



Contents list available at IJRED website

Int. Journal of Renewable Energy Development (IJRED)

Journal homepage: <http://ejournal.undip.ac.id/index.php/ijred>

Research Article

Copper and Lead Ions Removal by Electrocoagulation: Process Performance and Implications for Energy Consumption

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ABSTRACT. Electroplating wastewater contains high amount of heavy metals that can cause serious problems to humans and the environment. Therefore, it is necessary to remove heavy metals from electroplating wastewater. The aim of this research was to examine the electrocoagulation (EC) process for removing the copper (Cu) and lead (Pb) ions from wastewater using aluminum electrodes. It also analyzes the removal efficiency and energy requirement rate of the EC method for heavy metals removal from wastewater. Regarding this matter, the operational parameters of the EC process were varied, including time (20–40 min), current density (40–80 A/m²), pH (3–11), and initial concentration of heavy metals. The concentration of heavy metals ions was analyzed using the atomic absorption spectroscopy (AAS) method. The results showed that the concentration of lead and copper ions decreased with the increase in EC time. The current density was observed as a notable parameter. High current density has an effect on increasing energy consumption. On the other hand, the performance of the electrocoagulation process decreased at low pH. The higher initial concentration of heavy metals resulted in higher removal efficiency than the lower concentration. The removal efficiency of copper and lead ions was 89.88% and 98.76%, respectively, at 40 min with electrocoagulation treatment of 80 A/m² current density and pH 9. At this condition, the specific amounts of dissolved electrodes were 0.2201 kg/m³. and the energy consumption was 21.6 kWh/m³. The kinetic study showed that the removal of the ions follows the first-order model.

Keywords: Heavy metals; electrocoagulation; energy consumption; kinetics; copper; lead

Article History: Received: 16th July 2020; Revised: 15th Dec 2020; Accepted: 20th January 2021; Available online: 17th February 2021

How to Cite This Article: Prasetyaningrum, A., Ariyanti, D., Widayat, W., Jos, B. (2021) Copper and Lead Ions Removal by Electrocoagulation: Process Performance and Implications for Energy Consumption. *Int. Journal of Renewable Energy Development*, 10(3), 415-424. <https://doi.org/10.14710/ijred.2021.31665>

1. Introduction

The metal coating industry disposes of large amounts of liquid wastewater that contains a variety of hazardous and toxic heavy metals, namely alkaline cleaning agents, oil, fat, copper, chromium, nickel, cyanide, zinc, and degreasing solvents (Akbal and Camcı 2011; Hunsom *et al.* 2005; Prasetyaningrum *et al.* 2019). If they are released into the environment with no treatments, most of those metals can harm the environment. Common methods such as chemical, physical, and biological processes can be utilized to treat heavy metals present in wastewater. Those methods include the utilization of NaOH in the precipitation process and Al₂(SO₄)₃ or FeSO₄ in the coagulation process with subsequent time-consuming sedimentation, ion exchange, biosorption, precipitation, adsorption, reverse osmosis, filtration, and membrane collection (Reverberi *et al.* 2014; Adhoum *et al.* 2004; Tchobanoglous *et al.* 2003).

The precipitation method is a simple and common process to remove the heavy metals in wastewater. Basically, this method is conducted using chemical coagulants, mostly aluminum or iron salts, to convert the

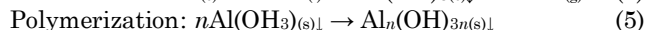
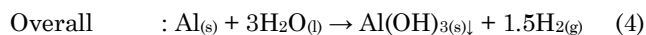
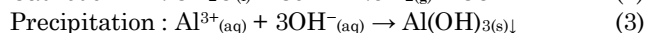
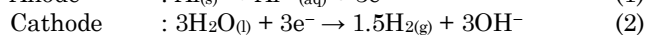
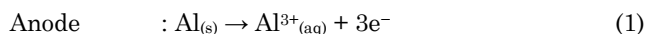
pollutants into colloidal material and regulate the pH of wastewater (Agridiotis *et al.* 2007). However, that method produces byproducts as secondary pollutants that may harm the environment (Shahedi *et al.* 2020).

One of the promising process to treat heavy metal content in the wastewater is electrocoagulation (EC). Compared to the other methods, there are several points of interest of the EC process, such as: having high-quality effluents, low energy consumption, low dissolved solids, and low sludge formation (Moradi *et al.* 2021; Samsami *et al.* 2020; Chen 2004; Zhu *et al.* 2005). In addition, it is very environmentally compatible, versatile, cost-effective (Cotillas *et al.* 2014), and provides opportunities to apply inherent safety principles (Fabiano *et al.* 2014) without executing strict safety standards (Abrahamsen *et al.* 2013).

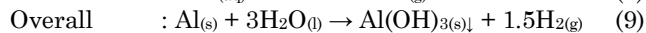
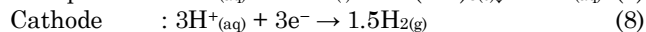
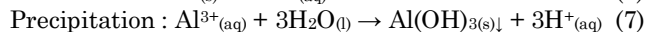
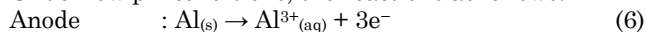
During the EC process, no chemicals were included to form coagulant. Besides, it was governed by the electric field, generally direct current (DC) charges, to the wastewater solutions through sacrificial electrodes made of iron or aluminum (Akbal and Camcı 2011; Shahedi *et al.* 2020; Chen 2004). The air is oxidized to produce oxygen gas and hydrogen ions (H⁺) due to the difference in

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electrical potential of the cathodic and anodic electrodes. Furthermore, the oxidized metal produces additional cations, with an occurrence of air reduction at the cathode to produce hydroxyl ions (OH⁻) and hydrogen gas simultaneously. Aluminum (Al) was chosen as the electrode for this study because it shows higher performance than the iron (Fe) electrode (Chen *et al.* 2018; Prasetyaningrum *et al.* 2018). The redox reaction in an aluminum electrode system under basic conditions can be summarized in the reaction equations below (Eq. (1)–(9)) (Pulkka *et al.* 2014).



