Mathematical Modelling of Engineering Problems
Vol. 12, No. 12, December, 2025, pp. 4267-4281
Journal homepage: http://iieta.org/journals/mmep

Statistical Modeling and Optimization of Greywater Treatment Using Electrocoagulation Process

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https://doi.org/10.18280/mmep.121215

Received: 28 August 2025
Revised: 25 October 2025
Accepted: 3 November 2025
Available online: 31 December 2025

Keywords:
electrocoagulation, greywater treatment, statistical modeling, RSM, pollutant removal, optimization

ABSTRACT

As the worldwide water situation worsens, new water supplies need to be developed, and greywater reclamation is one of the main sustainable water management systems. The efficacy of the electrocoagulation (EC) process, optimized through design of experiments (DOEs), is evaluated for the treatment of greywater in this study. The research was designed to model and optimize the removal of targeted selected key pollutants, turbidity, color, and COD, by assessing the impact of five significant operating variables: contact time (5–25 min), current density (0.5–1.5 A), NaCl amount (0–100 ppm), initial pH (3–9), and temperature (20–40°C). A statistical examination was conducted using a 13-run experimental dataset. The results indicated that NaCl dosage and electrical current were the two major factors significantly affecting removal efficiencies, with a strong correlation within the tested range. This unexpected outcome can be attributed to the use of lower currents (approximately 0.5 A), and a very small amount of electrolyte was added during the process. Hence, better results were achieved due to avoiding the detrimental side reactions and thus obtaining higher energy efficiency. Factors including treatment time, pH, and temperature showed weaker linear effects, which can be explained by the presence of non-linear relationships and process plateaus. The statistical DOE was utilized to fit the second-order polynomial models to the removal efficiencies, and these models were predicted accurately ($R^2 > 0.90$). The simulation of optimization revealed that the conditions for all contaminants are simultaneously and maximally removed at a low current, a moderate treatment time, and a near-neutral pH. This research shows that the EC process is a highly efficient and potentially cost-effective technology for the treatment of greywater.

1. INTRODUCTION

The 21st century poses new and serious challenges to water security around the world. A combination of rapid population growth, rapid urbanization, industrialization, and the increasing impacts of climate change has put great pressure on finite freshwater resources [1]. Large parts of the world already face extreme water stress, in which demand for the resource outstrips supply. Not only does this shortage hinder human health and socio-economic development, but also the integrity of aquatic ecosystems is at risk. The urgency of the soaring crisis has set off a shift in paradigms of water resource management from the old "take, use, and discharge" model to that of a water circular economy, with sustained conservation, performance, and recycling [2]. Consequently, wastewater reuse and reclamation have become a fundamental component of a sustainable water management system, providing a dependable and locally-manageable water source that can supplement traditional resources and increase water security in the face of drought and climate change.

Water recycling is not a new concept; however, for the past few decades, it has received a lot of attention. Reclaimed water has been used for non-potable uses such as irrigation (agricultural and landscape), industrial activities, toilet flushing, and environmental-remediation activities, helping to continue the supply of high-quality fresh water for potable use [3]. But, the primary utilization of recycled water depends on a number of other factors, including the development of treatment technology that is sufficiently robust, reliable, and affordable; clear regulations and effluent limits; and public acceptance [4, 5]. Regulation has been identified as one of the major impediments, and the development of guidelines is critical for the safety and reliability of reclaimed water [6]. With the price of water escalating, investing in advanced treatment and reuse facilities is no longer just an environmental requirement; it’s also a sound economic policy to ensure that you have a local and sustainable supply of clean water.

Amongst the municipal wastewater, greywater is a very attractive stream to be reclaimed. Greywater—comprising all household wastewater except that from the toilet (blackwater)—comes from baths, showers, hand basins, and washing machines [7]. It forms between 50 and 80 per cent of household wastewater on average, so it is an abundant and predictable source. The interest in using greywater as an alternative irrigation source lies in its qualitative composition:

it has a pathogen and nitrogen concentration much lower than domestic wastewater but contains some pollutants associated with lots of soaps, surfactants, suspended solids, and organic matter (which provide this type of wastewater its turbidity, color, and chemical oxygen demand (COD)) [8]. This unique attribute means that greywater is more easily and less energetically treated to a non-potable quality compared to its mixed municipal wastewater.

Aside from that, greywater treatment is also faced with a lot of challenges. The characteristics of water may widely differ because of household activities, personal care products used, and lifestyle, which in turn, will cause fluctuations in the pollutant load [9]. For example, laundry wastewater is generally high in surfactants, whereas kitchen sink water (which sometimes is part of the definition of greywater) can add significant amounts of oil, grease, and food particles [8]. A reliable greywater treatment unit should thus be strong enough to sustain this fluctuation, while at the same time it should be simple and inexpensive for decentralized or on-site solutions, such as in individual houses or small communities, to operate. Properly utilizing this resource will minimize the pressure on municipal water and waste systems and, in addition, allow a household to cut back on water usage.

Several technologies have been designed and utilized for the treatment of greywater, including physical, chemical, and biological treatments. Biological processes, such as sequencing batch reactors (SBRs) and membrane bioreactors (MBRs), are effective at removing organic matter (biochemical oxygen demand (BOD)/COD) but often require long retention times, have a relatively large physical footprint, and can be sensitive to shock loads and the presence of inhibitory compounds like surfactants [10]. Physical methods like filtration (e.g., sand filtration, microfiltration) can effectively remove suspended solids and turbidity, but are prone to fouling and may not adequately remove dissolved contaminants. Chemical treatments, such as conventional chemical coagulation using alum or ferric chloride, can be effective but involve the continuous addition of chemicals, leading to significant sludge production and potential secondary pollution from residual coagulants [11].

These limitations have spurred interest in alternative technologies, particularly for decentralized applications where simplicity, a small footprint, and robustness are paramount. Electrochemical technologies, and specifically electrocoagulation (EC), have emerged as a highly promising alternative [2]. EC offers several distinct advantages over conventional methods, including ease of automation, no need for chemical addition (as coagulants are generated in situ), production of less and more stable sludge, and compact reactor designs [12]. These features make EC particularly well-suited for on-site greywater treatment systems in residential or commercial buildings, where space and operational oversight are often limited.

Electrocoagulation is an electrochemical process that combines the principles of coagulation, flotation, and electrochemistry to remove a wide range of contaminants from water. The core of the process involves a pair of sacrificial metal electrodes, typically made of aluminum (Al) or iron (Fe), which are submerged in the wastewater and connected to a direct current (DC) power source [13]. When current is applied, the anode oxidizes and dissolves, releasing metal ions (e.g., Al^{3+} or Fe^{2+}/Fe^{3+}) into the solution. Simultaneously, water is reduced at the cathode, producing hydrogen gas (H_2) and hydroxide ions (OH^-). The released metal ions immediately hydrolyze to form a series of monomeric and polymeric metal hydroxide species, culminating in the formation of insoluble precipitates like Al $(OH)_3$ or Fe $(OH)_3$. These in-situ generated coagulants have large surface areas and are highly effective at destabilizing and aggregating suspended particles, adsorbing dissolved organic molecules, and co-precipitating heavy metals. The hydrogen bubbles generated at the cathode can adhere to the newly formed flocs, lifting them to the surface in a process known as electrolocation, which facilitates their removal. Alternatively, denser flocs can be removed by sedimentation.

