

Treatment of Landfill Leachate via Electrocoagulation and Electro-Fenton Processes: A Comparative Study

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ABSTRACT: *This study aims to compare the effectiveness of Electrocoagulation (EC) and electro-Fenton (EF) processes in the treatment of high-strength storage leachate. The effect of operating parameters, including initial pH, contact time, and mass ratio of COD: H₂O₂, on Chemical Oxygen Demand (COD) and Total Organic Carbon (TOC) removal efficiencies of leachate was investigated. For this purpose, a jacketed reactor in which the electrochemical process is performed using monopolar-bonded iron electrodes is designed. As a result of the experimental studies, the optimum operating parameters of the EC process were determined as follows: pH 7, current density 150A/m² and reaction time 30 minutes. Under these conditions, 37.87% COD and 47.36% TOC were removed respectively. Due to the lack of expected results in the treatment of this wastewater in the EC process, treatability studies were carried out with the EF process. As a result of the study, optimum conditions were found to be pH 3, current density 150 A/m², H₂O₂= 500 mg/L (KOl: H₂O₂≅1), and working time 10 min. A higher COD (71.7%) and TOC removal (90.87%) have been obtained with the EF process under optimum conditions. The operating costs of electrocoagulation and electro-Fenton processes under optimum conditions were calculated as 2.26 and 1.78 €/m³, respectively. Experimental findings revealed that, unlike the EC process, the EF process can be a good option for landfill leachate treatment in terms of providing less treatment time, less sludge, more cost-effectiveness, and necessary discharge limits.*

KEYWORDS: *COD; Electrocoagulation; Electro-Fenton; Landfill leachate; TOC.*

INTRODUCTION

Leachate is a mixture of important organic and inorganic pollutants, including resistant components, xenobiotic organic compounds, inorganic micropollutants, heavy metals, and other toxic substances, and contains a variety of pathogens that can potentially contaminate ground and surface waters [1,2]. The leachate composition ranges in the leachate pollution investigations are presented in Table 1 [3-8].

The amount and composition of landfill leachate are affected by waste type and compaction, landfill hydrology, climate, and particularly landfill age [9,10]. The biggest risk associated with leachate production is the contamination of surface waters. Therefore, storage leachate must be treated to meet local receiver discharge standards for toxicity or adverse environmental effects [11, 12].

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Table 1: Leachate composition ranges

Parameter	pH	COD (mg/L)	BOD (mg/L)	TKN (mg/L)	SS (mg/L)	NH ₄ ⁺ -N (mg/L)	Cl ⁻ (mg/L)	SO ₄ ²⁻ (mg/L)	BOD/COD
Range	5-11	100- 71000	3-25000	0.2- 13000	13-5000	10-13.000	150-5000	10-8000	0.04-0.7

(COD: Chemical Oxygen Demand, BOD: Biological Oxygen Demand, TKN: Total Kjeldahl Nitrogen, SS: Suspended Solids, NH₄⁺-N: Ammonium Nitrogen, Cl⁻: Chloride, SO₄²⁻: Sulphate)

Table 2: Limit values for direct discharge of leachate or for discharge of leachate to surface water after in-situ treatment

Parametres	Turkey		Netherlands	Germany	Ireland	France
	a	b				
pH	6-9	6-9	-	-	7.52	-
Suspended Solids (SS) (mg/L)	^a 200	^b 100	30	20	-	100
Oil and Grease (mg/L)	^a 20	^b 10	-	-	-	-
Chemical Oxygen Demand (COD) (mg/L)	^a 700	^b 500	75-150	200	141	120
Total Kjeldahl Nitrogen (TKN) (mg/L)	^a 20	^b 15	20	70	261	30
Adsorbable Organic Halogens (AOX) (mg/L)	-	-	-	0.5	-	-
Total Phosphorus (TP) (mg/L)	^a 2	^b 1	-	3	-	-

(a: Composite sample 2 hours b: Composite sample 24 hours)

A suitable treatment method should be efficient, easy to use, inexpensive, and environmentally friendly, especially in terms of operating and maintenance costs, producing less energy and less sludge. The techniques applied should be practical for treating large volumes of stabilized leachate [13,14]. There are many pollutant indicator parameters such as Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD), and total organic carbon (TOC), which are of great importance for the type of treatment technology to be applied. The increase in non-degradable organic leachate components especially residual COD) is mainly a function of dilution. OD₅/COD and COD/TOC ratios are common indicators of the biodegradability of organic compounds and oxidized organic carbon [15,16]. The EU, as well as certain European countries, has limits for the discharge of leachate for periods. Council Directive 1999/31/EC on the landfill of waste [17], Waste Framework Directive 2008/98/EC [18], Council Directive 91/271/EEC concerning urban wastewater treatment [19], and Water Framework Directive 2000/60/EC [20] are among the major European regulations governing the storage and leachate management. In Turkey, the limit values stipulated for "Discharge of wastewater to infrastructure facilities and surface waters" specified in the Water Pollution Control Regulation are applied for the discharge of leachate to the city wastewater sewage system after pre-treatment [21]. The limit values of some parametric pollutants for direct discharge of leachate or for discharge of leachate to surface

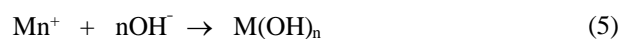
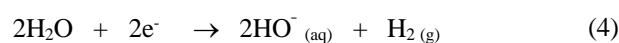
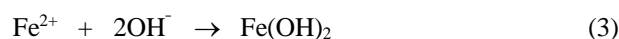
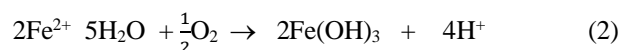
water after on-site treatment for Turkey and other European countries are presented in Table 2 [22-26].

For treatment of landfill leachate, coagulation [27,28], electrocoagulation [29,30], ozonation [31,32], adsorption [33,34], reverse osmosis [35,36], ion exchange [37,38], membrane processes [39,40], Fenton [41,42], electro-Fenton [43,44] and photo-Fenton [45,46], various treatment technologies have been applied, including advanced oxidation technical processes based on. It is difficult to obtain high treatment efficiency and effluent quality by using any of these methods alone. Combinations of two or more physicochemical treatments or biological treatment techniques are widely used in landfill leachate treatment. However, in the presence of toxic and persistent organic substances in wastewater, biological processes are unable to do their job. Although physicochemical methods are effective in the treatment of these wastewaters, they are not preferred because of the transfer of pollutants from one environment to another. However, these techniques have disadvantages such as high operating costs and low pollutant efficiency. Advanced oxidation processes, such as electrochemical treatment, are among the most effective treatment technologies for the removal of complex and degradation-resistant organic pollutants. In studies on leachate treatment, processes based on electrochemical processes seem to be the most effective approaches with low operating costs and high treatment efficiency when compared to traditional processes [47-50]. In the treatment of strong wastewater such as leachate due to the presence

of non-biodegradable pollutants, their complex structure, and high pollutant concentrations, studies in which electrochemical methods such as electrocoagulation (EC) and Electro-Fenton (EF) carried out together have attracted great interest in recent years and good results have been obtained in the removal of polluting parameters because these processes can efficiently fractionate a range of organic substances [51-54].

