COD removal from leachate by electrocoagulation process: treatment with monopolar electrodes in parallel connection
Mehtap Tanyol, Aysenur Ogedey and Ensar Oguz

ABSTRACT
This study examines the removal of chemical oxygen demand (COD) from landfill leachate generated from the municipal landfill site of Bingol, Turkey. The effect of parameters such as current density, pH, and inter-electrode distance during the electrocoagulation (EC) process on COD removal of the process was investigated. Moreover, for COD removal, the energy consumption and operating costs were calculated for iron electrode under the EC conditions. COD removal efficiency was 72.13% at the current density of 16 mA m⁻², pH of 8.05, and the inter-electrode distance of 9 mm at the detention time of 60 min with iron electrode and the COD concentration was reduced from 6,100 mg L⁻¹ to 1,700 mg L⁻¹ by EC. The highest value of the electrical energy and electrode consumptions per kg of COD in the optimum conditions were determined as 0.055 kWh kg⁻¹ COD and 3.43 kg kg⁻¹ COD and the highest operating cost value was found to be 1.41 US$ kg⁻¹ COD for 0–60 min time intervals.

INTRODUCTION
From the beginning of the last century, in urban cities of developing countries, growing populations, economic growth and increase in community living have expedited the generation rate of solid waste (Al-Khatib et al. 2010). Landfilling is a frequently used method for eliminating solid wastes in urban areas of low- and middle-income countries (Meunier et al. 2006). The main disadvantage of this method created during their operation display a danger to the environment as they may transport toxic contaminants to water resources. Leachate is the most challenging wastewater with a high content of dissolved organic matter (DOM), and may go out of the fill with aqueous surroundings. As is well known, leachate contaminated groundwater must be addressed. Therefore, leachate treatment is limited due to the contamination of leachate variable composition and high amount of refractory compounds (containing organic and inorganic matters, micropollutants like as xenobiotic organic chemicals and heavy metals) (Labanowski et al. 2010). These are affected by the style of waste and the age of the landfill. Moreover, this feature is affected by climate and precipitation conditions. Leachate produced from landfills can be categorized into young leachate and old leachate. Young leachate (i.e. with less than 5 years) is characterized by low molecular weight distributions and high organic content (chemical oxygen demand (COD) of >10,000 mg L⁻¹; ammonia-nitrogen (NH₃-N) between 500 and 6,000 mg L⁻¹; biochemical oxygen demand (BOD₅)/COD ratio of 0.4–0.7; and COD/NH₃-N ratio above 10:1); while old leachate (i.e. with more than 10 years) is generally characterized by low organic compounds (BOD₅/COD ratio of less than 0.1) due to the fact that these compounds are refractory to biodegradation (Riera-Torres & Gutierrez 2010; Wang et al. 2016; Ye et al. 2016; Zhong et al. 2017). Leachate removal techniques are categorized in four main classes: in the first of these, leachate is transferred by recycling or combined treatment with domestic wastewater. Second is biodegradation using aerobic or anaerobic processes. The third uses physico-chemical methods: sedimentation/ flotation and air stripping, chemical oxidation, Fenton oxidation, coagulation/flocculation, adsorption, and chemical precipitation. The last one uses membrane processes (Bouhezila et al. 2011). Recently, physico-chemical techniques have been the most widespread used in the treatment of landfill leachate. Among these, coagulation is a simple and widely used
method under suitable chemicals and ambient conditions (Zhao et al. 2011). During this process, rapid mixing of the coagulant chemicals with the wastewater follows by flocculation and settling. Unfortunately, coagulation has some disadvantages such as coagulant requirement, corrosion problems due to reducing pH, and problems with high produced sludge (Karhu et al. 2012). Currently, chemical coagulation (CC) is not used due to disadvantages such as high costs of repair and problems of sludge processing (Vasudevan et al. 2009; Bouhezila et al. 2011). Compared with classical CC, electrocoagulation (EC) is a better technology for phase separation that relies on the extrication of cations from the disintegration of suitable sacrificial anode via reactions for the removal of pollutants from wastewaters. Therefore, this process is implemented efficiently to various water and wastewater treatment problems (Canizares et al. 2008; Emamjomeh & Sivakumar 2009).

