

COD and Color Removal from Landfill Leachate by photo-electro-Fenton Process

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This study investigated the effects of operational parameters such as pH (2.5-5.0), current density (10-50 A/m²), H₂O₂ concentration (250-1500 mg/L) and distance between electrodes (1.0-1.5 cm) on COD and color removal from the leachate by the photo-electro-Fenton method. Under determined optimum experimental conditions (pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV lamp: 16W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm), maximum 83.84% COD removal and 84.46% color removal from leachate were achieved with an electrical energy consumption of 11.02 and 5.51 kWh/m³, respectively. Maximum removal efficiencies for COD and color were reached after 90 and 45 minutes of treatment, respectively.

Keywords: COD, color, removal, photo-electro-Fenton, leachate, cost.

1. INTRODUCTION

Leachate water is water with a very high pollution load, created by the infiltration of rainwater through the solid waste stored in the solid waste landfill, and the moisture and water in the composition of the waste. The composition of the wastes in landfills may change as a result of biochemical reactions such as aerobic, anaerobic, methanogenic and stabilization phases occurred over time. The amount of leachate produced is mainly affected by precipitation, evapotranspiration, runoff, groundwater infiltration, and the degree of compression within the sanitary landfill [1]. Pollutants in the leachate can be divided into four main groups. These are dissolved organic substances, inorganics, heavy metals and xenobiotic organic compounds. In other words, the most important pollutant parameters used in the identification of leachate components are: pH, suspended solids, biochemical oxygen demand (BOD), chemical oxygen demand (COD), ammonium (NH₄⁺), total nitrogen (TN), chloride, phosphorus, heavy metals and alkalinity, and due to these contaminant components and can pose a serious threat to the environment if it reaches water bodies without treatment. For example, excess nitrogen presence in water

bodies can trigger algae growth and the dissolved oxygen level can be depleted as a result of eutrophication. Exposure to high concentrations of ammonium nitrogen may cause acute toxicity to aquatic organisms [1,2]. Due to their high level of pollutant properties, leachate must be treated before being introduced into receiving environments.

Biological, physical and chemical processes, and a combination of biological and physical-chemical processes are often used in the treatment of effluents [1]. Although biological treatment is widely used in the treatment of leachate containing high amounts of organic matter due to its reliability and high cost-effectiveness, and it is more efficient in the treatment of young leachate characterized by high BOD/COD ratios [1]. Physical-chemical processes such as coagulation-flocculation [3,4], chemical precipitation, adsorption [5], membrane filtration [4,6] ion exchange, chemical oxidation/advanced oxidation processes (AOPs) [3] and electrochemical methods can also be used for the treatment of leachate [1,4].

In recent years, interest in the use of oxidation processes (AOPs) has increased. These processes provide mineralization of inorganic and organic pollutants that are difficult to break down into final products such as carbon dioxide and water. One of these processes, Fenton oxidation ($\text{Fe}^{2+}/\text{H}_2\text{O}_2$) is widely used for these purposes. It is an environmentally friendly process that provides high COD removal efficiency, and the main mechanism of the Fenton system is based on OH^\bullet oxidation, one of the strongest oxidizing ($E = 2.73 \text{ V}$) agents [1,7]. Fenton reactions take place by electron transfer between H_2O_2 and a metal ion such as iron (Fe^{2+}) which acts as a catalyst. As a result of this reaction, hydroxyl radicals are produced which are very effective in converting pollutants into CO_2 , water and inorganic salts as final products. The classical Fenton process includes the following reaction sequence [8,9,10].



The formation of hydroxyl radicals (Eq. (1)) is very rapid. The net reaction (1-7) can generally be described as the decomposition of H_2O_2 in the presence of iron as a catalyst.



Equation (8) states that the Fenton reaction is completed under acidic conditions, that is, the presence of H^+ ions is necessary for the decomposition of H_2O_2 . Iron acts as a catalyst in the above reactions by changing its form between Fe^{2+} and Fe^{3+} [8].

The photo-electro-Fenton process is similar to the electro-Fenton process. The only difference between them is ultraviolet (UV) irradiation sent into the same reactor. Thus, while 2 moles of OH^\bullet radicals are produced from 1 mol of H_2O_2 (Eq. (12)), the reaction can be accelerated by the re-formation of Fe^{2+} from iron complexes (Eqs. (10-11)) that can also be formed [11]. The photo-electro-Fenton process takes place according to the following reactions.