EC is an electrochemical technology that combines the advantages of conventional coagulation, flotation, and adsorption in water and wastewater treatment [55]. The mechanism of EC depends on the chemical properties and conductivity of the medium. In addition, properties such as pH, size of colloidal particles in the environment, applied current density, type and concentration of electrolyte, spacing between electrodes, and concentration of chemical species also affect the EC process [56]. The process is dependent upon the dissolution of the anode electrode. Dissolved electrodes form a coagulant species that destabilizes pollutants in water and wastewater and traps them as suspended particles [57]. When an electric current is applied, positive ions move to the cathode, and negative ions move to the anode. Oxidation reactions occur at the anode, and reduction reactions occur at the cathode. Metal ions resulting from anode dissolution and subsequent hydrolysis, act as coagulants that help attenuate laden pollutants and cause them to agglomerate together [58]. Iron (Fe) and aluminum (Al) electrodes are the anode materials widely used in the literature in the electrocoagulation process due to the formation of multivalent ions, their low cost, availability, and efficiency due to various hydrolysis products [59,60]. Al and Fe ions produced at the anode react with the hydroxide ions produced at the cathode, forming various dissolved Al-Fe hydrolysis products (complexes) and Al-Fe (OH)_{3(s)} hydroxide solids [61]. When iron electrodes are used during the EC process, ferric hydroxide (Fe (OH)₃) is formed by the reactions given below (Eq. 1-2-3). Ion and H₂ gas are formed from the OH⁻ (Eq. 4) reaction at the cathode. In addition, H₂ produced at the cathode and O₂ produced at the anode cause flotation (electro-flotation) of the suspended particles, and insoluble particles precipitated by filtration (sedimentation) can be removed [62]. Electrogenerated metal cations destabilize the colloidal particles by forming polyvalent poly hydroxide complexes that promote aggregation, while hydrogen formation

at the cathode promotes the mixing of components and aggregation. Coagulation/flocculation of suspended solids leads to the formation and collapse of high-density flocs due to polyhydroxides [63,64]. Finally, in most of the solutions, the production of polyhydroxide proceeds in harmony depending on the anode material (M) used and the number of electrons (n) involved in the reaction (Eq. (5)) [65].



Electrochemical processes based on iron electrodes and iron catalysts include EC, EF, and a combination of both methods. Combination techniques reduce energy consumption compared to conventional EC [66,67]. Another electrochemical process related to EC is EF, in which the Fe electrode is used as the Fe²⁺ source and the other necessary reagent, hydrogen peroxide (H₂O₂), is added to the reaction from the outside. In fact, with the addition of hydrogen peroxide, the electrocoagulation process is converted to the EC process [68]. Therefore, subsequent reactions involving Fe²⁺, Fe³⁺, and H₂O₂ take place and hydroxyl radicals are formed. These radicals then attack organic compounds and rapidly decompose organic substrates in wastewater [69]. EF technology is one of the Electrochemical Advanced Oxidation Processes (EAOPs) that is of great interest for wastewater treatment due to its simple operation, relatively low cost, and high degradation efficiency against organic pollutants when compared with conventional wastewater treatment methods [70]. The reaction kinetics is mainly controlled by the hydrogen peroxide generation rate. Hydroxyl radicals can oxidize a wide variety of organic compounds [71]. In this process, continuous electrogeneration of H₂O₂ at a suitable cathode takes place (Eq. (6)) with the addition of an iron catalyst to the treated solution to produce bulk oxidant •OH via the Fenton reaction [72]. In the EF process, •OH radicals from both anodic and cathodic sources work together to deeply oxidize organic pollutants in wastewater,

Table 3: General characteristics of leachate

Parameters	Collection Pool Output Concentration (a)	Collection Pool Inlet Concentration (b)	Average (a)
pH	8-8.5	7.5-8.5	8.25
Conductivity (mS/cm)	7.5-9.5	5.5-11	8.5
Chloride (mg/L)	1650- 1750	1400-1500	1700
Oxidation Reduction Potential-ORP (mV)	40-(-120)	90-290	-
COD (mg/L)	1400-3000	370-500	2200
AOX (mg/L)	50±10	5±3	50
TOC (mg/L)	400-500	300-350	425
NH ₄ -N (mg/L)	256-290	207	273
NO ₂ -N (mg/L)	0.8- 1.01	0.7	0.9
NO ₃ -N (mg/L)	89-109	15	99



(a)

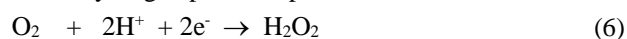


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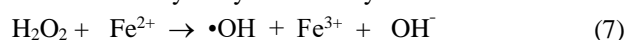
Fig.1: Landfill collection pool leachate inlet (b) raw leachate sample used in the experimental study

either through H atom abstraction or through the addition of OH to multiple bonds and intermediate radicals (Eq. (7)) [73]. Electrochemical reduction of Fe³⁺ on the cathode helps to regenerate the Fe²⁺ catalyst (Eq. 8) [74]. Finally, full mineralization is reached. These types of pollutants are broken down by oxidation and coagulation, which can reduce the amount of electricity used. One advantage of EF over the classical chemical Fenton reagent is that the Fe³⁺ ions formed by the Fenton Reaction (7) can be reduced at the cathode to regenerate Fe²⁺ ions according to the Reaction (8) [75].

In-situ hydrogen peroxide production at the cathode:



Production of hydroxyl radicals by Fenton reaction:



Regeneration of ferric ions at the cathode:



EC and EF processes and COD and TOC removal from real landfill leachate have not been studied much in the literature as a comparative study. In this study, leachate treatment, which is difficult to treat due to its complex structures and high pollutant concentrations, was

examined by using EC and EF techniques, which are one of the electrochemical treatment methods. The performances of the EC and EF processes were evaluated based on COD, TOC removal. The effects of experimental parameters such as initial pH, current density, and operating time on removal were investigated. Additionally, the operating cost-effectiveness of the sequential EC and EF process has also been analyzed. The results of this study aim to provide an important theoretical reference for improving the performance of processes used in the treatment of landfill leachate of the sequential EC-EF process.

