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Optimizing copper removal from synthetic water using electrocoagulation and response surface methodology

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ABSTRACT

The purification of water and wastewater requires a lot of energy and large amounts of chemicals can still be used with conventional techniques. The electrocoagulation (EC) method, an electrochemical treatment approach, has been suggested as a more cost-effective and environmentally friendly alternative. In this study, the removal of copper from synthetic water was investigated using EC technique. Box-Behnken Design (BBD) and Response Surface Methodology (RSM) were applied to optimize operating parameters such as current density, electrolysis time and initial pH. Analysis of variance (ANOVA) was used to assess the effect of factors and their interactions, and multiple regression analysis was used to fit it to a second-order polynomial equation. According to the results, current density had the greatest impact on copper removal. A current density of 7.24 mA/cm², a reaction time of 27.43 minutes, and an initial pH value of 7.56 were determined to be optimal conditions. Under these optimal conditions, the copper removal efficiency was 97.5%. Therefore, EC combines with RSM is an efficient treatment approach for copper-contaminated water.

Keywords: Wastewater treatment, Heavy metals, Electrochemical methods, Atomic absorption spectrophotometry (AAS), Box-Behnken Design (BBD), Response Surface Methodology (RSM).

INTRODUCTION

Copper (Cu) is one of the most important elements and frequently used metals in various industrial and agricultural applications (Horstkotte et al., 2012 ; Al-Saydeh et al., 2017). This widespread use of copper has resulted in its endless presence (Sharma et al., 2009) in ground and surface water, mainly through releases from industrial effluents, posing a serious environmental problem (Abdelaziz et al., 2022).

Copper is an important nutrient, but excessive amounts can lead to stomach and digestive issues, liver and kidney damage, and anaemia (Pandey & Madhuri, 2014). Furthermore, copper is not biodegradable,

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poisonous, and easy to aggregate at low levels in living organic entities in general and the human body. This can lead to serious diseases such as cancer, nerve damage, and kidney failure, and can even be fatal in high concentrations (Al-Saydeh et al., 2017).

To date, numerous physicochemical treatment techniques are employed to remove copper pollution in water. These methods include adsorption (Gros et al., 2011) using natural and modified adsorbents, cementation (Nassef and El-Twaeel, 2015), membrane filtration (Ferrer et al., 2016), electrochemical methods (Caprarescu et al., 2015; Dong et al., 2017), and photocatalysis (Kanakaraju et al., 2017). Each treatment technique has benefits but also shows some disadvantages. For example, adsorption, although very efficient and inexpensive, limits the concentration of copper ions, while membrane filtration, although clean, can be expensive; likewise, photocatalysis is simple and environmentally friendly, but time-consuming (Ab Hamid et al., 2022)

Electrocoagulation has been widely used to remove heavy metals, including Cu, from industrial effluent (Akbal and Camcı, 2010; Rincón and La Motta, 2014). Recently, EC has attracted more attention due to the following advantages: high-quality effluents, low energy consumption, low-dissolved solids, and low sludge formation (Chen, 2004; Moradi et al., 2021). In addition, it is very environmentally compatible, versatile, and 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). EC removes copper ions from water using electrical current and is based on the generation of coagulant in situ by dissolving a metal anode made of Al, Fe, or a hybrid Al/Fe electrode. The anode generates metal ions, while the cathode produces hydrogen gas, which can help drive the agglomerate particles out of the water (Ab Hamid et al., 2022). The main reactions occurring in the EC process for iron electrodes are given in Equations 1-5.

At the electrodes

Oxidation: $\mathrm{Fe_s \rightarrow Fe^{2+}_{(aq)} + 2e^-}$ (1)

Water reduction: $\mathrm{2H_2O + 2e^- \rightarrow H_2 + 2OH^-}$ (2)

Within the solution

$\mathrm{Fe^{2+} + O_2 + 2H_2O \rightarrow Fe^{3+} + 4OH^-}$ (3)

$\mathrm{Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2}$ (4)

$\mathrm{4Fe(OH)_2 + O_2 + 2H_2O \rightarrow 4Fe(OH)_3}$ (5)

The $\mathrm{Fe(OH)_2}$ and $\mathrm{Fe(OH)_3}$ species are advantageous for the rapid adsorption of pollutants due to their large specific surface areas (Ano et al., 2020; Drogui et al., 2007). The metal ions are removed from water by precipitation, co-precipitation and primarily through adsorption on iron hydroxides (Meunier et al., 2006).

