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Electrocoagulation Technology Based on Aluminum–Zinc Electrodes as an Environmentally Friendly Method for the Removal of Cr^{3+} and Cd^{2+} from Batik Wastewater

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A B S T R A C T

Industrial batik wastewater has the potential to contain heavy metals such as Cr^{3+} and Cd^{2+} at high concentrations, which can contaminate aquatic environments due to their toxic, persistent, and non-biodegradable nature. Therefore, an effective treatment technology is required. One promising method is electrocoagulation, a redox (reduction–oxidation) process that utilizes the electrolytic dissolution of Al–Zn electrodes to produce $Al(OH)_3$ coagulant. This coagulant exhibits a high capacity to adsorb, destabilize, and precipitate heavy metal ions from solution. This study aims to analyze the effects of voltage and electrolysis time variations on the performance of the electrocoagulation process for reducing heavy metal concentrations in batik wastewater under batch operation. The electrocoagulation process was conducted at voltages of 10–50 V, with electrolysis times of 80-160 minutes and an electrode spacing of 2 cm. The optimal conditions were 50 V and 160 minutes of electrolysis, resulting in decreases in Cr^{3+} concentration from 3.78 mg/L to 0.031 mg/L, Cd^{2+} from 2.55 mg/L to 0.021 mg/L, and TSS from 203.77 mg/L to 39 mg/L. The advantages of electrocoagulation technology lie in its ability to generate coagulants without the addition of external chemicals, the formation of more stable flocs, and higher efficiency in contaminant separation.

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 batik industry is one of the textile industry subsectors that holds a strategic position in the development of Indonesia's national creative industry. Its role extends beyond contributing to economic growth and job creation, as it also serves a cultural function, preserving cultural heritage and strengthening Indonesia's national identity. In its production practices, batik is generally classified into three main types based on the manufacturing technique, namely *batik tulis* (hand-drawn batik), *batik cap* (stamped batik), and *batik printing* [1]. The advancement of production technology, particularly in stamped and printed batik, has significantly increased production capacity and consequently led to higher consumption of process chemicals. Synthetic dyes are more widely used because they are economically advantageous, exhibit high colour stability, and enable greater production efficiency than natural dyes [2]. However, synthetic dyes such as indigosol, naphthol, and indanthrene have relatively low fixation efficiency on textile fibers, resulting in a substantial portion of their residues being discharged with wastewater [3]. Batik industrial wastewater is generated at several stages of the production process, including dyeing, washing, and wax removal (pelorodan), producing effluents with complex, diverse pollutant characteristics [4].

Batik effluent may contain heavy metal ions such as Cd^{2+}, Co^{2+}, Cr^{3+}, Cu^{2+}, and Pb^{2+}, which originate from components of synthetic dyes and additives that function as colour fixatives or enhancers [5]. The presence of heavy metals in batik wastewater is not solely due to dyes. Still, it is influenced by processing stages, such as chromium-based mordanting treatments (afterchrom) for specific fibre types, impurities within textile fibres, and the use of supporting chemicals, such as salts, sodium hydroxide, and oxidizing or reducing agents. Dye compounds containing heavy metals are generally persistent in aquatic environments and may be toxic to aquatic organisms when accumulated at high concentrations [6]. Chromium and cadmium are considered high-risk heavy metals due to their reactivity and toxicity. Exposure to Cr^{6+} is highly reactive and capable of forming harmful complexes, and it has been reported to be associated with respiratory disorders and an increased risk of lung cancer. Cadmium is known to be nephrotoxic and may cause bone tissue damage under chronic exposure. Both metals are classified as carcinogenic and toxic heavy metals [7]. In addition to heavy metals, batik wastewater also contains high concentrations of Total Suspended Solids (TSS), which originate from dye particles, wax residues, fibres, and undissolved chemicals. Elevated TSS levels increase turbidity, reduce light penetration, disrupt photosynthesis, and promote sedimentation that may adversely affect aquatic habitat quality [8]. Without proper treatment, batik wastewater can cause environmental pollution and pose risks to human health.

Electrocoagulation has been widely developed as an alternative wastewater treatment method because it is considered more efficient in reducing heavy metal content while requiring relatively minimal chemical additives compared with conventional methods [9]. This process operates by using an electric current to initiate electrochemical reactions at the electrode surface, thereby generating coagulants *in situ* without the need for external chemical addition [10]. The flocs formed during the process can adsorb and bind contaminants in wastewater, which are subsequently removed by sedimentation or flotation. The performance of electrocoagulation is strongly influenced by the electrode material used, as metal ions are released through oxidation–reduction reactions driven by the electrical potential difference within the system [11].

