

## TREATMENT OF BATIK INDUSTRY WASTE WITH A COMBINATION OF ELECTROCOAGULATION AND PHOTOCATALYSIS

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### ABSTRACT

This study successfully investigated the treatment of batik industry waste with a combination of electrocoagulation and photocatalysis. The use of an aluminum plate as an anode, a stainless steel 316 plate as a cathode, and titanium dioxide (TiO<sub>2</sub>) coated on aluminum plates, the main materials in this method, resulted in the simultaneous decolorization of Remazol Red dye, elimination of 2,4,6-trichlorophenol, and reduction of hexavalent chromium (Cr[VI]). For the TiO<sub>2</sub> coating, a TiO<sub>2</sub> suspension was applied to a sanded aluminum plate. Optimal performance of the combination of electrocoagulation and photocatalysis was achieved under operating conditions of 15 V DC electric voltage, an initial pH value of 7, and the use of an aeration flow. Under the operating conditions, 600 ml batik waste containing 5 mg/L 2,4,6-trichlorophenol, 3 mg/L Cr(VI), and 390 PtCo Remazol Red dye were eliminated simultaneously within 4 hours with conversions of approximately 58%, 100%, and 97%, respectively.

*Keywords:* 2,4,6-trichlorophenol; Cr(VI); Electrocoagulation; Photocatalysis; Remazol red

### 1. INTRODUCTION

Because of the philosophy behind and meaning associated with Indonesian batik, this tradition is considered an intangible cultural heritage of humanity. Since this designation by the United Nations Educational, Scientific and Cultural Organization (UNESCO), Indonesian batik has been commanding high prices around the world. Along with the increasing popularity of Indonesian batik has been a significant growth in the number and types of batik industries: from small-scale home industries to large-scale factories. This has had a negative effect on the environment. Many of the rivers near batik factories are polluted from the disposal of improperly processed industrial batik waste (Zaenuri, 2014).

Yogyakarta and Pekalongan are examples of cities that are well known for their batik industries. In an analysis of the liquid waste from the Batik Indah Yogyakarta industry, Saptarini (2009) found color waste in the amount of 324 PtCo, which was in excess of the color quality standard limit (50 PtCo, KepMen LH No.51 MENLH 1995). Although dyes do not directly have toxic effects, their by products can be categorized as dangerous substances (Brosillon et al., 2005). The total phenol content in the Brengi River in Pekalongan was found to be 0.031 mg/L

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(Yustiara et al., 2014), and the total chromium (Cr) was found to be 0.09 mg/L (Wijayanti et al., 2014). Phenol itself is a toxic carcinogenic compound that is very harmful to humans (Wong et al., 2011). The concentrations of heavy metals such as Cr(VI), although small, should be of concern. Aquatic organisms can absorb some of the compounds in water, and these compounds can accumulate in their bodies in as much as 100 or 1.000 times higher than the moisture content found in water (Cowen & Bruland, 1985).

Because of its diversity (color, phenolic compounds, and Cr metals), batik waste needs to be treated through a simultaneous processing method for its. This study proposed the use of a combination of electrocoagulation and photocatalysis for the simultaneous treatment of batik waste. Electrocoagulation is a continuous coagulation process that uses direct current through electrochemical events and electrolyte decomposition, with one of the electrodes being aluminum or iron (Mkpenie & Abakedi, 2015). Photocatalysis is used for degrading organic compounds, e.g., phenolic compounds and hexavalent chromium (Cr[VI]), and reducing the content of the metal ions in batik waste simultaneously with the help of ultraviolet (UV) light. Titanium dioxide (TiO<sub>2</sub>) in the form of nanoparticles is a commonly used semiconductor in photocatalysis (Slamet et al., 2007).