Some of the studies on advanced oxidation processes and leachate treatment in the literature are summarized below. Carluccio et al. obtained 82% COD removal under optimum conditions (pH: 3, time: 120 minutes, $\text{H}_2\text{O}_2/\text{Fe}^{2+}$: 1, $\text{H}_2\text{O}_2 = \text{Fe}^{2+}$: 3500 mg/L) in their study by Fenton oxidation from leachate water [12]. Gulsen and Turan achieved 85% COD removal under optimum conditions (pH: 2.5, H_2O_2 : 1200 mg/L,) in their COD removal study by Fenton oxidation from leachate pre-treated with an anaerobic fluidized bed reactor [13]. Lopez et al. achieved 60% COD removal under optimum conditions (pH: 3, Fe^{2+} : 830 mg/L, H_2O_2 : 10000 mg/L, treatment time: 2 hours) in their pre-treatment study performed by Fenton oxidation from leachate water [14]. Lin and Chang achieved 70.4% COD removal under optimum conditions (pH: 4, current: 2.5A, H_2O_2 : 1000 mg/L, treatment time: 31 minutes) in their treatment study performed by electro-Fenton method from leachate water [15]. Cortez et al. obtained maximum 60.9% COD removal in their study by Fenton oxidation from leachate under optimum conditions ($[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$: 3, Fe^{2+} : 4 mmol/L, pH: 3, treatment time: 40 minutes) [16]. Guo et al. achieved 60.8% COD removal under optimum conditions (pH: 3, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$: 20 g/L, H_2O_2 : 20 mL/L) in their study of COD removal from leachate with the Fenton method [17]. Kochany and Lipeczynska-Kochany, in their pre-treatment study with Fenton oxidation from leachate, under optimum conditions (pH: 3.5, Fe^{2+} : 56 mg/L, H_2O_2 : 650 mg/L, $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ molar ratio: 19.1) they achieved 66% COD removal [18]. Wang et al. applied the Fenton oxidation process to biologically treated leachate. Under optimum conditions (COD input: 220 mg/L, Fe^{2+} : 4.5 mmol/L, H_2O_2 : 5.4 mmol/L, $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ molar ratio: 1.2) they achieved 56% COD removal [19]. Primo et al. achieved 58% and 77% COD removal from the leachate by Fenton and photo-Fenton processes under optimum experimental conditions, respectively (COD input: 3300-4400 mg/L, Fe^{2+} : 2000 mg/L, H_2O_2 : 15000 mg/L, UV source: 150 W) [20].

The present study; investigated the effects of initial pH, current density, H_2O_2 concentration and distance between electrodes on COD and color removal from leachate by photo-electro-Fenton, revealed the optimum experimental conditions, and performed a cost analysis.

2. MATERIALS AND METHODS

2.1 Leachate characteristics

The leachate was obtained from the landfill located in Samsun. The landfill, where waste storage operations have been carried out regularly since 2008, is included in the middle-aged solid waste class with this feature, and approximately 300 m³ of leachate is generated daily at the site. Approximately 800-900 tons of garbage is discharged daily to the site. The characterization of the leachate is shown in Table 1.

Table 1. Characteristic of wastewater

Parameter	Level
pH	7.91-8.20
COD (mg/L)	7,983-8,150
Conductivity (mS/cm)	20-40
Color (Pt-Co)	1,887-1,980

2.2 Photoreactor and experimental procedure

Experiments were carried out in a cylindrical photoreactor with an inner diameter of 7.70 cm and height of 48.5 cm. One 16W power low-pressure mercury vapor lamp (UV-C, 254 nm) and 1 Anode (Iron), and 1 Cathode (Stainless Steel-304) electrode were placed inside the photoreactor. The electrode dimensions were 4.6 cm x 29.6 cm x 0.2 cm (width x height x thickness) and the active anode surface area was 267.498 cm². The electrodes were connected directly to the current power supply (GW GPC-3060D DC Power Supply - 30V, 6A) in monopolar parallel mode. Figure 1 shows the electrochemical system, Figure 2 shows the top view of the photoreactor.

