Study of the Use of Aluminum (Al) Electrodes in the Electrocoagulation Method for Reducing COD and BOD in Leachate Water

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Abstract. Electrocoagulation is an electrochemical method that uses sacrificial electrodes to remove wastewater. The most common electrode material is aluminum, which serves as both the cathode and the anode. The electrodes are connected in bipolar mode and a power source is used to provide alternating current. Various operating parameters that can influence removal efficiency were investigated, including spacing between the electrodes, applied voltage, electrolysis time, initial BOD and COD concentrations. This study examined the efficacy of employing aluminum (Al) electrodes in the electrocoagulation procedure to lower the concentration of chemical and biological oxygen demand in leachate. This research was carried out by varying the voltage (8, 10, and 12 Volts), time (10, 20, and 30 min), and distance between the electrodes (1, 2, and 3 cm). The results showed that the combination of 12 Volt voltage, 30 min, and the greatest COD reduction percentage of 80.8% was obtained with a 2 cm gap between the electrodes, while the BOD reduction reached 82.3%. This research provides important insights into the leachate pollution treatment potential of electrocoagulation using aluminum electrodes. The optimal results can be used as a basis for further development of efficient and environmentally friendly leachate treatment.

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https://doi.org/10.1051/e3sconf/202451903037

1 Introduction

Waste management has become a major concern worldwide. Waste contains both organic and inorganic pollutants. Although waste naturally undergoes decomposition, the resulting decomposition products tend to dissolve within waste piles. Consequently, a liquid known as leachate is formed. The leachate contains organic and heavy metals. Rainwater serves as a solvent agent, carrying pollutants resulting from the decomposition of waste into leachate containment ponds. If not effectively managed, the pollutants in leachate have the potential to contaminate soil, groundwater, and surface water sources around the Final Disposal Site (TPA) [1].

Water quality is typically evaluated using two popular metrics: BOD and COD. The Republic of Indonesia's Government Regulation No. 82 of 2001 covers certain water quality indicators including these criteria. Studies of BOD and COD in water are commonly used to quantify the quantity of dissolved organic matter. The organic compound's need for oxygen to decompose is determined by calculating the chemical oxygen demand of water. In contrast, BOD gauges the amount of dissolved oxygen required to oxidize or break down nearly all soluble and suspended organic compounds in water. High COD and BOD values in leachate indicate a significant level of organic pollution that must be addressed before water can be safely discharged into the environment [2].

Electrocoagulation is a water treatment method based on electrochemical principles. Anode active coagulants, which are metal ions, are released into the solution during electrolysis, whereas cathode electrolysis releases hydrogen gas. The electrocoagulation method in waste treatment was chosen because it does not require the use of chemicals, eliminating the need for excessive chemical handling steps. A key component of the electrocoagulation method is the electrode used. In previous studies, aluminum (Al) electrodes have shown potential as effective electrodes in the electrocoagulation process. Further investigation is necessary to maximize the utilization of aluminum electrodes in lowering the COD and BOD levels in leachate. This study aimed to explore the effectiveness of using aluminum electrodes (Al) in the electrocoagulation process by varying several key parameters, namely voltage, time, and electrode distance. By identifying the most effective combination of parameters, this research is expected to provide valuable insights into the development of more efficient and environmentally friendly leachate treatment technologies [3].

2 Research Methods

The study was conducted at the University Sumatera Utara's Water Quality Laboratory in the Environmental Engineering Department. This study was conducted from January 2023 to June 2023. The samples utilized in this study were leachate water obtained from the Terjun Kota Medan Landfill. Sample collection was performed by grab sampling. The final concentrations of BOD and COD in the leachate water were measured using batch processing equipment after the samples were treated using electrocoagulation. The variables in this study included voltage (8, 10, and 12 volts), time (10, 20, and 30 min), and distance (1, 2, and 3 cm).

The samples were then introduced into a 1 L glass reactor. The electrode used was an aluminum (Al) plate measuring 4 cm × 10 cm × 0.35 mm. Subsequently, an electric direct current (DC) was applied to the electrode at a predetermined voltage, time, and distance.

