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Electrocoagulation-based removal of lead from wastewater: Process optimization using response surface methodology

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ABSTRACT

Heavy metal contamination, particularly from lead (Pb), represents a critical environmental and public health concern owing to its persistence, high toxicity, and pronounced tendency for bioaccumulation in ecological and human systems. In this context, the present study investigates the effectiveness of electrocoagulation (EC) as a treatment technology for the removal of Pb^{2+} from aqueous solutions, coupled with a systematic process optimization approach. A Box-Behnken experimental design was applied within the response surface methodology (RSM) framework. It was used to evaluate the individual and interactive effects of key operational parameters, including pH, current density, and reaction time, on Pb^{2+} removal efficiency. The results clearly indicate that pH is the most influential variable, with optimal removal performance observed under near-neutral conditions (pH 7–8), where the formation of metal hydroxide flocs is maximized. In comparison, current density and reaction time exhibited moderate yet complementary influences, primarily through their roles in enhancing coagulant generation and facilitating floc growth and aggregation. The developed quadratic model demonstrated high statistical significance and excellent predictive capability, as reflected by a coefficient of determination (R^2) of 0.993. The strong agreement between experimental observations and model predictions confirms the robustness and reliability of the optimization framework. Under the identified optimal conditions – pH 7.6, current density 55 A/m², and reaction time 38.5 minutes – the experimental Pb^{2+} removal efficiency reached 98.35%, closely aligning with the model-predicted value of 99.82%. The minimal deviation between these values further validates the accuracy of the model and its suitability for predictive and optimization purposes. The findings of this study demonstrate that electrocoagulation is an efficient, cost-effective, and environmentally sustainable method for the removal of Pb^{2+} from contaminated water. Moreover, the integration of EC with advanced statistical optimization techniques such as RSM provides a powerful and reliable framework for enhancing process performance. These results support the potential application of EC technology in real wastewater treatment systems and contribute to the development of optimized strategies for heavy metal remediation.

Keywords: electrocoagulation, lead removal, response surface methodology, Box-Behnken design, wastewater treatment.

INTRODUCTION

Heavy metal contamination in aquatic environments has emerged as a critical global environmental and public health issue due to the persistence, toxicity, and non-biodegradable nature of these pollutants. Among them, lead (Pb) is of particular concern because of its extensive industrial applications and severe toxicological impacts. Pb has been classified as a priority pollutant by the United States Environmental Protection Agency (USEPA) and is associated with a wide range of adverse health effects, including neurological disorders, renal dysfunction, hematological abnormalities, and developmental impairments, especially in children (Fu and Wang, 2011; Tchounwou et al., 2012). Notably, even at trace concentrations (e.g., <0.01 mg/L), Pb can bioaccumulate in living organisms and biomagnify through food chains, resulting in long-term ecological and human health risks.

Pb contamination in aquatic environments is largely associated with industrial activities,

including battery manufacturing, electroplating, mining, smelting, and pigment production (Fu and Wang, 2011; Tchounwou et al., 2012; Jaishankar et al., 2014). Rapid industrialization, particularly in developing regions of Asia and Africa, has significantly increased the discharge of Pb-containing effluents into water bodies (Ali et al., 2019; WHO, 2022). Reported Pb concentrations in industrial wastewater and contaminated surface waters range from trace levels (μg/L) to several mg/L, frequently exceeding drinking water standards such as the WHO guideline of 0.01 mg/L (Fu and Wang, 2011; WHO, 2022). In developing countries, including Vietnam, Pb pollution is further intensified by urban runoff, untreated industrial discharges, and inadequate wastewater treatment infrastructure, posing serious environmental and public health risks (Anh et al., 2021; Le and Nguyen, 2024; Thi et al., 2025).

The removal of Pb from aqueous systems remains technically challenging due to its non-biodegradable nature and complex chemical behavior in water environments (Fu and Wang, 2011; Tchounwou et al., 2012). Conventional treatment methods, including chemical precipitation, ion exchange, adsorption, and membrane filtration, have been widely applied for heavy metal removal (Barakat, 2011; Ali et al., 2019). However, these approaches often exhibit several limitations, such as high operational costs, reduced efficiency at low metal concentrations, generation of secondary waste (e.g., sludge), and sensitivity to water chemistry conditions (Barakat, 2011; Fu and Wang, 2011). For instance, adsorption using activated carbon, biochar, and various nanomaterials has demonstrated removal efficiencies exceeding 90%, but this process mainly transfers Pb from the liquid phase to a solid matrix, raising concerns regarding adsorbent regeneration and disposal (Mohan et al., 2014; Crini and Lichtfouse, 2019). Similarly, membrane-based technologies such as reverse osmosis and nanofiltration can achieve high removal efficiencies (>95%); however, their practical application is often limited by membrane fouling, high energy consumption, and operational costs (Shannon et al., 2008; Ali et al., 2019).

In this context, electrochemical technologies have gained increasing attention as promising alternatives for wastewater treatment due to their high efficiency, operational simplicity, and reduced reliance on chemical reagents. Among these, electrocoagulation (EC) has emerged as an effective technique for removing heavy metals, including Pb, from aqueous solutions (Mollah et al., 2001; Garcia-Segura et al., 2017). The EC process involves the anodic dissolution of sacrificial electrodes, typically aluminum or iron, under an applied electric current, leading to the in situ generation of metal hydroxide coagulants. These species facilitate contaminant removal through multiple mechanisms, including adsorption, charge neutralization, and co-precipitation. Compared with conventional coagulation, EC minimizes the need for external chemical additives and produces lower volumes of sludge, making it an environmentally sustainable and cost-effective treatment option (Chen, 2004).

Recent studies further demonstrate the high efficiency and scalability of EC systems for heavy metal removal. For instance, Tuan et al. (2025) reported arsenic removal efficiencies of up to 99.93% at an initial concentration of 100 ppb and 99.41% at 300 ppb under optimized conditions of pH, current density, and reaction time using response surface methodology (RSM). The developed models exhibited strong predictive performance (R^2 = 0.997 and 0.802), highlighting the capability of EC systems to achieve near-complete contaminant removal under optimized operating conditions. These findings underscore the potential of electrocoagulation as a reliable, efficient, and scalable technology for treating heavy metal-contaminated water, particularly under near-neutral pH and moderate current densities.

