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Dissolved Lead Removal from Soil-washing Process Using Electrocoagulation

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Soil-washing using ethylenediamine-tetraacetate (EDTA) is an effective method for the remediation of lead (Pb) contaminated land. In practice, it is necessary to manage wastewater from this remediation process. The Electrocoagulation technique is an alternative method to remove Pb from soil-washing wastewater. The primary purpose of this study is to determine optimum conditions for the electrocoagulation process of Pb removal from soil-washing wastewater. This study used an electrochemical batch reactor with a monopolar parallel circuit. Based on the initial research, the Pb concentration in soil-washing wastewater was 3600 mg/L. Several parameters were used to obtain the optimal condition for Pb removal: operational voltage, type of electrode used, and time. The result showed that the optimum condition for Pb removal is on the operational voltage of 7 volt (V), using aluminium electrode pair, at an operating time of 80 min within the Pb removal efficiency of 96%. Furthermore, the kinetics study showed the highest Pb precipitation was 0.041/min following the first-order model. Using these optimal parameters, the Pb precipitation and removal efficiencies for real soil-washing wastewater were 0.0416/min and 96.7%, respectively. The electrocoagulation method is efficient for simultaneously removing lead from polluted effluents.

Keywords: soil remediation, EDTA, electrocoagulation, lead removal.

Introduction

Pb concentration in soil-washing wastewater was generated from the remediation process of contaminated soil from smelting used batteries. Pb is an in-degradable heavy metal that can accumulate in organisms. In addition, lead is a toxic heavy metal; consequently, it will cause disease or other bodily issues. The presence of this metal in the human body can have profound effects, particularly on children, including reduced intelligence, anaemia, stunted growth, decreased immunity, autism symptoms, and even premature death (González-Grijalva et al., 2019; Fu and Xi, 2020; Balali-Mood et al., 2021). Soil-washing is a widely used soil remediation method in developed countries (Liu et al., 2020). This method is carried out because it is effective in terms of metal removal efficiency and is environmentally friendly. Due to the use of the solvent, however, this technique is expensive. The solvent that is often used for lead metal is EDTA. Therefore, wastewater is treated to reduce the use of solvents in the soil-washing process. The are several methods to remove heavy metals from soil-washing wastewater, including chemical deposition (Wang and Chen, 2019), adsorption (Futalan et al., 2019), Advance Oxidation Process (Brandão et al., 2019), and electrocoagulation (Ye et al., 2016; Xie et al., 2020). The chemical deposition method is less desirable than the other methods as it creates problems handling large amounts of sludge. Therefore, electrocoagulation becomes a preferred alternative for the soil-washing wastewater treatment method. The electrocoagulation method has several advantages, including high metal removal efficiency, relatively low cost, low sludge production, minimum chemicals, and easy operation (Ebba et al., 2022).