The anodic and cathodic reactions formed from aluminum electrodes are as follows:

Cathode reaction for Alumunium:

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

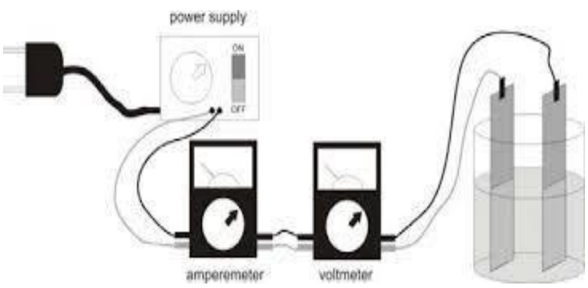
Anode reaction for Alumunium:

$$Al \rightarrow Al^{3+} + 3e^- \quad (2)$$

Overall reaction

$$2Al + 6H_2O \rightarrow 2Al(OH)_3 + 3H_2 \quad (3)$$

The leachate samples resulting from the electrocoagulation process were tested for COD and BOD levels. BOD testing was conducted according to SNI 06-6989.72:2009 on biochemical oxygen demand, whereas COD testing was performed using spectrophotometry. The testing data were then analyzed using Design Expert software with the Box–Behnken approach to ascertain how the research factors affected the effectiveness of wastewater COD and BOD reduction.



Figs. 1. Electrocoagulation Process

3 Results and Discussion

Using the Box–Behnken design (BBD) experimental design, this experiment was carried out in batch mode. Three independent variables were optimized using BBD, ensuring a more efficient experiment by utilizing fewer runs/units while accurately predicting optimum values [4]. Table 1 and 2 presents the experimental data.

Table 1. The Experimental Data (COD)

Running	Voltage (Volt)	Time (minute)	Distance (cm)	Initial COD Concentration (mg/L)	Final COD Concentration (mg/L)	Removal Efficiency (%)
1	10	10	1	1160	413,1	64,4
2	10	10	3		428,4	63,1
3	8	20	1		497,0	57,2
4	12	20	1		231,6	80,0
5	10	20	2		377,6	67,4
6	8	20	3		521,1	55,1
7	10	20	2		374,4	67,7
8	8	10	2		547,0	52,8
9	10	30	1		352,3	69,6
10	12	10	2		325,5	71,9
11	10	30	3		358,2	69,1
12	12	20	3		233,2	79,9
13	10	20	2		369,0	68,2
14	8	30	2		462,2	60,2
15	12	30	2		223,2	80,8

Table 2. The Experimental Data (BOD)

Running	Voltage (Volt)	Time (minute)	Distance (cm)	Initial BOD Concentration (mg/L)	Final BOD Concentration (mg/L)	Removal Efficiency (%)
1	10	10	1	104	43,2	58,5
2	10	10	3		44,1	57,6
3	8	20	1		46,0	55,7
4	12	20	1		20,8	80,0
5	10	20	2		41,4	60,2
6	8	20	3		54,9	47,2
7	10	20	2		40,5	61,1
8	8	10	2		59,1	43,2
9	10	30	1		30,6	70,6
10	12	10	2		30,6	70,6
11	10	30	3		33,2	68,1
12	12	20	3		21,2	79,6
13	10	20	2		40,5	61,1
14	8	30	2		45,1	56,6
15	12	30	2		18,4	82,3

Table 1 and 2 demonstrates that the 15th run, which had a 12-voltage, a 30-minute duration, and 2-centimeter distance, had maximum removal efficiencies of 80.8% and 82.3% for BOD and COD, respectively. The eighth test, which used an 8-volt voltage, a 10-minute duration, and a 2-centimeter distance, produced the lowest removal efficiencies for COD and BOD, which were 52.8% and 43.2%, respectively. The final COD concentrations obtained in the 3rd and 15th runs were below the quality standard of 300 mg/L. Furthermore, the final BOD concentrations obtained in all the runs were below the quality standard of 100 mg/L. The quality standards referred to in this study were stipulated in PERMENLHK No. 59 of 2016.

a) Response Model Selection Analysis

The analysis of response model selection was carried out to choose the statistical model most suitable for the data in the study, which was then analyzed to understand the impact of several variables on the reduction of COD and BOD levels. A number of factors, including the Model Summary Statistic, Lack of Fit, and Sequential Model Sum of Squares, must be considered when selecting a statistical model in RSM. In the Design-Expert 13 application, four types of models are available: linear, 2FI (2-factor interactions), quadratic, and cubic [5].