EC is an advanced electrochemical water treatment technique. It relies on the in situ generation of coagulant species through the controlled dissolution of sacrificial metal electrodes, typically aluminum or iron, under an applied electric current. Unlike conventional chemical coagulation, which requires the addition of external coagulants, EC produces active coagulant species directly within the aqueous medium via anodic oxidation, thereby enhancing process efficiency while minimizing chemical handling and secondary pollution (Mollah et al., 2001; Chen, 2004; Khandegar and Saroha, 2013).

In aluminum-based EC systems, the fundamental mechanism involves the electrochemical oxidation of the aluminum anode, leading to the release of Al^{3+} ions into the solution. Simultaneously, reduction reactions occur at the cathode, where water molecules are reduced to generate hydrogen gas (H_2) and hydroxide ions (OH^-). The hydrogen gas formed at the cathode contributes to flotation by attaching to suspended particles,

while the hydroxide ions facilitate the hydrolysis of aluminum species. The released Al^{3+} ions undergo rapid hydrolysis in the aqueous environment, forming a range of monomeric and polymeric hydroxo complexes, which ultimately precipitate as amorphous aluminum hydroxide, $Al(OH)_3$. These hydroxide species possess high specific surface area and strong adsorption capacity, making them highly effective for contaminant removal (Mollah et al., 2001; Chen, 2004).

The principal electrochemical and hydrolysis reactions governing the EC process can be expressed as follows:

$$Al \rightarrow Al^{3+} + 3e^- \quad (1)$$

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

$$Al^{3+}3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad (3)$$

The amorphous $Al(OH)_3$ flocs generated through these reactions act as highly efficient coagulant agents. Their removal mechanisms are multifaceted, encompassing adsorption onto floc surfaces, charge neutralization of destabilized colloidal particles, and co-precipitation with dissolved contaminants. In the case of heavy metals such as Pb^{2+}, removal occurs predominantly through the formation of insoluble metal–hydroxide complexes and their subsequent incorporation into the growing floc matrix. Additionally, sweep flocculation plays a critical role, whereby contaminants are entrapped within the bulk of precipitated hydroxides and removed via sedimentation or flotation (Figure 1) (Mollah et al., 2001; Kobya et al., 2003).

Previous studies have consistently demonstrated the effectiveness of EC for the removal of heavy metals from wastewater. For instance, Akbal & Camcı (2012) reported removal efficiencies exceeding 95–99% for Pb, Cu, and Zn using aluminum electrodes under optimized conditions. Similarly, Khandegar and Saroha (2013) emphasized the advantages of EC, including operational simplicity, low sludge production, and high removal efficiencies across a broad range of wastewater compositions. More recent investigations have extended the application of EC to complex and real wastewater matrices, with reported efficiencies typically above 90% for various heavy metals, thereby highlighting its strong potential for large-scale implementation (Sadik et al., 2019; Bhagawati et al., 2022). Nevertheless, the performance of EC systems is highly dependent on key operational parameters such as pH, current density, electrode configuration, and reaction time, all of which can significantly influence coagulant generation and pollutant removal mechanisms. This variability underscores the need for systematic optimization to achieve consistent and optimal treatment performance.

To address this challenge, statistical optimization techniques, particularly response surface methodology (RSM), have been widely employed to evaluate the interactive effects of multiple variables and to determine optimal operating conditions. RSM is a statistical technique used to model and optimize processes influenced by multiple variables. Among these approaches, the Box-Behnken design (BBD), a three-level design, enables efficient estimation

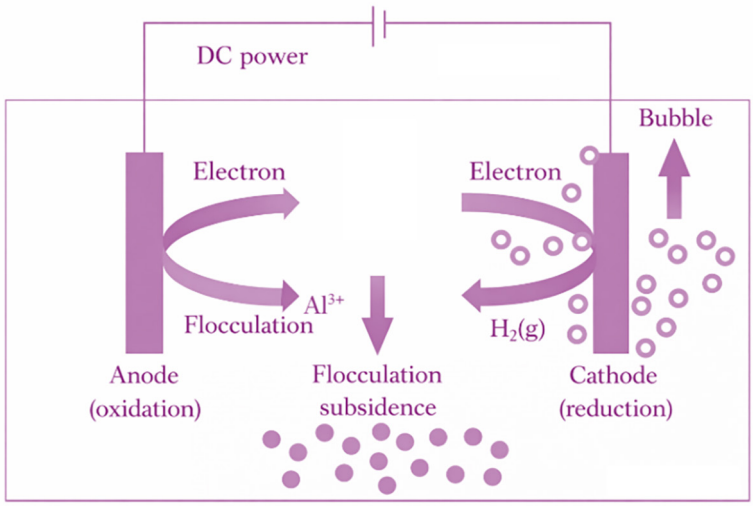

Figure 1. Conceptual diagram of the electrocoagulation process

of quadratic models while minimizing the number of experimental runs. BBD is therefore recognized as an efficient and reliable experimental design for three-factor systems, typically requiring only 15–17 runs while effectively capturing second-order interactions (Li et al., 2013; Montgomery, 2017; Rajewski and Dobrzyńska, 2021 Nurjaman et al., 2025). Despite these advances, studies integrating EC with advanced statistical optimization techniques for Pb removal under varying operational conditions remain limited. In contrast to previous electrocoagulation studies that often focus on limited parameter ranges or purely optimization-driven outcomes, this study provides a comprehensive evaluation of both individual and interactive effects of key operational variables under practically relevant conditions. Furthermore, the integration of response surface methodology with rigorous experimental validation yields a highly accurate predictive model and identifies optimal conditions, thereby enhancing the reliability and scalability of electrocoagulation for Pb removal in real-world applications.

Therefore, this study aims to systematically investigate the efficient removal of Pb^{2+} ions from wastewater using an EC system integrated with a statistical process optimization framework. The research is designed to provide both mechanistic understanding and practical guidance for improving treatment performance under controlled operating conditions. In particular, the study seeks to achieve several interrelated objectives. First, it aims to comprehensively evaluate the effects of key operational parameters – namely pH, current density, and reaction time – on Pb^{2+} removal efficiency, with a focus on elucidating their individual contributions and potential interaction effects within the electrocoagulation process. Second, it endeavors to develop a robust and predictive empirical model using RSM based on a Box-Behnken experimental design, thereby enabling quantitative assessment of process behavior and facilitating accurate prediction of system performance across the experimental domain. Third, the study aims to identify the optimal combination of operating conditions that maximizes Pb^{2+} removal efficiency while maintaining practical feasibility in terms of energy consumption and operational simplicity.

