Optimization of the electrochemical oxidation of textile wastewater by graphite electrodes by response surface methodology and artificial neural network

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ABSTRACT

In this study, electrochemical oxidation of combed fabric dyeing wastewater was investigated using graphite electrodes. The response surface methodology (RSM) was used to design the experiments via the central composite design (CCD). The planned experiments were done to track color changes and chemical oxygen demand (COD) removal. The experimental results were used to develop optimization models using RSM and the artificial neural network (ANN) and they were compared. The developed models by the two methods were in good agreement with the experimental results. The optimum conditions were found at 150 A/m², pH 5, and 120 min. The removal efficiencies for color and COD reached 96.6% and 77.69%, respectively. The operating cost at the optimum conditions was also estimated. The energy and the cost of 1 m³ of wastewater required 34.9 kWh and 2.58 US$, respectively. The graphite electrodes can be successfully utilized for treatment of combed fabric dyeing wastewater with reasonable cost.

Key words: artificial neural network, combed fabric dyeing wastewater response surface method, electro oxidation, graphite electrodes

HIGHLIGHTS

- Electrochemical oxidation of combed fabric dyeing wastewater was investigated using graphite electrodes.
- The optimum conditions were found at 150 A/m², pH 5, and 120 min.
- The removal efficiencies for the color and COD reached 96.60% and 77.69%, respectively.

1. INTRODUCTION

The textile industry is a competitive industry that has a crucial role in any developed/developing community. Unfortunately, the textile industries produce a large amount of wastewater (GilPavas et al. 2020). According to Neill et al. (1999), a large amount of water (125–250) L is required for 1 kg of the finished product (Neill et al. 1999). Thus the textile industry produces enormous wastewater quantities with dyestuff, surfactants, and additives, which have dangerous effects on the environment and humans (Aravind et al. 2016; Saleh et al. 2019a).

In the last decade, several methods were examined to treat textile wastewater. Yagub and colleagues (2014) reviewed the use of adsorption in dye removal from the aqueous solutions (Yagub et al. 2014). The regeneration of the adsorbent and the production of secondary pollutants have limited the process (Yalvaç et al. 2020). Membrane technology was also applied to treat and recycle textile wastewater (Cengiz Yatmaz et al. 2017; Nadeem et al. 2019). The initial cost and the fouling issues had not favored the membrane filtration (Lin et al. 2016). Coagulation and flocculation methods were also employed (Verma et al. 2012), but the possibility of generating an extra sludge had become a significant concern (Amaral-Silva et al. 2016). According to Soares et al. (2017), biological treatment may not be sufficient if applied alone (Soares et al. 2017). Advanced oxidation processes (AOPs) were implemented (Carvalho & Carvalho 2017). Despite their high performances, AOPs may generate other pollutants (Gagol et al. 2018). In regard, the electrochemical techniques have become more favorable because of their functionality, simplicity, safety, eco-friendliness, and low costs (Brillas & Martínez-Huitle 2015). The electrochemical treatment can degrade the pollutant in the textile wastewater via direct or indirect
oxidation (Radjenovic & Sedlak 2015). The electrochemical techniques mainly include electrocoagulation/electro flotation, electro oxidation, and electro reduction (Brillas & Martínez-Huitle 2015).

Electro oxidation (EO) has become the most popular electrochemical technique because it eliminates the redox chemicals, facilitates the reaction controls by managing the current or potential, and increases the onsite treatment possibility (Panizza & Cerisola 2005). The material of the anode can influence the electrochemical mechanism and the produced material (Särkkä et al. 2015). Lead and lead oxide (Awad & Abo Galwa 2005), boron-doped diamond (Koparal et al. 2007; Zhu et al. 2018; Kuchtová et al. 2020), dimensionally stable anode (DSA) (Tavares et al. 2012; Zhang et al. 2012), and activated carbon cloth (ACC) (Cukierman 2013) electrodes were used as a base for the electrode in the EO process. Graphite electrodes had the attention of the researchers because of their low cost, low resistivity, chemical inertness and good conductivity (Kariyajjanavar et al. 2013a, 2013b). The degradations of the dyes by graphite electrodes can occur via direct or indirect oxidation (Hamza et al. 2011). The direct oxidation occurs at the graphite surface or electron transfer directly to the graphite anode. The presence of chloride in the textile wastewater effluents makes indirect oxidation the dominant degradation mechanism. The oxidation of chloride at the graphite anode yields active species that can oxidize the pollutants (Isik et al. 2020).

