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"Batik" Industry Wastewater Treatment via Coagulation-Flocculation Process and Adsorption Using Teak Sawdust Based Activated Carbon

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Abstract

Untreated wastewater of Batik industry can pollute the environment because it contains metal compound, COD, BOD, which are higher than the allowable values. Therefore, a treatment of this wastewater prior discharging to water stream (i.e. river) is very important. This research aims to investigate the use of Teak sawdust as activated carbon, and also the effect of adsorbent concentration, adsorption contact time, as well as coagulation-flocculation-adsorption sequencing process to the level of COD, BOD, and Zn in Batik wastewater. The Batik wastewater used for this research obtained from Batik industry in Rembang, which mostly used naphtol as the coloring agent. The wastewater was initially treated by coagulation-flocculation process, followed by adsorption process. The coagulant-flocculant used in this research was 1 g/L of alum and 3 g/L of lime. Whereas, the adsorbent used was activated carbon made from Teak sawdust with variation of concentrations: 10, 16, 23, and 26 g/L. Whereas, the adsorption contact times were 20, 40, 100, 160, and 220 minutes. The results showed that the coagulation-flocculation process was able to decrease the levels of COD, BOD, and Zn by 73.28%, 73.62%, and 79.21% respectively. Additionally, the adsorption process by activated carbon also further decreased the levels of COD, BOD, and Zn significantly. Based on the results, the optimum concentration of activated that gave the best result was 26 g/L with 220 minutes contact time. Overall, the combination of coagulation-flocculation and adsorption sequencing process was able to decrease the level of COD, BOD, and Zn up to 96.69%, 96.90%, and 91.90% respectively.

INTRODUCTION

Batik is one of Indonesian cultural heritage. It is a traditional textile art originating from the island of Java. In Rembang city, Central Java, there are Batik Tourism villages, in which the majority of the population are making Batik as their main source of livehood. Batik making process usually produce liquid waste, which mainly sourced from the coloring process. The use of chemicals in the coloring process causes the liquid waste has characteristic such as high levels of COD, BOD, and Zn metals with concentration of 6972.01 mg/L, 2161.62 mg/L, and 56 mg/L respectively. If this liquid waste is directly discharged into the water without proper treatment, it can reduce the water quality index, and thus negatively affect the environment especially the water ecosystem (Mohan et al., 2005). Therefore, Batik wastewater must be treated to comply with the water quality standards set by the government. The standard level for discharged wastewater set by the government is maximum BOD 150mg/l, COD 300mg/l and Zn 5.0 mg/l (Perda Jawa Tengah, 2012).

Several methods have been developed to reduce the level of COD and BOD pollution in Batik wastewater. According to Meena et al. (2013), treating Batik wastewater using coagulation-

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flocculation followed by adsorption process is considered to be the most economical method. Coagulation-flocculation and adsorption sequencing process is superior because of their low cost and simple equipment design. To accelerate the coagulation-flocculation process, coagulants are needed. Type of coagulant commonly used for wastewater treatment is aluminum sulfate ($Al_2(SO_4)_{3}.{}_{14}H_2O$), which is due to its economical potential compared to other coagulants (Ignasius, 2014). Nevertheless, coagulation-flocculation process is not enough to reduce the level of COD, BOD and Zn metal in Batik wastewater. Further process is needed, such as adsorption. Adsorption is considered as the most promising alternative method due to its simplicity, low cost, and can be applied at even low concentrations (Chafidz, 2018). The adsorption process using activated carbon as the adsorbent can absorb or reduce the level of COD, BOD and Zn metals (Yuan & Liu, 2013). Adsorbent material can be obtained from biomass waste that is processed into activated carbon. One of the biomass waste that has high cellulose content is teak sawdust with 50.90% of cellulose (Lukmandaru et al., 2016). The quality of activated carbon is not only influenced by the characteristics of raw materials, but also influenced by the method of activation (Patil & Kulkarni, 2012). NaCl is chosen as an activator because it is easy available, low cost, harmless, and non-toxic. The use of NaCl activators after the carbonization process helps to expand the surface area of the pores and helps remove the impurities formed during carbonization, because these impurities can cover the pores of the activated carbon (Hartini et al., 2014)

MATERIALS AND METHODS

Materials

Batik liquid waste used for this research was obtained from "Rosyta Batik" in Rembang city, Central Java. Teak sawdust is obtained from furniture home industry in Jepara. The sodium chloride (NaCl) with the purity of 99.5% purchased from Merck was obtained from Chemical Engineering Laboratory Universitas Negeri Semarang. Aluminum sulfate (Al2(SO4)3.14H2O) and lime (CaO) was technical grade was obtained from Indrasari.