Previous studies used EC to assess the effect of physicochemical parameters on copper reduction in aqueous solutions (Kim et al., 2020; Vasudevan et al., 2012a). However, these studies were mainly concerned with the effect of a single factor, so they could not show the interactions of the parameters on copper reduction in aqueous systems. This gap in scientific knowledge could be overcome by using Response Surface Methodology (RSM). RSM use mathematical and statistical tools to simultaneously assess the combined effects of multiple factors on a response variable (Anderson and Whitcomb, 2016; Montgomery, 2017). An effective understanding of the correlation of several experimental factors and their influence on copper reduction under limited experimental conditions is likely when RSM is used (Anderson and Whitcomb, 2016).

This study aims to evaluate the performance of the electrocoagulation (EC) process in treating copper-contaminated synthetic water through an experimental design approach. The specific objectives of this research are as follows: i) to develop a model for copper removal and explore the influence of factors (initial pH, current density, and electrolysis time) on treatment efficacy using the Box-Behnken design (BBD), ii) to identify the optimal conditions that result in enhanced copper reduction.

MATERIALS AND METHODS

Preparation of copper solution

A stock solution of 1000 mg/L copper solution was synthesized by dissolving approximately 2.51 g of copper (II) sulfate ($CuSO_4$) in 1000 mL distilled water. The stock solution was diluted to make 5 mg/L synthetic solutions of copper to be treated in the reactor for removal experiments.

Batch experimentation and analysis

The removal of copper was performed in a 1,25 L laboratory-scale EC system (Figure 1) consisting of a pair of iron electrodes and a DC power supply. Before each run, both electrodes were washed with a brush prior immersed in HCl (0.1 N) solution to remove compounds deposited on the surface of the electrodes, then rinsed with tap water. The pH of the solution was adjusted to the desired level with hydrochloric acid or sodium hydroxide. During EC experiments, the solution was agitated continuously using a magnetic stirrer at 120 rpm. The operation started when the current density was adjusted to a desired value and stopped with the power generator turned off. After the reaction, the water was collected and filtered to remove the flocs. The copper concentration in the filtered water was determined using atomic absorption spectrophotometry (AAS). The copper removal rate Y (%) was calculated as described in Equation (1).

$$Y(\%) = \left(1 - \frac{\mathrm{C}}{C_0}\right) \times 100 \qquad (6)$$

Experimental design

Box–Behnken Design was used to determine the optimal copper removal percentage. A total of 15 experiments were performed to investigate the effect of initial pH, current density, and electrolysis time at three levels (-1, 0, +1) (Table 1) on the percentage of Cu(II) removal. These three factors labelled A, B, and C respectively, were investigated at three levels (− 1, 0, + 1) as shown in Table 1.

The EC experiments were conducted at ambient temperature with a fixed interelectrode distance of 1 cm, conductivity of (2.3 mS/cm) and 120 rpm, and the remaining concentration of Cu(II) ions was then determined as previously described. The following equation is a second-order polynomial equation that includes the independent factors and the dependent response.

$$Y = \beta_0 + \sum\beta_i X_i + \sum\beta_{ii} X_i^2 + \sum\beta_{ij} X_i X_j + \varepsilon \qquad (7)$$

Where Y is the expected response, β_0 is the intercept term, β_i, β_{ii} and β_{ij} are the linear, quadratic, and interaction impacts, respectively, X_i and X_j are the independent variables and ε is the error. Design Expert software was used for the statistical analysis.

