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The effect of cathode material, current density and anode–cathode gap on the COD and color removal from leachate by the electro-Fenton method

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ABSTRACT

In this study, the effects of different cathode electrodes (stainless-steel, graphite, carbon cloth), current density (25–125 A/m^2), the distance between electrodes (0.5–1.5 cm) on chemical oxygen demand (COD) and color removal from leachate by the electro-Fenton process were investigated. Under the optimum experimental conditions (pH: 3, current density: 100 A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, the distance between electrodes: 1.5 cm) a maximum COD removal of 82.5% was achieved with an electrical energy consumption of 0.75 kWh/m^3. The optimum conditions for color removal were pH: 3, current density: 75A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, the distance between electrodes 1.5 cm. Under these conditions, a maximum color removal of 69% from leachate was achieved with an electrical energy consumption of 0.108 kWh/m^3. According to the Water Pollution and Control Regulation in force in our country, it is understood that 700 mg/L and 290 Pt-Co values cannot be provided for COD and color parameters, respectively. This result concluded that this advanced oxidation process alone was not sufficient for the landfill leachate used in this study.

Keywords: Chemical oxygen demand (COD); Color; Electro-Fenton; Leachate; Removal

1. Introduction

Leachate is a liquid mixture consisting of highly organic and inorganic pollutants formed as a result of rainwater leaching from solid wastes, natural moisture, and biochemical reactions occurring in wastes. If leachates are safely collected and not discharged; it can be a potential source of pollution that threatens soil, surface water, and groundwater [1,2]. Leachate treatment technologies include biological and physicochemical methods or integrated systems in which these methods are used together [3].

Although biological treatment systems provide high biochemical oxygen demand removal efficiency, they are often inadequate to break down high molecular weight fractions and color removal. Physicochemical treatment processes (coagulation–flocculation, chemical oxidation, air stripping, membrane processes, and adsorption on activated carbon) are also used separately or in combination with biological treatment processes. However, these techniques have disadvantages such as high operating costs and low pollutant efficiency [4]. Advanced oxidation processes such as electrochemical treatment are among the most effective purification technologies for the removal of resistant organic pollutants and color [5]. The electro-Fenton process, which is one of the advanced oxidation processes, is expressed by the reaction of Fe^{2+} ion with hydrogen peroxide under acidic conditions.

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As given by Eq. (1), the iron ion initiates the decomposition of H_2O_2, catalyzes, and thus forms hydroxyl radicals ($OH^•$) [6].

$$Fe^{+2} + H_2O_2 \rightarrow Fe^{+3} + OH^• + OH^- \ (k = 76\ M^{-1}\ s^{-1}) \quad (1)$$

Hydroxyl radicals allow many organic and inorganic pollutants to be converted to much less toxicological or completely harmless final products such as CO_2 and H_2O [7]. Mostly, there are two types of electro-Fenton (EF) processes used. The first one H_2O_2 and iron are supplied to the reactor from outside and insoluble cathode material is used, the second one only H_2O_2 is added to the reactor externally, iron ions (Fe^{+2}) are obtained from the iron material dissolved in the reactor [1]. Apart from these, there are also systems in which H_2O_2 is produced at the cathode [8]. Electro-Fenton processes are the systems where the same mechanism as the Fenton process occurs but the H_2O_2 or Fe^{+2} ions are obtained by passing an electric current through the anode and cathode materials [9].

$$H_2O_2 + OH^• \rightarrow H_2O + (HO_2)^• \quad (2)$$

$$Fe^{+2} + OH^• \rightarrow Fe^{+3} + OH^- \quad (3)$$

Eq. (2) shows that with the effect of excess H_2O_2 may be present in the medium, the strong oxidizing hydroxyl radicals with less oxidative properties $(HO_2)^•$ radicals will be converted, Eq. (3) shows that similarly, excess Fe^{+2} ions will completely eliminate the hydroxyl radicals [10]. These reaction equations indicate that it is very important to determine the reaction rates at the ideal ratios by optimization studies.