EXPERIMENTAL SECTION

Leachate

In the study, 5 leachate samples were taken for reproducible sample analysis from the Sivas (Turkey) landfill leachate collection pool inlet and outlet, and experimental studies were performed on these leachate samples (Fig. 1ab). In the study, the characterization of the leachate samples taken from the Sivas (Turkey) landfill collection pool outlet and pool inlet is given in Table 3.

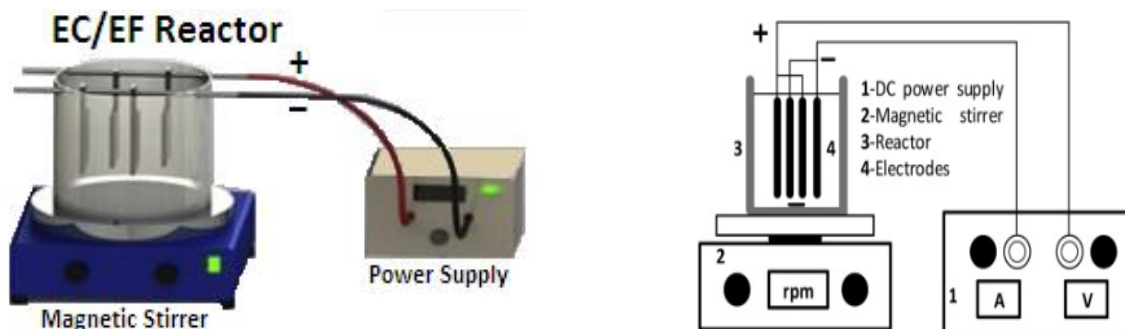


Fig. 2: Schematic view of the experimental setup

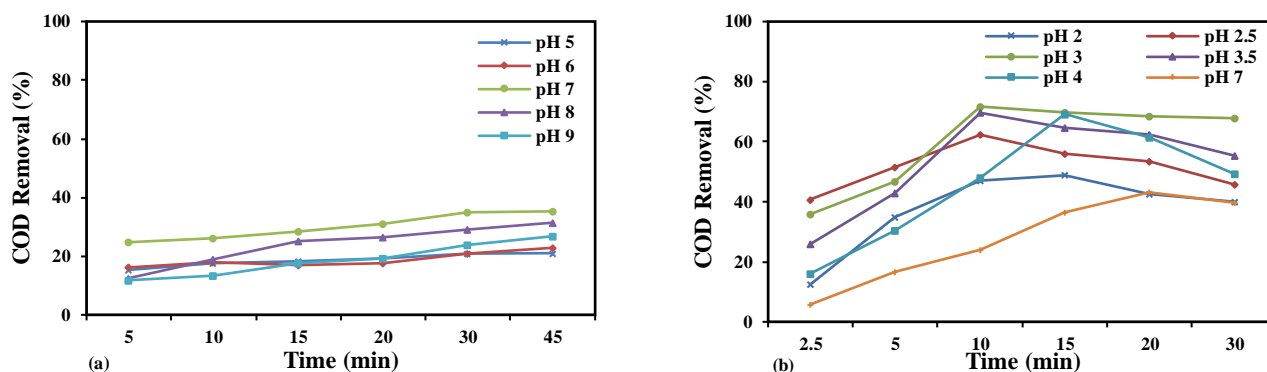


Fig. 3: The effect of pH on COD removal efficiency in EC (Fig a) and EF (Fig b) processes depending on electrolysis time (EC: current density 100 A/m^2 and electrolysis time 45 min, EF: 150 A/m^2 , H_2O_2 500 mg/L, electrolysis time 30 min)

Average values were taken as basis in wastewater samples taken from the outlet of the collection pool where leachate accumulated. When the COD and TOC values were examined, it was seen that these values were lower than the normal leachate characteristics when compared with the normal solid waste landfill leachate characterization mentioned in other studies. Thus, it is thought that the leachate coming into the collection pond is formed by the accumulation of water formed by precipitation and passes into the surface flow. As can be seen, the collection pool waste values are above the discharge standards with the average COD (2200 mg/L) and TOC (425 mg/L) concentrations. Also, landfill leachate has a high electrical conductivity value due to its high chloride (anion) concentration, which allows electrochemical oxidation without the addition of more electrolytes.

Electrochemical method and experimental study

The experiments were carried out in a 100x100x130mm double-walled reactor made of plexiglass. Current and voltage control was provided by a digital power supply

(Fig. 2). 1000 mL of wastewater was used for each experiment in the reactor. Monopolar parallel connected 4 electrodes were used in the reactor. The electrodes were placed 20 mm apart and completely immersed in the electrolyte. Iron (Fe) plates with the dimensions of 50x70x2 mm and an active surface area of 210 cm^2 were used as the electrode material. Before and after the experiment, the electrodes were immersed in a solution mixture of HCl acid (35% v/v) and hexamethylenetetramine ($(\text{CH}_2)_6\text{N}_4$ 2.80% v/v) in order to purify the residues that may accumulate on the surface. For pH adjustment, 0.1 M H_2SO_4 and NaOH solutions were used. To limit the effect of the hydrogen peroxide used in the EF experiments, the pH was increased to around 10-11. During the experiment, the current and voltage from the power source were monitored and recorded. In the experimental study, it was studied at a constant current, and the average voltage value was taken by recording the voltage throughout the experiment. The average voltage value was used in the energy consumption calculations. Anode and cathode electrode consumptions were weighed separately, but cost calculations were made considering the total electrode

consumption in the calculations. 1000 mL of wastewater was placed in the reactor and the stirring speed was set to 250 rpm. After the desired current and voltage adjustment was made on the power supply, the electrochemical treatment process was started. pH, conductivity, ORP, temperature, COD and TOC measurements were made in the samples filtered (0.45 μ m filter) and centrifuged at certain time intervals.