Statistical analysis and model development

Statistical analysis of the experimental data and analysis of variance (ANOVA) were performed using Design Expert software. The interactions between the independent variables and the responses were determined by the ANOVA test. The coefficient of determination (R^2) was used to express the quality of the fitted polynomial model and its statistical significance was checked by Fisher's F-test in the same software. The model conditions were used by the probe. > F (P-value: probability) with 95% confidence level. Based on the effects of the independent factors, three-dimensional plots and their respective contour plots were constructed. To assess the prediction accuracy, the coefficient of variation (CV) was calculated using Equation 8:

$$\mathrm{CV} = \frac{\mathrm{Y_{exp}} - \mathrm{Y_{cal}}}{\mathrm{Y_{exp}}} \times 100 \qquad (8)$$

Figure 1: Experimental Set-up for Electrocoagulation. (1) magnetic stirrer, (2) DC power supply, (3) wires, (4) electrode, (5) Electrocoagulation cell, (6) magnetic stirrer bar.

Table 1: Experimental factors and their levels for the reduction of Cu(II) using EC.

Variables	**Units**	**Factors**	**Levels**		
			− 1	**0**	**+ 1**
pH		A	5	7	9
Current density	mA/cm^2	B	4	6	8
Time	min	C	10	20	30

RESULTS

Model fitting

To study copper removal from synthetic solutions, a quadratic model was developed using regression analysis and regression coefficient estimation based on data from a Box-Behnken Design matrix (BBD). The model considered three process parameters: pH, electrolysis time, and current, coded as A, B, and C, respectively.

The results of Box Benhken design experiments for studying the effects of three independent variables on copper removal percentage are presented in Table 2 along with the predicted and observed values.

The following equation represents the quadratic model describing the response function with regression coefficients for copper removal from synthetic solution:

$$\mathrm{Y} = 95.4 + 2.185\mathrm{A} + 6.011\mathrm{B} + 3.744\mathrm{C} - 1.2\mathrm{AB} + 1.04\mathrm{AC} - 2.173\mathrm{BC} - 2.696\mathrm{A}^2 - 0.154B^2 - 2.274\mathrm{C}^2 \quad (9)$$

Where Y is the removal efficiency percentage, A the pH, B the current density (mA/cm^2) and C the reaction time (min).

Analysis of variance (ANOVA)

Statistical Fisher's test using ANOVA was performed to evaluate the significance of the quadratic polynomial model. The ANOVA results, presented in Table 3, show that the F value of 29.67 for the lack of fit implies that it is not significant relative to pure experimental error. The non-significant lack of fit is also good as the primary goal was that the model should fit the experimental data model obtained and was able to provide a good estimate of the response of the system in the studied area. Table 3 also shows that the variable pH (A), current density (B), time (C), the interaction term BC, and quadratic terms A^2 and C^2 are all significant model terms ($P < 0.05$) in the process.

Figure 2 illustrates the relationship between the predicted and observed values and R^2 of the linear plot was 0,982. The regression analysis resulted in a coefficient of determination R^2) value of 0.982, indicating that the model does not explain only 1.8% of the total variation. The adjusted determination coefficient (AdjR^2=0,948) was also high, implying that the model has high significance. The coefficient of variation (C.V.) was 1.49%, indicating that the experiments were more precise.

Effect of Process Variables on the copper removal

The effect of the three factors on the response variable is shown in Figure 3. Unlike the traditional trial and error method, the prediction profiler provides an efficient way to change one variable and keep others constant to study the individual impact on copper removal efficiency.

Current density, pH and reaction time are important parameters in the electrocoagulation process, each playing an important role in treatment efficiency and contaminant removal. The main effects of the three factors (pH, current density, and reaction time) on copper removal are shown in Figure 4. Figure 4a shows the relationship between pH and Cu(II) removal. A positive correlation is evident, wherein an increase in pH corresponds to a higher percentage of copper removal. At a pH of 8 the maximum of copper was eliminated, reaching 96.8%.

Figure 4b shows the influence of current density on copper removal. The trend indicates an increase in copper removal percentage with increasing current density with a maximum copper elimination rate of 100% achieved at a current density of 8 mA/cm^2.