Chemical oxygen demand (COD) and color removal from wastewater consisting of textile, milk products, sugar, and flour industry effluents with advanced oxidation processes were investigated. Underdetermined operating conditions (temperature: 25°C and pH: 8), by using ozonation at a dosage of 300 mg/h administered for 10 min, it was achieved a color removal of 100% and a COD removal 96%. By using a UV/H_2O_2 process, they achieved color and COD removal of 91% and 82%, respectively. Also, when the Fenton process was applied, while color and COD removal of 79% and 60%, respectively in 60 min, by using the photo-Fenton process, approximately 100% removal efficiencies were achieved for both parameters. Furthermore, the electricity consumption of all processes used in the study was compared and the energy requirement was obtained from the lowest to the highest during $UV/Fe/H_2O_2 < H_2O_2 < O_3 < UV/H_2O_2 < UV$ process [11].

Kurt et al. [12] tried the electro-Fenton process in the treatment of leather industry wastewater and investigated the effect of initial pH, H_2O_2, current intensity, and electrolysis time on COD removal efficiency. In their electrolysis conditions (1,670 mg/L H_2O_2, 4 W electrical power, pH: 3 and reaction time of 10 min) they achieved a COD removal of 72%.

In this study, the effects of different cathode electrodes, current density, and the distance between electrodes on COD and color removal from leachate were investigated. The optimum experimental conditions were determined. Also, electrical energy consumptions were calculated within the scope of cost analysis.

2. Materials and methods

2.1. Leachate and features

The leachate used in the experiments was obtained from the site of Samsun solid waste landfill facility which is located and operated approximately 10 km from Samsun City Center. The storage area consists of 3 lots, the first stage with a total lot area of 5.6 ha, the second stage with a total lot area of 4 ha, and the third stage with a total lot area of 6.9 ha. Since May 2008, the landfill of waste storage operations is carried out. Landfill enters the middle-aged landfill class with this feature. 800–900 tons of waste is stored in the field per day. In 2016, a total of 269,327 tons of waste was disposed of in the field. From 2008 to June 2017, the amount of waste disposed of was 1,840,256 tons. 300 m^3 of leachate per day is formed in the field. The leachate that occurred in the field is sent to Samsun East Advanced Treatment Plant with tankers of 30 m^3 capacity (10 tankers per day) every day. The characteristics of the leachate used in the experiments are shown in Table 1.

2.2. Electrochemical system

For EF experiments, a rectangular Plexiglas reactor with dimensions of 74 mm × 83 mm × 140 mm was used. The useful volume of the reactor was 860 cm^3. All experiments were conducted with 0–45 min EF times. The dimension of electrodes 45 mm width × 55 mm length × 2 mm thickness was used in the experiment. The electrodes were placed in the reactor in monopolar parallel mode and vertical position and connected to a digital direct current power source (GW GPC-3060D DC power supply – 30 V, 6 A). The electrochemical system used in this study illustrated in Fig. 1.

2.3. Experimental procedure and analytical methods

In each stage of the experiment, 2 anode and 2 cathode electrodes were used. The surface area of the active anode was 94.64 cm^2. In each experiment, the EF reactor was filled with 750 mL wastewater and agitated continuously at 250 rpm with a cylindrical magnetic bar. In all experiments, concentrated sulfuric acid (98%) (H_2SO_4, Merck) and 1 M sodium hydroxide (NaOH) was used for pH adjustment. pH adjustment of wastewater was done with the Thermo Scientific Orion 4-Star brand and model pH meter. After adjusting the pH of the leachate to the desired value, COD and color measurements were made in

Table 1
The characterization of leachate

Parameter	Level
pH	7.50–7.90
COD (mg/L)	7,150–9,000
Conductivity (μS/cm)	20–40
Color (Pt-Co)	2,102–3,596

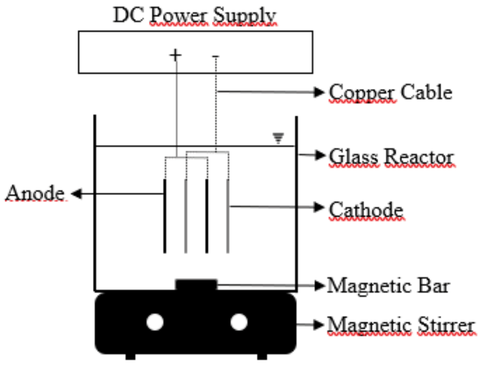

Fig. 1. Schematic diagram of the electrochemical set-up.