EC is not a new technology (Holt et al. 2005). Fundamental aspects of the EC process for studies that are targeted interactions and relationships between the three principles are electrochemistry, coagulation, and flocculation. According to the green chemistry, electrochemical technologies are promising methods because of its simple and efficient mechanism and operation (Siedlecka et al. 2012), for different types of wastewater such as textile effluents (Saravanan et al. 2010), urban wastewaters, and from metalworking industry (Canizares et al. 2008), paper industry, and tannery industry (Canizares et al. 2008; Bouhezila et al. 2011). It is known that the EC process widely uses aluminum or iron as electrodes and their cations are soluble in sacrificial anodes during the application of a direct current (El-Taweel et al. 2015). The metal hydroxides formed have a tendency to coagulate the liquid impurities, and the hydrogen bubbles created at the cathode allow flotation of foam containing the organic matter. This process mainly consists of three successive stages: (i) production of metal ions by electrolytic reaction at anode (ii) the hydrogen bubbles created by an electric current at the cathode whereby formation of coagulants and destabilization of the contaminants takes place (iii) formation of the flocs from destabilized phases (Khemis et al. 2006; Ghernaout et al. 2009; Saravanan et al. 2010; El-Taweel et al. 2015). The main reactions occurring in an EC cell can be described by the following equations.

At the anode, metal is oxidized into cations:

\[
\text{Fe(s)} \rightarrow \text{Fe}^{2+}_{(aq)} + 2e^{-} \quad (1)
\]

\[
\text{Fe}^{2+}_{(aq)} + 2OH^{-} \rightarrow \text{Fe(OH)}_{2(s)} \quad (2)
\]

At the cathode, water is reduced into hydrogen gas and hydroxyl anions:

\[
2\text{H}_2\text{O} + 2e^{-} \rightarrow \text{H}_2(g) + 2\text{OH}^{-}_{(aq)} \quad (3)
\]

In the bulk solution:

\[
\text{Fe}^{2+}_{(aq)} + 2\text{OH}^{-}_{(aq)} \rightarrow \text{Fe(OH)}_{2(s)} + \text{H}_2(g) \quad (4)
\]

The objective of this work is the COD removal from raw leachate samples and to determine the influence of several parameters such as current density, pH, and inter-electrodes distance using the EC process. In addition, optimal operating cost analysis with respect to the electrical energy and electrode was calculated.

**MATERIALS AND METHODS**

**Leachate source and characteristics**

The municipal landfill site of Bingol, Turkey is approximately 13 km away from the city center. Activity area in the direction of Bingol-Mus (east of the province) has been operating since 2013 (active landfill) and covers a total surface area of 45,000 m² (available capacity of 100,000 m³). The leachate used in this study pumped through the bottom of the landfill pool. The samples in a closed container were stored at 4 °C temperature. The characteristics of raw leachate are presented in Table 1.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Raw Leachate</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>8.05 ± 0.25</td>
</tr>
<tr>
<td>Conductivity (mS/cm)</td>
<td>18.92 ± 3.12</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>314 ± 26</td>
</tr>
<tr>
<td>SS (mg L⁻¹)</td>
<td>175 ± 20</td>
</tr>
<tr>
<td>Total dissolved solids (mg L⁻¹)</td>
<td>7,644 ± 152</td>
</tr>
<tr>
<td>Color₅₅₅ (m⁻¹)</td>
<td>900 ± 56</td>
</tr>
<tr>
<td>Color₅₂₅ (m⁻¹)</td>
<td>705 ± 49</td>
</tr>
<tr>
<td>Color₄₅₀ (m⁻¹)</td>
<td>570 ± 45</td>
</tr>
<tr>
<td>BOD₅ (mg L⁻¹)</td>
<td>3,100 ± 148</td>
</tr>
<tr>
<td>BOD₅/COD</td>
<td>0.517 ± 0.12</td>
</tr>
<tr>
<td>Chloride (mg L⁻¹)</td>
<td>1,049 ± 85</td>
</tr>
<tr>
<td>TKN (mg L⁻¹)</td>
<td>8,960 ± 320</td>
</tr>
<tr>
<td>NH₃-N (mg L⁻¹)</td>
<td>1,626 ± 110</td>
</tr>
</tbody>
</table>

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Electrochemical reactor cell

