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Performance of the Electro-Fenton Process in Reducing COD and TSS in Cigarette Industry Wastewater

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A B S T R A C T

Cigarette industry wastewater contains organic compounds and suspended solids that may adversely affect receiving water bodies when discharged without adequate treatment. This study evaluated the effects of aeration time, electric current, and contact time on dissolved oxygen (DO), chemical oxygen demand (COD), and total suspended solids (TSS) concentrations during Electro-Fenton treatment. The experiment was conducted in batch mode using a 3 L reactor, with aeration times of 0, 5, and 10 minutes, electric currents of 3, 4, and 5 A, and contact times of 10, 15, 20, 25, and 30 minutes. The initial wastewater characteristics were pH 4.8, DO 0 mg/L, COD 1,017 mg/L, and TSS 56 mg/L. Aeration increased the initial DO concentration to approximately 5.3 mg/L after 5 minutes and 6.9–7.1 mg/L after 10 minutes. The lowest final COD concentration, approximately 150 mg/L, was obtained at an electric current of 5 A, an aeration time of 10 minutes, and a contact time of 30 minutes. The lowest TSS concentration, approximately 10 mg/L, was observed at a contact time of 10 minutes under several treatment conditions. Longer contact times generally increased the TSS concentration, which was associated with the continued formation of iron-containing solids and flocs. These findings demonstrate that aeration time, electric current, and contact time influence COD and TSS concentrations during Electro-Fenton treatment of cigarette industry wastewater.

Contribution to Sustainable Development Goals (SDGs):

SDG 6: Clean Water and Sanitation

SDG 9: Industry, Innovation, and Infrastructure

SDG 12: Responsible Consumption and Production

1. INTRODUCTION

1.1. Research Background

The cigarette industry contributes significantly to the Indonesian economy. However, its production activities generate wastewater containing high concentrations of organic pollutants, including biochemical oxygen demand (BOD), chemical oxygen demand (COD), phenolic compounds, and other substances that may cause pH imbalance. The discharge of inadequately treated wastewater can increase the oxygen demand in receiving water

bodies, reduce dissolved oxygen availability, disrupt aquatic ecosystems, and deteriorate environmental quality [1].

Cigarette industry wastewater is classified as complex organic wastewater because it may contain nicotine, phenols, tobacco-specific nitrosamines, heavy metals, and volatile organic hydrocarbons. Its high organic load and relatively low biodegradability make conventional biological treatment less effective. Toxic compounds such as nicotine and phenol may also inhibit microbial activity during biological treatment [2]. Therefore, a more effective treatment method is required to degrade complex organic pollutants and reduce the pollutant load before the wastewater is discharged into the environment.



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Advanced Oxidation Processes (AOPs) are wastewater treatment methods based on the generation of highly reactive hydroxyl radicals ($\bullet\text{OH}$). These radicals have a high oxidation potential and can degrade complex organic pollutants into simpler compounds [3]. The Electro-Fenton process occurs through the reaction between ferrous ions (Fe^{2+}) and hydrogen peroxide (H_2O_2), producing hydroxyl radicals as the main oxidizing species. In the system used in this study, H_2O_2 was added externally, while Fe^{2+} was generated by dissolution of the iron anode. The generated hydroxyl radicals subsequently oxidize organic compounds, whereas iron hydroxide precipitates formed during the process can assist the removal of suspended solids through coagulation, adsorption, flocculation, and sedimentation mechanisms [4].

Previous research demonstrated that Electro-Fenton treatment substantially reduced COD concentrations in textile wastewater at an electrical voltage of 6 V and an electrode distance of 2 cm [5]. However, the study did not specifically evaluate the role of dissolved oxygen in COD removal. Other studies have explained that dissolved oxygen is a critical factor in the oxygen reduction reaction and the subsequent formation of oxidizing agents [6]. Aeration was therefore applied as a pretreatment to increase the dissolved oxygen concentration before the wastewater entered the Electro-Fenton reactor. Variations in aeration time, electric current, and contact time were evaluated to determine their effects on DO levels and the removal efficiencies of COD and total suspended solids (TSS).