Beyond these specific objectives, the outcomes of this research are expected to contribute to the broader development of advanced electrochemical treatment technologies for heavy metal remediation. By integrating experimental investigation with statistical optimization, the study provides a comprehensive framework for enhancing process efficiency and reliability. The findings are anticipated to offer valuable insights into the design and optimization of cost-effective and environmentally sustainable wastewater treatment systems, particularly in resource-constrained settings where conventional treatment methods may be limited by economic or infrastructural constraints.

METHODOLOGY

Preparation of lead-contaminated synthetic wastewater

A synthetic lead-contaminated wastewater solution was prepared to ensure controlled and reproducible experimental conditions for evaluating the EC process. Specifically, a stock solution containing Pb^{2+} at a concentration of 100 mg/L was formulated by dissolving analytical-grade lead nitrate, $Pb(NO_3)_2$ (purity ≥95%), in deionized water. The use of deionized water minimized the influence of background ions and potential interferences, thereby allowing a more precise assessment of the EC process performance. To enhance the electrical conductivity of the solution and facilitate efficient current flow during electrolysis, sodium chloride (NaCl) was added at a concentration of 2 g/L. The presence of chloride ions not only improves conductivity but may also contribute to the formation of additional reactive species, thereby potentially enhancing pollutant removal efficiency.

The initial pH of the solution was carefully adjusted within the range of 4–10 using standardized solutions of 0.1 N hydrochloric acid (HCl) or sodium hydroxide (NaOH). This pH range was selected based on its relevance to practical wastewater conditions and its known influence on metal speciation, coagulant formation, and overall EC performance. The pH was measured using a calibrated pH meter prior to the commencement of each experiment and re-measured upon completion to evaluate any variations induced by electrochemical reactions during treatment.

The efficiency of Pb^{2+} removal was quantified by comparing the initial concentration (L_o) with the residual concentration (L_t) at a given reaction

time. The removal efficiency (%) was calculated using the following expression:

$$Pb\ removal\ efficiency\ (\%) = \\ = \frac{L_0 - L_t}{L_0} \times 100 \quad (4)$$

where: L_0 (mg/L) denotes the initial concentration of Pb^{2+} and L_t (mg/L) represents the concentration remaining in solution at time t.

Residual lead concentrations were determined using appropriate analytical technique of atomic absorption spectrophotometry (AAS), ensuring high accuracy and reliability of the measurements. All experiments were conducted in triplicate under identical operating conditions to ensure reproducibility and statistical robustness. The reported results correspond to the mean values of these replicates, and standard deviations were monitored to assess experimental variability.

To systematically investigate the effects of key operational parameters and to identify optimal conditions for Pb^{2+} removal, the experimental design and process optimization were performed using RSM implemented in Design-Expert software. This approach enables the evaluation of individual and interactive effects of variables, as well as the development of predictive models and response surfaces for process optimization with a reduced number of experimental runs.

Experimental setup of the electrocoagulation system

The EC experiments were conducted using a laboratory-scale batch reactor system specifically designed to ensure operational stability, reproducibility, and precise control of key process parameters. As illustrated in Figure 2, the experimental system consisted of a 10 L rectangular acrylic reactor, selected for its chemical inertness, transparency, and ease of observation during operation. The reactor was equipped with a magnetic stirring system. This ensured continuous and homogeneous mixing of the electrolyte solution. As a result, mass transfer was enhanced, localized concentration gradients were minimized, and uniform floc formation was promoted throughout the treatment process.

A regulated direct current (DC) power supply (Alexan, adjustable within the range of 10–30 V and 0–5 A) was employed to deliver a stable electrical input to the system. This enabled accurate control of the applied current and voltage, which are critical parameters influencing electrode dissolution, coagulant generation, and overall removal efficiency. The electrode assembly comprised parallel plate electrodes made of aluminum (anode) and iron (cathode), each with dimensions of 21 × 10 × 3 mm. These materials were selected based on their widespread use in EC applications and their proven effectiveness in generating metal hydroxide flocs for contaminant removal.

The electrodes were vertically positioned within the reactor at an inter-electrode distance of 2.5 cm, a configuration chosen to balance electrical resistance and effective coagulant distribution. The immersion depth of the electrodes was maintained at approximately 20 cm to ensure sufficient contact with the electrolyte solution while maximizing the active surface area available for electrochemical reactions. Electrical connections between the electrodes and the power supply were established using insulated conductive wires with alligator clips, allowing flexible adjustment and secure attachment during operation. This configuration facilitated precise control over the applied current density and ensured consistent experimental conditions across different runs.

Electrocoagulation experiments were conducted in batch mode. The principal operating parameters (initial pH, current density, and reaction time) were systematically varied. The selected ranges of pH (4–10), current density (30–90 A/m^2), and reaction time (20–60 min) were determined based on literature evidence and preliminary experimental screening conducted by the research team to ensure both practical relevance and effective optimization. All other conditions, including electrolyte concentration, electrode configuration, and mixing speed, were kept constant to isolate both individual and interactive effects. This approach enabled a comprehensive evaluation of process performance and facilitated subsequent optimization.

During each experimental run, liquid samples were withdrawn at predetermined time intervals to monitor the progression of Pb^{2+} removal. The collected samples were immediately filtered to remove suspended flocs and prevent further reactions prior to analysis. Residual lead concentrations were then quantified using atomic absorption spectroscopy (AA-7000F, Japan), a highly sensitive and reliable analytical technique widely

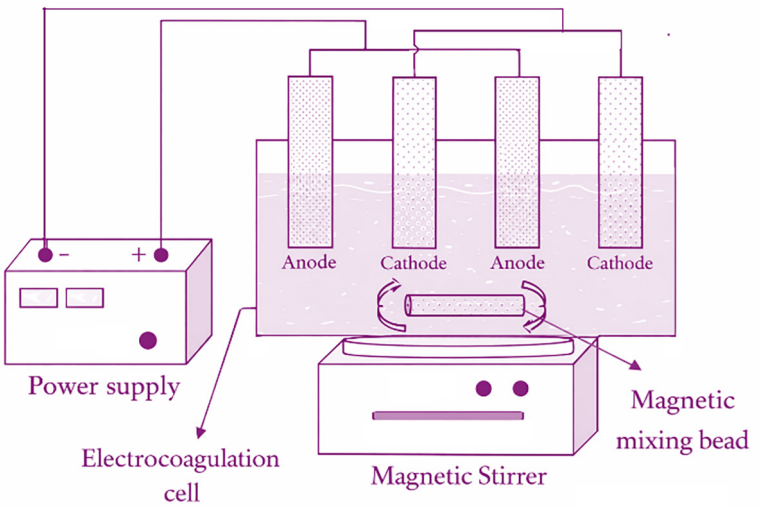

Figure 2. Schematic diagram of the electrocoagulation system

employed for trace metal determination. Calibration procedures and quality control measures were implemented to ensure the accuracy and precision of the analytical results.