The design of the batch monopolar EC reactor used in the present study is shown in Figure 1. The electrochemical reactor cell was made from Plexiglas material and is 1 L in volume. The electrodes sets (anode and cathode) consisted of four parallel pieces of iron (Fe) and aluminium (Al) plates each, with a dimension of 5 cm (width) \times 6 cm (length) \times 2 mm (depth), having a surface area of 30 cm$^2$. The electrodes were established on a perforated plastic plate placed in the cell. The batch monopolar electrochemical cell consisted of four electrodes that were all separately connected to the DC power supply (AA Tech ADC-3303D, 0–3 A, 0–60 V). Two anodes and two cathodes were used in the electrode assembly. The two Fe electrodes connected to the positive output of the power supply were consumed during the experiments while two Al cathodes, which were insoluble, were connected to the negative outlet of the power supply. A working volume of 500 mL of leachate was used for all experiments. Samples of 10 mL were drawn at 15, 30, 45, and 60 min intervals and monitored for residual COD concentration.

![Figure 1](https://iwaponline.com/wst/article-pdf/77/1/177/211756/wst077010177.pdf)

The electrochemical reactor of experimental design.

Experimental techniques of landfill leachate

All chemical compounds used in the study were of analytical grade. Physicochemical parameters such as conductivity and pH were measured using a pH meter (Thermo Orion 420A), while turbidity was measured with a digital portable turbidimeter (HACH 2100P). COD was determined with the use of closed reflux method. BOD$_5$ was fixed by a 5-day BOD$_5$ test according to standard methods. The color was measured according to standards of European Norm EN ISO 7887 using a UV/Vis spectrophotometer (Shimadzu 1800 UV/Vis). Measurements of total solids (TS), suspended solids (SS), ammonia nitrogen (NH$_3$-N), total Kjeldahl nitrogen (TKN), and chloride ion concentrations were also performed by the standard methods (APHA 2005).

Batch EC experiments

All EC experiments were conducted in a batch reactor with a magnetic stirrer using 500 mL of leachate. The pH was adjusted by adding 1 N sulfuric acid solution or 1 N sodium hydroxide solution for each experiment. Iron and aluminium electrodes with total area of 30 cm$^2$ were used.
as the anode and the cathode, respectively. The experiments were performed at room temperature (25°C ± 2°C) and without any electrolyte addition. Before each study, electrodes were immersed in the dilute sulfuric acid solution to remove any adherent oxides or impurities on the electrode surface and then washed with distilled water and dried for the next use. The magnetic stirrer was placed above the cell and set at constant 200 rpm. During the experiments, 10 mL sample was collected using a peristaltic pump (Filitec, BT100) at different time intervals from the electrochemical cell for analysis. Operational time was selected as 60 min. Consequently they were analyzed for residual concentrations of COD. The current densities studied were 8, 12, and 16 mA m⁻². The efficiency of the COD removal was analyzed at four pH values 4, 6, 8, and 10. Finally, the impact of inter-electrodes distance on the process was investigated at 3, 6, 9, and 12 mm. After sedimentation for 45 min, the supernatant was taken for COD analysis.

RESULTS AND DISCUSSION

The effect of current density on removal of COD

The effect of current density on removal of COD was investigated using three different current densities. Results are shown in Figure 2.