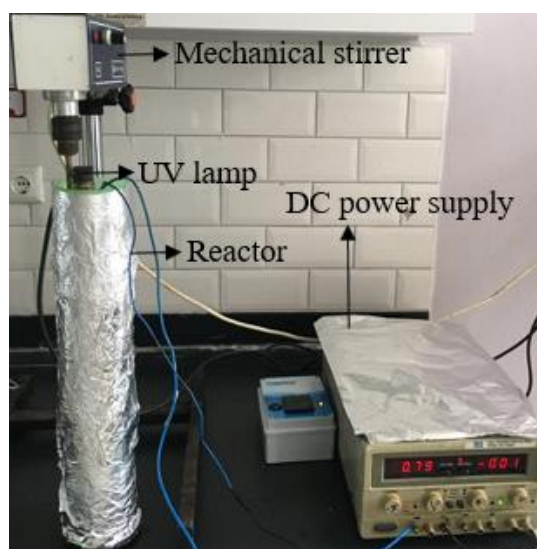


Figure 1. Experimental setup

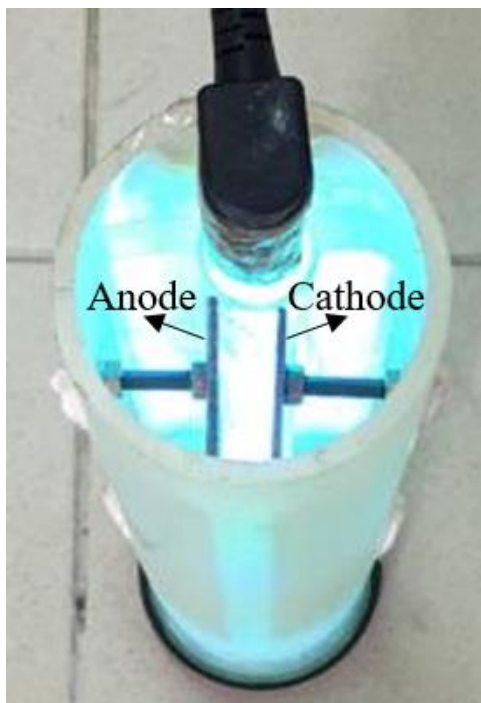


Figure 2. The top view of the photoreactor

2.3 Experimental procedure and analytical methods

To remove the suspended solids that may affect the electrochemical process, the wastewater was subjected to a pre-filtration process using a sieve with a diameter of 63 μm [21]. In each experiment, the photoreactor was filled with 2.2 L of wastewater and operated in batch mode. Stirring was carried out mechanically at 250 rpm from the top of the reactor. While Fe^{2+} was produced from the iron anode in the reactor, H_2O_2 was added externally. pH measurements were made with Thermo Scientific Orion 4 Star brand and model pH meter. After the wastewater was put into the photoreactor, it was mixed for a few minutes and then the voltage meter and UV lamp were operated simultaneously after the desired amount of H_2O_2 was added. The moment the UV lamp was turned on was considered the start time of the experiment. Taking samples at periodic intervals (1, 3, 5, 10, 20, 30, 45, 60, 90, 120, 150 minutes) and centrifuging at 9000 rpm for 10 minutes, then using a spectrophotometer (Merck Spectroquant Nova 60A brand and model) COD and color measurements were made.

All of the experiments and analyzes were made according to Standard Methods prepared for water and wastewater [22]. The Removal efficiencies were calculated by Equation (13).

$$\text{Removal efficiency (\%)} = \frac{C_0 - C_t}{C_0} \times 100 \quad (13)$$

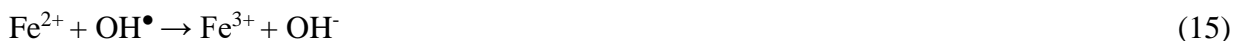
C_0 = Initial COD concentration (mg/L)

C_t = COD concentration at time t (mg/L)

3. RESULTS AND DISCUSSION

3.1 Effect of current density on COD and color removal

Current density is an important parameter that affects the efficiency of Fenton processes [23,24]. To examine the effect of current density on COD and color removal, current densities ranging from 10 to 50 A/m² were studied. Figure 3a and Figure 3b show COD and color removal efficiencies, respectively. Maximum 79.75% COD and 80.57% color efficiencies were achieved at 10 A/m² after 90th and 45th minutes of treatment, with output pH values of 3.79 and 3.48, respectively. While the maximum color removal efficiencies were 70.87%, 65.03%, 55.33%, 45.62% for the current densities ranging from 20 to 50 A/m², the maximum COD removal efficiencies were 69.53%, 57.26%, 49.08%, 38.80%. By increasing the current density from 10 A/m² to 50 A/m², the maximum COD removal decreased from 79.75% to 38.86%, and the maximum color removal from 80.57% to 45.62%. The fact that the increase in the amount of Fe²⁺ ions dissolved from the anode per unit time according to Equation (14) when the current density increases, contributes to the formation of COD and color over time can be considered as the reason for this case. This decrease in COD removal can also be explained by the increasing current that causes side reactions such as the conversion of OH• radicals to OH⁻ ions (Equation 15).