Electrocoagulation enables the degradation of various types of waste, such as dyes, organic compounds, and metals. However, a great deal of electrical energy is required for processing a large amount of pollutant in a short amount of time. This will certainly affect the cost of processing. Like electrocoagulation, photocatalysis has been analyzed for its ability to degrade various types of waste. That electrocoagulation requires only UV rays has attracted a great deal of attention because of the potential for lower processing costs (Jawad et al., 2016). However, a problem with photocatalysis is its ability to process waste in shaded conditions. In such a situation, the UV rays are unable to penetrate the waste to trigger photocatalytic materials (semiconductor materials such as titanium dioxide [TiO<sub>2</sub>]). Given the advantages and disadvantages of each process, the combination of electrocoagulation and photocatalysis has the potential for achieving the cost-effective simultaneous degradation of waste containing dyes, organic compounds, and metals in a timely manner.

The combination of electrocoagulation and photocatalysis for the simultaneous treatment of plural types of waste (dyes, organic compounds, and metals) has been rarely studied. Jati and Aviandharie (2015) performed a combination of electrocoagulation and photocatalysis to reduce Cr(VI) metal waste. Electrocoagulation was performed first; then photocatalysis was done in different containers. Escobar et al. (2016) optimized total organic carbon (TOC) removal for the treatment of lithographic wastewater by a process of electrocoagulation followed by photocatalytic. In the current study, the simultaneous processing of batik waste containing dyes, organic compounds, and metals was achieved through a combination of electrocoagulation and photocatalysis. The study thus provides an effective, environmentally friendly, and relatively low-cost method for processing batik industry plural waste.

## 2. EXPERIMENTAL METHODS

### 2.1. Preparation of Aluminum Plates Coated with Titanium Dioxide Suspension

The size of the aluminum plate used as the suspension medium for the TiO<sub>2</sub> was 13 cm × 5 cm × 0,1 cm. The aluminum plate was sanded with 80 CW sandpaper, a somewhat rough grade, to achieve good results. It was immersed in a glass beaker of water (H<sub>2</sub>O). The soaked plate was then sonicated for 15 minutes at room temperature. Next, the plate was removed from the beaker and dried with a hair dryer.

Evonic Industries AG was the source for the TiO<sub>2</sub> P25 (21% rutile and 79% anatase). The suspension was made with 2.5 g TiO<sub>2</sub> dispersed in 500 mL H<sub>2</sub>O. The suspension was stirred

with a magnetic stirrer for  $\pm 30$  minutes. Next, 1.25 mL tetraethyl orthosilicate (TEOS) was added to the suspension. The pH value of the suspension was then adjusted by adding drops of 10 M 65% nitric acid ( $\text{HNO}_3$ ) (supplied by Merck) until the pH value of the suspension reached 2–3. Finally, the suspension was sonicated for 15 minutes.

A brush was used to coat the sanded aluminum plate with the  $\text{TiO}_2$  suspension. The coating was done until a thick layer of  $\text{TiO}_2$  was formed. The final product was then dried in an oven for 30 minutes at 100–110°C.

## 2.2. Electrocoagulation and Photocatalysis

Electrocoagulation and photocatalytic were tested for their effectiveness in the simultaneous treatment of the synthetic waste from Remazol Red dyes (color), 2,4,6-trichlorophenol (TCP), and Cr(VI). The waste contained 390 PtCo color, 5 mg/L TCP, and 3 mg/L Cr(VI), which was dissolved in 600 mL demineralized  $\text{H}_2\text{O}$ . The Pyrex batch reactor had a capacity of  $\pm 1000$  mL. For the activity test, two aluminum anodes and one stainless steel 316 cathode were used. The standard operating conditions for this activity test were the use of 15 V electric current from DC Power Supply, air flow from an aerator, a UV lamp 4W Goldstar, initial pH value of 7, and 4 hours reaction time. Figure 1 shows the scheme for the electrocoagulation and photocatalysis reactor. The changes in color and TCP concentrations were analyzed with a UV-Vis Spectroquant Pharo 300 spectrophotometer, and the changes in the Cr(VI) concentration were analyzed with a Spectro DR 5000 UV-Vis spectrophotometer.