From Figure 2(a), it can be seen that the percentage of COD removal was comparatively low in the current densities of 8 mA m⁻² and 12 mA m⁻². Figure 2(a) demonstrates that the percentage of COD removal has increased by increasing the current density. At the same time, in Figure 2(b), quite low removal efficiencies were obtained in the experiments where low current density values (8 and 12 mA m⁻²) were applied. This situation can be caused by the minimum solubility of iron hydroxide in these current densities (Pulkka et al. 2014). It is clear that most of the particles are negatively charged in water, while alike charges avoid aggregation through electrostatic propulsion (Fernandes et al. 2015). It could be seen that the COD of raw leachate decreased to 2,000 mg L⁻¹ from its initial concentration of 6,100 mg L⁻¹ during EC. This decrease may be because of the Fe²⁺ ions released by anodic dissolution destabilizing to form oxhydroxides that are beneficial coagulants. Increasing current density speeds up the removal of pollutants due to the increase of bubble size and the flotation of H₂ (Wang et al. 2016). It can be seen from Figure 2(b) at 15 min and 60 min that COD removal efficiencies were 18.03% for 8 mA m⁻² and 20.49% for 12 mA m⁻². When the current density comes to 16 mA m⁻², COD removal reached 67.21%. According to the results, the current density of 16 mA m⁻² was selected as the best condition for the EC experiment. Because of the well-known influence of current density on the Fe electrode, significance of this parameter was commonly highlighted in landfill leachate treatment by some authors. Oumar et al. (2016) studied treatment of landfill leachate using integration of biofiltration (BF) and EC processes for the treatment of sanitary landfill leachate. Different current densities (3 mA cm⁻²–30 mA cm⁻²) and also treatment times (5, 10, 15, 20, and 30 min) were investigated. COD removal results are as follows: low COD removal (29%) was obtained after 30 min of EC treatment with low current density (3 mA cm⁻²). For higher current densities (10 mA cm⁻²–30 mA cm⁻²), a rapid decrease of residual COD concentration was achieved during the first 10 min (69–73% of total COD removal) and after that, the residual COD was decreased.
slowly until the end of the experiment. Based on these results, the current density of 10 mA cm$^{-2}$ and 30 min were selected as the best conditions for the EC treatment. Oumar et al. (2017) studied the effects of anode type, current density, and treatment time on the efficiency of the EC process for organic matter removal. The rate of COD removal was relatively high while current density was 5 mA cm$^{-2}$ for Fe electrodes in the EC process. At this current density, over 90% of total COD was removed within the first 15 min. After that, COD decreased very slowly until the end of the experiment. For the next step of the experiments, the following operating conditions were chosen: 8 mA cm$^{-2}$ and 20 min for iron. Under these conditions, 70% and 65% of the residual COD was removed using an iron anode. Li et al. (2011) also examined the removal efficiency of COD from landfill leachate by EC. The investigated current densities were 1.98, 2.98, 3.97, 4.96, and 5.95 mA cm$^{-2}$. The effect of current density was on the removal efficiencies by the next conditions: 6.8 pH and 30 min electrolysis time for Fe electrode. The removal efficiency of COD at current density of 1.98 to 4.96 mA cm$^{-2}$ increased from 23.6 to 93%, but it remained virtually unchanged considerably at higher current density conditions.

The color and NH$_3$-N values obtained from the experiments are presented in Table 2. According to Table 2, the removal efficiency of color and NH$_3$-N were evaluated under different EC conditions. Current density, pH, and the electrode material are very important parameters in the EC process. For NH$_3$-N removal, the maximum efficiency of the reactor was obtained at the current density of 16 mA m$^{-2}$, pH of 10, inter-electrode distance of 6 mm, and treatment time of 60 min. It may be because NH$_3$ is the predominant specie in the alkaline medium (pH ≥ 10). In this case, ammonia adsorbs at the electrode surface at higher potential values, and is oxidized to give N$_2$, while NH$_4^+$ dominant species at pH ≤ 7 electrochemically decomposed to produce nitrogen (Li et al. 2011). Another result can be seen in Table 2 that the maximum color removal efficiencies were obtained in the experiments at current density of 12 mA m$^{-2}$, pH of 8.05, inter-electrode distance of 12 mm, and treatment time of 60 min. For the initial values of pH 4 and 6, color removal efficiencies were determined as 44.44% and 16.67%, respectively, in our study. It is understood from the results that NH$_3$-N removal increases by increasing the pH values; however, color removal efficiencies of the EC process is quite limited at decreased pH values.

### Operating of process cost assessment

In the EC process, the quantity of dissolved metal is connected to the quantity of electricity penetrated by the solution (Ye et al. 2016). One of the most important parameters that must be detected to appraise a method of wastewater treatment is the cost of electrochemical experiments. The overall cost is based substantially on the factors of containing material (mainly electrodes) cost and operating cost (mainly electrical energy). Generally, energy consumption analysis can be calculated as (Li et al. 2011):

$$E = U \times I \times t_{EC}$$  \hspace{1cm} (5)

where $E$ is the electrical energy consumed (kW h), $U$ is the cell potential (volt), $I$ is the current (A), and $t_{EC}$ is the time of EC process (h). The current efficiency ($\Phi$) of the EC method related to the production of Fe$^{2+}$ ions was calculated with the following equation:

$$\Phi = \frac{\Delta m_{exp}}{\Delta m_{theo}} \times 100$$  \hspace{1cm} (6)

In this equation, $\Delta m_{exp}$ is based on the comparison of experimental weight loss of electrodes during the EC process. Also, $\Delta m_{theo}$ is the theoretical amount of metal dissolution dissolved in the solution by induced current in the electrolysis cell and can be calculated according to Faraday’s law:

$$M_{theo} = \frac{M_w \times I \times t_{EC}}{z_F \times F}$$  \hspace{1cm} (7)