Aluminium is one of the most widely used electrode materials in electrocoagulation processes because of its ability to release 3 equivalents of charge per mole, producing Al^{3+} ions that are highly effective in forming hydroxide flocs [12]. In addition, aluminium is relatively easy to obtain, cost-effective, and stable under various operating conditions. The use of aluminium combined with zinc has been explored to enhance system performance, as zinc offers good electrical conductivity and relatively high corrosion resistance [13]. Electrochemically, zinc has a more negative potential, which can support reaction stability and accelerate contaminant precipitation. In this system, aluminium primarily forms hydroxide flocs that trap pollutants, while zinc helps maintain current stability and optimise ion release during the treatment process [14]. Based on these considerations, this study aims to evaluate the effectiveness of electrocoagulation using aluminium–zinc electrode pairs in reducing the concentrations of heavy metal ions Cr^{3+} and Cd^{2+} in batik wastewater, and to examine changes in Total Suspended Solids (TSS) during the treatment process.

1.2. Literature Review

The working principle of electrocoagulation is based on electrochemical reactions that occur when an electric current is passed through a solution via a pair of electrodes. At the aluminium anode, oxidation occurs, causing the metal to dissolve and releasing Al^{3+} ions into the solution. These Al^{3+} ions subsequently react with hydroxide ions (OH^-) produced at the cathode, forming aluminium hydroxide ($Al(OH)_3$). This compound forms flocs with a high surface area and functions as an *in situ* coagulant. The generated flocs can adsorb and bind suspended particles and heavy metal ions in solution, forming larger aggregates that can subsequently be separated by sedimentation.

I.2.1 Aluminum and Zinc Electrodes

The selection of electrode material plays a crucial role in determining process performance, as it influences coagulant formation, system stability, and contaminant removal efficiency. Aluminium is widely used as an electrode material due to its ability to generate $Al(OH)_3$ species that act as effective *in situ* coagulants under specific operating conditions. In addition, aluminium is economical, widely available, and suitable for wastewater treatment applications [12]. In an electrocoagulation reactor, direct current (DC) induces oxidation at the anode and reduction at the cathode, producing metal ions that hydrolyze to form hydroxide flocs, which act as *in situ* coagulants. The combination of aluminum and zinc electrodes is selected because aluminum effectively generates coagulants. In contrast, zinc provides good corrosion resistance and system stability, thereby enhancing the efficiency of heavy metal removal [13].

I.2.2 Electrochemical Reactions

The reactions occurring in the system are as follows:
At the aluminum anode, an oxidation reaction occurs that releases Al^{3+} ions into the solution [15]:

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^-$$

The Al^{3+} ions formed then react with hydroxide ions to produce aluminium hydroxide:

$$Al^{3+}{}_{(aq)} + 3OH^-{}_{(aq)} \rightarrow Al(OH)_{3\,(aq)}$$

Aluminium hydroxide ($Al(OH)_3$) acts as a coagulant, adsorbing heavy metal ions in solution and facilitating the precipitation of pollutants.

In addition to metal ion reduction reactions, the cathode also undergoes water reduction, producing hydroxide ions (OH^-) and hydrogen gas (H_2)) [16]:

$$2H_2O_{(l)} + 2e^- \rightarrow 2OH^-{}_{(aq)} + H_{2(g)}$$

The hydroxide ions generated at the cathode can further react with metal ions in solution to form insoluble hydroxide compounds, which subsequently precipitate [17].

$$Cr^{3+}{}_{(aq)} + 3e^- \rightarrow Cr_{(s)}$$

$$Cr^{3+} + 3OH^-{}_{(aq)} \rightarrow Cr(OH)_{3(s)}$$

Ion kadmium (Cd^{2+}):

$$Cd^{2+}{}_{(aq)} + 3e^- \rightarrow Cd_{(s)}$$

$$Cd^{2+}{}_{(aq)} + 2OH^-{}_{(aq)} \rightarrow Cd(OH)_{2(s)}$$

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1.3. Research Objective

This study aims to analyze the effect of electrolysis time and applied voltage on the removal efficiency of Cd^{2+} and Cr^{3+} in batik wastewater using Al–Zn electrodes, as well as to evaluate its compliance with the wastewater quality standards stipulated in the Regulation of the Minister of Environment and Forestry of the Regulatiom of the Minister of Environment and Forestry of the Republic of Indonesian No. 16/MENLHK/SETJEN/KUM.1/4/2019.

