Application of continuous system electrocoagulation method for textile industry wastewater treatment

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Abstract. Wastewater of textile industry containing any pollutants, turbid and strong color appearance. Electrocoagulation (EC) is a wastewater treatment alternative, which are some factors: distance of electrode, voltage application, and time operation. This research was determined the optimum voltage density and flow rate by continuous system EC. The couples of Al electrode was immersing in the EC chamber by various distance: 1,5; 3.0; 4.5; 6 cm, and applied 18 volts. The wastewater was passed to the EC chamber by ascending in various flow rate: 1,0; 1.5; 2.0; 2.5 mL s⁻¹. The product was evaluated according to the regulations PermenLH RI no 5/2014, including: COD, TSS, pH, DO, Conductivity, and Al concentration. The result optimum voltage density application is 12 Vcm⁻¹ (18 volts /1.5 cm), and a flow rate 1 mL s⁻¹. COD Removal is 91.80% (1.683 to 138 mgL⁻¹; fulfil the regulations), TSS removal is 87.83% (937 to 114 mg L⁻¹, (not fulfil the regulations), color degradation is 83.98% (239, 14 to 38,31 mgL⁻¹PtCo). The others are pH increase in the range of tolerant, DO increase up to 8,2 mg L⁻¹, Conductivity increase up to 5350 μs cm⁻¹, and Al increase up to 2,71 mgL⁻¹.

1. Introduction
Processes in the textile industry involve a variety of chemicals such as dyes, napto, sodium hydroxide and other materials. The remaining chemicals can become a source of contaminants and are disposed of with the wastewater stream. The quality of textile industrial wastewater is COD 1100 mgL⁻¹, color 1500 mg L⁻¹ Pt-Co, and pH 8.8 [1]. The standard parameters of textile wastewater according to Indonesian government regulations are COD 150 mgL⁻¹, TSS 50 mgL⁻¹ and pH 6-9 [2].

The practice of textile wastewater treatment in industry generally uses chemical coagulation and flocculation methods, or in combination with activated sludge treatment methods. This method requires a large area of land for a long time and new problems arise, namely the emergence of a lot of sludge. Therefore we need a better processing method. One of the promising alternative methods of wastewater treatment is the electrocoagulation method. Electrocoagulation (EC) method is more efficient and inexpensive to treat wastewater, the equipment is simple and easy to operate and does not require additional chemicals, besides that does not require a large space [3].

Wastewater treatment methods the EC uses a coagulant generated electrolysis. Electrolysis is carried out in a chamber containing waste water, and one or more pairs of metal sheets (plates) called electrodes (cathode and anode) are dipped into it, then electrified. Iron plate, aluminum plate, titanium plate,
stainless steel can be used as electrodes [4]. If an aluminum (Al) electrode is used, Al oxidation will occur at the anode, which will form a coagulant in the form of Al\(^{3+}\) ions based on the reaction:

\[
\text{At the anode: } \text{Al (s)} \rightarrow \text{Al}^{3+} (\text{aq}) + 3\text{e}^- \quad (\text{Al oxidation})
\]

\[
\text{At the cathode: } 3\text{H}_2\text{O} + 3\text{e}^- \rightarrow 3/2\text{H}_2 + 3\text{OH}^- \quad (\text{Water reduction})
\]

The Al\(^{3+}\) ion then undergoes hydrolysis to form [Al(H\(_2\)O)\(_5\)OH\(^+\)]; [Al\(_2\)(OH)\(_4\)]\(^{4+}\); [Al (OH)\(_4\)]\(^-\); [Al\(_6\)(OH)\(_{15}\)]\(^{3+}\], and other polyaluminum ions [5]. These ions will neutralize the pollutant charge in the wastewater and form aggregates. Aggregate will be pushed up and float due to the encouragement of hydrogen gas that is formed.

