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Research Article

Efficiency of Electrocoagulation for Laboratory Wastewater Treatment Using Aluminum Electrodes

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Abstract

Environmental analysis of waste generated from residual samples in a testing laboratory is necessary to mitigate its impact. This study aims to assess the performance of electrocoagulation (EC) using aluminum (Al) electrodes in reducing Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), and pH level in laboratory wastewater. This investigation successfully employed an EC process utilizing Al electrodes in both anode and cathode configurations. The experimental conditions include Voltage (10, and 20 V), contact time (15, 30, 45, and 60 minutes), and the electrode configuration (monopolar and bipolar). The results indicated that a bipolar configuration of Al electrode relatively outperformed a monopolar configuration. Optimal condition was achieved at 20 V, and contact time of 60 minutes. Results showed COD removal efficiency up to 96.15% reducing COD from 627.45 to 24.183 mg/L, TSS removal efficiency up to 92.45%, lowering TSS from 53 to 4 mg/L. While pH increased during the process, it remained within acceptable limits. This substantial reduction in pollutants significantly improved water quality, surpassing regulatory standards. The results suggest that EC is a promising approach for achieving sustainable treatment for laboratory wastewater.

Keywords: Laboratory wastewater, Electrocoagulation, Aluminum electrode, COD removal, TSS removal

1. Introduction

Despite the growing number of commercial testing laboratories, there is a lack of adequate laboratory waste management. It is believed that a significant portion of laboratories registered on the official website of Indonesian Ministry of Environment and Forestry do not have adequate waste treatment facilities. Most of these laboratories still rely on third-party services for waste disposal, with annual disposal being the common practice. Due to the extended storage period of laboratory waste, which is often composed of chemical residue, there is a heightened risk of environmental pollution. Considering this, to mitigate the potential for contamination, effective waste treatment is necessary [1]. A variety of techniques are available for wastewater treatment, incorporating methods such as adsorption and ion exchange [2, 3] as well as membrane technologies [4-6], electrodialysis [7], Electrocoagulation (EC) [8].

The EC process is widely recognized as an environmentally friendly approach and one of the most effective methods for wastewater treatment. EC emerged as promising wastewater technology due to superior efficiency, reduced risks associated with chemical handling [9] lower sludge production [10], and rapid treatment compared to conventional chemical coagulation methods [9]. EC is influenced by several factors, namely applied voltage, electrode distance, electrode configuration, solution pH, electrode materials, wastewater characteristic, and contact time [11]. Extensive research has been conducted on the application of electrocoagulation for wastewater treatment. Table 1 Provides a comprehensive overview of these

studies, emphasizing the impact of operational parameters such as electrode material, current density, and pH on treatment efficiency. Electrocoagulation processes frequently make use of aluminum and iron electrodes, largely due to their affordability, commercial accessibility, and the desirable characteristics of their hydroxide, including low toxicity and high valence, which contribute to effective pollutant removal. Despite these factors, aluminum is the preferred material due to its stability, handling convenience, and solubility [12]. According to Huang et al. [13] the electrodes reactions for aluminum are given in equations (1-5)

Cathode reaction for aluminum:

$$2H_2O + 2e^- \longrightarrow 2OH^- + H_2 \quad (1)$$

$$2H_2O + O_2 + 4e^- \longrightarrow OH^- + H_2 \quad (2)$$

Anode reaction for aluminum:

$$Al \longrightarrow Al^{3+} + 3e^- \quad (3)$$

$$Al^{3+} + 3OH^- \longrightarrow Al(OH)_3 \quad (4)$$

The oxidation of aluminum results in the formation of Al^{3+} ions, which subsequently precipitate as aluminum hydroxide.

Overall reaction:

$$2\,Al + 6H_2O \longrightarrow 2Al(OH)_3 + 3H_2 \quad (5)$$

Aluminum hydroxide ($Al(OH)_3$), formed from Al^{3+} and OH^- ions, acted as coagulants to destabilized pollutant [8], the amorphous $Al(OH)_3$ generated exhibits a substantial surface area, enabling the efficient adsorption of dissolved organic matter and colloidal particles. These particles can be efficiently separated from the aqueous phase through floatation [14]. The formation of hydrogen bubbles at cathode increases fluid agitation and aids in contaminant adsorption, decreasing their buoyant density. Consequently, the floatation separation process is enhanced [15].