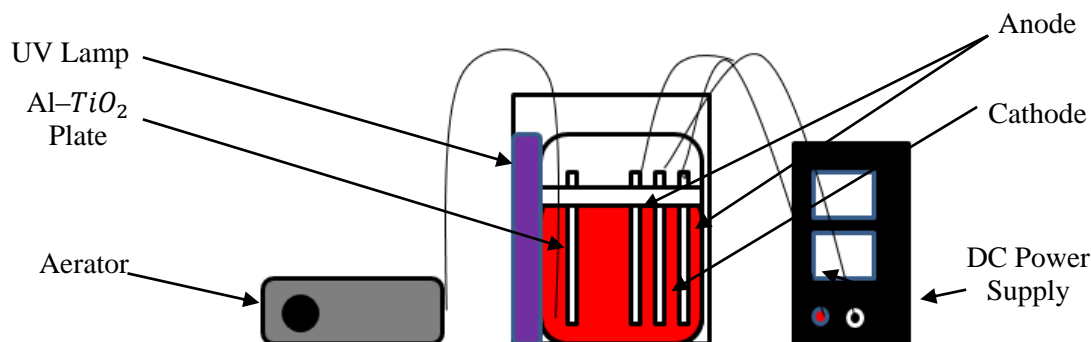


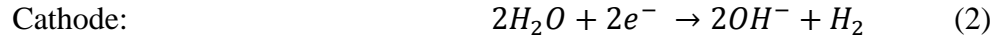
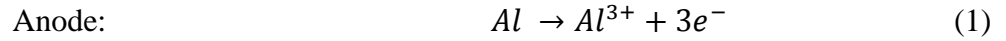
Figure 1 Electrocoagulation and photocatalysis reactor scheme

## 3. RESULTS AND DISCUSSION

### 3.1. Effects of Electrocoagulation and Photocatalysis on Color Concentration

The results of the effects of the combination of electrocoagulation and photocatalysis on plural waste, especially color concentration, can be seen in Figure 2. Figures 2 and 3 illustrate the superior results with the combination of electrocoagulation and photocatalysis (96.93%) over just electrocoagulation (90.27%) for color removal. The reason is that photocatalysis is a process that can also decolorize dyes (Mukhlis et al., 2013). When the electrocoagulation process began, several reactions occurred simultaneously. The metal ions ( $\text{Al}^{3+}$ ) were released from the anode into the  $\text{H}_2\text{O}$ . At the cathode surface, the  $\text{H}_2\text{O}$  was hydrolyzed to hydrogen gas ( $\text{H}_2$ ) and hydroxyl ion ( $\text{OH}^-$ ). Meanwhile, free-flowing electrons from the anodes destabilized the charged surfaces on the dissolved solids (dyes) and oils emulsified. Flocs containing aluminum hydroxide ( $\text{Al}[\text{OH}]_3$ ) coagulant, dissolved solids (dyes), heavy metals, and other contaminants formed.

In an electrochemical system with aluminum anodes, the possible electrode reactions are as follows:



The forming of the  $Al(OH)_3$  coagulant:

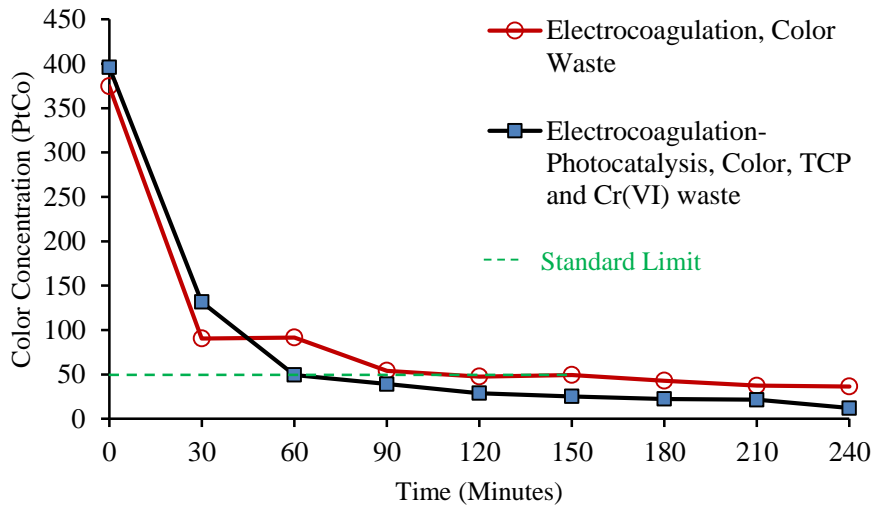
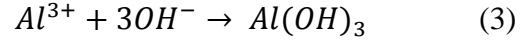