### Table 2 | Effect of current density, pH and inter-electrode distance on color and NH$_3$-N removal efficiencies by EC

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Current density (mA m$^{-2}$)</th>
<th>pH</th>
<th>Inter-electrode distance (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>8</td>
<td>12</td>
<td>16</td>
</tr>
<tr>
<td>Color$_{re}$ (%)</td>
<td>83.89</td>
<td>97.78</td>
<td>95.0</td>
</tr>
<tr>
<td>NH$_3$-N (%)</td>
<td>4.98</td>
<td>6.64</td>
<td>9.41</td>
</tr>
</tbody>
</table>

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where $M_{\text{theo}}$ is the electrode consumption (kg m$^{-3}$), $M_w$ is the molecular mass of iron (55.85 g mol$^{-1}$), $z$ is the number of electron transferred ($z_{\text{Fe}} = 2$), $I$ is the current (A), and $F$ is Faraday’s constant (96,485 C mol$^{-1}$). In this instance, the operating cost relied on the electrode material and electrical energy was calculated using Equation (8) (Canizares et al. 2008; Palahouane et al. 2015).

Operating cost = a $C_{\text{energy}}$ + b $C_{\text{electrode}}$  

Coefficient (a) was given by the National Office for Water and Electricity according to the Turkish market in July 2016 was 0.110 US$/kWh$, and (b) prices for Fe electrode material obtained by the national market (2016) is 3.074 US$/kg$, respectively (Demirbas & Kobya 2017; Elazzouzi et al. 2017). $C_{\text{energy}}$ and $C_{\text{electrode}}$ were calculated from Equations (9) and (10) to find out the energy consumption per kg of COD removed:

$$C_{\text{energy}} (\text{kWh/kg COD}) = \frac{U \times I \times t_{EC}}{V \times C_i \times R_e}$$  

(9)

$$C_{\text{electrode}} (\text{kg/kg COD}) = \frac{I \times t_{EC} \times M_w \times \Phi_{\text{Fe}}}{z \times F \times V \times C_i \times R_e}$$  

(10)

where $V$ is volume (m$^3$) of the wastewater, $C_i$ (kg m$^{-3}$) is COD concentration of wastewater in the input stream, and $R_e$ is COD removal efficiency. Figure 3 shows the energy consumption per kg of COD in the EC process. Figure 4 demonstrates the influence of operating costs using the electrochemical method to remove COD from leachate.

According to Figure 3, energy and electrode consumptions per kg of COD to current density 16 mA m$^{-2}$, optimum pH 8.05 and different times (0, 15, 45, and 60 min) were determined as 0.027 kWh kg$^{-1}$ COD and 0.042 kg kg$^{-1}$ COD, 0.031 kWh kg$^{-1}$ COD and 0.096 kg kg$^{-1}$ COD, 0.05 kWh kg$^{-1}$ COD and 2.35 kg kg$^{-1}$ COD, 0.055 kWh kg$^{-1}$ COD and 3.45 kg kg$^{-1}$ COD, respectively. In this case, the energy and electrode consumptions increased with the increase in the operating time from 15 to 60 min. Figure 4 shows the cost estimation analysis of the EC as a function of time.

As seen in Figure 4, operating cost values were 0.17 US$ kg$^{-1}$ COD, 0.40 US$ kg$^{-1}$ COD, 0.97 US$ kg$^{-1}$ COD, 1.41 US$ kg$^{-1}$ COD for 0–60 min time intervals, respectively. It is known that the operating cost of COD removal is increased due to the electrolysis durations.

**Effect of pH on removal COD**

A very important factor in the EC process is the operating pH value influencing the performance on pollutant removal. The effect of different initial pHs on the removal of COD is shown in Figure 5. The influence of pH under acid, alkaline, and natural conditions on the efficiency of leachate of pH 4.0, 6.0, 8.05 (natural pH) and 10.0 was investigated at current density of 16 mA m$^{-2}$.

As shown in Figure 5(a), the percentage of COD removal increased with increasing pH from 4 to 8.05, but decreased at pH 10. COD removal efficiencies after 60 min of electrolysis for the different pH values are shown in Figure 5(b). The removal efficiencies of COD are represented as function of time.