Table 1. Overview of documented research on electrocoagulation in water treatment.

Waste water source	Pollutant concentration (mg/L)	Electrode	Treatment time (min)	Removal efficiency (%)	References
Fish Processing	520 (COD) 262 (TSS)	Aluminum and Iron	120	96.34 (COD) 73.22 (TSS)	Munawarah, et al. [16]
Oleochemical	87000 (COD) 983 (TSS)	Aluminum	30	62.64 (COD) 66.12 (TSS)	Azli & Azoddein [17]
Domestic Wastewater	472 (COD)	Aluminum and Iron	27	94	Oktiawan et al. [18]
Tanney Industry	4162.3 (COD) 1825 (BOD	Aluminum	24	56.8 (COD 69.2 (BOD)	Aguilar-Ascón, et al. [19]
Ketchup Industry	12032 (COD) 847.07 (TSS)	Aluminum	20	81.86 (COD) 82.61 (TSS)	Syaifuddin & Bagastyo [20]
Cork Boiling	271 (TSS)	Aluminum	60	99	Silva et al. [21]
Textile Dyeing	369-397 (COD)	Copper and Aluminum	20	73.8 ± 1.57	Nam et al. [22]
Oil rigs drilling	1568 (TSS)	Aluminum and Iron	60	67	Ale-Tayeb et al. [23]
Furniture industry	23552 ± 5649 (COD)	Stainless-steel and Iron	30	92.5 ± 0.6	Vicente et al. [24]
Hospital	4533 (COD)	Iron	90	80.78	Rangseesuriyachai et al. [25]

In an effort to address the issue of laboratory waste management, the objective of this study was to evaluate the performance of the electrocoagulation process in treating laboratory wastewater by systematically investigating the impact of key operational parameters such as applied voltage, contact time, and electrode configuration.

2. Materials and methods

2.1 Laboratory wastewater characteristics

Laboratory wastewater samples were obtained from an environmental laboratory located in Cimahi, West Java. These samples consisted of residual samples from analytical instrument, leftover reagents, and waste generated during the analysis process. A total of 16 experiments were conducted, each using 500 mL of sample. The characteristics of the laboratory wastewater prior to EC are summarized in table 2.

Table 2. Characteristics of Laboratory Wastewater

Parameter	Value	Discharge limits
pH	1.77	6.0 – 9.0
COD (mg/L)	627.45	150.0
TSS (mg/L)	53.00	50.0

The discharge limits were adopted from Indonesian standards listed on PERMEN LHK RI No, P.5/MENLHK/SETJEN/KUM. 15/10/2014. The sample was kept at 4 °C to prevent degradation.

2.2 Equipment and instrument

The EC setup consisted of a 1000 mL beaker as the reaction vessel, a Direct Current (DC) power supply (ATTEN PPS3005S), Al electrodes (10 × 4 × 0.659 cm), a pH meter (Mettler Toledo FG2), an oven (Memmert UN55) for electrode drying and TSS measurement, a spectrophotometer (Shimadzu UV-VIS 1800) for COD measurement, and an ultrasonic cleaner (Dealta D68H) for electrode maintenance.

2.3 Operating conditions and preparations.

Al electrodes (10 × 4 × 0.659 cm) were used in this study. The electrode was subjected to a series of preparation steps, including polishing, ultrasonic cleaning, sanding, acetone rinsing, and oven drying at 100 °C for 60 minutes. After being dried to a constant weight and stored in a desiccator, the electrodes were connected to a DC power supply and positioned vertically and parallel., with 2 cm inter-electrode distance. To minimize Ohmic resistance [26], smaller inter-electrode distance is generally preferred. For each experiment, 500 mL of wastewater was treated in 1000 mL beaker, as depicted in figure 1.

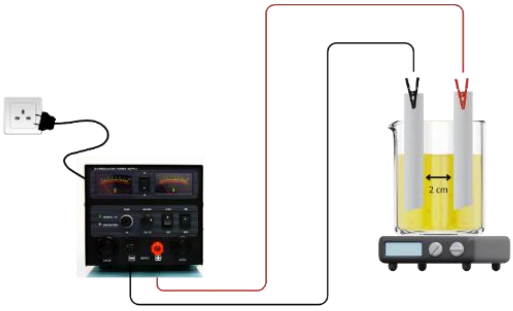

Figure 1. Electrocoagulation setup.

