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Research Article

Comparative Investigation of COD and Color Removal From Real Textile Wastewater by Electro-Fenton and Photo-Electro-Fenton Methods: The Effect of Operational Parameters and Cost Analysis

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With the rapid growth and development of the textile industry in the world, textile wastewater production has also increased in parallel to this. In this study, the effects of pH (2.0–4.5), current density (10–60 A/m^2), and hydrogen peroxide (H_2O_2) concentrations (250–2000 mg/L) on COD and color removal from textile wastewater by electro-Fenton (EF) and photo-electro-Fenton (PEF) methods were comparatively investigated. Under the optimum conditions for the EF method (pH: 2.0, current density: 30 A/m^2, H_2O_2: 1500 mg/L, interelectrode distance: 1.5 cm, stirring speed: 250 rpm, experimental time: 30 min), 84.13% COD and 79.49% color removal were obtained. For the PEF method, under optimum conditions (pH: 2.0, current density: 20 A/m^2, H_2O_2: 1000 mg/L, UV lamp: 16 W, distance between electrodes: 1.5 cm, stirring speed: 250 rpm, experimental time 60 min), 92.06% COD and 84.62% color removal were obtained. The total treatment costs for the EF and PEF methods were calculated as 13.116 \$/m^3 and 9.122 \$/m^3 of wastewater, respectively.

Keywords: COD; color; textile wastewater; treatment

1. Introduction

In parallel with the rapid increase in industrialization in the world, the increasing use of hazardous and toxic chemicals poses a danger to our environment [1]. The textile industry ranks second in terms of water use in the world. In proportion to this, 20% of the wastewater produced in the world originates from the textile industry [2]. The composition of textile wastewater includes a wide range of pollutants such as colorants, dyes, nitrates, toxic metals such as lead, cadmium, and mercury [3], BOD, COD, total dissolved solids [4], total suspended solids [5], a wide range of organic and inorganic pollutants such as sodium hydroxide, sodium sulfate, calcium chloride, nitrite, urea, ethyl alcohol [6], phenol, oil, sulfate, and phosphate [7], volatile compounds, and surfactants [8]. The high pollution concentrations in textile wastewater make it necessary to treat these waters before discharging them into the receiving environment [9]. The mixing of textile wastewater into surface water resources without treatment is very harmful to aquatic organisms, humans, and the ecosystem [10, 11]. The treatment methods of textile wastewater include adsorption [12–14], chemical coagulation [15], membrane filtration [16–18], ion exchange [19], electrochemical processes [20, 21], and advanced oxidation processes (AOPs) [22–24].

The basis of AOPs is the removal of pollutants by oxidation by producing hydroxyl radicals ($OH^{\bullet}$). Due to its high oxidation capacity (2.80 V), $OH^{\bullet}$ is very effective in removing organic pollutants from wastewater [25–27]. AOPs stand out with advantages such as ease of operation, low cost, and environmental friendliness compared to other treatment processes [28]. AOPs include UV-based processes (UV, UV/H_2O_2, UV/TiO_2, UV/H_2O_2/TiO_2), ozone-based systems (O_3, O_3/UV, O_3/H_2O_2), Fenton (Fe^{+2}/H_2O_2), and

photo-Fenton ($Fe^{+2}/H_2O_2/UV$) [29]. Nowadays, Fenton-based AOPs (electro-Fenton [EF], photo-electro-Fenton [PEF]) are effectively used to remove organics from wastewater [30]. The basis of the Fenton processes is the formation of $OH^\bullet$ from H_2O_2 under the catalyst Fe^{+2} [30, 31]. The basic Fenton reactions are given below [32].

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^\bullet + OH^-, \quad (1)$$

$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} + HO_2^\bullet + H^+, \quad (2)$$

$$Fe^{2+} + OH^\bullet \longrightarrow Fe^{3+} + OH^-, \quad (3)$$

$$OH^\bullet + OH^\bullet \longrightarrow H_2O_2, \quad (4)$$

$$H_2O_2 + OH^\bullet \longrightarrow H_2O + HO_2^\bullet, \quad (5)$$

$$OH^\bullet + HO_2^\bullet \longrightarrow H_2O + O_2, \quad (6)$$

The formation of $OH^\bullet$ as a result of the reaction of Fe^{+2} ions with H_2O_2 is the basis of Fenton reactions (equation (1)). In the case of the use of Fe^{+3} ions, $HO_2^\bullet$ is formed, which has a lower oxidation capacity than $OH^\bullet$ (equation (2)). Apart from these two reactions, other reactions can also occur in wastewater or solution (equations (3)–(6)).

Combining electric current with Fenton reagents is called EF. Integrating EF systems with UV light source is called PEF. The basic PEF reactions are given below [32].

$$Fe(OH)^{2+} + h\nu \longrightarrow Fe^{2+} + OH^\bullet \ (\lambda < 580\,nm), \quad (7)$$

$$Fe(RCO_2)^{2+} + h\nu \longrightarrow Fe^{2+} + CO_2 + R^\bullet \ (\lambda < 450\,nm), \quad (8)$$

$$H_2O_2 + h\nu \longrightarrow 2OH^\bullet \ (\lambda < 300\,nm). \quad (9)$$

The integration of EF systems with UV increases the degradation capacity of pollutants [33].

Some of the studies conducted with AOPs from textile wastewater are given below.

Su et al. [34], in their study conducted with the Fenton method on textile wastewater, obtained 86.7% COD and 97% color removal under optimum conditions (pH: 3, $[COD]:[Fe^{2+}]:[H_2O_2]$ = 1:0.95:7.94). Blanco et al. [35], in their study conducted with textile wastewater (pH: 3, H_2O_2: 1650 mg/L, Fe^{+2}: 216 mg/L), obtained 64% TOC and 70% COD removal. Guimarães et al. [36], in their study conducted with the UV/H_2O_2 process with Reactive Blue 19 textile dye, obtained 91% color removal (H_2O_2: 500 mg/L, experimental time: 3 h). Eslami et al. [37], in their study from textile wastewater using the EF method (pH: 3, H_2O_2: 1978 mg/L, experimental time: 60 min), achieved 70.6% COD removal. Patil and Raut [38], in their treatment study from textile wastewater using the Fenton process, obtained 98% color removal under optimum conditions (pH: 3, H_2O_2: 0.1 mL/L, experimental time: 30 min).