Under low pH conditions, the reactions as follows:



EC process was widely applied for treatment process of various types of wastewater (Moradi *et al.* 2020; Al-Shannag *et al.* 2013), pulp and paper mill industries (Al-Shannag *et al.* 2013; Pandey and Thakur 2020), olive mills (Flores *et al.* 2018), textile processing (Bener *et al.* 2019), manufacture of potato chips (Kobya *et al.* 2006), baker's yeast production (Kobya and Delipinar 2008), and pigments industries (Martinez-Huitle and Brillas 2009). Numerous studies have proven that the EC process has high effectivity in the removal of heavy metal ions from industrial/synthesized wastewater (Zini *et al.* 2020; Akbal and Camcı 2011; Adhoum *et al.* 2004; Basha *et al.* 2008).

There are several reports on the EC process utilization in pollutants removal from wastewater; however, the research which studies the influence of process parameters on the EC process performance and energy requirements is still limited. The objective of this study is to analyze parameters that affect EC process performance and energy consumption in Cu and Pb ions removal from synthesized wastewater by utilizing the EC process. The kinetic models of Cu and Pb removal in the time of the EC process are also interesting to be explored. Furthermore, the data obtained from the reaction kinetics model and energy consumptions are useful for designing large-scale EC processes.

2. Materials and Methods

2.1. Experimental setup

The process has been carried out using synthesized wastewater containing an initial Lead (Pb) and Copper (Cu) concentration of 10.92 ppm 147.6 ppm, respectively. The initial wastewater concentration samples were referred to the composition of Pb and Cu from an industrial electro-plant located at Juwana, Pati, Central Java, Indonesia. Wastewater samples utilized in this study were prepared using Pb(NO₃)₂ (≥99.5%, Merck Cat No. 107398) and CuSO₄ (99%>, Merck Cat No. 102790).

The initial wastewater concentration samples were referred to the composition of Cu and Pb from the wastewater of industrial electroplating located at Juwana, Pati, Central Java, Indonesia.

The EC reactor for the experiments was filled with 600 mL of the wastewater solution. Aluminum electrodes were utilized as the cathodes and anodes with width, length, and thickness were 5 cm, 10 cm, and 1 mm, respectively. A power supply with direct-current (DC) and voltage ranging at 0–30 V, current density ranging at 0–8 A and controlled by a potentiostat were connected to the aluminum electrodes, at an ambient temperature of 27±1 °C. The samples (5–7 mL) were analyzed periodically. The filtrate was taken to measure the concentration of lead and copper ions. After the experiment, the EC reactor was rinsed using HCl solution and washed using distilled water. Therefore, the heavy metal ions, as well as the original and treated wastewater, were filtered before analyzing the concentrations. Atomic Absorption Spectrophotometer (AA-7000 Shimadzu) was used to measure the concentration of Cu and Pb ions in the filtrate. Moreover, the adjustment of the pH value of wastewater to the desired value was conducted using 0.1 M NaOH (E. Merck Cat. No. 104698) or 0.1 M H₂SO₄ (E. Merck Cat. No. 100317). The pH was measured using portable pH meters (HI 8424 Hanna). The experiment was conducted in triplicate to minimize experimental errors.

2.2. Characterization

Following the EC process, the sample was strained in order to separate the deposits/precipitates. The concentration of Cu and Pb ions were tested by Atomic Absorption Spectrophotometer (AA-7000, Shimadzu). Orion Star (A3225) conductivity meter was used to analyze the wastewater electrical conductivity (σ).

The percent (%) removal efficiency of copper and lead ions removal were calculated using Eq. (10) as follows:

$$\eta(\%) = \frac{C_0 - C_t}{C_0} \times 100 \quad (10)$$

where C_0 represents the concentration at initial condition, and C_t is the treated concentration of Pb and Cu ions during the EC process, respectively.

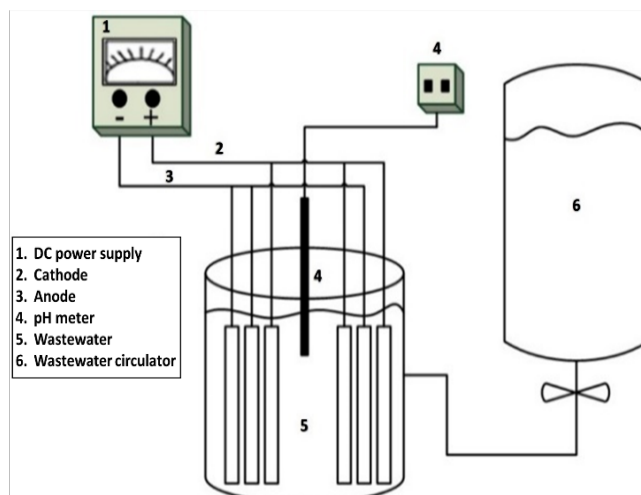


Fig. 1 The experimental setup of electrocoagulation process

The energy requirements per unit volume were calculated using Eq. (11) as follows:

$$E = \frac{P \times I \times t}{V} \tag{11}$$

where E represents the specific energy requirement (kWh/m³), t is the time of EC process (h), P represents the applied voltage (Volt), I denotes the direct current (A), and V denotes the volume of the treated wastewater (m³). The expenditure on the electrical energy requirements and the mass loss of electrodes greatly influence the total cost of wastewater treatment with the EC process.

The mass losses of the aluminum electrodes m_{Al} can be calculated using Eq. (12), which follows Faraday's law.

$$m_{Al} = \frac{I \times t \times M_{wAl}}{z_{Al} \times F \times V} \tag{12}$$

Where m_{Al} denotes the dissolved electrode (kg/m³), I denotes the direct electrical current (A), t represents the time of the EC process (s), M_{wAl} denotes the Al molecular weight (27 g/mol), z_{Al} denotes the Al chemical equivalence ($z_{Al} = 3$), F denotes the Faraday constant ($F = 96500$ C/mol), and V denotes the treated wastewater volume (m³).