The combed fabric dyeing wastewater has a high chlorine concentration (14,515 mg/L). Thus, the graphite electrode can be considered an attractive solution. This study aims to explore the removal efficiency of color and COD from the combed fabric dyeing wastewater by graphite electrodes. The affecting parameters (pH, time, and applied current) were optimized. The traditional methods of optimization are not able to describe the complicated interaction among the variables and the responses (Taherkhani et al. 2018). The response surface methodology (RSM) is a mix of statistical and mathematical techniques that could be used to solve this issue (Körbahti & Demirbüken 2017; Gadekar & Ahammed 2019). The RSM starts with experiment design, followed by model fitting and verification, and ends with determining the optimum conditions step (Darvishmotevallí et al. 2019). In this study, the central composite design (CCD) was used to design the experiments. Furthermore, the artificial neural network (ANN) was used to model and optimize the electrooxidation process. ANN is another modeling technique that has the advantage of the ability to create a non-linear relationship between the affecting factors and the proposed responses without any prior knowledge of the relationship nature (Elfghi 2016; Yabalak & Yilmaz 2019). The design matrix and the final pH values obtained from RSM were utilized to build an artificial neural network. In this way, the modeling results had a more precise prediction. So, the results from the two methods were compared and examined statistically. Finally, the operational cost of the electrooxidation of the combed fabric dyeing wastewater by the graphite electrodes at the optimum conditions was analyzed.

2. MATERIAL AND METHOD

2.1. Material

Combed fabric dyeing wastewater was collected from textile factory in Gaziantep province, Turkey. The wastewater was characterized based on the American public health association standard methods (Rice et al. 2017). The characteristics are shown in Table 1.

| Parameter                  | Unit | Value |
|----------------------------|------|-------|
| COD                        | mg/L | 833   |
| TSS                        | mg/L | 12    |
| Sulphate (SO₄)             | mg/L | 164   |
| Total Nitrogen             | mg/L | 51.3  |
| Total Kjeldahl Nitrogen    | mg/l | 47.8  |
| pH                         | –    | 6.9   |
| Conductivity               | μS/cm| 3,030 |
| Color                      | Pt-Co| 2,213 |
| Chloride                   | mg/L | 1,451 |
2.2. Experimental setup

The electrooxidation experiments were conducted in a batch reactor (Figure 1). The reactor consists of a 400 mL borosilicate glass reactor, two graphite electrodes with 5 cm width \( \times \) 8 cm height \( \times \) 2 mm thickness, a digital DC power supply (AATech ADC-3303D, the maximum voltage of 30 V) with two connecting wires, and a magnetic stirrer (Wisd -Wisestir MSH-20A) with a Teflon-covered magnetic stirring bar. 200 mL of the textile wastewater was inserted inside the borosilicate glass reactor, followed by placing the electrode pairs with a 2 cm distance between them and connected to the DC power supply with the wires. The stirring process at 300 rpm continued simultaneously with the electrooxidation experiments.

At certain times, the samples were collected and centrifuged at 6,000 rpm for 5 min. The pH and the conductivity were measured by pH/Cond. 340i Handheld multi meter, WTW. Color changes were noticed by UV-visible spectrophotometer (Hach DR 3900) using Platinum–Cobalt (Pt-Co) method following the Standard Method No. 2120 (Rice et al. 2017). The chemical oxygen demand (COD) was measured using the closed reflux method-titrimetric method following Standard Method No. 5220C (Rice et al. 2017). The concentration of the chloride before and after the electro oxidation process was measured by argentometric method following Standard Method No. 4500B (Rice et al. 2017). The removal efficiency was calculated using Equation (1).

\[
\text{Removal (\%)} = \frac{\text{Initial Concentration} - \text{Final Concentration}}{\text{Initial Concentration}}
\]  

(1)

2.3. RSM and ANN

Central composite design (CCD) as an approach in response surface methodology (RSM) was used to optimize the affecting parameter. CCD is a statistical method for multivariate nonlinear model development. The developed model can be used to explore the relationship between the parameters. CCD is also applied for regression model equations calculation. The applied current, pH, and time were used as model variables to design the experiments by Design Expert Version 11.0 [Stat-Ease]. The removal efficiencies obtained from the preliminary studies, \( \alpha \)-values for the low and maximum level, and the economic perspectives were the basis for the ranges of the variables selection process. The ranges of the factors were selected based on many criteria. Table 2

![Experimental setup for the electrooxidation.](image)

Table 2 | Independent variables ranges

| Variable | Unit | Factor | Low | High | \(- \alpha\) | \(+ \alpha\) |
|----------|------|--------|-----|------|-------------|-------------|
| Current  | A/m² | A      | 50  | 150  | 15.91       | 184.09      |
| pH       | –    | B      | 5   | 9    | 3.64        | 10.36       |
| Time     | Min  | C      | 60  | 120  | 39.55       | 140.45      |

Figure 1 | Experimental setup for the electrooxidation.
represents the variables and their ranges. In total, 20 experiments were carried out to track the effects of the parameter changes on COD and color.

ANN modeling was integrated with the RSM using the pH values at the end of the experiments. In the ANN, the network type was a feed-forward artificial neural network. The network included an input layer, a hidden layer, and an output layer. The experiment matrix in the CCD was used as a base for the ANN modeling. The input for the ANN was the experiment conditions in addition to the final pH. The output layer was the removal efficiencies for color and COD. The neurons required in the hidden layer were determined by trial and error to obtain the maximum regression with minimum errors. In this study four neurons were used. The designed ANN is shown in Figure 2.