As can be seen from Figure 3a, COD removal efficiencies increased over time for all current density values during the first 90 minutes. This situation can be explained by the increased OH• radicals in the reactor. The electrical energy consumptions after 90 minutes of treatment were 11.02, 11.31, 11.73, 12.37, 13.20 kWh/m³ for 10, 20, 30, 40, 50 A/m², respectively. Figure 3b shows the effect of current density on color removal. The electrical energy consumptions after 90 minutes of treatment were 5.51, 5.69, 5.92, 6.19, 6.60 kWh/m³ for current densities of 10, 20, 30, 40, 50 A/m², respectively. Color removal efficiencies for all current densities decreased overtime after 45 minutes. This situation can be explained by the decrease of OH• radicals in the reactor and the increase in color due to the iron ions formed. The current density of 10 A/m², which provided maximum COD and color removals and the lowest electrical energy consumption, was accepted as optimum and was used in subsequent experiments.

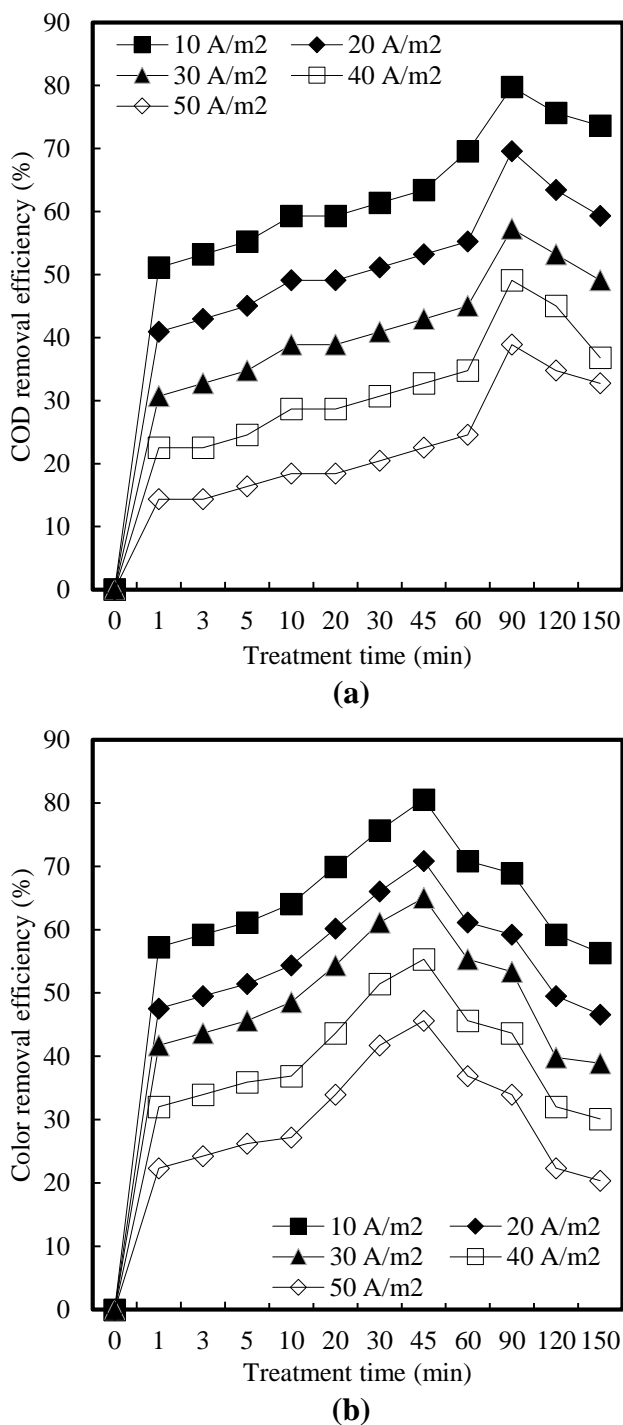
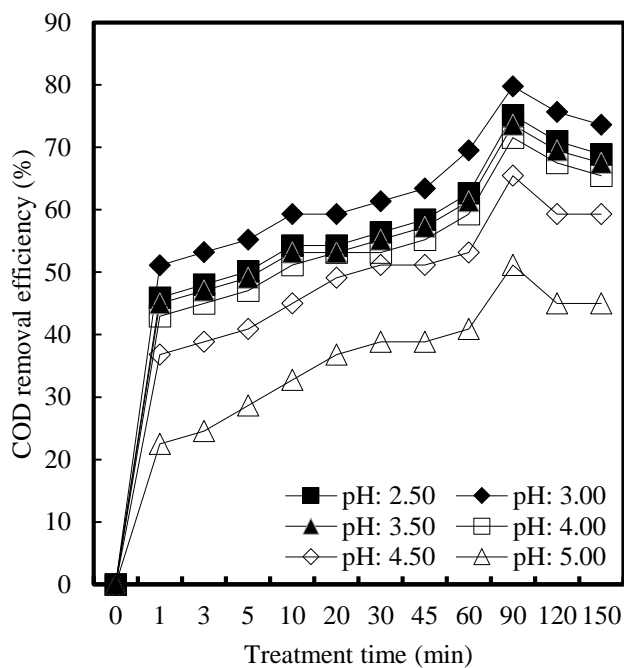