Electrocoagulation is an electrochemical process involving redox reactions. A chemical phenomenon related to the charge is transferred in solution or on the electrode surface. However, there is very little information on applying this method for soil-washing wastewater. In general, wastewater treatment using electrocoagulation has been tested. The removal efficiency it produces is almost perfect, where on average, the researchers show results that exceed 90% (Bouguerra et al., 2015; Khan et al., 2021; Chegeni et al., 2021). Kobya and Demirbas (2015) investigated the remediation of Zn and NaCN in acid waste from the electroplating industry with electrocoagulation. The maximum removal efficiency was 100% after 60 minutes with a current of 60 A/m^2 and a pH of 9.5. Another study by McBeath et al. (2021) tested Ar from groundwater for community clean water with As(V) almost completely removed to a final measured concentration of 0.1 µg/L. Shakers et al. (2020) used copper electrodes to remove Ni and Cr(VI) from artificial and native wastewater with parameters including pH, electrode spacing, current density, and hydraulic retention time (HRT) studied in batches and continuous modes. The removal efficiency reached 99.96% for Ni and 98% for Cr(VI) at a current intensity of 5–10 mA/cm^2, pH 9.2, and a distance between electrodes of 4 cm. On the other hand, electrocoagulation could only remove 44%-69% of Pb from wastewater after washing the soil using EDTA (Pociecha and Lestan, 2010; Pociecha and Lestan, 2012; Silva et al., 2018). The three used the original wastewater to investigate the technology they developed. Pociecha and Lestan (2010) used an Al anode and a stainless-steel cathode connected to an electric current with an intensity of 10.6V for 30 minutes; the average efficiency was only able to reduce half of the initial concentration of 170 mg/L solution. In another study, Pociecha and Lestan (2012) also tested other plate materials of the graphite type, but this time the Pb concentration in the soil used as the experimental sample reached 5,330 mg/kg. The elimination results were no better than his previous experiments. This experiment used a larger power supply and exposure time than before in alkaline conditions (pH=9.3), but this variation only obtained a 44% Pb removal. In another study, Silva et al. (2018) used a 0.1M $NaNO_3$ solution in the anodic compartment, while other cathodic supporting electrolytes, in the form of 0.1M $NaNO_3$, 0.1M EDTA, and 0.1M citric acid, were used in the catholyte reservoir. In addition, reverse polarity and direct current were used to remove Pb^{2+} from the sample. According to the study, RP and citric acid promote Pb^{2+} to migrate effectively through the soil by electromigration. However, electrocoagulation using Al electrodes to completely remove Pb^{2+} from the liquid in environmental remediation technologies waste after 60 minutes only gave an average efficiency of 69%. *Table 1* displays the specifics of the previous studies on electrocoagulation in general and wastewater from soil-washing.

Previous studies recommend investigating various factors related to electrocoagulation for wastewater from

Table 1. *Comparison of previous studies and this research*

Wastewater	Power Supply	Plat Material	Density	Initial Concentration	pH	Exposure Time	Removal Efficiency	Reference
The general use of electrocoagulation								
Synthetic solutions	30V	Al	2.67 mA/cm^2	500 mg/L	5	30 min	99%	Bouguerra et al., 2015
Synthetic solutions	-	Plexiglas	2.32 mA/cm^2	41 mg/L	6	13 min	97%	Khan et al., 2021
Synthetic solution	25V	Al	300 mA	168 mg/L	9	40 min	90%	Chegeni et al., 2021
The use of electrocoagulation for wastewater from soil-washing								
Real wastewater	10.6V	Al and stainless-steel	32 mA/cm^2	170 mg/L	7.52	30 min	53%	Pociecha and Lestan, 2010
Real wastewater	17.3V	Graphite	4.4 mA/cm^2	5330 mg/L	9.3	105 min	44%	Pociecha and Lestan 2012
Synthetic solutions	30V	Stainless steel	350 A/m^2	15 mg/L	10.0	20 min	97%	da Mota et al., 2015
Real wastewater	1.0V/cm	Graphite	-	0.1 mg/L	9.60	60 min	69%	Silva et al., 2018

soil-washing. Numerous studies have demonstrated the success of this technique in removing heavy metals from various wastewaters, allowing it to provide higher efficiency. Therefore, this study aims to obtain the optimal parameters of the electrocoagulation process to achieve high Pb removal efficiency as part of the remediation of Pb-contaminated soil. This is the first report on the optimum conditions for electrocoagulation as part of remediation for Pb-contaminated soil. Wastewater generated from the soil-washing process in East Java, Indonesia, was used as the sample in this study with an acidic pH of 3. We hypothesised that the negatively charged EDTA-heavy metal complex would absorb the flock of various positively charged Pb hydroxide monomers and polymers formed during electrocoagulation and could then be removed from the soil wash solution. First, we investigated the optimal operating voltage, operating duration, and electrode type for the electrocoagulation process of Pb removal from wastewater that forms Pb-EDTA complexes at an acidic pH. These optimum conditions were validated using actual soil-washing wastewater samples. Second, kinetic analysis was also carried out to determine the rate for the removal reaction using zero-first-second order; and first-order models.