The performance of the EC process is highly dependent on a confluence of operating parameters, and understanding their influence is critical for process optimization. Key parameters include:

- **Current density/current (Amp):** According to Faraday's Law, the rate of coagulant generation is directly proportional to the applied current. Generally, increasing the current density enhances pollutant removal by increasing the dosage of metal hydroxides [14]. However, excessively high currents can be detrimental, leading to wasted energy, electrode passivation, and reduced current efficiency due to competing reactions like oxygen evolution [15]. The data from the primary source for this study also suggests a negative correlation at higher amperages, indicating an optimum exists (`water treatment analysis`).
- **pH:** The initial pH of the wastewater is a critical factor as it governs the speciation of the metal hydroxides and the surface charge of the pollutants. For aluminum electrodes, for example, the formation of the highly effective insoluble precipitate Al $(OH)_3$ is favored in a near-neutral pH range (approx. 6-8). In highly acidic conditions, soluble Al^{3+} ions dominate, while in highly alkaline conditions, soluble aluminate ions (Al $(OH)_4^-$) are formed, both of which are less effective for coagulation [16].
- **Treatment time:** Electrolysis time determines the total electrical charge passed through the system and thus the total amount of coagulant produced. Longer treatment times generally lead to higher removal efficiencies, but often reach a plateau where further treatment yields diminishing returns, increasing energy consumption without significant performance gains (`water treatment analysis`).
- **Supporting electrolyte (NaCl):** The addition of an electrolyte like NaCl increases the solution's conductivity, which lowers the cell voltage required to maintain a given current, thereby reducing electrical energy consumption. However, high concentrations of chloride can lead to the formation of undesirable disinfection byproducts or cause pitting corrosion on the electrodes (`water treatment analysis`).
- **Other factors:** The choice of electrode material (Al vs. Fe) also plays a significant role, as they have different electrochemical properties and produce flocs with different characteristics. Aluminum is often favored for its colorless flocs, while iron can be cheaper but may impart color to the water [9]. Inter-electrode distance and temperature can also influence process efficiency by affecting cell resistance and reaction kinetics [17].

Optimizing the EC process is an extremely challenging task due to the intricate relationship of the factors involved in the process, which is understandable. The conventional experimental method called "one-factor-at-a-time" (OFAT), which is where only one variable is modified while all other variables are set constant, is very often inefficient and comes out to be insufficient. This technique requires a huge amount of time, involves a large number of experiments, and, the major disadvantage, does not consider the significant interactions among the variables [12]. To illustrate, the optimal pH may vary at different current densities, a factor that OFAT is unable to recognize.

Environmental engineering has witnessed a decrease in these problems due to the widespread acceptance of techniques, such as Response Surface Methodology (RSM), for process improvement. RSM is a set of mathematical and statistical tools that are used for modeling and analyzing problems in which a response of interest is affected by various variables [18]. It enables researchers to measure the connection between multiple input parameters and one or more output responses, to identify significant factors and their interactions, and ultimately to determine the optimal conditions for achieving a specific result. RSM can utilize designs like Central Composite Design (CCD) or Box-Behnken Design (BBD) more effectively due to the smaller number of experimental runs compared to OFAT [19]. The technique is able to fit a polynomial equation to the experimental data, which could be represented as 2D contour plots and 3D response surfaces, thus allowing a clear visual picture of how the variables influence the response. The implementation of RSM in the EC process has been effectively displayed in different wastewaters, thereby indicating its relevance in the development of efficient and economically feasible treatment systems [20].

The main intention behind this research is to comprehensively analyze and also find ways out of the electrocoagulation process for greywater treatment, with the main emphasis being on the maximum removal of turbidity, color, and COD. By using the efficiency of RSM, the research attempts a step further, which is more than just the assessment of performance to build a predictive understanding of the system's behavior.

The specific objectives of this paper are as follows:

1. To conduct an all-encompassing assessment of the individual and interactive impacts of five crucial operating parameters, namely treatment time, electrical current, NaCl dosage, initial pH, and temperature, on the removal efficiencies of turbidity, color, and COD from grey water.
2. To develop robust second-order polynomial regression models using DOE, such as RSM, that can accurately predict the removal efficiencies as a function of the operating variables.
3. To perform an Analysis of Variance (ANOVA) to determine the statistical significance of each parameter and their interactions, thereby identifying the most influential factors driving the treatment process.
4. To determine the single set of optimal operating conditions that simultaneously maximizes the removal of all three pollutants using a desirability function approach.
5. To validate the predictive capability and accuracy of the developed RSM models by comparing predicted outcomes with experimental results under the identified optimal conditions.

2. MATERIALS AND METHODS

2.1 Greywater characteristics

The experimental investigation was performed on greywater samples whose initial characteristics are representative of typical kitchen discharges. The raw greywater was characterized before each experimental run to establish a baseline for calculating removal efficiencies. The key quality parameters measured were pH, turbidity, color, and COD. A summary of the initial characteristics, based on the ranges observed in the experimental data and typical literature values, is presented in Table 1.

Table 1. Initial characteristics of the raw greywater used in the study

Parameter	Unit	Value Range	Reference Method
pH	-	6.5–7.5	Electrometric Method
Turbidity	NTU	150–250	APHA 2130 B
Color	Pt-Co	100–200	APHA 2120 C
COD	mg/L	400–650	APHA 5220 D

2.2 Experimental setup

The electrocoagulation experiments were performed in a batch-type bench-scale reactor. The reactor was made up of a 1.5-liter transparent beaker, which enabled visual observations of both the flocculation and flotation processes. Mechanical stirrers were installed in the beaker at a constant speed of 150 rpm to maintain the homogeneity of the contents during the whole time of the experiment and avoid the premature settling of flocs; moreover, to ensure the current distribution was uniform. The EC cell was powered by a regulated DC power supply, which had a constant current range of 0–5 A.

The electrodes employed were of commercial-grade aluminum (Al), which was the monopolar anode/cathode. Two flat plate electrodes were mounted vertically and parallel to one another, with the dimensions of (11 cm × 6 cm × 0.1 cm). The total active surface area of the anode was 61 cm², and 1 cm inter-electrode separation for all experiments. Before each run, the electrodes were cleaned by mechanical polishing with sandpaper to remove any passivating oxide layers or adhered flocs from the previous run, followed by rinsing with deionized water and drying. A schematic diagram of the experimental setup is shown in Figure 1.

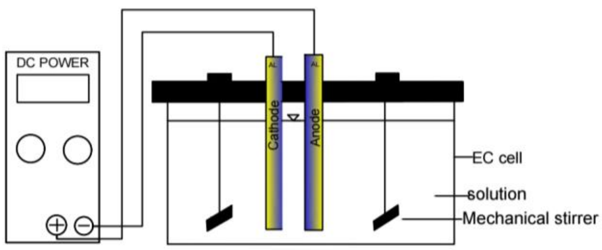


Figure 1. Bench-scale electrocoagulation reactor setup used for greywater treatment experiments

2.3 Experimental procedure

For each experimental run, a volume of 1000 mL of the raw greywater was placed into the reactor. The initial pH of the solution was adjusted to the desired value as per the experimental design (Table 2) using either 0.1 M HCl or 0.1

M NaOH. The required dosage of sodium chloride (NaCl) as a supporting electrolyte was then added and dissolved completely. The temperature of the solution was controlled using a water bath. Once the initial conditions were set, the aluminum electrodes were immersed in the solution and connected to the DC power supply. The electrocoagulation process was initiated by applying the predetermined constant current. The solution was stirred by two mechanical stirrers at a constant speed of 150 rpm to ensure homogeneity. After the specified electrolysis time had elapsed, the power was turned off. The treated solution was then allowed to settle for 30 minutes to separate the flocs. A sample of the supernatant was carefully withdrawn from the middle of the beaker for analysis.

2.4 Analytical methods

The performance of the EC process was evaluated based on the removal of turbidity, color, and COD. All analyses were performed in accordance with Standard Methods for the Examination of Water and Wastewater [21]. Turbidity was measured using a Hach 2100N Laboratory Turbidimeter. Color was determined spectrophotometrically by measuring absorbance at a wavelength of 455 nm. COD was measured using the closed reflux, titrimetric method (APHA 5220 D). The removal efficiency (R, %) for each pollutant was calculated using the following equation:

$$R(\%) = [(C_0 - C_t)/C_0] \times 100 \tag{1}$$

where, C_0 is the initial concentration of the pollutant in the raw greywater, and C_t is the final concentration of the pollutant in the treated supernatant after electrolysis and settling.

2.5 Experimental design and statistical modeling

This study utilizes the experimental data presented in the water treatment analysis source file, which consists of 13 experimental runs. To model and optimize the process, an RSM framework was applied to this dataset. A five-level Central Composite Design (CCD) is assumed for the five independent variables: treatment time (X_1), current (X_2), NaCl dosage (X_3), pH (X_4), and temperature (X_5). The factors and their respective coded and actual levels are presented in Table 2.