Blanco et al. [39], in their pollutant removal study from textile wastewater using the photo-Fenton method (pH: 2.7, Fe^{+2}: 216 mg/L, H_2O_2: 4950 mg/L, T: 35°C), achieved 79% COD and 75% TOC removal. Torrades and García-Montaño [40], in their study using Fenton and photo-Fenton methods from textile wastewater, achieved 62.9% and 76.3% COD removal under optimum conditions (pH: 3, $[Fe^{+2}]$: 1.79 mM, $[H_2O_2]$: 73.5 mM, experimental time: 120 min), respectively. Cetinkaya et al. [41], in their study conducted with the classical Fenton method from textile wastewater, achieved 95% color removal (pH: 3, Fe^{+2}: 0.10 g/L, H_2O_2: 2.20 g/L, experimental time: 90 min). Ilhan et al. [42], in their study conducted with the Fenton process from textile wastewater, achieved 98.6% color, 84.6% COD, and 78.6% TOC removal (pH: 3.2, Fe^{+2}: 1093 mg/L, H_2O_2: 1600 mg/L, experimental time: 150 min). Kaur et al. [43], in their study using the EF method ($[FeSO_4]$: 0.53 mM, current: 0.32 A, experimental time: 90 min), from textile wastewater, achieved 90.30% color and 100% COD removal. Trigueros et al. [44], in their study using the photo-Fenton method from textile wastewater, achieved 99% color and 93% COD removal under optimum conditions (pH: 2.8, Fe^{+3}: 170 mg/L, H_2O_2: 8000 mg/L, experimental time: 240 min, mixing speed: 150 rpm), respectively. Beyazıt and Atmaca [45], in their study conducted with the PEF method from leachate, achieved maximum 84.46% color and 83.84% COD removal efficiencies under optimum conditions (current density: 10 A/m^2, pH: 3, H_2O_2: 1000 mg/L, stirring speed: 250 rpm, UV lamp: 16 W, distance between electrodes: 1.0 cm).

In this study, the effects of some important operating parameters (pH, current density, H_2O_2 concentration) on COD and color removal from textile wastewater with EF and PEF methods were analyzed comparatively. In addition, optimum experimental conditions were determined, and detailed cost analyses were made for electrical energy consumption and per m^3 of wastewater. Additionally, a kinetic study was conducted.

2. Materials and Methods

2.1. Wastewater and Properties. The textile wastewater used in the experiments was supplied by a company operating in the Organized Industrial Zone of Fatsa district of Ordu in the field of printing textile products. The visual of the textile wastewater is given in Figure 1, and the properties of the wastewater are given in Table 1.

2.2. Experimental Setup. All experiments were carried out in a cylindrical Plexiglas reactor. The properties of the reactor and electrodes used in the experiments are given in Table 2.

Electrodes consisting of one anode and one cathode were placed inside the reactor. For PEF experiments, a 16 W low-pressure mercury vapor lamp (UV-C lamp) was also placed inside the reactor. The wavelength of the UV-C lamp is 254.3 nM. The electrodes inside the reactor were positioned in monopolar parallel mode and connected to a voltage meter (GW GPC3060D DC 30 V, 6 A) at the beginning of the experiments. It has been determined that the photoreactor with its original design features operates with high performance and efficiency that will not hinder the use of the PEF process. Another advantage of the photoreactor is that it can

Figure 1: Image of textile wastewater.

Table 1: Wastewater properties.

Parameter	Value
pH	6.16
Conductivity (μS/cm)	6145
Color (Pt-Co)	975
COD (mg/L)	2520
Phenol (mg/L)	< 0.1
Total chromium (mg/L)	< 0.01

Table 2: Reactor and electrode properties.

Reactor size (inner diameter × outer diameter × height) (cm)	7.70 × 8.25 × 48.50
Useful volume (dm^3)	2.258
Anode: iron, cathode: steel	1 pcs, 1 pcs
Electrode dimensions (width × height × thickness) (cm)	4.6 × 29.6 × 0.2
Active anode surface area (cm^2)	267.498

work in high volumes and provide a homogeneous mixture of wastewater from the center of the reactor. The top view of the reactor is shown in Figure 2, and the experimental setup is shown in Figure 3.

2.3. Experimental Procedure and Analytical System. To remove suspended solids that may adversely affect the AOPs, the wastewater was subjected to a prefiltration process with a 0.063-mm-diameter sieve [46]. For all experiments, 2.2 L of wastewater was first added to the reactor and mixed at 250 rpm with a mechanical stirrer in batch mode. While Fe^{+2} ions were produced at the anode with the help of an electric current, H_2O_2 was added to the system externally. Against possible oxidation on the electrode surfaces, the electrodes were kept in concentrated HCl after all experiments and then rinsed with pure water. H_2SO_4 (Sigma-Aldrich) and NaOH (Merck) were used to adjust the pH of the wastewater.

The pH measurements of the wastewater were carried out with a pH meter of the OHAUS300 Starter brand and model. Then, the reactor was filled with wastewater, and after mixing for a few minutes, the desired amount of H_2O_2 was added, and the voltage meter and UV lamp were operated simultaneously.

Samples were taken at periodic intervals (2, 4, 6, 8, 10, 15, 20, 25, 30, 35, 40, 45, 60, 90, 120, 150 min) and centrifuged at 9000 rpm for 10 min with a NÜVE NF 800R brand and model centrifuge device, and then color and COD measurements were made using a spectrophotometer (Merck Spectroquant Nova 60A brand and model). All EF and PEF experiments were carried out according to standard methods prepared for water and wastewater treatment. COD analyses in wastewater samples were performed according to the closed reflux colorimetric method (Standard Methods, 5220 D) [47]. COD removal efficiencies were calculated with the help of equation (10).

$$\text{Percentage of removal efficiency}\,(\%) = \frac{C_o - C_t}{C_o} \times 100, \tag{10}$$

C_o = initial COD concentration ($mg.L^{-1}$), C_t = COD concentration at time t ($mg.L^{-1}$).

Color measurements were obtained with Pt-Co units using Standard Methods for the Examination of Water and Wastewater (2120 B) [47]. In this direction, the absorbance value of the samples taken was measured at a wavelength of 340 nm and then multiplied by the necessary multiplication factor to determine the color values.

