Comparative Study of Chemical Coagulation and Electrocoagulation for the Treatment of Real Textile Wastewater: Optimization and Operating Cost Estimation

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ABSTRACT: Pollutants derived from real textile wastewater present a high environmental risk. This work involves the study of the removal of chemical oxygen demand (COD), color, and turbidity from Tunisian real textile wastewater by two different water treatment technologies: chemical coagulation (CC) and electrocoagulation (EC). A comparative study between these two methods was conducted based on the separation performance and operating cost (OC). The effects of different operational parameters including electrolysis time (t), voltage, and pH for EC and the coagulant concentration, initial pH, and time of slow mixing (t_sm) for CC were studied using response surface methodology. The developed quadratic models for the responses were in good agreement with the experimental data. The experiments proved the efficiency of both chemical and electrochemical techniques for the treatment of textile effluent. Indeed, by using EC, the reduction efficiencies of COD, color, and turbidity were 63.05, 99.07, and 96.31%, respectively, under optimal conditions (pH 9, t = 36.26 min, and voltage 4 V). For CC treatment, the achieved removal efficiencies of COD, color, and turbidity were 54.02, 96.21, and 93.7%, respectively, at pH 8.57, a coagulant concentration of 204.75 mg/L, and a t_sm of 28.41 min as optimal operating conditions. The OC obtained for EC and CC was about 0.47 and 0.2 USD/m³, respectively. Even if the OC of the EC process was higher as compared to the CC process, the treated water obtained by EC meets the Tunisian Standards (NT 106.03 and NT 09-14) for textile wastewater discharge into the environment and demonstrates a high potential for its reuse in various industrial activities. EC technology can be integrated into a wastewater management system that ensures a zero liquid discharge of wastewater into the environment.

1. INTRODUCTION

The amount of freshwater available for global use decreases, and water scarcity is becoming one of the greatest challenges of our time. Water crisis is especially pronounced in Africa. Nowadays, 13 African countries are experiencing severe water shortages.1 Water scarcity threatens several industries that have become vital to global fabric trade, and those industries are considered the ones with the most water consumption. Textile industrial processing needs a massive supply of clean water estimated to be ~200–250 L of water per kg of cotton cloth.2 Thus, the textile industry has been the epicenter of important pollutant sources, mainly coming from sizing, scouring, bleaching, mercerizing, dyeing, printing, and finishing processes.3 The major contaminant in the textile effluents is the coloring material in addition to dissolved organic and inorganic matter such as salts, toxic substances such as heavy metals, and so on.4 The dyes in textile wastewater pose the biggest threat to the environment because of their high toxicity.4

Therefore, industrial effluents have to be treated properly to ensure that there is a significant reduction in the level of pollutants according to the local standards. Consequently, the treated water could be discharged safely into the environment or reused in the industrial processes to promote sustainability and reduce the water burden for current and future generations. Currently, wastewater treatment and reuse can reduce the enormous gap between the increasing water demand and limited water supply. As a result, the choice of appropriate technology for wastewater treatment is a crucial issue because it directly affects the quality of obtained water.5

Depending on the treatment technology and effluent streams, the wastewater treatment plant can aggravate the environmental burden by generating a dischargeable effluent. Therefore, an assessment has to be done to evaluate the environmental impact.
of wastewater treatment technologies and, thus, choose the appropriate techniques.

Many treatment technologies have been adapted to treat textile effluent such as biological technologies, membrane-based methods (e.g., ultrafiltration and reverse osmosis), advanced oxidation processes (AOPs), and adsorption.\(^{26}\)

Biological treatments are time-consuming, especially for bacteria adapting, and require relatively high-volume reactors; furthermore, the organisms are sensitive to pH, temperature, and oxygen, especially under aerobic conditions and toxic components. In addition, some dyes are not biodegradable.\(^{8}\) Adsorption method requires a prefiltration step to remove suspended solids (SSs) and is limited by adsorbent capacity; also, the regeneration of the adsorbent is a challenging issue.\(^{9,10}\) Although the AOPs can remove most of the pollutants,\(^{5,6}\) they are expensive and involve several operational difficulties and is a complex technology that could limit their large-scale use.\(^{11,12}\) Moreover, the cited technologies are among the highest energy consumers compared to other technologies of wastewater treatment.\(^{13,14}\)

Recently, physicochemical methods such as chemical coagulation (CC) and electrochemical techniques, such as electrocoagulation (EC), have been widely used as alternative methods over the aforementioned technique thanks to their advantages including small equipment space and less sludge production.\(^{15-21}\)

In the CC process, colloidal particles are destabilized by the addition of chemicals called coagulants. At first, an augmentation of the ionic strength promotes double-layer compression, then a neutralization of the particle surface takes place by adsorbing counter anions.\(^{18,22}\) The CC process is characterized by the direct addition of a coagulant and adjustment of solution pH. The colloids are mostly negatively charged and stable in water solution. Therefore, we need some cations that interact with colloids, neutralize their charges and destabilize the water solution. Hydrolyzing metal salts are the more effective and used coagulants in water purification. Many researchers have employed different aluminum and iron(III) salts such as aluminum sulfate \(\text{Al}_2(\text{SO}_4)_3\) and ferric chloride \((\text{FeCl}_3)\), with the goal of removing impurities in industrial textile effluents.\(^{23}\) In the CC process, the destabilized particles form bigger aggregates that can be removed later through flocculation/decantation or filtration.

EC consists of an \textit{in situ} formation of hydrolyzable metal coagulants through electrolytic oxidation of metal anodes (commonly aluminum (Al) or iron (Fe)).\(^{24}\) In both EC and CC, the Al or Fe hydroxides are efficient coagulants and have a large surface area that can adsorb and remove dissolved contaminants such as color, chemical oxygen demand (COD), biochemical oxygen demand (BOD\(_5\)), SSs, and turbidity.\(^{25}\)

In the EC process, the removal of coagulated particles can be done by sedimentation or through electro-floation by lifting them to the surface with \(\text{H}_2(\text{g})\) bubbles generated at the cathode.\(^{26}\)

In the case of the Al anode, at a fixed electric current or potential, EC can be represented as

\[
\text{Al(s)} \rightarrow \text{Al}^{3+} (\text{aq}) + 3e^- 
\]

(1)

The reduction of water to form \(\text{H}_2(\text{g})\) may happen at the cathode

\[
\text{H}_2\text{O} + e^- \rightarrow \frac{1}{2}\text{H}_2(\text{g}) + \text{OH}^- (\text{aq}) 
\]

(2)

Metal ions (i.e., \(\text{Al}^{3+}\)) formed at the anode react with hydroxyl ions \((\text{OH}^-)\) present in water and are liberated from the cathode to produce various monomeric and polymeric species, which (at an appropriate pH) can form a wide range of coagulating species and metal hydroxides.\(^{27}\)

The comparison between the results observed in CC and EC is not clear because both similar and opposite efficiencies can be found in the literature.\(^{28-37}\) This variation depends on many factors such as effluent characteristics, their nature (real or synthetic wastewater), and the specific parameters of each technique. Despite the different efficacies obtained, CC and EC have successfully been used for the treatment of different types of wastewater such as landfill leachate wastewater, diary wastewater, slaughterhouse wastewater, and textile wastewater.\(^{38-40}\)

Besides, the majority of research is dedicated mainly to study the effect of operational parameters on the efficiency of pollutant removal from industrial wastewater; however, a few of them have been focused on the possibility of reusing it in various industrial activities. The primary goal of this study is to obtain treated wastewater suitable to be reused in the process. The wastewater effluent from the textile factory is a mixture of dyes and other pollutants. Companies within this industry were massive water consumers utilized for cleaning the raw material, and for the different steps of textile manufacturing. Two treatment methods EC and CC will be used and compared to each other under optimal conditions. The comparison is based on the effectiveness of treatment to obtain treated water suitable for discharging safely into the environment or recycling in the same process. Moreover, operating cost (OC) will be evaluated for both processes and considered an important criterion for comparison.