Tables 3 and 4 display the findings of the analysis regarding the choice of the mercury removal efficiency model with reference to the Sequential Model Sum of Squares. The accepted model based on this criterion has a P-value of less than 5% (significant) [6,7].

Table 3. Sequential Model Sum of Squares (COD parameters)

Source	**Sum of Squares**	**df**	**Mean Square**	**F-value**	**p-value**	
Mean vs Total	1003.42	1	1003.42			
Linear vs Mean	**3.94**	**3**	**1.31**	**198.39**	**< 0.0001**	**Suggested**
2FI vs Linear	0.0051	3	0.0017	0.1990	0.8942	
Quadratic vs 2FI	0.0289	3	0.0096	1.24	0.3873	
Cubic vs Quadratic	0.0377	3	0.0126	24.04	0.0402	Aliased
Residual	0.0010	2	0.0005			
Total	1007.43	15	67.16			

Table 4. Sequential Model Sum of Squares (BOD parameters)

Source	**Sum of Squares**	**df**	**Mean Square**	**F-value**	**p-value**	
Mean vs Total	944.84	1	944.84			
Linear vs Mean	7.21	3	2.40	101.61	< 0.0001	
2FI vs Linear	0.1039	3	0.0346	1.77	0.2300	
Quadratic vs 2FI	**0.1253**	**3**	**0.0418**	**6.73**	**0.0331**	**Suggested**
Cubic vs Quadratic	0.0290	3	0.0097	9.37	0.0980	Aliased
Residual	0.0021	2	0.0010			

Total	1007.43	15	67.16			

Tables 5 and 6 displays the models selected using the lack-of-fit analysis. The model selected based on lack-of-fit tests has a P-value > 0.05 (not significant), which indicates that it is appropriate for the answer [8,9].

Table 5. Lack of Fit Tests (COD Parameters)

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Linear	**0.0717**	**9**	**0.0080**	**15.22**	**0.0632**	**Suggested**
2FI	0.0666	6	0.0111	21.22	0.0457	
Quadratic	0.0377	3	0.0126	24.04	0.0402	
Cubic	0.0000	0				Aliased
Pure Error	0.0010	2	0.0005			
Linear	**0.0717**	**9**	**0.0080**	**15.22**	**0.0632**	**Suggested**
2FI	0.0666	6	0.0111	21.22	0.0457	

Table 6. Lack of Fit Tests (BOD Parameters)

Source	Sum of Squares	df	Mean Square	F-value	p-value	
Linear	0.2582	9	0.0287	27.83	0.0352	
2FI	0.1543	6	0.0257	24.95	0.0390	
Quadratic	**0.0290**	**3**	**0.0097**	**9.37**	**0.0980**	**Suggested**
Cubic	0.0000	0				Aliased
Pure Error	0.0021	2	0.0010			

Tables 7 and 8 show the model selection based on summary statistical analysis. The third study is a summary of the mathematical response model of mercury removal efficiency. The best model prioritizes the values of adjusted R^2 and forecast R^2 [10,11].