Figure 5(b) shows that the COD removal reached from 37.71% to 65.57% when pH was increased from 4 to 6 and then decreased to 54.01% at pH 10. It was found that the highest removal efficiency of COD was obtained at natural pH conditions, whereas in acidic or alkaline ambient the efficiency of COD removal decreased. These results could
be explained by the Fe(OH)$_3$ flocs under acidic conditions (initial pH 4–6) and depends on the initial pH. Fe(OH)$_3$ coagulate occurs in stronger acidic media than Fe(OH)$_2$. Considering this, for the Fe electrode, the quantity of the insoluble iron hydroxide drastically increased with pH ranging from 4 to 7. While the pH increase in the acidic medium was due to the formation of hydrogen at the cathode, the pH fall was due to the formation of hydroxide precipitates that were released near the anode and secondary reactions such as water oxidation and chlorine production and hydrolysis (Fernandes et al. 2014). At high pH values (pH > 9.0) a part of Fe(OH)$_3$ was dissolved as Fe(OH)$_4$ and a small amount of pollutants could be separated (Pulkka et al. 2014). Similar results were obtained by a number of authors. According to An et al. (2017) the initial pH was an essential factor for the EC process. The authors studied the effect of initial pH (between 3 and 11) to determine the removal efficiency of COD from wastewater. Different pH conditions create distinct kinds of flocs. The green Fe(OH)$_2$ flocs were constituted at acidic pH, while the green Fe(OH)$_2$ and yellow Fe(OH)$_3$ colloids were generated at neutral pH, and the red-brown Fe(OH)$_3$ flocs occurred at alkaline pH. EC treatment was more effective under alkaline conditions because the Fe(OH)$_2$ was less efficient than Fe(OH)$_3$ in flocculation or enmeshment.

Demirbas & Kobya (2017) studied treatment performances and operating costs of real metalworking fluid wastewater (MWFW) using CC and EC processes. They observed the effect of initial pH in the wastewater on the removal of COD within the pH range of 3–8 at a current density of 80 A m$^{-2}$ and an operating time of 25 min in the EC process. Between 60 and 90% of COD removals at pH of 3–7 were obtained with the Fe electrode in the EC. Maximum COD removal efficiency was observed at pH 7 for the Fe electrode. Fernandes et al. (2014) investigated the influence of the pH on the removal of COD in combined EC/electro-oxidation processes. EC experiments were performed with Fe electrodes, and the impact of the initial pH and electrolysis time was studied. EC trials at initial pH 6 or the natural pH (8.5) for a period of 2 or 3 h at an implemented current intensity of 2.5 A were performed. According to the results, COD removal at an initial pH 6 was appropriate for the process performance for 2 h of EC. However, for 3 h EC experiments, the effect of pH was insignificant as

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**Figure 5** | Effect of initial pH on (a) COD concentration, (b) COD removal, and (c) medium pH as function of time (initial COD = 6,100 mg L$^{-1}$, current density = 16 mA m$^{-2}$, inter-electrode distance = 12 mm, stirring speed = 200 rpm, temperature = 25 °C, V = 0.5 L).
the amount of COD remained unchanged for both of the initial pH values examined. Linares-Hernandez et al. (2010) investigated the eradication of permanent organic compounds in industrial wastewater using a mix of EC and electrooxidation processes. A batch monopolar electrochemical reactor was constructed with Fe electrodes for the EC stage. A current density of 200–800 A m$^{-2}$ was applied at a range of pH values between 2 and 12. After the experiments, the COD removal rate increased as the pH increased from 2 to 8, but decreased for higher values. These results indicated that the COD removal was similar to previous studies, which decreased dramatically at pH values greater than 10. The time taken to reduce COD shortened by about half (from 900 to 425 mg L$^{-1}$) with EC lasting approximately 30 min. The reduction of COD as a function of time was achieved under optimum conditions of pH 8 and current density of 800 A m$^{-2}$.

**Effect of inter-electrode distances removal of COD**

During EC, the distance between the electrodes is very important not only for the electrode assembly but also for the required electrode area. In a reactor formed as monopolar electrodes in a parallel connection (MP–P), COD removal efficiency varies with the distance between the electrodes. To determine the impact of electrode distances on the process, the reactor was arranged 3, 6, 9, and 12 mm apart under the same experimental conditions. The influence of inter-electrode distances on COD removal during EC is shown in Figure 6.