Figure 3. Effect of current density on (a) COD removal; (b) color removal (pH: 3, H₂O₂: 500 mg/L, UV: 16W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm)

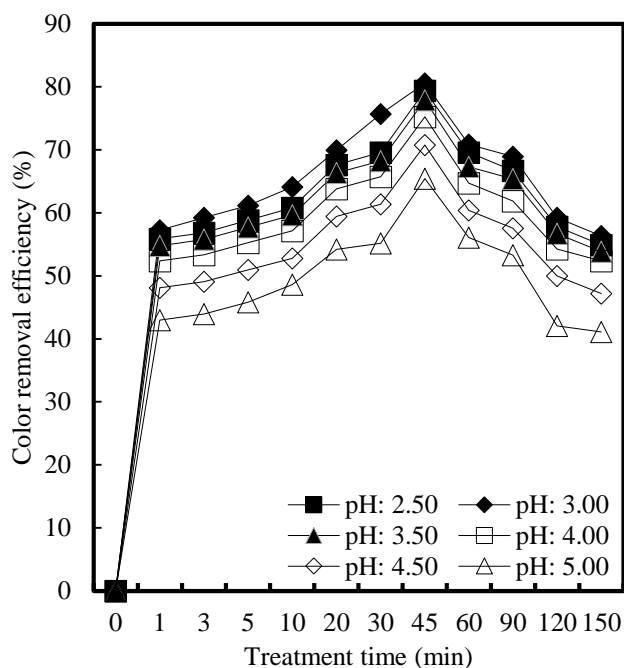
3.2 Effect of pH on COD and color removal

To examine the effect of pH on COD and color removal, pH values varying from 2.5 to 5 were studied. As can be seen from Figure 4a and Figure 4b, the maximum 79.75% COD and 80.57% color removal efficiencies were reached at pH 3 after 90th and 45th minutes of treatment, respectively. The

final pH values obtained after 90th and 45th minutes were 3.79 and 3.48, respectively. While the maximum COD removal efficiencies for pH 2.5, 3.0, 3.5, 4.0, 4.5, 5.0 were 75.16%, 79.75%, 73.62%, 71.57%, 65.44% and 51.13%, the maximum color removal efficiencies were 79.38, 80.57%, 77.86%, 75.23%, 70.73% and 65.40%.



(a)



(b)

Figure 4. Effect of pH on (a) COD removal; (b) color removal (Current density: 10 A/m², H₂O₂: 500 mg/L, UV: 16W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm)

By increasing the pH from 3 to 5, COD removal decreased from 79.75% to 51.13%, and color removal from 80.57% to 65.40% after 90 minutes of treatment. This can be explained as follows: dissolved iron ions begin to precipitate as Fe^{3+} , $\text{Fe}(\text{OH})_3$ above pH 3, and the formed $\text{Fe}(\text{OH})_3$ can only lower the dissolved Fe^{3+} concentration and accumulate on the electrode surfaces, preventing the regeneration of Fe^{2+} that may occur with UV effect. Similarly, Guo et al. reported in their study by Fenton oxidation from leachate that the COD removal decreased from 72% to 52% by increasing the pH from 3 to 5 [17]. The electrical energy consumption obtained after 90 minutes of treatment for pH 2.5, 3.0, 3.5, 4.0, 4.5, 5.0, were 11.04, 11.02, 11.04, 11.06, 11.05, 11.08 kWh/m^3 , respectively. As seen in Figure 4b, the color removal efficiencies increased for all pH values during the first 45 minutes. This situation can be explained by the increasing OH^\bullet radical in the reactor over time. The electrical energy consumption obtained after 45 minutes of treatment were 5.51, 5.50, 5.50, 5.52, 5.52, 5.53 kWh/m^3 for pH 2.5, 3.0, 3.5, 4.0, 4.5, 5.0, respectively.