2. MATERIALS AND METHODS

This study used batik wastewater originating from the dyeing and washing processes of fabric as the research sample. The electrocoagulation process was conducted using an experimental setup consisting of a batch reactor in the form of a 1000 mL glass beaker, equipped with two electrodes: aluminium as the anode and zinc as the cathode. The zinc plate was 8 mm thick, while the aluminium plate was 5 mm thick. Both electrodes were installed in parallel within the reactor, with an inter-electrode distance of 2 cm, and connected to a DC power supply via conducting cables. The electrocoagulation apparatus used in this study is illustrated in Figure 1.

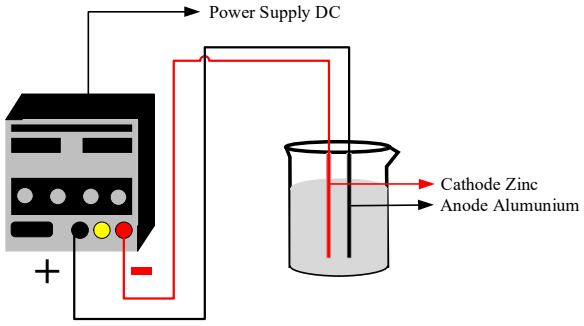

Figure 1. Electrocoagulation Apparatus Setup

This study was conducted using a batch electrocoagulation method to reduce the concentrations of heavy metal ions Cr^{3+} and Cd^{2+} as well as Total Suspended Solids (TSS) in batik wastewater. The applied voltage varied from 10 to 50 volts, and the contact time varied from 80 to 160 minutes. The distance between the electrodes was maintained constant at 2 cm throughout all experimental runs. A total of 1000 mL of batik wastewater was introduced into the batch reactor. Aluminium and zinc electrodes were installed inside the reactor and connected to a DC power supply. The voltage was adjusted according to the predetermined variations, and the electrocoagulation process was carried out according to the specified contact times.

After the electrocoagulation process was completed, the mixture was allowed to settle for 60 minutes to facilitate sedimentation, allowing the formed flocs to separate from the filtrate. The obtained filtrate was subsequently analyzed to determine the concentrations of Cr^{3+} and Cd^{2+} using Atomic Absorption Spectroscopy (AAS), while TSS levels were measured according to applicable standard testing procedures. The wastewater parameter values obtained after the electrocoagulation treatment were then compared with the textile industry wastewater quality standards.

3. RESULT AND DISCUSSION

3.1. Analysis of Batik Wastewater Characteristics

The batik industrial wastewater was analyzed for its heavy metal ion content, specifically Cr^{3+} and Cd^{2+}, using the Atomic Absorption Spectrophotometer (AAS) method. In addition, Total Suspended Solids (TSS) were measured as a supporting parameter for evaluating wastewater quality. The initial characteristics of the batik wastewater before electrocoagulation treatment are presented in Table 1.

Table 1. Initial Analysis Results of Batik Industrial Wastewater

No	Parameter	Initial Concentration (mg/L)	Quality Standard (mg/L)
1	Cr	3.78	1.0
2	Cd	2.55	0.1
3	Cu	0.013	0.5
4	Pb	0.08	0.1
5	Hg	0.021	0.01
6	Fe	0.015	2.0
7	TSS	203.77	50

(Source: BSPJI, Surabaya)

Based on Table 1, the initial characterisation results indicate that although several parameters remain within permissible limits, the concentrations of certain heavy metals exceed established standards. The chromium (Cr) concentration of 3.78 mg/L exceeds the allowable limit of 1 mg/L, while cadmium (Cd) at 2.55 mg/L surpasses the standard limit of 0.1 mg/L. Therefore, further treatment is required to reduce pollutant concentrations and minimize potential risks to aquatic environmental quality.

3.2 Effect of Voltage and Electrocoagulation Time on Cr^{3+} and Cd^{2+} Concentrations

The analysis results indicate that the concentrations of heavy metal ions Cr^{3+} and Cd^{2+} decrease with increasing voltage and electrocoagulation time. The data presented in Table 2 show that higher voltage and longer electrolysis duration result in lower residual concentrations of both metals in the wastewater.