Figure 2 Profile of color concentrations under various conditions over time

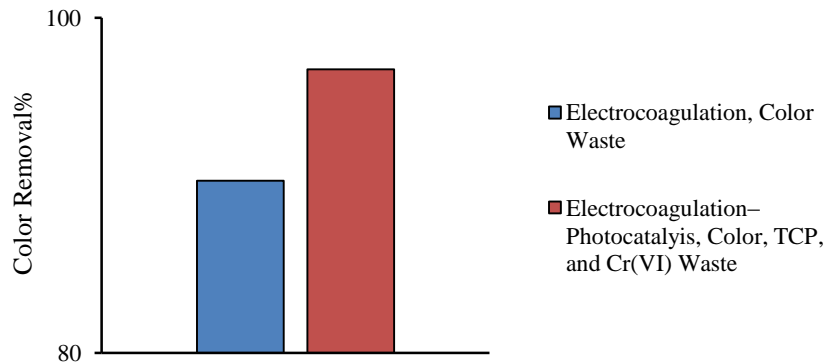
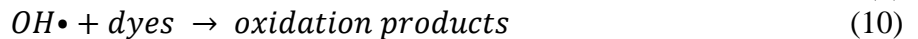


Figure 3 Comparison of color removal performance under various conditions (t = 240 minutes)

In this test, dyes were also degraded with hydroxyl ions produced from photocatalysis (Jawad et al., 2015) for which the reaction was as follows:



### 3.2. Effects of Electrocoagulation and Photocatalysis on Total Organic Carbon Concentrations

The results of the effects of the combination of electrocoagulation and photocatalysis on plural waste, especially TCP concentrations, can be seen in Figure 4 below.

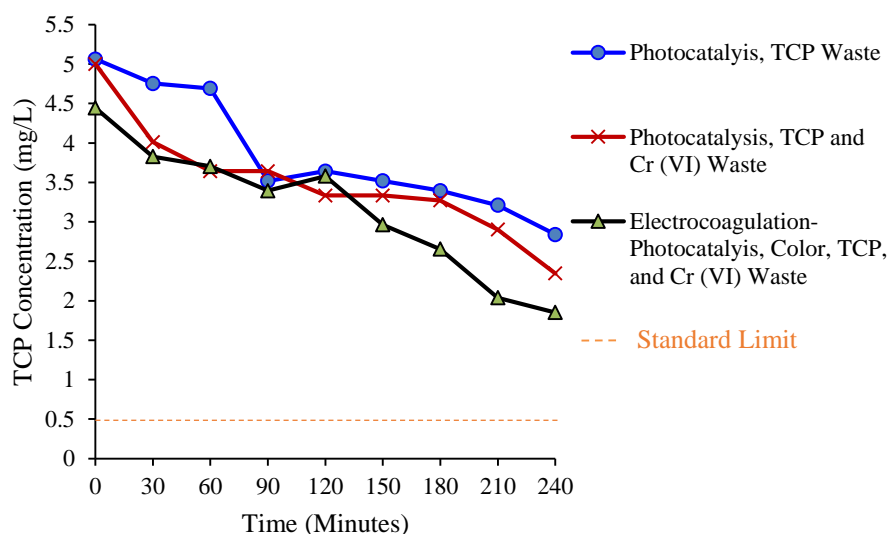


Figure 4 Profile of total organic carbon concentrations under various conditions over time