2. MATERIALS AND METHODS

2.1. Textile Wastewater Characteristics. This study was carried out using real textile wastewater from the textile manufacture in Monastir City, one of the most significant provinces of Tunisia in terms of the textile industry. This factory focuses on the production of denim jeans and generates an average of 300 m\(^3\)/day of effluents as a result of prewashing, washing, and finishing steps. The water used in the industrial processes was purchased from the local sanitation and the wastewater generated was discharged into the public sewage system. The main textile wastewater characteristics are listed in Table 1.

| Table 1. Characteristics of the Real Textile Wastewater |
|----------------|-------------|
| parameter       | value       | discharge limits\(^a\) |
| pH              | 6           | 6.5–9                  |
| COD\(^b\) (mg/L)| 404         | 160                    |
| BOD\(_5\) \(^c\) | 120         |                        |
| SSs\(^d\) (mg/L)| 325         | <35                    |
| SO\(_4^{2-}\) (mg/L)| 500      | 400                    |
| Cl\(^-\) (mg/L) | 401         |                        |
| electric conductivity (mS/cm)| 3.39 | 5.00                  |
| turbidity (NTU\(^e\)) | 64 | <5                    |
| color (at 669 nm)| blue       | 0.518                  |

\(^a\) Tunisian Standards NT 106.03 and NT09.14.\(^41\) \(^b\) COD: chemical oxygen demand. \(^c\) BOD\(_5\): biochemical oxygen demand. \(^d\) SSs: suspended solids. \(^e\) NTU: nephelometric turbid unit.
The effluent examined in this study has some characteristics that exceed discharge limits such as COD, BOD$_5$, and turbidity (Table 1). The discharge limits were adopted according to the Tunisian Standards NT 106.03 and NT 09-14.41 The textile wastewater was stored at 4 °C to avoid degradation. To remove the large SSs, the wastewater was filtered with a prefiltration grid before being used in the study.

2.2. Experimental Apparatus and Operating Conditions. EC experiments were carried out using a batch reactor with a capacity of 500 mL. Two aluminum plate electrodes of dimension 90 × 35 × 1 (mm × mm × mm) with a purity of 98% were used and connected in a monopolar arrangement to a direct current (DC) power source (GPS-3303) operated at the potentiostatic mode, in which required voltage was fixed. During experiments, a 5 cm height electrode was immersed into the solution. The anode and cathode were placed vertically and parallel to each other with the interelectrode distance set at 1 cm. In fact, the choice of interelectrode distance was based on the bibliographic study.42−44 The increase of electrode gap leads to an increase in the Ohmic resistance43 at a constant anodic surface area and conductivity of the solution (eq 3):

$$R_{\text{Ohm}} = \frac{d}{S \cdot k}$$  (3)

where $d$ is the interelectrode distance, $k$ is the water conductivity, and $S$ is the active anode surface (m²). At constant voltage, when the Ohmic resistance between electrodes increases, the current passed through electrodes decreases, and the rate of anodic dissolution decreases.

The anodic oxidation and cathodic reduction were started by switching the DC power supply on for the desired electrolysis time ($t$). The reactor was placed on a magnetic stirrer set at 150 rpm to maintain uniform shaking. In order to remove the layer from the electrodes, the electrode surfaces were rinsed with 0.1 M HCl after each experiment and then washed with distilled water and dried.

In each experiment, 400 mL of real textile wastewater was placed in the reactor at ambient temperature. The pH was adjusted to the desired value by adding a required volume of sodium hydroxide or sulfuric acid. It should be mentioned that no additional electrolyte (as the supporting electrolyte) was added during the EC process.

For CC, the experiments were conducted on the Jar test apparatus. Different combinations of pH and coagulant concentration were tested. Commercial-grade aluminum sulfate Al$_2$(SO$_4$)$_3$ (alum) was used as the coagulant. The choice of this coagulant has been well studied.18,23 First, aluminum sulfate has been adopted to make a comparison with EC employing Al electrodes. Second, it is an effective and cheap coagulant. A stock solution was prepared with 2 g/L of concentration. Then, this solution was used to prepare the dilute solutions fixed in this study. The aluminum coagulant concentrations were varied from 97.73 to 602.27 mg/L to determine the optimum concentration for pollutant removal. 400 mL of each solution was put in six parallel jars with six speed-controlled paddles at room temperature. The samples were stirred for 3 min at 200 rpm. These values were close for the majority of research works.23−25 Then, it was followed by further slow mixing at 45 rpm; the duration of this step will be optimized during this project.

For EC and CC processes, at the end of each experiment, before settling, a sample was withdrawn for SSs analysis. After that, the formed flocs were allowed to settle for 60 min. After settling, the supernatant of each treated sample was withdrawn and analyzed to evaluate removal efficiencies in terms of COD, color, and turbidity.

EC and CC processes were subjected to optimization. Three parameters were optimized including electrolysis time ($t$), voltage, and initial pH for EC and coagulant concentration, time of slow mixing step ($t_{\text{mix}}$), and initial pH for CC.

2.3. Analytical Methods. Turbidity, conductivity, and pH measurements in supernatant liquid were done by a turbidimeter (TU-2016), conductivity meter (EZDO 7200), and pH meter (Adwa AD 1000), respectively. The COD and SS measurements were conducted based on Standard Methods for the Examination of Water and Wastewater.45 A colorimetric method with closed reflux using potassium dichromate as an oxidant was developed for the measurement of COD. The sample was incubated for 120 min at 150 °C in a thermoreactor (ALB64). A UV/visible spectrophotometer (Shimadzu UV-1700) was used for COD analysis at 620 nm wavelength and for color analysis at the wavelength of maximum absorbance band of textile wastewater, that is, at 669 nm ($\lambda_{\text{max}}$) (Figure 1).

The amount of Al metal residue induced by the coagulant itself or by the dissolution of the electrode in the liquid, under optimized conditions, was determined using inductively coupled plasma atomic emission spectrometry (HORIBA JOBIN YVON). Sulfate ion was analyzed by a gravimetric method according to Standard Methods for the Examination of Water and Wastewater.45 The color, COD, and turbidity removal efficiencies (%) were calculated using the following formulas:

$$\text{color removal} = \frac{A_0 - A_t}{A_0} \times 100\%$$  (4)

$$\text{COD removal} = \frac{\text{COD}_0 - \text{COD}_t}{\text{COD}_0} \times 100\%$$  (5)

$$TU = \frac{TU_0 - TU_t}{TU_0} \times 100\%$$  (6)

where $A_0$ and $A_t$ represent the initial and final absorbance of textile wastewater, respectively. COD$_0$ and COD$_t$ are the initial
The electrode consumption for the EC process (in kg/m³) is calculated using eq 9, according to Faraday’s law:

\[ C_{\text{elec}} = \frac{ItM}{nFV} \]

where \( I \) is the electric current (A), \( t \) is the time of electrolysis (s), \( M \) is the molar mass of aluminum (26.98 g/mol), \( n \) is the number of electrons transferred (\( z = 3 \) for Al), \( F \) is Faraday’s constant (96,500 C/mol), and \( V \) is solution volume (m³).

The total OC is calculated based on economic data obtained from the Tunisian market in 2020, as shown in Table 2.

The energy consumption in the EC process is used for the removal of pollutants from textile wastewater by EC and CC processes (kW h/m³), respectively; consumed electrode, chemical intake, and sludge transportation and disposal, respectively.