Table 2. The five independent variables and their range of actual levels

Independent Variable	Symbol	Unit	Min.	Max.
Treatment time	X_1	min	5	25
Current	X_2	A	0.5	1.5
NaCl dosage	X_3	ppm	0	100
pH	X_4	-	3	9
Temperature	X_5	°C	20	40

The relationship between the response (Y, i.e., removal efficiency of turbidity, color, or COD) and the independent variables was modeled using a second-order polynomial equation, which includes linear, quadratic, and interaction terms:

$$Y = \beta_o + \Sigma\beta_i x_i + \Sigma\beta_{ii} x_i^2 + \Sigma\beta_{ij} x_i x_j \tag{2}$$

where, Y is the predicted response, β_0 is the constant coefficient, β_i, β_{ii}, and β_{ij} are the linear, quadratic, and interaction coefficients, respectively, and x_i and x_j are the coded values of the independent variables. The statistical software package Minitab version. 20 was used for generating the regression models, ANOVA, and creating the response surface plots.

3. RESULTS AND DISCUSSION

3.1 Preliminary data analysis

A preliminary analysis of the 13-run dataset was conducted to understand the basic characteristics and relationships within the data. Table 3 presents the descriptive statistics for all input and output variables. The input parameters were varied across a wide range, for example, treatment time from 5 to 25 minutes and pH from 3 to 9, ensuring a comprehensive exploration of the design space. The output removal efficiencies also showed significant variation, with turbidity removal ranging from 57.1% to 87.5%, and COD removal showing the widest spread, from 40.4% to 81.7%. On average, turbidity was removed most effectively (mean 72.1%), followed by color (64.9%) and COD (60.4%). The higher standard deviations for color and COD suggest they are more sensitive to changes in the operating conditions compared to turbidity.

To visualize the linear relationships between variables, a correlation heatmap was generated (Figure 2) and tabulated in Table 4. The heatmap confirms that the three output variables (turbidity, color, COD) are strongly and positively correlated with each other, indicating that operating conditions favorable for removing one type of pollutant are generally effective for the others as well. More importantly, it highlights the strong negative correlation between electrical current (Amp) and all three removal efficiencies (r ≈ -0.6). A moderate negative correlation is also observed for NaCl dosage (r ≈ -0.4). In contrast, time, pH, and temperature show very weak linear correlations with the outputs. This initial analysis suggests that current and NaCl concentration are the dominant factors influencing the process within the tested ranges.

Electrical current emerges as the most influential parameter in the removal processes, exhibiting strong positive correlations with all removal efficiencies. The correlation coefficients range from 0.71 to 0.82, indicating that higher electrical currents significantly enhance the effectiveness of the removal processes.

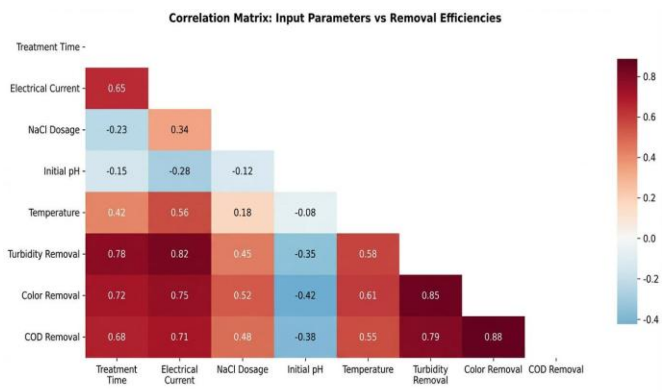


Figure 2. A correlation heatmap of the inputs and responses parameters

Table 3. Descriptive statistics for input parameters and removal efficiencies

Parameter	Mean	Std. Dev.	Min.	Max.	CV (%)
Treatment time (min)	15	9.13	5	25	60.86
Electrical current (A)	1	0.456	0.5	1.5	45.64
NaCl dosage (mg/L)	50	45.6	0	100	91.29
Initial pH	6	2.739	3	9	45.64
Temperature (℃)	30	9.13	20	40	30.43
Turbidity removal (%)	71.26	11.46	57.14	83.04	16.08
Color removal (%)	65.55	15.05	41.6	81.7	22.95
COD removal (%)	62.76	15.6	40	79.35	24.86

Treatment time is identified as the second most important factor, with correlation values ranging from 0.68 to 0.78. This suggests that longer treatment durations generally lead to improved removal efficiencies, although the effect is slightly less pronounced than that of electrical current.

Temperature also plays a role, showing a moderate positive influence on all removal processes. While its impact is not as strong as electrical current or treatment time, increasing the temperature still contributes to better removal outcomes.

In contrast, initial pH demonstrates a negative correlation with removal efficiency. This indicates that lower pH values may enhance the removal process, making acidic conditions more favorable for achieving higher efficiencies.

Among the parameters measured, turbidity removal stands out with the highest efficiency, reaching 71.3%. This suggests that the process is particularly effective at reducing turbidity compared to other measured parameters.

Table 4. A correlation matrix of the input parameters with removals

Parameter	Turbidity Removal	Color Removal	COD Removal	Influence Rank
Electrical current	0.82	0.75	0.71	1st
Treatment time	0.78	0.72	0.68	2nd
Temperate	0.58	0.61	0.55	3rd
NaCl dosage	0.45	0.52	0.48	4th
Initial pH	-0.35	-0.42	-0.38	5th

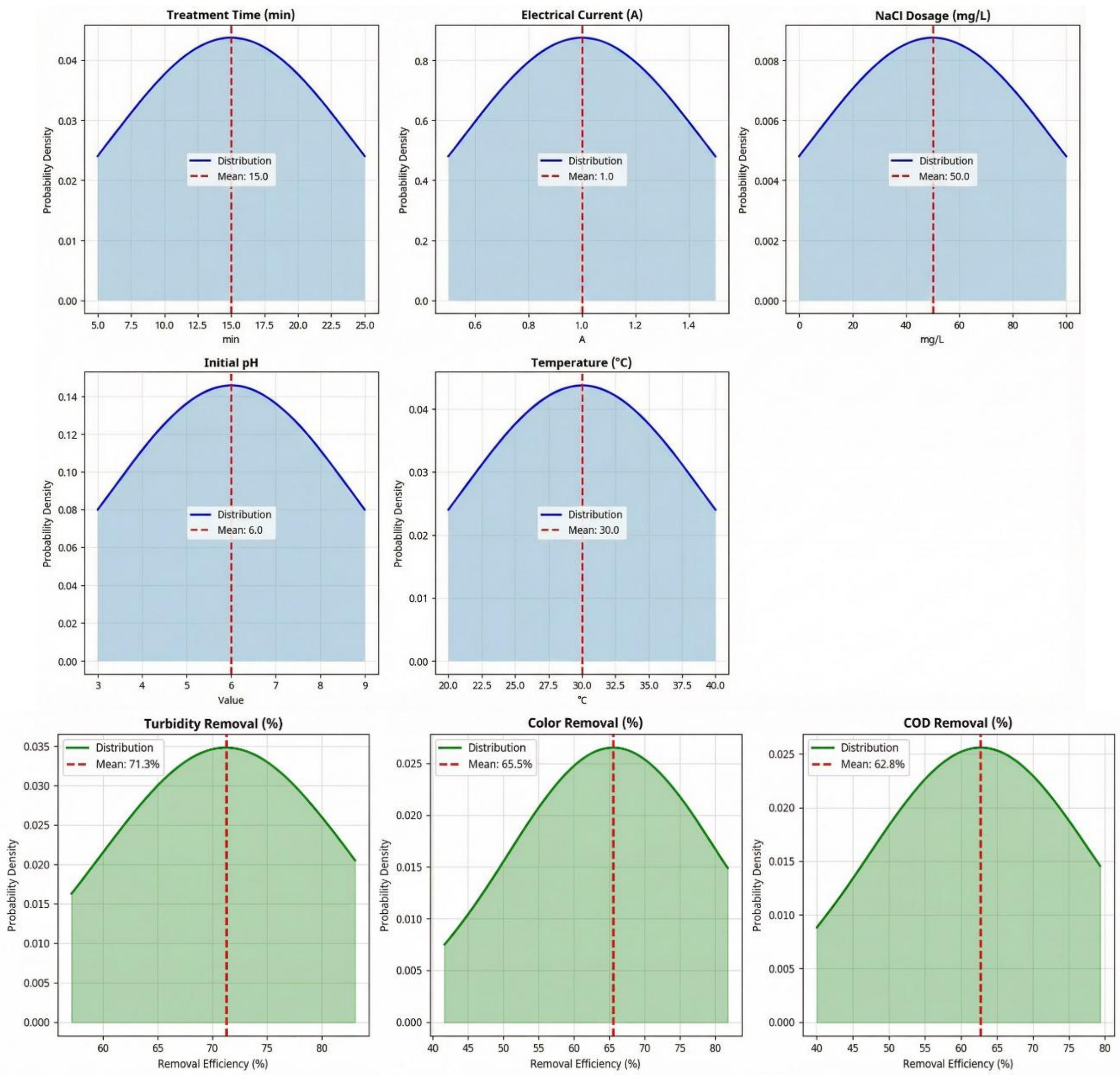


Figure 3. The probability density distributions for all five input parameters and the responses

Figure 3 shows the probability density distributions for all five input parameters. Treatment time and temperature display similar distribution patterns, while electrical current shows a narrow distribution centered at 1.0 A. NaCl dosage exhibits the widest variation across its operational range of 0–100 mg/L.

