Study of reduction of iron (Fe), copper (Cu) and lead (Pb) concents in leachate using electrocoagulation method with aluminum (Al) electrode

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Abstract. Leachate can be defined as the liquid resulting from the decomposition of waste and infiltration of rainwater in the landfill. Leachate has unique characteristics, it has a high content of organic, metals, acids, dissolved salts and microorganisms and due to the complex leachate content it requires processing in treating leachate. Electrocoagulation is one of technology that can be applied to remove organic and heavy metal content in leachate. In this study, it was seen how the effect of voltage and contact time using aluminum electrodes on decreasing the concentration of Fe, Cu and Pb in leachate then calculating the reduction efficiency of Fe, Cu and Pb. The treatment of electric voltages given in this study were 4, 8 and 12 volts with a contact time of 15, 30 and 45 minutes. The results showed metal removal reached 0.030 mg/L, 0.007 mg/L and 0.010 mg/L and with efficiencies of 99.476%, 99.364% and 98.214% for Fe, Cu and Pb parameters. The decrease in metal content by 21.8% is influenced by voltage and 3.74% is influenced by the contact time when tested using the Pearson Correlation Test. These results indicate that the leachate treatment process with electrocoagulation processing has great efficiency.

1. Introduction
Leachate can be broadly defined as the liquid resulting from the decomposition of waste and the infiltration of rainwater in the landfill. The quality of leachate varies greatly depending on the composition of the waste, the age of the waste, the climate, and the hydrogeology [1]. Leachate has distinctive characteristics, namely high organic content, metals, acids, dissolved salts, and microorganisms. These characteristics cause leachate to be very dangerous for the environment with a fairly high potential for contamination. Leachate can seep into the soil causing soil and groundwater pollution directly because in leachate there are various organic and inorganic chemical compounds as well as a number of pathogens [2]. To reduce the potential for leachate pollution to the environment, it is necessary to take leachate processing efforts at the TPA (Final Processing Site).

The leachate treatment of most of the landfills in Indonesia uses pond system technology, namely using storage ponds, anaerobic ponds, aerobic ponds, and stabilization ponds. The weakness of the processing is the relatively long residence time, which is between 30-50 days and the pool building requires a large area of land. To overcome these problems, one alternative is electrocoagulation technology.
Electrocoagulation is a technology that can be applied to remove organic and heavy metal content contained in leachate. The principle of the electrocoagulation technique is by flowing an electric current to an electrode plate to produce ions that can act like a coagulant by binding pollutants in water [3]. According to [4] the advantages of electrocoagulation processing technology are that it has a relatively low retention time and does not require large land for processing.

The purpose of this study is to analyze the decrease of heavy metal levels in landfill leachate by using the batch system electrocoagulation method, which is then seen also the effect of the variables that will be used in the partial electrocoagulation process, namely contact time and electric voltage measurements.

2. Research Methods
This study used a batch system electrocoagulation technique with an Aluminum (Al) electrode plate mounted with a monopolar system. The electrode plates used are four plates arranged with a distance between the electrodes of 2 cm. The leachate sample is then put into the reactor which has been assembled and turned on the DC (Direct Current) source and wait for the electrocoagulation process according to variations in electric voltage of 4, 8 and 12 volts and contact time of 15, 30 and 45 minutes. This method will see a comparison of the efficiency of the contact time and voltage from the results of the research that has been done then will analyze the effect of the electric voltage and contact time partially by using the Spearman correlation test. In this study, experimental data will be presented in the form of tables and graphs.

The removal efficiency of each parameter is calculated by comparing the influent and effluent efficiency expressed in percent (%) as follows:

\[
\text{Efficiency} = \frac{C_1 - C_2}{C_1} \times 100\%
\]

\[E = \text{Efficiency}\]
\[C_1 = \text{Concentration before treatment}\]
\[C_2 = \text{Concentration after treatment (Metcalf & Eddy, 2003)}\]

3. Result and Discussion

3.1. Leachate Characteristics
The water used in this study is water sourced from the inlet of the leachate reservoir at the TPA.

| No | Parameter | Standard | Concentration |
|----|-----------|----------|---------------|
| 1  | Fe        | mg/l     | 5.72          |
| 2  | Cu        | mg/l     | 1.1           |
| 3  | Pb        | mg/l     | 0.56          |