This study aims to evaluate the effects of aeration time, electric current, and contact time on the performance of the Electro-Fenton process in treating cigarette industry wastewater.

1.2. Literature Review

1.2.1. Cigarette Industry Wastewater

Cigarette industry wastewater is generated by various production activities, including primary processing, clove processing, secondary processing, raw material washing, tobacco extraction, and equipment cleaning. These activities produce wastewater containing a high organic pollutant load and relatively low biodegradability, which may pose a risk of environmental pollution when discharged without adequate treatment [7]. Organic compounds commonly found in cigarette industry wastewater include nicotine, phenols, and other substances that can increase Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD), thereby reducing the quality of receiving water bodies [1].

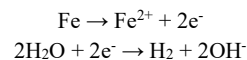
Cigarette industry wastewater is classified as complex organic wastewater because it generally has high COD concentrations and a low biodegradation rate [8], [9]. It may also contain toxic components, including nicotine, tobacco-specific nitrosamines, heavy metals, and volatile organic hydrocarbons [2]. The presence of these substances increases the complexity of the wastewater and may inhibit the activity of microorganisms in conventional biological treatment systems. The combination of a high organic load, low biodegradability, and toxic compounds makes cigarette industry wastewater difficult to treat using conventional methods alone.

Therefore, wastewater from the cigarette industry must be treated before being discharged into the environment. According to the Regulation of the Minister of Environment of the Republic of Indonesia Number 5 of 2014, the maximum allowable concentrations for Category I cigarette industry wastewater are

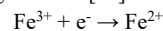
300 mg/L for COD, 150 mg/L for BOD, 100 mg/L for TSS, 0.5 mg/L for phenol, 3.0 mg/L for ammonia, and 5.0 mg/L for oil and grease, with a permitted pH range of 6.0-9.0 [10]. These standards indicate the need for an effective treatment technology capable of reducing organic pollutants and suspended solids to meet the applicable wastewater discharge requirements.

1.2.2. Electro-Fenton Process

The Electro-Fenton process is an electrochemical development of the conventional Fenton process that combines electrochemical reactions with the formation of hydroxyl radicals ($\bullet\text{OH}$). In the conventional Fenton reaction, ferrous ions (Fe^{2+}) activate hydrogen peroxide (H_2O_2) to produce hydroxyl radicals, which have a high oxidation potential and are capable of degrading complex organic compounds [11], [12]. The principal Fenton reaction is expressed as follows:



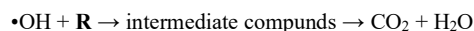
The released Fe^{2+} subsequently reacts with H_2O_2 to produce hydroxyl radicals. These radicals oxidize organic compounds non-selectively and convert them into smaller intermediate compounds, which may undergo further oxidation into simpler products. At the same time, Fe^{3+} produced during the Fenton reaction can be reduced back to Fe^{2+} through electron transfer, as shown in the following reaction [13]:



The regeneration of Fe^{2+} supports the continuity of the $\text{Fe}^{2+}/\text{Fe}^{3+}$ cycle and maintains hydroxyl-radical formation during treatment. This mechanism can improve iron utilization and reduce the need for large amounts of externally supplied iron compounds [6]. In addition to organic oxidation, iron ions may undergo hydrolysis, forming iron hydroxide precipitates. These precipitates can facilitate the removal of suspended particles through coagulation, adsorption, flocculation, and sedimentation.

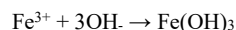
1.2.3. COD and TSS Removal Mechanism

COD removal in the Electro-Fenton process primarily occurs through the oxidation of organic compounds by hydroxyl radicals ($\bullet\text{OH}$). These radicals have a high oxidation potential and react non-selectively with complex organic pollutants, including nicotine, phenols, and other compounds contained in cigarette industry wastewater [3], [11]. The oxidation process breaks complex molecular structures into smaller intermediate compounds, which may subsequently undergo further oxidation into simpler products. The reaction can be generally represented as follows:



Where **R** represents organic compounds contained in the wastewater. The degradation process occurs gradually rather than through direct mineralization. As the concentration of oxidizable organic compounds decreases, the amount of oxygen required for chemical oxidation also decreases, resulting in a lower COD concentration.