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Table 2. Economic Data Used for Calculating the OC

| a (USD/kWh) | b_{elec} (USD/kg) | b_{coag} (USD/kg) | c (USD/L_{chemical}) | d (USD/kg_{sludge}/km) |
|-------------|-------------------|-------------------|----------------------|------------------------|
| 0.0875      | 2.7334            | 0.7021            | 0.0454               | 0.0004                 |

Table 3. Removal of COD, Color, and Turbidity for EC and CC Processes in the Preliminary Study

| trial | conditions | % COD | % color | % TU | % COD | % color | % TU |
|-------|------------|-------|---------|------|-------|---------|------|
| 1     | pH 7, voltage = 9 V, electrolysis time, \((t) = 6.36 \text{ min}\) | pH 7, coagulant concentration = 0.696 g/L, coagulation time = 6.36 min | 16.46 | 96.72 | 96.50 | 13.34 | 46.87 | 39.30 |
| 2     | pH 7, voltage = 9 V, electrolysis time, \((t) = 40 \text{ min}\) | pH 7, coagulant concentration = 1.9 g/L, coagulation time = 40 min | 43.69 | 96.68 | 88.66 | 10.20 | 41.69 | 32.70 |
| 3     | pH 7, voltage = 9 V, electrolysis time, \((t) = 73.63 \text{ min}\) | pH 7, coagulant concentration = 3.9 g/L, coagulation time = 73.63 min | 15.59 | 93.185 | 83.81 | 4.06 | 15.3 | 17.33 |

\(\% \text{ COD} \), \(\% \text{ color} \), and \(\% \text{ TU} \) are given in Table 3. Experimental values are evaluated using eqs 10 and 11, respectively:

\[ C_{\text{energyEC}} = \frac{U \int I \, dt}{V} \]

(10)

\[ C_{\text{energyCC}} = \frac{N_p \rho N_f d_f^4}{V} \]

(11)

where \( U \) is the electrical potential (V), \( N_p \) is the power number (dimensionless), \( \rho \) is the liquid density (kg/m³), \( N \) is the impeller rotational speed (rpm), and \( d_f \) is the impeller diameter (m). The power number is a function of the Reynolds number \((Re)\) and the geometry of the agitator. In this study, a flat-blade turbine with two blades is used as the agitator.

2.4. OC Analysis. The EC and CC processes require an OC, which includes electrical energy, material (electrode for EC), chemicals, and sludge transportation and disposal. The OCs for EC and CC (unit price/m³) could be calculated using eqs 7 and 8, respectively:

\[ OC = a \cdot C_{\text{energyEC}} + b_{\text{elec}} \cdot C_{\text{elec}} + c \cdot C_{\text{chemical}} + d \cdot C_{\text{sludge}} / D \]

(7)

\[ OC = a \cdot C_{\text{energyCC}} + b_{\text{coag}} \cdot C_{\text{coag}} + c \cdot C_{\text{chemical}} + d \cdot C_{\text{sludge}} / D \]

(8)

where \( C_{\text{energyEC}} \) and \( C_{\text{energyCC}} \) are the amounts of energy consumed in EC and CC processes (kW h/m³), respectively; \( C_{\text{elec}} \) is the electrode consumption (kg_{electrode}/m³); \( C_{\text{coag}} \) is the coagulant consumption (kg/m³); \( C_{\text{chemical}} \) is the chemical consumption for pH adjustment (L_{chemicals}/m³); \( C_{\text{sludge}} \) is the amount of produced sludge (kg_{sludge}/m³); \( D \) is the sludge transport distance (km); and \( a \) (unit price/kWh), \( b_{\text{elec}} \) (unit price/kg_{electrode}), \( b_{\text{coag}} \) (unit price of coagulant/kg), \( c \) (unit price/L_{chemicals}), and \( d \) (unit price/kg_{sludge}/km) are the unit prices of electrical energy, consumed electrode, chemical intake, and sludge transportation and disposal, respectively.

The electrode consumption for the EC process (in kg/m³) is calculated using eq 9, according to Faraday’s law:

\[ C_{\text{elec}} = \frac{ItM}{nFV} \]

(9)

where \( I \) is the electric current (A), \( t \) is the time of electrolysis (s), \( M \) is the molar mass of aluminum (26.98 g/mol), \( n \) is the number of electrons transferred (\( z = 3 \) for Al), \( F \) is Faraday’s constant (96,500 C/mol), and \( V \) is solution volume (m³).

The total OC is calculated based on economic data obtained from the Tunisian market in 2020, as shown in Table 2.

The energy consumption in the EC process is used for electrolytic dissolution of the sacrificial aluminum anode, whereas for the CC process, electricity is used for agitation. The amounts of electrical energy consumed (kWh/m³) during the removal of pollutants from textile wastewater by EC and CC are evaluated using eqs 10 and 11, respectively.
attitude of the pH in the chemical and the electrochemical experiments. On one hand, the pH value rises with time in the EC process because of the formation of aluminum hydroxide (Al(OH)₃(s)) as the final product. On the other hand, this value decreases in the CC process as a result of the acid characteristic of the aluminum solutions used. Therefore, the initial pH value fixed in CC should be higher than those taken into EC. Moreover, the highest values of aluminum obtained by EC are no longer suitable in CC. Thus, we can conclude that lower aluminum concentrations should be applied for the CC process compared to the EC process.

Finally, by referring to various preliminary experiments and with the goal to optimize both technologies, it was decided to determine the lower and upper conditions limits of each technique separately.

2.6.2. Experimental Design. To determine the optimum conditions for EC and CC, the experiments were performed using a central composite design (CCD) as a second-order design of response surface methodology (RSM). RSM is a collection of mathematical and statistical techniques used in general for the optimization of treatment processes. Three factors are studied: initial pH ($A$), electrolysis time ($B$), and applied voltage ($C$) for EC and initial pH ($A$), coagulant dose ($B$), and time of the slow mixing step ($C$) for CC. The corresponding responses for both techniques are the percentage removal of COD, color, and turbidity and OC.

The coded and actual values and ranges for each independent variable for both processes are shown in Table 4. By referring to the literature and well-established preliminary studies, the lower and upper limits are fixed.

### Table 4. Experimental Range and Levels of the Independent Variables Used in RSM

| Variable | $-\alpha$ | $-1$ | $0$ | $+1$ | $+\alpha$ |
|----------|-----------|-----|-----|------|---------|
| EC Process |          |     |     |      |         |
| $A$: pH  | 2         | 4   | 7   | 10   | 12      |
| $B$: electrolysis time ($t$) (min) | 6.36 | 20 | 40 | 60 | 73.63 |
| $C$: voltage (V) | 4 | 6 | 9 | 12 | 14 |
| CC Process |          |     |     |      |         |
| $A$: pH  | 6         | 7   | 8.5 | 10   | 11      |
| $B$: coagulant dose [coag] (mg/L) | 97.73 | 200 | 350 | 500 | 602.27 |
| $C$: time of slow mixing ($t_{ms}$) (min) | 12 | 15 | 20 | 25 | 28.41 |

The experimental design, analysis of variance (ANOVA), and mathematical modeling were performed using Minitab 19 (trial version). The design includes 20 experiments for each process: eight cubic points, six central points, and six axial points. Second-order polynomial models were used to predict the optimal conditions (eq 12):

$$ Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \sum_{j=1}^{i} \beta_{ij} X_i^2 + \sum_{i=1}^{k} \sum_{j=1}^{i} \sum_{l=1}^{i} \beta_{ijk} X_i X_j + \epsilon $$

(12)

where $k$ is the number of factors studied and optimized in the experiment, $\beta_0$ is the offset term, $\beta_i$ is the linear effect of the input variables, $X_i$ and $X_j$ are independent variables, and $\beta_{ij}$ is the quadratic parameter coefficient of the input variable $X_i$. Also, $\beta_{ijk}$ is the linear interaction effect between the input variables $X_i$ and $X_j$, and $\epsilon$ is the random error of experiments.