According to Figure 6(a), as the distance between the electrodes increases, the COD removal increased owing to the amount of metal ions dissolved. The differences of COD removal efficiency after 60 min of EC are presented in Figure 6(b). The COD removal efficiency obtained using varied electrode distances is shown in Figure 6(b). At the beginning of the EC, anodic oxidation occurs with the current applied to the electrodes. Depending on the time, metal electrolytes on the anode will form a very thin film and an extra resistivity, which will be increased as the electrode distance increases. In this case, ohmic losses will occur and inhibit the anodic oxidation (Ghosh et al. 2008; Raju et al. 2008). The results from the present study indicate that with the increment of inter-electrode distance, the percentage of COD removal also increased. COD removal efficiencies were 38.53% for 3 mm, 61.48% for 6 mm, and 72.13% for 9 mm. The initial COD concentration (6,100 mg L$^{-1}$) was reduced to 1,700 mg L$^{-1}$ by EC for 9 mm. However, the COD removal rate decreased to 67.21% as the inter-electrode distance increased from 9 to 12 mm. Similar results have been discussed by a number of authors. Ghosh et al. (2008) reported that at higher inter-electrode distances, alongside the interval of collection of suspended particles, adsorption of contaminants would be low. Bouhezila et al. (2011) investigated the effect of the inter-electrode interval on removal of COD with the EC process. It was observed that the distance between electrodes were ranging from 0.5 to 2.8 cm, the removal of COD increased by 10% after 15 min of treatment time. When the electrode distance was 2.8 cm, the COD removal ratio increased about 70%. Ricordel & Djelal (2014) investigated the effect of the electrode gap in the EC treatment on leachates at two distinct electrode gap values as 2–4 cm apart for both the lowest (25 A m$^{-2}$) and highest (95 A m$^{-2}$). These authors have found that after 210 min EC, the differences of COD removal efficiency were not substantial.
between 2 and 4 cm gaps for both current densities tested. They also found that increasing the electrode gap increased the power consumption per COD removal for the required current supply. Therefore, they preferred that the electrode gap value for this treatment was 2 cm. Zhang et al. (2013) studied the effect of distance between electrodes for total phosphate (TP) removal. According to the results of experimental conditions and measured parameters, TP removal efficiency was increased by increasing the electrode gap from 15 to 25 mm, but then it decreased with further increase in the inter-electrode distance. Terrazas et al. (2010) studied the impact of the gap between anode and cathode for turbidity removal. The preferred gap values were 2 and 3 cm for this treatment. The results showed an increase in energy consumption (ranging from 0.68 kWh m$^{-3}$ to 1.16 kWh m$^{-3}$) with an increase in cell potential, but the percentage of turbidity removal decreased with an increase in gap rise. The turbidity increased initially and started to decrease after 5 min, but the highest turbidity removal (92% in the present case) was achieved after just about an hour, in contrast to the mini-cell, which only demanded 30 min.

**CONCLUSION**

In many parts of engineering, the EC process is used to efficiently remove the organic or inorganic pollutants. Our study evaluated the effectiveness of the EC process for the removal of COD from the municipal landfill site of Bingol, Turkey. The influence of several parameters such as current density, pH, and inter-electrodes distance on the percentage of COD removal were investigated. The results indicate that the percentage of COD removal increased when the current density rises from 8 to 16 mA m$^{-2}$. The optimum current density was obtained at 16 mA m$^{-2}$ at the electrolysis time of 60 min. COD removal at four pH values (4, 6, 8.05, and 10) was investigated during 60 min of EC time. Even though the highest removal of COD was formed at pH 8.05, as the pH rises to 10, removal rate was lower due to a number of complicated reactions around electrodes at high alkaline pH. In addition, inter-electrode distance changing from 3 to 12 mm had a great effect on COD removal rate. The COD removal efficiency increases with electrode gap ranging from 3 to 9 mm, and decreased with a further increase in the inter-electrode distance up to 12 mm. The optimum electrode gap was chosen as 9 mm. According to these results, the COD removal rate of 72.13% was obtained at the optimum current density of 16 mA m$^{-2}$, optimum pH of 8.05 (natural pH), and optimum inter-electrode distance of 9 mm using EC process. The highest value of energy and electrode consumptions per kg of COD were determined as 0.055 kWh kg$^{-1}$ COD and 3.43 kg kg$^{-1}$ COD at optimum conditions. In this case, the energy and electrode consumptions increased with an increase in the operating time from 0 to 60 min. The highest operating cost value for COD removal efficiencies by the EC process were observed at 1.41 US$/kg COD for 0–60 min time intervals.

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