3.3 Effect of H_2O_2 on COD and color removal

To examine the effect of H_2O_2 concentration on COD and color removal from leachate, H_2O_2 concentrations ranging from 250 to 1500 mg/L were studied. The results are given in Figure 5a and Figure 5b, respectively. The maximum 83.84% COD and 84.46% color removal were achieved at the H_2O_2 concentration of 1000 mg/L after 90th and 45th of treatment minutes, respectively, with final pH values of 3.55 and 3.29. The maximum COD removal efficiencies for 250, 500, 750, 1250, 1500 mg/L H_2O_2 concentrations were 53.16%, 79.75%, 81.80%, 75.67% and 69.53%, while the maximum color removal efficiencies were 56.32%, 80.57%, 82.52%, 77.63% and 70.87%. When increased H_2O_2 concentration from 250 mg/L to 1000 mg/L, COD removal increased from 53.16% to 83.84%. This situation may be associated with increasing OH^\bullet radicals in the environment with increasing H_2O_2 concentrations. Guo et al. reported in a study, which was on oxidation of leachate by Fenton, that when the H_2O_2 concentration was increased from 5 mL/L to 20 mL/L, COD removal increased 62% to 70% [17]. A similar correlation was obtained by Bali in color removal from Direct Red 28 (DR28) dye by the UV/ H_2O_2 process, indicating that color removal increased from 64.2% to 85.7% by increasing the initial H_2O_2 concentration from 5 mM to 60 mM [25]. As can be seen from Figure 5a, COD removal decreased from 83.84% to 69.53% by increasing H_2O_2 concentration from 1000 mg/L to 1500 mg/L. This can be explained by the scavenging effect of excess H_2O_2 on hydroxyl radicals according to Equation (3). The electrical energy consumed for 1000 mg/L H_2O_2 concentration at which 83.84% maximum COD removal was achieved, was 11.02 kWh/m^3 . By increasing H_2O_2 concentration from 250 mg/L to 1000 mg/L, after 45 minutes of treatment the color removal efficiency increased from 53.16% to 84.46%. On the other hand, with increasing the H_2O_2 concentration from 1000 mg/L to 1500 mg/L, the color removal efficiency decreased from 84.46% to 70.87%. The amount of electrical energy consumed after 45 minutes of treatment for 1000 mg/L H_2O_2 concentration at which 84.46% maximum color removal was achieved was 5.51 kWh/m^3 . A concentration of 1000 mg/L H_2O_2 , which provides the maximum COD and color removal with the lowest electrical energy consumption, was considered optimum for subsequent experiments.

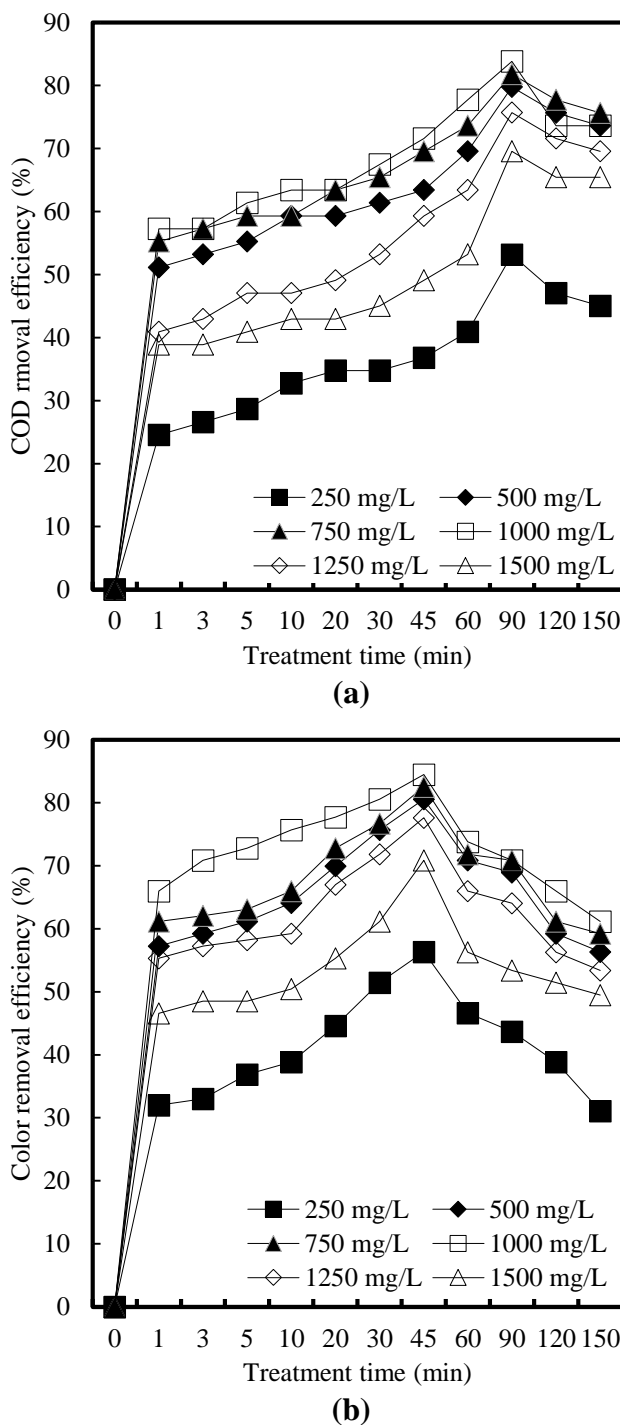
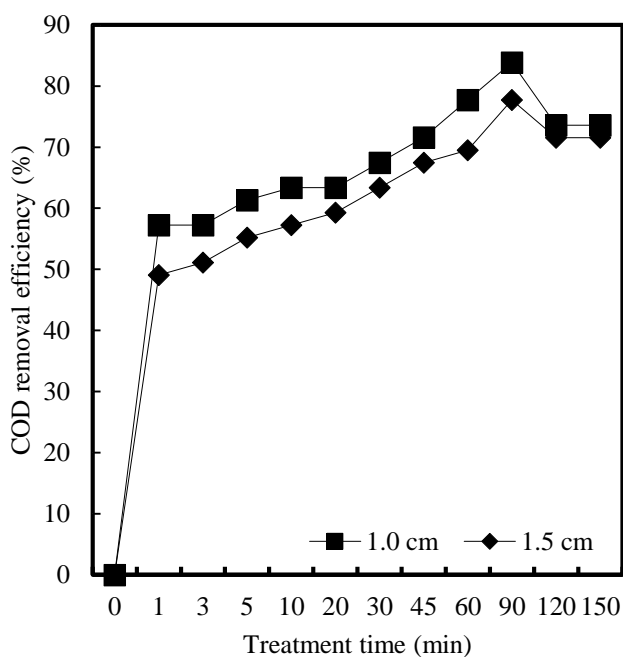


