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# **Comparison of Different Volume Reactor for Batik Wastewater Pre-Treatment with Ozonation to Improve Biodegradability**

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#### Abstract

Batik is a unique ethnic and heritage textile from Indonesia with low biodegradability (BOD<sub>5</sub>/COD) and demanding biological wastewater treatment. However, in its application a high biodegradability value is needed. To increase biodegradability the ozonation process can be used as pre-treatment. The purpose of this study is to determine the effectiveness of ozone pre-treatment in color removal, COD removal, and changes in biodegradability. This study was divided into two types of the reactor with a volume of 2 L and 16 L with each ozone dose of 4 mg/min and 40 mg/min. The results of color removal in the reactor with a volume of 2 L showed a value of 85% for color removal and 34.6% for COD removal. The reduction in efficiency occurred in the color removal to 65% in reactor 16 L. The variables used are the location of the sample point, volume reactor and the time of sampling. The removal of COD with a volume of 16 L was not evenly distributed at each altitude of 43; 35; and 33% (50; 100; 150 cm). Biodegradability increased from 0.143 to 0,49 (2L reactor) and 0.4-0.45 (16L reactor), this allows for the application of higher biological wastewater treatment. Which the NH<sub>3</sub>-N and total phenol removal efficiencies of 12.9%-31.4% and 3-21.2%, respectively.

Keywords: Pre-treatment ozone, batik wastewater, biodegradability, COD removal, color removal

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## **INTRODUCTION**

Batik is one of the textile products that have increased rapidly because nowadays batik as a fashion starting to develop. In addition, batik designated as one of Indonesia's original cultural heritage by UNESCO. This added to the people's love for this type of fashion increased. Along with the increasing number of batik consumers, existing batik that primarily produce byproduct including solid waste and wastewater. The batik industry is included in the textile industry, which consumes the most water in its production process, because of which wastewaterproduced reaches 3919–8159 L/pc (Handayani et al., 2019) or 17.93 L/pc (Handayani et al., 2018). Wastewater from textile industry is one of the basic environmental problems (Xu et al., 2018; Apritama et al., 2020). The most commonly used batik wastewater treatment is conventional sedimentation or storing waste in a reservoir. Some of the ingredients in batik industrial wastewater that have the potential directly discharged into the water body cause water pollution are the ingredients of color, organic, suspended solids and high heavy metals (Tambunan et al., 2018). Eutrophication impacts of excess batik wastewater because of inputs on waterbody and ecosystems (Handayani et al., 2018; Yoshanti et al., 2017). Textile wastewater treatment such as biological treatment, adsorption, advanced oxidation processes, coagulation and flocculation, membrane filtration, membrane technology, have been apllied (Kristianto et al., 2018; Hastuti et al., 2020).

Characteristics of the batik wastewater have low biodegradability. The biodegradability of batik wastewater only showed of BOD5/COD (341.25 mg/L / 4092 mg/L) only 0.08 (Khalik et al., 2015). Another study of Subramaniam et al., (2016) result that the biodegradability index is only 0.015. The natural dye used in batik wastewater has biodegradability between 0.15-0.4, is better than the synthetic dyes with an average of 0.05 (He et al., 2017). This result shows that batik wastewater has very low biodegradability value and difficult to apply to the biological treatment process. Some examples of previous research with biological processing are not enough to meet the quality standards of batik wastewater discharge. Combination Up-flow Anaerobic Reactor (UAR)-Activated Sludge (AS) only remove COD efficiency of 74.98 -91.66% ranged between 139 mg/L and 514 mg/L (Yuliasni et al., 2018).

Aerobic condition only removed BOD 80-85% within the range concentration of 1,000-2,000 mg/L (Suhartini et al., 2019). In anaerobic treatment, which using bio-ball media only removed COD by 90.99% with initial COD of 2220 mg/L and final COD of 200 mg/L (Aliyuddin et al., 2018). Ozone pre-treatment with anoxic-aerobic reactor also be successful in increasing the efficiency of Endek wastewater treatment and meets the quality standard (Suryawan et al., 2019). Another study also increase the COD and color removal from Reactive Black 5 wastewater with combination of ozone pre-treatment and Moving Bed Biofilm Reactor (MBBR) (Pratiwi et al., 2018).

One treatment, which recommended as a pretreatment before biological treatment, is the ozonation. Ozone is a technology that produces OH radicals that can break down the long chain organics into short chains in wastewater, especially batik wastewater that has low biodegradability. Biodegradability also showed of the chemical transformation processes (Ahmadi et al., 2017). In its application, treatment applicable on a small scale or large scale. For this reason, the purpose of this study was to determine the degradation of color, COD, and improvement biodegradability of batik wastewater on a laboratory scale (2 L) and large scale (16 L). The characteristics used are artificial wastewater that has been adapted to the characteristics of the original wastewater.

#### METHOD Meterials

# Materials

The ozone dosage was determined by the iodometric method, according to the Standard Methods No 2350 E procedure. The wastewater used in this experiment is real wastewater from the storage resulting from the batik printing process in one of the batik industries located in Cimahi City, West Java Province. The reactor used in this study was divided into two types, namely small scale with a volume of 2 L and an upscale with volume 16 L. The initial value of COD batik wastewater is 846 mg/L. The initial batik wastewater BOD<sub>5</sub>/COD is 0.49.