3. Results and Discussion

3.1. Effect of pH on COD and Color Removal. The pH value of wastewater is one of the main factors affecting the treatment efficiency in Fenton systems. Typically, acidic pH values (2–4) are preferred for pollutant removal with Fenton systems [25]. In pollutant removal studies with Fenton systems, the optimum pH value has been reported to be around 2–3 [48–51]. Iron hydroxide complexes are formed at alkaline pH values, which reduces the oxidation capacity. To investigate the effect of pH on the EF process, other operating parameters (current density: 30 A/m^2, H_2O_2: 1000 mg/L, stirring speed: 250 rpm, distance between electrodes: 1.5 cm) were kept constant and the effect of different pH values (2–4.5) was investigated. For pH 2, 2.5, 3.0, 3.5, 4.0, 4.5 values, COD removal efficiencies were found as 80.16%, 78.57%, 76.59%, 71.83%, 62.09%, and 43.25%, respectively, while color removal efficiencies were found as 66.67%, 63.59%, 60.51%, 55.38%, 50.26%, and 40.00%, respectively. At the end of the 30th minute, when maximum pollutant removal was achieved, the consumed electrical energy amounts were calculated as 0.147, 0.166, 0.184, 0.239, 0.239, and 0.276 kWh/m^3, respectively. The obtained results are consistent with studies reporting that the optimum pH value is 2 [52–56]. The pH effect on COD and color removal with the EF method is shown in Figures 4(a) and 4(b), respectively. When Figures 4(a) and

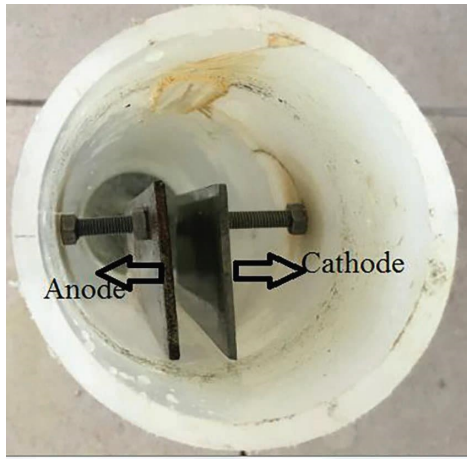

Figure 2: Top view of the reactor.

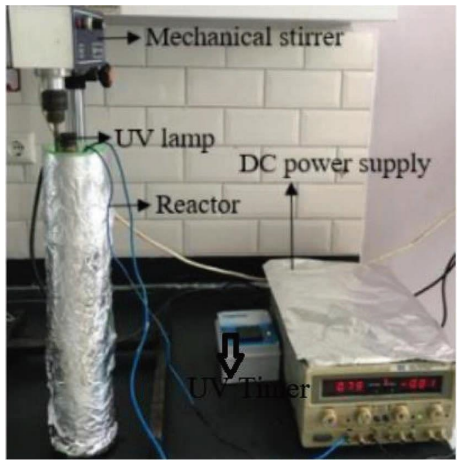

Figure 3: Experimental setup.

4(b) are examined together, it is seen that COD and color removal were not observed since sufficient $OH^{\bullet}$ was not formed in the environment during the first 4 min. There was a noticeable increase in COD and color removal efficiencies by the sixth minute. This situation can be associated with the increasing $OH^{\bullet}$ in the environment. By increasing the pH from 2 to 4.5, COD removal decreased from 80.16% to 43.25%, and color removal decreased from 66.67% to 40.00% after 30 min. The reason why COD removal decreased after 30 min is the slow dissolution of organic matter coagulated with $Fe(OH)_n$ flocs. Similar results were also reported in a previous work by Altin and Lin and Chang [57, 58]. Altin [57], in his study on COD and color removal from leachate using the PEF method, observed that COD and color removal decreased slightly after 20 min. David et al. [59], in their color removal study with the EF method from distillery wastewater, reported that the color removal efficiency decreased from 43% to 38% by increasing the pH from 3 to 5. Gümüş and Akbal [60] reported in their study on COD removal from synthetic phenolic wastewater using the EF method that COD removal decreased from 87.50% to 62.50% as the pH was increased from 3 to 7.

To investigate the effect of pH on COD and color removal by PEF process, other operating conditions were kept constant (current density: 10 A/m^2, H_2O_2: 1000 mg/L, UV lamp: 16 W, stirring speed: 250 rpm, distance between electrodes: 1.5 cm) and pH (2–4.5) values were tested. For pH 2, 2.5, 3.0, 3.5, 4.0, and 4.5 values, COD removal efficiencies were found as 80.16%, 78.57%, 76.59%, 71.83%, 62.09%, and 43.25%, respectively, while color removal efficiencies were found as 66.67%, 63.59%, 60.51%, 55.38%, 50.26%, and 40.00%, respectively. The electrical energy consumption in this minute was calculated as 7.322, 7.331, 7.344, 7.356, 7.380, and 7.405 kWh/m^3, respectively. Figures 5(a) and 5(b) show the pH effect on COD and color removal with the PEF method. The results are consistent with previous studies. Beyazıt and Atmaca [45], in their study from leachate with the PEF method, found that by increasing the pH from 3 to 5, COD removal decreased from 79.75% to 51.13%, and color removal decreased from 80.57% to 65.40%. Beyazıt and Atmaca [61], in their study conducted with the PEF method on MDF wastewater, reported that COD removal decreased from 98.13% to 58.41% and color removal decreased from 78.50% to 51.65% by increasing the pH from 3 to 5. For PEF experiments, the pH 2 value, at which the maximum removal efficiency was achieved, was found to be optimum. The pH 2 value was taken as the basis for optimizing the subsequent operating parameters.

3.2. Effect of Current Density on COD and Color Removal. Current density is one of the important operating parameters affecting the performance of Fenton systems. As the current density value increases, the amount of $OH^{\bullet}$ formed in the environment increases. In addition, current density is a parameter that needs to be optimized since it affects the amount of electrical energy consumed [62]. To investigate the effect of current density on COD and color removal, other operating conditions (pH: 2, H_2O_2: 1000 mg/L, stirring speed: 250 rpm, distance between electrodes: 1.5 cm) were kept constant and 10, 20, 30, 40, 50, and 60 A/m^2 values were tested. At the end of the 30th minute, the maximum COD removal efficiencies for 10, 20, 30, 40, 50, and 60 A/m^2 values were found to be 41.47%, 54.37%, 80.16%, 77.38%, 75.40%, and 71.43%, respectively, while the color removal efficiencies were found to be 35.38%, 40.51%, 66.67%, 61.54%, 56.41%, and 48.72%, respectively. The amount of electrical energy consumed at the end of the 30th minute was calculated as 0.018, 0.074, 0.147, 0.344, 0.583, and 0.884 kWh/m^3 for 10, 20, 30, 40, 50, and 60 A/m^2 current density values, respectively. By increasing the current density from 10 A/m^2 to 30 A/m^2, COD removal increased from 41.47% to 80.16% and color removal increased from 35.38% to 66.67%. This is related to the increase in the concentration of $OH^{\bullet}$ formed in the medium and the increased amount of iron ions in the medium (equation (11)) [25].