Figure 5. Effect of H₂O₂ on (a) COD removal; (b) color removal (pH: 3, current density: 10 A/m², UV: 16W, stirring rate: 250 rpm, distance between electrodes 1.0 cm)

3.4 Effect of distance between the electrodes on COD and color removal

The distance between electrodes is an important parameter that affects the efficiency of Fenton systems. If the distance between electrodes is too short, electrically generated Fe²⁺ ions are oxidized to Fe³⁺, which reduces the efficiency of Fenton reactions. On the other hand, if this distance is too large,

the electrical energy consumed increases due to the increased ohmic resistance between the electrodes. [26]. For both reasons, it is very important to examine the distance between electrodes in electrochemical treatment studies. The effect of distance between electrodes (1.0 and 1.5 cm) on COD and color removal from the leachate was examined under final optimum experimental conditions (pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV lamp: 16W, stirring rate: 250 rpm). Figure 6a and Figure 6b show the changes in the removal efficiencies. Maximum COD removal decreased from 83.84% to 77.71% after 90 minutes of treatment when the distance between electrodes was increased from 1.0 cm to 1.5 cm. After 90 minutes of treatment, in which the maximum COD removal efficiency was achieved, the electrical energy consumption was 11.02 kWh/m³ and 11.04 kWh/m³ for the distances between the electrodes of 1.0 and 1.5 cm, respectively. A similar correlation was found in a study by Asaithambi et al. which was on COD and color removal from leachate by photo-electro-Fenton. Under their optimum experimental conditions (COD input: 2000 mg/L, pH: 3, current density: 30 A/m², H₂O₂: 300 mg/L, UV lamp: 32W, test duration: 4 hours); when the distance between electrodes was increased from 0.75 to 3 cm, COD removal decreased from 97% to 69.50% and electrical energy consumption increased from 3.1 to 6.5 kWh/m³ [27]. As seen in Figure 6b, the maximum color removal decreased from 84.46% to 79.58% at the end of 45 minutes by increasing the distance between electrodes from 1.0 cm to 1.5 cm. After 90 minutes of treatment, in which the maximum color removal efficiency was achieved, the electrical energy consumption was 5.51 kWh/m³ and 5.53 kWh/m³ for the distances between the electrodes of 1.0 and 1.5 cm, respectively. At the end of the 90th and 45th minutes when the maximum COD and color removal was achieved, wastewater's final pH values were 3.55 and 3.29, respectively.