Methods

Characteristics of Wastewater

This study obtained lead-contaminated soil from smelting used batteries in East Java. Based on the initial research, the lead concentration in the soil sample was 51 861.7 mg/kg. The soil was washed using 0.1 M EDTA with a solid-liquid ratio of 1:10 and a pH of 3. This process was performed for 3 hours at room temperature within a stirring speed of 75 rpm. The electrocoagulation method used the filtrate produced from this process for wastewater treatment samples. The lead concentration measurement in wastewater before and after the electrocoagulation process was carried out using the AA-7000 series of Atomic Absorption Spectrophotometers (AAS) made by Shimadzu Corporation (Japan). It was then validated using certified reference material (BAT) IM-16, and pH was measured using an Oakton EW-35419-23 pH meter made in Melbourne-Australia. In contrast, conductivity and TDS were calculated using a HI8732 portable conductivity TDS meter made by Hanna Instruments, USA. The data in the result and discussion section was obtained from 3 repetitions. Before being processed using the electrocoagulation method, the wastewater sample was characterised based on several parameters, including temperature, pH, conductivity, TDS, turbidity, TSS, DO, and Pb concentration, as described in *Table 2*. Based on this analysis, it was found that these parameters were above the quality standard required in the Regulation of the Environmental Minister of the Indonesian Republic No. 5 of 2014 concerning Wastewater Quality Standards. Therefore, wastewater treatment is highly needed.

Table 2. *Wastewater characterisation of soil-washing*

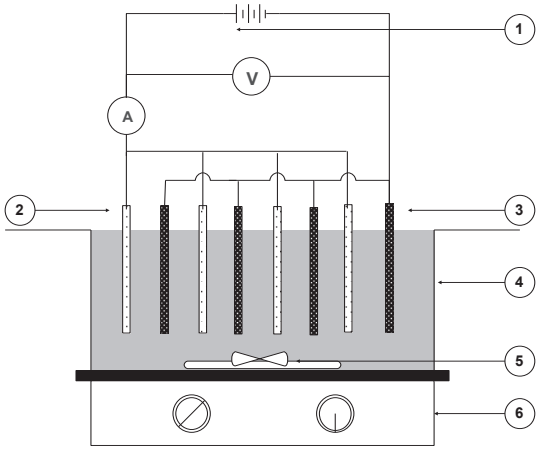
Parameter	Unit	Measured Value	[a]Standard
Temperature (T)	°C	24.8 ± 0.12	-
Turbidity	NTU	63.3 ± 1.53	-
Total Dissolve Solid (TDS)	mg/L	12 930 ± 7.07	4000
Total Suspended Solid (TSS)	mg/L	26.7 ± 0.58	400
Conductivity	mS/cm	17.6 ± 0.07	-
Potential of Hydrogen (pH)	-	3.2	6.0 - 7.0
Dissolve Oxygen (DO)	mg O_2/L	6.8 ± 0.26	-
Pb metal	mg/L	4548 ± 47.5	1.0

[a]Regulation of the Environmental Minister of the Indonesian Republic No. 5 of 2014

Electrocoagulation Set Up and Procedure

The electrocoagulation process was carried out using a batch reactor with a capacity of 1 L. The reactor is plexiglass with dimensions of 20x10x9.5 cm, as shown in *Fig. 1*. This reactor was operated with four parallel pairs of electrodes measuring 5.5x8.5x1 mm, with a distance of 1 cm between each pair. A power supply with a voltage range of 0 to 30V and a current capacity of 0 to 5A supplied the direct current (DC). In addition, the bottom of the reactor is fitted with a magnetic stirrer for constant 60 rpm stirring to homogenise the solution and reduce residual voltage.