$$Fe^{3+} + e^{-} \longrightarrow Fe^{2+}. \quad (11)$$

The experimental results obtained are consistent with the literature. Kuleyin et al. [25], in their pollutant removal study from textile wastewater with the EF process, found that

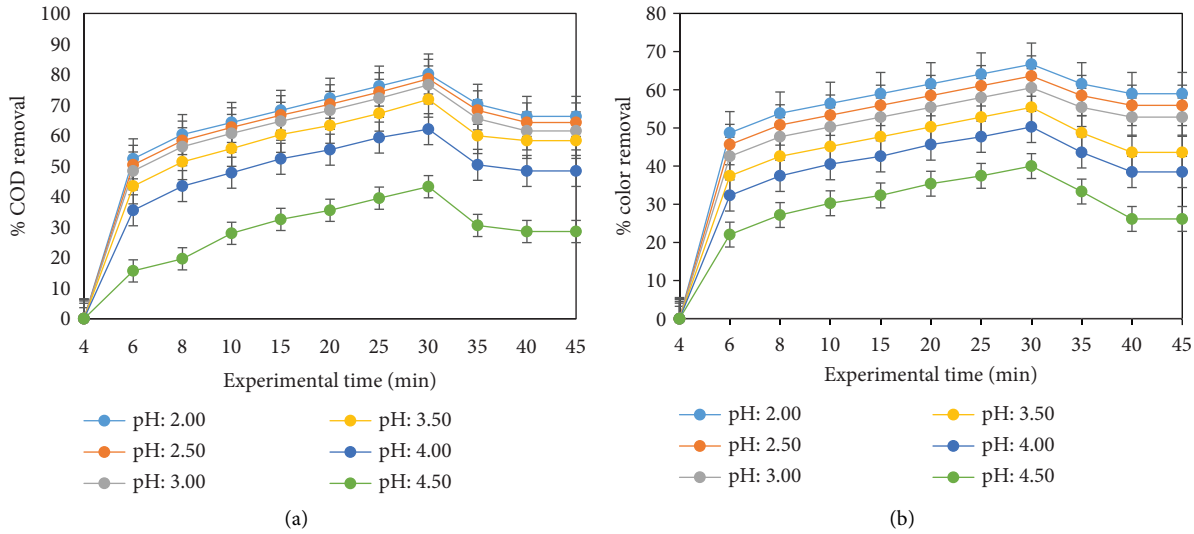

FIGURE 4: (a) The effect of pH on COD removal by the EF method (current density: 30 A/m^2, H_2O_2: 1000 mg/L, mixing speed: 250 rpm, interelectrode distance: 1.5 cm). (b) The effect of pH on color removal by the EF method (current density: 30 A/m^2, H_2O_2: 1000 mg/L, mixing speed: 250 rpm, interelectrode distance: 1.5 cm).

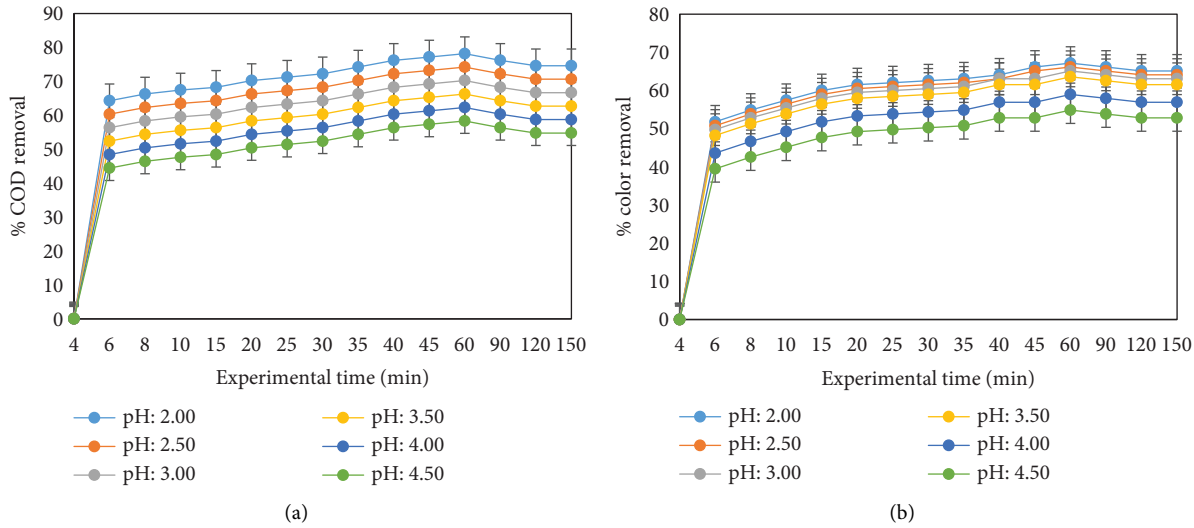

FIGURE 5: (a) The effect of pH on COD removal by the PEF method (current density: 10 A/m^2, H_2O_2: 1000 mg/L, UV lamp: 16 W, mixing speed: 250 rpm, interelectrode distance: 1.5 cm). (b) The effect of pH on color removal by the PEF method (current density: 10 A/m^2, H_2O_2: 1000 mg/L, UV lamp: 16 W, mixing speed: 250 rpm, interelectrode distance: 1.5 cm).

by increasing the current density value from 6.5 mA/cm^2 to 10 mA/cm^2, color removal increased from 88% to 93% and COD removal increased from 78% to 90%. Gümüş and Akbal [60], in their study conducted on phenolic wastewater using the EF method, found that COD removal increased from 87.50% to 96.88% by increasing the current density value from 1 mA/cm^2 to 5 mA/cm^2. By increasing the current density value from 30 A/m^2 to 60 A/m^2, COD removal decreased from 80.16%, 71.43% and color removal decreased from 66.67% to 48.72%. Atmaca and Beyazıt [63], in their COD removal study from leachate using the EF method, found that by increasing the current density value from 100 A/m^2 to 125 A/m^2, the COD removal efficiency decreased from 68% to 46%. This situation can be explained by the side reactions of $OH^\bullet$ according to equation (12) [62].

$$2OH^\bullet \longrightarrow O_2 + 2H^+ + 2e^-. \quad (12)$$

Figures 6(a) and 6(b) show the effect of current density on COD and color removal, respectively.