All experiments were conducted under controlled laboratory conditions, with ambient temperature maintained within the range of 25–30 °C to minimize the influence of temperature fluctuations on electrochemical reactions and solution properties. This controlled environment, combined with standardized operational procedures, ensured high reproducibility and reliability of the experimental data, thereby providing a robust basis for subsequent modeling and optimization analyses.

RESULTS AND DISCUSSION

Electrocoagulation of Pb^{2+} Wastewater

EC experiments were systematically conducted using synthetic lead-contaminated wastewater with an initial Pb^{2+} concentration of 100 mg/L, representing a relatively high contamination level to rigorously evaluate the treatment efficiency and robustness of the process. The experimental program was designed following a structured statistical approach, resulting in a total of 15 experimental runs corresponding to different combinations of key operational parameters. These runs were established based on a RSM framework, enabling efficient exploration of both the individual and interactive effects of variables such as pH, current density, and reaction time on Pb^{2+} removal performance.

To ensure the reliability, accuracy, and reproducibility of the obtained results, each experimental condition was performed in triplicate under identical operating parameters. The reported values represent the arithmetic mean of these replicates, while variability among measurements was carefully monitored to assess experimental consistency. This replication strategy minimizes random errors and enhances the statistical robustness of the dataset, which is essential for subsequent modeling, optimization, and interpretation.

Throughout the experimental campaign, strict control of operational conditions – including solution composition, electrode configuration, mixing intensity, and temperature – was maintained to isolate the effects of the investigated variables. The systematic design and execution of experiments allowed for the generation of a comprehensive dataset suitable for developing predictive models and identifying optimal operating conditions for maximum Pb^{2+} removal efficiency.

The detailed experimental design matrix, including the levels of independent variables and the corresponding response values (Pb^{2+} removal efficiency), is presented in Table 1. This table provides a complete overview of the experimental conditions and serves as the basis for subsequent statistical analysis, model development, and process optimization.

The statistical significance and adequacy of the developed response surface model for Pb^{2+} removal efficiency were rigorously evaluated using analysis of variance (ANOVA). This approach enables the quantitative assessment of the relative contributions of model terms, including linear, interaction,

Table 1. Box–Behnken design and results for Pb^{2+} removal

RUN	pH	Current density (A/m²)	Reaction time (min)	EFFLUENT (mg/L)	Lead removal (%)
1	4	30	40	13.2	86.8
2	10	30	40	2.5	97.5
3	4	90	40	14.0	86
4	10	90	40	2.8	97.2
5	7	30	20	1.2	98.8
6	7	30	60	0.9	99.1
7	7	90	20	6.2	93.8
8	7	90	60	1.2	98.8
9	4	60	20	12.8	87.2
10	10	60	20	3.8	96.2
11	4	60	60	11.5	88.5
12	10	60	60	1.5	98.5
13	7	60	40	0.7	99.3
14	7	60	40	1.3	98.7
15	7	60	40	1.0	99

and quadratic effects, as well as the identification of statistically significant factors influencing process performance. In particular, ANOVA provides key statistical indicators such as the F-value, p-value, coefficient of determination (R^2), adjusted R^2, and lack-of-fit test, which collectively determine the reliability and predictive capability of the model.

A high model F-value accompanied by a low p-value ($p < 0.05$) indicates that the regression model is statistically significant, while non-significant lack-of-fit ($p > 0.05$) confirms that the model adequately represents the experimental data without systematic deviation. Furthermore, the closeness between the predicted and adjusted R^2 values reflects the robustness of the model and its suitability for predicting Pb^{2+} removal efficiency under varying operating conditions. The significance of individual terms, including main factors (e.g., pH, current density, and reaction time) and their interactions, was also examined to elucidate their respective roles in governing the electrocoagulation process.

Generally, the ANOVA results provide a comprehensive statistical validation of the developed model and support its application for process optimization and interpretation of parameter interactions. The detailed ANOVA table summarizing the sources of variation, degrees of freedom, sum of squares, mean squares, F-values, and corresponding p-values for Pb^{2+} removal efficiency is presented in Table 2.

Table 2. Analysis of variance (ANOVA) for lead removal performance

Source	Sum of squares	df	Mean square	F-value	p-value	
Model	367.61	9	40.85	53.78	0.0002	significant
X-pH	129.52	1	129.52	170.53	< 0.0001	
Y-current density	3.76	1	3.76	4.95	0.0767	
Z-reaction time	0.2554	1	0.2554	0.3362	0.5872	
XY	0.0625	1	0.0625	0.0823	0.7857	
XZ	0.2500	1	0.2500	0.3292	0.5910	
YZ	5.52	1	5.52	7.27	0.0430	
X^2	136.27	1	136.27	179.42	< 0.0001	
Y^2	4.07	1	4.07	5.36	0.0685	
Z^2	0.3900	1	0.3900	0.5135	0.5057	
Residual	3.80	5	0.7595			
Lack of fit	3.62	3	1.21	13.40	0.0703	not significant

As presented in Table 2, the developed quadratic regression model for Pb^{2+} removal efficiency is statistically highly significant, as evidenced by a model F-value of 53.78. This large F-value indicates that the variation explained by the model is substantially greater than the unexplained residual variation. Moreover, the associated probability value ($p = 0.0002$) suggests that there is only a 0.02% likelihood that such a high F-value could arise purely from random noise. This extremely low p-value confirms the robustness, reliability, and statistical validity of the proposed model for describing the electrocoagulation process under the investigated conditions.