3. RESULT AND DISCUSSION

3.1. Color removal

CCD was used to study the effect of the independent variables on color removal. The results were tested by linear, 2FI, quadratic, and cubic models. Based on their regression coefficients, the linear model was selected to represent the results. Table 3 shows the regression coefficients for the tested models.

The prediction ability of the developed model was also examined by the analysis of variance (ANOVA). According to Table 4, the model has a large F-value (101.97) and a p-value smaller than 0.0001. These values show that the model is significant and can be used in color removal prediction. The model was also found to be non-significant in the lack of fit test. The model has a predicted $R^2$ of 0.91 that reasonably agreed with the adjusted $R^2$ of 0.94 since the difference is less than 0.20. Also, the model has an adequate precision since the model ratio 31.42 is larger than 4.

The removal of color can be expected using Equation (2).

\[
\text{Color Removal} \ (%) = 74.99 + 0.07 \times \text{Current} - 0.17 \times \text{pH} + 0.09 \times \text{Time}
\]  

![Figure 2](image) | The designed ANN.

| Source | Sequential p-value | Lack of Fit p-value | Adjusted $R^2$ | Predicted $R^2$ | Note |
|--------|--------------------|---------------------|----------------|----------------|------|
| Linear | 1.22E-10           | 0.11                | 0.94           | 0.91           | Suggested |
| 2FI    | 0.18               | 0.13                | 0.95           | 0.87           | – |
| Quadratic | 0.50            | 0.10                | 0.95           | 0.82           | – |
| Cubic  | 0.62               | 0.02                | 0.94           | –1.77          | Aliased |
3.2. COD removal

COD removal by the electro-oxidation method was also examined and modeled by the CCD method. The experimental results were fitted to the linear, 2FI, quadratic, and cubic models. The quadratic model had the highest regression values (Table 5). The quadratic model was reduced to improve the regression factor and to have a better data description. The reduced quadratic model had an adjusted $R^2$ of 0.97, while the predicted $R^2$ 0.95. The reduced quadratic model had adequate precision with a ratio of 44.12.

Analysis of variance test (ANOVA) was also applied for COD removal model. The model was found to be significant with F-value and $p$-value of 138.01 and $<0.0001$, respectively. The model was also examined by lack of fit test. According to F-value (4.62), the model was good of a fit, and the lack of fit is not significant. The significances of model terms were investigated and shown in Table 6.

Accordingly, the developed model can be used to predict the removal efficiency of COD, as shown in Equation (3).

$$\text{COD Removal} \text{ (°)} = -3.482 + 1.068 \times \text{Current} - 1.825 \times \text{pH} + 0.115 \times \text{Time} - 0.004 \times \text{Current}^2$$  \hspace{1cm} (3)

3.3. ANN results

The used ANN to predict color and COD removal is shown in Figure 2. The developed ANN was tested by Levenberg Marquartz (LM) algorithm with log sigmoidal as a transform function (Hammoudi et al. 2019). Table 7 shows the performance of the network and the errors calculation based on different indicators.

**Table 4** | ANOVA and the lack of fit tests results for color removal

| Source | Sum of squares | df | Mean | F-value | $p$-value |
|--------|----------------|----|------|---------|-----------|
| Model  | 250.11         | 3.00 | 85.37 | 101.97  | 1.22E-10 |
| A-Current | 146.76        | 1.00 | 146.76 | 179.50  | 4.10E-10 |
| B-pH   | 1.63           | 1.00 | 1.63  | 1.99    | 0.18      |
| C-Time | 101.72         | 1.00 | 101.72 | 124.42  | 5.88E-09 |
| Residual | 13.08         | 16.00 | 0.82  | –       | –         |
| Lack of Fit | 11.45       | 11.00 | 1.04  | 3.19    | 0.10      |
| Pure Error | 1.63        | 5.00 | 0.33  | –       | –         |

**Table 5** | Regression coefficients for the tested models for COD removal

| Source | Sequential p-Value | Lack of Fit p-value | Adjusted $R^2$ | Predicted $R^2$ | Note |
|--------|--------------------|---------------------|----------------|-----------------|------|
| Linear | 0.16E-03           | 0.20E-03            | 0.65           | 0.50            | –    |
| 2FI    | 0.96               | 0.11E-03            | 0.58           | 0.46            | –    |
| Quadratic | 5.53E-07       | 0.07                | 0.97           | 0.91            | Suggested |
| Cubic  | 0.50               | 0.02                | 0.97           | –0.23           | Aliased |

Reduced Quadratic model: Adjusted $R^2$ 0.97, Predicted $R^2$ 0.95.