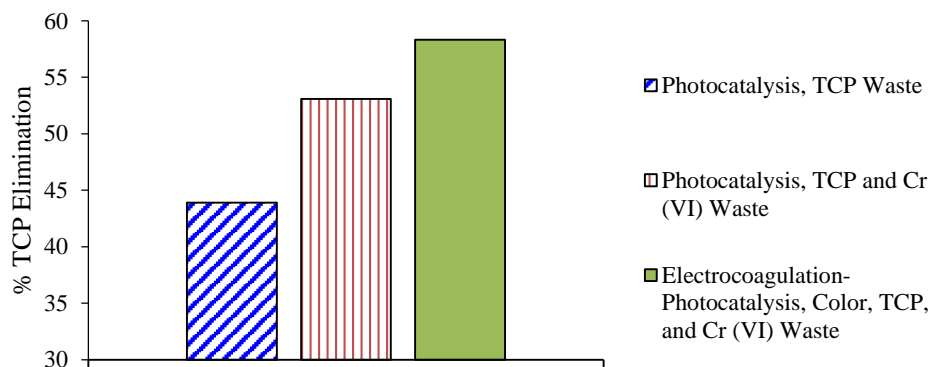


Figure 5 Comparison of total organic carbon elimination performance under various conditions (t = 240 minutes)

During photocatalysis, organic compounds, such as TCP, were degraded by the hydroxyl ions produced from the process. The reactions were the same with Equations 4–10. The only difference was the type of pollutant. However, in some cases, the photocatalysis could not be performed optimally because of recombination (Heltina et al., 2015); consequently, the reactions in the Equation 4 hole ( $h^+$ ) and the electron ( $e^-$ ) were not formed maximally. This usually occurs because of the inactivity of the  $e^-$ , thus facilitating recombination. Figures 4 and 5 illustrate the higher elimination rate (53.09%) with the simultaneous degradation of TCP and Cr(VI) (plural) waste over the rate (43.90%) with the degradation of TCP waste only. The reason is that the electrons were used to reduce the metal ions Cr(VI). However, with a combination of electrocoagulation and photocatalysis, a higher elimination rate (58.33%) of TCP was achieved because the formed  $Al(OH)_3$  coagulant also eliminated the TCP through electrocoagulation (Mohammed, 2007).

The aeration rates were chosen to improve the performance of the photocatalysis. A consideration with the aeration treatment was the possibility of contact between the TCP and the TiO<sub>2</sub>-coated aluminum plate because of the turbulence created by the effect of the agitation (Lam et al., 2009). The aeration treatment also increased the amount of dissolved oxygen in the waste sample. The oxygen took on the role of electron scavenger so that the recombination rate in the photocatalysis could be reduced. The phenomenon also caused an increase in the OH groups ( $OH\bullet$ ) that resulted from the reaction process between the oxygen and the electrons (Nawawi et al., 2017). As the number of OH groups ( $OH\bullet$ ) increased, the number of pollutants that could be attacked by the OH group ( $OH\bullet$ ) increased. The mechanism of TCP degradation by photocatalysis was predicted to be the same as that proposed by Huanhuan et al. (2013)

### 3.3. Effect of Electrocoagulation and Photocatalysis on Hexavalent Chromium Concentrations

The results of the effects of electrocoagulation and photocatalysis combination on plural waste, especially the Cr(VI) concentration, can be seen in Figure 6 below.

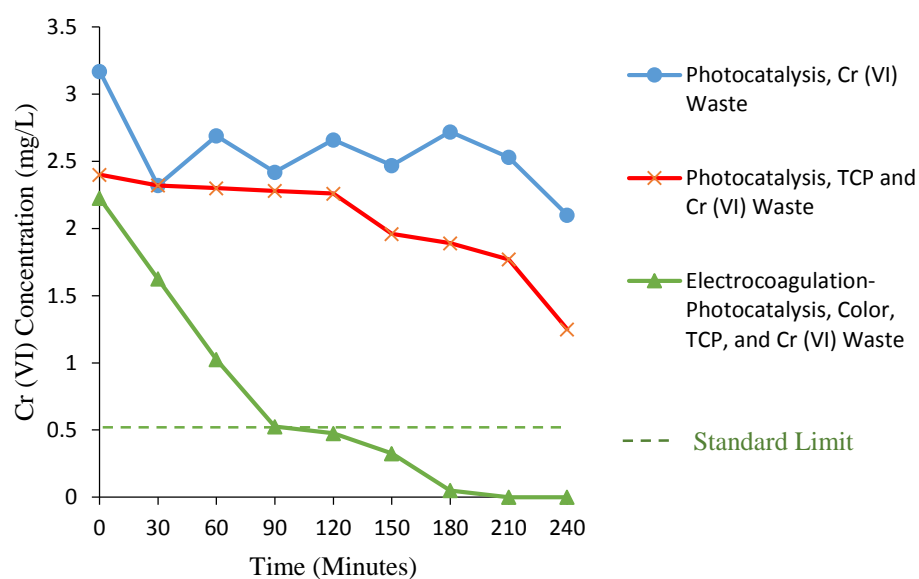


Figure 6 Profile of Hexavalent Chromium concentration under various conditions over time