The EC batch system is a flowless process. Meanwhile, a continuous system is an EC process in which wastewater flows [6]. The EC batch system process is carried out in an electrolysis chamber in which there are two direct electric current conductors, namely the electrodes, which are immersed in a to stationary electrolyte solution, while the continuous system of waste water is flowed continuously into the chamber contained in the electric conducting electrode[7].

By applying 10 volts, and 2 cm electrode distance, electrolysis time of 25 minutes in municipal wastewater treatment can reduce COD by 80.70% and reduce TSS by 60.38% [8]. The EC method can also reduce COD up to 95% in electrolysis for 4 hours, and can also reduce the metal content up to 99% after 30 minutes by applying a current density of 102 Am\(^-2\) [9]. Both of them are EC batch system. In this research, EC textile water was done in continuous system, by applied 18 volt and flow rate variation from 1-2.5 ms\(^{-1}\).

2. Material and Method

2.1. Materials and Tools

The materials used include demine water, hydrochloric acid (HCl) (1: 1), digestion solution (K\(_2\)Cr\(_2\)O\(_7\)) 0.1 N, acetylene gas, nitrous oxide gas (N\(_2\)O), standard solution of KHP, buffer solution pH 10.01; buffer solution pH 4.01; buffer solution 7.00; 0.01 M KCl standard solution), concentrated Ag\(_2\)SO\(_4\) and H\(_2\)SO\(_4\),aluminum standard 1000 mgL\(^{-1}\) solution and textile industry wastewater samples.

The equipment used is a waste reservoir, an EC chamber, a heater, a timer, a glass device, a filter with a pore size of 0.45 µm (equipped with a filter holder and pump), a glass funnel, a DC Power Supply, desiccator, pH meter, conductometer, DO, COD reactor, oven, analytical balance, crocodile clamp, vacuum equipment, pH meter, 15 x 15 cm aluminum plate, and AAS.

2.2. Sample and analysis

The sample used in this research is textile industrial wastewater in Cibitung, Bekasi, West Java. The textile industrial wastewater treatment process is carried out by the continuous system electrocoagulation method at various electrode distances and the waste flow rate with the application of a fixed electric potential of 18 volts. Before and after wastewater treatment, the wastewater was tested including: pH, DO, DHL, COD, TSS, and color analysis and dissolved Al levels.

Measurement of pH SNI 06-6989.11-2004 and DO by electrometric method, COD analysis was carried out by closed reflux method refers to SNI 6989.2: 2009, DHL measurement method SNI 06-6989.1-2004 SNI 06-6989.1-2004, TSS by the gravimetric method SNI 06-6989.3-2004, color analysis refers to SNI.6989.80: 201. Analysis of Al using AAS method refers to SNI 06-6989.34-2005.

2.3. EC experiment tool setup

The experimental equipment consists of: a wastewater reservoir, EC chamber, Al plate as an electrode, direct current (DC) power supply, and a reservoir for treated water, as well as current connector cables. The EC chamber is 20 cm x 15 cm x 15 cm made of plastic, and is partitioned into 3 compartments. The first and the third compartment has measuring 3 cm x 15 cm x 15 cm. The first component is used for trapping oil / grease, and the third is used for deposition. The second compartment measures: 10 cm x 14 cm x 12 cm or the volume is 1680 mL (effective volume) for electrolysis. Within this compartment
are given 8 slots 1.5 cm apart for attaching the electrode plate. The inlet channel is located at the bottom of the first compartment and the outlet channel is located at the top. So that the wastewater travels in the electrolysis chamber from the bottom up (ascending). In the wastewater reservoir, a water tap is installed to regulate the flow rate of treated wastewater. DC Power supply applied as a constant 18 volt. Figure 1 shows the continuous system EC experimental apparatus setting.

2.4. Experimental variable setting
The independent variables carried out in this EC process include: electric voltage, electrolysis contact time and electrode distance. The electric voltage applied is constant 18 volts. The wastewater is flowed into the chamber with a rate variation of 1; 1.5; 2.0; 2.5 ml s\(^{-1}\). The variation of the electrode distance applied was 1.5; 3.0; 4.5 and 6.0 cm, each treatment combination was repeated twice so that a total of 32 times running experiments.