**Table 6** | ANOVA and the lack of fit tests results for COD removal

| Source | Sum of squares | df | Mean square | F-value | $p$-value |
|--------|----------------|----|-------------|---------|-----------|
| Model  | 4,857.24       | 4.00 | 1,214.31    | 138.01  | 1.22E-11 |
| A-Current | 3,181.33      | 1.00 | 3,181.33    | 361.56  | 6.55E-12 |
| B-pH   | 181.63         | 1.00 | 181.63      | 20.64   | 0.39E-03 |
| C-Time | 161.60         | 1.00 | 161.60      | 18.37   | 0.65E-03 |
| $A^2$  | 1,332.68       | 1.00 | 1,332.68    | 151.46  | 3.06E-09 |
| Residual | 131.98        | 15.00 | 8.80       | –       | –         |
| Lack of Fit | 119.10       | 10.00 | 11.91      | 4.62    | 0.05      |
| Pure Error | 12.89         | 5.00 | 2.58        | –       | –         |

Uncorrected Proof
The MSE and RMSE for the ANN models (color and COD) were lower than the RSM models. The lower value means higher description and prediction capabilities (Gadekar & Ahammed 2019). MAPE test is a method to determine the accuracy of the model (Mohamed 2019). Lower values for the MAPE are favorable and mean the model is more accurate. In this case, the ANN models have lower values. ANN models have higher regression coefficients \( R^2 \) (Saleh et al. 2019b). To ensure that the models are validated in describing the electro oxidation process, the experiments conducted before and after the modeling process. The experimental results and the modeled results are shown in Table 8.

### 3.4. Parameters effects

The effects of current density, pH, and time on the electrochemical oxidation of combed fabric dyeing wastewater were explored. Combed fabric dyeing wastewater was exposed to varied current densities (50–150 A/m²). The current density is linearly proportional to color removal (Figure 3(a) and 3(b)). The maximum color removal (95%) occurred at the current 150 A/m², while the minimum color removal was 82% at the current density of 50 A/m². Figure 3(c) and 3(d) show the contour map for the removal of color using electrooxidation process with the changes in the current, pH, and time.

### Table 7 | Performance indicator for color and COD removal models

| Performance | Color Removal | COD removal |
|-------------|---------------|-------------|
|             | RSM           | ANN         | RSM           | ANN         |
| MSE         | 0.65          | 0.15        | 6.6           | 1.24        |
| RMSE        | 0.81          | 0.39        | 2.56          | 1.11        |
| MAPE        | 0.73          | 0.26        | 4.71          | 0.93        |
| \( R^2 \) predicted | 0.91      | 0.98        | 0.95          | 0.97        |

### Table 8 | The design matrix among the experiments results and the expected data

| Run | Current (A/m²) | pH | Time (min) | Color Removal (%) | Experimental | RSM | ANN | COD removal (%) | Experimental | RSM | ANN |
|-----|----------------|----|------------|-------------------|--------------|-----|-----|----------------|--------------|-----|-----|
| 1   | 100.00         | 10.36 | 90.00     | 88.61            | 87.94        | 88.80 | 52.89 | 56.58        | 52.88        |
| 2   | 15.91          | 7.00  | 90.00     | 81.92            | 83.01        | 82.48 | 8.26  | 10.10        | 8.31         |
| 3   | 100.00         | 7.00  | 90.00     | 88.34            | 88.52        | 88.37 | 59.80 | 62.72        | 62.64        |
| 4   | 100.00         | 7.00  | 90.00     | 88.40            | 88.52        | 88.37 | 62.81 | 62.72        | 62.64        |
| 5   | 50.00          | 5.00  | 120.00    | 88.39            | 88.32        | 88.14 | 43.18 | 45.01        | 43.18        |
| 6   | 100.00         | 7.00  | 90.00     | 88.32            | 88.52        | 88.37 | 62.81 | 62.72        | 62.64        |
| 7   | 150.00         | 9.00  | 60.00     | 89.38            | 88.73        | 89.37 | 62.00 | 61.36        | 61.37        |
| 8   | 50.00          | 5.00  | 60.00     | 82.83            | 82.86        | 82.44 | 44.63 | 38.13        | 48.41        |
| 9   | 150.00         | 5.00  | 120.00    | 96.60            | 94.88        | 96.59 | 77.69 | 75.54        | 77.69        |
| 10  | 150.00         | 5.00  | 60.00     | 90.06            | 89.42        | 89.68 | 67.98 | 68.66        | 67.98        |
| 11  | 100.00         | 7.00  | 39.55     | 82.91            | 83.93        | 82.80 | 58.68 | 56.93        | 58.66        |
| 12  | 184.09         | 7.00  | 90.00     | 92.91            | 94.04        | 92.80 | 59.71 | 61.43        | 59.71        |
| 13  | 100.00         | 7.00  | 90.00     | 88.36            | 88.52        | 88.37 | 64.81 | 62.72        | 63.45        |
| 14  | 100.00         | 3.64  | 90.00     | 88.79            | 89.11        | 89.29 | 63.84 | 68.85        | 63.81        |
| 15  | 50.00          | 9.00  | 60.00     | 82.83            | 82.17        | 82.55 | 28.72 | 30.84        | 28.85        |
| 16  | 50.00          | 9.00  | 120.00    | 88.48            | 87.63        | 88.33 | 40.29 | 37.72        | 40.27        |
| 17  | 100.00         | 7.00  | 90.00     | 88.34            | 88.52        | 88.37 | 62.81 | 62.72        | 62.64        |
| 18  | 100.00         | 7.00  | 140.45    | 92.50            | 93.11        | 92.51 | 69.42 | 68.50        | 69.38        |
| 19  | 150.00         | 9.00  | 120.00    | 92.77            | 94.19        | 92.76 | 71.07 | 68.24        | 71.07        |
| 20  | 100.00         | 7.00  | 90.00     | 89.75            | 88.52        | 88.37 | 62.81 | 62.72        | 62.64        |
The removal efficiency of COD also increased with the current density (Figure 4(a)). The relationship between COD degradation and the applied current was found to be quadratic (Figure 4(b)). The maximum COD degradation at higher current density was 76%, while the minimum was 31% at a lower value (Figure 4(c)). As shown in Table 1, the textile wastewater contains chloride with concentration of 1,451 mg/L. The chloride concentration decreased 70 mg/L at the end of the reaction. In the electrooxidation method, chloride (Cl\(^{-}\)) can be converted to different forms (Periyasamy & Muthuchamy 2018). Equation (4) presents the generation of chlorine (Cl\(_2\)) at the anode side, while Equation (5) shows the reaction at the cathode side (Rajkumar & Muthukumar 2017).

