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Removal of methyl red in wastewater by activated carbon derived from rice husk

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Abstract

The textile industry is the largest industry contributing to the wastewater pollution and is a concern for the environment today. This pollution occurs when wastewater from the textile industry discharges the water into the river and it will affect both aquatic and human life. There are several ways to overcome this pollution problem but the current method is very expensive and its effectiveness depends on the colour of the dye. In this research, activated carbon of rice husk was used as an adsorbent to remove the methyl red dye from the wastewater. To detect the effectiveness of rice husk, two materials were used which were raw rice husk and also activated carbon to remove methyl red dyes from wastewater. There are four parameters used to investigate the optimum level which are the effect of initial dye concentrations ranges from (2 mg/L-10 mg/L), contact time (20-100 minutes), pH (2-10) and adsorbent dosage (0.5-2.5 g). The percentage of dye is calculated and recorded to see the comparison between raw rice husk and also activated carbon of rice husk. The results show that the rice husk can be used as a potential adsorbent to remove methyl red from the waste water.

1. INTRODUCTION

Water is very important in our life, therefore, it needs to be maintained and improving its quality must be continuous. Non point sources and point sources contribute in contaminating valuable water resources. The results of the investigation indicated that groundwater and surface water has been found to be contaminated in many areas (Ahmaruzzaman and Gupta, 2011). Water is important for living things, including human, animals and plants. Today, clean water becomes less due to anthropogenic activities. To minimize the pollution problem it needs to be investigated seriously and appropriate action needs to be taken to ensure the problem is solved. There are many other environmental pollution but water pollution is a pollution that attracts scientists and researchers around the world (Pirkarami and Olya, 2014). There are several activities that contribute to water pollution, for example; the pollution comes from industries such as mining, painting, agriculture, and textiles. The sources of this pollution, especially textiles will eventually cause water pollution. The major contributor to many of the country's economy is textile industry, encompassing small and large-scale operations around the world. The textile industry is the largest industry in the world where it is capable of providing apparel to human beings. The textile process is classified as an industry that uses high water resources, fuel and chemicals which capable of producing high waste. The textile industry is the main source of producing dye in wastewater. The most serious thing is that when the textile industry is a major cause of water body pollution due to the untreated waste disposal (Donmez, 2002). The textile processing industry is not only dependent on the dyes but it depends on the various types of substances such as heavy metals which are capable of affecting the wastewater. There is a long sequence of processing stage that requires chemical, water and energy inputs and generates waste at every level. Among materials from the processing of textile are liquid, gas and solid waste which could harm the aquatic life and human when the wastewater is discharged into the river (Uma *et al*., 2009). Thus, this study aims to remove methyl red dye by using an environmentally friendly material,

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rice husk that was carbon activated as a potential adsorbent in dye removal.

2. MATERIALS AND METHODS

2.1 Raw material collection and preparation

Rick husk was obtained from the production of rice mill at Kota Bharu. The rice husk was washed using distilled water to clean the dirt. After that it was dried in the oven at 105°C for 2 hours. Then, it was ground using grinder and sieved until the rice husk become powder. The rice husk was kept in the different air-tight container for further use.

2.2 Preparation of activated carbon

500 g of rice husk material sample was stirred in 500 ml of 0.2 M nitric acid, HNO_3 for 24 hours at room temperature. The sample of rice husk was put in the furnace for 1 hour and it was cooled before the sample was weighed. Then, the sample of rice husk was washed with distilled water using filter paper. This sample was washed until the pH was 7. The washed samples were put in the oven at 55 °C for 24 hours. The rice husk becomes activated carbon. The sample was weighed after dried in the oven for 24 hours. This activated carbon was stored in desiccator (Bharathi and Ramesh, 2013).

2.3 Preparation of methyl red solution

Methyl red dye was used in this study to investigate the efficiency of rice husk as an adsorbent. The solution was prepared by dissolving 1 g of dye in 1 L of distilled water to get the concentration of 1000 mg/L. This dilution was prepared to get the 2, 4, 6, 8, and 10 ppm of methyl red dye concentration for standard series solution.