![Figure 1. Experimental Setting Apparatus.](image)

2.5. Data processing
The data of each parameter in each experiment is plot versus the flow rate, then the trend is studied and the effectiveness of the treatment is evaluated based on the parameters of the textile wastewater standard PermenLH No. 5/2014 Annex XLII regarding the textile industry wastewater quality standards. Pollutant removal efficiency, especially for COD, TSS, and color parameters is calculated based on the formula:

\[
E_{\text{fisien}} \text{ Penyisihan polutan } x = \frac{[X]_0 - [X]_t}{[X]_0} \times 100 \%
\]

\([x]_0 = \text{concentration of pollutant before EC, and [x]_t = concentration of pollutants after EC}\)

3. Result and Discussion

3.1. Characteristics of Textile Wastewater
The characteristics of the textile wastewater sample analysis results can be seen in full in Table 1. The results of the measurement of wastewater obtained an average pH of 6.91 indicates that textile wastewater is neutral and is within the permissible minimum and maximum pH limits. Textile wastewater can be acidic if the cloth immersion process to remove the starch layer uses H\(_2\)SO\(_4\) or HCl; or alkaline when immersed using NaOH solution[10].

DO was measured of 7.6 mgL\(^{-1}\), which is quite high, indicating that the activity of decomposing microbes in wastewater is quite low. The initial value of DHL in wastewas very high at 5030 µs cm\(^{-1}\). In comparison, the conductivity of river water is generally between 200-1000 µs cm\(^{-1}\), and saline water has a conductivity between 1000 - 10,000 µs cm\(^{-1}\) [11]. Thus, wastewater samples are included in the saline water category. The high conductivity of textile wastewater is due to the large number of dissolved salts. The textile industry process requires chemicals at the immersion, cooking and bleaching
stages during the dyeing stage. At this stage, a strong acid or base is needed, chlorine, non-chlorine oxidizing agents in the form of dissolved salts [10].

| No. | Parameter | Unit     | Results | Quality standards [3] |
|-----|-----------|----------|---------|-----------------------|
| 1   | pH        | -        | 6.91    | 6.0-9.0               |
| 2   | DO        | mgL⁻¹    | 7.60    | -                     |
| 3   | DHL       | µs cm⁻¹  | 5030    | -                     |
| 4   | COD       | mgL⁻¹    | 1683    | 150.0                 |
| 5   | TSS       | mgL⁻¹    | 937     | 50.0                  |
| 6   | Al³⁺      | mg L⁻¹   | 0.02    | -                     |
| 7   | Color     | mgL⁻¹ PtCo | 239.14  | -                     |

The COD level was 1683 mgL⁻¹. COD levels are very high, exceeding environmental quality standards (150 mgL⁻¹). COD represents the presence of organic and inorganic pollutants which can be oxidized by K₂Cr₂O₇. The wax coating on the fabric, residual dyes, hypochlorite can cause high COD.

The TSS measurement results were quite high, namely 937 mgL⁻¹, One of the materials involved in the manufacture of cloth is starch (amylum), and dye [10], both of which are water-soluble but non-ionizing organic substances. Textile wastewater sample had a color of 239.14 PtCo, meaning that the textile wastewater appeared to be dark. According to Mastika [12] water naturally has a color scale of <50 PtCo, peat groundwater has a brownish color between 38 – 701 mgL⁻¹ PtCo [13]. The color density of textile wastewater is caused by the content of organic pollutants, especially textile dyes, and possibly chromium metal ions. and iron [10].

3.2. Voltage density and residence time of wastewater
The amount of electricity that affects the EC process is the electric current, as in electrolysis, where the amount of substance produced by electrolysis is directly proportional to the current strength. However, electric current cannot be forced to flow with a certain strength, because the strength of the electric current to flow is greatly influenced by the characteristics of wastewater.

