ORIGINAL PAPER

Assessment of treatment performance in COD, TOC and AOX removal from landfill leachate by electro-Fenton technique

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Abstract

Landfill leachate is a mixture of highly concentrated organic and inorganic pollutants and needs to be treated properly due to their toxicity and serious adverse effects on the environment. Recently, advanced oxidation processes have been successfully applied for leachate treatment. In this study, the removal of chemical oxygen demand (COD), total organic carbon (TOC) and absorbable organic halogens (AOX) in waste leachate stabilized by the electro-Fenton (EF) process, which is one of the advanced oxidation processes, was investigated. For this purpose, a jacketed reactor in which the electrochemical process takes place was designed and iron electrodes were used for maintenance and cost efficiency in this process. Within the scope of the study, the efficiency of the important process parameters, such as pH, current density, catalyst amount, oxidant dose and electrolysis time, was determined and their values were optimized. Optimum values were found to be as pH 3, current density 150 A/m^2, hydrogen peroxide amount 500 mg/L and electrolysis time 10 min. The best removal efficiencies were obtained as 71.7% COD, 97.4% AOX and 90.87% TOC under optimum conditions. In addition, within the scope of this study, the collapsibility and operating costs of the sludge having formed as a result of the EF process were also examined. It has been observed that especially the sludge formed as a result of the EF process has good settling properties. When the results were evaluated, it was confirmed that the EF process can be used efficiently to break down leachate organics in terms of wastewater quality and discharge limits.

Keywords Landfill leachate · Electro-Fenton (EF) · COD · TOC · AOX · Removal

Introduction

Landfill is one of the most common methods for the disposal of municipal solid waste (MSW) all over the world because of its economic advantages. One of the most important problems in sanitary landfills is leachate management. Leachate waters are waters containing dissolved and suspended form of physical, chemical and biological pollution. Leachate water consists of a complex mixture of organic and inorganic compounds with a distinctive odour. Leachate water may contain organic and inorganic ions and micro-pollutants in addition to metals. For this reason, landfill leachate generated in solid waste landfills is one of the most difficult and expensive wastewaters to treat owing to the high amount of organic and inorganic pollutants in its structure. It is often referred to as "difficult wastewater" and has a more concentrated pollution load when compared to many domestic and industrial wastewaters. They are highly toxic and carcinogenic wastewaters and can cause significant harm to the environment and human health (Baderna et al. 2019; Kapelewska et al. 2019). When the stored waste reaches its saturation capacity, pollutants such as organic substances, dissolved salts and heavy metals from garbage in the landfill are transported and leak out. The resulting leachate can pass from the soil to the ground and surface waters and affect the quality of the ground and surface water in that place. Transport of dissolved substances in leachate is an important source of pollutants for surface and groundwater. Since leachate affects the aquatic ecosystem and human health, it must be kept in a landfill or treated before being released into the aquatic ecosystem. Many parameters such as chemical oxygen demand (COD), biological oxygen demand (BOD),

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absorbable organic halogens (AOX) and total nitrogen (TN) are important for the type of treatment technology to be applied. In the early stages, leachate contains significant amounts of BOI_5, COD, total dissolved solids (TDS), nutrients and heavy metals. Collecting leachate and returning it to the landfill is an effective method used in leachate treatment. When the leachate is rotated, the components in the leachate are reduced by the biological activity and other physical processes and chemical reactions that occur in the landfill. The increase in non-degradable organic leachate components (especially residual COD and AOX) is mainly a function of dilution. The BOD_5/COD and COD/TOC ratios are common indicators of organic compound biodegradability and the oxidized state of organic carbon. In particular, if the COD exceeds the receiving environment discharge limits, significant problems arise. In studies on the treatment of leachate, it has been reported that almost half of the pollutants, including many inorganic pollutants, are below the direct discharge limits. It has been reported that the parameters exceeding the discharge limits are predominantly organic substances (COD, BOD_5 and AOX) and nitrogen (mainly NH_4-N and NO_3-N) (Liu et al. 2015; Mor et al. 2018; Vaccari et al. 2018; Yu et al. 2020). The European Union (EU), as well as certain European countries, have limits for the discharge of leachate for time periods. Council Directive 1999/31/EC of on the landfill of waste (EC 1999), Council Directive 91/271/EEC concerning urban wastewater treatment (EC 1991), Water Framework Directive 2000/60/EC (EC 2000) and Waste Framework Directive 2008/98/EC (EC 2008) are among the major European regulations governing the storage and leachate management. In Turkey, the limit values stipulated for "Discharge of wastewater to wastewater 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 (WPCR 2004). 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 1 (Stegmann et al. 2003; Mukherjee et al. 2014; Brennan et al. 2016; Pajooh et al. 2017).