TSS removal occurs through a different mechanism involving coagulation, adsorption, flocculation, and sedimentation. Fenton-based treatment does not only oxidize dissolved organic compounds but also produces iron species that can capture suspended particles [14]. During the Electro-Fenton process, Fe^{2+} and Fe^{3+} undergo hydrolysis and form iron hydroxide precipitates, including $\text{Fe}(\text{OH})_2$ and $\text{Fe}(\text{OH})_3$. The formation of ferric hydroxide can be represented by the following reaction [15]:



The iron hydroxide precipitates act as coagulants by neutralizing the surface charges of colloidal particles, adsorbing pollutants, and entrapping suspended solids. The destabilized particles subsequently combine with the precipitates to form larger flocs that can be separated through sedimentation or filtration [14][16]. Therefore, TSS removal is mainly determined by the formation, aggregation, and settling of iron hydroxide flocs.

1.2.4. Effect of Operating Conditions on Electro-Fenton Performance

The performance of the Electro-Fenton process is influenced by the interaction among electric current, aeration time, and contact time. These operating conditions affect the availability of ferrous ions, dissolved oxygen, hydrogen peroxide, hydroxyl radicals, and iron hydroxide flocs. Therefore, changes in one factor do not always produce a proportional increase in COD and TSS removal because the process involves simultaneous oxidation and coagulation mechanisms.

Electric current controls the rate of electrochemical reactions and the dissolution of the iron anode. Increasing the electric current can increase the release of Fe^{2+} into the wastewater, thereby providing more catalyst for the Fenton reaction and supporting hydroxyl-radical formation. Previous studies have shown that electric current is an important factor affecting COD removal in Electro-Fenton treatment [4], [17]. However, excessive current may accelerate iron dissolution, increase hydrogen evolution, promote competing reactions, and generate excessive iron hydroxide solids. Consequently, a higher electric current does not always result in better treatment performance, particularly for TSS removal.

Aeration time affects the initial dissolved oxygen concentration and oxygen mass transfer into the wastewater. A longer aeration period generally increases the opportunity for oxygen to dissolve in the liquid phase until it approaches saturation [18]. Dissolved oxygen may participate in the oxygen reduction reaction at the cathode and support the formation of H_2O_2 and hydroxyl radicals [6], [19]. In the system used in this study, H_2O_2 was externally added; therefore, aeration mainly functioned to increase the initial DO concentration and potentially support the availability of oxidizing species. Nevertheless, increasing aeration time does not necessarily improve pollutant removal continuously because the process is also controlled by Fe^{2+} availability, H_2O_2 concentration, pH, electric current, and reaction time.

Contact time determines the duration available for organic oxidation, iron dissolution, floc formation, and particle sedimentation. A longer contact time generally provides more opportunity for hydroxyl radicals to degrade organic compounds, potentially improving COD removal [17]. However, prolonged electrolysis may cause excessive anode dissolution and increase the formation of iron hydroxide solids. If these solids do not settle completely, they may remain suspended, thereby increasing the measured TSS concentration [14], [16]. Therefore, the optimum contact time for COD removal may differ from that required for TSS removal.

1.3. Research Objective

This study aims to evaluate the effects of aeration time, electric current, and contact time on the performance of the Electro-Fenton process in reducing Chemical Oxygen Demand (COD) and Total Suspended Solids (TSS) in cigarette industry wastewater.

2. MATERIALS AND METHODS

The wastewater used in this study was collected from a cigarette industry located in East Java, Indonesia. The experimental equipment consisted of an aeration reactor, a batch Electro-Fenton reactor, a direct-current power supply, an aerator, a magnetic stirrer, and iron electrodes. Hydrogen peroxide (H_2O_2 , 50%) was used as the oxidizing agent. For each 3 L batch of wastewater, 11 mL of 50% H_2O_2 solution was added at the beginning of the Electro-Fenton treatment. No pH adjustment was performed before or during the Electro-Fenton treatment, and the process was allowed to proceed at the wastewater's natural pH. Commercial iron plates were employed as the anode and cathode. Each electrode measured 15×10 cm, with a submerged surface area of approximately 300 cm^2 per electrode, for a total of 600 cm^2 . The distance between the electrodes was maintained at 4 cm.