3. RESULTS AND DISCUSSION

The experimental results obtained by CCD were regressed and fitted to the RSM model. The percentage removals of color, COD, turbidity, and the OC (USD/m³) are presented by second-order polynomial equations: 13 and 13 for EC and CC processes, respectively, in terms of uncoded factors. Experimental and predicted response data obtained from the CCD approach are presented in Tables 5 and 6 for EC and CC, respectively.

The positive sign in front of the coefficient indicates a synergistic effect, whereas the negative sign indicates an antagonistic effect. Considering the EC process and by referring to the coefficients in eqs 13–16, the pH and electrolysis time ($t$) had a positive effect on the removal of COD, color, and turbidity. However, the applied voltage had a negative influence on COD removal and OC. In the CC process (eqs 17–20), the positive sign of the pH coefficient denotes a positive influence on COD, color and turbidity removal, and OC.

$$\text{COD removal (\%)} = 78 + 7.16A + 0.694B - 17.3C - 0.028A^2 - 0.02090B^2 + 0.483C^2 - 0.1311AB - 0.107AC + 0.2239BC \quad(13)$$

$$\text{color removal (\%)} = 59.9 + 2.12A + 0.695B + 4.41C + 0.1555A^2 + 0.00105B^2 - 0.066C^2 - 0.0537AB - 0.2799AC - 0.0551BC \quad(14)$$

$$\text{TU (\%)} = 25.1 + 2.83A + 1.38B + 10.03C + 0.209A^2 + 0.00234B^2 - 0.244C^2 - 0.074AB - 0.363AC - 0.1485BC \quad(15)$$

$$\text{OC (USD/m}^3\text{)} = 3.595 - 0.398A - 0.137B - 0.0476C + 0.0214A^2 + (2.860 \times 10^{-6})B^2 + 0.0190C^2 + 0.00079AB + 0.0063AC + 0.00969BC \quad(16)$$

$$\text{COD removal (\%)} = -609 + 84.2A + 0.79B + 15.9C - 2.711A^2 - 0.000224B^2 - 0.0545C^2 - 0.0464AB - 1.153AC - 0.01021BC \quad(17)$$

$$\text{color removal (\%)} = -214.5 + 66.5A - 0.059B + 0.71C - 3.498A^2 - 0.00013B^2 + 0.0228C^2 + 0.0142AB - 0.2444AC + 0.00203BC19 \quad(18)$$

$$\text{TU (\%)} = -504 + 121.6A - 0.152B + 5.06C - 6.18A^2 - 0.000219B^2 - 0.002C^2 + 0.0282AB - 0.656AC + 0.00330BC19 \quad(19)$$

$$\text{OC (USD/m}^3\text{)} = 0.020105 + 0.00018A + 0.000755B + 0.000189C + 0.00424A^2 - (5.1860 \times 10^{-6})B^2 + 0.00038C^2 - (1.1111 \times 10^{-6})AB - 0.000167AC - (1.66667 \times 10^{-6})BC \quad(20)$$

3.1. Statistical ANOVA. The statistical model ANOVA was used to validate the experimental data obtained for EC and CC, respectively. The fit of the model to the experimental data was evaluated through the coefficient of determination ($R^2$) and...
adjusted determination coefficient (R^2_adj) complemented by the graphic plot of predicted values by the model versus observed experimental values (the result of a particular run) as shown in Figures 2a–d and 3a–d for EC and CC, respectively. The plot indicates an adequate agreement between the applied model data and the experimental data.

The results show high reliability in the estimation of OC and the removal of COD, turbidity, and color. R^2 values were found to be 0.92, 0.892, 0.952, and 0.989 for the EC process and 0.837, 0.846, 0.893, and 0.99 for the CC process on COD, color, and turbidity removal and OC (USD/m^3), respectively. It can be deduced that only 0.081 (COD removal), 0.109 (color removal), 0.048 (turbidity removal), and 0.011 (OC) in the response variable were not explained by the models in the EC process; however, 0.16 (COD removal), 0.154 (color removal), 0.107 (turbidity removal), and 0.001 (OC) in the response variable were not explained by the models of the CC process. The R^2_adj were also high and equal to 0.914, 0.815, 0.914, and 0.980 for the EC process and 0.753, 0.781, 0.813, and 0.99 for the CC process on COD, color, and turbidity removal and OC.
respectively, which is in a good agreement with $R^2$ values implying a high significance of the quadratic model and ensures that the model is adequately accurate and reliable. ANOVA results for four models are given in Tables 7 and 8. Values of “Prob > F” were observed to be less than 0.0500, indicating that the model terms are significant. Considering the EC process, the terms that are found to be significant are $AB$, $BC$, $B^2$, and $C^2$ for the COD removal model; $B$, $C$, $AB$, $AC$, $BC$, and $A^2$ for color removal; $B$, $C$, $AB$, $BC$, $AC$, and $C^2$ for turbidity removal; and $B$, $C$, $BC$, $A^2$, and $C^2$ for the OC model. In the case of the CC process, model terms that are considered to be significant are $B$, $AB$, $AC$, $BC$, $A^2$, and $B^2$ for the COD removal model; $A$ and $A^2$
### Table 7. ANOVA Results of Models Obtained for the Effect of Operating Conditions on the Response of the EC Process

| source | df | % COD | % color | % TU | OC (USD/m³) | df | % COD | % color | % TU | OC (USD/m³) | F-value | p-value |
|--------|----|-------|---------|------|-------------|----|-------|---------|------|-------------|---------|---------|
| model  | 9  | 9     | 9       | 9    | 386.7       | 9  | 48.06 | 277.3   | 3.21 | 13.15       | 23.54   | 105.51  |
| A      | 1  | 1     | 1       | 1    | 37.58       | 9  | 16.79 | 26.89   | 0.012 | 1.28        | 3.6     | 2.28    |
| B      | 1  | 1     | 1       | 1    | 78.61       | 9  | 45.07 | 450.2   | 11.24 | 2.67        | 10.08   | 38.2    |
| C      | 1  | 1     | 1       | 1    | 19.26       | 9  | 109.69| 993.6   | 14.04 | 0.655       | 23.51   | 84.31   |
| AB     | 1  | 1     | 1       | 1    | 495.07      | 9  | 82.98 | 157.6   | 0.018 | 16.83       | 17.78   | 13.38   |
| AC     | 1  | 1     | 1       | 1    | 7.48        | 9  | 50.77 | 85.39   | 0.0257| 0.254       | 10.88   | 7.24    |
| BC     | 1  | 1     | 1       | 1    | 1444.1      | 9  | 87.42 | 635.1   | 2.70  | 49.11       | 18.73   | 53.89   |
| A²     | 2  | 1     | 1       | 1    | 0.93        | 9  | 28.21 | 51.15   | 0.5365| 0.032       | 6.05    | 4.34    |
| B²     | 2  | 1     | 1       | 1    | 1007.1      | 9  | 2.54  | 12.62   | 0.0000| 34.25       | 0.544   | 1.07    |
| C²     | 2  | 1     | 1       | 1    | 272.85      | 9  | 5.08  | 69.58   | 0.4215| 9.28        | 1.09    | 5.9     |

### Table 8. ANOVA Results of Models Obtained for the Effect of Operating Conditions on the Response of the CC Process

| source | df | % COD | % color | % TU | OC (USD/m³) | F-value | p-value |
|--------|----|-------|---------|------|-------------|---------|---------|
| model  | 9  | 9     | 9       | 9    | 365.56      | 90027   | 0.0006  |
| A      | 1  | 1     | 1       | 1    | 48.06       | 0.000   |
| B      | 1  | 1     | 1       | 1    | 412.41      | 0.000   |
| C      | 1  | 1     | 1       | 1    | 64.62       | 0.000   |
| AB     | 1  | 1     | 1       | 1    | 872.77      | 0.000   |
| AC     | 1  | 1     | 1       | 1    | 598.63      | 0.000   |
| BC     | 1  | 1     | 1       | 1    | 494.44      | 0.000   |
| A²     | 1  | 1     | 1       | 1    | 536.23      | 0.000   |
| B²     | 1  | 1     | 1       | 1    | 365.89      | 0.000   |
| C²     | 1  | 1     | 1       | 1    | 26.78       | 0.000   |
for the color removal and turbidity models, and A and B for the OC model.