#### Methods

In a 2 L reactor, the reactor height is 50 cm while the 16 L reactor height is 200 cm. In each reactor, air flowed from the aerator by passing ozone generator through the flow meter. Each ozone dose on a small scale is 4mg/min and 40 mg/min, while on an upscale of 40 mg/min. Color and COD parameters are measured once every 30 min utes within 240 minutes of treatment. For BOD parameters measured at the beginning and in the end of treatment. COD, color, and BOD measurement in upscale reactor measured at different heights, namely at the altitude of 50, 100 and 150 cm. The parameters tested were organic ingredients in the form of COD, color, and BOD. COD examination using a closed reflux method spectrophotometric refers to SNI 06-6989.2-2004. For color analysis, refer to spectrophotometric SNI M-03-1989-F. Whereas for BOD analysis use the standard SNI 6989.72:2009. For ammonia-nitrogen (NH<sub>3</sub>-N) and total phenol parameters measured at the beginning and end of pre-treatment. NH<sub>3</sub>-N and total phenol examination refers to SNI 06-6989.30-2005 and SNI 06-6989.21-2004 respectively.



(a) 2 L pre-treatement ozone reactor (b) 16 L pre-treatement ozone reactor

Figure 1. Schematic pre-treatment reactor with volumes of 2 L (a) and 16 L (b)

#### **RESULTS AND DISCUSSION**

#### **Color and COD removal**

In the 12 minutes of ozonation, the color change has not occurred significantly. The initial concentration of batik wastewater was 3319.55 Pt-Co. The Platinum-Cobalt (Pt-Co) color scale measures the "yellowness" of a liquid and is well suited for quality

control and contamination detection. Based on the Pt-Co unit, color removal in 2 L reactor that is around 85% (Fig 2a). The results of visual color removal can be seen in Figure 3. The ozone dose of 4 mg/min and 40 mg/min only accelerates the color removal process in 0-120 minutes. The initial value of COD batik wastewater is 846 mg/L with COD removal is around 34.6% (Fig 2b).

Study of Estikarini et al., (2016) in textile wastewater, it was successfully removed COD with initial characteristics of 1,096 mg/L to 377 mg/L for 105 minutes with an ozone dose of 32 mg/minute. Another wastewater treatment with pre-treatment for increase removal applied with biological treatment (Malik et al., 2019; Wu et al., 2018).



Figure 2. Color (a) and COD (b) removal with 2 L ozone pre-treatment reactor



Figure 3. Visual results of batik wastewater treatment with ozone pre-treatment

Based on the Pt-Co color unit the color removal efficiency in 16 L reactor around 65% in 240 minutes treatment (Fig 4a). Each port has almost the same results in color removals. Color degradation begins to experience saturation at 120 minutes, similar to the 2 L reactor. The results of 16 L reactor color removal efficiency decreased from the scale of 2 L. To improve the quality another technology was success to remove color is combined process of chemical coagulation and electrocoagulation (Ali et al., 2019).

The final results of the COD removal showed 43; 35; and 33% at an altitude of 50; 100; 150 cm (Fig 4b).

This result shows the COD removal value is high in the most oxidative area, which is at the height of 50 cm. At the height of 50 cm is an area adjacent to the diffuser where ozone and wastewater contact is quite strong. The results are shown in the wastewater that Sandu using ozone on a pilot scale was COD removal much faster in the first 9 minutes and then decreased (Sandu et al., 2004). Larger ozone doses (6.9 g O3) result in better COD removal, but also more ozone is becoming residual (Suryawan et al., 2021). Total COD removal was 61.0-77.4%, of which 25.5-40.4% was oxidized (Suryawan et al., 2021).



Figure 4. Color and COD removal with 16 L ozone pre-treatment reactor

The reaction of ozone with water is as follows:

 $O_3 + H_2O \rightarrow HO^+ + OH^ HO^+ + OH^- \rightarrow 2H_2O$  $O_3 + HO_2 \rightarrow HO + 2O_2$  $HO + HO_2 \rightarrow H_2O + O_2$ 

# **Biodegradability (BOD/COD)**

Increased biodegradability in all experiments was followed by the measurement of changes in the  $BOD_5/COD$  ratio. To apply ozone as a pretreatment for biological treatment along with oxidation so that it can be processed by increasing efficiency. The purpose of the pretreatment process is not to mineralize the compound (convert to  $CO_2$  and  $H_2O$ ), but instead, convert not biodegradable compounds into biodegradable compounds in conventional biological treatment processes.

Table 1. Results of measurement of BOD<sub>5</sub>/COD values in each experiment.