Based on Figure 2, the concentration of Cr^{3+} decreases as the electrolysis time increases and the applied voltage becomes higher. The best condition was obtained at 50 volts and 160 minutes, where the lowest Cr^{3+} concentration reached 0.031 mg/L, which complies with the applicable wastewater quality standard

Table 2. Effect of Voltage and Electrocoagulation Time on the Final Concentrations of Cr^{3+} and Cd^{2+}

Voltage (Volt)	Electrolysis Time (min)	Final Cr^{3+} Concentration (mg/L)	Final Cd^{2+} Concentration (mg/L)
10	80	0.603	0.381
	100	0.528	0.326
	120	0.4233	0.26
	140	0.3891	0.214
	160	0.3019	0.2
20	80	0.577	0.376
	100	0.46	0.296
	120	0.391	0.25
	140	0.3345	0.173
	160	0.2709	0.148
30	80	0.571	0.367
	100	0.44	0.281
	120	0.38	0.23
	140	0.27	0.163
	160	0.1684	0.118
40	80	0.52	0.354
	100	0.422	0.266
	120	0.337	0.183
	140	0.2119	0.117
	160	0.1092	0.084
50	80	0.488	0.327
	100	0.344	0.224
	120	0.18	0.118
	140	0.096	0.066
	160	0.031	0.021

(Source: BSPJI, Surabaya)

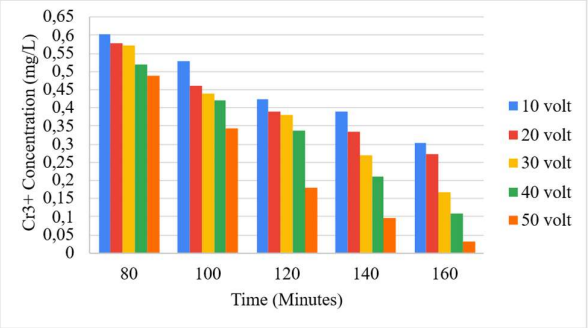

Figure 2. Relationship Between Electrolysis Time and Voltage on Cr^{3+} Concentration

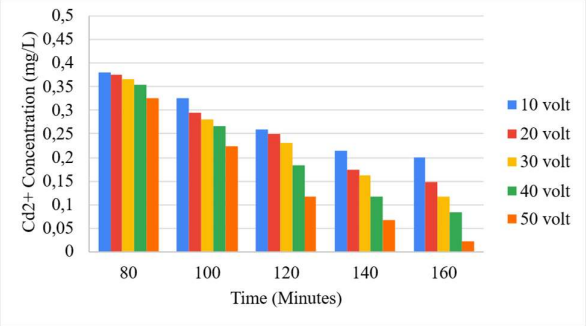

Figure 3. Relationship Between Electrolysis Time and Voltage on Cd^{2+}

Based on Figures 2 and 3, the concentrations of Cr^{3+} and Cd^{2+} decrease with increasing electrolysis time and applied voltage. The optimal conditions were observed at 50 V and 160 minutes, resulting in the lowest concentrations of Cr^{3+} (0.031 mg/L) and Cd^{2+} (0.021 mg/L), which meet the required quality standards. The concentrations of Cr^{3+} and Cd^{2+} decrease with increasing applied voltage, indicating that this parameter significantly influences electrocoagulation efficiency. Higher voltage increases the electrical current through the system, thereby accelerating oxidation–reduction reactions and enhancing the formation of $Al(OH)_3$, a coagulant that plays an important role in adsorbing and binding heavy metal ions. Similarly, the concentrations of Cr^{3+} and Cd^{2+} decrease with increasing electrolysis time, demonstrating that longer electrolysis duration also significantly affects electrocoagulation efficiency. Prolonged electrolysis allows more extensive electrochemical reactions to occur, leading to greater formation of the $Al(OH)_3$ coagulant, which can adsorb and bind heavy metal ions. These results demonstrate that electrocoagulation using aluminum–zinc electrodes is effective in reducing Cr^{3+} and Cd^{2+} concentrations to levels that comply with environmental standards.