Table 9 gives a statistical summary of the quadratic order polynomial regression models for EC and CC. Besides $R^2$ and $R_{adj}^2$ values, other statistical parameters such as $p$-value, $F$-value, and adequate precision were also used to evaluate the competence of developed models. The values of adequate precision are 12.59, 13.70, 19.18, and 36.17 for EC and 10.12, 11.46, and 189.68 for CC on COD, color, and turbidity removal and OC (USD/m$^3$). High $R^2$ values and smaller standard deviation values indicate the better predicting response of the model developed. As can be seen from Table 9, the quadratic models for our response were significant at the 5% confidence level because $p$ values were less than 0.05. The "$F$-value" of the developed model was found to be 13.5, 10.3, 23.54, and 105.51 for the EC process and 8.5, 8.00, 10.24, and 2424.45 for the CC process on COD, color, and turbidity removal and OC (USD/m$^3$), respectively, depicting that these models are statistically significant.

Table 9. Statistical Summary of the Developed Model for EC and CC

| Response | $R^2$ | $R_{adj}^2$ | $p$-value | Prob > $F$ | $F$-value | Adequate Precision | Standard Deviation |
|----------|-------|-------------|-----------|-------------|-----------|-------------------|-------------------|
| COD (%)  | 0.92  | 0.852       | 0.0002    | 13.15       | 12.59     | 5.42              |
| color (%)| 0.892 | 0.815       | 0.0006    | 10.30       | 13.7      | 2.16              |
| TU (%)   | 0.952 | 0.914       | <0.0001   | 23.54       | 19.18     | 3.43              |
| OC (USD/m$^3$) | 0.989 | 0.979       | <0.0001   | 105.51      | 36.18     | 0.1745            |

| Response | $R^2$ | $R_{adj}^2$ | $p$-value | Prob > $F$ | $F$-value | Adequate Precision | Standard Deviation |
|----------|-------|-------------|-----------|-------------|-----------|-------------------|-------------------|
| COD (%)  | 0.837 | 0.753       | 0.0027    | 8.50        | 10.47     | 7.22              |
| color (%)| 0.846 | 0.781       | 0.0023    | 8.00        | 10.12     | 6.46              |
| TU (%)   | 0.893 | 0.813       | 0.0006    | 10.24       | 11.46     | 9.92              |
| OC (USD/m$^3$) | 0.99  | 0.99        | <0.0001   | 2424.45     | 189.68    | 0.0026            |

Figure 4. 3D response surface plot for COD, turbidity, and color removal; the effect of pH, electrolysis time ($t$), and voltage = 9 V (EC process).
3.2. Effect of Operating Parameters on Removal Efficiency. A three-dimensional (3D) graphical representation of the models’ responses was drawn to analyze the combined effects of operating parameters such as pH with reaction time and pH with voltage for the EC process, and coagulant concentration with pH for the CC process.

3.2.1. EC Process. Figures 4−6 give the variation of COD, color, and turbidity removal with initial pH and electrolysis time (t), t with voltage, and pH and voltage, respectively. The third parameter (i.e., the one not plotted in each of the figures) is set at its central level.

From Figure 4, the best COD removal efficiency was obtained in alkaline and acidic media. Indeed, for a pH of 10 and a t between 27 and 40 min, the obtained removal was about 43%; also, for a pH of 4 and t between 47 and 57 min, the removal was about 42.5% approximately. The same trend was depicted for color. In fact, the color efficiency was more enhanced at acidic medium for a pH 4−5 under a t more than 50 min as the removal of color was about 97%; besides, in alkaline medium (pH > 8) and under a t less than 30 min, the removal efficiency of color is higher than 97%. It is noted also from Figure 4 that the turbidity reaches more than 95% starting from pH 7 onward under a t less than 25 min. Mohamad Zailani et al.50 reported that the highest removal efficiency of turbidity was obtained for a pH < 6 and increases with pH more than 9 at t = 20 min on the treatment of landfill leachate using an Al electrode. Vidal et al.51 observed complete decolonization after 5 min for both initial pH values equal to 4 and 8.5. Deghles and Kurt52 found a high removal efficiency of color and COD at acidic medium with Al electrode on the treatment of tannery wastewater. Izadi et al.53 treated paper-recycling wastewater using Al electrodes and showed an optimal removal efficiency of COD and color when the pH of the wastewater was between 5 and 10. Sontaya et al.54 studied the removal of COD from palm mill wastewater by EC. The results showed a maximum COD removal of about 89.2%, at an applied voltage of 12 V and t = 15 min in both acidic and basic conditions. Linares-Hernández et al.55 analyzed the Al species associated with the aqueous pH. At a pH below 3.5, Al^{3+} is the predominant species. However, at a pH of 4−9.5, the predominant Al chemical species is Al(OH)_{3(aq)} which has a large surface area useful for the rapid adsorption of soluble organic compounds and also trapping of colloidal particles. At a

Figure 5. 3D response surface plot for COD, turbidity, and color removal; the effect of voltage and electrolysis time (t), pH 7 (EC process).
pH value greater than 10, the dominant specie is $\text{Al(OH)}_4^-$, which obviously does not coagulate the pollutants, and hence, there is no reduction in COD and color. The same results were found by other researchers.\textsuperscript{56,57} Besides, the high removal efficiency obtained for COD and color at alkaline and acidic medium could suggest that is not only the EC by direct anodic oxidation of Al electrode implicated in the pollutant removal, but also suggest that the indirect oxidation by oxidants such as hypochlorite or chlorine could happen. In fact, the chloride anions present in the raw wastewater (Table 1) may be oxidized into chlorine (eq 21). The abovementioned can generate HOCl (eq 22) and hypochlorite ions (eq 23). These species, because of their high oxidative potentials, can take part in oxidizing soluble organic matter.\textsuperscript{58,59}

\[
\begin{align*}
2\text{Cl}^- & \rightarrow \text{Cl}_2 + 2e^- \quad \text{(21)} \\
\text{Cl}_2 + \text{H}_2\text{O} & \rightarrow \text{HOCl} + \text{Cl}^- + \text{H}^+ \quad \text{(22)} \\
\text{HClO} & \rightarrow \text{ClO}^- + \text{H}^+ \quad \text{(23)}
\end{align*}
\]

Figure 5 presents the interaction impact of voltage and electrolysis time (t) on COD, color, and turbidity removal at pH 7. At a constant Ohmic resistance (eq 3) between the electrodes, increasing voltage value leads to an increase in the current passing through the electrodes. Thereby, the rate of Al\textsuperscript{3+} ions released from the respective electrode increases according to Faraday’s law (eq 9). The dissolved metal ions can form a wide range of metal hydroxides that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants. Voltage is expected to exhibit a strong effect on removal efficiency. For the color and turbidity treatment, the best removal was depicted at a higher voltage value and a shorter treatment time or at a low value of voltage and a high treatment time. In the case of COD removal, we observe a different behavior compared to color and turbidity removal. Therefore, (i) for $t < 35$ min, increasing the voltage decreases the percentage removal of COD, whereas (ii) for $t$ between 35 and 45 min, the COD removal decreases and then increases slightly with increasing voltage and (iii) for $t > 45$ min, the COD removal efficiency increases continuously with increasing voltage. Indeed, at $t = 30$ min, the COD removal percentage decreases from 50 to 35% with increasing voltage. While, at $t = 50$ min, the COD removal efficiency increases from 39 to approximately 50% with the increase of applied voltage.