3. RESULT AND DISCUSSION

3.1. Characteristics of Cigarette Industry Wastewater

The cigarette industry wastewater used in this study was first characterized to determine its initial conditions prior to treatment. The parameters observed included COD, TSS, pH, temperature, and dissolved oxygen (DO). The results of the initial characterization are presented in Table 1. Based on these results, the wastewater exhibited a high organic pollutant load and a pH tending toward the acidic range, thus requiring further treatment before discharge into the environment.

Table 1. Initial Characteristics of Cigarette Industry Wastewater

pH	DO (mg/L)	COD (mg/L)	TSS (mg/L)
4,8	0	1017	56

Based on Table 1, the cigarette industry wastewater had an acidic pH of 4.8, which was below the permitted range of 6.0-9.0. The acidic condition indicates that the wastewater requires further treatment before discharge and may also influence the chemical reactions occurring during the Electro-Fenton process.

The COD concentration reached 1,017 mg/L, substantially exceeding the applicable discharge standard of 300 mg/L. This high COD value reflects a considerable organic pollutant load and indicates that a large amount of oxygen would be required to oxidize the organic compounds contained in the wastewater. In addition, the DO concentration was 0 mg/L, indicating that dissolved oxygen was absent in the untreated wastewater. This condition further confirms the high oxygen demand caused by the organic matter present.

Meanwhile, the TSS concentration was 56 mg/L, which remained below the standard limit of 100 mg/L. Although the TSS concentration complied with the standard, the elevated COD, acidic pH, and absence of dissolved oxygen demonstrate that the

wastewater still required effective treatment before being discharged into the environment.

3.2. Effect of Operating Conditions on COD Concentration

The electro-Fenton process reduced the initial COD concentration of cigarette industry wastewater from 1,017 mg/L under various operating conditions. The final COD concentration was influenced by electric current, aeration time, and contact time. In general, increasing the contact time improved COD removal because a longer reaction period provided more opportunity for hydroxyl radicals to oxidize organic compounds in the wastewater. However, the effect of electric current and aeration was not linear under all treatment conditions.

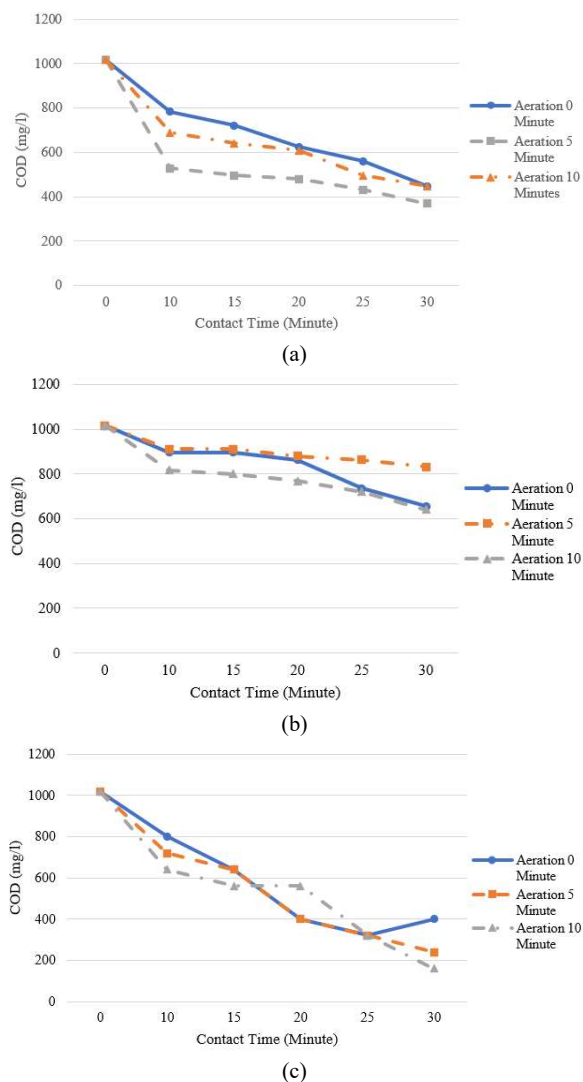


Fig.1 Effect of Contact Time and Aeration Time on COD Concentration at Different Electric Currents: (a) 3 A, (b) 4 A, and (c) 5 A