\[
2\text{Cl}^- \rightarrow \text{Cl}_2 + 2e^- \quad (4)
\]

\[
2\text{H}_2\text{O} + 2e^- \rightarrow \text{H}_2 + 2\text{OH}^- \quad (5)
\]

The generated chlorine reacts with water to form the hypochlorous acid (Equation (6)) (Sirés et al. 2014). The hypochlorous acid is a weak acid, which dissociates into hypochlorite and hydrogen ion, as shown in Equation

![Figure 3](http://iwaponline.com/wst/article-pdf/doi/10.2166/wst.2021.240/904495/wst2021240.pdf)

**Figure 3** | (a) Surface color removal response for current and pH effects, (b) surface color removal response for current and time, (c) Contour response for current and pH effects on color removal, (d) Contour response for current and time effects on color removal.
The increases in the removal of both color (Figure 3(c)) and COD (Figure 4(b)) with the increases in the current density can be related to the generation rate of hypochlorite. At higher current density, the generation rate of hypochlorite increases consequently, the degradation of color and COD also raised, as shown in Equation (8).

\[
OCl^- + \text{Pollutant} \rightarrow \text{Product} + \text{CO}_2 + \text{H}_2\text{O} + \text{Cl}^- \tag{8}
\]

The effect of pH was also optimized. Different experiments were conducted at different pH values (5–9). The removal efficiencies of color and COD had inverse relationships with pH (Figures 3(c) and 4(c)). The maximum removals for color and COD were found to be at pH 5, while the lowest values were at pH 9. At alkali medium, the ionizations of chlorine/hypochlorite ions are low (Isik et al. 2020). In addition to that, the productions of

\[
\text{Cl}_2 + \text{H}_2\text{O} \rightarrow \text{HOCl} + \text{H}^+ + \text{Cl}^- \tag{6}
\]

\[
\text{HOCl} \rightarrow \text{H}^+ + \text{OCl}^- \tag{7}
\]
chlorate or perchlorate are more favorable (Equation (9)). In the acidic medium, the generations of chlorine/chloride from the hypochlorous acid are higher (Equation (6)). The presence of chloride/chlorine in the wastewater increases the degradation efficiency since it can oxidize the organic at the anode or in the solution (Mussa et al. 2015). Thus the oxidation of COD and color at lower pH values (pH 5) are higher.

\[
6OCl^- + 3H_2O \rightarrow \frac{3}{2}O_2 + 6H^+ + 2ClO_2^- + 4Cl^- + 6e^- \tag{9}
\]

The optimum time required to degrade color and COD was found to be 120 min (Figures 3(d) and 4(d)). The removal efficiencies increased with the increases in the time. At 60 min, the removal efficiencies for color and COD were 89 and 68%, respectively. The removal efficiencies increased to reach 95% for color and 76% for COD at 90 min.

The optimum conditions for textile wastewater treatment by graphite electrode were determined. The maximum color and COD removal was when the applied current reached 150 A/cm², pH was 5, and the reaction continued to 120 min. At the optimum conditions, color was removed with a percentage of 96.6% and COD was degraded with a percentage of 77.7%. The electro oxidation of textile wastewater by the graphite electrodes had successfully reduced color and COD to meet the Turkish discharge standards of textile industry wastewaters (Hukuk ve Mevzuat Genel Müdürlüğü 2004). Table 9 shows the initial and final concentration among with the standard concentration.