Analytical method

The pH, conductivity (Elmetron CPC-505), Oxidation Reduction Potential-ORP (Hanna 2211), temperature, COD and TOC (Apollo 9000) measurements of the leachate used in the studies were made according to the analysis methods specified in the standard methods [76]. COD measurements were made according to the "Close Reflux" Colorimetric Method expressed in the Standard Methods, and a calibration curve was prepared from the prepared potassium hydrogen phthalate (C₈H₅KO₄) standard solution (50-1000 mg O₂/L COD). TOC measurements were made using the TOC device at an oven temperature of 680 °C and using dry air according to the "High Temperature Combustion" method. A calibration curve was created with potassium hydrogen phthalate standard solution to be used in TOC analysis. The COD, TOC, and (R %) removal efficiency were calculated according to the equation (Eq. 9) given below.

$$R(\%) = \frac{C_i - C_e}{C_i} \times 100 \quad (9)$$

In this equation, C_i and C_e are the initial and final concentrations of pollutants (mg/L) in feed solution and leachate streams.

RESULTS AND DISCUSSION

Effect of pH

Since the pH value is considered as one of the parameters affecting the treatment efficiency of the method, first of all, the optimum pH value, which provides the removal of COD and TOC in the EC and EF methods, was investigated. Electrical conductivity and pH are important parameters in electrochemical processes. The lower these values are, the lower the current efficiency and the higher the applied potentials to avoid passivation of the electrode are. As the pH of the solution increases, the electrode potential is expected to increase linearly in the negative direction. With the increase of the ambient

pH value, both the passive zone narrows and the passive corrosion current density increases. The pH of the environment affects chemical processes such as dissolution, precipitation, redox and retention reactions between wastewater and leachate [77]. In order to determine the effect of initial pH in the EC process, experiments were carried out using iron electrodes in the pH 5-9 range, with a constant current density of 100 A/m² and during 45 min of electrolysis. As for EF experimental studies, experiments were carried out by changing the initial pH value of the leachate in the range of 2-7. Current density is taken as 150 A/m², H₂O₂ concentration as 500 mg/L, and electrolysis time as 30 min. According to the results obtained, the effect of pH on COD removal in the EC and EF processes has been presented in Fig. 3 below.

The removal mechanism in the EC process can be expressed by flotation with H₂ formed at the cathode and with the formation of metal-hydroxy complexes that will form according to the pH value of the wastewater sample adsorption of pollutants from the wastewater and flocculation mechanism. The Fe²⁺ and Fe³⁺ ions formed in the solution as a result of the use of iron electrodes in the electrocoagulation process and the anodic dissolution of these materials form monomeric and polymeric iron hydroxide complex types depending on the pH. These complexes tend to polymerize in the pH range of 4-7, resulting in many monomeric and polymeric types of iron [78,79]. As a result of the formation of iron complexes at various pHs, the pollutants whose charge balance is disturbed combine with the iron hydroxide species and form a precipitate.

The COD removal efficiency in the EC process is highly dependent on the initial pH. In acidic conditions, iron species are more soluble than at neutral pH, and therefore coagulation of pollutants is well accomplished at the natural pH of the wastewater, which is close to neutral [80]. In the study, the highest COD removal efficiency was found to be 34.94% at pH 7 during 30 min of operation. It is seen that the COD removal efficiency decreases at pH values below or above 7 (Fig 3a). In many studies, it has been reported that various operating parameters such as electric current and electrolytes increase the dissolution of the anode and accordingly the pH of the solution increases [81,82]. Several reports have shown that precipitation of metallic species depletes the alkalinity produced at the cathode, thus resulting

in a relatively stable pH under alkaline conditions [83]. As a result of these results, it was decided to choose the initial pH of the leachate as pH 7, which is the value with the highest removal efficiency in both parameters. These results were in agreement with other studies investigating the effect of pH on EC activity [84-86].

It is known that Fenton reactions are highly dependent on solution pH compared to other oxidation processes and generally occur in an acidic environment. An acidic environment is a favorable condition for H_2O_2 production. It has been stated that the most suitable pH range for Fenton oxidation is 2.5-5 [87,88]. As can be seen in Fig. 3b, high removal efficiencies were obtained in the removal of pollutants at low pH values. The highest COD removal efficiency of 71.7% was obtained at pH 3 at 10 min. In cases where the initial pH was >3 , yield reductions were observed. This may be due to the reaction occurring on the anode surface and the increase in the H_2O_2 concentration in the environment. In the study, as the electrolysis time increases, there is an increase in the removal efficiency of all pollutants. While there was a rapid removal efficiency for COD, especially in the 0-15min. range, there was a partial increase after 15 min. and no significant change was observed. This can be explained especially by the rapidity of the reactions in the electrolysis environment and the dependence of the reaction on pH.

As a result of these results, it was decided to choose the pH of the leachate as pH 3, which is the value with the highest removal efficiency. This is an expected pH value when the studies on the EF process are evaluated and is consistent with the results in the literature [89-92]. The reason why the yields are high, especially at low pH (acidic conditions) can be considered that the iron electrode used as a result of Fenton reactions is in soluble form in acidic conditions and it decomposes H_2O_2 and forms $OH\cdot$ radicals [93]. Also, at $pH < 3$ in cases hydrogen peroxide will remain stable with respect to oxonium ion formation [94]. Due to Fe^{2+} regeneration, the Fenton process becomes less effective at $pH < 3$ due to the reaction between Fe^{3+} and H_2O_2 [95]. When Fe^{2+} ions rise above pH: 5-6, they form hydroxyl complexes instead of hydroxyl radicals ($OH\cdot$), and H_2O_2 decomposes under basic conditions and loses its oxidation ability [96]. With the increase in pH, the iron ions in the environment are converted into Fe^{3+} hydroxide form, which is formed under basic conditions and has a precipitating feature, reducing

the efficiency of the system. Since at higher pH values $Fe(OH)_3$ will precipitate, it causes H_2O_2 to decompose into H_2O and O_2 . As a result, the oxidation efficiency is significantly reduced. If the pH values are close to neutral, however, iron ions are mostly found in the form of hydroxyl complexes (such as $Fe(OH)_2$, $Fe(OH)^-$). Therefore the fact that Fe (II) oxidation depends on the ambient pH can be attributed to the oxidation of not only Fe (II) but also $(Fe(OH)^-)$ [97].