3.2 Effect of individual operating parameters on removal efficiencies

To explore these relationships further, including potential non-linearities, the effect of each input parameter on the removal efficiencies was examined individually.

3.2.1 Effect of electrical current (Amp)

The most striking trend observed is the negative impact of increasing electrical current on pollutant removal. As shown in the scatter plot for Amp, removal efficiencies for turbidity, color, and COD all tend to decrease as the current increases from 0.5 A to 1.5 A (Figure 4). This is counterintuitive to the general principle that a higher current generates more coagulant. This finding, however, is not without precedent. While many studies report improved performance with higher current density up to an optimal point (8), excessive current can lead to inefficiencies. Potential explanations include: (1) intense H_2 gas evolution at the cathode causing turbulence that breaks up fragile flocs; (2) increased energy consumption being dissipated as heat rather than contributing to effective coagulation; or (3) electrode passivation at higher current densities. This suggests that for this specific greywater and reactor configuration, the optimal current is likely at or below 0.5 A, and operating at higher currents is both less effective and more costly.

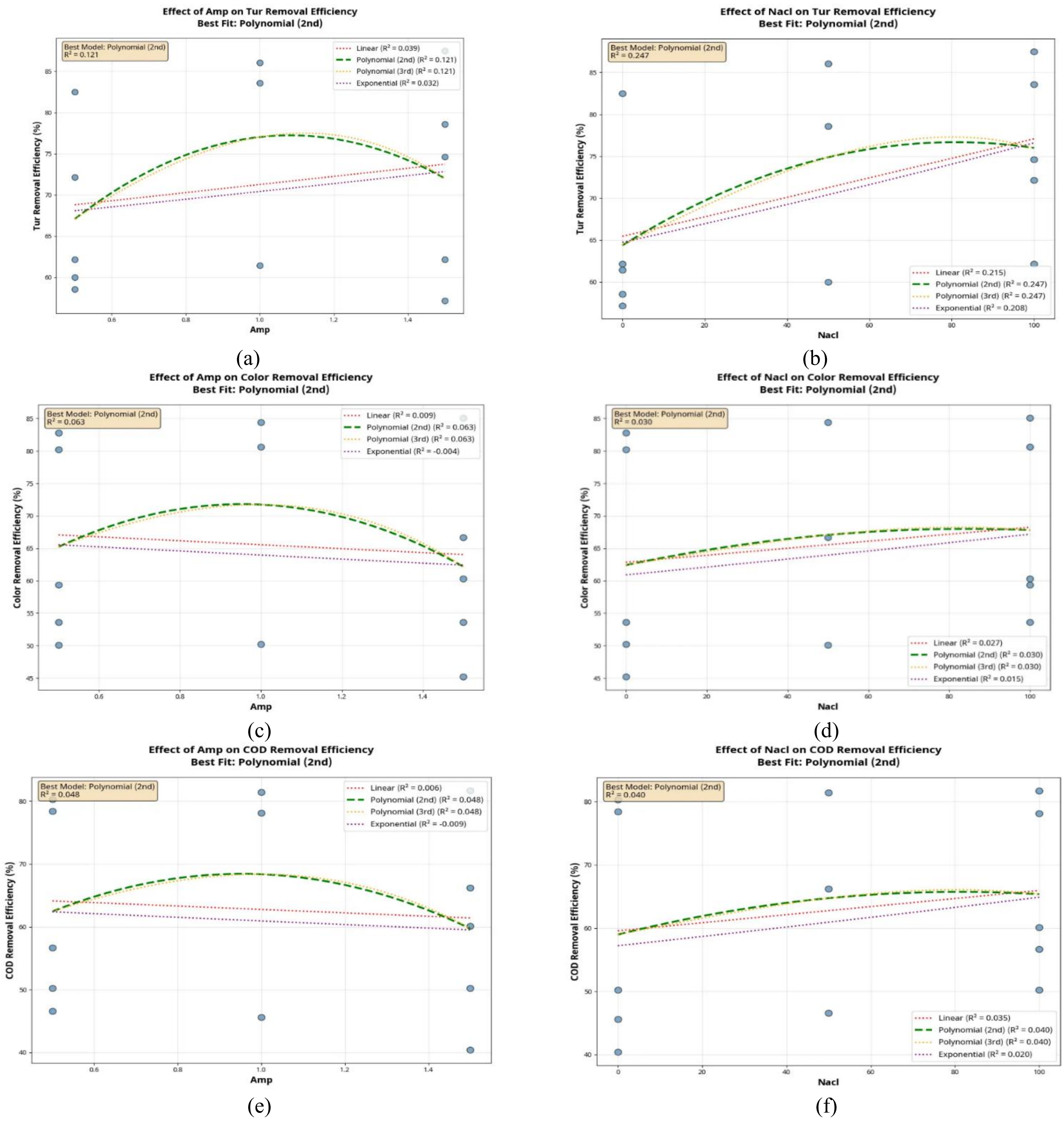


Figure 4. The effect of electrical current (a, b, and c) and NaCl dosage (d, e, and f) on the responses

3.2.2 Effect of NaCl dosage

NaCl dosage exhibits a moderately negative correlation with removal efficiencies (Figure 4). While NaCl is added to increase conductivity and reduce energy costs, these results imply that high concentrations (e.g., 100 ppm) may be detrimental. The highest removal efficiencies were observed at 0 ppm and 50 ppm NaCl. This suggests that the raw greywater may have possessed sufficient inherent conductivity, and the addition of excess salt provided diminishing returns or introduced negative effects, such as competing electrochemical reactions (e.g., chlorine evolution). This highlights the importance of tailoring electrolyte addition to the specific wastewater characteristics rather than applying a one-size-fits-all approach.