### 3.5. Anode efficiency and cost analysis

The operation cost is the key factor in treating the wastewater. In this study, a cost analysis was accomplished to determine the feasibility of use electro oxidation treatment of the textile wastewater by graphite electrode. The efficiency of the graphite electrode based on the specific energy was determined using Equations (10)–(12) (Dizge et al. 2018; Ukundimana et al. 2018).

\[
SEG \text{ (kWh/m}^3) = \frac{V_o \times I \times t}{1000 \times V} \tag{10}
\]

\[
SEG \text{ (kWh/kg color) } = \frac{V_o \times I \times t}{1000 \times V \times (Color_i - Color_f)} \tag{11}
\]

\[
SEG \text{ (kWh/kg COD) } = \frac{V_o \times I \times t}{1000 \times V \times (COD_i - COD_f)} \tag{12}
\]

where; SEG is the graphite electrode specific energy, \(V_o\), I, t, and V are the applied voltage (V), current (A), time (h), and wastewater volume (m³), respectively. \(COD_i\) and \(COD_f\) are the initial and final chemical oxygen demand (kg/L).

The treatment cost for 1 m³ of the textile wastewater was estimated by multiplying the SEG (kWh/m³) by the cost of the kilowatt per hour. According to the Turkish Electricity Distribution Company the cost of 1kWh is about 0.074 US$. Table 10 presents the cost analysis of the treatment of textile wastewater by the electro oxidation at the optimum conditions.

The results shown in Table 9 shows that the treatment of the textile wastewater by the electro oxidation method when the graphite was the electrode is efficient. The energy and the cost of 1 m³ of wastewater required just 34.90 kWh and 2.58 US$, respectively. These results are lower than the study obtained by Isik and colleagues (2020) who used activated carbon cloth as electrodes (Isik et al. 2020). A comparison with other works are shown in Table 11.

### Table 9 | Initial, final, and the standard values for the different parameters

| Unit       | Initial Value | Final value | Standard |
|------------|---------------|-------------|----------|
| Color      | Pt-Co         | 2,213       | 75.2     | 260      |
| pH         | -             | 6.9         | 6.5      | 6–9      |
| COD        | mg/L          | 833         | 185.8    | 200      |
4. CONCLUSION

The textile industry is a competitive industry that has a crucial role in any developed/developing community. Unfortunately, the textile industries produce a large amount of wastewater. The textile wastewater has large quantities of dyestuff, which have dangerous effects on the environment and humans. In this study, the treatment of combed fabric dyeing wastewater by graphite electrode was explored for the removal of color and COD. The affecting parameters (pH, time, and current) were optimized using the response surface methodology (RSM) and artificial neural network (ANN). As expected, the developed ANN has described the results more precisely. Based on the two models, the optimum conditions were found to be 150 A/m², pH 5, and 120 min. The removal efficiencies for color and COD reached 96.6% and 77.69%, respectively. The operating cost at the optimum conditions was also estimated. The energy and the cost of 1 m³ of wastewater required just 34.90 kWh and 2.58 US$, respectively. Although an efficient treatment application was utilized successfully to treat combed fabric dyeing wastewater, the graphite electrodes still suffer from carbon corrosion problems, which affects the electro-oxidation process performance.

ACKNOWLEDGEMENT

This project supported by Scientific Research Projects Council of Mersin University (Project number: 2021-1-TP2-4262).

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.
REFERENCES

Amaral-Silva, N., Martins, R., Paiva, C., Castro-Silva, S. & Quinta-Ferreira, R. 2016 A new winery wastewater treatment approach during vintage periods integrating ferric coagulation Fenton reaction and activated sludge. Journal of Environmental Chemical Engineering 4, 2207–2215.

Aravind, P., Subramanyan, V., Ferro, S. & Gopalakrishnan, R. 2016 Eco-friendly and facile integrated biological-cum-photo assisted electrooxidation process for degradation of textile wastewater. Water Research 93, 230e241.

Awad, H. & Abo Galwa, N. 2005 Electrochemical degradation of Acid Blue and Basic Brown dyes on Pb/PbO2 electrode in the presence of different conductive electrolyte and effect of various operating factors. Chemosphere 61, 1327–1335.

Brillas, E. & Martínez-Huitle, C. 2015 Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review. Applied Catalysis B: Environmental 166–167, 603–645.

Carvalho, S. & Carvalho, N. 2017 Dye degradation by green heterogeneous Fenton catalysts prepared in presence of Camellia sinensis. Journal of Environmental Technology 187, 82–88.

Cengiz Yatmaz, H., Dizge, N. & Kurt, M. S. 2017 Combination of photocatalytic and membrane distillation hybrid processes for reactive dyes treatment. Environmental Technology 38 (21), 2745–2751.

Cukierman, A. 2013 Development and environmental applications of activated carbon cloths. ISRN Chemical Engineering 261523.

Darvishmotevali, M., Zarei, A., Moradnia, M., Noorisepehr, M. & Mohammadi, H. 2019 Optimization of saline wastewater treatment using electrochemical oxidation process: prediction by RSM method. MethodX 6, 1101–1113.

Dizge, N., Akarsu, C., Ozay, Y., Gulsen, H. & Adiguzel, S. 2018 Sono-assisted electrocoagulation and crossflow membrane processes for brewery wastewater treatment. Journal of Water Process Engineering 21, 52–60.