Effect of current density

In the electrocoagulation process, the current density is an important parameter in advanced oxidation processes (EAOP) as it controls the reaction rate and the amount of OH^- produced [98]. Higher applied current density means higher applied voltage in the electrochemical system. In order to have a minimum power consumption, the applied current density must be restricted. Total energy consumption and operating costs also need to be considered in order to achieve maximum removal. When more current density is applied than required, cost increase as may occur, and sludge formation will increase in parallel ratio. In the studies, it was stated that the removal efficiency decreased with the increase in the amount of excess iron at high current density in electrochemical treatment. This situation is explained by the fact that the excess catalyst in the environment reacts with the hydroxyl radical and prevents the oxidation of the pollutant, as explained in the literature [30,99]. If the value of the current density is low, it requires a longer time to remove the pollutant, requiring larger facilities and operating costs. To maximize contaminant removal efficiency with minimal power consumption, the current density must be kept at an optimum level [100]. Therefore, depending on the amount of current applied to the system, there is a relationship between the amount of metal dissolved in the anode electrodes. As more electrode material passes into the reaction solution with the increase of current density, it increases the rate of electrochemical reactions and metal hydroxide formations that disrupt the balance of pollutants in wastewater [101].

The effect of applied current density on COD removal for EC and EF processes is given in Fig. 4. The effect of current density was investigated in the range of 50-300 A/m^2 for both processes. Looking at the results, although the removal efficiency for COD increased slightly

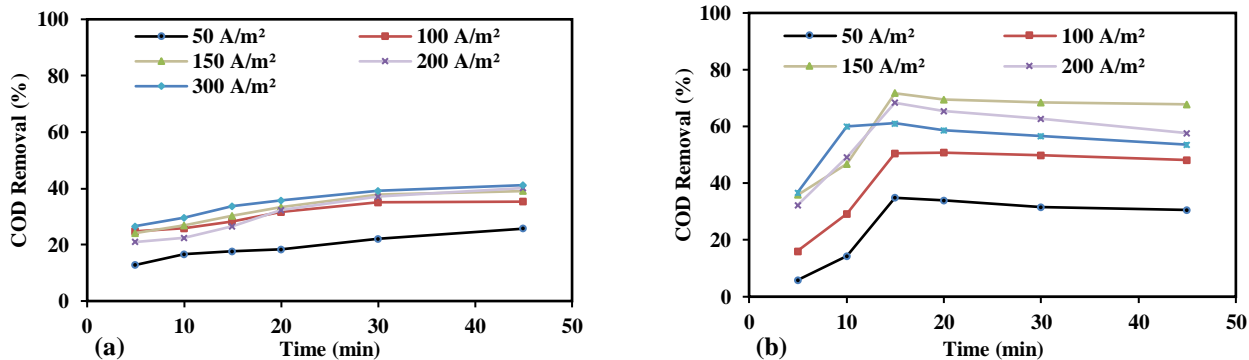


Fig. 4: Effect of current density on COD removal efficiency ((a) EC process; pH: 7.0 t: 45 min, (b) EF process; pH: 3, t: 45 min, H₂O₂: 500 mg/L, H₂O₂/COD: 0.9)

with the increase of current density, high removal efficiency could not be obtained. In the EC process, the COD removal efficiency increased by approximately 5.86% when the current density was increased from 50 to 300 A/m² due to the increase in the concentration of iron and hydroxide ions in the solution. The removal efficiencies were realized between 37.87 - 41.14% at current densities of 150-300 A/m². The rate of increase in the removal efficiency is slow after 100 A/m² and showed a very small difference after this current density. Considering the energy consumption of the process, the operating cost of the electrodes, and corrosion, the best value current density was determined as 150 A/m².

With hydroxide ions and polymeric species like other soluble species ferric hydroxy complexes can also be produced. These species may not transform fast enough to produce an insoluble Fe(OH)_{3(s)} and may prevent COD removal [102-104]. On the other hand, studies have shown that the gradual increase of side reactions such as the reduction of oxygen to water and hydrogen formation may be the main reasons [105,106]. In various studies, it has been reported that COD removal by EC process does not change significantly at higher current densities [107-109]. Some soluble and miscible organic compounds do not react at all with hydroxides and are not removed by EC, and the COD in the leachate is only partially removed as it is a mixture of various compounds [110]. As hydrophobic molecules are adsorbed on the surface of aggregates and high molecular weight compounds, they may show a weak charge density that can be more easily neutralized by EC. Therefore, low molecular weight organic substances may not be easily removed by electrocoagulation [111].

In the EF process, in which the effect of current density was examined, a better removal efficiency was obtained (Fig.4b). The highest COD removal efficiency was obtained as 71.7% at 150 A/m² current density. Considering the operating cost and corrosion of the electrodes, 150 mA/m² was chosen as the optimum current density. Obtaining the highest removal efficiencies at this current density means that the degree of anodic dissolution of the iron is increased, resulting in a greater amount of precipitate to remove contaminants. Higher current density causes hydrogen peroxide formation at the cathode by reducing oxygen at the anode, and accordingly, the formation of hydroxyl radicals, which are highly reactive in the environment and responsible for degradation, increases [112,113]. The degradation of organics by the EF process depends on the appropriate Fe²⁺ the concentration as well as the H₂O₂ concentration [114]. Electro-regeneration of iron ions at the cathode occurs more rapidly with increasing current, resulting in increased efficiency of Fenton chain reactions.

Effect of reaction time on removal efficiency

The removal of pollutants in the EC process is mostly proportional to the electrolysis time and the direct electric current density and the concentration of metal ions formed on the electrodes [115]. Electrolysis time is a very important measure in electrocoagulation as it determines how long water must be purified to meet the required criteria. The electrolysis time also affects the efficiency of the EC. Anodic electrodissoolution causes the release of coagulant species during electrolysis. With the dissolution of the anode and reduction of the cathode, the production of metal ions and more hydrogen bubbles occurs when the reaction