A closer examination of individual model terms reveals that pH (X) and its quadratic component (X^2) are highly significant ($p < 0.0001$), clearly indicating that pH is the dominant factor governing Pb^{2+} removal efficiency. This finding is consistent with the fundamental role of pH in controlling metal speciation, hydrolysis reactions, and the formation of aluminum hydroxide flocs, which are the primary agents responsible for contaminant removal in electrocoagulation systems. The strong significance of the quadratic term further suggests the presence of a non-linear relationship, implying the existence of an optimal pH range within which maximum removal efficiency can be achieved.

In addition to the main effects, the interaction between current density (Y) and reaction time (Z) (denoted as YZ) is statistically significant ($p = 0.0430$), highlighting a synergistic effect between these two parameters. This interaction suggests that the influence of current density on Pb^{2+} removal is dependent on the duration of electrolysis, reflecting the combined roles of coagulant generation rate and contact time in determining treatment performance. In contrast, the linear terms corresponding to Y and Z, as well as the interaction terms XY (pH × current density) and XZ (pH × reaction time), are not statistically significant ($p > 0.05$). This indicates that, within the selected experimental domain, their individual and pairwise contributions are relatively less pronounced compared to pH. Similarly, the quadratic terms Y^2 and Z^2 are also non-significant, suggesting that curvature effects associated with these variables are minimal over the studied range.

The adequacy of the model is further supported by the lack-of-fit test. The lack-of-fit F-value of 13.40, associated with a p-value of 0.0703 (greater than 0.05), indicates that the lack of fit is not statistically significant. This result confirms that the model provides an appropriate representation of the experimental data, with no systematic deviation between predicted and observed values. Consequently, the developed regression model can be considered reliable for both prediction and optimization purposes.

Based on the regression analysis, the relationship between Pb^{2+} removal efficiency and the independent variables is described by the final empirical model in terms of actual factors, as given in Equation 5:

$$\begin{aligned} Pb\ Removal = {} & 54.32083 + 10.90417 \cdot pH + \\ & + 0.025278 \cdot Current\ Density - 0.026042 \cdot \\ & \cdot Reaction\ time + 0.001389 \cdot pH \cdot \\ & \cdot\ Current\ Density + \ 0.004167 \cdot pH \cdot \\ & \cdot Reaction\ time + 0.001958 \cdot\ Current\ Density \cdot \\ & \cdot Reaction\ time - 0.675000 \cdot pH^2 - 0.001167 \cdot \\ & \cdot Current\ Density^2 - 0.000813 \cdot Reaction\ time^2 \end{aligned} \quad (5)$$

This equation quantitatively describes the combined linear, interaction, and quadratic effects of the operating parameters on Pb^{2+} removal efficiency, and it serves as a predictive tool for estimating system performance under various conditions within the experimental domain.

Furthermore, as illustrated in Figure 3, the model exhibits excellent predictive capability, as indicated by a very high coefficient of determination ($R^2 = 0.993$). This implies that 99.3% of the variability in Pb^{2+} removal efficiency can be explained by the model, leaving only a small fraction attributable to unexplained error. The close agreement between predicted and experimental values, derived using Equation 5, further validates the accuracy and reliability of the developed model. Such a high level of model performance underscores its suitability for process optimization and supports its application in scaling up and practical implementation of electrocoagulation for lead removal from wastewater.

Impact of operational parameters on Pb removal efficiency

The influence of key operational parameters on Pb^{2+} removal efficiency was systematically evaluated to elucidate the governing mechanisms of the EC process. Variations in removal performance as a function of the investigated factors – namely pH, current density, and reaction time – are illustrated in Figure 4 through response

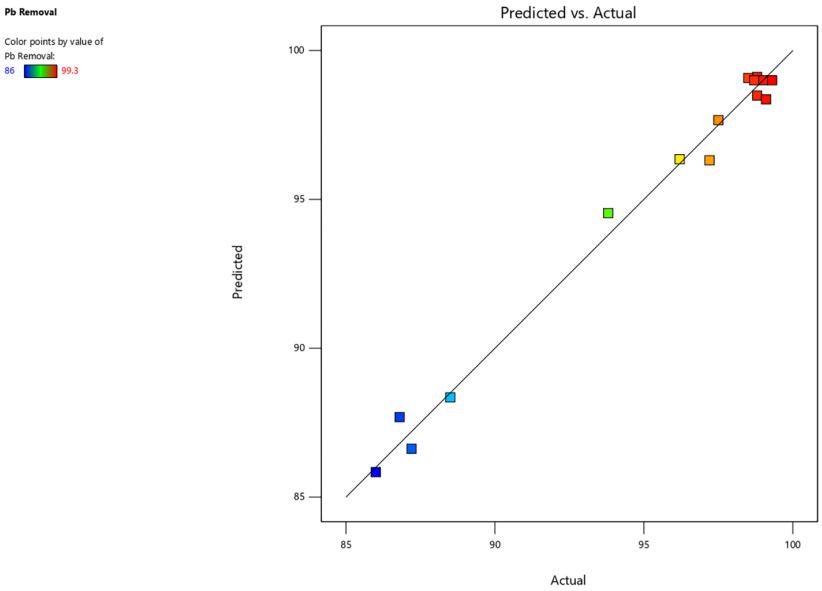

Figure 3. Experimental versus predicted values of Pb removal efficiency

surface plots generated from the developed regression model. These graphical representations provide valuable insight into both the individual effects of each parameter and their interactive relationships, thereby facilitating a deeper understanding of process behavior.

Figure 4 provides a comprehensive depiction of the individual effects of key operational parameters – namely pH, current density, and reaction time – on the percentage removal of Pb^{2+} during the EC process. The trends illustrated in the figure highlight the relative importance of each factor and offer mechanistic insights into the underlying processes governing heavy metal removal.

As shown in Figure 4(a), pH exerts a pronounced and dominant influence on Pb^{2+} removal efficiency. A substantial increase in removal efficiency is observed as the solution pH rises from acidic conditions toward near-neutral values, with maximum performance typically achieved within the pH range of approximately 7–8. This behavior can be attributed to the critical role of pH in regulating both the speciation of dissolved metal ions and the hydrolysis of aluminum species generated at the anode. Under near-neutral conditions, the formation of amorphous $Al(OH)_3$ flocs is significantly enhanced, providing a large surface area and abundant active sites for adsorption and co-precipitation of Pb^{2+} ions. In contrast, at lower pH values, the solubility of aluminum species remains high, limiting floc formation and consequently reducing removal efficiency. At excessively high pH levels, the possible formation of soluble aluminate species may also hinder effective coagulation, further supporting the existence of an optimal pH range.