Equipment

Equipment used for this paper were beakerglass, oven Ecocell 55, shaker Unimax 2010, furnace. The analysis methods and equipment used were spectrophotometer, FTIR Perkin Elmer Spectrum Version 10.4.00 , AAS PinAAcle 900F Singapura, SEM JED-2300 Analysis Station, BET Quantachrome Instruments Version 3.01and ICP-EOS.

Coagulation-Flocculation Pretreatment of Batik Waste

The Batik liquid waste was filtered and then put into a 2000 mL beaker glass. Alum 1 g/L was added and stirred at 100 rpm for 15 minutes. Then adding 3 g/L of lime and stirred at 60 rpm for 20 minutes. The mixture was idled for 24 hours and then filtered to separate the waste and sediment formed.

Synthesis of Activated Carbon

Carbonitation

Teak sawdust is washed and then dried using oven at 110°C for 4 hours, then sieved using 120 mesh sieve. The carbonization process was carried out using furnace at 700°C for 1 hour.

Activation

The carbon is then activated using 5 M NaCl and stirred for 30 minutes. The solution was filtered and the residue was dried using oven at 110°C for 4 hours. The dried residue (activated carbon) is washed with HCl and rinsed with distilled water until pH ±7 then dried in oven at 110°C for 4 hours.

Adsorption of COD, BOD, and Zn

The adsorption process was carried out with different activated carbon concentrations (i.e. 10, 16, 23 and 26 g/L) and contact times (i.e. 20, 40, 100, 160, and 220 minutes)

RESULTS AND DISCUSSIONS

Coagulation-Flocculation Process Analysis

In coagulation-flocculation process, coagulant aluminum sulfate ($Al_2(SO_4)_{3}.{}_{14}H_2O$) was added to break the stability of the colloidal particles. Lime (CaO) was added to enhance the formation of

Table 1. Results of coagulation-flocculation process towards Batik waste

	pH	COD (mg/L)	BOD (mg/L)	Zn (mg/L)
Standards	6.0-9.0	300	150	5.00
Before coagulation-flocculation	5.0	6972.01	2161.32	40.97
After coagulation-flocculation	6.0	1862.66	569.97	22.75
% reduction	-	73.28%	73.62%	44.47%

flock that will form deposit and increase the pH of the Batik wastewater to pH 6. Based on the literature, the optimum condition of coagulation-flocculation using aluminum sulfate coagulant was at pH 6 (Coniwanti, 2013). Additionally, the use of pH 6 in the treatment of Batik wastewater is also to comply with the established quality standards set by the government, which is the pH should be in the range of 6.0–9.0. The analysis results of COD, BOD, and Zn after the coagulation-flocculation process are shown in Table 1.

Based on Table 1, the coagulation-flocculation process was able to reduce the levels of COD, BOD, and Zn contained in Batik liquid waste. In this process, the contents of COD, BOD, and Zn metal were absorbed by solids or flock formed by the addition of coagulants. The reaction occured by the addition aluminum sulfate ($Al_2(SO_4)_3.14H_2O$) and lime (CaO) produced aluminum hydroxide ($Al(OH)_3$) which will settle as flock and $CaSO_4$ as calcium salt. The aluminum hydroxide acted as flock will surround the colloidal particles containing COD, BOD, and Zn metal ions. Due to their properties, colloidal particles will attach to the surface of $Al(OH)_3$ to neutralize the surface charge. Colloids containing COD, BOD, and Zn will be trapped when the flock is formed and remain as it is when settles (Said, et al., 2010). Coagulation-flocculation undergoes an optimum process at pH near neutral. This is because at acidic pH, the solubility of $Al(OH)_3$ will increase to form positive charges $[Al(OH)_2]^+$ and $[Al(OH)]^{2+}$. At alkaline pH, the solubility of $Al(OH)_3$ will increase to form a negative charge $[Al(OH)_4]^-$. The change in $Al(OH)_3$ solubility causes reduction of $Al(OH)_3$ ability to absorbs the surrounding particles.