2.2. Kinetics model

Two models, namely the first-order and pseudo-first-order models, were evaluated to describe the kinetic rate of Cu and Pb ions removal from wastewater. The degradation of Cu and Pb ions concentration is shown in the equation below (Eq. (13)).

$$-\frac{dC_t}{dt} = -r \tag{13}$$

Where C_t (mg/L) denotes the Cu and Pb ions concentration at EC time t (min) and $(-r)$ is the removal rate of Cu and Pb ions. In the first-order model equation, the value of $(-r)$ is described as $(-r=k_1.C_t)$, where k_1 is the kinetic rate constant of the first-order model. The integration of Eq. (13) at the boundary condition of $C_t=C_0$ at $t=0$ and $C_t=C_t$ at $t=t$ gives:

$$C_t = C_0 e^{-k_1 t} \tag{14}$$

The linearization of (Eq. (14)) gives:

$$\ln C_t = -k_1 t + \ln C_0 \tag{15}$$

By plotting Eq. (15), the value of k_1 can be obtained from the value of the slope.

For the pseudo-first-order model, the value of $(-r)$ is described as $(k_{app}(C_t - C_e))$. Therefore, Eq. (13) can be rewritten as follows:

$$-\frac{dC_t}{dt} = k_{app}(C_t - C_e) \tag{16}$$

where C_e is the Cu and Pb ions concentration at equilibrium condition. The integration of Eq. (16) with the boundary conditions of $C_t=C_0$ at $t=0$ and $C_t=C_t$ at $t=t$ is obtained as follows:

$$C(t) = C_e + (C_0 - C_e)e^{-k_{app}t} \tag{17}$$

where C_e denotes the Cu and Pb ions concentration at equilibrium, C_0 denotes the concentration at initial condition, and k_{app} represents the kinetic rate constant of the pseudo-first-order model.

3. Results and Discussions

3.1. The Effects of Parameters

3.1.1. The Effect of Current Density

Several researchers reported that one of the important parameters affecting the heavy metal ions removal using the EC process is current density (Das and Nandi 2019; Prasetyaningrum *et al.* 2019; Chen *et al.* 2018). Therefore, in this study, the current density was varied, including 40, 50, 60, 70, and 80 A/m², in order to investigate its effect on Cu and Pb ions removal. Fig. 2 shows the degradation profile of copper and lead ions during the EC process for 40 min at pH = 9 and $\sigma = 9.0$ mS/cm.

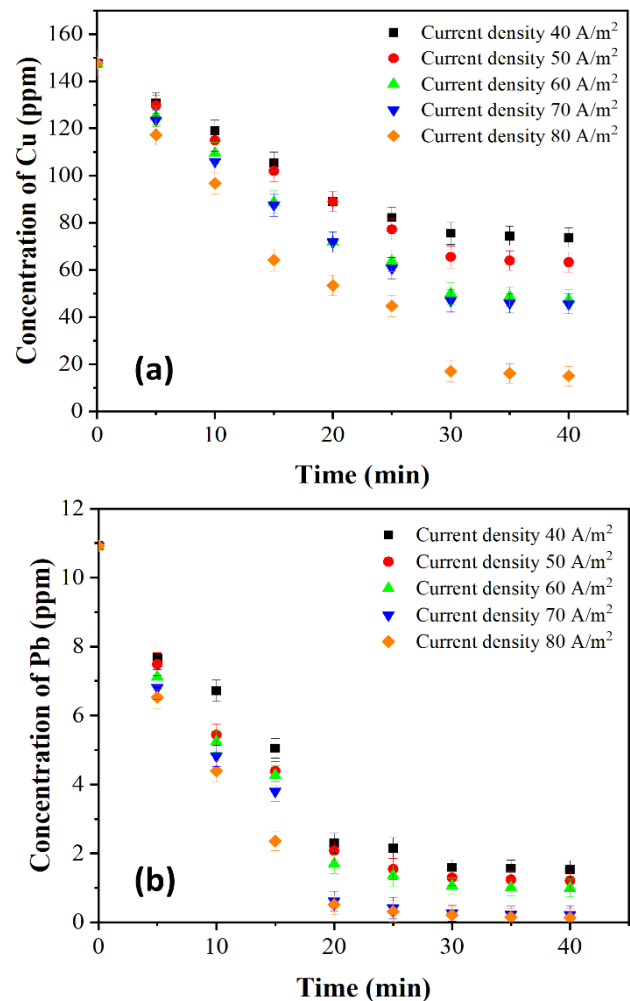


Fig. 2 Degradation profile of : (a) copper ions (b) lead ions during the EC process at a different current density

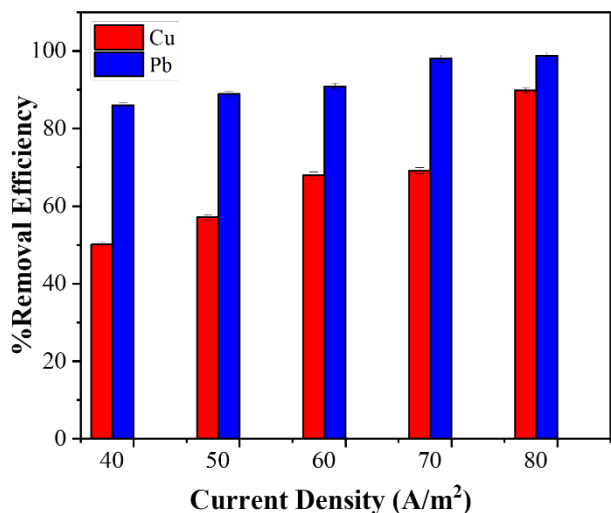


Fig. 3 The removal efficiencies of copper and lead ions at varied direct current density.