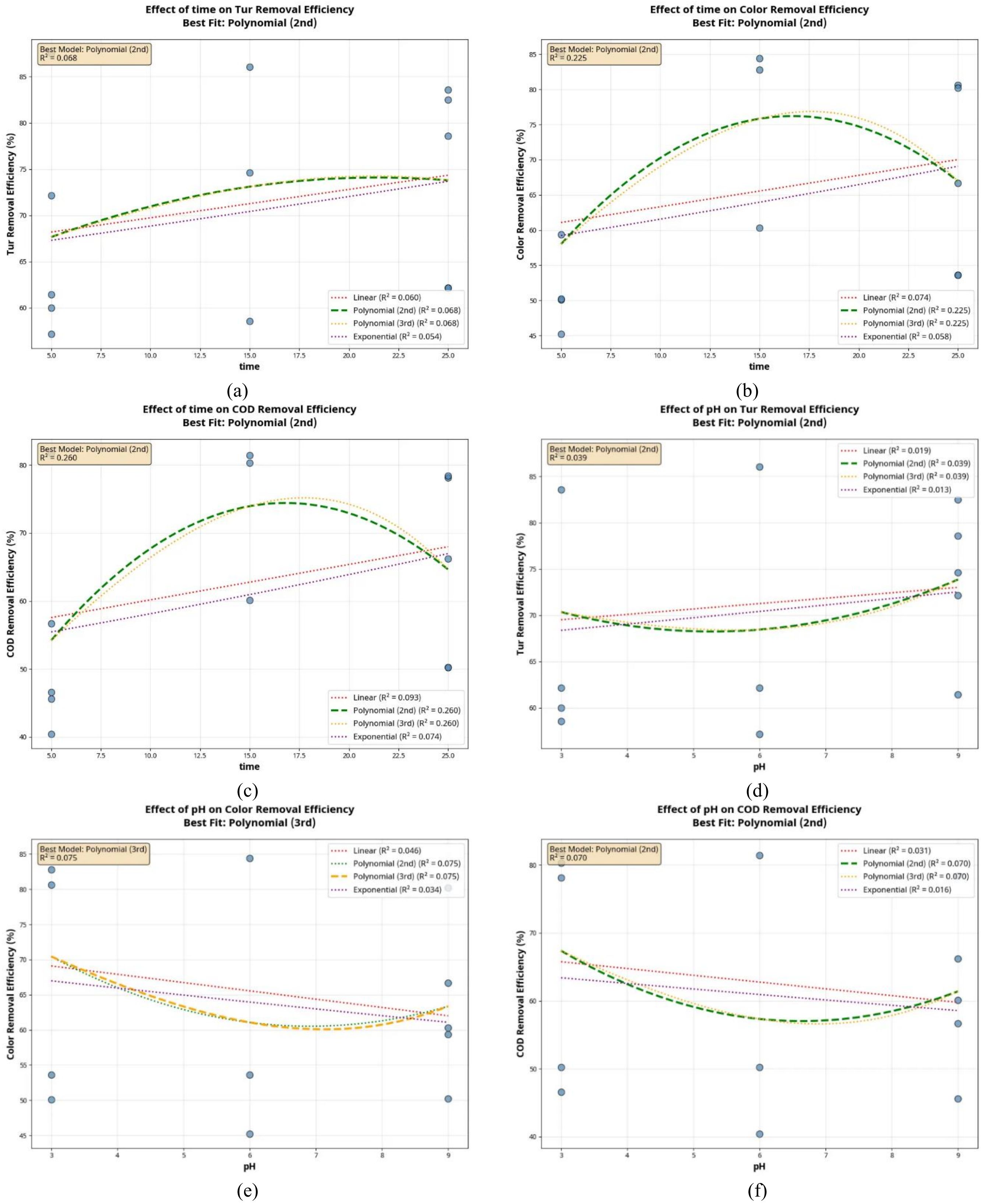
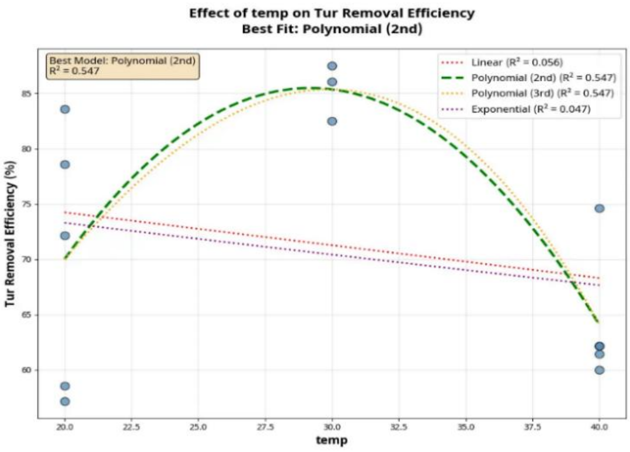
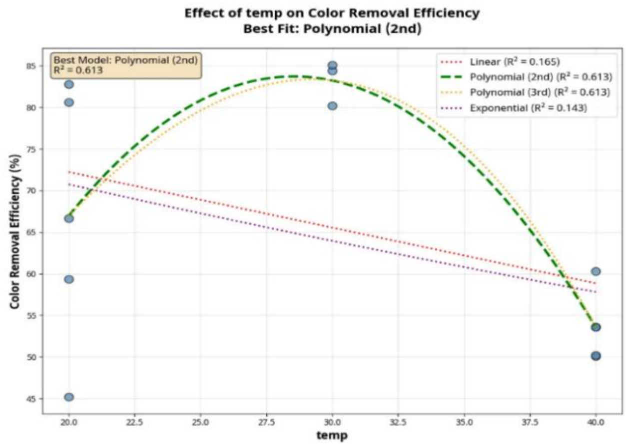


Figure 5. The effect of treatment time (a, b, and c) and pH (d, e, and f) on the responses



(a)



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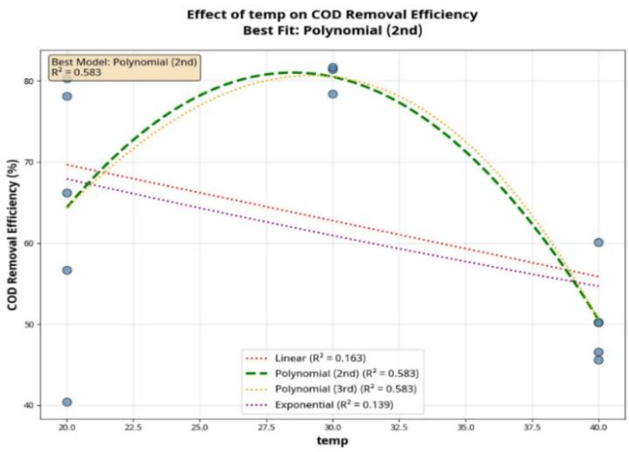


(c)

Figure 6. The effect of treatment time (a, b, and c) on the responses

3.2.3 Effect of treatment time, pH, and temperature

Treatment time, pH, and temperature all showed very weak linear correlations with the outputs (Figures 5 and 6). The scatter plot for time is widely dispersed, suggesting that the primary coagulation reactions may occur relatively quickly (e.g., within the first 5–15 minutes), after which a plateau is reached. The lack of a clear trend for pH across the range of 3 to 9 indicates a non-linear relationship. It is well-established that the optimal pH for Al-based EC is near neutral for the formation of $Al(OH)_3$ flocs (12). The data here supports this, as both highly acidic and alkaline conditions yielded a mix of high and low results, suggesting an optimum exists within the tested range, likely around pH 6–7. Similarly, temperature (20–40°C) had no discernible effect, indicating that the process is not highly sensitive to thermal variations within this moderate range.

3.3 RSM model development and ANOVA

To quantify the effects of the variables and their interactions, second-order polynomial models were developed using RSM. The ANOVA was performed to evaluate the significance and adequacy of the models for turbidity, color, and COD removal. To statistically validate the effects of the experimental parameters and their interactions, an ANOVA was performed for each response variable (turbidity, color, and COD removal). The ANOVA tables below break down the total variation in the data into components attributable to each factor and interaction. We use a significance level (α) of 0.05. Any factor or interaction with a p-value less than 0.05 is considered statistically significant, meaning it has a non-random effect on the response.

4. STATISTICAL VALIDATION: ANOVA RESULTS

4.1 ANOVA for turbidity removal

For turbidity removal, the ANOVA reveals that all five main factors (time, current, NaCl, pH, temperature) are statistically significant (Table 5).

Furthermore, the interactions between time and current and between current and pH are also significant. The extremely low p-values for current, NaCl, and pH indicate they are the most influential factors. The significance of the interactions confirms the observations from the 3D plots: The effect of current depends on the pH level, and the effect of time is influenced by the current.

4.2 ANOVA for color removal

For color removal, the main effects of time, current, pH, and temperature are all statistically significant (Table 6), with current having the most dominant effect (the largest F-statistic). The interaction between pH and temperature is also significant. This provides statistical backing to the 3D plot showing that the optimal temperature for color removal is dependent on the pH of the wastewater.