To investigate the effect of current density on COD and color removal by the PEF process, current density values of 10, 20, 30, 40, 50, and 60 A/m^2 were tried. The current density value of 20 A/m^2 at which maximum COD and color removal efficiencies were obtained was found to be optimum. At the end of 60 min, COD removal efficiencies for 10, 20, 30, 40, 50, and 60 A/m^2 values were found as 78.17%,

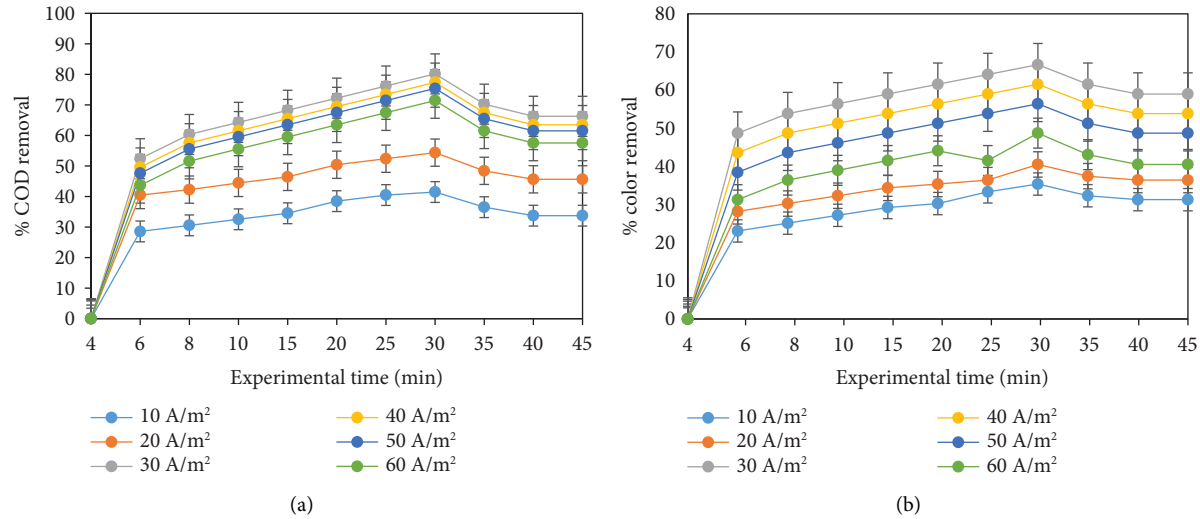

FIGURE 6: (a) The effect of current density on COD removal by the EF method (pH: 2, H_2O_2: 1000 mg/L, mixing speed: 250 rpm, interelectrode distance: 1.5 cm). (b) The effect of current density on color removal by the EF method (pH: 2, H_2O_2: 1000 mg/L, mixing speed: 250 rpm, interelectrode distance: 1.5 cm).

92.06%, 88.10%, 84.13%, 80.16%, and 76.19%, respectively, while color removal efficiencies were found as 67.18%, 84.62%, 82.56%, 80.51%, 78.46%, and 76.41%, respectively. At the end of the 60th minute, when maximum COD and color removal were obtained, the consumed electrical energy amounts were calculated as 7.319, 7.467, 7.601, 8.006, 8.559, and 9.037 kWh/m^3 for 10, 20, 30, 40, 50, and 60 A/m^2 values, respectively. By increasing the current density from 10 A/m^2 to 20 A/m^2, the COD removal efficiency increased from 78.17% to 92.06% and color removal increased from 67.18% to 84.62%. With increasing the current density value above 20 A/m^2, decreases were observed in the removal efficiencies. The obtained results are compatible with the literature. Crispim et al. [64], in their study on COD removal from leachate using the PEF method, found that by increasing the current density from 30 mA/cm^2 to 90 mA/cm^2, COD removal increased from 46% to 89% in 4 h of treatment. Beyazıt and Atmaca [61], in their study on COD removal from MDF wastewater using the PEF method, found that by increasing the current density from 10 A/m^2 to 50 A/m^2, COD removal decreased from 98.10% to 46.56% in 60 min of treatment. Figures 7(a) and 7(b) show the effects of current density on COD and color removal with the PEF process, respectively.

3.3. Effect of H_2O_2 Concentration on COD and Color Removal. Since the H_2O_2 concentration is proportional to the $OH^\bullet$ concentration produced in the medium, it is very important for the efficiency of Fenton processes [65]. H_2O_2 is the most important source of $OH^\bullet$ produced in Fenton processes. While low H_2O_2 concentration reduces pollutant removal efficiency, higher than optimum value reduces efficiency due to the scavenging effect. H_2O_2 constitutes the most significant item in operating expenses, and as H_2O_2 concentration increases, so does the total treatment cost. Therefore, H_2O_2 optimization is extremely important for both pollutant removal and cost. The effects of different H_2O_2 concentrations (250–2000 mg/L) on COD and color removal were investigated in EF and PEF experiments. For EF experiments, the maximum COD and color removal (84.13%, 79.49%) was achieved at the end of 30 min with an electrical energy consumption of 0.147 kWh/m^3 for a 1500 mg/L H_2O_2 value. While the COD removal efficiencies obtained for 250, 500, 750, 1000, 1250, 1500, 1750, and 2000 mg/L H_2O_2 values were found as 23.41%, 36.51%, 60.32%, 80.16%, 82.14%, 84.13%, 81.35%, and 80.75%, respectively, the color removal efficiencies were found as 17.95%, 28.21%, 43.59%, 66.67%, 69.23%, 79.49%, 68.72%, and 67.69%. By increasing the H_2O_2 concentration from 250 mg/L to 1500 mg/L, COD and color removals increased from 23.41% and 17.95% to 84.13% and 79.49%, respectively. This situation can be associated with the increased $OH^\bullet$ in the environment. By increasing the H_2O_2 concentration from 1500 mg/L to 2000 mg/L, COD removal decreased from 84.13% to 80.75% and color removal decreased from 79.49% to 67.69%. This situation is due to the excessive amount of H_2O_2 scavenging the $OH^\bullet$ formed according to equations (13) and (14) [62].

$$OH^\bullet + H_2O_2 \longrightarrow HO_2^\bullet + H_2O, \quad (13)$$

$$HO_2^\bullet + OH^\bullet \longrightarrow H_2O + O_2. \quad (14)$$

Similar results were found by Gümüş and Akbal [60] in their study on phenol and COD removal from synthetic phenolic wastewater using the EF method. They found that by increasing the H_2O_2 concentration from 250 mg/L to 1000 mg/L, phenol removal increased from 72.62% to 97.76%, while COD removal increased from 59.38% to 93.75%.