Fig. 1. *Electrocoagulation reactor*



1. Power Supply
2. Anode Electrode
3. Cathode Electrode
4. Electrocoagulation Cell
5. Magnetic Bar-Stirrer
6. Magnetic Stirrer
A Amperemeter
V Voltmeter

The electrode plate used in this study was aluminium and graphite. Graphite is widely used due to its low cost and advantageous properties for the electrocoagulation process, including its ability to conduct electricity and high thermal conductivity. In addition, it is relatively inert and of high purity compared to other metals (Meiramkulova et al., 2020). Before use, the electrodes were washed in multiple phases, including the following: in the first stage, nitric acid and demineralised water were used to eliminate impurities from the electrode (Dönmez et al. 2017), then in the second stage, the electrodes were rinsed using acetone to remove adhering oil. In the third stage, the electrodes were dried using an oven at 105°C and weighed to obtain the initial weight before electrocoagulation. The electrocoagulation process is carried out for 2 hours, where the samples were taken at certain time intervals and then filtered using a 0.45-micron filter. After filtration, these samples were analysed to determine the Pb content using AAS. The removal efficiency (RE) is calculated using *Eq. (1)*.

$$\mathrm{RE}\ (\%) = \frac{C0 - C(t)}{C0} \times 100\% \qquad (1)$$

Where: *RE* – Pb-removal efficiency (%); C_0 – the initial Pb concentration in wastewater (mg/L); $C_{(t)}$ – the Pb concentration at a specific time t (mg/L).

This study's first step is to obtain the optimum voltage by varying working voltages of 3V, 5V, and 7V. This optimum voltage was used for the second step to determine the optimum electrode plate by varying anode-cathode types and operating times. In this study, the anode-cath-

ode types used were Aluminium (Al)-Aluminium (Al), Aluminium (Al)-Graphite(C), and Graphite(C)-Graphite(C). At the same time, the operation time varied by sets of 10, 20, 30, 40, 60, 80, 100, 120, 150, and 180 min, respectively. This optimum voltage, electrode type, and operating time are used for real wastewater treatment.

Kinetics Model

To determine the reaction rate constants of Pb removal, this study employed kinetics analysis utilising various models such as zero-order, first-order, second-order, and first pseudo-order. The kinetic model was designed to measure the decrease of Pb concentration from wastewater in a continuous reactor. The Pb removal rate constant for the zero-order model was computed using *Eq. (2)* from Metcalf and Eddy's (2003) publication, where k denotes the reaction rate constant, C represents the Pb concentration, and n indicates the order of the reaction.

$$\frac{dC}{dt} = -k.C^n \quad (2)$$

Where: *dC* – decay Pb concentration (mg/L); *dt* – the operation time (min); *k* – the reaction rate constant (min^{-1} for first order, min^{-1} for pseudo first order, and $(mg/L)^{-1}min^1$ for the second order); *n* – order reaction.

Then the first-order model and the second-order response can be calculated using *Eq. (3)* and *Eq. (4)*, respectively. The first and pseudo-order kinetics can be written in *Eq. (5)* (Ouaissa et al., 2014; Subroto et al., 2022).

$$-\ln\frac{C_{(t)}}{C_0} = k.t \quad (3)$$

$$\frac{1}{C_{(t)}} - \frac{1}{C_0} = k.t \quad (4)$$

$$-\ln\frac{q_e - q_{(t)}}{q_e - q_0} = k.t \quad (5)$$

Where: q_0 – the initial Pb concentration (mg/L); q_e – the concentration of Pb at equilibrium (mg/L); $q_{(t)}$ – the concentration of Pb at time t (mg/L).