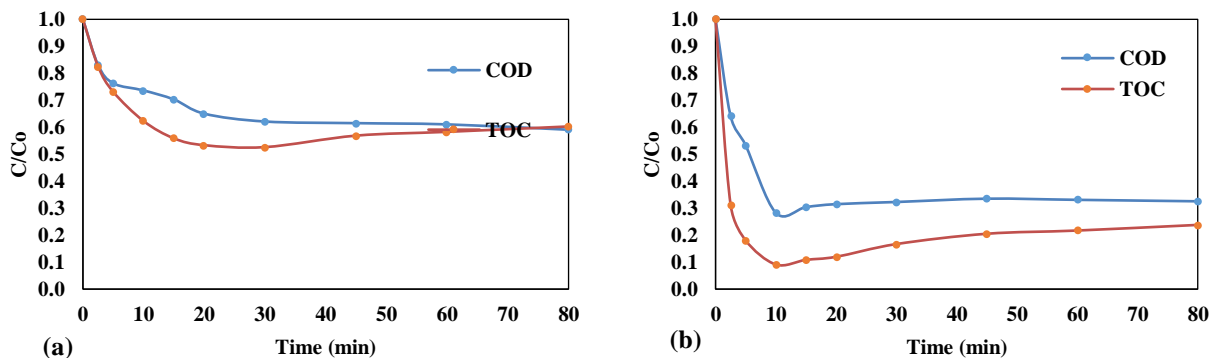


Fig. 5: In EC and EF processes the effect of electrolysis time on COD and TOC removal efficiency (a-EC; pH 7, 150 A/m², b-EF; pH:3, 150 A/m², H₂O₂: 500mg/L)

time is prolonged. These enhance EC and EF advances together, resulting in high removal of a pollutant [116,117]. If sufficient time is not provided for the reactions and the coagulation to occur immediately after, the dissolved electrode material and the coagulants it creates can form sludge that is difficult to filter in water. The COD and TOC removal efficiency depends on the number of hydroxide flocs increasing with the increase in current density and electrocoagulation time, and optimum reaction time should be provided for the formation of metal hydroxides [86]. However, prolonging the reaction time beyond the optimum condition may cause organic pollutants in wastewater to produce more toxic intermediates and decrease treatment efficiency [118]. Although a further increase in reaction time will result in a slight increase in removal efficiency, this is not always applicable due to high energy consumption as well as electrode consumption. The optimum reaction time in terms of process load and process efficiency is considered to be 20 to 30 minutes [119]. In order to examine the effect of the reaction time on the COD and TOC removal efficiency in the EC and EF processes, experiments were carried out at pH and current density, where the highest removal efficiency was obtained in the previous experiment results. In the study, the effect of reaction time on EC and EF processes was investigated in the range of 2.5-80 min (Fig. 5). As the electrolysis time increases, there is a direct proportional increase in the removal efficiency. During the EC treatment, it has been realized a sharp increase in the productivity of COD, and TOC removal when the reaction time was increased from 5 min to 20 min (Fig. 5a). The pollutant removal efficiencies reached the highest value at 30 min and there was no

significant change in the removal efficiency at longer times. The decrease in removal rate after 30 minutes may be due to cathodic passivation and the production of monomeric electrocoagulant species [120,121]. Due to the fact that the amount of precipitate formed is not high and a significant part of the COD is soluble, lower removal efficiencies are observed in shorter periods [51]. It is seen that the TOC removal efficiency also increases over time but tends to decrease after a certain point. The reason for this could be the bubble velocity formed in the medium with the increase in electrolysis time, the amount of decomposition of hydroxide species formed and the increase of pH M(OH)₄ flocs that will form. For this reason, the optimum electrolysis time was chosen as 30 min, in which the highest COD (37.87%) and TOC (47.36%) removal efficiencies were obtained. The results show that although the electrolysis time continues to increase, the removal of residual dissolved contaminants by EC alone is not sufficient. Determining the appropriate reaction time is critical to achieving higher performance efficiency in the EF process. In experiments investigating the effect of reaction time on EF, the highest COD and TOC removal efficiencies were achieved in 10 min reaction time (71.7% and 90.87%), and no significant change in removal efficiency was observed at longer times (Fig. 5b).

Some studies based on the EF process have reported that the optimum reaction time is less than 30 min [122,123]. It has also been reported that the mineralization of organics in landfill leachate is rapid during initial reaction times, oxidation of organic intermediates is slow and a similar initial rapid degradation in landfill leachate is largely due to readily degradable organics [124,125]. The COD

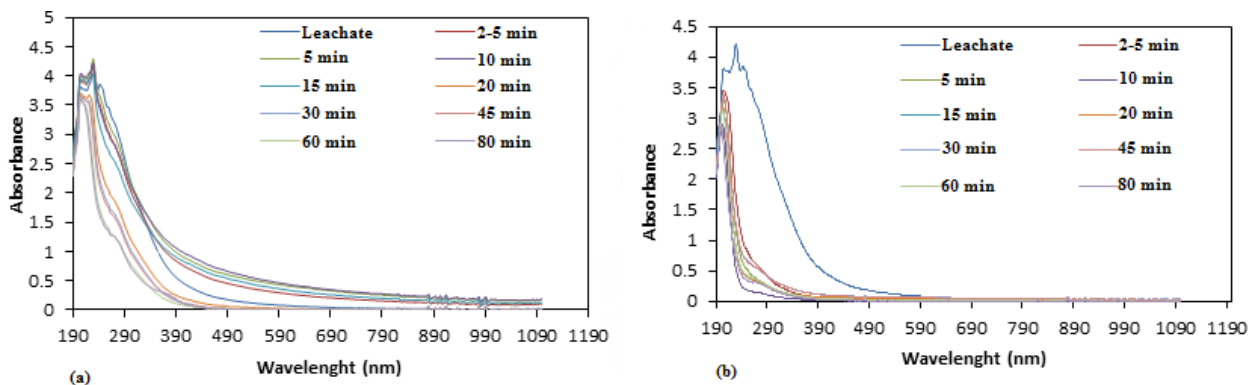
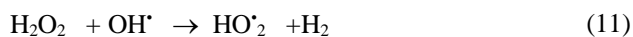


Fig. 6: Spectrum scanning at optimum conditions based on time in (a) EC and (b) EF processes

degradation recorded after 10 min can be in the form explained as the unchanged significantly, possibly due to turning H_2O_2 to H_2O and O_2 (Eq. 10-11) as the electrolysis time increases [126]. In addition, a rapid COD reduction in the first 10 min of the process may be due to the oxidation of easily degradable organic compounds. However, as time went on, the rate of degradation may have slowed gradually, possibly because of the formation of short-chain organic acids and hardly oxidizable by-products [127]. The early rapid degradation of organic compounds can be attributed to reactions between hydroxyl radicals and pollutants, then the reaction rate decreases as the pollutant concentration decreases and more intermediates are produced.



A sharp increase in TOC removal efficiency was observed by increasing the reaction time from 2.5 to 10 min. With further increase in electrolysis time, there is a steady increase in removal efficiency. The increase in fast and slow removal of TOC with increasing electrolysis time can be explained by the reaction between $\bullet\text{OH}$ and TOC in wastewater. At a constant TOC concentration, the increased electrolysis time may accelerate the formation of $\bullet\text{OH}$, leading to a rapid reaction with the TOC contaminant. However, under saturated Fe^{2+} conditions, stable OH formation reacts with insufficient pollutants; therefore, TOC removal efficiency gradually increases [128].