The reduction mechanisms of Cr^{3+} and Cd^{2+} generally occur through similar electrochemical processes. At the aluminium anode, oxidation generates Al^{3+} ions, while at the cathode water reduction produces OH^- ions. The interaction between these ions forms aluminium hydroxide ($Al(OH)_3$), which acts as an in situ coagulant with an amorphous floc structure and a large surface area. These flocs play a role in adsorption and coprecipitation processes, in which heavy metal ions are either bound to the floc surface or trapped within the formed precipitates. In the case of chromium, Cr^{3+} ions react with OH^- ions to form insoluble $Cr(OH)_3$ precipitates, thereby reducing the dissolved Cr^{3+} concentration [17]. This mechanism indicates that, in addition to adsorption by $Al(OH)_3$, direct precipitation also contributes significantly to chromium removal efficiency. Meanwhile, for cadmium, the increasing concentration of OH^- ions during the process accelerates the formation of $Cd(OH)_2$ precipitates, which are subsequently trapped within the flocs and separated through sedimentation or flotation mechanisms [18].

Increasing the applied voltage results in a higher electrical current in the system, thereby enhancing dissolution at the aluminium anode and increasing the amount of Al^{3+} ions released by oxidation. This condition leads to greater coagulant formation and increases the likelihood of interactions between flocs and heavy metal ions in the solution. In addition, higher voltage also increases hydrogen gas production at the cathode, generating small bubbles within the system. These bubbles assist mixing and increase particle-floc collision frequency, thereby accelerating aggregation and flotation-based separation. Furthermore, longer electrolysis time results in greater production of Al^{3+} and OH^- ions, allowing adsorption and precipitation processes to proceed more effectively and stably.

3.3 Effect of Voltage and Electrocoagulation Time on TSS Concentration

Based on Table 3, the TSS reduction analysis was conducted at a constant electrolysis time of 160 minutes, with voltage varying from 10 to 50 volts. The results indicate that increasing the applied voltage for the same duration improves TSS removal efficiency. This phenomenon is associated with the increase in electrical current and the intensity of electrochemical reactions,

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which enhance the formation of coagulants during the process. At the final stage of treatment, the TSS concentration was reduced to below the maximum limit of 50 mg/L, as specified in the textile industry wastewater quality standard, indicating that the applied electrocoagulation process is effective.

Table 3. Effect of Voltage and Electrocoagulation Time on TSS Concentration

Voltage (Volt)	Electrolysis Time (min)	Final Concentration (mg/L)
10	160	154
20	160	124
30	160	89
40	160	45
50	160	39

(Source: BSPJI, Surabaya)

Variations in voltage and contact time in the electrocoagulation process not only influence TSS removal efficiency but also affect electrode consumption during the reaction. At the aluminium electrode, oxidation reactions produce Al^{3+} ions, which subsequently react with OH^- ions in solution to form aluminium hydroxide, $Al(OH)_3$. This compound acts as a natural coagulant, effectively binding dyes and suspended particles in wastewater. The formation of this coagulant neutralizes the charges on small particles, thereby reducing repulsive forces and promoting aggregation into larger flocs. The resulting flocs are larger and denser than the original particles, allowing them to separate more easily from the liquid phase by sedimentation at the bottom of the reactor. This floc formation mechanism becomes the primary factor in reducing TSS levels during electrocoagulation [19]. As the applied voltage increases and the electrolysis time becomes longer, the amount of Al^{3+} ions generated also increases, leading to greater formation of $Al(OH)_3$ as a coagulant. This condition enhances the system's capability to capture and precipitate suspended particles more effectively. In addition, longer contact time provides greater opportunities for particles and flocs to interact and form more stable aggregate structures. The greater the electrical charge supplied to the system, the more intense the electrochemical reactions occurring on the electrode surfaces. This increase in reaction intensity directly enhances coagulant formation and particle aggregation efficiency, resulting in a more significant reduction in TSS concentration [20].

4. CONCLUSION

Electrocoagulation using aluminum–zinc electrodes proved effective in reducing the concentrations of heavy metal ions Cr^{3+} and Cd^{2+} as well as Total Suspended Solids (TSS) in batik wastewater. The increase in applied voltage and electrolysis time significantly affected the removal efficiency, with the optimal conditions at 50 volts and 160 minutes. Under these conditions, the concentrations of Cr^{3+}, Cd^{2+}, and TSS were successfully reduced to levels that comply with the applicable environmental quality standards. These results demonstrate that electrocoagulation has strong potential as an effective method for treating batik wastewater containing heavy metals.

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