The quantity of electricity that is easily determined by the EC experiment is the electric potential. Therefore an important parameter of EC performance can be determined by constructing the distance between the electrodes and applying a certain electric voltage. The application of a certain voltage and electrode distance can be expressed in a universally understandable term, namely voltage density. In addition to the voltage density, the success of EC is determined by time. In a continuous system, the time required for wastewater to be in the electrolysis chamber is determined by the flow rate and chamber volume. The general term associated with these two variables is the residence time or length of time the wastewater is in contact with the electrodes, which is known as "hydraulic retention time" and is abbreviated as HRT. Voltage density and HRT using the following formula:

\[ \text{Voltage density} = \frac{\text{Electric potential (volt)}}{\text{Electrode distance (cm)}} \]

\[ \text{HRT} = \frac{\text{Volume of EC chamber (mL)}}{\text{Flow rate (mL s}^{-1})} \]

Voltage density and HRT in the EC experiment in this study are summarized in Table 2.

| No. | Electrode distance (cm) | Voltage density (volts cm⁻¹) | Flow rate (mL s⁻¹) | HRT (seconds) | HRT (minute) |
|-----|-------------------------|----------------------------|-------------------|--------------|--------------|
| 1   | 1.5                     | 12                         | 1                 | 1680         | 28           |
| 2   | 3.0                     | 6                          | 1.5               | 1120         | 19           |
| 3   | 4.5                     | 4                          | 2.0               | 840          | 14           |
| 4   | 6.0                     | 3                          | 2.5               | 672          | 11           |

Note: Constant voltage 18 volts and effective volume chamber 1680 mL.
3.3. Results of Electrocoagulation Experiments

3.3.1. Relationship Between Flow Rate (HRT) and Changes in pH. During the EC process, the pH value has increased, but is still within the required pH quality limits of textile wastewater, namely 6.0-9.0 [2]. The trend of increasing the highest eluent pH was in the experiment with the highest voltage density, namely 12 Vcm$^{-1}$. This is due to the high voltage density the electric current that flows is also high so that the electrolysis of water forms more hydroxide ions. The increase in pH in various experiments is still within the specified limits. This is in accordance with Gupta that during the EC process there is a slight increase in pH due to the formation of OH$^-$ and H$_2$ gas [13].

3.3.2. Effect of flow rate (HRT) on DO at various voltage densities. Dissolved oxygen (DO) increases during EC. The highest increase occurred in the experiment with a voltage density of 12 volts cm$^{-1}$, from 7.55 mgL$^{-1}$ to 8.31 mg L$^{-1}$. The increase in DO can be caused by the oxidation of water near the anode based on the reaction: 2H$_2$O $\rightarrow$ O$_2$ + 4H$^+$ + 4e$. This is in accordance with the opinion of Bazzafshan et al. [14] that it is possible that water oxidation occurs at the secondary electrode. Water oxidation competes with aluminum electrode oxidation so that it is not as strong as water reduction at the cathode which produces H$_2$ gas and OH$^-$ ions. The EC process causes the movement of the water mass which makes it easier for dissolved oxygen. Figure 3 shows the relationship between DO and HRT levels in various voltage density experiments.

3.3.3. Effect of Flow Rate (HRT) on COD at various Voltage Densities. The COD decreases with the length of stay in the EC chamber. Figure 2A shows the pattern of COD reduction in HRT. The longer the HRT COD decreased, however, until the HRT of 30 minutes only one treatment could reach the quality standard (150 mgL$^{-1}$), namely the experiment with a density of 12 Vcm$^{-1}$. Figure 2B shows the difference in the amount of COD removal. The smaller of flow rate, the greater HRT and the longer the EC contact time so that more deposits occur. According to Faraday’s law, namely the amount of substance obtained from the electrode is directly proportional to the amount of current flowing in the electrolysis chamber.