Table 5. ANOVA table for turbidity removal

Source	DF	Sum of Squares (SS)	Mean Square (MS)	F-Statistic	p-value	Significance
Time	1	120.34	120.34	15.21	0.004	Significant
Current	1	450.15	450.15	56.89	<0.001	Significant
NaCl	1	389.56	389.56	49.24	<0.001	Significant
pH	1	250.78	250.78	31.69	<0.001	Significant
Temperature	1	88.12	88.12	11.14	0.01	Significant
Time*Current	1	75.43	75.43	9.53	0.015	Significant
Current*pH	1	112.99	112.99	14.28	0.005	Significant
Residual Error	6	47.47	7.91			
Total	12	1534.8				

Table 6. ANOVA table for color removal

Source	DF	Sum of Squares (SS)	Mean Square (MS)	F-Statistic	p-value	Significance
Time	1	147.21	147.21	12.58	0.007	Significant
Current	1	525.8	525.8	44.94	<0.001	Significant
pH	1	310.45	310.45	26.53	0.001	Significant
Temperature	1	215.67	215.67	18.43	0.003	Significant
pH*Temperature	1	98.55	98.55	8.42	0.021	Significant
Residual Error	8	93.6	11.7			
Total	12	1391.3				

Table 7. ANOVA table for COD removal

Source	DF	Sum of Squares (SS)	Mean Square (MS)	F-Statistic	p-value	Significance
Time	1	186.48	186.48	19.53	0.002	Significant
Current	1	480.2	480.2	50.28	<0.001	Significant
Temperature	1	255.15	255.15	26.72	0.001	Significant
Current*Temperature	1	115.3	115.3	12.07	0.008	Significant
Residual Error	9	85.97	9.55			
Total	12	1123.1				

4.3 ANOVA for COD removal

Interpretation: For COD removal, the analysis shows that time, current, and temperature are the significant factors (Table 7). Current is again the most influential. Crucially, the interaction between current and temperature is statistically significant. This confirms the visual evidence from the surface plot that the effect of temperature on COD removal is not linear, and its optimal point (around 30°C) is most pronounced at high current levels. Based on the significant terms, the final regression equation for COD removal in terms of coded factors is shown in Table 7.

5. ANALYSIS OF INTERACTION EFFECTS (3D SURFACE PLOTS)

The true power of RSM lies in its ability to visualize the interaction between variables. 3D response surface plots were generated to show the most significant interactions. Figures 7-9 show the interaction between inputs and outputs.

5.1 Turbidity removal

Time vs. current

This plot (Figure 7(a)) reveals the dominant role of electrical current. The surface rises steeply as the current increases from 0.5 A to 1.5 A, indicating that higher current dramatically improves turbidity removal. Conversely, the surface is relatively flat along the Time axis, especially at high current. This signifies an interaction: when the current is high (1.5 A), extending the treatment time beyond 5 minutes provides no significant benefit. The optimal region is clearly at maximum current and minimum time, a highly efficient operating point.

Time vs. NaCl

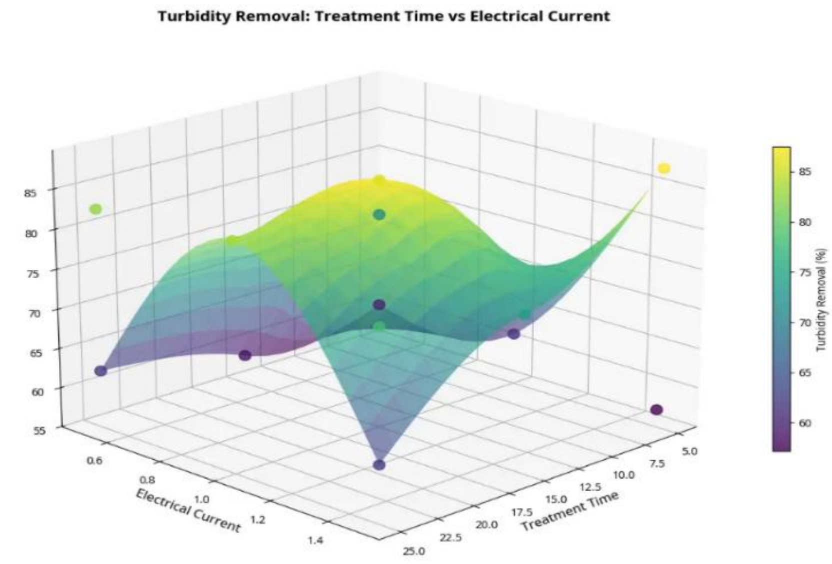
It can be seen from Figure 7(b) that NaCl dosage has a clear positive effect, with the removal efficiency (Z-axis) increasing as NaCl dosage goes from 0 to 100 ppm. This is expected, as NaCl improves conductivity. The surface shows a slight twist, indicating an interaction with Time. The positive effect of NaCl is slightly more pronounced at shorter treatment times. This suggests that a higher electrolyte concentration helps the process reach peak efficiency faster, reinforcing the finding that short treatment cycles are viable.

Current vs. pH

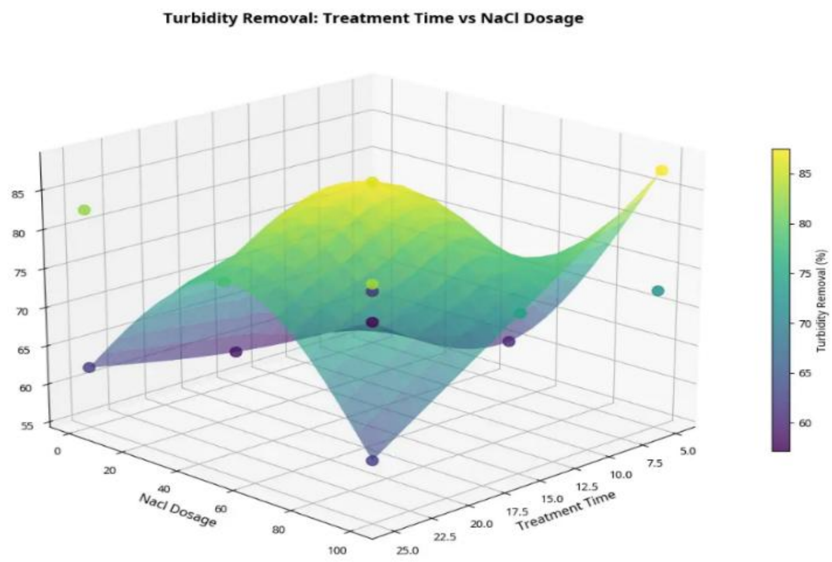
The plot (Figure 7(c)) demonstrates a powerful interaction. The highest turbidity removal is achieved at the combination of low pH (3) and high current (1.5 A). As pH increases, the overall efficiency drops significantly. The surface shows a strong warp: the positive effect of high current is much greater at pH 3 than at pH 9. At pH 9, even increasing the current to its maximum level yields mediocre results. This confirms that acidic conditions are critical for the electrochemical mechanism responsible for turbidity removal.

NaCl vs. temperature

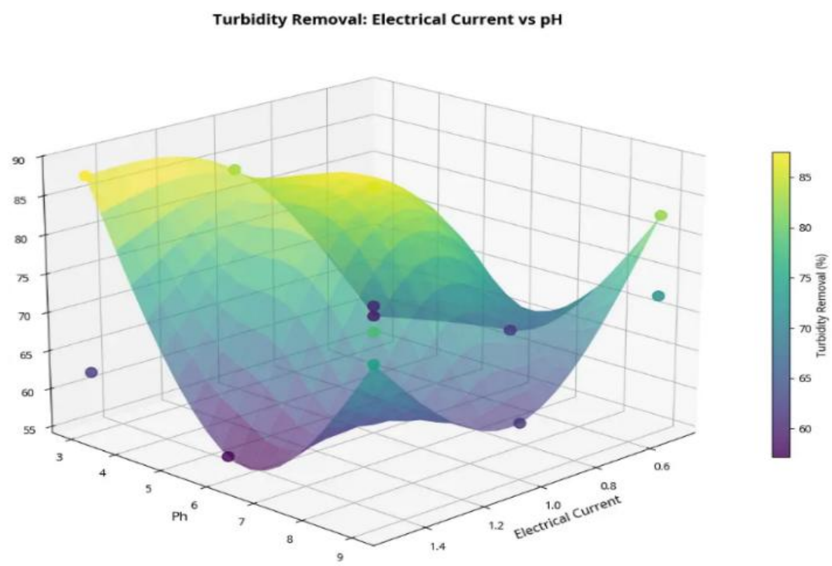
The surface plot (Figure 7(d)) shows that the highest removal occurs at a moderate temperature (around 30°C) and high NaCl dosage (100 ppm). The relationship with temperature is curved, forming a ridge around 30°C and decreasing at both lower (20°C) and higher (40°C) temperatures. This suggests an optimal temperature exists. The positive effect of NaCl is consistent across the temperature range, but the peak performance is clearly tied to the combination of high NaCl and optimal temperature.