The concentration of copper ion decreases from 147.63 ppm to 73.69, 63.27, 47.32, 45.57, and 14.94 ppm at the current density of 40, 50, 60, 70, and 80 A/m², respectively. On the other hand, the lead ion concentration decreases from 10.92 ppm to 2.29, 2.07, 1.69, 0.61, and 0.50 ppm at current density of 40, 50, 60, 70, and 80 A/m², respectively.

Fig. 3 shows the influence of varied current densities values on the efficiencies of Cu and Pb removal. The Cu removal efficiencies are 50.08%, 57.14%, 67.95%, 69.135, and 89.88% for current densities 40, 50, 60, 70 and 80 A/m² respectively. In addition, the removal efficiencies of lead are 86.01%, 88.92%, 90.91%, 98.01%, and 98.76% for current densities 40, 50, 60, 70 and 80 A/m², respectively. It is found that above 70 A/m², the removal efficiency of heavy metals ions remains unchanged for further higher current densities. Thus, further studies were conducted at 80 A/m².

Since the value of current density increases, it is found that the Cu and Pb ions removal efficiencies increase. This improvement is because of the generation of coagulant (Al³⁺ ions) in a higher amount at higher current density (Das and Nandi 2019). It means that the higher the value of current density, the higher the possibility of the anode dissolution. Therefore, high amounts of metal hydroxide flocs that can increase the efficiency in removing Cu and Pb ions from the solution are formed. Consequently, the removal efficiency of Cu and Pb ions from the solution increases. Besides, increasing the current density increases the number of charged electrons that can damage the stability of the molecular bonds in the pollutants (Prasetyaningrum *et al.* 2019). Therefore, the metal ions will easily form oxide complexes. This destabilization leads to the formation of oxide complexes, which further flocculated and precipitated.

This result is following some preceding studies. Chen *et al.* (2018) identified the current density as the essential factor that can affect the EC process performance. It further specifies the quantity of Al³⁺ and OH⁻ that were formed during the EC process and also increases the current density required to remove heavy metal ions. Beyazit (2014) reported that 100% of Cu removal was obtained at a current density of 90 A/m². Hossain *et al.*

(2013) reported that, by increasing the current density about 50 to 125 A/m² could increase the removal efficiency. This improvement is caused by the ion formation in a high amount on the electrodes. These ions can promote pollutant molecules destabilization. Another research reported by Nasrullah *et al.* (2012) explained that increasing current density in sewage treatment could increase the high removal efficiency of COD, BOD, and suspended solids. Pertaining to the difference between the results in this study and other studies is due to differences in the concentration of the wastewater solution and the performance of the equipment used during the EC process.

3.1.2. The Effect of pH

Moussa *et al.* (2017) reported that pH value of the wastewater solution is one of the important parameters affecting the EC process performance. The concentration degradation profiles of Cu and Pb ions at different pH value, including 3, 5, 7, 9, and 11, are presented in Fig. 4. The current density was maintained at 80 A/m².

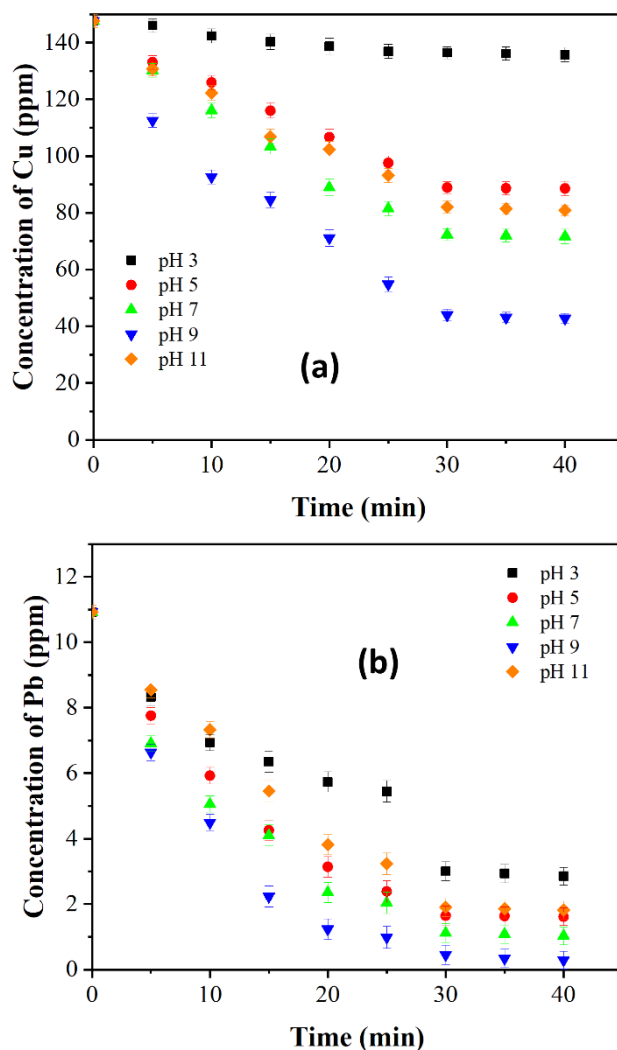


Fig. 4 Degradation profile of (a) copper ions (b) lead ions during the EC process at different pH

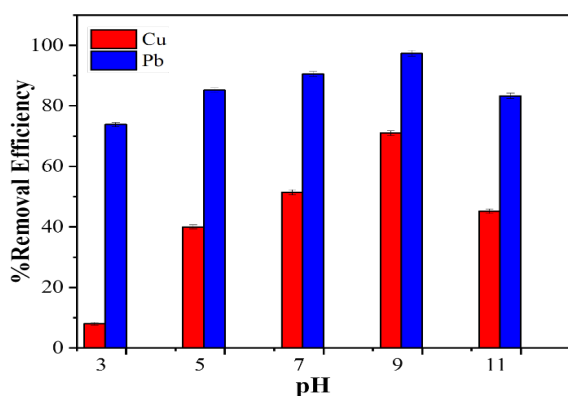


Fig. 5 Removal efficiencies of copper and lead ions at different pH

Besides, the removal efficiency of Cu and Pb ions at different pH value is shown in Fig. 5. As can be observed from Fig. 5, the pH value of wastewater significantly influences the removal of Cu and Pb ions. The Cu ion removal efficiencies are 8.02%, 40.01%, 51.44%, 71.03% and 45.21% at pH 3, 5, 7, 9 and 11, respectively. Surprisingly, the removal efficiencies of lead ions are 73.86%, 85.21%, 90.54%, 99.38% and 83.34% at pH 3, 5, 7, 9, and 11, respectively.