One aluminum serves as the cathode, while the other acted as the anode. The study was conducted in 16 batches, varying the EC process length (15, 30, 45, and 60 minutes), electrode configuration (Monopolar, and Bipolar), and applied voltage (10, and 20 V).

2.4 Analytical method

The pH measurements were conducted using pH meter. The quantification of COD and TSS was carried out using the methodologist outlined in Standard Methods for the Examination of Water and Wastewater [27]. COD was measured using a closed reflux colorimetric method with potassium dichromate ($K_2Cr_2O_7$) 0.01 N as the oxidant and 10.12 g silver sulfate (Ag_2SO_4) in 1000 mL of Sulfuric Acid (H_2SO_4) as the catalyst. Samples was digested at 150 °C for 120 minutes in a thermoreactor (CR2200 - WTW). COD analysis was performed using a UV/Visible spectrophotometer (Shimadzu UV-1800). TSS was measured using a filtration method with glass microfiber filter paper (Whatman 934-AH 1.5 µm). The filter paper was dried at 120 °C for 60 minutes to a constant weight.

All reagent used in this study were analytical grade (p.a.) from Sigma-Aldrich.

The percentage removal efficiencies of COD and TSS were calculated using equation 6 and 7, respectively [28]

$$\text{COD Removal} = \frac{\text{COD}_o - \text{COD}_t}{\text{COD}_o} \text{ x } 100\% \quad (6)$$

$$\text{TSS Removal} = \frac{\text{TSS}_o - \text{TSS}_t}{\text{TSS}_o} \text{ x } 100\% \quad (7)$$

Where COD_0 and CODt symbolize the initial and final COD concentrations (mg/L), respectively. Likewise, TSS_0 and TSSt represent the initial and final TSS concentrations (mg/L).

3. Results and discussion

3.1 Temperature changes during EC process

The temperature rise of the wastewater during treatment depends on factors like pH, voltage, and processing time. Figure 2 visually represents these temperature changes during the EC process. As shown in Figure 3, the EC process causes the temperature rise from 20 to 77 °C.

The release of Al^{3+} ions during EC leads to an increase in pH, which, in turn, raises the temperature and accelerate the reaction rate [29, 30]. Extended EC treatment can lead to a substantial increase in wastewater temperature. This temperature rise is primarily due to the exothermic nature of the electrochemical reactions involved [31].

Exothermic reactions release heat, raising the solution temperature. The heat generated by the process can lead into increase in wastewater temperature, potentially affecting both treatment efficiency and the quality of treated effluent [32]. Consequently, rigorous monitoring and control of temperature are necessary. Excessive temperature increase can affect the efficiency of the process and the quality of the treated effluent [33].

3.2. COD removal efficiency

According to the research conducted by Gusa et al. [34], the addition of electrodes results in more efficient outcomes. As seen on Figure 3, the concentration of COD removed increased significantly due to the addition of two electrodes. With the highest efficiency of COD removed at 96.14%, addition of two electrodes showed more efficient outcome. It is because by increasing the number of the electrodes, the active surface area also increases, thereby increasing the number of contaminants that adhere to the electrodes [35]. When there are additional plates placed between the anode and cathode, the EC process performance improves due to an increase in the amount of coagulants, which subsequently enhances removal efficiencies [36].

Figure 4 demonstrates the variation in COD removal efficiency as a function of applied voltage for various electrode configurations. In accordance with Faraday's law, an increase in applied voltage results in proportional increase in the quantity of coagulant generated, leading to enhanced COD removal [37], the applied voltage not only determines the coagulant dosage rate but also influences bubble production rate and size. This, in turn, affect mass transfer phenomena between pollutants, coagulants, and gas, thereby impacting floc formation [38]. Figure 4 shows that for a given contact time, the COD removal efficiency exhibited a slight increase with the elevation of electrical potential. Variations in the configuration of electrodes also led to improved efficiency at a higher electrical potential of 20 V. Although higher voltages resulted in enhanced treatment performance, they concurrently led to a substantial increase in energy consumption, as confirmed by previous research [39]. Our experimental findings further corroborate this, demonstrating a significant rise in energy consumption from 12.29 to 41.47 kWh/m^3 for the monopolar. configuration and from 23.25 to 114.8 kWh/m^3 for the bipolar configuration. Consequently, operational costs increased from Rp 10.44 to Rp 35.34 for monopolar and from Rp 19.76 to Rp 97.55 for bipolar electrode configurations, respectively. These results highlight the importance of balancing treatment performance with energy efficiency.