Many investigators have focused on the study of the applied voltage (or current density, CD) influence on pollutant removal. Three main trends have been observed between voltage and COD removal in the literature: (i) the COD removal increases with increasing applied voltage; (ii) the COD removal was found to be independent of applied voltage; and (iii) the COD removal decreases with increasing applied voltage. Givven et al.\textsuperscript{59} studied the removal of COD from simulated beet sugar factory wastewater. The results showed that the removal efficiency increased with voltage until 10 V and that above this value the COD removal remained constant. Similar results were reported by some researchers.\textsuperscript{54,60} Ates et al.\textsuperscript{61} investigated the effect of applied voltage on COD removal by using an Al electrode. Results showed that the increase in voltage did not affect significantly the COD removal efficiency. However, Jing et al.\textsuperscript{62} reported that the COD removal rate first increases from 49.41 to 77.46% as CD rises to 24.73 mA/cm\textsuperscript{2} and then decreases to 60.42% with the increase of CD.

The results of this study are in part in coherence with those obtained in the literature. Additionally, the decrease of COD removal by increasing the applied voltage can be explained by the fact that high voltage above the optimum value leads to a higher oxygen generation from the anode. Consequently, the competition between Al anode dissolution and bubbles formation can cause a reduction of Al(OH)\textsubscript{3}(s) formation, especially during the short time of treatment. Thus, it may lead to a decrease in COD removal. Moreover, Alam et al.\textsuperscript{63} have stated that a further increase in CD above a certain value causes an increase in the size of O\textsubscript{2}(g) and H\textsubscript{2}(g) bubbles. Therefore, large bubbles tend to remove the pollutant quickly to the surface, decreasing the possibility of capturing them.

Such results exhibit a different mechanism of COD removal than the color and turbidity and highlight the importance of electrolysis time as a key parameter to reaching a high COD removal. Applying a high voltage at a short time did not enhance COD removal. A high voltage and long electrolysis time were needed to achieve the best removal efficiency, which is about 60% at 60 min and 12 V.

Figure 6. 3D response surface plot for turbidity and color removal, the effect of voltage and pH, electrolysis time $(t) = 40$ min (EC process).
Figure 6 gives the interaction impact of voltage and pH on color and turbidity removal at $t = 40$ min. For a given value of pH, increasing voltage leads to a decrease in turbidity removal. The same trend is observed for color removal at pH $> 5$. Higher color and turbidity removal are obtained for a pH of more than 8 and the applied voltage between 6 and 7 V.

3.2.2. CC Process. The 3D surface graph, initial pH versus concentration of coagulant, depicted in Figure 7a shows that a significant interaction occurs between initial pH and coagulant concentration for COD removal as a response at 20 min of slow mixing ($t_{sm}$).

Given that, different parameters and phenomena can influence the coagulation processes. It is, therefore, difficult to determine an optimal coagulant concentration and initial pH value. Several optimum coagulant concentrations and initial pH values are found in the literature$^{33-35}$ because this process includes different phenomena in different pH ranges.$^{35}$ It is clear from Figure 7a that the COD removal is reduced at low coagulant concentration and initial pH. The domain of high COD removal (more than 50%) can be identified at coagulant concentrations between 400 and 500 mg/L and for pH values corresponding to 7–8.5. Similar findings were obtained by Guida et al.$^{64}$ on the treatment of five municipal wastewater using alum-coagulation for the treatment of COD. The results show a high removal efficiency of COD obtained at pH between 6 and 8 under a coagulant concentration of about 450 mg/L, then the percentage removal decreases for pH 10. Similar results were also reported by Joaquin and Nirmala.$^{65}$ Furthermore, Selçuk et al.$^{66}$ suggested an optimum pH of 6.0 for the elimination of 48% of COD with an aluminum concentration of 500 mg/L. Azbar et al.$^{67}$ presented an optimum pH of 8 for an aluminum concentration of 400 mg/L to obtain 59% COD removal. On the other hand, according to Lefebvre and Legube,$^{68}$ the elimination of organic matter is optimal for pH values between 4.5 and 5.5. However, it is very difficult to obtain such acidity on an industrial scale.

It is noted also from Figure 7a that the effect of coagulant concentration on COD removal is different at higher concentrations. In the CC technique, it is necessary to study the coagulant concentrations that must be added: if we add higher concentrations, that are higher than optimal concentrations, we will obtain different COD removals. This result is expected because coagulation occurs when the zeta potential approaches zero.$^{23}$ Therefore, an overdose of coagulant can cause the opposite effect. The positive charges, which are arranged on the surface of the particles, cause the redispersion of COD.
During the overinjection of the coagulant, the pH drops and leads to the formation of species more soluble than the Al(OH)$_3$.

Figure 7b shows that, for a concentration of aluminum coagulant of about 350 mg/L and a pH value between 7 and 8.5, the increase of the duration of slow mixing ($t_{sm}$) from 15 to 25 min leads to an increase in the COD removal. Moreover, Figure 7c shows the combined effects of slow mixing duration ($t_{sm}$) and concentration of aluminum coagulant. It can be observed that COD removal efficiency increases together with the increasing aluminum coagulant concentration at a low mixing duration ($t_{sm}$) of less than 20 min. Therefore, $t_{sm}$ is a parameter that presents a significant effect, especially at the beginning of the treatment ($t_{sm} < 20$ min) with increasing coagulant concentration. The obtained results illustrate that the maximum removal of COD (57%) was achieved in a dosage of alum coagulant 500 mg/L and slow mixing of 15 min at pH 8.5.

By considering color and turbidity removal from the aforementioned ANOVA analysis, illustrated in Figure 8, the initial pH is the most operational parameter in the coagulation process affecting the removal efficiency of color and turbidity. To eliminate the maximum pollution in wastewater, we should first determine the optimum pH of the coagulant. In our study, the color and turbidity removal was maximum in a pH between 6 and 8.5. In this range, Al(OH)$_3$ and Al(OH)$_2^+$ are charged positively. Insoluble Al(OH)$_3$ species are also predominant. Because the colloidal particles are negatively charged, these cations will lead to the neutralization of the solution charge.

Yet, it is noticed that the pH changes during the CC. This is due to the formation of Al(OH)$_3$. However, Al(OH)$_3$ precipitation is possible only when the initial pH is higher than 4. This range will depend on the coagulant concentration. Therefore, the pH and the coagulant concentration have closely combined effects for well-determined values (pH > 4 and a higher coagulant concentration). Figure 8a,b shows the

![Figure 8](https://example.com/figure8.png)

**Figure 8.** 3D surfaces plot for color and turbidity removal efficiency: (a, b) coagulant concentration vs pH at $t_{sm}$ 20 min and (c, d) $t_{sm}$ vs initial pH at coagulant concentration 350 mg/L.
combined effect of pH and coagulant concentration in the elimination of pollution in this wastewater.

The domain of high color and turbidity removal (more than 80%) can be identified at coagulant concentrations between 250 and 500 mg/L and for the corresponding pH values of 6–8.5. Various researchers indicated the important effect of coagulant concentration and pH values. Azbar et al.67 proposed an optimum pH of 8 for 400 mg/L of coagulant concentration. GilPavas et al.69 have also found 93% of turbidity removal for 600 mg/L of coagulant salt at pH 9.4. Merzouk et al.28 noticed that decolorization was the most effective in a pH range between 4.0 and 7.8. The highest efficiency (87% color) at pH 6.4 was, therefore, achieved when aluminum concentration was about 40 mg/L. The decrease of pH in the solution remains small due to the limited addition of aluminum (40 mg/L).