Figure 4c shows a direct correlation between reaction time and copper removal percentage. Higher response times correspond to a higher percentage of copper removal. It is noteworthy that the elimination rate peaks at 96.9% after a treatment period of 20 minutes

Pareto analysis (depicted in Figure 3) indicates the contribution of each factor to copper removal. Current density holds the most significant influence (58.13%), followed by reaction time (22.545%), while initial pH's effect is relatively minor (7.68%). This analysis offers insights into the relative importance of factors in the EC process for copper removal.

Optimization of Process Parameters

Response Surface and contour Plots

The individual effect contributed by each main variable; the response was also influenced by the interaction variables. To gain a better understanding of the interaction effects of variables on yields, contour plots for the measured responses were formed based on the model (see Equation 9).

Figure 5 shows the response surface plot of copper removal over the process parameters (current density, pH) with variable time fixed at center point (20 min). It exhibits that the percentage of copper removal increases due to both increases in the pH of the solution and current density.

Figure 6 shows the response surface plot of copper removal over the process parameters (current density, time) with variable pH fixed at center point (7). It exhibits that the percentage of copper removal increases due to both increases in the current density and reaction time.

Figure 7 shows the response surface plot of copper removal over the process parameters (pH, time) with variable current density fixed at center point (6 mA/cm^2). It exhibits that the percentage of copper removal increases due to both increases in the pH and reaction time.

As mentioned has an optimum amount with respect to current density, reaction time and pH. At 7,56 pH, 7,24 A current density and 27,43 min reaction time reached 100% (see Table 4)

Figure 2 shows that the predicted results match the experimental values satisfactorily. Thus the response surface method was successfully applied to maximize the copper removal.

Model validation

The accuracy of the model's predicted responses (Y_{pred}) was validated by additional experiments performed under optimal conditions. The experimentally determined value (Y_{exp}) for the removal of copper (Cu(II)) was 97.5%.

Table 2: Experimental design matrix and the response of experimental settings for the reduction of Cu(II) using EC.

Run	**pH**	**Current density (mA/cm^2)**	**Time (min)**	**Copper removal (%)**	
	A	**B**	**C**	**Exp**	**Pred**
1	5	8	20	98.2	97.58
2	9	4	20	89.3	89.92
3	7	8	30	100	100.56
4	7	8	10	97.99	97.41
5	9	6	30	98.6	97.40
6	7	4	30	92.3	92.88
7	7	6	20	94.2	95.40
8	7	6	20	96.9	95.40
9	9	8	20	98.9	99.55
10	5	6	30	90.88	90.95
11	5	4	20	83.8	83.15
12	7	4	10	81.6	81.05
13	7	6	20	95.1	95.40
14	9	6	10	87.9	87.83
15	5	6	10	84.34	97.58

Table 3: The analysis of variance (ANOVA) of the reduced quadratic model representing the reduction of Cu(II) using EC

Source	**Sum of Squares**	**Df**	**Mean square**	***F*-value**	***p*-value**	
Model	511.22	9	56.80	29.67	0.0008	**Significan**t
A-pH	38.19	1	38.19	19.95	0.0066	
B-Current density	289.08	1	289.08	151.00	<0.0001	
C-Time	112.13	1	112.13	58.57	0.0006	
AB	5.76	1	5.76	3.01	0.1433	
AC	4.33	1	4.33	2.26	0.1931	
BC	18.88	1	18.88	9.86	0.0257	
A^2	26.84	1	26.84	14.02	0.0134	
B^2	0.0873	1	0.0873	0.0456	0.8394	
C^2	19.09	1	19.09	9.97	0.0252	

Residual		9.57	5	1.91			
Lack of Fit		5.79	3	1.93	1.02	0.5293	**not significant**
Pure Error		3.78	2	1.89			
Cor Total		520.79	14				
Std. dev.	1.38		R^2				0.9816
Mean	92.67		Adj. R^2				0.9485
*C.V%	1.49		Adeq. precision				17.2698