Spectrum scanning in the ultraviolet (UV) region is widely used in various studies as a measurement method that proves the presence of organic compounds in water. Organic compounds are mostly observed in the UV region.

Spectrum scanning was performed in the wavelength range of 190-1100 nm of the output samples obtained in optimum conditions with the highest removal efficiency depending on time (Fig. 6). As can be seen from the figure, there is not much change in organic matter removal at some time intervals during EC. However, in the spectrum scans of some samples, removal is observed. This difference (decrease) in the spectrum scans of the samples at the beginning and end of the EC is due to the removal of contaminants from the leachate. The increase in the amount of ions formed at the anode with the increase of the current density can cause an increase in the bubble formation rate and amount, and on the other hand, the decrease in the bubble size can increase the removal efficiency. If in the EF process, in the spectrum scanning of the samples taken, both it was looked after whether organic substances were transformed into different compounds as a result of the reaction with OH^\bullet radicals, and an assessment has made in terms of optimum pH selection. It can be clearly seen from the spectrum scanning that the removal efficiency is higher in the EF process compared to the EC process. Determining the optimum electrolysis time (10 min) and pH (pH 3) values as optimum values according to the results obtained from both spectrum scanning and analysis in the removal with the EF process confirms the results.

Effect of H_2O_2 concentration

In EF reactions, the consumption of H_2O_2 is one of the most important factors limiting the effectiveness of the method. As the initial H_2O_2 concentration added to the system increases, the concentration of the hydroxyl radical, which is the main oxidizing agent, increases, increasing COD removal. Since hydroxyl radicals are very

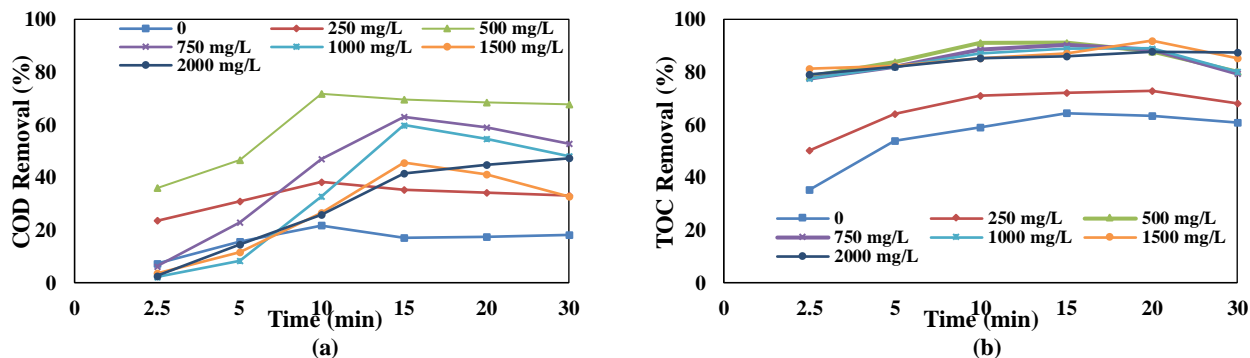


Fig. 7: The effect of H_2O_2 concentration on (a) COD, (b) TOC removal efficiency in the EF process ($\text{pH}:3$, current density: 150 A/m^2 , electrolysis time: 30 min)

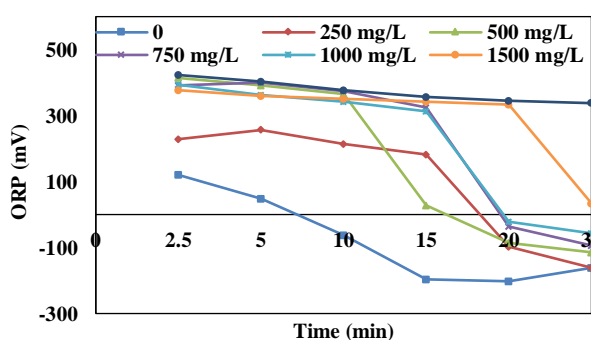
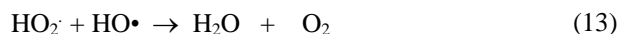


Fig. 8: Time-dependent ORP change in EF process

active and fast from the moment they are formed in the environment, they react with the organic substances in the environment in a short time and increase the removal efficiency [129]. This increase in removal rate can be explained by the increase in the initial concentration of H_2O_2 due to the increase in the number of hydroxyl radicals resulting from the rapid degradation of H_2O_2 [130]. However, when there is an excess of iron ions in the electrolyte solution, it can consume hydroxyl radicals and affect the degree of degradation [131]. In the study, the effect of different H_2O_2 concentrations (0.25-2.0 g/L) on removal efficiency at constant pH and current density is presented in Fig. 7.

When the experimental results were examined, the COD and TOC removals were 71.7% and 91.03%, respectively, at the oxidation time (10 minutes) where the highest removal efficiency was obtained and at the H_2O_2 concentration where sufficient oxidant concentration was provided (500 mg/L) (Fig. 7a-b). In similar studies, it has been reported that reduced feeding time increases the rate of COD removal [132–134]. It is seen that the removal efficiencies remained almost constant in the trials where higher concentration values (750-2000 mg/L H_2O_2) were

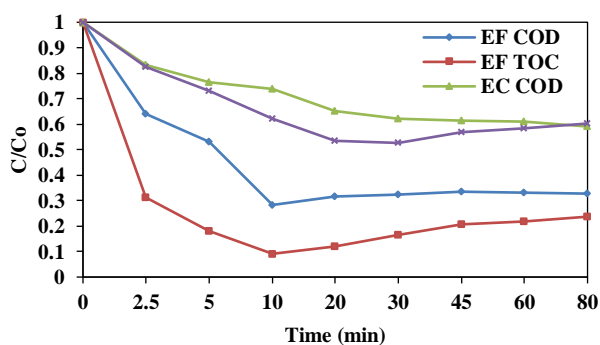
applied. Due to increased reactions between hydrogen peroxide and hydroxyl radicals, a slight decrease in removal efficiency may have occurred at higher hydrogen peroxide concentrations. The decrease in removal efficiency at high H_2O_2 doses may be due to the scavenging effect of H_2O_2 hydroxyl radicals (Eq. (12, 13)) and the recombination of hydroxyl radicals (Eq. (14)) [135].