The effects of H_2O_2 concentrations on COD and color removal are shown in Figures 8(a) and 8(b), respectively.

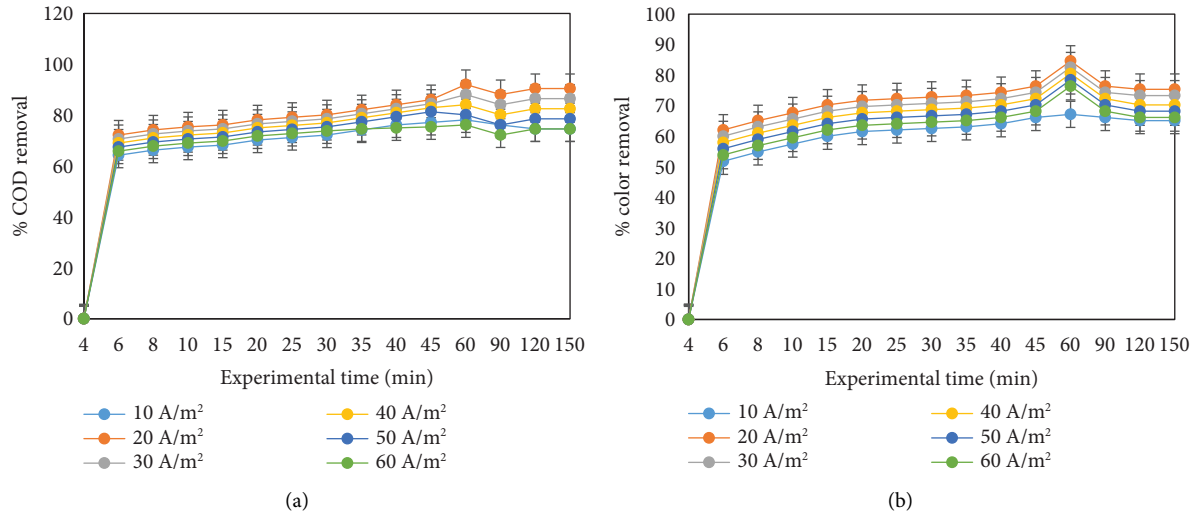

FIGURE 7: (a) The effect of current density on COD removal by the PEF method (pH: 2, H_2O_2: 1000 mg/L, UV lamp: 16 W, mixing speed: 250 rpm, interelectrode distance: 1.5 cm). (b) The effect of current density on color removal by the PEF method (pH: 2, H_2O_2: 1000 mg/L, UV lamp: 16 W, mixing speed: 250 rpm, interelectrode distance: 1.5 cm).

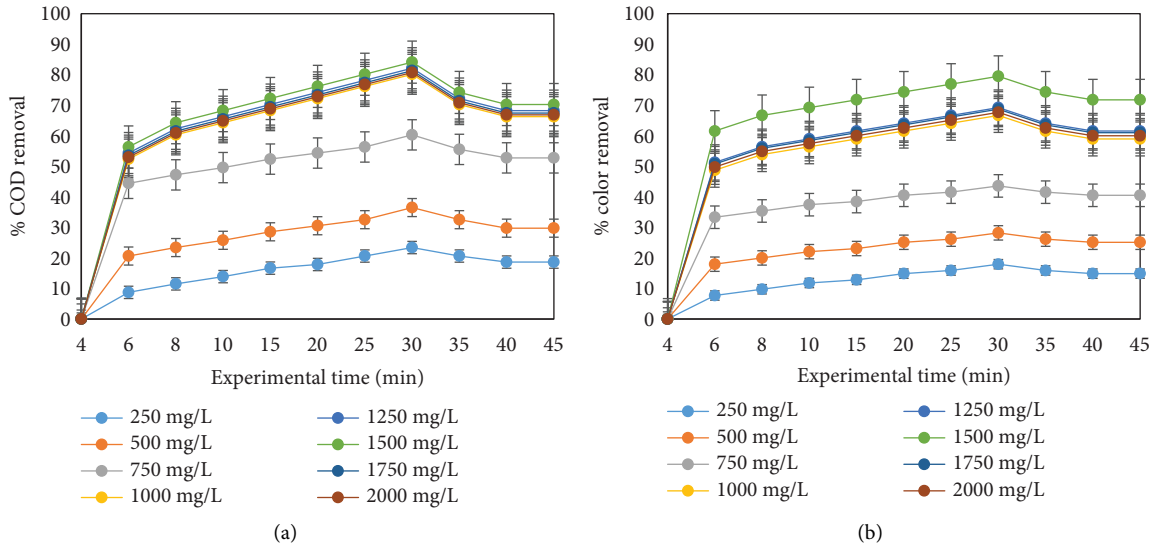

FIGURE 8: (a) The effect of H_2O_2 concentration on COD removal by the EF method (pH: 2, current density: 30 A/m^2, mixing speed: 250 rpm, interelectrode distance: 1.5 cm). (b) The effect of H_2O_2 concentration on color removal by the EF method (pH: 2, current density: 30 A/m^2, mixing speed: 250 rpm, interelectrode distance: 1.5 cm).

For PEF experiments, maximum COD and color removal were achieved at the end of 60 min with an electrical energy consumption of 7.490 kWh/m^3 and a concentration of 1000 mg/L H_2O_2. While the COD removal efficiencies obtained for 250, 500, 750, 1000, 1250, 1500, 1750, and 2000 mg/L H_2O_2 values were found to be 36.51%, 56.35%, 76.19%, 92.06%, 88.10%, 84.13%, 80.16%, and 78.17%, respectively, the color removal efficiencies were found to be 31.28%, 46.67%, 62.05%, 84.62%, 79.49%, 76.41%, 73.33%, and 69.23%, respectively. The effects of different H_2O_2 concentrations on COD and color removal are shown in Figures 9(a) and 9(b), respectively.