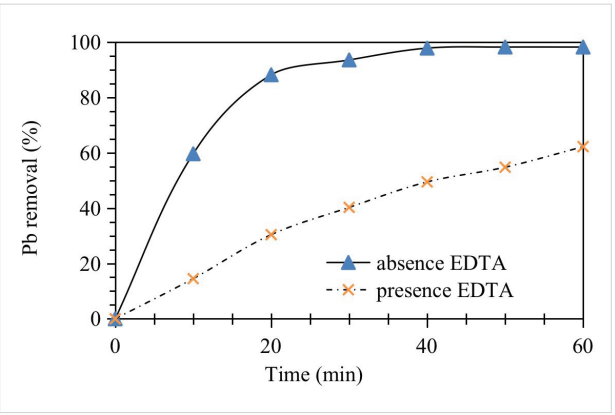
Results and Discussion

The Effect of EDTA on Desorbed Lead

EDTA is widely used for soil remediation because it can bind metal cations and has a lower physical and chemical impact on the soil matrix. However, this chelating agent will affect the Pb removal in the wastewater from the Soil-washing process. *Fig. 2* shows that the Pb removal efficiency in the presence of EDTA is lower than in the absence of EDTA. In the absence of EDTA during electrocoagulation, Pb ions rush towards the cathode surface, where Pb ions that undergo a reduction reaction are deposited on the cathode surface. As described in *Fig. 2*, after 40 min of processing, the Pb removal efficiency is constant at 98%, and the Pb removal rate is 42.228 mg/L/min. The Pb removal efficiency increases with longer operational time with EDTA. Notably, the Pb removal efficiency reaches 60% after 60 minutes of operation.

In the absence of EDTA during electrocoagulation, Pb is bounded in the demineralised water as a solvent. Demineralised water is a monodentate chelating agent compared to polydentate chelating agents, and it can act as a washing solution. In contrast, EDTA is a polydentate chelating agent, where Pb-EDTA produces a stable bond with the metal in the form of a chelate ring. The interaction of ligands and metal complexes comes from six donor atoms, which consist of 2 ni-

Fig. 2. *The effect of EDTA on lead removal*



trogen atoms and 4 oxygen atoms. It can form strong bonds in metal-EDTA complexes, especially those derived from 4 oxygen atoms from the carboxylate group (Eckert et al., 2022). Thus, the destruction of Pb in EDTA requires more time in the electrocoagulation process compared to when EDTA is not present. However, the efficiency is lower than that when EDTA is absent. The presence of EDTA in the wastewater causes Pb precipitation to occur much more slowly, at a rate of 19.294 mg/L/min.

The Effect of EDTA on Desorbed Lead

In the electrocoagulation process, operational voltage has a significant impact. As the operational voltage increased, the generated current and the dissolved electrode also increased. *Figure 3* displays the Pb removal for different operational voltages during the electrocoagulation process, indicating a significant decrease in Pb concentration with an increase in operation time of up to 80 minutes, after which the Pb reduction becomes insignificant. The Pb concentration at the optimum operation time for 3V is 2912 mg/L, while for 5V and 7V, it is 542 mg/L and 124 mg/L, respectively. An increase in operational voltage causes an increase in the oxidation of the aluminium metal. Therefore, the contaminant deposition will increase. As a result, pollutant removal will increase, as described in *Table 2*. The Pb release rate constant for different operational voltages, 3V, 5V, and 7V, is 13.522, 35.453, and 43.229 mg/L/min, respectively. The current density for the electrolysis process increased as the operational voltage increased. The current density for different operational voltages of 3V, 5V, and 7V, are 0.0027, 0.068, and 0.1 A/cm^2, respectively. The Al^{3+} ions release rate for different operational voltages of 3V, 5V, and 7V is 0.0004, 0.0105, and 0.0238 g/min, respectively. The two-way analysis of variance (ANOVA) analysis results indicate significant differences in Pb concentrations between the time and operational voltage variables with $p<0.05$.

Masthura (2020) states that the increase in deposit amount was directly proportional to the amount of oxidised metal resulting from the rise in operational voltage. The residue is produced because hydroxides are formed, which can neutralise electrostatic charges on dispersed particles and reduce the electrostatic repulsion between them, eventually reaching the van der Waals point (Kobya and Demirbas, 2015). In addition, an increase in operational voltage generated more current, forming bubbles, which indirectly affected the mixing process and electrode mass transfer (Niza et al., 2019). In addition, an increase in operational voltage causes a decrease in the bubble diameter, increasing the electrode's effective area (Koponen, 2015).