Elgí, F. 2016 A hybrid statistical approach for modeling and optimization of RON: a comparative study and combined application of response surface methodology (RSM) and artificial neural network (ANN) based on design of experiment (DOE). Chemical Engineering Research and Design 113, 264–272.

Gadekar, M. R. & Ahammed, M. M. 2019 Modelling dye removal by adsorption onto wood treatment residuals using combined response surface methodology-artificial neural network approach. Journal of Environmental Management 231, 241–248.

Gogol, M., Przyjazny, A. & Boczkaj, G. 2018 Wastewater treatment by means of advanced oxidation processes based on cavitation—a review. Chemical Engineering Journal 338, 599–627.

GilPavas, E., Dobrosz-Gómez, I. & Gómez-García, M.-Á. 2020 Efficient treatment for textile wastewater through sequential electrocoagulation, electrochemical oxidation and adsorption processes: optimization and toxicity assessment. Journal of Electroanalytical Chemistry 878, 114578.

Hammoudi, A., Moussaceb, K., Belebchouche, C. & Dahmoune, F. 2019 Comparison of artificial neural network (ANN) and response surface methodology (RSM) prediction in compressive strength of recycled concrete aggregates. Construction and Building Materials 209, 425–436.

Hamza, M., Ammar, S. & Abdelhed, R. 2011 Electrochemical oxidation of 1,3,5-trimethoxybenzene in aqueous solutions at gold oxide and lead dioxide electrodes. Electrochimica Acta 56, 3785–3789.

Huluk ve Mevzuat Genel Müdürlüğü KONTROLÜ YÖNETMELİĞİ, 2004 SU KIRILİŞİ KONTROLU KURALLARI. Resmi Gazete, Ankara.

Isik, Z., Unal, B. & Karagunduz, A. 2015 Electrochemical treatment of textile dye bath wastewater using activated carbon cloth electrodes. Avicenna Journal of Environmental Health Engineering 7 (1), 47–52.

Kariyajinanavar, P., Narayana, J. & Nayaka, Y. 2013a Electrochemical degradation of C.I. Vat brown I dye on carbon electrode. Advanced Chemistry Letters 1, 32–39.

Kariyajinanavar, P., Narayana, J. & Nayaka, Y. 2013b Degradation of textile dye C.I. Vat black 27 by electrochemical method by using carbon electrodes. Journal of Environmental Chemical Engineering 1 (4), 975–980.

Kaur, P., Imteaz, M. A., Sillanpaa, M., Sangal, V. K. & Kushwaha, J. P. 2020 Parametric optimization and MCR-ALS kinetic modeling of electro oxidation process for the treatment of textile wastewater. Chemometrics and Intelligent Laboratory Systems 205, 104027.

Kong, Y., Yuan, J., Wang, Z., Yao, S. & Chen, Z. 2009 Application of expanded graphite/attapulgite composite materials as electrode for treatment of textile wastewater. Applied Clay Science 46, 358–362.

Koparal, A. S., Yavuz, Y., Gurel, C. & Ogunveren, U. B. 2007 Electrochemical degradation and toxicity reduction of C.I. Basic Red 29 solution and textile wastewater by using diamond anode. Journal of Hazardous Materials 145, 100–108.

Kürbahri, B. K. & Demirbülbü, P. 2017 Electrochemical oxidation of resorcinol in aqueous medium using boron-doped diamond anode: reaction kinetics and process optimization with response surface methodology. Frontiers in Chemistry 5, 75.

Kuchková, G., Chýlková, J., Váňa, J., Vojs, M. & Dušek, L. 2020 Electrodioxidative decolorization and treatment of model wastewater containing acid dye 80 on boron doped diamond and platinum anodes. Journal of Electroanalytical Chemistry 863, 114036.

Lin, J., Ye, W., Baltaru, M.-C., Tang, Y. P., Bernstein, N. J., Gao, P., Balta, S., Vlad, M., Volodin, A., Sotto, A., Luis, P., Zydne, A. L. & Bruggen, B. V. d. 2016 Tight ultrafiltration membranes for enhanced separation of dyes and Na2SO4 during textile wastewater treatment. Journal of Membrane Science 514, 217–228.

Mohamed, Z. E. 2019 Using the artificial neural networks for prediction and validating solar radiation. Journal of the Egyptian Mathematical Society 27, 47.
Mussa, Z. H., Othman, M. R. & Abdullah, M. P. 2015 Electrochemical oxidation of landfill leachate: investigation of operational parameters and kinetics using graphite-PVC composite electrode as anode. Journal of the Brazilian Chemical Society 26 (5), 939–948.

Nadeem, K., Geyer, G. T., Keskinler, B. & Dizge, N. 2019 Investigation of segregated wastewater streams reusability with membrane process for textile industry. Journal of Cleaner Production 228, 1437–1445.