2.4 Preparation of blank solution

The blank solution of the adsorbent was

The blank solution of the adsorbent was prepared by using 50ml of methyl red dye solution transferred into a 100ml conical flask. The flask was then placed into a constant temperature 35°C orbital shaker at 300rpm and shaken for 24 hours. The reading obtained from blank solution was used as baseline for the optimization of the adsorption. The value of standard pH for blank is 7. The obtained reading from blank solution was expected to be zero

2.5 Optimization for adsorption

The effect of adsorbent dosage was determined by using adsorbent mole ratio of 0.5, 1.0, 1.5, 2.0 and 2.5 g activated carbon rice husk. This sample was added into 50 ml of the methyl red dye solution in the different conical flask and was put in orbital shaker at 130 rpm about 30 minutes to observe the changes of adsorption rate using the different of adsorbent dosage.

The effect of contact time was determined by setting the contact time in 20, 40, 60, 80 and 100 minutes. The effect of pH was analysed over the pH range from 2-10 by mixed with 0.5 g of adsorbent. The dilution from 1000 mg/L methyl red was prepared to get the 2, 4, 6, 8, and 10 mg/L of initial dye concentration (Bharathi and Ramesh, 2013).

2.6 Calculation of adsorption efficiency

The adsorption capacity and efficiency of rice husk as adsorbent was estimated by using this formula (Takdastan *et al*., 2016):

$$qe = \frac{v}{m} x \, (C_o - C) \quad \text{--------------- Eq. 1}$$

Where,
v = volume of solution (ml)
m = mass of adsorbent (mg)
C_o = initial concentration
C = final concentration

Meanwhile, the calculation for the percentage of dye removal was calculated by using the equation 2 (Takdastan *et al*., 2016):

$$E = C_o - \frac{C}{Co} x \, 100\% \quad \text{------------------ Eq. 2}$$

3. RESULTS AND DISCUSSION

3.1 Effect of contact time on dye adsorption

The result shows the comparison of the effectiveness between the raw material and activated carbon from rice husk to remove methyl red dye. This experiment was performed to determine the highest percentage removal of methyl red dye with different contact time. The contact time from 20 to 100 minutes and 0.5 g of adsorbent were used for 50 mL methyl red dye solution. Figure 1 shows the changes that occurred when the raw material and activated carbon of rice husk adsorption was carried out on methyl red (MR) dye with different time series. The graph shows that when using raw material there was an increase from 60 to 80 minutes where the percentage increase from 57.8% to 78.4%.

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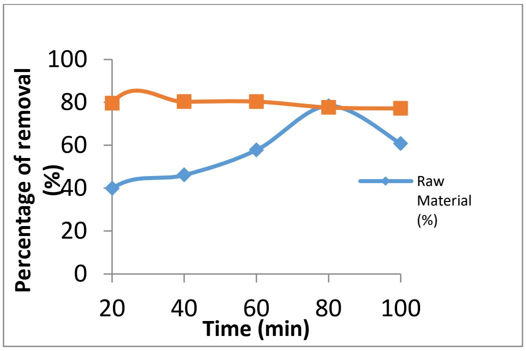

Figure 1: Effect of contact time for raw material and activated carbon.

This graph shows that from 20 minutes to 80 minutes, the adsorption using raw material increases rapidly from 39.84 % to 78.4% and then decreases when reach 100 minutes. Therefore, the optimum contact time was considered to be 80 minutes with 78.4% of percentage removal. When the concentration of dye did not achieve the equilibrium, the percentage removal of methyl red will decrease and the graph show that the increase of contact time, the adsorption capacity will decrease. Meanwhile activated carbon recorded 80.37% (min 60) of removal and reached equilibrium state. From that, it can be observed that raw material has less potential to remove the dye at the higher contact time. This result obtained shows that the activated carbon is more efficient compared to raw material of rice husk. This is due to the small, low volume pores that increase the surface area from activated carbon of rice husk and it was reported to have a highest sorption capacity compared to other agricultural wastes (Bharathi and Ramesh, 2013).

3.2 Effect of Initial Dye Concentration

The effect of initial dye concentration is shown in Figure 2. The different concentrations of dye were used to observe the percentage removal. From Figure 2, it shows that the adsorption of rice husk increases with the increase of methyl red concentration at the concentration 2 mg/L until 4 mg/L.

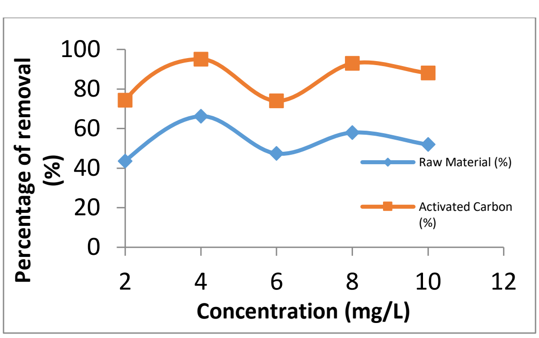

Figure 2: Effect of initial dye concentration for raw material and activated carbon.