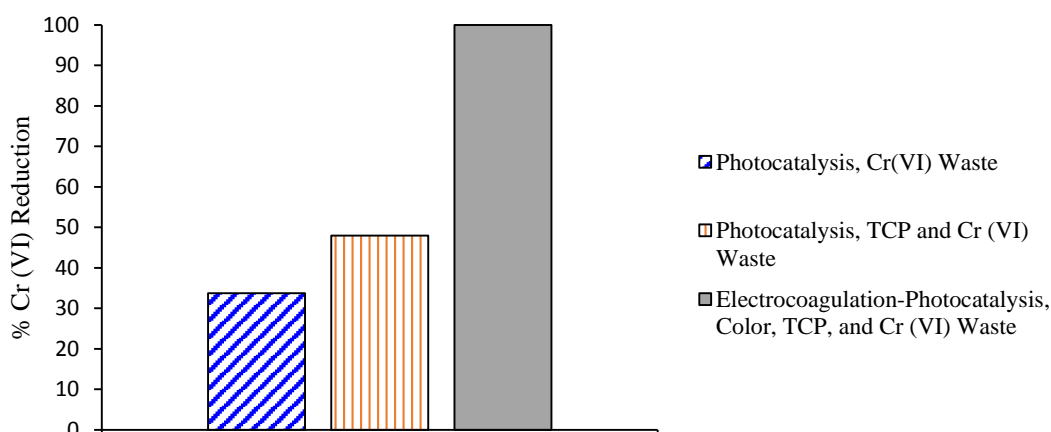
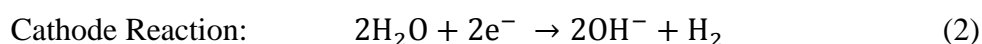
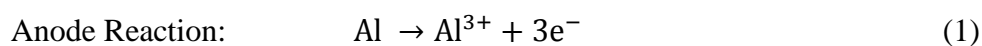
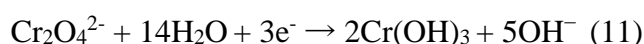


Figure 7 Comparison of Hexavalent Chromium reduction performance under various conditions (t = 240 minutes)

Figures 6 and 7 illustrate the similarities between the Cr(VI) reduction profile and the TCP elimination profile. The elimination rate (47.92%) with simultaneous TCP and Cr(VI) (plural) waste degradation was higher than the rate (33.75%) with TCP waste degradation only. The difference in this case was the improved Cr(VI) reduction, 100%, achieved through the combination of photocatalysis and electrocoagulation. The reason is that although the electron ( $e^-$ ) formed from the anodes generally reacts with  $H_2O$  to form  $H_2$ , it tended to react with the Cr(VI) in this study, thus reducing the metal ion to Cr(III)



Cr(VI) reduction reaction occurring at neutral pH:



## 5. CONCLUSION

The simultaneous processing of mixed waste containing 2,4,6-trichlorophenol, Cr(VI), and Remazol Red dye was achieved in this study. The use of an aluminum anode, a stainless steel 316 cathode, and  $\text{TiO}_2$  coated on an aluminum plate as a photocatalyst under operating conditions of 15 V DC electric voltage, initial pH value of 7, and the use of aeration flow was found to be optimal for the process. Under these operating conditions, 600 ml batik waste containing 5 mg/L 2,4,6-trichlorophenol, 3 mg/L Cr(VI), and 390 PtCo Remazol Red dye was eliminated simultaneously within 4 hours with conversions of approximately 58%, 100%, and 97%, respectively.

## 6. ACKNOWLEDGEMENT

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