As shown in Fig. 5, the Cu and Pb ions removal efficiency increases as the pH of wastewater increases. This improvement is caused by the increase in OH^- species as the pH value increases. As the OH^- species increases, the metal hydroxide formation as the coagulants or flocs increases. Therefore, the copper and lead ions removal increase as the pH value of wastewater increases from 3 to 9. Kim *et al.* (2020) stated that at acidic pH, the metal hydroxide is difficult to be generated because the OH^- ion is difficult to be formed. In contrast, the formation of metal hydroxides can be easily conducted in alkaline conditions. Hence, the efficiency of heavy metals removal increases.

However, as can be observed in Fig. 5, the efficiency of Cu and Pb ions removal decreases at the further alkaline condition. At the pH of 11, the removal efficiency of copper and lead decrease. It is true since the metal hydroxides formed as the coagulants can be dissociated into the soluble metal cations at higher than pH 9 (Kim *et al.* 2020). This fact confirms that the solubility of coagulants strongly depends on the acidity of the solution (Chen *et al.* 2018). On the other hand, at pH above 9, soluble compounds, such as $[\text{Al}(\text{OH})_4]^-$, were generated and may lack the process (Cañizares *et al.* 2009; Prasetyaningrum *et al.* 2019). Other researchers also reported that pH has an influence on the EC process performance, such as in the copper removal (Kim *et al.* 2014; Yılmaz *et al.* 2008) and removal of Cu, Ni, and Mn (Al-Aji *et al.* 2012). According to Merma *et al.* (2020), $\text{Al}(\text{OH})_3$ is precipitated at the pH range of 5 and 9.5, while at pH higher than 9.5, $[\text{Al}(\text{OH})_4]^-$ is the major species. Therefore, it is suggested that the decrease in the efficiency of Cu and Pb ions removal at pH 11 is due to the dissociation of metal hydroxides and the generation of $[\text{Al}(\text{OH})_4]^-$ species which is a soluble species.

As explained before, the value wastewater pH affects the aluminum hydroxide or polyaluminum hydroxymetallic compounds formation because those compounds are produced by the reaction of OH^- at the cathode and Al^{3+} at the anode. The flocs of aluminum hydroxide attended as a coagulant agent and have high

adsorption for Cu and Pb ions so that precipitation occurs. The adsorption phenomenon is caused by the link structure that forms between colloidal particles of aluminum particles. However, it should be noted that the efficiency of Cu removal at a pH of 3 is low. It confirms that at acidic conditions, the copper removal is low. This is because of the low formation of coagulant species at the acidic condition. Within this acidic condition, the concentrations of OH^- were not adequate to form the metal hydroxide species (Kim *et al.* 2020). As a result, it is not easy to produce enough OH^- ions for metal hydroxide production in an acidic condition. In line with this finding, Merma *et al.* (2020) explained that the stable component of aluminum at a pH lower than 5 is Al^{3+} . It means that the $\text{Al}(\text{OH})_3$ as the coagulant at EC treatment is difficult to be generated. Therefore, the efficiency of Cu removal at a pH of 3 is low because of the low formation of metal hydroxide since the metal hydroxide is the coagulant for the ion removal.

Regarding the lead removal, as can be observed, the removal efficiency of Pb is high at all varied pH values and seems to be independent of the pH value. This result is obtained because the lead concentration in the solution is low enough, which is 10.92 mg.L^{-1} . On the other hand, lead ions can be precipitated at a low pH range of 1 to 5 as $\text{Pb}(\text{OH})_2$, PbCO_3 , and $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$ (Bouguerra *et al.* 2015). It is suggested that in an acidic condition, the lead ion removal is due to the precipitation process, while in the alkaline condition, the lead ion removal is due to the EC process. Therefore, the lead removal at all varied pH is high enough.

3.2. The kinetic study of the EC process

3.2.1. Kinetics Model

The Cu and Pb ions removal rates have been studied using the first-order and pseudo-first-order models kinetics equations at varied current densities and pHs. The comparison of the kinetic rate parameters of the first-order and pseudo-first-order models for the removal rates of Cu and Pb ions at different current density is depicted in Table 1, and the comparison of data and model data using the kinetic models (Eq. (14) and (17)) is depicted in Fig. 6.

The obtained values indicate that the first-order and pseudo-second-order models are suitable to describe the kinetic model of Cu and Pb ions removal in the EC process. The values show that the models were both suitable in describing the kinetics model of the Cu and Pb ions removal in the EC process with R^2 values above 0.9. However, the first-order is used when the equilibrium concentration is low. In this experiment, the C_e values of Cu ions removal at 80 A/m^2 and the C_e values of Pb ions at 70 and 80 A/m^2 were zero. When the C_e value is zero, the pseudo-first-order becomes the first-order model (Prasetyaningrum *et al.* 2020). Besides, Chen *et al.* (2018) has reported that in some cases, when the pseudo-first-order model is forcibly used, the value of C_e might be negative. Therefore, the first-order model is more appropriate to predict the copper and lead ions removal process. According to Al-Shannag *et al.* (2015), the value of kinetic rate constants varies at different current densities. Meanwhile, this research shows the different parameters in the pH.