Sources: Research result, 2020

3.2. The Effect of Electric Voltage on the Decrease of Fe, Cu and Pb Logam Metal Content
The electrocoagulation process is carried out using variations in electrical voltage ranging from 4 volts, 8 volts and 12 volts. Based on the data obtained after the stress treatment was given, there was a significant decrease in the metal content of Fe, Cu and Pb as shown in the following figure:
Figure 1. Analysis of the Effect of Voltage on 15 Minutes Contact Time

Figure 2. Analysis of the Effect of Voltage on 30 Minutes Contact Time

Figure 3. Analysis of the Effect of Voltage on 45 Minutes Contact Time
Based on the picture above, it can be seen that there was a very significant decrease as a result of the increase in electrical voltage. Based on the analysis of the data on the decrease in the concentration of metal content, it can be seen that the voltage is inversely proportional to the concentration of metals in the leachate. The higher the voltage, the lower the metal content in the leachate. It can also be seen from the graph, that there was a very significant decrease as a result of the increase in electrical voltage. This is due to the rate of ion release by the electrode and the relatively slow floc formation due to the low voltage. In accordance with Bazrafshan [6] which says that the higher the voltage, the higher the number of Al³⁺ ions produced, so at high voltages, more deposits are produced to remove contaminants. Bazrafshan [6] also said that when the voltage increases, the bubble growth rate increases and the bubble size decreases, which is beneficial for increasing the efficiency of pollutant release.

In this study, statistical data analysis was carried out using the Spearman correlation test to see the effect of partial electric voltage on the decrease in the metal content of Fe, Cu and Pb in leachate. From this analysis, it was found that the correlation between electric voltage and metal content in this study was -0.812 and it was found that the correlation between electric voltage and metal content was negative, which means that if the voltage increases, the metal content will decrease. From the magnitude of the correlation, it can be seen that 65.93% decrease in metal content is influenced by electric voltage.

3.3. Effect of Contact Time on Reduction of Fe, Cu and Pb Content

The electrocoagulation process was carried out using variations of contact time for 15, 30 and 45 minutes. Based on the data obtained after the stress treatment was given, there was a significant decrease in the metal content of Fe, Cu and Pb as shown in the graph:

![Graph showing the effect of contact time on metal content](image)

**Figure 4.** Analysis of the Effect of Contact Time at a Voltage of 4 Volt
Based on the results of the data analysis of the reduction in the concentration of the metal content above, it can be seen how the contact time can affect the decrease in the metal content in leachate. Where the contact time is directly proportional to the removal of the metal concentration in the leachate. It can also be seen from the graph, that there was a significant decrease as a result of the increase in contact time. The longer the electrocoagulation process takes place, the lower the metal content in the leachate will increase. This is due to the increasing amount of time that the ions released by the electrodes due to the electric voltage will bind to contaminants in the form of heavy metals in water when forming flocs which then settle to the bottom of the reactor.

It can be seen in Figure 6 that the decrease in Fe, Cu, and Pb levels has constant levels at 12 volts for 15, 30, and 45 minutes. According to Al-Qodah [7] pollutant levels will decrease as the electrocoagulation time increases to its maximum and optimum time. According to Khandegar (2013)[4] when the contact time is longer, floc formation occurs which can help in increasing the efficiency of pollutant removal. When the efficiency of reducing pollutant is constant or does not increase, this is because the electrocoagulation process has reached the optimum time and at the optimum time the number of flocs formed is more than sufficient to reduce pollutants.

Figure 5. Analysis of the Effect of Contact Time at a Voltage of 8 Volt

Figure 6. Analysis of the Effect of Contact Time at a Voltage of 12 Volt
In this study, statistical data analysis was carried out using the Spearman correlation test to see the effect of partial electric voltage on the decrease in the metal content of Fe, Cu and Pb in leachate. From this analysis, it was found that the correlation between contact time and metal content in this study was -0.246 and it was found that the correlation between contact time and metal content was negative, which means that if the contact time increases, the metal content will decrease. From the magnitude of the correlation, it can be seen that 6.05% decrease in metal content is influenced by contact time.