The variation of applied voltage is also conducted in different given time; the results can be seen in table 3.

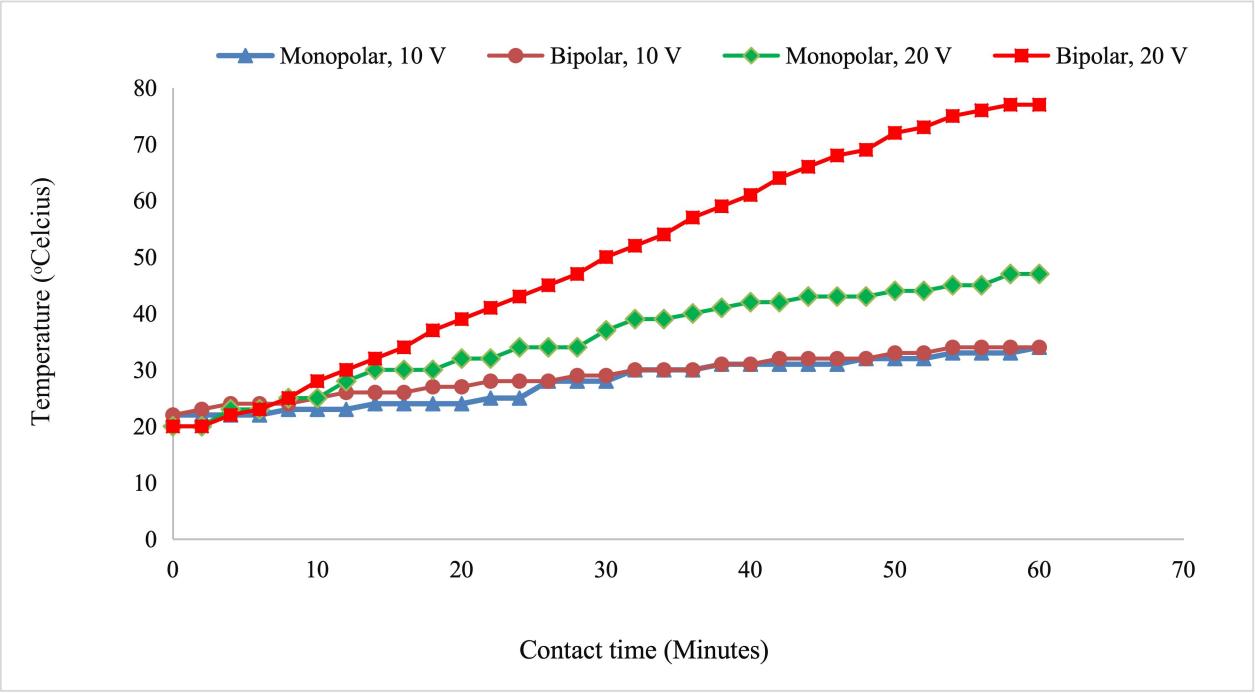

Figure 2. Changes in temperature over time for monopolar and bipolar electrode configurations during the electrocoagulation process, indicating the rise in temperature from 20 °C to 77 °C.