The increase in the $\text{H}_2\text{O}_2/\text{Fe(II)}$ molar concentration in the electro-Fenton oxidation causes parasitic reactions to dominate in the system. As the dose of H_2O_2 used as an oxidant increases, oxidation of organic substances occurs, but after a certain period of time, little or no change occurs in the environment. This process continues until the H_2O_2 threshold is reached. When the optimum dose of H_2O_2 is reached, sudden increased efficiencies in COD removal and toxicity reduction are observed. Continued addition of the oxidant after the cut-off point results in a rapid reduction in aquatic toxicity. In other words, the addition of H_2O_2 above the optimum dose does not increase the efficiency and causes positive interference in experimental studies such as COD for efficiency control. Therefore, a certain amount of H_2O_2 must be given in the EF process [136]. The time-dependent ORP change at different H_2O_2 concentrations in the EF process is given in Fig. 8. The ORP value represents a comprehensive indicator of the effect of current density, contaminants, and reaction time on the performance of the electro-oxidation system and can be used as an effective control factor to optimize

Table 4: Comparison of electrocoagulation and electro-fenton processes under optimum condition

Process	COD removal (%)	TOC removal (%)	ORP (mV)	pH	COD:H ₂ O ₂	Current density (A/m ²)	Time (min)	Operating cost(\$/m ³)
EC	37.87	47.36	-118.5	7	NA	150	30	2.26
EF	71.7	90.87	367.5	3	0.9	150	10	1.78

**Fig. 9: Comparison of COD-TOC removal efficiency in EC and EF processes at optimum conditions**

the electro-oxidation process [137]. It is a measure of the cleanliness of water and its ability to break down contaminants is measured in millivolts (mV). If the measurement result is positive, it indicates that oxidation, that is, rusting, and destructive/corrosive effects of this water are dominant, and a negative value indicates that this water has reduced, that is, antioxidant power [138]. As the oxidant concentration increases, the oxidation potential of the environment increases and becomes more stable. This situation can be explained by the increase in H₂O₂ concentration as explained before. ORP decreases with increasing current density and electrolysis time. As can be seen from the figure, this decrease in value accelerated especially with the increase in current density. This shows that the Fe electrode used as the anode in the EF process does not provide sufficient oxidation potential. During electrolysis, the iron dissolved in the anode passes into the environment as Fe²⁺, and the OH• ions formed especially at the cathode during the electrolysis of water rapidly degrade the H₂O₂ in the environment and the oxidation potential decreases.

Comparison of electrocoagulation and Electro-Fenton under optimum conditions

In this study, in which the treatability of leachate waters with EC and EF processes was investigated, different treatment efficiencies were obtained. In Table 4, the removal efficiency results, and total operating cost

values obtained in EC and EF processes under optimum conditions are given comparatively for the COD and TOC parameters. Total Operating cost was calculated using the following equation (Eq. (15)) based on electrode material, electrical energy, and chemical usage [139,140].

$$\text{Operating Cost } (\$/\text{m}^3\text{kg}) = 0.06C_{\text{energy}} + 0.6C_{\text{electrode}} + [1.0034\text{CH}_2\text{O}_2] \quad (15)$$

In this equation; C_{energy}: Energy consumption in EC and EF processes (kWh/m³), C_{electrode} is the amount of solute ferrous metal electrode in the EC and EF reactor (kg electrode/m³ of treated leachate water), CH₂O₂: It represents the amount of chemical consumed in the EF process (kg/m³).

In the optimum operating conditions of the EC process (pH 7, 150 A/m²), the COD and TOC removal efficiencies were obtained as 37.87% and 47.36%, respectively (Fig. 9). In order to increase the efficiency of the EC process, the EF process was applied. With the EF process, high treatment efficiency has been achieved in waters with high toxic and organic pollution. In the EF process, H₂O₂ has added as an oxidant. The COD and TOC removal efficiencies of the EF process at optimum operating conditions (pH 3, 150 A/m², H₂O₂: 500 mg/L) were obtained as 71.7% and 90.87%, respectively. When evaluated in terms of removal, it was determined that the COD and TOC removal efficiencies were higher in the EF process compared to the EC process. Considering the optimum conditions where the highest removal efficiencies of both processes are achieved, it is seen that the EF process is more economical when evaluated in terms of total operating cost and energy cost. Electro-Fenton has a lower overall operating cost due to the shorter electrolysis time, which results in less energy and electrode consumption.

CONCLUSIONS

This study investigated the performance of landfill leachate treatment in terms of COD and TOC removal using EC and EF processes. Different treatment efficiencies were obtained in this study, in which the treatment of leachate waters with EC and EF processes was investigated. Under

optimum operating conditions of the EC process (pH 7, 150 A/m², and 30 min), the COD and TOC removal efficiencies were 37.87% and 47.36%, respectively. Low treatment efficiencies have been achieved in leachate pollutant removal with the EC process. The EF process has been applied to increase the efficiency of the EC process. The COD and TOC removal efficiencies of 71.7% and 90.87%, respectively, were obtained under the optimum operating conditions of the EF process (pH 3, 150 A/m², H₂O₂: 0.5 g/L, 10 min). In the calculations made, the total operating cost for the COD and TOC parameters was 2.26 and 1.78 (\$/m³) for the EC and EF processes, based on electrode material, electrical energy, and chemical usage, respectively. According to the EC results, it can be concluded that the oxidation mechanism for the removal of free radicals and organic compounds produced in EF shows higher purification efficiency than the coagulation mechanism in toxic and organic polluted waters. It has been observed that EC treatment alone is not sufficient for leachate contaminant removal. Considering the optimum conditions where the highest removal efficiencies of both processes are obtained, it is seen that the EF process is more economical when evaluated in terms of total operating cost and energy cost. The study showed that the EF treatment can be effectively used as a post-treatment to improve the treatment efficiency of landfill leachate. Modified electrochemical treatment processes with EF-like (AOPs) can achieve high simultaneous organic removal in leachate treatment with complex wastewater characteristics.

Abbreviations

mV:	Millivolts
EAOP:	Advanced Oxidation Processes
UV:	Ultraviolet
ORP:	Oxidation Reduction Potential
EC:	Electrocoagulation
EF:	Electro-Fenton
COD:	Chemical Oxygen Demand
BOD:	Biological Oxygen Demand
AOX:	Adsorbable Organic Halogens
TP:	Total Phosphorus
TKN:	Total Kjeldahl Nitrogen
SS:	Suspended Solids
NH ₄ ⁺ -N:	Ammonium Nitrogen
Cl:	Chloride

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