The high value of aluminum concentration (250–500 mg/L) in this work in comparison to some research works and also to the theory18,23,28,35 can explain the rise of the pH range. Very similar results were obtained by other researchers.69,70 These later showed that the displacement or the widening of the optimal pH range of coagulation is mainly related to the concentration of aluminum salts.70

Although this parameter has a positive effect on the efficiency of coagulation, it is also mainly responsible for the presence of

Figure 9. Contour plots of OC (USD/m³) for the EC process: (a) electrolysis time (t) vs initial pH, (b) voltage vs initial pH, and (c) voltage vs electrolysis time (t).
residual aluminum in the treated water. In this way, finding an optimum coagulant concentration can avoid saturation of the water with aluminum(III) and, therefore, the appearance of dissolved aluminum.18 Letterman and Vanderbrook70 noted that residual aluminum is a significant problem in solutions that apply high concentrations of aluminum.

Figure 8c,d shows that, for a given coagulant concentration (i.e., 350 mg/L) and slow mixing (t_sm) duration, increasing pH leads to an increase in color and turbidity removal. It can be concluded that the use of Al\textsubscript{2}(SO\textsubscript{4})\textsubscript{3} as a coagulant in its optimal concentration for a suitable initial pH of the wastewater, gives a satisfactory removal of COD, color, and turbidity.

### 3.3. OC Assessment

Cost analyzing forecasts is a critical factor because they can help assess a company’s cost and determines the commercial applicability of the technology. The cost estimation analysis during the EC and CC processes includes the cost of electricity, chemical reagent, sludge disposal, and material as mentioned above in eqs 7 and 8. The effects of process parameters on OC (USD/m\textsuperscript{3}) are presented in Figures 9 and 10 for EC and CC, respectively. It could be inferred that OC was strictly dependent on electrolysis time (t) and voltage for the EC process, while it is influenced by the coagulant dosage in the case of the CC process. Figure 9 shows that for a given value of pH, increasing t and the applied voltage leads to an increase in the OC. The maximum of OC was about 4.59 USD/m\textsuperscript{3} obtained with a voltage equal to 12 V and t = 60 min. Applied voltage and t influence both the electrode consumption and electricity consumption for the EC process as depicted through eqs 9 and 10, respectively. This fact exhibits that energy consumption and electrode material cost were the highest contributors to the OC.

The aluminum coagulant concentration seemed to be the most operating parameter influencing the OC in the CC process as it is depicted in Figure 10. For a given pH and time of slow mixing (t_sm), increasing the concentration of coagulant leads to an increase in the OC.

The costs were varying between 0.314 and 4.59 and 0.113 and 0.467 USD/m\textsuperscript{3} for EC and CC, respectively. Figure 11 illustrates the percentage of OC contribution of electricity, electrode/coagulant consumption, sludge disposal, and chemical consumption to the total OC.

As expected, the cost of electricity and aluminum electrodes were the most contributors to the total operation cost in the EC process.
The experimental results closely agree with the conditions, the predicted removal efficiency of COD, color, and turbidity removal was set at the maximum values, while the OC was at a minimum.

The factors were set in a range study in each process. In order to validate the results of the model and to check its accuracy, lab practical tests were performed and compared to the predicted results under the optimum conditions in EC and CC. Table 10 summarizes the results of optimization and the comparison between the predicted and real performance of the EC and CC under the optimum conditions.

The results showed that the optimum conditions for the EC process were pH 9, an electrolysis time of 4 V, and t = 36.26 min; whereas they were pH 8.57, a coagulant concentration of 204.75 mg/L, and t = 28.41 min for the CC process. Under optimized conditions, the predicted removal efficiency of COD, color, and turbidity and OC were 62.58%, 99.33%, 98.34%, 0.4660 USD/m³ and 54.59% 96.60% 97.78% 0.2036 USD/m³ for EC and CC, respectively. The experimental results closely agree with the predicted values obtained using RSM. Therefore, these findings indicate that the models obtained are suitable for predicting and optimizing the EC and CC performance.

### 3.5. Comparative Study

At first sight, it is clearly obvious that the EC process is more performant in terms of COD, color, and turbidity removal compared to the CC process. However, the OC of EC was about 2 times higher than the OC of CC.

To ensure that the generated effluent meets the discharge standards, an analysis of residual aluminum and sulfate was done under optimal conditions. Aluminum concentration in treated water should be controlled because it is recognized for a long time as a toxic metal agent on gill-breathing and it may pose a potential risk to humans. Sulfate ion also has an adverse effect on the environment. A high concentration of sulfate will cause the death of invertebrates. In addition, it can be converted to hydrogen sulfide (H₂S) according to eq 24. The above-mentioned is a distasteful and highly toxic gas when it is released into the atmosphere.

$$\text{SO}_4^{2-} + 2\text{CH}_3\text{O} \rightarrow \text{H}_2\text{S} + 2\text{HCO}_3^-$$

Table 11 gives the results of residual aluminum, sulfate, and other pollutants in the treated solution under optimum conditions, compared to textile wastewater discharge standards. It is clear from Table 11 that the water treated by the EC process meets the local discharge standards in terms of residual aluminum and sulfate as well as for the other parameters. While, in the case of the CC process, the COD concentration slightly exceeds the discharge standards to the environment and has a high concentration of sulfate. In this technique, the optimum condition was 204.7 mg/L of aluminum sulfate, which corresponds to 172.4 mg/L of sulfate ion. Therefore, the relevant value of sulfate 678 mg/L in treated water becomes mainly from the wastewater. Thus, it has required a post-treatment to reduce the sulfate content. Such an alternative can add a supplement OC. We conclude also that this high concentration of sulfate in water can be reduced by the EC technique contrary to CC as reported by researchers.

Furthermore, the findings showed that less sludge formation was observed through the EC process. Similar results were found by Kobya et al. These scientists operated EC and CC processes

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**Table 10. Optimization Results**

| Conditions | EC Process | CC Process |
|------------|------------|------------|
| pH 9, electrolysis time (t) = 36.26 min, voltage = 4 V, CD = 57.1 A/m², desirability = 0.986 | % COD | % color | % TU | OC (USD/m³) |
| experimental values | 63.05 | 99.07 | 96.31 | 0.4705 |
| predicted values | 62.58 | 99.33 | 98.34 | 0.4660 |
| predicted error (%) | −0.75 | 0.26 | 2.06 | −0.95 |
| pH 8.57, coagulant concentration = 204.75 mg/L, t = 28.41 min, desirability = 0.898 | % COD | % Color | % TU | OC (USD/m³) |
| experimental values | 54.02 | 96.21 | 93.70 | 0.1975 |
| predicted values | 54.59 | 96.60 | 97.78 | 0.2036 |
| predicted error (%) | 1.04 | 0.40 | 4.17 | 3.02 |