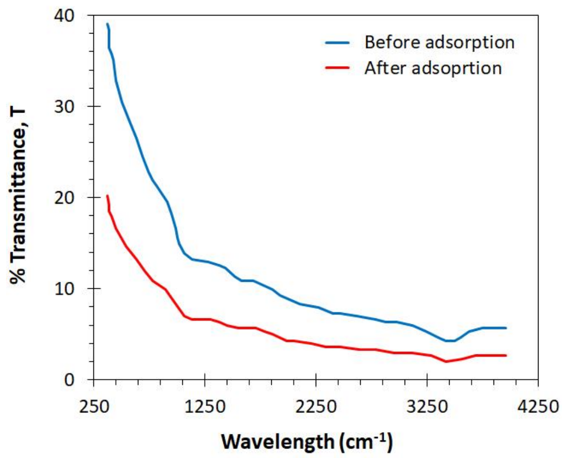
Characteristics of Teak sawdust based activated carbon

The Teak sawdust based activated carbon was analyzed using FTIR to determine the availability of functional groups. Functional cluster analysis is carried out with a wavelength of 370-4000 cm^{-1}. Based on Figure 1a, activated carbon before adsorption produces absorption peaks at 3332 cm^{-1}, 1701.23 cm^{-1}, 1612.38 and 877.40 cm^{-1}. The absorption peak of 3333 cm^{-1} indicates the presence of O-H bonds with alcohol or carboxylic acid groups. The absorption peak of 1701.23 cm^{-1} indicates the presence of a C=O bond with an aldehyde compound, while at 1612.38 cm^{-1} indicates the presence of a C=C bond with an alkene compound. The absorption peak 877.40 cm^{-1} indicates the presence of CH bonds with aromatic ring compound groups (Skoog, 1998). While the activated carbon after adsorption process showed a decrease in intensity at wave numbers 3436.07 cm^{-1}, 1701.55 cm^{-1}, 1619.38 cm^{-1} and 876.39 cm^{-1}, which indicating that OH, C=O, C=C and CH groups decreased as due to the adsorption process. The reduction in intensity and absorption peaks confirmed that adsorption type which occurred on the active sites was chemisorption (Astuti & Kurniawan, 2015).

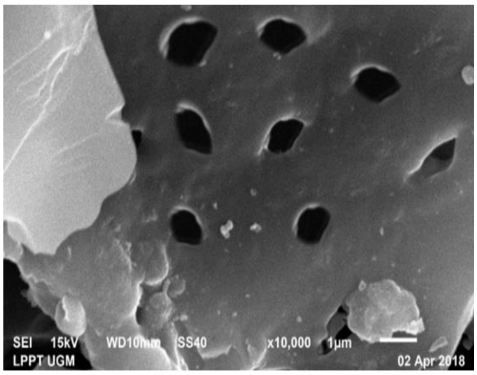
Additionally, surface morphology of Teak sawdust based activated carbon was also analyzed using a scanning electron microscopy (SEM). Figure 1b shows the SEM micrograph of the activated carbon prior to the adsorption process. As seen in Figure 1b, some of pores were opened, though some of them still covered with dirt. The formation of pores was due to the initial heating or carbonization. The activation by NaCl after the carbonization process helped to widen the surface area of the pores and helped to remove impurities formed during carbonization (Hartini et al., 2014). Additionally, the Brunauer Emmet Teller (BET) analysis was also used to determine the surface area of activated carbon through N_2 gas physisorption at 77.35 K using NOVA quantachrome apparatus. Based on BET analysis, the activated carbon has a surface area of 25.441 m^2/g.

The Influence of Activated Carbon Concentration at COD, BOD, and Zn Reduction

The effect of activated carbon concentration on decreasing levels of COD, BOD and Zn metal is shown in Figure 2. Based on Figure



(a)



(b)

Figure 1. (a) FTIR result and (b) SEM micrograph of Teak sawdust based activated Carbon

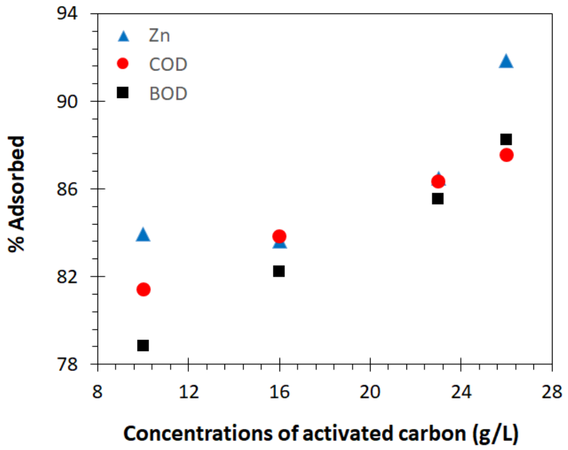


Figure 2. The effect of activated carbon concentrations on the reduction of COD, BOD, and Zn.