Similar results were found by Asaithambi et al. [66] in their study on COD removal from leachate using the PEF method. They found that by increasing the hydrogen peroxide concentration from 75 mg/L to 300 mg/L, COD removal increased from 58.50% to 97%. They reported that by increasing the hydrogen peroxide concentration from 300 mg/L to 450 mg/L, COD removal decreased from 97% to 80.50%.

3.4. Cost Calculation. The electrical energy and iron amounts consumed in the Fenton processes were calculated according to equations (15) and (16), respectively [67, 68]. In

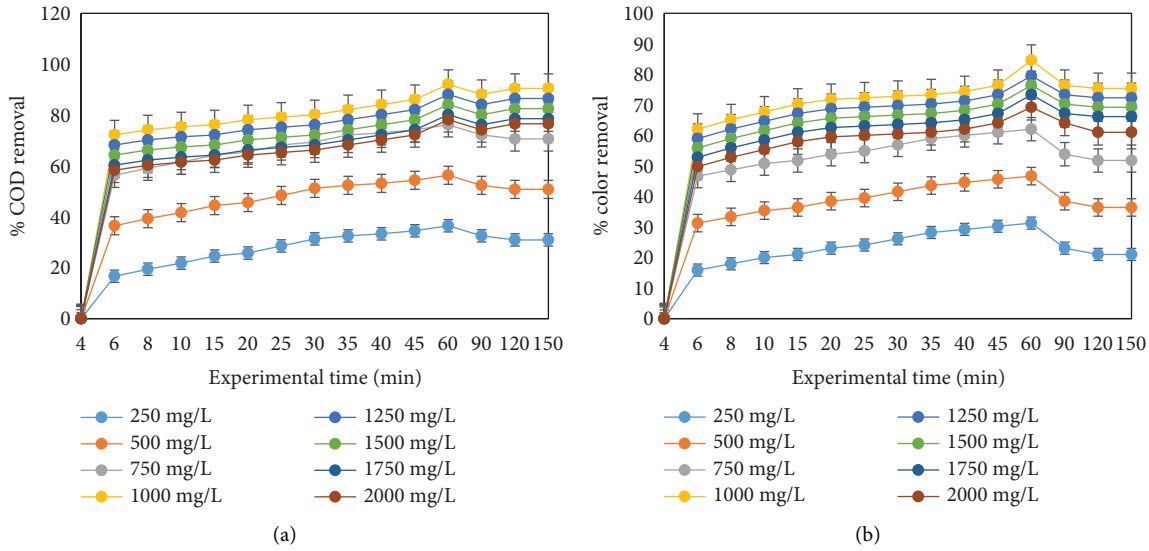

FIGURE 9: (a) The effect of H_2O_2 concentration on COD removal by the PEF method (pH: 2, current density: 20 A/m^2, UV lamp: 16 W, mixing speed: 250 rpm, interelectrode distance: 1.5 cm). (b) The effect of H_2O_2 on color removal by the PEF method (pH: 2, current density: 20 A/m^2, UV lamp: 16 W, mixing speed: 250 rpm, interelectrode distance: 1.5 cm).

addition, the electrical energy consumption from the UV lamp was calculated for the PEF process according to equation (17).

$$\text{EEC} = \frac{\text{Voltage} \times I \times t}{V}, \tag{15}$$

where EEC (kWh/m^3) represents the electrical energy consumption, I (A) represents the supplied current, t (sn) represents the experimental period, and V (L) represents the wastewater volume.

$$\Delta M = \frac{M \times I \times t}{z \times F \times V}, \tag{16}$$

where ΔM (g/m^3) represents the theoretical amount of iron consumed, M (g/mol) represents the molecular weight of iron, z represents the number of valence electrons, and F represents the Faraday constant.

$$\text{UV Lamp electrical energy consumption}\left(\text{kWh/m}^3\right) = \frac{\text{Power}\,(W) \times \text{Time}\,(\text{hour})}{\text{Wastewater} \times \text{volume}\left(\text{m}^3\right)}. \tag{17}$$

Tables 3 and 4 are not cited because they are cost calculations made by me.

Examples from different literature studies are given below:

Dobrosz-Gómez et al. [52], in their Fenton advanced oxidation study with textile wastewater contaminated with acid-black 194 dye, found the total operating cost to be 10.55$/m^3.

Guvenc et al. [69], in their study on COD removal from real chemical industry wastewater using the EF process, found the electrical energy consumption per kilogram of COD to be 54.5 kWh.

Çalık and Çiftçi [70] reported that in their treatment study of textile wastewater with Fenton and photo-Fenton processes, the total treatment cost varied between 9.56 and 16.88 Euro/m^3 for the Fenton process and between 13.46 and 20.13 Euro/m^3 for the photo-Fenton process.

Pillai and Gupta [21] found the total treatment cost to be 10.2$/m^3 in their study of batch electrochemical oxidation of textile wastewater.

Boyjoo et al. [71] found the treatment cost as 49.4$/m^3 as a result of photocatalytic treatment of shower water using a pilot-scale reactor.

In this study, the total treatment costs for the EF and PEF methods were calculated as 13.116 $/m^3 and 9.122 $/m^3 of wastewater, respectively. The amortization period is calculated as follows: The weight loss in the anode should not exceed 10% [72].

According to equation (16), electrodes need to be replaced every 19 days for the EF process and every 37 days for the PEF process.

More importantly, there is need for pre-pilot, pilot, and scale-up studies for EF and PEF system. Till date, all the studies reported in the literature was performed on

Table 3: Cost calculation for the EF process.

	Electrical energy cost ($/m^3)	Fe^{+2} cost ($/m^3)	H_2O_2 cost ($/m^3)	Total treatment cost ($/m^3 wastewater)
Initial pH				
2.0	0.006	0.110	8.66	8.776
2.5	0.007			8.777
3.0	0.007			8.777
3.5	0.010			8.780
4.0	0.010			8.780
4.5	0.012			8.782
Current density (A/m^2)				
10	0.0006	0.037	8.66	8.698
20	0.003	0.073		8.736
30	0.006	0.110		8.776
40	0.015	0.146		8.821
50	0.025	0.183		8.868
60	0.037	0.219		8.916
H_2O_2 (mg/L)				
250	0.006	0.110	2.171	2.287
500			4.342	4.458
750			6.486	6.602
1000			8.66	8.776
1250			10.83	10.946
1500			13.00	13.116
1750			15.17	15.286
2000			17.31	17.426

Note: Unit price of electrical energy: 0.042 $/kWh. Unit price of Fe^{2+}: 0.571 $/kg. Unit price of H_2O_2: 3.43 $/lt.