Table 1
Kinetic parameters of copper and lead ions removal at varied current densities

Heavy metal	CD (A/m ²)	First-order		Pseudo-first-order		
		k_1	R ²	k_{app}	C_e	R ²
Cu	40	2.05×10^{-2}	0.9834	4.27×10^{-2}	52.4996	0.9863
	50	2.41×10^{-2}	0.9917	3.45×10^{-2}	28.9792	0.9915
	60	3.21×10^{-2}	0.9915	4.04×10^{-2}	16.6083	0.9910
	70	3.35×10^{-2}	0.9909	4.21×10^{-2}	16.1524	0.9924
	80	5.93×10^{-2}	0.9825	5.35×10^{-2}	0	0.9793
Pb	40	5.78×10^{-2}	0.9782	6.25×10^{-2}	0.2128	0.9730
	50	6.53×10^{-2}	0.9796	7.65×10^{-2}	0.3853	0.9882
	60	7.11×10^{-2}	0.9786	8.01×10^{-2}	0.2481	0.9845
	70	1.12×10^{-1}	0.9759	9.31×10^{-2}	0	0.9718
	80	1.23×10^{-1}	0.9835	1.04×10^{-1}	0	0.9881

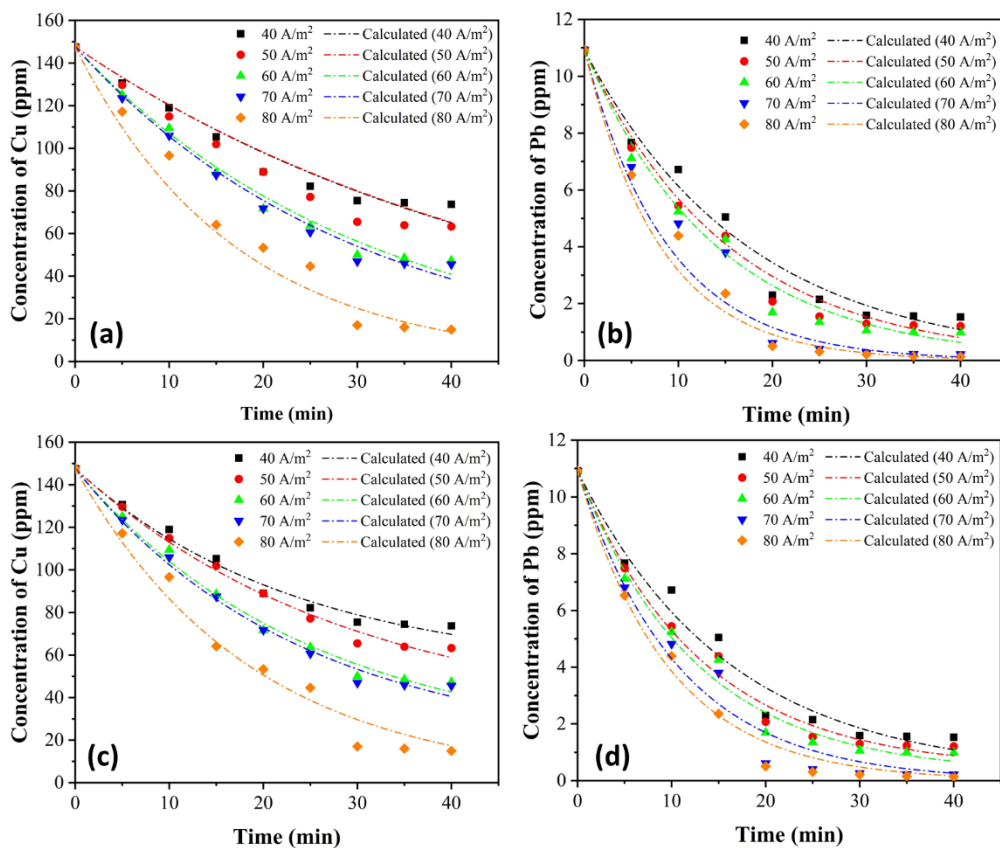


Fig. 6 The comparison of experimental and modelled data at varied current density for first-order of Cu (a) and Pb (b) and pseudo-first-order of Cu (c) and Pb (d)

Table 2
Kinetic parameters of copper and lead ions removal at different pH

Heavy metal	pH	First-order		Pseudo-first-order		
		k_1	R ²	k_{app}	C_e	R ²
Cu	3	2.53×10^{-3}	0.9710	4.51×10^{-2}	132.8654	0.9823
	5	1.49×10^{-2}	0.9872	3.41×10^{-2}	63.4878	0.9859
	7	2.13×10^{-2}	0.9847	4.36×10^{-2}	50.6771	0.9902
	9	3.58×10^{-2}	0.9870	5.73×10^{-2}	28.0163	0.9869
	11	1.74×10^{-2}	0.9874	3.89×10^{-2}	59.4721	0.9886
Pb	3	3.58×10^{-2}	0.9819	4.31×10^{-2}	1.0415	0.9534
	5	5.58×10^{-2}	0.9873	7.25×10^{-2}	0.8112	0.9974
	7	6.68×10^{-2}	0.9896	8.37×10^{-2}	0.5880	0.9920
	9	9.82×10^{-2}	0.9956	9.86×10^{-2}	0	0.9968
	11	4.96×10^{-2}	0.9905	4.86×10^{-2}	0	0.9886