raw wastewater for each experiment. Subsequently, H_2O_2 (35% Merck) was added to the wastewater introduced into the reactor and stirred with a magnetic stirrer at 250 rpm. During the experiments, samples were taken at periodic intervals 2.5, 5, 7.5, 10, 15, 20, 25, 30, 35, 40, and 45 min. Samples were centrifuged at 9,000 rpm for 10 min. COD and color analyzes were done with Merck Spectroquant Nova 60A brand model spectrophotometer. All experiments were carried out according to the Standard Methods Book for Water and Wastewater. Within all the processes, COD values were measured through the closed reflux colorimetric method (5520-D) with a Merck Spectroquant Nova 60A brand model spectrophotometer [13]. The COD removal efficiency was calculated by using Eq. (4):

$$\text{Removal efficiency } (\%) = \frac{C_0 - C_t}{C_0} \times 100 \qquad (4)$$

where C_0 is the initial concentration of COD (mg/L); C_t is the concentration of COD corresponding time (mg/L).

Color measurements were carried out according to the Pt-Co method in the Standard Methods book. The absorbance values of the samples at 340 nm wavelength were measured and the color values were calculated by multiplying by the multiplication factor [13]. According to Heidmann and Calmano [14], the conductivity of wastewater (20–40 mS/cm) used in this study was sufficient for transferring electrons from anode to cathode. Therefore, no electrolyte was used. Most studies show that pH values ranging between 3 and 4 [7,15,16] are most effective on the COD and color removal by the electro-Fenton method. Therefore, the initial pH of the wastewater used in this study was adjusted to 3 prior to the addition of Fenton reagents for all experiments. Also, initial pH values increased overtime at every stage of our experiments. Maximum efficiencies for COD and color were obtained after 25 and 7.5 min EF, respectively. The pH changes obtained accordingly are presented in sections 3.1, 3.2, and 3.3.

3. Results and discussion

3.1. Effect of cathode material on COD and color removal

The choice of the cathode material is one of the factors that directly affect the efficiency of electro-Fenton processes. Carbon-containing materials are widely used in electro-Fenton systems due to their electrochemical properties [17]. Also, the cathode material directly affects the consumption of electrical energy. Important factors in cathode material selection are high metallic conductivity, stability, and electrical energy consumption. The most commonly used cathode materials include stainless steel, graphite, and carbon cloth [18]. In all stages of this study, high purity (>99.9) iron electrodes were used as the anode. However, in this first step, three different cathode materials were used. The results were discussed comparatively. In order to investigate the effect of cathode material on COD and color removal keeping the other conditions constant (pH: 3, current density: 100 A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, the distance between electrodes: 1.5 cm) different cathode materials (stainless steel, graphite, carbon cloth) were used in the experiments. Figs. 2 and 3 show the effect of different cathode materials on COD and color removal, respectively. As shown in Fig. 2, COD removal was not observed in the first 10 min. This shows that a certain time is required for Fe^{2+} to react with H_2O_2 to form hydroxyl radicals. That is, there was no COD removal since there were not enough hydroxyl radicals in the medium during the first 10 min. Removal efficiencies were increased for all cathode types between 10 and 20 min. After the 20th min, the removal efficiency for the steel cathode reached a peak with 82.5% in the 25th min. The removal efficiency for the graphite at the 25th min was 64.4% and 49.6% for the carbon cloth. At this minute, the electricity consumptions calculated for steel cathode, graphite, and carbon cloth were 0.75, 0.91, and 1.17 kWh/m^3, respectively. There was no significant change in COD removal efficiencies between 25 and 30 min. After 30 min, COD removal efficiencies decreased to zero. This is an indication that there is no H_2O_2 in the medium. Fig. 3 shows the effects of different cathode materials on color removal. As can be seen from Fig. 3, maximum removal efficiencies were reached at the end of 5 min for all cathode types. At the end of the 5th min, the color removal efficiency was found 61% for steel cathode, 58% for graphite, and 31% for carbon cloth. There was no significant change in removal efficiency between 5 and 10 min. After 10 min, the color removal efficiency decreased to zero. This can be explained by the absence of the hydroxyl radical in the medium and the increasing iron ions in the medium coloring the water. At the end of the 5th min, electrical energy consumption was calculated as 0.13, 0.16, and 0.23 kWh/m^3 for steel cathode, graphite, and carbon cloth, respectively.