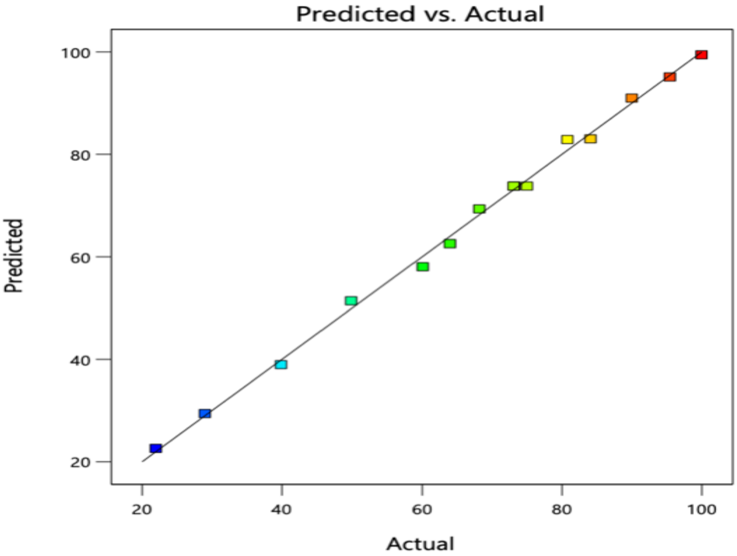

Figure 2: Actual versus Predicted values for Cu(II) removal (%).

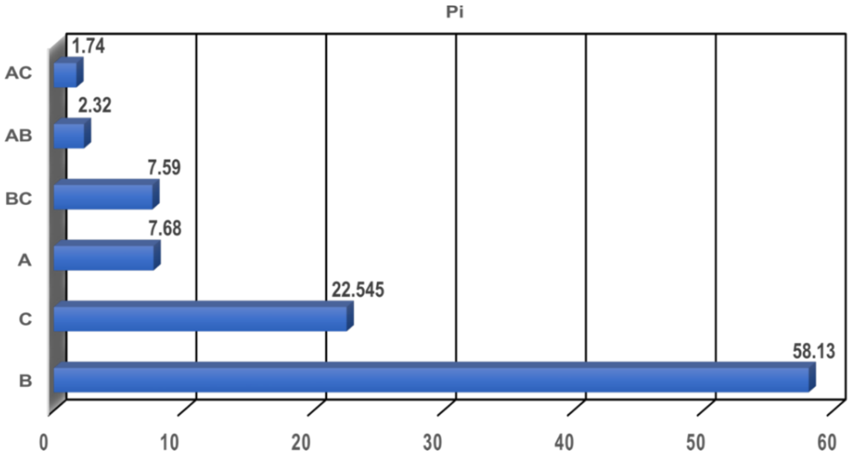

Figure 3: Contributions of the different factors and their interactions.

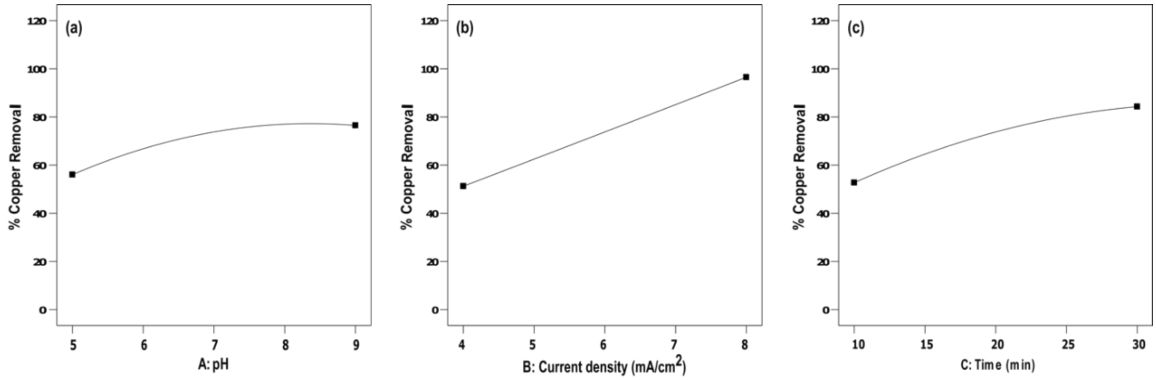

Figure 4: Effect of pH (a), current density (b), and time (c) on the removal of Cu(II).