The initial dye concentration of an effluent is important since activated carbon can only adsorb a fixed amount of dye (Bharathi and Ramesh, 2013). The graph in Figure 2 shows that the percentage removal of dye using raw material increased from 43.59 % to 66.24 % at 2 mg/L and 4 mg/L. After that, the percentage of removal decrease to 47.37 % at 6 mg/L. Raw material has reached a maximum concentration at a concentration of 4 mg / L as the surface of the raw material is no longer able to adsorb the excess dye. This is because raw materials do not have an active surface area to adsorb high amount of dyes. Besides it is due to the different structure of raw material from activated carbon and it did not produce interactions between raw material surface and the adsorbate (Guo *et al*., 2002). From this result, it shows that the optimum concentration for raw material is 66.24 % at 4 mg/L of initial dye concentration.

On the other hand, the adsorption capacity of activated carbon increased from 74.26 % to 95.1 % (2 to 4 mg/L). The highest percentage is the optimum result for effect of initial dye concentration. From this observation, it can be proven that the methyl red adsorption has to encounter the boundary layer film onto adsorbent surface and then finally, it has to diffuse into the porous structure of the adsorbent (Rahman and Akter, 2016). Maximum dye removal was recorded at 4 mg/L (95.1%).

3.3 Effect of Adsorbent Dosage

Figure 3 shows the percentage removal of wastewater sample using the adsorbent dosage from 0.5 g to 2.5 g. This experiment was carried out at 30°C of temperature with 130 rpm using orbital shaker and the contact time was 80 minutes.

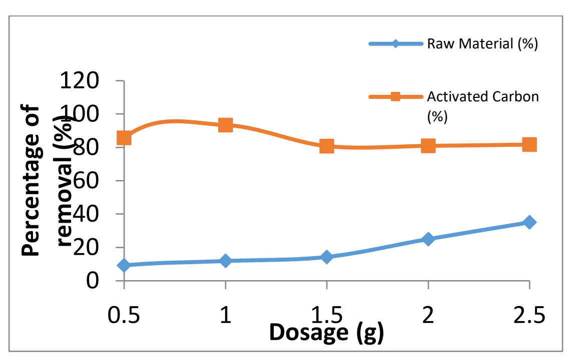

Figure 3: Effect of adsorbent dosage for raw material and activated carbon.

The graph demonstrates that the percentage of removal increased with increasing amount of dosage. This happened because initially the site of adsorbent could not effectively contact with adsorbate.

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When the adsorbent amount was higher, the adsorbent site of the raw rice husk was available for removing certain amount of methyl red dye from wastewater sample. The result shows that the percentage removal increases when the amount of dosage increases. From Figure 3, 0.5 g of dosage can remove 9.19 % of dye and this reading changed to 35.03 % when the dosage was 2.5 g. Raw material requires high dosage to ensure that the adsorption done on the dye is effective. The optimum dosage to remove dye is 2.5 g of raw rice husk.

Figure 3 also shows that the percentage of removal was slightly increased from 0.5 g to 1 g which is 85.71 % to 93.39 %. Meanwhile, from 1.5 g the graph showed decreased adsorption which is 80.77 %. This is due to the saturation occurred at 1.5 g. From the observation, the optimum value of percentage removal is at 1 g. The increase in dye removal in dye removal percentage with adsorbent dosage can be attributed to increase adsorbent surface area and availability of more adsorption sites (Afrah and Abdulhussein, 2015). From the graph it can be observed that the at beginning of the process the rate of the dye removal percentage by activated carbon from rice husk was fast compared to the raw rice husk.

3.4 Effect of pH

The pH value of the solution is an important parameter for the adsorption process. The variety of pH can show the significant influence to this experiment. In general, pH value may enhance or depress the uptake of adsorption for removal the dye. This is attributed to the change of adsorbent surface with the change of pH value (Gupta *et al.*, 2012). Figure 4 shows the relationship between pH values and the percentage removal of dye from wastewater sample using raw material and activated carbon of rice husk.