Figure 4(b) illustrates the effect of current density on Pb^{2+} removal efficiency, revealing a generally increasing trend, albeit less pronounced compared to that of pH. As current density increases, the rate of anodic dissolution of the aluminum electrode is enhanced, leading to a higher production of coagulant species (Al^{3+}) and, consequently, an increased formation of hydroxide flocs. Additionally, higher current density promotes the generation of hydrogen bubbles at the cathode, which can facilitate the flotation of flocculated particles. However, the observed improvement in removal efficiency occurs at a relatively gradual rate, indicating diminishing returns beyond a certain threshold. This suggests that excessively high current densities may not be economically optimal, as they increase energy consumption without yielding proportional gains in treatment performance.

The influence of reaction time is presented in Figure 4(c), where Pb^{2+} removal efficiency initially increases with increasing electrolysis duration, followed by a plateau or slight decline at extended reaction times. The initial rise in efficiency can be attributed to the progressive accumulation of coagulant species and the increased opportunity for interaction between Pb^{2+} ions and hydroxide

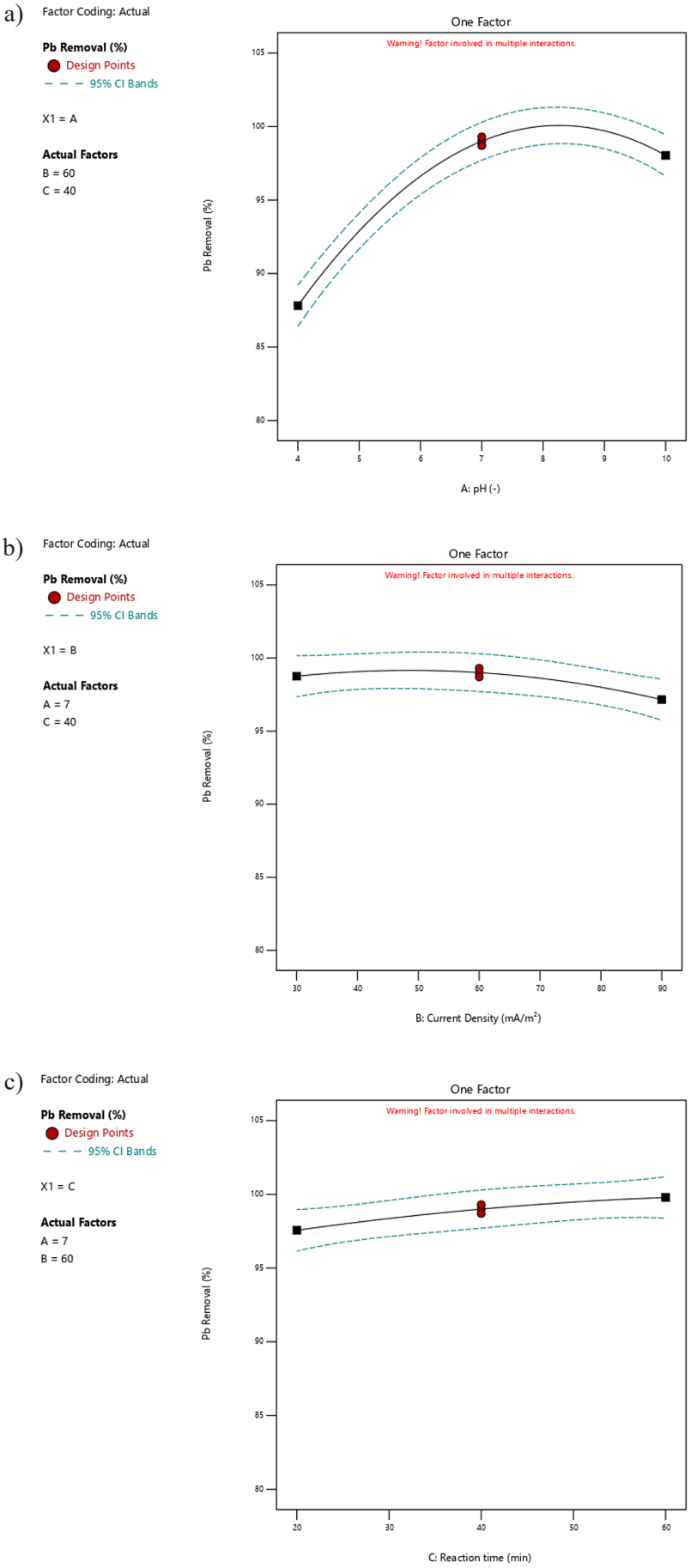

Figure 4. Variation in Pb removal efficiency as a function of (a) pH, (b) current density, and (c) reaction time

flocs. Adequate reaction time allows for the growth, aggregation, and stabilization of flocs, thereby enhancing pollutant removal. However, at longer durations, the marginal improvement diminishes, and in some cases, a slight decrease in efficiency may occur. This phenomenon may be associated with floc breakage, re-dispersion of previously captured particles, or saturation of available adsorption sites, which collectively limit further removal.

Taken together, the results presented in Figure 4 clearly demonstrate that pH is the most influential parameter affecting Pb^{2+} removal efficiency, followed by current density and reaction time within the investigated operational range. These findings are consistent with both the statistical analysis and the fundamental electrochemical mechanisms of the EC process, underscoring the critical importance of optimizing pH conditions while balancing energy input and treatment duration for efficient and sustainable wastewater treatment.

Optimization via design of experiments approach

Figure 5 presents the three-dimensional response surface plots and corresponding contour diagrams that describe the interactive effects of pH, current density, and reaction time on Pb^{2+} removal efficiency during the EC process. These plots, generated from the developed quadratic regression model, provide a comprehensive visualization of how simultaneous variations in two operational parameters influence system performance while the third parameter is held constant. Such analysis is essential for identifying optimal operating regions and understanding the synergistic or antagonistic relationships among process variables.