**Table 11. Residual Aluminum, Sulfate, and Other Pollutants in Treated Water under Optimum Conditions**

| Parameter | Value |
|-----------|-------|
| aluminum (mg/L) | raw wastewater 0.461, EC process 0.245, CC process 0.1975 |
| sulfate (mg/L) | 500 |
| COD (mg/L) | 404 |
| SS (mg/L) | 325a |
| turbidity (NTU) | 64.03 |
| pH | 6.5 < pH < 9 |
| electric conductivity (mS/cm) | 3.39 |

aThe wastewater was filtered before treatment. bFinal pH after treatment. cSludge produced.
| refs | wastewater characteristics | optimal conditions | % pollutant removal | sludge produced (kg/m³) | OC |
|------|---------------------------|--------------------|-------------------|------------------------|----|
| 28   | synthetic textile wastewater: disperse red dye: 235 mg/L, turbidity: 0.15 NTU | CC process, Alum: 40 mg/L pH 6.4, high stirring speed: 180 rpm, slower stirring speed: 60 rpm | 87 % color | n.d | 0.32 ($/kg dye) |
|      |                           | EC process, Al−Al pH 6−9, t = 14 min, CD = 31.25 mA/cm² | 95 % color | n.d | 0.52 ($/kg dye) |
| 33   | textile wastewater, COD: 2031 mg/L, turbidity: 671 NTU | EC process, Al−Al−Al pH 5.5, t = 10 min, CD = 30 A/m² | 65 % COD, 83 % turbidity | 0.168 | 0.4 $/m³ |
|      |                           | CC process, Al₂(SO₄)₃: 1500 mg/L pH 6, t = 20 min, high stirring speed: 250 rpm, slower stirring speed: 50 rpm | 59 % COD, 90 % turbidity | 0.355 | 0.75 $/m³ |
| 69   | industrial textile wastewater: COD: 720 mg/L, turbidity: 161 NTU, color (λ_max = 600 nm): Abs: n.d | CC process, Alum: 600 mg/L pH 9.3 | 53 % COD, 97 % turbidity | 0.456 | 0.41 USD/m³ |
| 80   | synthetic textile wastewater: AR18 dye, color (λ_max = 508 nm): Abs: n.d | EC process, Al−Al pH 7, CD = 26 A/m², t = 40 min | 51 % color | 0.8 | n.d |
| 81   | textile wastewater, COD: 1550 mg/L, color: Abs 0.466 | CC process, Al₂(SO₄)₃: 1100 mg/L pH 6.5−7 | 33.4 % COD, 75.6 % color | 1.7 | n.d |
|      |                           | Fenton process, Fe dosage: 1093 mg/L, H₂O₂ dosage: 1600 mg/L pH 3.2, t = 150 min | 84.6 % COD, 98.6 % color | 0.792 | n.d |
| 82   | textile wastewater, COD: 990 mg/L, turbidity: 396 NTU | EC process, Al−Al pH 4.57, t = 10 min, CD = 4 mA/cm² | 97.1 % COD, 96 % turbidity, 98.5 % color | 1.44 | 0.283 US$/m³ |
| 83   | synthetic textile wastewater: dye concentration: 200 mg/L, color (λ_max = 480 nm): Abs: n.d | EC process, Al−Al pH 7.1, t = 5 min, CD = 15.5 A/cm² | 87.5 ± 0.5 % color | 0.315 $/m³ | n.d |
| 78   | textile wastewater: COD: 668 mg/L, color (λ_max = 600 nm), Abs: 0.212 | CC process, Al₂(OH)₃: 100 mg/L pH 7.1 | 24 ± 1 % color | n.d | n.d |
|      |                           | coagulation with FeSO₄: 450 mg/L pH 12, high stirring speed: 100 rpm, slower stirring speed: 40 rpm | 62 % COD, 99 % color | n.d | n.d |
| 84   | synthetic textile wastewater: reactive yellow 86, indanthrene blue RS, basic GR 4 and reactive yellow 145 dyes, turbidity: 303 NTU | EC process, Al−Al t = 90 min, pH 7.56, CD = 625 A/m² | 43 % COD, 97 % color | n.d | n.d |
|      |                           | CC process, Al₂(SO₄)₃: 450 mg/L pH 7.3, t = 5 min | 98 % color | n.d | n.d |
|      |                           | ozonation (O₃) astro, 15 min, pH 11, 20 mg O₃/min | maximum 96 %, color | 0.256 | 0.4705 USD/m³ |
|      |                           | EC process, Al−Al t = 36.26 min, voltage = 4 V, CD = 57.1 A/m² | 63.05 % COD, 96.31 % turbidity, 99.07 % color | 0.395 | 0.1975 USD/m³ |
|      |                           | CC process, Al₂(SO₄)₃: 204.75 mg/L pH 8.57, t = 24 h | 54.02 % COD, 96.21 % turbidity, 96.21 % color | 0.395 | 0.1975 USD/m³ |

This work: textile wastewater, COD: 2031 mg/L, turbidity: 64 NTU, color (λ_max = 669 nm), Abs: 0.518
for the treatment of textile wastewater, resulting in removal efficiency of COD and turbidity of about 65.83 and 59.90% for EC and CC, respectively. The generated sludge was about 0.168 kg/m³ for the EC process using aluminum anode, pH 7, and CD of 30 A/m² and 0.355 kg/m³ for CC using 1500 mg/L of Al₂SO₄·18 H₂O as a coagulant agent and pH 7.

Besides, the sludge generated should be removed before being released into the environment. In fact, sludge transportation and disposal were considered in the evaluation of OC.

Moreover, textile manufacturing consumed a considerable amount of fresh water about 300 m³/day as aforementioned, making treatment options that allow for water reuse attractive. Therefore, the effluent of treated water by the EC process is more suitable to be recycled in the industrial process to promote sustainability of textile manufacture and reduce the water burden. The idea of industrial water reuse offers a solution to reduce water consumption from natural resources, especially in water-scarce regions such as in the Middle East and North Africa, and ensure a sustainable water supply.

3.6. Comparison with Literature. As mentioned above, there are many techniques reported in the literature for textile wastewater treatment among them CC, EC, and other technologies such as AOPs. Nevertheless, few investigations deal with the comparison between EC and CC for textile wastewater based on OC.

It is revealed from Table 12 that EC and CC proved a high removal efficiency in terms of color and turbidity; however, the EC method seemed to be more efficient in COD removal. The same results were found in our work. Besides, the OC comparison between studies is not straightforward. Indeed, the cost analyses may be conducted by considering different terms from one study to another (e.g., considering chemical consumption and electricity and not sludge disposal and discharge). In our study, the OC was higher in the EC process compared to the CC process under the optimum conditions. This result was in harmony with those obtained by Merzouk et al. These researchers compared EC and CC processes to treat synthetic textile wastewater. They established that the OCs for EC and CC process are calculated at 0.52 and 0.32 $/kg dye, respectively (considering coagulant and power requirements for mixing for the CC process and the energy consumption and the mass consumption of the electrodes for EC). Unlike, Kobya et al. compared the CC and EC processes for real textile wastewater treatment. They noticed that by using the CC process, the OC was two times higher compared to EC. The OC includes all expenses associated with energy, electrode material, chemical costs, maintenance, sludge transportation and disposal, and labor. Nevertheless, the OC of EC and CC processes remained lower compared to other techniques such as AOPs.

Moreover, as depicted in Table 12, the amount of sludge produced by EC is small when compared to CC.

4. CONCLUSIONS

In the present study, optimization of CC and EC for the purification of textile wastewater was investigated using RSM coupled with CCD. The obtained model and the experimental data were in good agreement. The optimum levels of key parameters are a pH of 9, applied voltage of 4 V, residence time (t) of 36.26 min for EC and a pH of 8.57, coagulant concentration of 204 mg/L, and time of slowing mixing (t_m) of 28.41 min for the CC process. Under the optimum conditions, the experimental removal of COD, color, and turbidity and OC (USD/m³) were 63.05, 99.07, 96.31, 0.4705 USD/m³ and 54.02, 96.21, 97.78, 0.1975 USD/m³ for EC and CC, respectively, which are very close to predicted values. Both techniques are proven to be effective for pollutant removal. Even if the OC of the EC process was two times higher than that of the CC process, the treated water obtained by the EC process meets the discharge standards of textile effluents and it was more suitable to be recycled and reused in the textile industry.

The results highlight the applicability of the EC treatment process in the treatment of various industrial effluents, especially for real textile wastewater. EC provides a new direction for water recycling, which is meaningful for both the environment and industries providing a solution for sustainable water management in the future. Moreover, to make the EC process widespread on an industrial scale, further works should offer a better understanding of EC mechanisms especially related to COD removal.

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Notes

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