Table 7. Model Summary Statistics (COD Parameters)

Source	Standard Deviation	R²	R² Adjusted	R² Predicted	PRESS	
Linear	**0.0813**	**0.9819**	**0.9769**	**0.9636**	**0.1459**	**Suggested**
2FI	0.0920	0.9831	0.9704	0.9204	0.3192	
Quadratic	0.0881	0.9903	0.9729	0.8487	0.6063	
Cubic	0.0229	0.9997	0.9982		*	Aliased

Table 8. Model Summary Statistics (BOD Parameters)

Source	Standard Deviation	R²	R² Adjusted	R² Predicted	PRESS	
Linear	0.1538	0.9652	0.9557	0.9336	0.4960	
2FI	0.1398	0.9791	0.9634	0.9231	0.5744	
Quadratic	**0.0788**	**0.9958**	**0.9884**	**0.9373**	**0.4683**	**Suggested**
Cubic	0.0321	0.9997	0.9981		*	Aliased

b) Analysis of Variance (ANOVA)

Each independent variable's impact on the response is ascertained through the application of analysis of variance (ANOVA). The analysis of variance (ANOVA).

Table 9. Results of ANOVA Analysis for COD Parameter

Source	Standard Deviation	R²	R² Adjusted	R² Predicted	PRESS	
Model	3.94	3	1.31	198.39	< 0.0001	significant
A-Voltage	3.57	1	3.57	540.32	< 0.0001	
B-Times	0.3543	1	0.3543	53.59	< 0.0001	
C-Distance	0.0084	1	0.0084	1.27	0.2834	
Residual	0.0727	11	0.0066			
Lack of Fit	0.0717	9	0.0080	15.22	0.0632	not significant
Pure Error	0.0010	2	0.0005			
Cor Total	4.01	14				

Table 10. Coefficient of Variation for COD Parameter

Standard deviation	0.0813		**R²**	0.9819
Mean	8.18		**R² Adjusted**	0.9769
C.V. %	0.9942		**R² Predicted**	0.9636
			Adeq Precision	41.8531

Table 11. Results of ANOVA Analysis for BOD Parameter

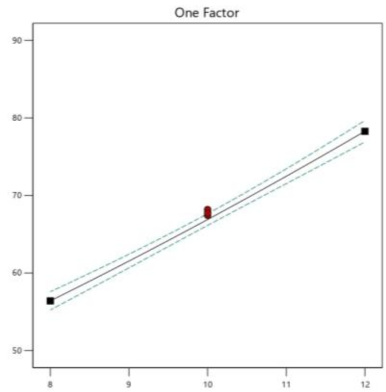
Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	7.44	9	0.8270	133.22	< 0.0001	significant
A-Voltage	5.97	1	5.97	962.39	< 0.0001	
B-Times	1.16	1	1.16	186.21	< 0.0001	
C-Distance	0.0835	1	0.0835	13.46	0.0145	
AB	0.0204	1	0.0204	3.28	0.1299	
AC	0.0816	1	0.0816	13.14	0.0151	
BC	0.0020	1	0.0020	0.3217	0.5951	
A²	0.0285	1	0.0285	4.59	0.0851	
B²	0.0003	1	0.0003	0.0468	0.8372	
C²	0.1042	1	0.1042	16.78	0.0094	
Residual	0.0310	5	0.0062			
Lack of Fit	0.0290	3	0.0097	9.37	0.0980	not significant
Pure Error	0.0021	2	0.0010			
Cor Total	7.47	14				

Table 12. Coefficient of Variation for BOD Parameter

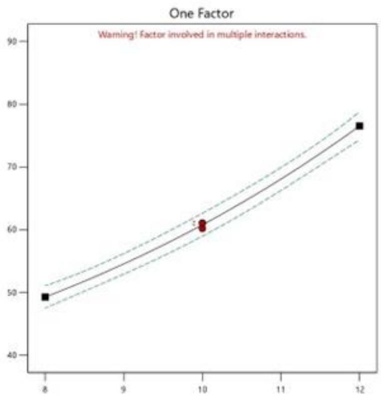
Standard deviation	0.0788		**R²**	0.9958
Mean	7.94		**R² Adjusted**	0.9884
C.V. %	0.9927		**R² Predicted**	0.9373
			Adeq Precision	38.6837

When all variables are assumed to accurately describe the variation in the dependent variable, the coefficient of determination (R^2) shows how well the data points fit the regression line. R^2 values often lie between 0 and 1. A closer R^2 value to 1 indicates a greater influence, and vice versa. If the independent and dependent variables have no relationship, the R^2 value is negative [12,13].