When Figs. 2 and 3 were investigated together, steel was used as the cathode material in the following experiments since the lowest electrical energy consumption, and highest COD and color removal were obtained by steel cathode. The initial pH values of 3 increased to 3.87 and 3.17 in COD and color removal studies, respectively.

3.2. Effect of current density on COD and color removal

Current density is an important parameter that affects the operating cost of the electro-Fenton process and the efficiency of the treatment by affecting the solubility of the iron from the anode electrode [19]. The performance of electro-Fenton systems is significantly influenced by the current density, which is the driving force of electron transfer, and consequently

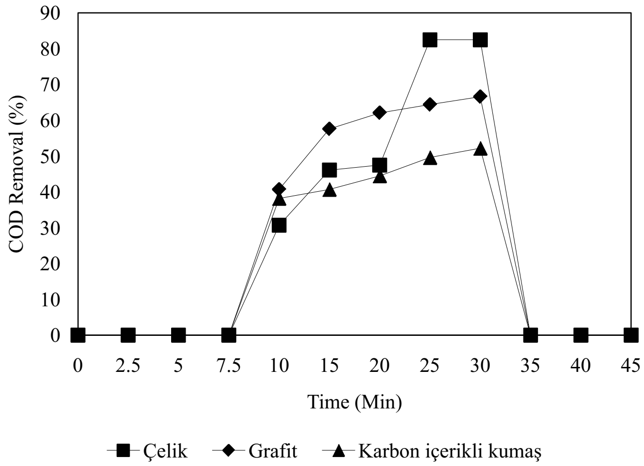

Fig. 2. The effect of cathode material on COD removal (conditions: anode: iron, pH: 3; current density: 100 A/m^2; H_2O_2: 2,000 mg/L; stirring rate: 250 rpm; the distance of electrodes: 1.5 cm).

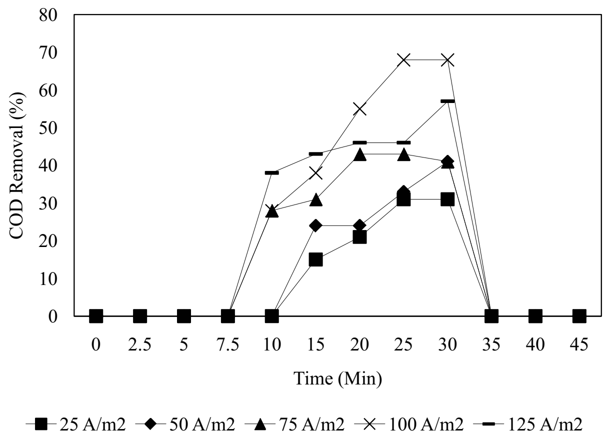

Fig. 4. The effect of current density on COD removal (conditions: anode: iron; cathode: steel; pH: 3; H_2O_2: 2,000 mg/L; stirring rate: 250 rpm; the distance of electrodes: 1.5 cm).

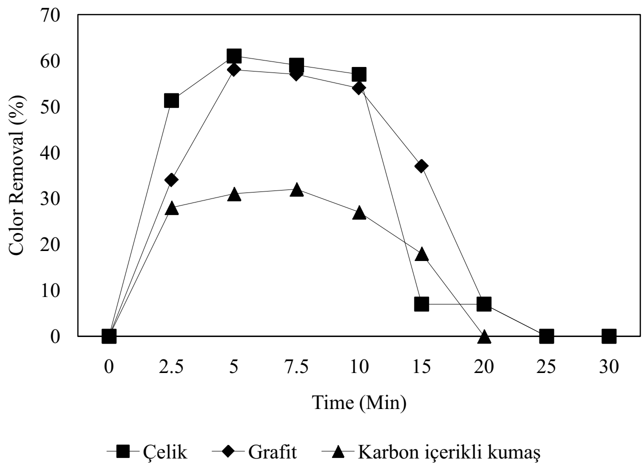

Fig. 3. The effect of cathode material on color removal (conditions: anode: iron; pH: 3; current density: 100 A/m^2; H_2O_2: 2,000 mg/L; stirring rate: 250 rpm; the distance of electrodes: 1.5 cm).

corresponds to the production rate of H_2O_2. The high current density increases the amount of hydroxyl radical produced in the solution. Furthermore, higher current density leads to faster Fe^{2+} regeneration (Eq. (5)) and increases the efficiency of Fenton chain reactions [20].