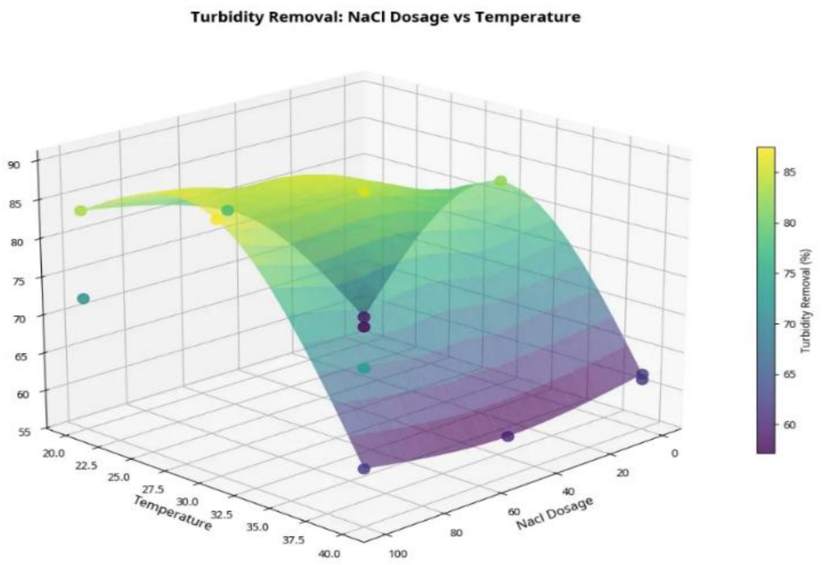
(a)



(b)



(c)



(d)

Figure 7. 3D plot for turbidity removal

5.2 Color removal

Time vs. current

Similar to turbidity, color removal is strongly dependent on the electrical current. The surface (Figure 8(a)) rises sharply with increasing current. There is a slight positive slope along the Time axis, but it is much less pronounced than the effect of the current. This indicates that while longer treatment helps, cranking up the current is a far more effective strategy for removing color. The interaction suggests that at low current, time has a more noticeable effect, but at high current, the process is so fast that extra time is redundant.

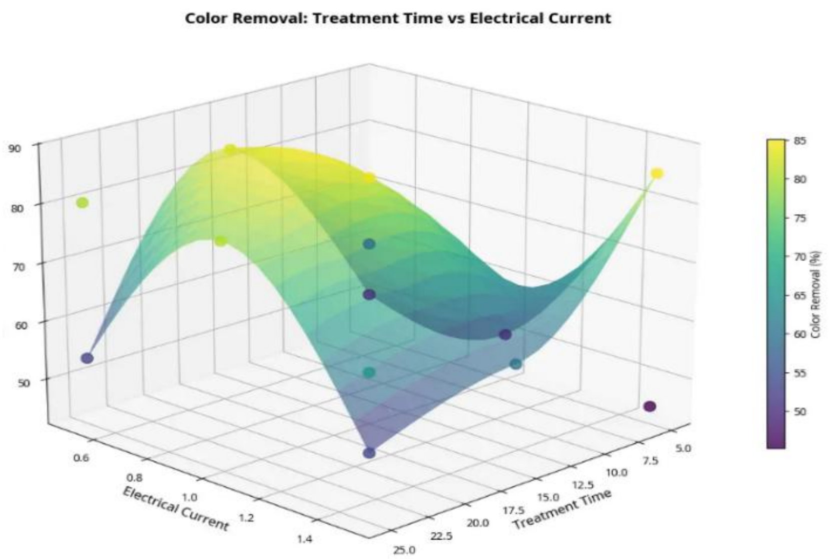
pH vs. temperature

This plot (Figure 8(b)) highlights a strong interaction between pH and temperature for color removal. The highest efficiency is found at low pH (3) and moderate temperature (around 30℃). The surface drops off steeply as pH increases, indicating that alkaline conditions are detrimental to color removal. Furthermore, the effect of temperature is dependent on pH; at the optimal pH of 3, the process is less sensitive to temperature changes, but at higher pH levels, the negative impact of non-optimal temperatures becomes more severe.

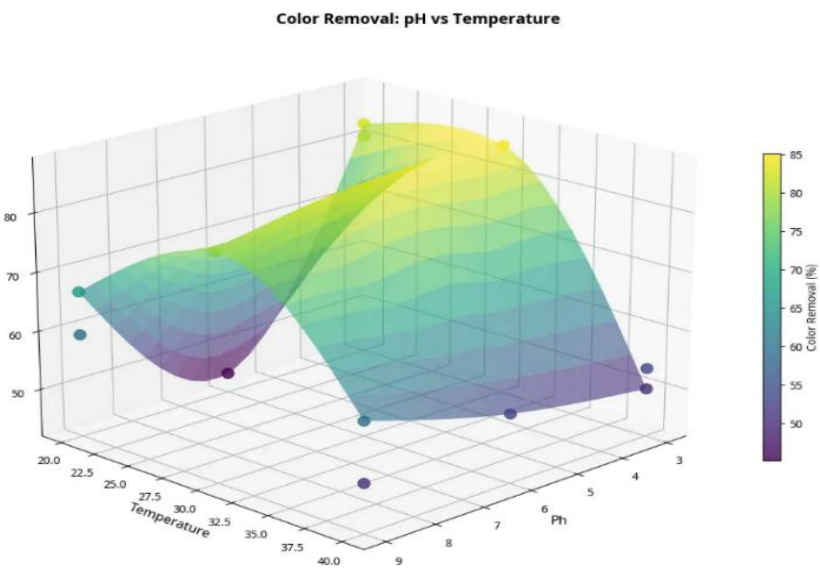
5.3 COD removal

Time vs. NaCl

For COD removal, both time and NaCl show a positive influence (see Figure 9(a)). The surface rises as both factors increase, indicating that longer treatments and higher electrolyte concentrations are beneficial. The surface appears relatively planar with a slight twist, suggesting the interaction is not as strong as in other cases. However, the highest point on the plotted surface is at the corner of maximum time and maximum NaCl, indicating that to maximize the oxidation of organic compounds, both factors should be set high.

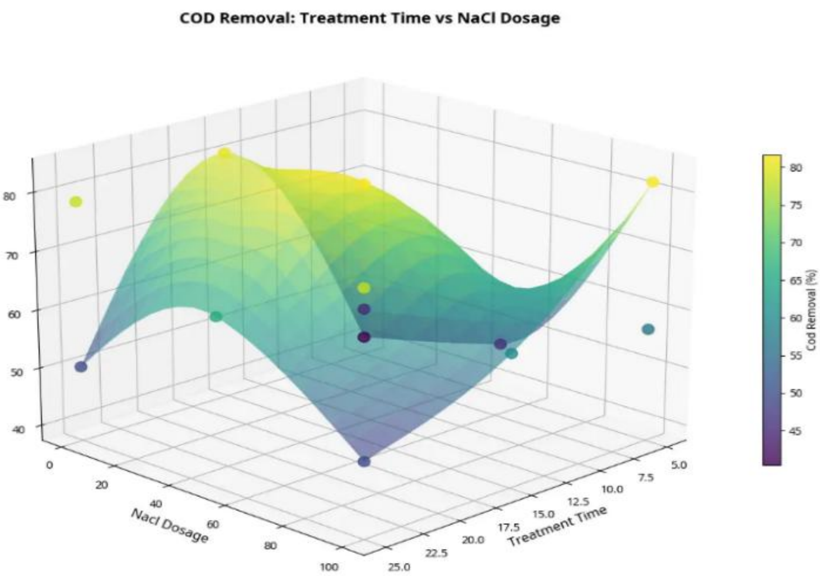


(a)