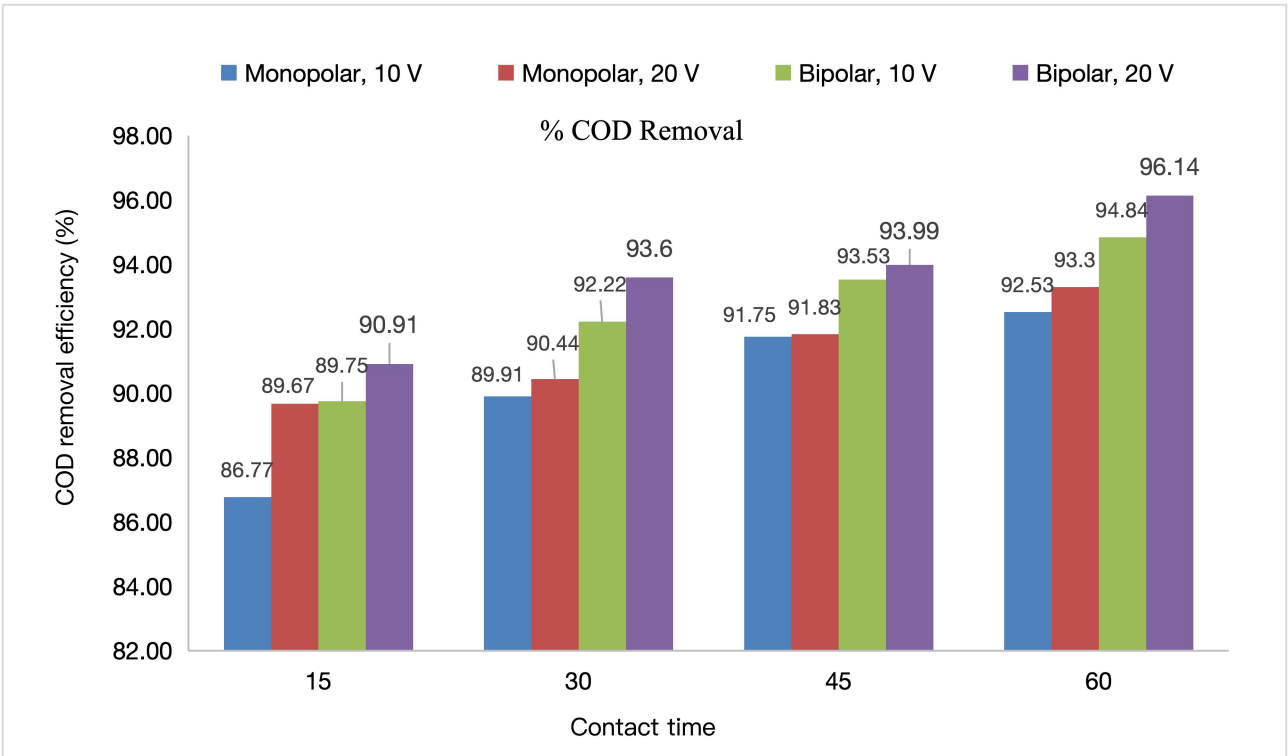

Figure 3. Comparison of COD removal efficiency under varying electrode configuration, showing the increase in efficiency from 92.53 to 94.84% and from 93.3 to 96.14% with addition of two electrodes.

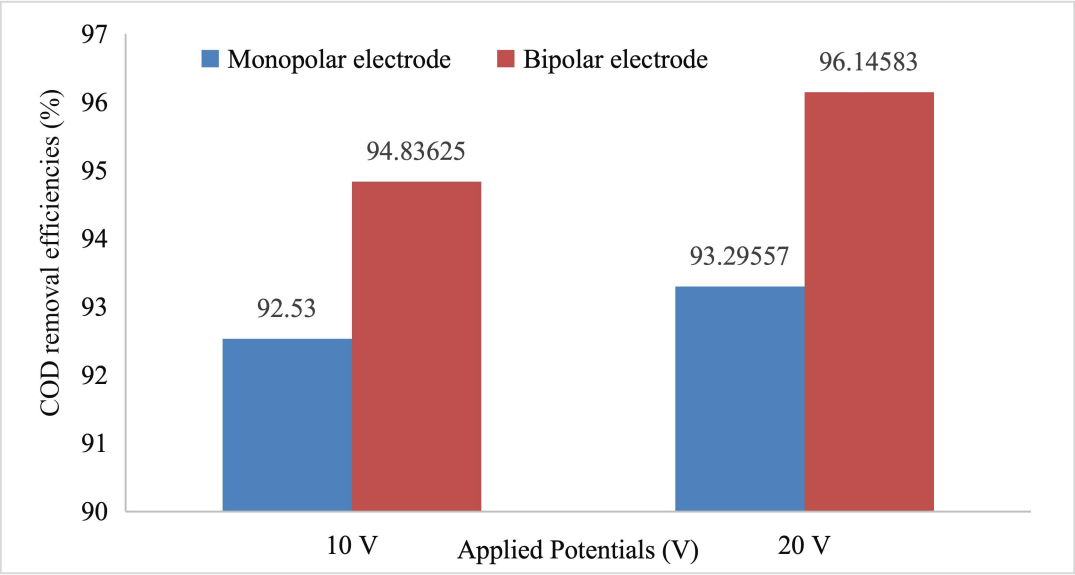

Figure 4. Variation of COD (Chemical Oxygen Demand) removal efficiency as a function of applied voltage for different configuration of electrodes used in the treatment process.