As illustrated in Figure 5(a), the combined effect of pH and current density reveals a pronounced dependence of Pb^{2+} removal efficiency on pH, particularly within the near-neutral range (approximately pH 7–8). The response surface exhibits a steep gradient along the pH axis, indicating that even small changes in pH can significantly affect removal performance. In contrast, current density exerts a secondary but consistently positive influence, as higher values enhance the rate of anodic dissolution and coagulant generation. The maximum removal efficiency is achieved at elevated pH levels in conjunction with moderate-to-high current densities, highlighting the dominant role of pH in controlling metal speciation and hydroxide precipitation, while current density primarily governs the kinetics of coagulant production. This observation is consistent with previous studies, which have reported optimal Pb^{2+} removal under near-neutral conditions due to the enhanced formation of insoluble metal hydroxides and increased adsorption capacity of flocs (Mollah et al., 2001; Kobya et al., 2003).

Figure 5(b) illustrates the interaction between pH and reaction time, revealing a strong synergistic relationship between these two parameters. Pb^{2+} removal efficiency increases markedly as both pH and reaction time increase, with the highest efficiencies observed at near-neutral pH combined with extended electrolysis durations. The pronounced curvature of the response surface indicates significant quadratic effects, suggesting that sufficient reaction time is essential to allow complete hydrolysis of aluminum species, growth of hydroxide flocs, and effective capture of Pb^{2+} ions. Longer reaction times facilitate enhanced aggregation, sweep flocculation, and stabilization of flocs, thereby improving removal efficiency. These findings are in agreement with the work of Holt et al. (2005) and Garcia-Segura et al. (2017), who emphasized the importance of adequate electrolysis duration in promoting floc development and pollutant removal, particularly under optimal pH conditions.

In contrast, Figure 5(c) presents the interaction between current density and reaction time at a constant pH, showing a comparatively weaker combined effect on Pb^{2+} removal efficiency. Although a gradual increase in removal efficiency is observed with increasing current density and reaction time, the response surface appears relatively flat compared to those involving pH, indicating limited interaction between these two parameters within the studied range. This suggests that once a sufficient rate of coagulant generation and adequate contact time have been achieved, further increases in current density or reaction time result in diminishing returns. From a practical perspective, this behavior highlights the importance of optimizing these parameters to balance treatment efficiency and energy consumption. Similar observations have been reported in previous EC studies, where excessive current density was found to increase operational costs without

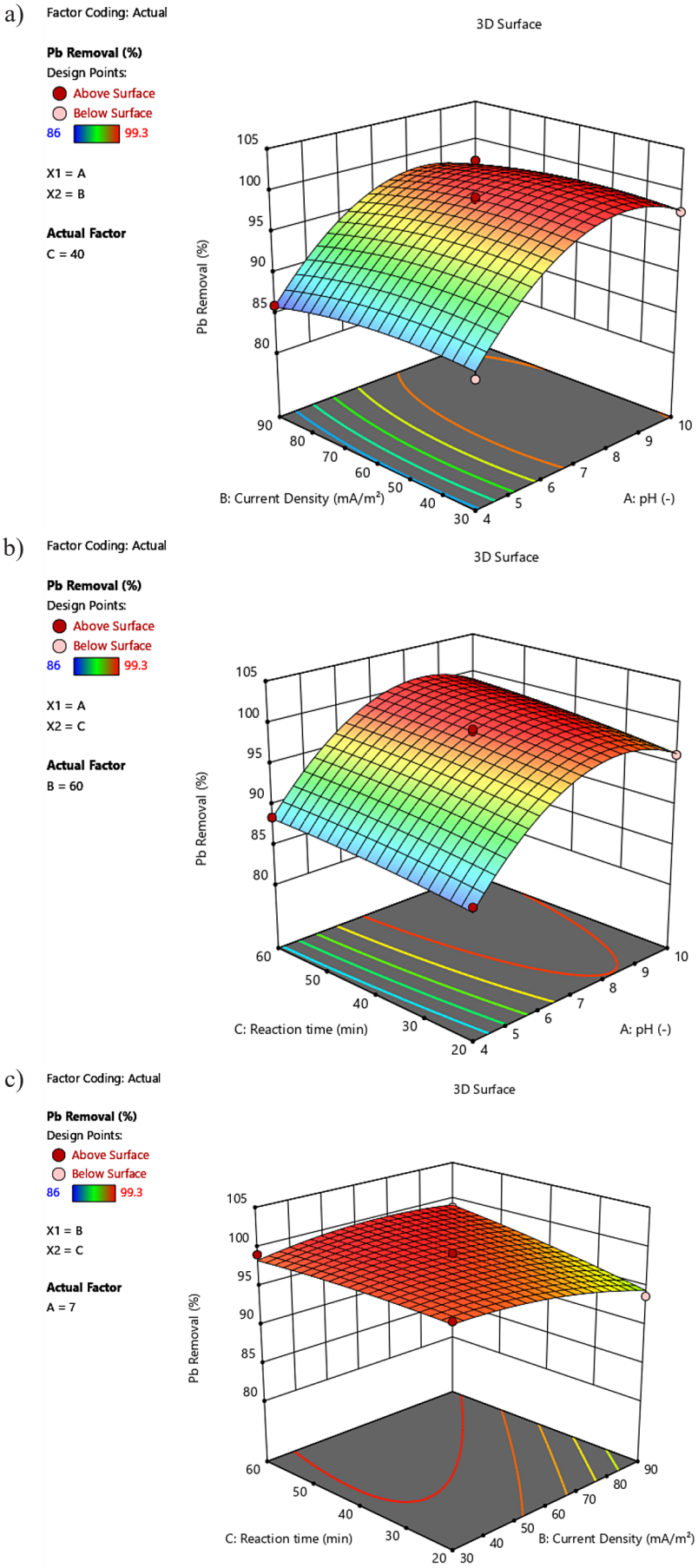

Figure 5. Three-dimensional plots depicting the interaction between operating variables (pH, current density, and reaction time) on Pb removal efficiency

yielding proportional improvements in pollutant removal (Chen, 2004; Sadik, 2019).

As a whole, the response surface analysis clearly demonstrates that pH is the most influential parameter governing Pb^{2+} removal efficiency, while current density and reaction time play complementary roles in enhancing process performance. The hierarchical importance of these factors – pH as the primary controlling variable, followed by current density and reaction time – aligns well with the fundamental electrochemical and physicochemical mechanisms of electrocoagulation, as well as with findings reported in the broader literature on heavy metal removal. These insights provide a robust scientific basis for process optimization and contribute to the effective design and scale-up of EC systems for wastewater treatment applications.