(a)

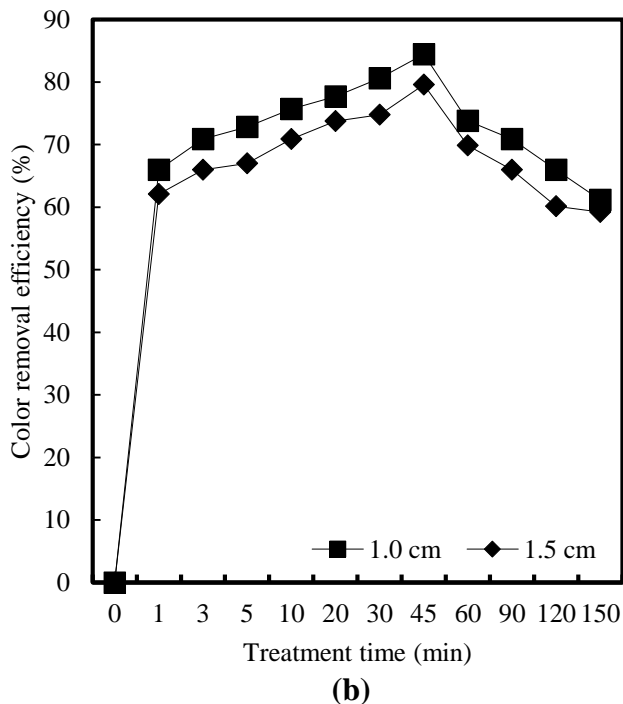


Figure 6. Effect of distance between electrodes on (a) COD removal; (b) color removal (Anode: Iron, Cathode: Steel, pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV: 16W, stirring rate: 250 rpm)

3.5 Operational cost analysis

Cost calculations were calculated for the optimum experimental conditions obtained. Electrical energy from voltage meter, anode material and UV source energy consumptions were calculated by Equation (16), Equation (17) and Equation (18), respectively [28,29].

$$E = \frac{UxIxt}{V} \tag{16}$$

E = Electrical energy consumption (kWh/m³)

U = Voltage (V)

I = Applied current (A)

t = Treatment time (sec)

V = Wastewater volume (L)

$$\Delta M = \frac{IxtxM}{zxFxV} \tag{17}$$

ΔM = Theoretically consumed Fe²⁺ amount (g Fe²⁺/m³)

I = Applied current (A)

t = Treatment time (sec)

M = molecular weight of iron (g/mol)

Z = number of valence electrons of iron (2)

V = Wastewater volume (L)

$$\text{UV source energy consumption (kWh/m}^3\text{)} = \frac{\text{Power (W)} \times \text{Time (saat)}}{\text{Wastewater volume (m}^3\text{)}} \quad (18)$$

The cost calculations obtained using Equations (16) - (18) are as follows: for maximum COD removal (Anode: Iron, Cathode: Steel, pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV lamp: 16 W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm) electricity energy consumption was 11.02 kWh/m³, anode consumption: 192 g/m³, H₂O₂ consumption: 2.52 L/m³. The total treatment cost (electrical energy consumption + anode consumption + H₂O₂ consumption) was found to be 309.83 TL/m³ ≈ 41.3 US \$/m³. For maximum color removal (Anode: Iron, Cathode: Steel, pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV lamp: 16 W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm) electricity energy consumption was 5.51 kWh/m³, anode consumption: 96 g/m³, H₂O₂ consumption: 2.52 L/m³. The total treatment cost (electrical energy consumption + anode consumption + H₂O₂ consumption) was found to be ≈ 40.8 US \$/m³.

5. CONCLUSIONS

In this study, the effects of current density, pH, H₂O₂ concentration and distance between electrodes on COD and color removal from leachate by the photo-electro-Fenton method were investigated. While maximum 83.84% COD removal were obtained under optimum conditions (initial COD: 8150 mg/L, anode: iron, cathode: steel-304, pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV lamp: 16 W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm, treatment time: 90 minutes), under similar conditions (initial color: 1905 Pt-Co, anode: iron, cathode: steel-304, pH: 3, current density: 10 A/m², H₂O₂: 1000 mg/L, UV lamp: 16 W, stirring rate: 250 rpm, distance between electrodes: 1.0 cm, treatment time: 45 minutes), the achieved maximum color removal was 84.46%. The [H₂O₂]/[Fe²⁺] molar ratios were calculated as 8.53 and 17.06 for maximum COD and color removal, indicating much more H₂O₂ consumption for color removal. Total treatment cost was calculated as 309.83 TL/m³ ≈ 41.3 US \$/m³ for COD removal, and 306.11 TL/m³ ≈ 40.8 US \$/m³ for color removal. The formation of more hydroxyl radicals in the reactor with the effect of UV light will reduce the need for H₂O₂ to be added externally. However, according to the standards given for the discharge of leachate in Turkey (COD: max. 700 mg/L, color: max. 280 Pt-Co); the photo-electro-Fenton used in the present study method was not found to be very enough for removing both COD and color from leachate. Therefore, the application of photo-electro-Fenton with other treatment methods as pre-treatment or post-treatment will be more effective for achieving more high COD and color removal efficiencies in the treatment of leachate containing high amounts of pollutants.

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