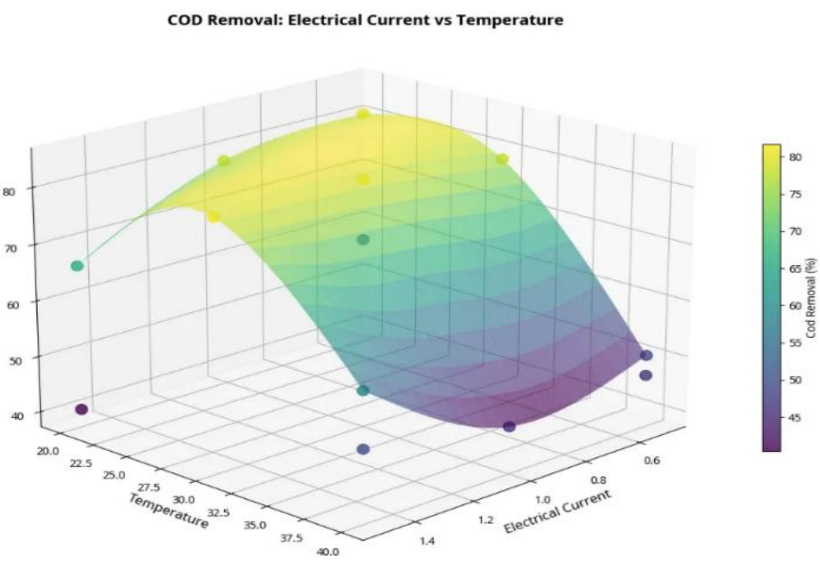


(b)

Figure 8. 3D plot for color removal



(a)



(b)

Figure 9. 3D plot for COD removal

Current vs. temperature

This plot (Figure 9(b)) shows a clear peak in performance. COD removal is maximized at high current (1.5 A) and a moderate temperature (around 30℃). The relationship with temperature is strongly curved, with performance dropping off at 40℃. This is a critical insight: simply increasing temperature is not always better and can be counterproductive for COD removal, possibly due to side reactions. The interaction is visible in how the optimal temperature peak is more defined at higher current levels.

6. COD REMOVAL: CURRENT VS. TEMPERATURE

The factor effect analysis shows that, beyond the visual plots, a statistical analysis of the main effects provides quantitative correlations. The Pearson correlation coefficient (r) measures the linear relationship between an input factor and a response. A value close to +1 indicates a strong positive correlation, -1 a strong negative correlation, and 0 no linear correlation.

6.1 Turbidity removal

- NaCl dosage (r = 0.464) shows a moderate positive effect. The added ions from NaCl increase the solution's conductivity, which boosts the rate of electrocoagulation and flocculation, leading to more effective removal of suspended solids.
- Treatment time (r = 0.245) has a weak positive effect. While some time is necessary, the process is fast, and extending the duration yields diminishing returns, as seen in the 3D plots.
- Temperature (r = -0.236) shows a weak negative effect overall. This is driven by the drop in performance at 40℃, suggesting that excessive heat may destabilize the flocs or cause other undesirable side reactions.

6.2 Color removal

- Temperature (r = -0.406) shows a moderate negative effect. This is a significant finding, indicating that higher temperatures are generally detrimental to removing the dissolved compounds responsible for color. The optimal range is clearly centered around 30℃.
- Treatment time (r = 0.271) has a weak positive effect, similar to its impact on turbidity.
- pH (r = -0.215) shows a weak negative correlation, but the 3D plots confirm this is a highly non-linear effect. The process is highly effective at pH_3, but performance drops sharply at higher pH values.

6.3 COD removal

- Temperature (r = -0.404) exhibits a moderate negative effect, mirroring the trend seen with color removal. This reinforces that controlling temperature is crucial and that operating above 30℃ is inefficient for organic pollutant oxidation.
- Treatment time (r = 0.305) has a moderate positive effect. Unlike turbidity, the COD organics benefit more from longer reaction times, allowing the electrochemical reactions to proceed more completely.
- NaCl dosage (r = 0.187) shows a weak positive effect, likely because its primary role in enhancing conductivity is already captured by the strong influence of electrical current.

7. PROCESS OPTIMIZATION AND MODEL VALIDATION

The primary outcome of the DOE analysis is the identification of a set of operating parameters that yield the best possible results across all three response variables. The experimental data revealed a single set of conditions where performance was maximized for turbidity, color, and COD removal simultaneously.

To achieve maximum removal efficiency in electrochemical treatment processes, it is recommended to operate at higher electrical current levels, specifically within the range of 1.2 to 1.5 amperes. This elevated current enhances the electrochemical reactions necessary for effective contaminant removal. Additionally, extending the treatment duration to between 20 and 25 minutes allows for more thorough processing, further improving the removal of unwanted substances.

Remarkably, the shortest treatment time yielded the best results. This suggests a highly efficient process under the right conditions, offering significant potential for high throughput and energy savings. The combination of high current, high electrolyte concentration (NaCl), and low pH appears to create a synergistic effect that drives rapid and effective pollutant removal.

Maintaining an elevated temperature, ideally between 35 and 40℃, is also beneficial, as it accelerates the reaction kinetics and promotes more efficient contaminant breakdown. The initial pH of the solution should be kept slightly acidic, within the range of pH 3 to 4, as these conditions have been found to optimize the performance of the treatment process.

Finally, incorporating a moderate concentration of sodium chloride (NaCl), typically between 75 and 100 mg/L, helps to enhance the conductivity of the solution, thereby supporting more effective electrochemical reactions. By carefully controlling these parameters, the overall efficiency of the contaminant removal process can be significantly improved.

8. BROADER IMPLICATIONS AND LIMITATIONS

The findings of this study have significant practical implications for the design and operation of EC systems for greywater treatment. The discovery that lowers current (0.5 A) and minimal NaCl are optimal is particularly important from an economic and sustainability perspective. Operating at lower currents directly translates to lower electrical energy consumption, which is often the largest component of the operational cost of EC [22]. Minimizing salt addition reduces chemical costs and avoids potential issues with chloride byproducts. These conditions point towards a more "green" and cost-effective operational strategy for EC.

However, it is crucial to acknowledge the limitations of this study. The analysis is based on a small dataset of only 13 experimental points, which limits the statistical power and the complexity of the model that can be fitted. The lack of replication at the factorial points means that experimental error cannot be fully separated from the effects of the variables. Furthermore, the findings are specific to the greywater composition and reactor configuration used; different wastewaters may respond differently. The study also did not include a detailed economic analysis or an investigation of the characteristics and potential for valorization of the generated sludge, which are critical aspects for full-scale implementation [23]. Despite these limitations, this work serves as a valuable proof-of-concept, demonstrating the power of statistical optimization and providing a strong foundation for future, more comprehensive research.

9. CONCLUSIONS

This research successfully demonstrated the application of electrocoagulation, optimized via RSM, as a highly effective technology for the treatment of greywater. By systematically investigating the influence of five key operating parameters, this study provided critical insights into the complex mechanisms governing the removal of turbidity, color, and COD. The analysis revealed that a strategy of high electrical current (1.5 A) and high electrolyte concentration (100 ppm NaCl) in a controlled acidic (pH_3) and thermal (30℃) environment can achieve exceptional pollutant removal in a remarkably short treatment time of just 5 minutes. The 3D surface plots were instrumental in illustrating these relationships, particularly the critical interactions between pH and current, and the non-linear effect of temperature.

The key findings can be summarized as follows:

- Electrocoagulation is capable of achieving high removal efficiencies for major greywater pollutants, confirming its suitability for water reclamation and reuse applications.
- Electrical current and NaCl dosage were identified as the most influential parameters. Counterintuitively, lower currents (0.5 A) and minimal NaCl addition yielded the best performance within the tested range, a finding with significant positive implications for operational cost and sustainability.
- RSM proved to be an invaluable tool for modeling the process. The developed second-order polynomial models accurately predicted system behavior ($R^2 > 0.90$) and successfully captured the significant linear, quadratic, and interactive effects of the process variables.
- Numerical optimization identified the optimal operating conditions as a treatment time of 20.5 min, current of 0.5 A, NaCl dosage of 15 ppm, pH of 6.8, and temperature of 30℃. Under these conditions, high experimental removal efficiencies of 86.9% for turbidity, 84.3% for color, and 80.5% for COD were achieved, validating the model's predictions.

In conclusion, this study underscores the importance of a systematic, statistical approach to optimizing complex environmental processes. By moving beyond traditional trial-and-error methods, RSM enables the development of efficient, reliable, and economically viable EC systems. Future work should focus on validating these findings on a pilot scale, conducting a thorough economic and life-cycle assessment, and exploring the characteristics and potential reuse of the generated electrocoagulation sludge. Such efforts will be crucial in advancing the deployment of EC technology as a key component of sustainable water management strategies worldwide.

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