c) Influence of Voltage Variation on COD Removal Efficiency



d) Influence of Voltage Variation on BOD Removal Efficiency

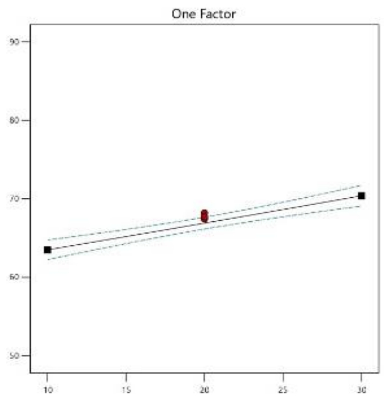


Figs 2. Graphic Analysis of the Effect of Voltage on the Efficiency of Removal COD and BOD

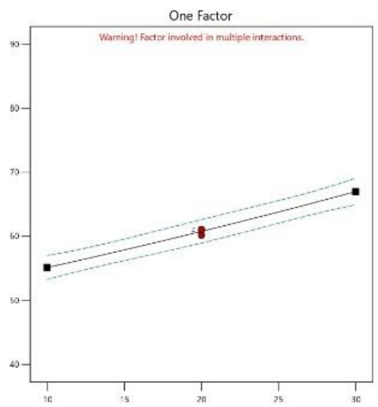
In this study, the voltage was varied as 8, 10, and 12 volts. Based on Figure 2, observations show that the efficiency of BOD and COD reduction increases with voltage. When an electric voltage was applied continuously, the electrode released additional Al^{3+}, which promoted the growth of $Al(OH)_3$ flocs. A coagulant called $Al(OH)^{3+}$ aids in the collection and absorption of various organic and inorganic pollutants present in wastewater, forming complex compounds with large molecular weights that are easily precipitated. The increased formation of precipitates leads to a decrease in the pollutant concentrations in the leachate [14]. This is because a reduction in BOD values causes a decrease in the concentration of organic compounds in the wastewater [15]. Research conducted by [16] also supports the idea that the applied voltage significantly influences the electrocoagulation process in reducing the BOD concentration in the leachate. Increasing the voltage and longer processing times affect the O and H elements produced from the oxidation reaction at the electrode, resulting in a greater capacity to reduce organic substances and COD in leachate [17]. This condition may arise because of the increase in electric voltage, leading to increased oxidation at the anode, producing Al^{3+} ions that subsequently bind to negatively charged organic substances (COD), forming large floccules that settle gravitationally, resulting in a further decrease in the COD concentration in the leachate [18].

This study is consistent with the research by [19], where increasing all three voltages resulted in smaller final concentrations of COD and BOD. During the electrocoagulation process, more flocs develop and stick to the electrode as the electric voltage increases. The elimination % attained increased with increasing applied voltage [20]. The more flocs formed, the better the electrocoagulation process [21]. At voltages of 8, 10, and 12 volts, the formed flocs float to the surface of the solution. This is because mixing occurs between the flocs and hydrogen gas produced by the reaction of the two electrodes. The electrode mass transfer and solution mixing are significantly influenced by the voltage, which also directly regulates the coagulant dose and bubble formation rate, according to [22]. Thus, to understand the connection between voltage and electrocoagulation performance, a number of studies have been carried out. According to the study findings, the efficacy of pollutant removal increases when voltage is applied. The quantity of power flowing through the electrolyte solution determines how much dissolved metal or precipitate is present. The study by [23], which claims that wastewater may have BOD and COD eliminated using the electrocoagulation procedure, also shows a similar tendency.

e) Influence of Time Variation on COD Removal Efficiency



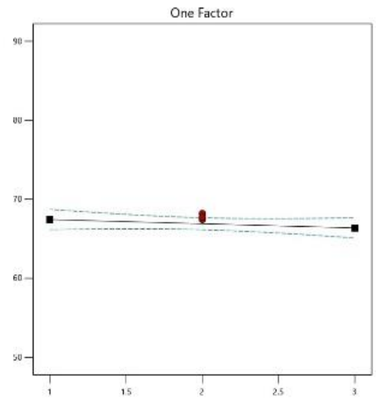
f) Influence of Time Variation on BOD Removal Efficiency