3.4. Iron (Fe) Allowance Analysis
The efficiency of removing iron (Fe) concentration in leachate at various electric voltages of 4, 8 and 12 volts with contact times of 15, 30 and 45 minutes can be seen from the following figure:

![Figure 7. Analysis of the Effect of Voltage and Contact Time on Efficiency of Fe Metal Concentration Reduction](image)

The greatest efficiency in reducing the concentration of Fe with Al electrodes was in the 12 volt voltage treatment with a contact time of 45 minutes, which was 99.47%. From Figure 7 it can be seen that there is an increase in the efficiency of decreasing the concentration of ferrous metal as the voltage and contact time increase. The greater the voltage applied during the electrocoagulation process, the reduction value and the rate of reduction of pollutant levels in water will increase. The increase in stress is directly proportional to the number and mass of flocs produced in binding contaminants in water (Setianingrum et al., 2016)[8]. It can also be seen that the longer the contact time, the more efficient the reduction of Fe metal content will be. The contact time is directly proportional to the electric voltage, where the longer the processing time and the greater the voltage applied to the electrocoagulation process, the more efficient the removal of pollutants in water will be, where more ions will be formed for contaminant binding in the water. The electrochemical process that occurs in this study produces a coagulant in the form of Al(OH)₃ which functions to bind pollutants in the form of Fe²⁺ ions which then form AlFe(OH)₃ compounds so that this can reduce iron levels contained in leachate. The reactions that occur can bind these pollutants are $\text{Al(OH)}_3(aq) + \text{Fe}^{2+}(aq) \rightarrow \text{AlFe(OH)}_3(s)$.

3.5. Copper (Cu) Allowance Analysis
The efficiency of removal of copper (Cu) concentration in leachate at various electrical voltages of 4, 8 and 12 volts with contact times of 15, 30 and 45 minutes can be seen from the following figure:
The greatest efficiency for reducing the concentration of Cu with Fe electrodes was at 12 volts with a contact time of 45 minutes, which was 99.36%. The longer the electrocoagulation process and the greater the electric voltage given, the higher the efficiency value. The greater the voltage applied to the electrode, the greater the power for the electrode, releasing Al$^{3+}$ ions from the anode by the oxidation process which will bind to OH$^-$ ions from the water reduction process at the cathode which then forms a precipitate in the form of Al(OH)$_3$ which settles to the bottom of the reactor. So, the increasing electric voltage during the electrocoagulation process, more flocs will form and stick to the cathode, in which case the redox reaction that occurs at the electrode goes well so that the efficiency of reducing pollutant levels increases. Coagulant in the form of Al(OH)$_3$ which serves to bind pollutants in the form of Cu$^{2+}$ ions which then form AlCu(OH)$_3$ compounds so that this can reduce copper levels contained in leachate. The reactions that occur for the binding of these pollutants are $Al(OH)_3(s) + Cu^{2+}(aq) \rightarrow AlCu(OH)_3(s)$

3.6. Lead (Pb) Allowance Analysis
The efficiency of removal of copper (Pb) concentration in leachate at various electrical voltages of 4, 8 and 12 volts with contact times of 15, 30 and 45 minutes can be seen from the figure below:
Figure 9. Analysis of the Effect of Voltage and Contact Time on Efficiency of Pb Metal Concentration Reduction

The greatest efficiency for reducing the concentration of Cu with Fe electrodes was at 12 volts with a contact time of 45 minutes, which was 98.21%. The efficiency of the electrocoagulation process is directly related to the concentration of hydroxide (OH⁻) and metal ions produced by the electrode, with the longer time given during processing, the binding process of hydroxy and metal ions into coagulants is increasing. Thus the level of decrease in the content of lead (Pb) in the water will increase. The electrochemical process that occurs in this study produces a coagulant in the form of Al(OH)₃ which functions to bind lead pollutants in the form of Pb²⁺ ions which then form AlPb(OH)₃ compounds so that this can reduce the levels of lead contained in leachate. The reactions that occur for the binding of these pollutants are \( \text{Al(OH)}_3(s) + \text{Pb}^{2+}(aq) \rightarrow [\text{AlPb(OH)}_3]_3(s) \).

4. Summary
Electrocoagulation is a treatment process that is quite effective in removing Fe, Cu and Pb content in leachate. The effect of electric voltage and contact time based on statistical analysis is 65.93% and 6.05%, respectively, on the decrease in metal content in leachate. The efficiency of reducing the content of heavy metals Fe, Cu and Pb by electrocoagulation method using Aluminum (Al) electrodes obtained the highest results when given an electric voltage treatment of 12 Volts with a contact time of 45 minutes, namely 99.48% for Fe, 99, 37% for Cu parameters and 98.21% for Pb parameters.

Acknowledgments
Authors wishing to acknowledge assistance or encouragement from colleagues, special work by technical staff.

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