The decrease in COD concentration in the EC was due to the coagulation of waste organic pollutants in the EC chamber. Based on the double layer theory, the reduction in COD is due to the floc formed by organic pollutant ions binds to positive coagulant ions. The positively charged coagulant will absorb waste negative ions such as nitrite, phosphate, and other organic compounds then form a floc which helps the COD reduction process [15]. The hydrogen gas generated at the cathode helps contaminants float or lift, so they separate from the water. This causes the reduction of dissolved organic or dissolved materials including Al(OH)$_3$ floc which binds organic waste and captures organic pollutants [16]. In textile waste, common organic pollutants are dissolved starch and dyes.

The change in percent COD removal is greater with the greater the voltage density applied Figure 2B. The maximum COD removal at HRT 30 minutes and the voltage density of 12 Vcm$^{-1}$ reached 91.76%, from 1683 mg L$^{-1}$ to 138 mgL$^{-1}$, these results have met the quality standard. Another study applying 18 kV with the aid of an oxygen flow of 0.5 L min$^{-1}$ on textile waste only reduced COD by 76.50% [17]. According to Hariyono, the best textile wastewater treatment at 12 volts and a contact time of 60 minutes can reduce COD by up to 88.9% [1]. According to Setianingrum et al. applying a voltage of 10 V and an electrode distance of 2 cm (voltage density 5 Vcm$^{-1}$) can reduce COD from 428 mgL$^{-1}$ to 54 mgL$^{-1}$ (87.38%) [18]. Reducing COD up to 97.9%, using high electric currents [19]. A high concentration of initial pollutants will result in a high percentage of removal of pollutants. At the initial concentration the reaction rate is very fast, resulting in a large percentage of pollutant removal, and at the initial low concentration of pollutants, the reaction rate is lower and only results in a small percent removal of pollutants. According to the kinetics theory the rate of degradation is directly proportional to the concentration; the higher the concentration, the higher rate. The pollutant concentration will be decrease exponentially[20].

3.3.4. Effect of Flow Rate (HRT) on TSS at Various Current Densities. Textile wastewater prior to EC processing has a TSS value of 937 mgL$^{-1}$. Figure 3A shows the pattern of changes in TSS due to differences in HRT for various voltage density applications. From the graph, it can be seen that the smaller the flow rate (the greater the HRT), the greater the decrease in the TSS value. This is because
the greater the contact time, the greater the potential for decomposition of the Al electrode to release Al\(^{3+}\) so that the Al(OH)\(_3\) floc is also greater.

The results of processing textile wastewater with EC were quite optimal in reducing the TSS parameter from 937 mgL\(^{-1}\) became 114 mgL\(^{-1}\). However, after going through the EC process, none of the TSS values met the maximum quality standard of 50 mgL\(^{-1}\) [2]. Figure 3B shows the percent change in TSS removal. The best TSS removal at a voltage density of 12 Vcm\(^{-1}\) with a waste flow rate of 1 mL s\(^{-1}\), HRT 30 minutes. The maximum allowance was 87.72% with a final TSS value of 475 mgL\(^{-1}\) [17]. The difference in the application of the voltage density and time gives a difference in the TSS removal results, this is due to the difference in the amount of electric current that flows.