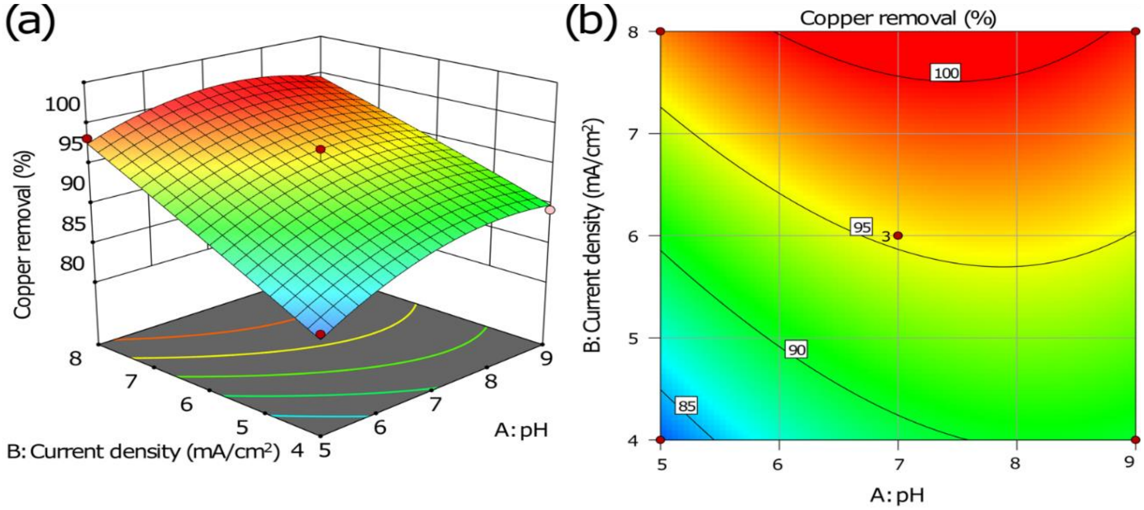

Figure 5: Surface plot (a) and contour plot (b) of Cu(II) removal as a function of current density and pH at time=20 min.

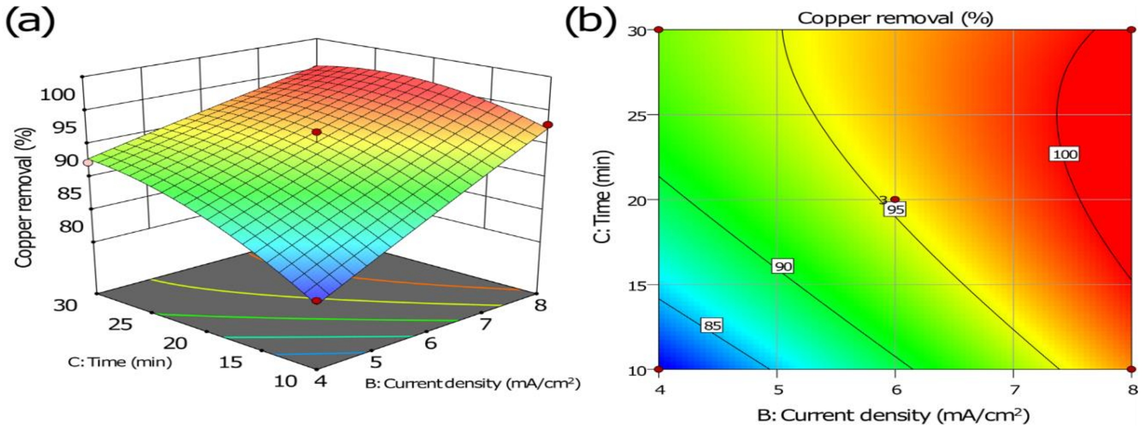

Figure 6: Surface plot (a) and contour plot (b) of Cu(II) removal as a function of Time and current density at pH=7.