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (5)$$

In EF processes, two effects of current density can be mentioned. EF reactor with appropriate electrodes, the cathode and anode allow H_2O_2/Fe^{+2} production [21]. Zhang et al. [22] in their study, it was determined that when the current was increased from 2.5 to 3 A, COD removal efficiency decreased from 87.2% to 79.3% [22]. On the other hand, Hou et al. [23] reported that when the current density was increased from 10 to 25 mA/cm^2, total organic carbon removal increased from 51.4% to 72.3% [23]. Therefore, the applied current density needs to be optimized for the balance between the desired efficiency and energy costs. In this study, in order to investigate the effect of current density on COD and color removal, the effect of different current densities (25–125 A/m^2) was examined provided that other conditions (anode: iron, cathode: steel, pH: 3, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, the distance of electrodes: 1.5 cm) were kept constant. Figs. 4 and 5 show the effect of different current densities on COD and color removal, respectively. As shown in Fig. 4, COD removal was not observed during the first 7.5 min. This indicates that a certain period of time must elapse for the formation of hydroxyl radicals in the medium. At the end of 10 min, the removal efficiencies were 0%, 0%, 28%, 28%, and 38% for the current density ranging from 25, 50, 75, 100, and 125 A/m^2, respectively. For all current densities, the removal efficiencies increased between 10 and 25 min to a maximum. This is an indication that hydroxyl radicals increase in the medium. At the end of the 25th min, the removal efficiencies were 31%, 33%, 43%, 68%, and 46% for the current density ranging from 25 to 125 A/m^2, respectively. By increasing current density from 25 to 100 A/m^2, COD removal increased from 31% to 68%. Several studies confirm this correlation. For example, Asaithambi et al. [24] found that COD removal increased from 37.5% to 100% by increasing the current density from 7 to 35 A/m^2 in their study in relation to leachate by the electro-Fenton method. Atmaca [1] found that COD removal increased from 45% to 70% when the current intensity was increased from 1 to 3A. This may be related to the formation of an increased amount of hydroxyl radical in solution with increasing current density [25]. At the end of the 25th min, electrical energy consumption (for 25–125 A/m^2) was 0.06, 0.22, 0.39, 0.59, and 0.95 kWh/m^3, respectively. After 30 min, for all current densities, the removal efficiencies decreased to zero. This is an indication that there is no H_2O_2 in the medium. As a result, the highest removal efficiency was reached at the end of 25 min at a current density of 100 A/m^2 and the optimum current density value was found to be 100 A/m^2.

Fig. 5 shows the effect of current density on color removal. As the current density increased from 25 A/m^2 to 75 A/m^2, the color removal efficiency increased from 54% to 69%. Atmaca [1] reported that color removal increased from 74% to 91% when the current intensity was increased from 1 to 2.5 A. The highest color removal efficiency (69%) was achieved with an electrical energy consumption of 0.11 kWh/m^3 at 75 A/m^2 at the end of 7.5 min EF. At the end of this EF time, the removal efficiencies obtained for the current densities of 25, 50, 100, and 125 A/m^2 were 54%, 39%, 59%, and 62%, respectively and electrical energy consumption was 0.01, 0.05, 0.22, and 0.30 kWh/m^3, respectively. After 7.5 min, color removal efficiencies were reduced for all current densities. These results showed that the optimum current density value for color removal from the leachate was 75 A/m^2. The initial pH values of 3 increased to 3.42 and 3.29 in COD and color removal studies, respectively.

3.3. Effect of distance between electrodes on COD and color removal

In the electro-Fenton process, the distance between electrodes is another important parameter affecting pollutant removal efficiency. If the distance between electrodes is too close, the electron transfer cannot take place very well and the Fe^{2+} ions are oxidized to Fe^{3+} according to Eq. (6). As Fe^{2+} ions to react with H_2O_2 and thus, the formation of hydroxyl radicals decreases, the pollutant removal efficiency decreases [26].