The result shows the increment from pH 2 until pH 6 which is from 21.79 % to 40.9 % of dye removal in raw material. The optimum pH of this parameter was 6 for raw material. The ability of adsorbent to adsorb adsorbate was affected by acidity condition of adsorbent surface. From the graph it shows that the percentage removal of dye increases at the beginning. Based on previous research, if the acidity of solution is lower it will give potential to adsorb more dyes from water because when the water is in acidic solution it will produce salt as a factor that can react with dye. This percentage increases at pH 2 until pH 6, after that the decrement occurs when the acidity condition was changed (Taha *et al.*, 2012).

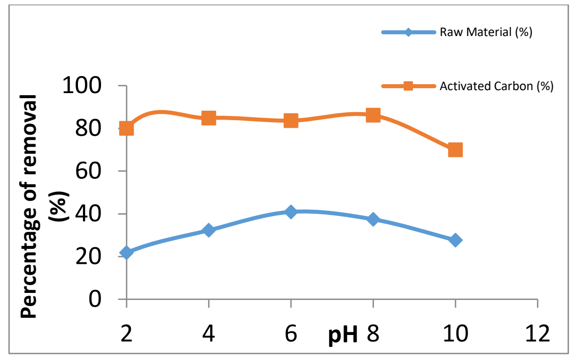

Figure 4: Effect of pH for raw material and activated carbon.

The result shows the increasing of removal percentage from pH 2 to pH 8 which is 80 % to 86.9 % from the activated carbon. Meanwhile the lowest adsorption capacity occurs at pH 10 with 69.91 %. Based on previous researches, it explains that the highly effective pH in removing this dye is the pH in which is acidic. There are two types of acid dissociation which is strong acid and weak acid in water. Strong acids and weak acids have the same chemical properties but the rate of reaction and the weakness of the weak acidic electricity are lower. The weak acid is partly partially acidized in water to produce hydrogen ions, H^+, with low concentrations. Contrary to the result of this study, this may be due to the activated carbon state remained at pH 6 where it reacts to the surface of the coloured solution. In addition, when reached at pH 10, the adsorption decreases. The increasing in the activated carbon adsorption of the dye with decreasing of pH values is due to the attraction between the azo dye and excess H^+ ions in the solution (Afrah and Abdulhussein, 2015).

Table 1 shows that all of the parameters tested ie: time, concentration, dosage and pH had significant difference ($p<0.05$) between raw material and activated carbon of rice husk.

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Table 1: Significant difference between raw material and activated carbon.

Time	Activated carbon	Raw Material
Mean	79.01	56.426
Variance	2.27635	212.80383
Observations	5	5
Hypothesized Mean Difference	0	
df	4	
t Stat	3.443386	
P(T<=t) one-tail	0.013106	
t Critical one-tail	2.131847	
P(T<=t) two-tail	0.026212	
t Critical two-tail	2.776445	
Concentration	**Activated carbon**	**Raw Material**
Mean	84.86	53.414
Variance	102.43205	80.17953
Observations	5	5
Hypothesized Mean Difference	0	
df	8	
t Stat	5.203389	
P(T<=t) one-tail	0.000410	
t Critical one-tail	1.859548	
P(T<=t) two-tail	0.000819	
t Critical two-tail	2.306004	
Dosage	**Activated carbon**	**Raw Material**
Mean	84.504	19.07
Variance	28.72458	115.65555
Observations	5	5
Hypothesized Mean Difference	0	
df	6	
t Stat	12.176844	
P(T<=t) one-tail	0.000009	
t Critical one-tail	1.943180	
P(T<=t) two-tail	0.000019	
t Critical two-tail	2.446912	
pH	**Activated carbon**	**Raw Material**
Mean	80.888	32.016
Variance	42.83477	57.89393
Observations	5	5
Hypothesized Mean Difference	0	
df	8	
t Stat	10.888511	
P(T<=t) one-tail	0.000002	
t Critical one-tail	1.859548	
P(T<=t) two-tail	0.000004	
t Critical two-tail	2.306004	

4. CONCLUSION

The present investigation showed that the agriculture waste of rice husk can effectively be used as an activated carbon for removal of dye from wastewater under optimum parameter. This effectiveness of rice husk activated carbon was proven when this study was compared with raw rice husk. From the result, it shows that the removal percentage from activated carbon is higher than raw material of rice husk. This is due to the activated carbon that has extended surface area, high adsorption capacity, microporous structure and special surface reactivity.

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