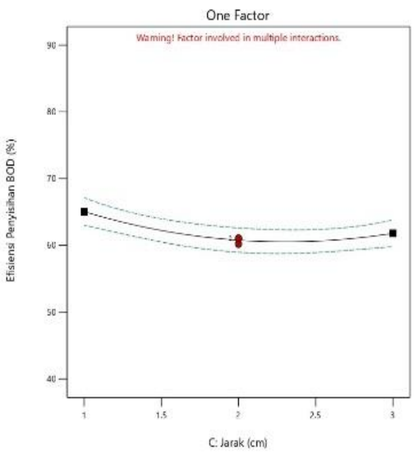
Figs 3. Graph of the Analysis of the Effect of Time on the Efficiency of COD and BOD Removal

The contact times used in this study were 10, 20, and 30 min. Figure 3 illustrates this point: the more time spent, the more effective are the COD and BOD elimination. This is because more Al^{3+} is generated when the electrocoagulation process runs longer, leading to an increased formation of $Al(OH)_3$ that binds and forms large flocs that then float on the liquid owing to the oxidation and reduction processes. Additionally, O_2 and H_2 gases were formed at the electrodes, contributing to the formation of these flocs [24]. The reduction in COD values in leachate occurs because organic materials undergo changes that make them unstable, which is induced by coagulants and the electric field during the electrocoagulation process. As a result, the bonds between organic molecules are disrupted, causing these molecules to be absorbed by coagulant flocs and then settle at the bottom of the electrocoagulation tank [25]. The decrease in BOD values occurs because positive charges attract negative ions, and when negative and positive charges meet, an attractive interaction produces a strong bond. This results in the formation of coagulants that, in turn, form flocs capable of reducing the content of organic compounds in the leachate [26].

g) Influence of Electrode Distance Variation on COD Removal Efficiency



h) Influence of Electrode Distance Variation on BOD Removal Efficiency



Figs 4. Graph of the Analysis of the Effect of Electrode Distance on the Efficiency of COD and BOD Removal

The electrode distance was varied as 1, 2, and 3 cm in this investigation. Figure 4 illustrates that the best removal efficiency occurred at an electrode distance of 2 cm. A distance of 1 cm yielded the best removal effectiveness for the BOD parameter. This suggests that the removal of BOD and COD by electrocoagulation benefits more from the reduced electrode distance. The electrical resistance in a solution is directly proportional to the distance between the electrodes, which influences the distribution of electric current in the solution [27]. As a result, the electric current may be distributed unevenly, with some areas of the solution receiving more current than others. The speed of electron transmission between the cathode, where the reduction process occurs, and the anode, which receives electrons, is also influenced by the electrode distance. Therefore, in line with the research by [28], electrocoagulation with a small electrode distance will result in maximum removal efficiency.

A decrease in the processing efficiency occurs when the distance between the electrodes is increased, causing a significant increase in the electrical resistance, leading to decreased conductivity. The interaction between molecules weakens when the distance between the electrodes exceeds 2 cm because of the suboptimal electric field, resulting in fewer electrochemical reactions to form $Al(OH)_3$ coagulants. Consequently, pollutants cannot bind to the coagulant; in other words, they are not treated [29].

4 Conclusion

The maximum percentage of COD removal was 80.8% during testing at 12 volts for 30 min at a distance of 2 cm between the electrodes, while the maximum BOD removal was 82.3%. Accordingly, the effectiveness of COD and BOD removal was significantly affected by the voltage, duration, and electrode spacing. The efficiency with which COD and BOD are removed increases with the voltage. COD and BOD removal efficiencies increased with longer electrocoagulation contact times. The effectiveness of BOD and COD removal decreased with increasing electrode distance.

5 Acknowledgments

This research work is supported by TALENTA Research Universitas Sumatera Utara in the 2022 budget Year. The date 4 August 2022, Number: 20/UN5.2.1.4/PPM/2022 which giving me an opportunity to be part of this grant

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