$$Fe^{2+} \rightarrow Fe^{3+} + e^- \tag{6}$$

As the distance between electrodes increases, the electrical resistance between the electrodes increases, resulting in longer electrolysis time and lower contaminant removal efficiency [27]. On the other hand, increases in the distance between electrodes increase the consumption of electrical energy [18]. Zhang et al. found that COD removal efficiency decreased from 80.8% to 71.8% by increasing the distance between electrodes from 2.1 to 2.8 cm in their study with leachate, COD removal efficiency increased from 73.6% to 80.4%, increasing the distance between electrodes from 0.7 to 1.3 cm in the same study. In this study, the effect of electrode distance on COD and color removal was investigated for optimum experimental conditions. Figs. 6 and 7 show the effect of distance between electrodes on COD and color removal, respectively. To investigate the effect of distance between electrodes on COD removal, 0.5, 1, and 1.5 cm distances were tested under experimental conditions. As can be seen from Fig. 6, COD removal efficiency increased from 32% to 68% at the end of 25 min by increasing the distance between electrodes from 0.5 to 1.5 cm. This can be explained by the fact that the electron transfer does not perform very well if the distance between the electrodes is too small. At the end of 25 min, electrical energy consumption was 0.48, 0.56, and 0.60 kWh/m^3 for 0.5, 1, and 1.5 cm distances, respectively. While there was no significant change in removal efficiencies for all distances between 25–30 min, removal efficiencies decreased to zero after 30 min. The effect of the distance between electrodes on color removal was also investigated for optimum experimental conditions (anode: iron, cathode: steel, pH: 3, current density: 75 A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm). As can be seen from Fig. 7, the color removal efficiency increased from 40% to 0.08 kWh/m^3 with a maximum electrical energy consumption of 69% by increasing the distance between electrodes from 0.5 to 1.5 cm at the end of the 7.5 min EF period. After 7.5 min, the color removal efficiency for all distances decreased to zero. This can be explained by the excess iron ions coloring in water and the absence of hydroxyl radicals. The initial pH values of 3 increased to 3.42 and 3.29 in COD and color removal studies, respectively. Tsai et al. [28] in their study with leachate, investigated the COD removal using iron and aluminum anodes and cathode copper. They achieved a COD removal of 41.8% with Fe–Cu electrodes in 10 V voltage and 20 min of operating time, and a COD removal of 39.6% efficiency when Al–Cu electrodes were used. Altin [16] investigated the performance of the photo-electro-Fenton process in the treatment of leachate. COD, color, and phosphate removal efficiencies were determined in their study. Also, electrocoagulation, electro-Fenton, UV/H_2O_2, and the photo-electro-Fenton processes were compared. In their study, the highest pollutant removal efficiencies were obtained by the photo-electro-Fenton process. At optimum operating conditions (pH: 3, H_2O_2 concentration: 3,000 mg/L, current strength: 2.5 A and operating time: 20 min), the contaminant removal efficiencies were 94% for COD, 97% for color and 96% for phosphate, respectively [16]. In this study, it was investigated the efficiency of the electro-Fenton process in the treatment of leachate. Under optimal conditions, the contaminant removal efficiencies were 82.5% for COD, 69% for color, respectively. It was understood that the efficiencies obtained with this study were similar to the literature.

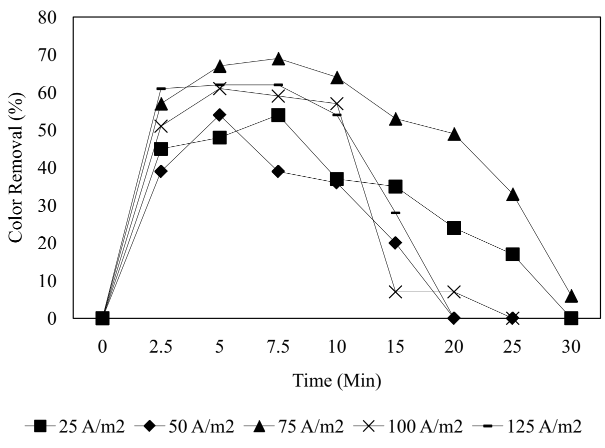

Fig. 5. The effect of current density on color removal (conditions: anode: iron; cathode: steel; pH: 3; H_2O_2: 2,000 mg/L; stirring rate: 250 rpm; the distance of electrodes: 1.5 cm).

3.4. Cost analysis and calculations

Electrical energy consumption was calculated using Eq. (7) [29] and anode consumption was calculated using Eq. (8) [30].