Table 3 shown that increasing applied voltage resulted in a modest improvement in the outcome of each experiment. This is credited to the fact that higher voltage leads to increased oxidation of aluminum, resulting in an increased quantity of precipitate for pollutant removal [40].

As contact time increases allows for greater formation of metal ions and hydroxide flocs, which in turn enhances the removal pollutant from the wastewater [41]. Conversely, with shortened contact time, there is insufficient time for both the Al^{3+} to dissolve from the anode which act as a destabilizing agent [42], and hydrogen gas at cathode in the solution [43].

Figure 5 depict more efficient outcomes of the EC process under prolonged contact time. Prolonging the contact time from 15 to 60 minutes led to a slightly enhancement in COD removal, increasing from 86.77 to 92.53% for the monopolar configuration and from 89.75 to 94.84% for the bipolar configuration. When given higher voltage, the amount of COD removal showed a slight increase from 89.67 to 93.3% for monopolar configuration, and from 90.91 to 96.14% for bipolar configuration. This is ascribed to the fact that the electrolysis duration significantly impacts the production of requisite ion concentrations from the electrodes. These ions serve as precursors for adsorbent formation. Simultaneously, the electrolytic process generates gas bubbles at both electrodes, which enhance the upward transport of destabilized contaminants within the solution [8].

3.3. TSS Removal efficiency

The effectiveness of pollutant removal through electrolysis depends on several factors, including metal ions present during the process, type of electrode used, electrical current strength and electric potential, and the duration of the process [44]. Table 4 summarizes the results of the experiments conducted to investigate the influence of various factors, such as electrode configuration, applied voltage, and contact time, on TSS removal efficiency. The data suggests that the highest TSS removal was achieved using a bipolar configuration at an applied voltage of 20 V and a contact time of 60 minutes, among the various factors tested based on the experiment results.

This is evidenced by a TSS reduction of 49 mg/L, resulting in efficiency of 92.45%. These results can be attributed to several factors, including the addition of two extra electrodes, which increased the active surface area, thus enhancing pollutant removal [35], while the extended reaction time provided

sufficient time for the formation of a larger number of metal ions and the evolution of a greater volume of gasses [45], both of which contributed to the improved pollutant removal efficiency.

Table 3. Results showing the impact of applied voltage on COD removal efficiency across different experimental setups.

Contact time (Minutes)	Electrode configuration	Applied voltage (V)	COD removal efficiency (%)
15	Monopolar	10	86.77
30	Monopolar	10	89.91
45	Monopolar	10	91.75
15	Monopolar	20	89.67
30	Monopolar	20	90.44
45	Monopolar	20	91.83
15	Bipolar	10	89.75
30	Bipolar	10	92.22
45	Bipolar	10	93.53
15	Bipolar	20	90.91
30	Bipolar	20	93.60
45	Bipolar	20	93.99

Additionally, increasing the applied voltage accelerated the EC process. The elevated current density facilitated a higher rate of metal ion release, leading to increased precipitate formation and improve pollutant removal [46]. Therefore, the use of two additional electrodes and a higher voltage yielded better results compared to the other configurations at all tested time intervals. This suggests that the configuration and operating parameters significantly influence the performance of the EC process [11].

3.4. pH change

The EC process typically results in an increase in solution pH. This pH elevation is attributed to electrolytic reactions at the aluminum cathode. The reaction at the cathode causing the reduction of water (H_2O) generated hydroxide ions (OH^-) and hydrogen gas (H_2). Several studies have reported an increase in solution pH during the EC process [47-50], observed that solutions with initial pH below pH 9 underwent pH increases. In this study, shown by Figure 6 it was also observed that the solution pH increased. The most significant pH increase was observed in the experiment using four plates at 20 V. The most suitable pH for the process, 7.73, was observed at a contact time of 45 minutes. Increasing the contact time to 60 minutes caused a notable rise in pH, approaching an alkaline value of 8.92. This result is related to the fact that as the contact time and applied voltage in the EC process increase, the OH^- ions and H_2 gas generated at the cathode also increases. Consequently, prolonged EC contact time may lead to a highly alkaline pH (> 9), which can pose potential hazards [39].