Table 4: Cost calculation for the PEF process.

	Electrical energy cost ($/m^3)	Fe^{+2} cost ($/m^3)	H_2O_2 cost ($/m^3)	Total treatment cost ($/m^3 wastewater)
Initial pH				
2.0	0.310	0.073	8.66	9.043
2.5	0.310			9.043
3.0	0.311			9.044
3.5	0.311			9.044
4.0	0.312			9.045
4.5	0.313			9.046
Current density (A/m^2)				
10	0.310	0.073	8.66	9.043
20	0.316	0.146		9.122
30	0.321	0.219		9.200
40	0.339	0.293		9.292
50	0.362	0.366		9.388
60	0.382	0.439		9.481
H_2O_2 (mg/L)				
250	0.316	0.146	2.171	2.633
500			4.342	4.804
750			6.486	6.948
1000			8.660	9.122
1250			10.83	11.292
1500			13.00	13.462
1750			15.17	15.632
2000			17.31	17.772

Note: Unit price of electrical energy: 0.042 $/kWh. Unit price of Fe^{2+}: 0.571 $/kg. Unit price of H_2O_2: 3.43 $/lt.

laboratory scale; as such, full-scale studies are required to understand the behavior of EF system on large-scale treatment reactors as well as optimization works with full cost and environmental impact. Major attention should be devoted in the future to the identification of reaction intermediates, development of rate expressions based on established reaction mechanisms, identification of scale-up parameters, and criteria for cost-effectiveness.

Table 5: First- and second-order reaction kinetics constants and correlation coefficients of COD and color.

Treatment processes	COD removal first-order kinetics k (min^{-1}) (r^2)	COD removal second-order kinetics k (min^{-1}) (r^2)	Color removal first-order kinetics k (min^{-1}) (r^2)	Color removal second order kinetics k (min^{-1}) (r^2)
EF	0.1606 (0.9913)	0.0002 (0.949)	0.1001 (0.9952)	0.0004 (0.9859)
PEF	0.0975 (0.8161)	0.0002 (0.6484)	0.0639 (0.8019)	0.0003 (0.6859)

Hence, it is advisable to use EF and PEF methods as pretreatment to reduce the toxicity to a certain level, beyond which biological treatment can be employed.

3.5. Economic Evaluation. The operating costs of the EF and PEF processes were found by calculating the values of Fenton reagents (Fe^{+2}, H_2O_2) and electrical energy consumption. The Fe^{+2} cost was assumed to be \$0.571/kg, H_2O_2 cost was \$3.43/L, and electrical energy consumption cost was \$0.042/kWh. The total treatment cost for the optimum conditions of the EF process was calculated as \$13.116/m^3, while the total treatment cost for the optimum conditions of the PEF process was calculated as \$9.122/m^3. The most important cost component is H_2O_2. The significant reduction in H_2O_2 consumption in the PEF process significantly reduced the total treatment cost. In addition, the treatment cost per kg of COD was found to be \$6.558/kg COD for the EF process and \$3.93/kg COD for the PEF process.

3.6. Kinetic Study. First- and second-order reaction kinetics were used to study COD and color removal kinetics for EF and PEF methods.

$$\text{First} - \text{order reaction kinetics: } \frac{dC}{dt} = -k_1 C,$$
$$\text{Second} - \text{order reaction kinetics: } \frac{dC}{dt} = -k_2 C^2, \tag{18}$$

where C is the concentrations of COD and color, k_1 and k_2 represent the apparent kinetic rate constants of first- and second-order reaction kinetics, respectively, and t is the experimental time.

The following kinetic equations are obtained by integrating equations (19) and (20).

$$C_t = C_o e_{1^t}^{-k}, \tag{19}$$

$$\frac{1}{C_t} = \frac{1}{C_0} + k_2 t, \tag{20}$$

where C_t is the concentration of COD and color at reaction time t.

Depending on the first- and second-order reaction kinetics for COD and color removal in EF and PEF processes, k_1, k_2 values were calculated, regression analyses (r^2) were performed, and the results are summarized in Table 5.

When the r^2 values obtained using optimum experimental conditions were compared, it was found that the r^2 values based on first-order reaction kinetics for COD removal for EF and PEF processes were 0.9913 and 0.8161, respectively, and for color removal, they were 0.9952 and 0.8019, respectively. These results demonstrate that COD and color removal from textile wastewater by the EF and PEF processes conform to first-order kinetics.

4. Conclusion

- In this study, the treatability of textile wastewater with EF and PEF processes were investigated. In all experiments, current density, initial pH, and H_2O_2 concentration values were optimized. To optimize the pH value, experiments were conducted in the range of (2–4.5) and the optimum pH value was found to be 2.0. It was determined that COD and color removal efficiencies decreased at pH values higher than 2.
- It was found that the current density value is an important parameter in pollutant removal efficiency. For the EF process, it was found that COD and color removal increased by increasing the current density value up to 30 A/m^2, and decreased at higher current density values. The increase in current density generally increased COD and color removal efficiencies. On the other hand, increasing the current density increased the amount of electrical energy consumed. The optimum current density value for the PEF process was found to be 20 A/m^2.
- The results showed that H_2O_2 concentration has a significant effect on the performance of EF and PEF systems. In addition, the H_2O_2 value is the most important parameter among the cost components. A 1500 mg/L H_2O_2 value was found to be optimum for the EF process, and 1000 mg/L H_2O_2 value for the PEF process. The significant reduction of H_2O_2 consumed in the PEF process is one of the important advantages of the method.
- For the EF process, a maximum of 84.13% COD and 79.49% color removal was achieved after 30 min of treatment, while for the PEF process, a maximum of 92.06% COD and 84.62% color removal was achieved after 60 min of treatment. When the EF and PEF processes were compared, it was determined that the PEF process was more effective in terms of both pollutant removal and cost. As a result of the treatment carried out with the PEF process, the discharge standards (COD: 300 mg/L, color: 280 mg/L) in the water quality control regulation were met. It should also be noted that as a result of the treatment, the biodegradability (BOD/COD) rate of the wastewater increases significantly, and this constitutes a significant

advantage for biological treatment. It was concluded that the optimized PEF process would be useful for large-scale applications in the treatment of textile industry wastewater.

Data Availability Statement

Data are available upon request from the author.

Conflicts of Interest

The author declares no conflicts of interest.

Author Contributions

Kasım Atmaca: writing–original draft, investigation, visualization, methodology, data analysis, and editing.

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