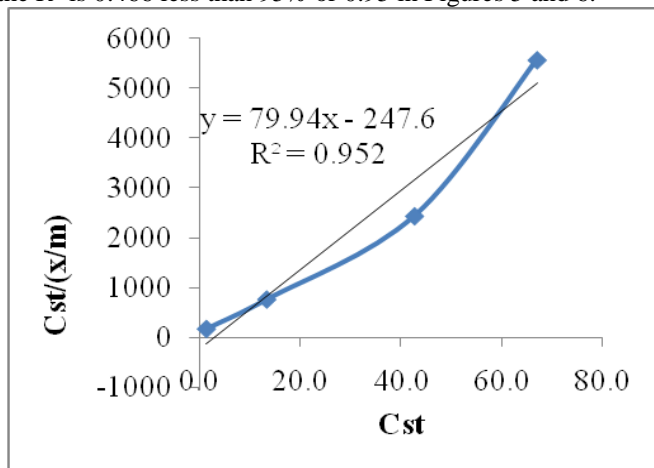


Figure 5: Langmuir isotherm curve

This shows that the membrane can only adsorb one adsorbing molecule for each molecule of the cellulose nata de pina membrane. This membrane has homogeneous surface rather than heterogeneous. In addition, adsorption will only occur in one layer (monolayer).

Maximum adsorption power $(x/m)_{max}$ is calculated based on the value of the slope of the Langmuir adsorption isotherm curve $y = 79,94x - 247,6$. Based on the y value in the line equation, a gradient of $1/(x/m) = 79,94$ is obtained. So the $(x/m)_{max}$ value obtained based on calculations is 0.0125 g/g or equivalent to 12.5 mg per gram of adsorbent. So every 1 gram of adsorbent is able to adsorb up to 12.5 mg of RBB dye.

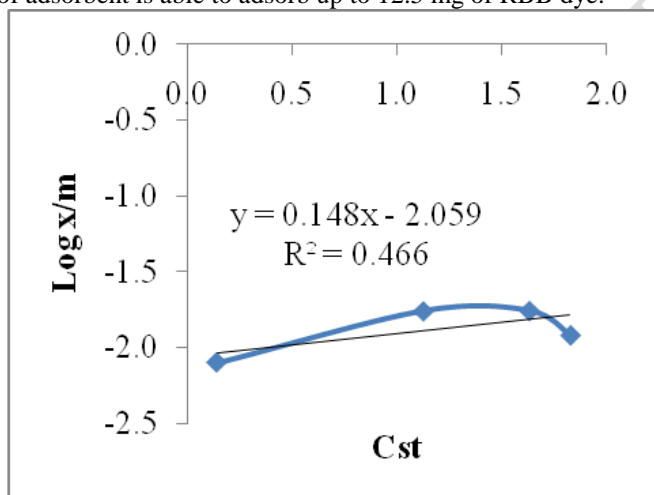


Figure 6: Freundlich isotherm curve

IV. CONCLUSION

Based on the objectives and research results described above, it can be concluded that the cellulose nata de pina membrane in this study under optimum conditions is pH 2, concentration of 60 mg/L, and contact time for 60 minutes has a percent efficiency of adsorption 31.69119%. The RBB adsorption isotherm pattern by the nata de pina cellulose membrane meets the Langmuir adsorption isotherm pattern

with $R^2 = 0.952$ with a maximum membrane of 12.5 mg per gram of adsorbent.

V. SUGGESTION

Based on the results of the research and discussion described above, the thing that can be suggested is the need for further research to test the adsorption power of the cellulose nata de pina membrane in the actual textile dye waste.

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