The figure shows changes in COD levels during the Electro-Fenton process at electrical current of 3 A, 4 A, and 5 A with aeration times of 0, 5, and 10 minutes. In general, COD levels decrease as contact time increases, although the rate of decrease varies with electric current and aeration time. At a current of 3 A, as shown in Figure (a), the COD decrease occurred gradually across all aeration time variations. After a contact time of 30 minutes, the final COD concentrations for 0-minute and 5-minute

aeration reached approximately 450 mg/L and 430 mg/L, respectively, while 10-minute aeration resulted in the lowest final COD concentration, at approximately 350 mg/L.

At an electric current of 4 A in Figure (b), the COD reduction tends to be slower compared to other current intensities. The final COD concentration after a 30-minute contact time was recorded at approximately 650 mg/L with 0 minutes of aeration, 820 mg/L with 5 minutes of aeration, and 630 mg/L with 10 minutes of aeration.

The slower COD reduction observed at 4 A may be associated with the non-linear interaction among iron dissolution, H_2O_2 availability, and hydroxyl-radical formation during the Electro-Fenton process. Although increasing the electric current can enhance Fe^{2+} generation from the iron anode, a higher current does not necessarily result in a proportional increase in COD removal. Excessive electrochemical reactions may promote competing reactions, hydrogen evolution, and the formation of iron hydroxide solids, which can reduce the effective utilization of reactive species for organic oxidation [4], [17]. Therefore, the lower COD reduction at 4 A compared with 3 A and 5 A may reflect an imbalance between Fe^{2+} generation and oxidant availability under these experimental conditions rather than a direct linear effect of electric current.

Meanwhile, the use of a current strength of 5 A in Figure (c) resulted in the greatest reduction in COD. At the end of the 30-minute contact time, the COD concentration with 0 minutes of aeration was around 400 mg/L, while 5 minutes and 10 minutes of aeration resulted in COD concentrations of approximately 240 mg/L and 150 mg/L, respectively. These results indicate that a 5 A current with 10 minutes of aeration yielded the lowest final COD concentration among the treatment variations.

The reduction in COD observed in this study is comparable with the performance reported for Electro-Fenton treatment applied to other types of industrial wastewater. In the present study, the COD concentration decreased from 1,017 mg/L to approximately 150 mg/L under a current of 5 A, 10 minutes of aeration, and 30 minutes of contact time. Ramadeandra et al. [4] reported a 77.14% COD reduction in batik wastewater at a current of 5 A and a contact time of 55 minutes. Priyadi and Ratni [5] reported a COD reduction of 94.1% in textile wastewater after 180 minutes of Electro-Fenton treatment. Similarly, Ghjair and Abbar [17] achieved approximately 96.27% COD removal from hospital wastewater in 46.67 minutes. Although direct comparison should be interpreted with caution due to differences in wastewater characteristics, H_2O_2 dosage, electrode configuration, and operating conditions, these findings indicate that the Electro-Fenton process applied to cigarette industry wastewater achieved substantial COD reduction in a relatively short contact time.

3.3. Effect of Operating Conditions on TSS Concentration

The electro-Fenton process reduced the initial TSS concentration from 56 mg/L under various combinations of electric current, aeration time, and contact time. In general, the lowest TSS concentrations were obtained at a contact time of 10 min. However, prolonging the treatment beyond this point tended to increase the TSS concentration, indicating that longer electrolysis did not necessarily improve suspended-solid removal.