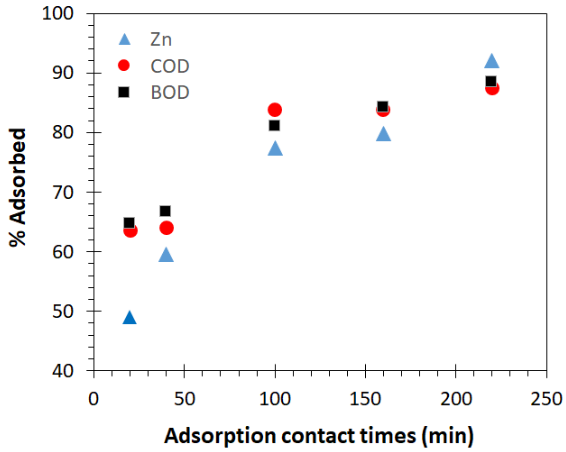


Figure 3. Effect of contact time towards COD, BOD, and Zn reduction.

2, the increase of activated carbon concentration added will affect the amount of COD, BOD, and Zn absorbed. The concentration of activated carbon at 26 g/L relatively absorbs highest amount of COD, BOD, and Zn. This is because the increase of activated carbon concentration is proportional to number of particles which causes larger active site resulted in better adsorption process until the equilibrium condition is reached. However, for further addition of activated carbon will influenced by its equilibrium state.

The effect of adsorption contact time on the levels of COD, BOD and Zn metal is shown in Figure 3. The figure shows that the Zn content rapidly decreased in the adsortion contact time of 20 to 100 minutes. It is because there were still many empty active sites to adsorb the solution before reaching equilibrium state (Owamah et al., 2013). The longer duration of adsorption process means more adsorbent will interact and collide with Zn solution, which resulted in the greater adsorption (Anggrenistia et al., 2015). It was proposed in this study that the equilibrium state was not reached under 220 minutes of adsorption contact time. Therefore, the optimum contact time of Zn adsorbtion was 220 minutes.

Additionally, the results of COD, BOD, and Zn after coagulation-flocculation and adsorption sequencing process are presented in Table 2. As seen in the Table, the coagulation-flocculation and adsorption sequencing process was able to reduce the levels of COD, BOD, and Zn up to 96.69%, 96.90%, and 91.90% respectively. The combination of coagulation-flocculation and adsorption sequencing processes on Batik liquid waste was able to comply the minimum quality standards set by the government.

Table 2. Analysis COD, BOD, and Zn after coagulation-flocculation and adsorption sequencing processes

	pH	COD (mg/L)	BOD (mg/L)	Zn (mg/L)
Standards	6.0-9.0	300	150	5.00
Before coagulation-flocculation	5.0	6972.01	2161.32	40.97
After coagulation-flocculation	6.0	1862.66	569.97	22.75
After adsorption	6.0	230.9	67.04	1.842
% Reduction	-	96.69%	96.60%	91.90%

CONCLUSION

The coagulation - flocculation and adsorption sequencing process was carried out to reduce the levels of COD, BOD, and Zn contained in the wastewater produced by the Batik industry in Rembang city, Central Java. The coagulation-flocculation process was able to decrease the level of COD, BOD, and Zn by 73.28%, 73.62%, and 79.21% respectively. Additionally, the adsorption process by activated carbon also further decreased the levels of COD, BOD, and Zn significantly. Based on the results, the optimum concentration of activated that gave the best result was 26 g/L with 220 minutes contact time. Overall, the combination of coagulation-flocculation and adsorption sequencing process was able to decrease the level of COD, BOD, and Zn up to 96.69%, 96.90%, and 91.90% respectively. Additionally, the BET analysis shows that teak sawdust activated carbon surface area was 25.44 m^2/g.

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