To determine the optimal operating conditions for maximizing Pb^{2+} removal efficiency, a quadratic model derived from RSM was employed. This model was selected based on its statistical significance, adequacy, and superior predictive performance, as confirmed by the ANOVA results and model diagnostics. The optimization objective was defined to achieve the highest possible Pb^{2+} removal efficiency within the experimental domain, while maintaining practical and energy-efficient operating conditions. In particular, the optimization strategy sought to minimize unnecessary electrical input and avoid excessive reaction times, thereby enhancing the economic feasibility of the electrocoagulation (EC) process.

The model predicted that optimal performance could be achieved at a solution pH of 7.6, a current density of 55 A/m^2, and a reaction time of 38.5 minutes. These conditions correspond to a near-neutral pH environment, which is known to favor the formation of amorphous aluminum hydroxide flocs with high adsorption capacity, combined with a moderate current density that ensures sufficient coagulant generation without excessive energy consumption. The selected reaction time provides adequate duration for floc formation, aggregation, and pollutant capture, while avoiding inefficiencies associated with prolonged electrolysis. The optimal pH range (7–8) identified in this study aligns with findings reported by Kobya et al. (2003) and Garcia-Segura et al. (2017), who attributed enhanced removal efficiency to the formation of amorphous $Al(OH)_3$ with high adsorption capacity. However, compared to these studies, the present work demonstrates slightly higher efficiency (>98%), likely due to optimized interaction effects captured through RSM.

To validate the reliability of the model, confirmatory experiments were conducted under the predicted optimal conditions. The results, summarized in Table 3, demonstrate a high degree of agreement between the experimentally observed Pb^{2+} removal efficiency and the model-predicted value. Specifically, the experimental removal efficiency reached 98.35%, while the model predicted a slightly higher value of 99.82%, resulting in an absolute deviation of only 1.47%. This small discrepancy indicates excellent model accuracy and confirms the robustness of the quadratic model in capturing the complex relationships among the process variables.

As shown in Table 3, the close correspondence between experimental and predicted values highlights the strong predictive capability of the developed model. The low absolute error further confirms that the model can reliably estimate Pb^{2+} removal efficiency within the studied parameter space. This level of agreement not only validates the statistical soundness of the quadratic model but also demonstrates its practical applicability for process optimization.

Collectively, the optimization results underscore the effectiveness of the electrocoagulation process in achieving high Pb^{2+} removal efficiency under well-defined operating conditions. The successful integration of experimental data with RSM-based modeling provides a powerful framework for process design, optimization, and potential scale-up, offering valuable guidance for the implementation of EC technology in real-world wastewater treatment applications.

Table 3. Optimal operating conditions and corresponding Pb removal efficiency for electrocoagulation treatment

Parameter / Response Optimal conditions	Value
pH	7.6
Current density (A/m^2)	55
Reaction time (min)	38.5
Pb removal efficiency (%)	
Experimental (actual)	98.35
Model-predicted	99.82
Absolute difference	1.47

CONCLUSIONS

This study systematically demonstrated the high efficiency and practical potential of EC as an advanced treatment technology for the removal of Pb^{2+} from aqueous solutions. Under optimized operating conditions, the EC process achieved removal efficiencies exceeding 98%, highlighting its strong capability for addressing heavy metal contamination in wastewater systems. The superior performance observed in this study can be attributed to the in situ generation of highly reactive metal hydroxide flocs, which facilitate multiple removal mechanisms, including adsorption, charge neutralization, and co-precipitation.

The integration of RSM with a Box-Behnken experimental design proved to be a robust and efficient approach for investigating the complex interactions among key operational variables. This statistical framework enabled the systematic evaluation of both individual and combined effects of pH, current density, and reaction time on Pb^{2+} removal efficiency, while minimizing the number of experimental runs required. The use of RSM not only facilitated the development of an empirical predictive model but also provided valuable insights into the nonlinear behavior and interaction effects inherent in the electrocoagulation process.

Among the investigated parameters, pH was identified as the most influential factor governing Pb^{2+} removal efficiency. Optimal performance was consistently observed under near-neutral conditions (pH ≈ 7–8), which favor the formation of amorphous aluminum hydroxide species with high adsorption capacity and strong affinity for metal ions. In contrast, current density and reaction time exhibited secondary yet complementary roles. Current density primarily controlled the rate of anodic dissolution and coagulant generation, while reaction time determined the extent of floc formation, growth, and pollutant capture. The interplay among these variables underscores the importance of balanced operating conditions to achieve both high removal efficiency and energy-efficient operation.

The developed quadratic regression model exhibited high statistical significance and excellent predictive capability, as reflected by a coefficient of determination (R^2) of 0.993. The strong agreement between experimental observations and model predictions confirms the reliability and robustness of the modeling approach. Furthermore, the non-significant lack-of-fit and the consistency of residuals provide additional evidence that the model adequately represents the experimental system and can be confidently used for prediction and optimization purposes.

The optimal operating conditions were determined to be a solution pH of 7.6, a current density of 55 A/m², and a reaction time of 38.5 minutes. Under these conditions, an experimental Pb^{2+} removal efficiency of 98.35% was achieved, which closely aligns with the model-predicted value of 99.82%. The minimal deviation between these values further validates the accuracy of the model and demonstrates its applicability for guiding process optimization and potential scale-up. Such a high level of agreement is indicative of a well-calibrated model capable of capturing the essential dynamics of the EC process.

The findings of this study highlight the significant potential of electrocoagulation as a cost-effective, efficient, and environmentally sustainable technology for the treatment of Pb-contaminated wastewater. Compared to conventional treatment methods, EC offers advantages such as reduced chemical consumption, simple operation, and the ability to handle a wide range of contaminants. Moreover, the successful application of RSM underscores the importance of employing systematic statistical optimization tools to enhance process performance and operational efficiency.

For future research, several aspects warrant further investigation to facilitate the practical implementation of EC technology. These include the application of the process to real and complex wastewater matrices, comprehensive evaluation of energy consumption and cost efficiency, assessment of electrode passivation and long-term durability, and the exploration of hybrid systems combining EC with other treatment technologies (e.g., electro-Fenton, membrane filtration, or adsorption). Addressing these challenges will be essential for improving the scalability, sustainability, and industrial applicability of electrocoagulation systems in wastewater treatment.

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