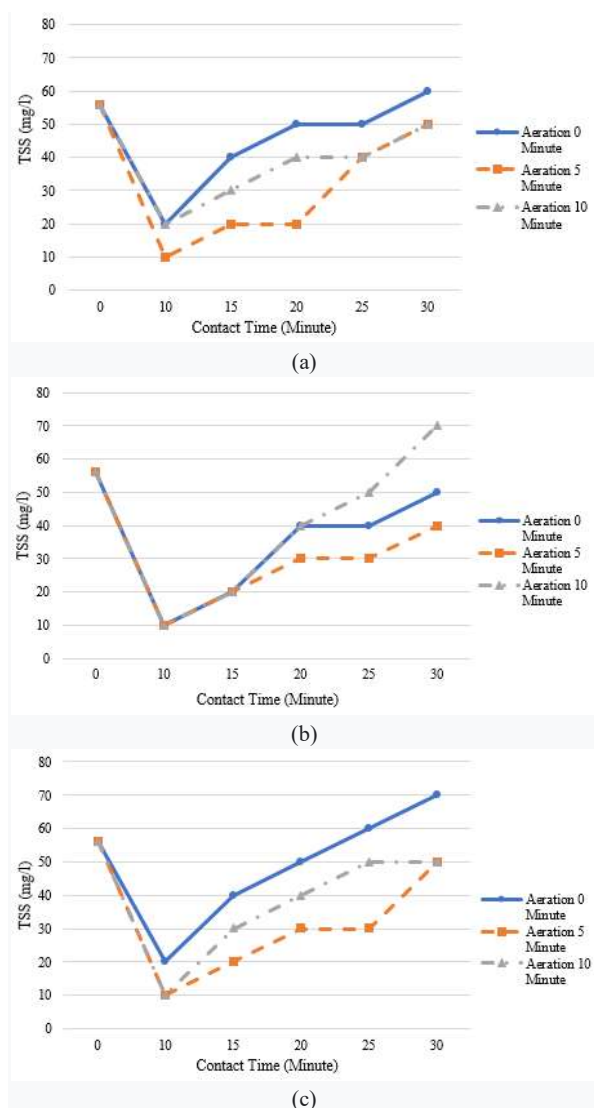


Fig.2 Effect of Contact Time and Aeration Time on TSS Concentration at Different Electric Currents: (a) 3 A, (b) 4 A, and (c) 5 A

The figure shows changes in TSS concentration during the electro-Fenton process at current densities of 3 A, 4 A, and 5 A, with aeration times of 0, 5, and 10 minutes. Across all treatment variations, the initial TSS concentration of 56 mg/L decreased sharply within 10 minutes of contact time. The lowest TSS concentration, approximately 10 mg/L, was observed under several conditions, particularly during the 5-minute and 10-minute aeration treatments at certain current strengths. However, after 10 minutes of contact time, the TSS concentration tended to rise again until the end of the process.

At a current strength of 3 A, the TSS concentration after a 30-minute contact time reached approximately 55 mg/L for 0 minutes of aeration and approximately 45 mg/L for 5 and 10 minutes of aeration. At a current strength of 4 A, the final TSS concentrations were approximately 50 mg/L, 40 mg/L, and 70 mg/L for aeration times of 0, 5, and 10 minutes, respectively. Meanwhile, at a current strength of 5 A, the final TSS concentration reached approximately 70 mg/L under non-aerated conditions and approximately 40 mg/L after 5 and 10 minutes of aeration. Overall, a contact time of 10 minutes resulted in the

lowest TSS concentration, whereas longer contact times caused the TSS concentration to rise again. This pattern indicates that extending the process time does not always improve TSS removal, as it may be accompanied by the formation of increasing amounts of solids and iron flocs.

4. CONCLUSION

The Electro-Fenton process effectively reduced COD and TSS concentrations in cigarette-industry wastewater under the operating conditions investigated. The lowest final COD concentration, approximately 150 mg/L, was obtained at an electric current of 5 A, an aeration time of 10 minutes, and a contact time of 30 minutes. Meanwhile, the lowest TSS concentration, approximately 10 mg/L, was obtained at a contact time of 10 minutes under several combinations of electric current and aeration time. Prolonging the contact time beyond 10 minutes generally increased the TSS concentration, which was associated with the continued formation of iron-containing solids and flocs. These results indicate that electric current, aeration time, and contact time play important roles in determining the performance of the Electro-Fenton process. Future studies should evaluate energy consumption, electrode loss, and operational costs across different electric currents to assess the technical and economic feasibility of the process for large-scale cigarette-industry wastewater treatment.

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