Category	BOD <sub>5</sub> /COD
Initial batik wastewater	0.143
After 2 L ozonation pre-treatment	0.49
After 16 L ozonation pre-treatment	
50 cm*	0.45
100 cm*	0.40
150 cm*	0.44

\*50 cm, 100 cm, 150 cm measured from the bottom up

The biodegradability of batik wastewater showed an increase of from about 0.143 to about 0.49 within 240 minutes for 2 L volume reactor. In upscale reactor at an altitude of 150 cm, an increase in BOD5/COD was lower than the height of 50 and 100 cm. The results of the treatment of dyes depend on the composition of the wastewater and consequently the degradation process by microorganisms. Other results obtained after ozonation, the samples became more easily decomposed where textile wastewater indicated a low  $BOD_5/COD$  ratio (0.143), and these samples have nonbiodegradable, which can be associated with toxicity and/or solubility of low compounds present in wastewater samples. After 240 minutes of ozonation, this ratio reached a value of 0.4-0.49, which could ensure a good sample of biodegradability. Study of artificial wastewater with RB5 could improve the biodegradability from 0.2-0.3 to 0.4-0.6 (Suryawan et al., 2021). Real wastewater from ethnical textile also improved 0.18 to 0.32 (Ulucan-Altuntas et al., 2018). The combination of permanganate and ozone has also increased the biodegradability of textile wastewater to 0.33-0.68 (Liang et al., 2018).

The use of ozone oxidation is following by microbiological degradation that can provide more economical and effective process condition than oxidation or single biodegradation. However, dyes and organic matter are not completely damaged in the ozonation treatment scheme as pre-treatment and the formation of intermediates and final products must be observed (Someshi et al., 2010).

### NH<sub>3</sub>-N and Total Phenol removal

Ammonia-Nitrogen (NH<sub>3</sub>-N) on a 2L scale reactor showed a low efficiency of 31.4% (Table 1). The removal of NH<sub>3</sub>-N in the 16L reactor did not have a significant difference with the 2L reactor. The highest efficiency value occurs at an altitude of 50 cm of 30.9%. Another alternative treatment is adsorption with wastepaper sludge, empty fruit bunch fibre, and flamboyant pods (*Delonix Regia*) that can reduce NH<sub>3</sub>-N of 49.3\%, 79.5% and 74.3 %, respectively (Daud et al., 2018a; Nasir et al., 2018, Daud et al., 2018b)

Table 2. Results of measurement of NH<sub>3</sub>-N and Total Phenol efficiency in each experiment

	Removal effi	moval efficiency (%)	
Reactor	Ammonia-	Tatal	
	Nitrogen	Dhonol	
	(NH3-N)	Filehol	
2L	31.4	16.4	
16L (50 cm <sup>*</sup> )	30.9	21.2	
16L 100 cm <sup>*</sup> )	24.3	3.0	
16L (150 cm*)	12.9	9.3	

\*50 cm, 100 cm, 150 cm measured from the bottom up

A larger reactor volume will result in a smaller total reduction of phenol due to the larger volume capacity of wastewater. In the biological treatment, high ammonia (NH<sub>3</sub>-N) tend to be toxic for the bacteria and biological wastewater treatment (Yuzer et al., 2012). NH<sub>3</sub>-N decrease level of free oxygen present in water cam impact aquatic life (Musa et al., 2019). High ammonia content can also cause eutrophication in water bodies (Septiariva, 2021; Afifah et al., 2020). The 2L scale reactor, total phenol removal efficiency was only 16.4% with effluent concentration of 0.51 mg/L. The 16L reactor the largest total phenol removal measurement occurred at an altitude of 50 cm of 21.2%. Phenol compounds is toxic compound that difficult to remove and mostly produced by the textile industry. Accumulation of phenol in environment released over a long period of time can toxic effects on humans and animals (Sable et al., 2018). Phenol treatment can be applied by conventional treatment with advanced treatment. Wastewater treatment with advanced treatments such as ozone applications can be applied to reduce phenol but requires high energy costs (Villegas et al., 2016).

Improving textile wastewater such as batik wastewater effluent quality, post-treatment treatment is needed. A combination of adsorbs and ozone technology was applied to textile waste treatment (Khamparia & Jaspal, 2017). For cheaper post-treatment technology is phytoremidiation. Some researchers have succeeded to treated batik and textiles wastewater using phytoremidiation technology (Safauldeen et al., 2019; Imron et al., 2019, Effendi et al., 2018; Tangahu et al., 2019). Hybrid technologies such as membranes have also been used for post-treatment textile wastewater (Bengani-Lutz et al., 2017)

# CONCLUSION

The results of color and COD removal at each reactor are quite high, but still cannot measure up the standard quality. However, the optimum increase in biodegradability is from 0.143 to 0.4-0.49. The results of 16 L reactor color removal efficiency decreased from the scale of 2 L. The final results of the COD removal showed 43; 35; and 33% at an altitude of 50; 100; 150 cm. The removal of NH3-N in the 16L reactor did not have a significant difference with the 2L reactor. The highest efficiency value occurs at an altitude of 50 cm of 30.9%. The 2L scale reactor, total phenol removal efficiency was only 16.4% with effluent concentration of 0.51 mg/L. The 16L reactor the largest total phenol removal measurement occurred at an altitude of 50 cm of 21.2%. This can make ozone technology to be used as a pre-treatment in biological wastewater treatment. This application is highly recommended for developing countries that use biological treatment as the main treatment in textile industry.

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