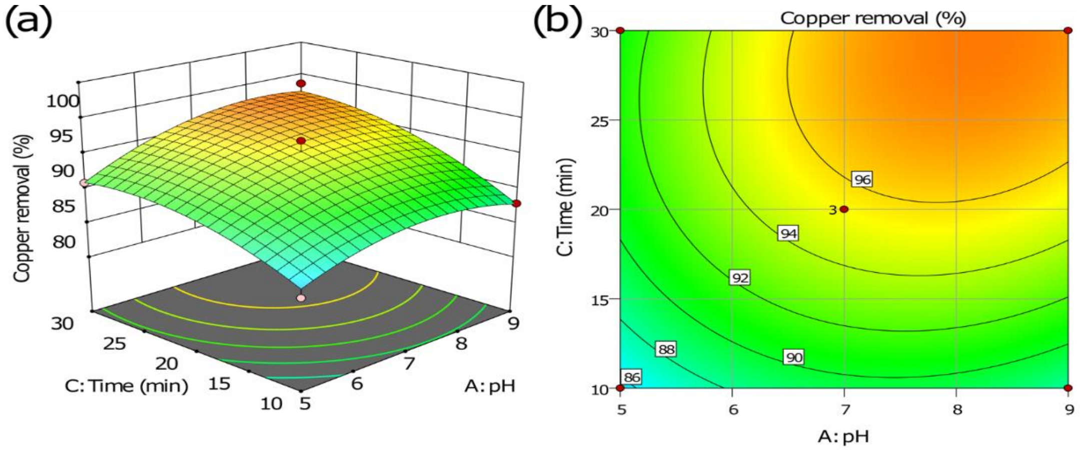

Figure 7: Surface plot (a) and contour plot (b) of Cu(II) removal as a function of Time and pH at current density = 6 mA/cm^2.

Table 4: Optimized values of the process parameters obtained with the Design expert software.

Solution	**A**	**B (mA/cm^2)**	**C (min)**	**Removal rate (%)**	**Desirability (%)**
Cu(II)	7.56	7.24	27.43	100	1

DISCUSSION

The results of the multiple regression analysis show that the independent variables A, B and C have a significant impact on the percentage of copper removal. F-value and p-value are 29.67 and 0.0008, respectively. These F-values, which are above the critical value in the Fisher-Snedecor table (Fc = 5.99) with a p-value < 5%, show that the models are valid and robust (Briton et al., 2018).

The fitted R-squared value of 0.982 suggests that the model accounts for 98% of the variation in the dependent variable. This value is close to 1, indicating that the regression model correlates well with the experimental response (Bao et al., 2022). The model's coefficient values show that B is the most significant variable influencing the percentage of copper removal, with a coefficient of + 6,011. This finding aligns with the results reported in a previous study by Nguyen et al. (2019). This indicates that as the value of B increases, the value of the copper removal percentage also increases. A has a coefficient of + 2.185, indicating that it also has a significant positive impact on copper removal percentage. C has a coefficient of + 3.744, indicating that it has a positive effect on the percentage of copper removal, but not as strong as B. The interaction term BC has a negative effect coefficient of -2.173, indicating that the combined values of B and C have a negative effect. This negatively affects the percentage of copper removal. The quadratic terms A^2 and C^2 also have negative coefficients, indicating that the effect of A and C on percent copper removal can decrease as their values increase beyond a certain point. Overall, the results indicate that current density (B) has the greatest impact on Cu(II) removal, followed by electrolysis time (C) and pH (A). The negative coefficient of the interaction terms suggests that the optimal conditions for Cu(II) removal can be achieved by finding an equilibrium between B and C. The negative coefficients of the squared terms show that the effects of A and C on Cu(II) removal can decrease after an optimal value is reached.

The results showed that the removal efficiency of the percentage of copper removal was affected by the pH of the solution. The findings showed that the efficiency of copper ion removal was improved by increasing the pH from 5 to 9. The removal efficiency of Cu(II) increased to 4.37% (2 x 2.185) on average. This improvement is due to an increase in OH^- species with increasing pH. The formation of metal hydroxide as coagulants or flocs increases with increasing OH^-. This increases the removal of copper ions as the pH of the effluent rises. According to Vasudevan et al., when using an iron anode, reducing the pH of the solution can lead to a reduction in the efficiency of copper removal, as the oxidation of ferrous iron (Fe II) to ferric iron (Fe III). On the other hand, neutral or slightly alkaline pH promotes the oxidation of Fe (II) to Fe (III) and facilitates complex polymerization. This process results in the formation of hydroxylated colloidal polymers and an insoluble precipitate of hydrated ferric oxide, which can improve copper removal efficiency.