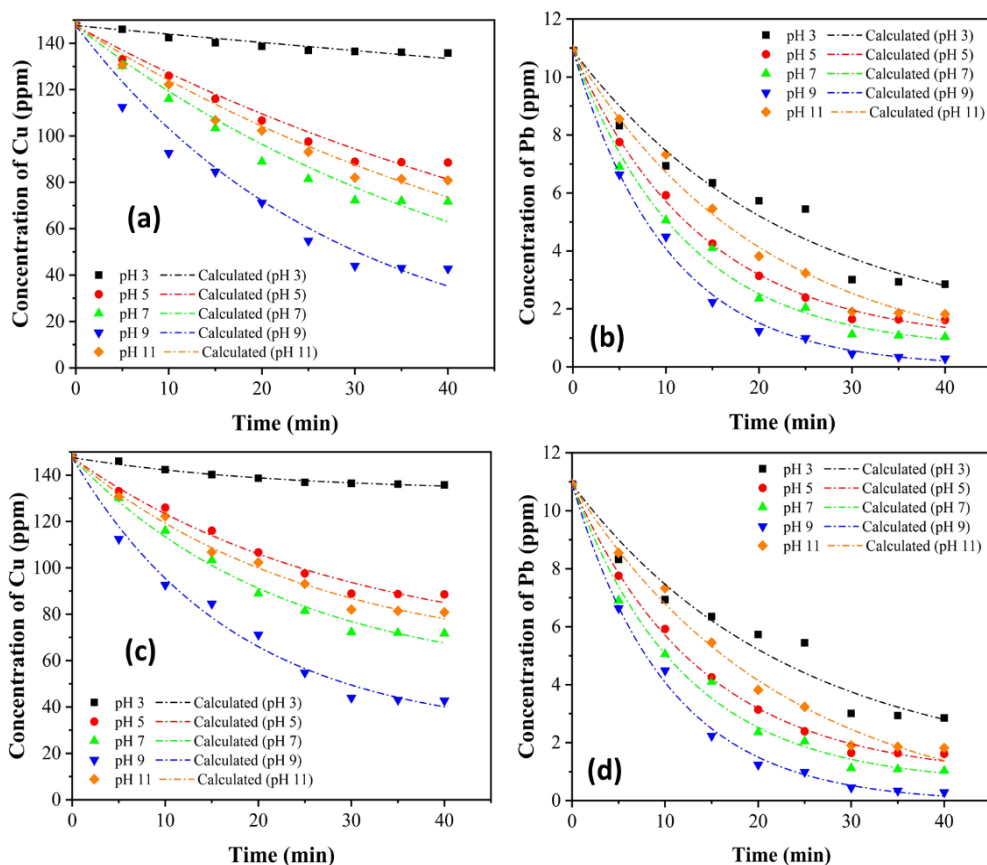


Fig. 7 The comparison of experimental and modeled data at varied pH for first-order of Cu (a) and Pb (b) and pseudo-first-order of Cu (c) and Pb (d)

Table 2 displays the kinetic rate parameters of the first-order and pseudo-first-order models at varied pH values, while the comparison of the experimental and model data using Eq. (14) and (17) is depicted in Fig. 7. This shows that the copper removal using the EC process fits properly with R² values above 0.95. On the other hand, the C_e values of lead ions removal at pH of 9 and 11 were zero. In this case, we conclude that the first-order model is more appropriate to describe the Pb ions removal process.

Some researchers modeled the kinetic rates of the EC process of metal ions removal in the adsorption step. Yilmaz *et al.* (2008) found a pseudo-second-order homogeneous reaction model for boron removal by EC using aluminum anode. Vasudevan *et al.* (2009) and Xi *et al.* (2015) found that the first and second-order models appropriately fitted with the simultaneous removal of mercury, lead, nickel, and cadmium. It is related to the varying coagulant ions properties, the pollutants nature, and the operating parameters.

3.2.2. Effect of concentration of Cu²⁺ and Pb²⁺ on the kinetic of the EC process

The correlation of kinetic rate constant and the initial concentration of Cu²⁺ and Pb²⁺ can be written as follows (Eq. (18) – (20))

$$k = f(C_{Cu}, C_{Pb}) \tag{18}$$

$$k = \alpha C_{Cu}^a C_{Pb}^b \tag{19}$$

$$\ln k = \ln \alpha + a \ln C_{Cu} + b \ln C_{Pb} \tag{20}$$

The value of *k* is the kinetic rate constant of the electrocoagulation process from the first-order model, while α , *a*, and *b* are the constants. The determination of those constants can be derived using the multiple regression analysis (MRA) as reported elsewhere (Prasetyaningrum *et al.* 2020).

The multiple regression analysis is a method that is able to determine the constant values α , *a*, and *b*. These constants are determined simultaneously. Using the MRA methods, the obtained values of α , *a* and *b* are 4.295×10⁻⁷, -0.1200, and -0.6616, respectively, with R² is 0.9477. Therefore, the overall equation for modeling the relation correlating the kinetic rate constant and the concentration of Cu²⁺ and Pb²⁺ can be written as follows (Eq. (21)).

$$k = 4.295 \times 10^{-7} C_{Cu}^{-0.1200} C_{Pb}^{-0.6616} \tag{21}$$

As can be observed from Eq. (21), the exponent value of C_{Cu} and C_{Pb} is negative. It indicates that the concentration of Cu dan Pb has a negative effect on the value of the kinetic rate constant. The higher the Cu dan Pb concentrations, the lower the kinetic rate constant value. This finding is in line with the result of the previously reported study. The initial concentration of the heavy metals is negatively proportional to the kinetic rate

constant (Prasetyaningrum *et al.* 2020). In addition, Eq. (21) shows that the exponent value of C_{Cu} is higher than the exponent value of C_{Pb} . It indicates that the value of the kinetic rate constant is more influenced by the concentration of Cu rather than be influenced by the concentration of Pb. The different concentration of Cu^{2+} and Pb^{2+} are 35, 75, 100, 147.63 ppm for Cu^{2+} and 2.5, 5, 7.5, 10.92 ppm for Pb^{2+} for removal efficiency 88.2%, 90.12%, 92.10%, 89.88% for Cu and 90.43%, 96.12%, 99.34% and 98.76% for Pb, respectively. Some research about the effect of initial concentration has been studied. Al-Aji *et al.* (2012) have reported that a high value in the initial concentration of heavy metals was reduced in relatively less time than the low concentration. The EC treatment is more effective at the beginning of the process when the concentration is relatively low. Al-Aji *et al.* (2012) have reported that using the EC process for 40 min at the current density of 8 mA/cm², the Cu ions concentrations in the wastewater were reduced from initial Cu concentration of 50, 100, 150, and 250 mg/L were reduced to 1.9, 2.55, 4, and 7.9 mg/L with a removal efficiency of 96.2%, 97.45%, 97.3%, and 96.84%, respectively. Dermentzis *et al.* (2016) have reported relatively the same results. For a high concentration, a longer time for the removal process is required, but a high initial concentration was more effective. Kim *et al.* (2014) had reported that the initial concentration of Cu and Pb ions had no effect on removal efficiency when the concentrations were 10 and 50 mg/L. Nevertheless, the value of the initial concentration had significantly affected the removal efficiency when it was 100 mg/L. Therefore, the higher value of the initial concentration results in a higher removal efficiency than the lower concentration.