Fig. 3. *The effect of operational time on the Pb concentration*

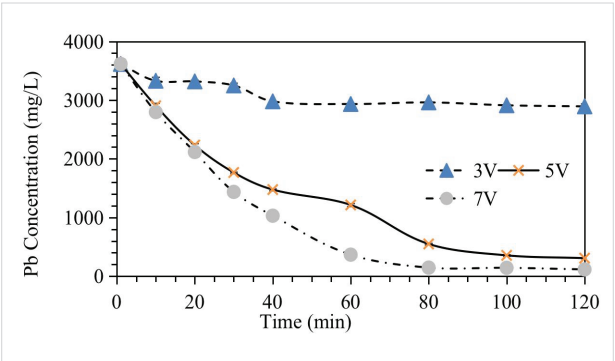


Table 2. *Pb precipitation and Al^{3+} ion release rate for different operational voltage*

Operational Voltage (V)	Pb Precipitation (mg/L/min)	Al^{3+} Ion Release Rate (g/min)
3	13.522	0.0004
5	35.453	0.0105
7	43.229	0.0238

The Effect of the Electrode Types

Figure 4 illustrates that when a pair of aluminium electrodes (Al-Al) were used, the optimal reduction in Pb concentration was achieved at 3476 mg/L after 80 minutes of operational time, with a Pb precipitation rate of 43.643 mg/L/min and a Pb removal efficiency of 96%. On the other hand, the graphite electrode pair (C-C) reached its optimal condition after 150 minutes with a Pb concentration reduction of 3460 mg/L, a Pb precipitation rate of 24.497 mg/L/min, and a Pb removal efficiency of 96%. However, when using aluminium-graphite (Al-C), electrodes did not reach its optimal condition even after 180 minutes, with only a 2600 mg/L Pb reduction. The Pb precipitation rate for the first 20 minutes is 48.175 mg/L/min, then decreased to 10.504 mg/L/min with a Pb removal efficiency of 82%. The two-way ANOVA analysis results showed significant differences in Pb concentrations for the different types of electrodes with $p<0.05$. In addition,

as shown in *Figure 4*, the Al-C electrode exhibited two consecutive slopes, with a reaction rate of 0.0202/min for the first 20 minutes, followed by a slower rate of 0.0069/min until the 30th minute until it reached equilibrium. Furthermore, *Table 3* demonstrates that the reaction rate constant for the first order was 0.0236 and 0.041/min for the C-C and Al-Al electrodes, respectively.

The current density affects the number of metal ions released by the electrode. The current density is directly proportional to the amount of metal ion dissociation in the electrolysis process (Moussa et al. 2017). However, excessively high current density leads to the wastage of electrical energy, which is why determining the optimum current density is crucial for this process. The increase in Al hydroxide due to the dissolution of the aluminium electrode is directly proportional to the current density, and this release rate can help estimate the replacement time for the electrode (Ningsih et al., 2022). The surface adsorption characteristic of the graphite electrode, the first precipitation stage, occurs without any obstacle until the saturation stage is reached. As the absorption capacity weakens, Adeogun and Balakrishnan (2017) stated that eliminating Rhodamine by using the electrocoagulation process follows two stages of absorption: surface absorption and the intra-particulate diffusion stage. Meanwhile, Graphite-Graphite (C-C) has a lower reaction rate constant than the Al-Al electrode. The graphite electrodes consist of compressed grains with a larger internal area than other metals. As a result, this electrode can absorb hydrogen or oxygen gas formed from redox reactions (Yuvaraj and Santhanaraj, 2014).