Current optimization is an important factor to consider to improve the removal efficiency of Cu ions during electrocoagulation. The results show that the current density has a clearly positive effect on the Cu(II) removal from the synthetic wastewater. When the current density was increased from 4 to 8 mA/cm^2 at a fixed electrolysis time of 30 minutes, the removal rate improved by approximately 12.022% (2x 6.011). This indicates that increasing the current density can improve the performance of the EC process in removing Cu(II) from wastewater. The results obtained are consistent with the results of other researchers. They reported that by increasing the current density according to Faraday's law, large amounts of hydroxide ions and dihydrogen bubbles are generated (Al Aji et al., 2012; Beiramzadeh et al., 2022). This large amount of hydroxide ions would precipitate a large amount of Cu(II) directly to $Cu(OH)_2$ or could form more iron hydroxides ($Fe(OH)_2$ and $Fe(OH)_3$) removing a large amount of Cu(II) through adsorption, complex formation and co-precipitation (Meunier et al., 2006; Drogui et al., 2011; Kessentini et al., 2019). In addition, a large number of small gas bubbles (H_2) pull the metals contained in the flakes to the free surface of the reactor by the flotation effect (Burboa-Charis et al., 2019).

Electrolysis time is another critical factor affecting copper removal efficiency. The positive effect (b_3 = +3.744) shows that extending the electrolysis time from 10 to 30 minutes improves the removal rate by around 7.488% (2x 3.744). These results are fully consistent with previous studies by Bhagawan et al. (2014) and Aljaberi and Hawaas (2023). The increase in removal rate could be due to the production of more metal hydroxides, which occurs due to Faraday's law and increases the Cu (II) adsorption sites. In addition, the contact time between the adsorbents (metal hydroxides) and the cation Cu(II) is determined by the electrolysis time. A longer contact time allows more metal cations to be adsorbed on the surface of the metal hydroxide (Ano et al., 2023). According to Equation 8, the observed low coefficient of variation (CV) of 2.58% underlines the robustness and consistency of the experimental results (Ntakiyiruta et al., 2022).

Conclusion

This study investigated the removal of copper from synthetic water using a combination of electrocoagulation (EC) and RSM. The study systematically examined the individual and combined effects of initial pH, current density, and electrolysis time on Cu(II). The findings revealed that the current density exerted the most significant influence on the copper removal process, and notable interactions were observed between current density and reaction time. The statistical analysis indicated a high degree of correlation, with a coefficient of determination (R^2) value of 0.982 and adjusted determination coefficient (AdjR^2 = 0,948) showing that the quadratic polynomial regression model could properly interpret the experimental data.

The study successfully identified the optimal conditions for achieving maximum copper removal efficiency: an initial pH of 7.56, a current density of 7.24 mA/cm^2, and a reaction time of 27.42 min. Under these conditions, the copper removal efficiency reached an impressive 97.5%.

The results show that EC is an efficient method for removing copper from synthetic wastewater. The use of response surface methodology (RSM) provides a systematic approach for optimizing the EC process variables and maximizing copper removal efficiency. The combination of EC and RSM offers a promising approach to the treatment of copper-contaminated wastewater. However, further research is required to address the applicability and effectiveness of electrocoagulation for removing copper from real wastewater should be investigated.

COMPETING INTERESTS

The authors declare that they have no competing interests.

AUTHORS' CONTRIBUTIONS

AD: Conceived, designed and performed the experiments; analyzed and interpreted the data and wrote the paper. AT: Conceived and designed the experiments; interpreted the data; contributed reagents, materials, analysis tools or data; revised the paper. ADi participated in paper revision, data analysis and interpretation. DK participated in data analysis and paper revision.

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