Neill, C. O., Hawkes, F., Hawkes, D., Lourenço, N., Pinheiro, H. & Delee, W. 1999 Colour in textile effluents-sources, measurement, discharge consents and simulation: a review. Journal of Chemical Technology & Biotechnology 74, 1009–1018.

Panizza, M. & Cerisola, G. 2005 Application of diamond electrodes to electrochemical processes. Electrochimica Acta 51, 191–199.

Periyasamy, S. & Muthuchamy, M. 2018 Electrochemical oxidation of paracetamol in water by graphite anode: effect of pH, electrolyte concentration and current density. Journal of Environmental Chemical Engineering 6, 7558–7567.

Radjenovic, J. & Sedlak, D. 2015 Challenges and opportunities for electrochemical processes as next-generation technologies for the treatment of contaminated water. Environmental Science & Technology 49 (19), 11292–11302.

Rajkumar, K. & Muthukumar, M. 2017 Response surface optimization of electro-oxidation process for the treatment of C.I. Reactive Yellow 186 dye: reaction pathways. Applied Water Science 7, 637–652.

Rice, E., Baird, R. & Eaton, A. 2017 Standard Methods for the Examination of Water and Wastewater. American Public Health Association, American Water Works Association, Water Environment Federation, USA.

Saleh, M., Yalvaç, M., Arslan, H. & Gün, M. 2019a Malachite Green dye removal from aqueous solutions using invader centaurea solstitialis plant and optimization by response surface method: kinetic, isotherm, and thermodynamic study. European Journal of Environmental Science and Technology 17, 755–768.

Saleh, M., Yalvaç, M. & Arslan, H. 2019b Optimization of remazol brilliant blue R adsorption onto xanthium italicum using the response surface method. Karbala International Journal of Modern Science 8 (1(8)).

Särkkä, H., Bhatnagar, A. & Sillanpää, M. 2015 Recent developments of electro-oxidation in water treatment – A review. Journal of Electroanalytical Chemistry 754, 46–56.

Sireš, I., Brillas, E., Oturan, M., Rodrigo, M. & Panizza, M. 2014 Electrochemical advanced oxidation processes: today and tomorrow. A review. Environmental Science and Pollution Research International 21 (14), 8356–8367.

Soares, P. Souza, R., Soler, J., Silva, T., Guelli, S., Boaventura, R. & Vilar, V. 2017 Remediation of a synthetic textile wastewater from polyester-cotton dyeing combining biological and photochemical oxidation processes. Separation and Purification Technology 172, 450–462.

Taherkhani, S., Darvishmotavalli, M., Bina, B., Karimyan, K., Fallahi, A. & Karimi, H. 2018 Dataset on photodegradation of tetracycline antibiotic with zinc stannate nanoflower in aqueous solution-application of response surface methodology. Data Brief 19, 1997–2005.

Tavares, M., da Silva, L., Sales Solano, A., Tonholo, J., Martinez-Huitle, C. & Zanta, C. 2012 Electrochemical oxidation of methyl red using Ti/Ru0.3Ti0.7O2 and Ti/Pt anodes. Chemical Engineering Journal 204–206, 141–150.

Ukundimana, Z., Omwene, P., Gengec, E., Can, O. & Kobya, M. 2018 Electrooxidation as post treatment of ultrafiltration effluent in a landfill leachate MBR treatment plant: effects of BDD, Pt and DSA anode types. Electrochimica Acta 286, 252–263.

Verma, A., Dash, R. & Bhunia, P. 2012 A review on chemical coagulation/flocculation technologies for removal of color from textile wastewaters. Journal of Environmental Management 95 (1), 154–168.

Yabalak, E. & Yilmaz, Ö. 2019 Eco-friendly approach to mineralise 2-nitroaniline using subcritical water oxidation method: use of ANN and RSM in the optimisation and modeling of the process. Journal of the Iranian Chemical Society 16, 117–126.

Yagub, M., Sen, T., Afroz, S. & Ang, H. 2014 Dye and its removal from aqueous solution by adsorption: a review. Advances in Colloid and Interface Science 209, 172–184.

Yalvaç, M., Arslan, H., Saleh, M., Gün, M. & Hekim, M. Ş. 2020 Utilizing of bio-adsorbent in zero waste concept: adsorption study of crystal violet onto the centaurea solstitialis and verbascum thapsus plants. Pamukkale University Journal of Engineering Sciences.

Zhang, O., Lin, B., Chen, Y., Gao, B., Fu, L. & Li, B. 2012 Electrochemical and photoelectrochemical characteristics of tinbo5 nanosheet electrode. Electrochimica Acta 81, 74–82.

Zhu, C., Jiang, C., Chen, S., Mei, R., Wang, X., Cao, J., Ma, L., Zhou, B., Wei, Q., Ouyang, G., Yu, Z. & Zhou, K. 2018 Ultrasound enhanced electrochemical oxidation of Alizarin Red S on boron doped diamond(BDD) anode:effect of degradation process parameters. Chemosphere 209, 685–695.

First received 19 March 2021; accepted in revised form 13 June 2021. Available online 23 June 2021.