To evaluate the effects of contact time, electrode configuration, and applied voltage on the efficiency of TSS and COD removal, a two-way Analysis of Variance (ANOVA) without replication was utilized. This statistical method was deemed appropriate as each experimental batch was subjected to a single test. Two hypotheses were formulated: the null hypothesis, which asserts that there are no significant differences between the means of the samples, and the alternative hypothesis, which posits that significant differences exist. The results of the ANOVA analysis provided calculated F-values and critical F-values for both the electrode configuration and the contact time/applied voltage factors. The calculated F-values were 2.8444 and 171.76, respectively, which surpassed their corresponding critical F-values of 2.0148 and 3.3158.

Consequently, the null hypothesis was rejected, indicating that variations in electrode configuration, contact time, and applied voltage had a significant impact on the efficiency of TSS and COD removal. Moreover, the p-values associated with the electrode configuration and contact time/applied voltage factors were determined to be 0.0072 and 4×10^{-17}, respectively.

Table 4. TSS removal efficiency under various condition.

Contact time (Minutes)	Electrode configuration	Applied voltage (V)	TSS removed (mg/L)	Efficiency (%)
15	Monopolar	10	38.60	72.83
30	Monopolar	10	41.80	78.87
45	Monopolar	10	44.00	83.02
60	Monopolar	10	46.20	87.17
15	Monopolar	20	40.00	75.47
30	Monopolar	20	42.00	79.25
45	Monopolar	20	45.00	84.91
60	Monopolar	20	46.80	88.30
15	Bipolar	10	41.60	78.49
30	Bipolar	10	43.40	81.89
45	Bipolar	10	45.60	86.04
60	Bipolar	10	47.40	89.43
15	Bipolar	20	44.80	84.53
30	Bipolar	20	46.20	87.17
45	Bipolar	20	47.80	90.19
60	Bipolar	20	49.00	92.45

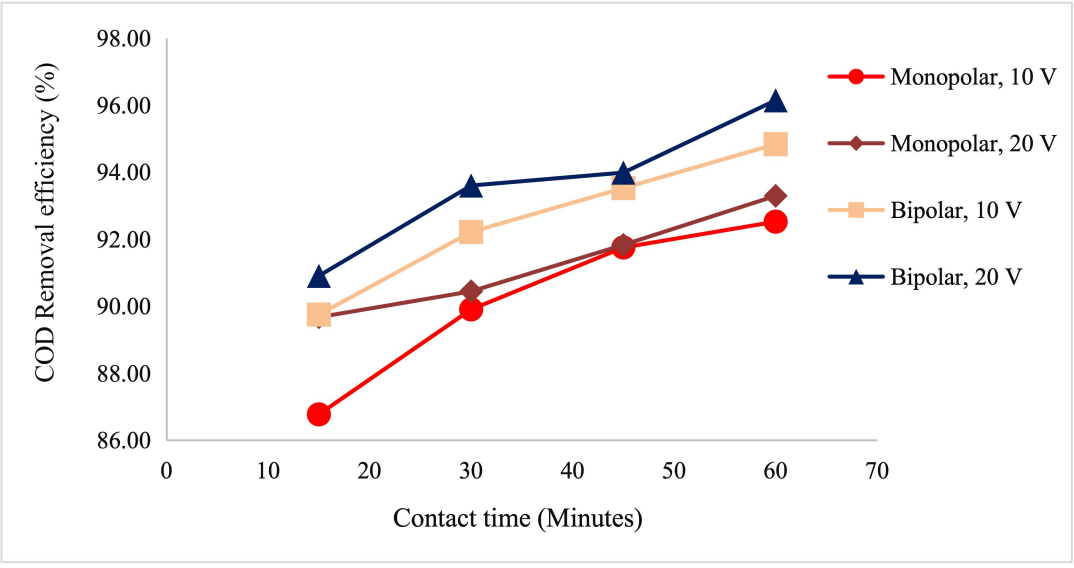

Figure 5. Effect of contact time on COD removal efficiency, demonstrating how longer contact times enhance pollutant removal rates.