3.3.5. Increased Water Conductivity EC Results. The conductivity of the EC treated water increases with a steady increasing pattern. In textile wastewater before processing it has a high electrical conductivity value of 5030 µs cm\(^{-1}\). The higher the voltage density and HRT, the higher the conductivity value will be. The greatest change in conductivity was in the experiment with a voltage density of 12 Vcm\(^{-1}\) with an HRT of 30 minutes, reaching 5360 µs cm\(^{-1}\). The pattern of change is the same for the difference in voltage density but with a different quantity. The increase in conductivity is a consequence of dissolving the aluminum electrode into the water which does not coagulate. Another possibility is that textile waste contains dyestuffs with an azochromophore group and an auxochrome group. The auxochrome groups are –NO\(_2\), -NH\(_2\), -COOH, -SO\(_3\)H and –OH which are soluble in water are polar [20]. The auxochrome groups can be released during EC to become ions which contribute to the increase in conductivity.
3.3.6. **Dissolved aluminum content.** Faraday's law states that the number of grams of mass equivalent of a substance attached to, released, dissolved, or reacted at an electrode is equal to the Faraday amount of electric charge transferred through the electrolyte. The denser the electrode spacing or the higher of voltage density and the smaller the flow rate (higher of HRT), the greater the dissolved Al content. The initial dissolved Al level was 0.02 mg L\(^{-1}\), increasing to 2.71 mg L\(^{-1}\). The highest increase in dissolved Al levels was at a density distance of 1.5 cm and the smallest flow rate was 1 ml s\(^{-1}\) or HRT 30 minutes.

3.3.7. **Effect of Flow Rate (HRT) on Color Removal.** Figure 4A shows a color reduction pattern on the PtCo scale, and 4B shows a graph of the difference in the percentage of color reduction. The closer the electrode spacing or the greater the voltage density and the smaller the flow rate (greater HRT), the color value has the greatest decrease.

The best treatment is the voltage density of 12 V cm\(^{-1}\) with HRT of 30 minutes resulting in a color reduction from 305.5 to 48.5 on the PtCo scale (84.26%). This result is lower than Haryono et al (2018) [1] which was able to reduce the color by up to 93.3% (from 1500 to 100 mg L\(^{-1}\) PtCo), on the application of 12 volts and time of 60 minutes. This is due to different treatment conditions, and different initial color concentrations. Applying 10 volts of electricity and 2 cm of electrode distance (density 5 volts cm\(^{-1}\)) for 60 minutes can reduce the color from 2733 to 75.5 PtCo (97.24%) [18].

The color reduction in EC can be caused by two things, namely the adsorption process followed by coagulation, and the damage to the chemical structure of the dye. Adsorption occurs due to electrostatic and van der Waals forces which cause the dye to leave the waste solution and join the surface of the solid (coagulant). This is evidenced by the color of the solid that floats apart from the solution. The second possibility is the damage to the chemical structure of the dye due to EC. Textile dyes are organic compounds containing chromophore groups and auxochrome groups that determine color. The main chromophore groups are the azo group (\(-\text{N} = \text{N}-\)), and an ethylene group (\(-\text{C} = \text{C}-\)), which can cause color. The auxochrome groups are - NO\(_2\), -NH\(_2\), -COOH, -SO\(_3\)H, and water-soluble -OH is polar [20]. During the EC process the auxochrome groups can be released due to electrolysis[21]. It can be proved that in fact if the EC was carried out for a long time it produced a colorless floc.

![Figure 4. Color reduction vs HRT](image)

**A** Pattern of decrease in color intensity. **B** Pattern of change in% color removal

4. **Conclusion**

The application of voltage density (voltage and electrode distance) and flow rate (HRT) affects the water quality of the EC results. The results of this study showed the best treatment at a voltage density of 12 V cm\(^{-1}\) (18 Volt, 1.5 cm) and HRT 30 minutes (flow rate 1 mL s\(^{-1}\)). The COD reduction is 91.80% from 1683 mg L\(^{-1}\) to 138 mg L\(^{-1}\) which have met the quality standards and TSS decreased 87.83% from 937 mg L\(^{-1}\) to 114 mg L\(^{-1}\) who have not met the quality standard (> 50 mg L\(^{-1}\)), and are able to reduce color 84.26%, namely from 305.5 to 48.5 mg L\(^{-1}\) PtCo scale. The others are pH increase in the range of tolerant, DO increase up to 8.2 mg L\(^{-1}\), Conductivity increase up to 5350 \(\mu\)S cm\(^{-1}\), and Al increase up to 2.71 mg L\(^{-1}\).
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