Kinetics Model

The kinetic models are observed using several models. The fitting of the k-value between actual data and the kinetic model is described in *Fig. 5a, 5b, 5c, and 5d*, respectively. It shows that the best fit of actual data and model is depicted in *Fig. 5b*, which follows the first-order kinetic model. Whereas *Fig. 5a, Fig. 5c*, and *Fig. 5d* demonstrate that several points of actual data significantly differ from the model. This result concluded that the k-value follows the first-order model, as depicted in *Fig. 5b* in this research, it was found that the Pb removal reaction rate constant varies depending on the operational voltage used. The rate constant is 0.002/min for 3V, 0.021/min for 5V, and 0.041/min for 7V.

Fig. 4. *The effect of operational time on the Pb concentration*

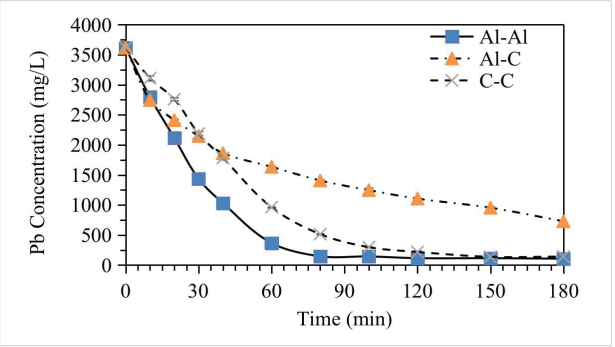


Fig. 5. *Fitting actual data and (a) zero, (b) first, (c) second, (d) first pseudo, order kinetic model*

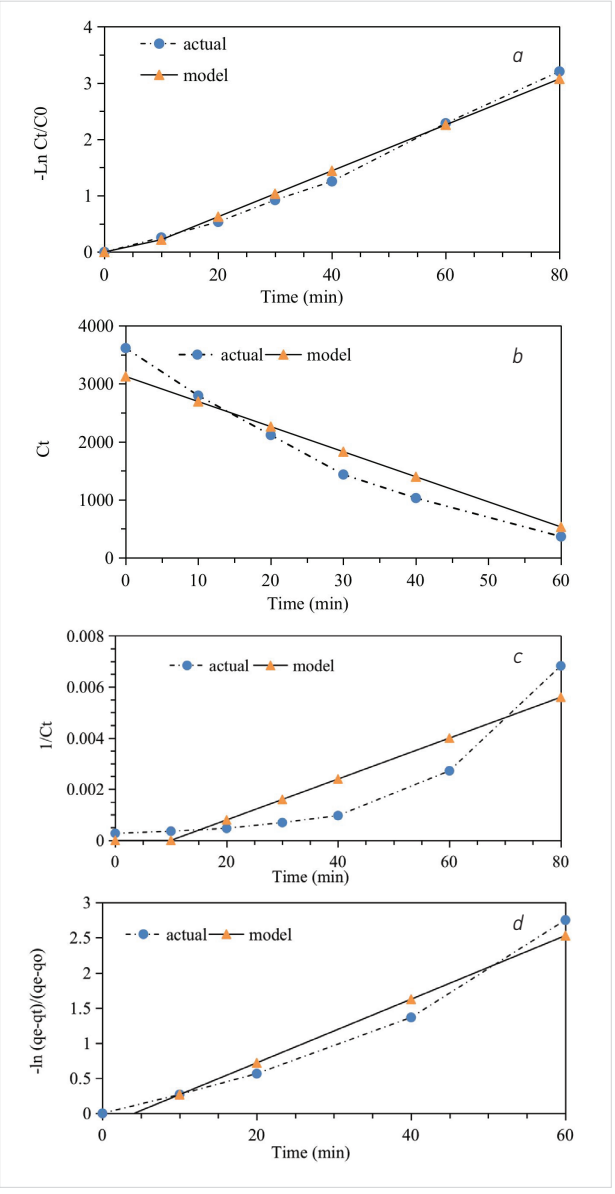


Table 3. *Example of multicolumn table format. If the title of the table is more than one row, the second row should be formatted using a hanging indent following the above limit*