The p-values obtained from the statistical analysis, all of which were less than 0.05, indicated that the observed differences in the efficiency of the electrocoagulation process are statistically significant. This confirms that the variation in electrode configuration, contact time, and applied voltage have a substantial impact on the treatment process.

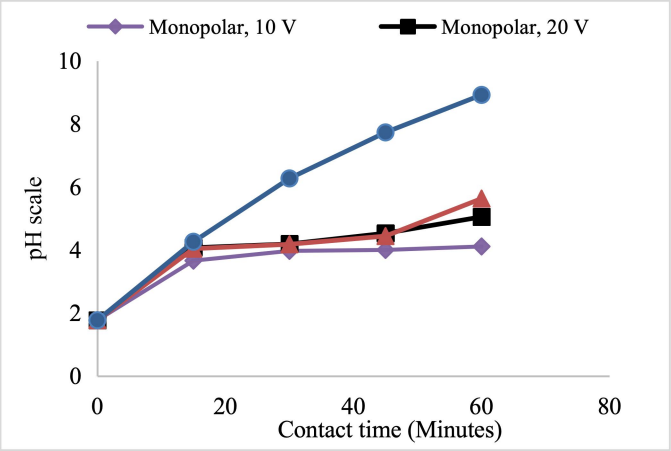

Figure 6. Change in pH levels during the electrocoagulation process, demonstrating the increase in pH over time with varying configurations. The most significant pH increase was observed with four plates at 20 V, reaching a peak of 8.92 after 60 minutes of contact time, indicating a trend towards alkalinity as the electrocoagulation process progresses.

4. Limitations

Scale Limitations: The experiments were conducted on a laboratory scale, which may not fully represent the complexities and variabilities present in full-scale industrial wastewater treatment applications. The results may not be directly transferable to larger systems without further validation.

Electrode Material Constraints: The study focused solely on aluminum electrodes, which, while effective, may have limitations in terms of corrosion and longevity compared to other materials. Future studies could explore alternative electrode materials to assess their efficiency and durability.

Environmental Variability: The wastewater samples used were collected from a specific environmental laboratory, which may not reflect the diverse characteristics of wastewater from different sources. Variations in composition could affect the generalizability of the findings.

Limited Parameter Exploration: Although several parameters such as voltage, contact time, and electrode configuration were varied, other factors like temperature fluctuations and wastewater composition were not extensively studied. These factors could influence the electrocoagulation process and its efficiency.

Long-term Performance: While this study successfully demonstrated the effectiveness of electrocoagulation in treating laboratory wastewater, it is important to note that the longevity and stability of the aluminum electrodes over multiple treatment cycles were not examined. Future research should focus on evaluating these aspects to determine how electrode performance may change with prolonged use. Understanding the durability of electrodes will be crucial for optimizing operational parameters and ensuring sustainable application in industrial wastewater treatment systems.

5. Conclusion

EC proved to be an effective method for laboratory wastewater treatment, achieving compliance with Ministry of Environment and Forestry Regulation No.5/2014 standards for COD, TSS, and pH. Optimal pollutant removal was achieved using bipolar electrode configuration at 20 V. Significant COD and TSS removal were achieved within 60-minutes contact time, with COD removal efficiencies ranging from 86.77 to 96.15%, initial COD level of 627.45 mg/L were reduced to a minimum of 24.18 mg/L. Similarly, TSS removal efficiencies ranged from 72.83 to 92.45%, reducing initial TSS level of 53 mg/L to a minimum of 4 mg/L. Meanwhile, optimal pH was achieved at 45-minute contact time with a value of 7.73. The findings of this study suggest that the proposed methodology could be adapted for full-scale implementation in industrial wastewater treatment facilities.

Authors Contribution

Anceu Murniati (AM) Conceptualization, Methodology, Supervision, Manuscript Review, and Editing; Ranti Nur Aprillianti (RNA) Restu Muchammad Ibrahim (RTI) Data Analysis, Manuscript Drafting; Maulida Rahayu (MR) Data Analysis; Arie Hardian (AH) Supervision, Manuscript Review and Editing.

Conflicts of Interest

There are no conflicts of interest reported by the writers.

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Data Availability statement

The data presented in this study are available on request from the corresponding authors.

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