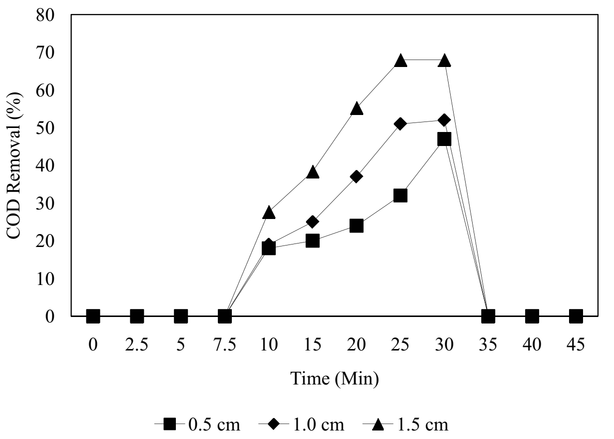

Fig. 6. The effect of distance between the electrodes on COD removal (conditions: anode: iron; cathode: steel; pH: 3; H_2O_2: 2,000 mg/L; stirring rate: 250 rpm).

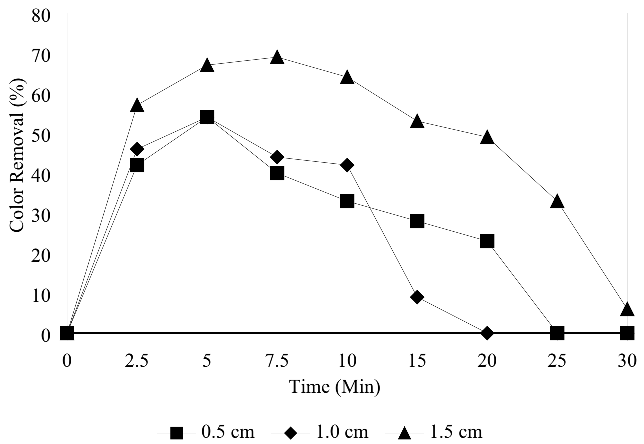

Fig. 7. The effect of distance between the electrodes on color removal (conditions: anode: iron; cathode: steel; pH: 3; H_2O_2: 2,000 mg/L; stirring rate: 250 rpm).

$$E = \frac{UxIxt}{V} \tag{7}$$

where E is the energy consumption (kWh/m^3); U is the applied voltage (V); I is the applied current (A); t is the experimental time (s); V is the wastewater volume (L).

$$\Delta M = \frac{IxtxM}{zxFxV} \tag{8}$$

where ΔM is the theoretical (produced g Fe^{2+}/m^3 treated wastewater); I is the applied current (A); t is the experimental time (s); M is the iron molecular weight (g/mol); Z is the oxidation number of iron (2); V is the wastewater volume (L).

Under optimal conditions (anode: iron, cathode: steel, pH: 3, current density: 100 A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, distance between electrodes: 1.5 cm) electricity consumption was 0.75 kWh/m^3, anode consumption was 555 g/m^3, H_2O_2 consumption was 5.05 L/m^3. The total cost (electrical energy + anode consumption + H_2O_2 consumption) was calculated as 71 \$/m^3. The cost per kilogram of COD was found to be \$12.03 (\$12.03/1 kg COD).

4. Conclusions

In this study, the effects of some operating parameters (cathode material, current density, the distance between electrodes) on COD, and color removal from leachate were investigated separately. Optimum conditions for COD and color removal were determined. The optimum conditions for COD removal were found pH: 3, current density: 100 A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, distance between electrodes: 1.5 cm. In these conditions, a maximum COD removal of 82.5% was achieved with electrical energy consumption of 0.75 kWh/m^3 from leachate. Optimum conditions for color removal were determined as pH: 3, current density: 75 A/m^2, H_2O_2 concentration: 2,000 mg/L, stirring rate: 250 rpm, distance between electrodes: 1.5 cm. Under these conditions, a maximum color removal of 69% was achieved with electrical energy consumption of 0.08 kWh/m^3 from leachate. Although the results showed that the electro-Fenton method can be applied successfully in COD and color removal from leachate, it was determined that the treated leachate did not provide discharge limits prevailing in Turkey. On the other hand, since there are limited studies on the effect of different cathode materials on the COD and color removal from leachate by electro-Fenton, it is thought that this study will shed light on future studies.

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