Type of Electrode	Zero Order		First Order		First Pseudo Order	
	k	R^2	k	R^2	k	R^2
Al-Al	43.643	0.9344	0.0410	0.9878	0.0380	0.9816
C-C	24.497	0.8708	0.0236	0.9877	0.0250	0.9904
Al-C	48.175	0.9921	0.0202	0.9990	0.0099	0.9969
	10.504	0.9149	0.0069	0.9763	0.0075	0.9789

Application for Real Wastewater

Electrocoagulation can effectively remove Pb from Soil-washing wastewater, as Pb concentration decreases with increasing operational time due to increased H_2 gas and OH-ions (*Fig. 6*). This leads to greater Pb deposition on the electrodes and improved Pb removal efficiency. Based on the kinetic study, the reaction rate follows the first-order kinetic model and is categorised as a non-spontaneous reaction. The reaction rate is determined by one reactant, and there is no interaction with other parameters. The Pb removal process does not depend on the generation of Al, but it depends on the electrophoretic velocity of the colloidal particle towards the anode. Aluminium affects the Pb electrocoagulation process in the Soil-washing waste, but the concentration of this reactant does not contribute to how fast the reaction takes place. Similar results were shown in the study, where the process of taking Pb from the solution using solar energy electrocoagulation followed the first-order kinetic model. It indicates that a more complex wastewater composition does not affect the occurrence of colloidal destabilisation. Similar results occurred in the electrocoagulation process for the Mn, Ni and Zn metals removal in the electroplating wastewater, where a more complex composition does not affect the metal removal efficiency (Pokhrel 2017).

Fig. 6. *Pb removal as a function of the operational time for the real wastewater*

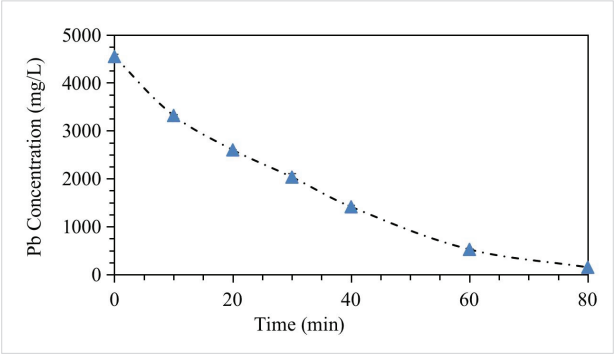


Fig. 6 illustrates how the Pb concentration changes over time during electrocoagulation. The Pb removal efficiency is shown in *Fig. 9*, and it was found to be 96.7% for an operational time of 80 min and a reaction rate constant of 0.0416/min. The Pb removal efficiency for the real soil-washing wastewater is slightly higher than that for the synthetic wastewater, with efficiencies of 96% and a reaction rate constant of 0.0407/min.

Conclusions

This study observes several parameters that significantly affect the Pb removal by the electrocoagulation process, including operational voltage, time, and electrode type. The optimum parameters for the electrocoagulation process using artificial wastewater are the working voltage of 7V, using aluminium electrode pair, at an operating time of 80 min within the Pb desorbed efficiency is 96%. The Pb removal rate constant of the EDTA complex when using artificial wastewater is 0.0407/min. This efficiency value is higher than the previous study using an electrocoagulation method. Moreover, when the optimal parameter was used for the electrocoagulation process with real wastewater from the soil-washing process, there was no significant difference in the results. The Pb removal rate constant is 0.0416/min, and the Pb removal efficiency is 96.7%, even though the wastewater has a complex composition. Based on the kinetic study, the reaction rate followed the first-order kinetic model and is categorised as a non-spontaneous reaction. Future research could investigate the same variables on other heavy metals such as Cd, Cr, Cu, Ni, and Zn to make this topic more comprehensive. It is also recommended to obtain detailed reports on energy consumption and finance to develop larger-scale models and analyse experimental data using 3 dimension models in the future.

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