3.3. Energy and Electrodes Consumptions

The dissolved electrode amount and electrical energy requirement per volume of wastewater were examined in this study. Furthermore, the correlation of removal efficiencies at different CD and EC times also studied. As shown in Table 3, the energy consumption as well as the number of dissolved electrodes depend on the electrocoagulation time and current density, as it increases along with the addition of electrocoagulation time and current density.

The application of current density from 40 to 80 A/m² will increase copper removal efficiency from 50.08 to 89.88%. However, under these conditions, energy requirements increase from 4.44 to 21.60 kW h/m³. Furthermore, the removal of lead ions increases from 86.01 to 98.76%, and the energy consumption increases from 4.44 to 21.60 at the CD of 40 to 80 A/m². At 40 min of the EC process, the number of dissolved electrodes increases from 0.1398 kg/m³ to 0.2201 kg/m³ at the current density of 40 to 80 A/m². The results of this study indicate that the current density should not be carried out below 80 mA/cm², and the time of the EC process is ranged between 20 to 40 min. In these conditions, the specific energy consumptions are ranged to the level of 10.80–21.60 kWh/m³, and the specific amounts of dissolved electrodes are ranged to the level of 0.0988–0.2201 kg/m³. The results of this study show a relationship between an increase in specific energy requirements and the dissolution of certain electrodes.

Table 3

Removal efficiencies, amount electrodes dissolved, and electrical energy requirements at varied current densities at pH 9

t (min)	CD (A/m ²)	E (kWhr/m ³)	mAl (kg/m ³)	Removal efficiency (%)	
				Cu ²⁺	Pb ²⁺
20	40	2.00	0.0654	39.76	78.98
	50	3.63	0.0768	39.76	80.98
	60	5.82	0.0844	51.40	84.48
	70	8.33	0.0951	51.40	94.41
	80	10.80	0.0988	63.86	95.37
40	40	4.44	0.1398	50.08	86.01
	50	6.80	0.1492	57.14	88.92
	60	12.71	0.1686	67.95	90.91
	70	16.33	0.1902	69.13	98.01
	80	21.60	0.2201	89.88	98.76

The electrical energy requirement and the amount of the dissolved electrode in the EC process affect the economic factors. Some studies have reported that the current density is a notable factor in EC performance. Yilmaz *et al.* (2008) reported that the energy consumption of boron removal increased with increasing current density. Chen *et al.* (2018) reported an efficient removal of Zn²⁺ with controllable current density. Meanwhile, according to Al-Shannag *et al.* (2013), the current density and electrocoagulation time can be used to control the number of specific power consumption and the specific dissolution of electrodes. By applying current density 40 A/m² and electrocoagulation time 45 minutes, the energy consumption was recorded 6.25 kWh/m³. Eventhough detailed cost analysis is needed to validate the feasibility electrocoagulation process. They reported that for 95% removal of Zn²⁺, the energy consumption was 0.25 kWh/m³ at a current density of 83 A/m². Compared to these reports, it can be seen that the energy consumption obtained in this study is different. The difference in energy consumption is due to the different operating parameters. Akbal and Camcı (2011) explained that energy consumption depends on several operating parameters, especially the electrode materials and wastewater conductivity.

It can be seen from Table 3, the higher the value of current density, the higher the value of energy consumption. The number of electrodes dissolved per unit volume also shows the same results. This data can be used to support the process design activity related to the EC for heavy metal removal.

Finally, the amount of energy and electricity requirements during the EC greatly influences the operating cost. Some researchers have developed the empirical equation to calculate the operating cost. Ozyonar and Karagozoglu (2011) calculated the operating cost with electrodes and electrical cost. They assumed that both the costs of electrode and energy consumption were taken as the main cost items expressed as follows:

$$OC = aC_{Energy} + bC_{Electrode} \quad (22)$$

where OC is the operating cost (\$), a is the unit price of electrical energy price (\$/kWh), C_{Energy} is the energy consumption (kWh), which can be calculated from Eq. (11), b is the unit price electrode material price (\$/kg), and $C_{Electrode}$ is the electrode consumption which can be

calculated from Eq. (12). More complex, Kobya *et al.* (2010) included the chemicals consumed in the process as the main cost item to the equation. Similar to the assessment result, the cost of EC process depends on the energy consumption used during the process.

4. Conclusion

This research studied the removal process of Cu and Pb ions from synthesized electroplating wastewater using a batch EC process. Current density is a significant key parameter for reducing wastewater using the EC process. The efficiency of Cu and Pb ions removal increased as the current density increased. The removal of heavy metal ions significantly improves with the residence time of the EC process and current density. The applied current density should not be higher than 80 A/m², and the EC time should be ranged at 20 to 40 min to minimize the energy requirement, even though the removal efficiency was maintained in a high number. In addition, the pH value of the wastewater should be maintained at pH 7–9 to obtain optimal removal of Cu and Pb ions. At this condition, the specific energy consumption is ranged from 10.80–21.60 kWh/m³ and the dissolved electrodes of 0.0988–0.2201 kg/m³. The correlation between the kinetic rate constant and the concentration of Cu²⁺ and Pb²⁺ is expressed as $k = 4.295 \times 10^{-7} C_{Cu}^{-0.1200} C_{Pb}^{-0.6616}$. In conclusion, the EC process can be used as one of the alternative methods for copper and lead ions removal from electroplating wastewater. Further research and development in the pilot scale size in continuous mode along with sludge management study are necessary to support the development of EC process to be implemented in the industrial electroplating wastewater treatment.

Acknowledgments

We would like to express our gratitude to Diponegoro University, Indonesia for funding research and facilities.

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