AOX, which is important in terms of the type of treatment technology and one of the parameters that we examined in our study, is a parameter that determines the total amount of organically bound halogens in wastewater. AOX is a parameter that should be considered because it contains chemicals that have mutagenic and carcinogenic effects on human health and is abundant in the environment. AOX compounds are hydrophobic and persistent. They often accumulate in the food chain. They are highly toxic, and some species have been found to be carcinogenic and mutagenic (Kaczmarczyk and Niemirycz 2005; Vinder and Simonic 2012). Organohalogens are organic compounds containing one of the 7A group elements such as fluorine, chlorine, bromine or iodine. As it is known, although the control of organohalogen compounds is important all over the world, these compounds are classified as pollutants that are dangerous for the environment and need to be monitored. When the available literature resources are examined, it has been seen that very few studies have been conducted on the determination and monitoring of organohalogen concentrations in Turkey. It is known that various studies are carried out in the world on monitoring and evaluation of chlorinated organic compounds. With regard to AOX removal, numerous studies have been cited reporting that the efficiency of biological processes in removing these compounds from wastewater is less than 50% even at a hydraulic retention time of 15 h (Ribeiro et al. 2020a). Studies conducted in Finland reported that AOX welded production, whose main sources are the pulp industry, PVC production and waste incineration, is responsible for approximately 50% of the total organic halogen emissions to the environment (El-Hadj et al. 2007). It has been reported that the average concentration of AOX, which is a measure of the organic halogen load in a sampling site such as wastewater or sewage waste soil, in sewage wastes is approximately 37 g/L (Schowanek et al. 1996). 56% of the priority pollutants identified by the United

Table 1 Limit values for direct discharge of leachate or for discharge of leachate to surface water after in situ treatment

Parameters	Turkey		The Netherlands	Germany	Ireland	France
pH	6–9	6–9	–	–	7.52	–
Suspended solids (SS) (mg/L)	[a]200	[b]100	30	–	–	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 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	–	–

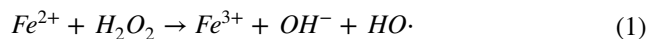([a]Composite sample 2 H, [b]composite sample 24 H)

*Existing discharge standards for the AOX parameter in Turkey are not included in the table, since they are not covered by the regulation
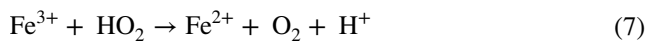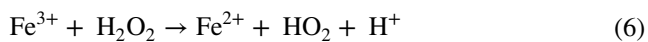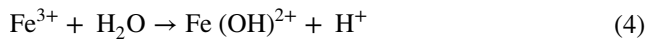
States Environmental Protection Agency (USEPA) and 23 persistent organic pollutants controlled by the Stockholm Convention are all reported to be AOX compounds (Xie et al. 2016). Due to the harmful effects of organic halogens, AOX removal studies have been carried out in leachate and other industrial wastewater (Lei et al. 2007; Nooten et al. 2008; Goi et al. 2009; Xie et al. 2017; Milh et al. 2020; Ribeiro et al. 2021; Xu et al. 2021). Due to the pollutant parameters described above, leachate treatment requires a very difficult and complex process compared to domestic wastewater. Therefore, it is necessary to check the generation of leachate water for toxicity or adverse environmental effects and to treat it with appropriate treatment methods to meet local discharge standards (Qi et al. 2018; Tenodi et al. 2020). Because of its high chemical stability and/or low biodegradability, using physical, chemical and biological treatment methods alone in leachate treatment is difficult to obtain the desired treatment efficiency and wastewater quality. For this, a combination of these methods is usually recommended for leachate treatment (Muller et al. 2015; El-Gohary et al. 2016; Saleem et al. 2018; Miao et al. 2019). Regarding leachate treatment in the literature, different treatment processes such as air stripping (De et al. 2019; Smaoui et al. 2020), coagulation–flocculation (Taoufik et al. 2018; Djeffal et al. 2021; Cheng et al. 2021), adsorption (Augusto et al. 2019; Reshadi et al. 2020; Ren et al. 2020), membrane processes (Chen et al. 2021; Shu et al. 2021; Nazia et al. 2021; Farinelli et al. 2021; Feng et al. 2021) chemical or electrochemical oxidation (de Oliveira et al. 2019; Mahtab et al. 2020; Pierpaoli et al. 2021; Wu et al. 2021) and ozonation (Hoffmann et al. 2020; Yang et al. 2021; Aziz et al. 2021) were studied. These techniques have proved suitable for the removal of these parameters in leachate, including humic, fulvic acid, heavy metals, polychlorinated biphenyls (PCBs), organohalogen compounds (AOXs) and some other persistent organic pollutants (Kamuriddin et al. 2017).

Advanced oxidation processes (EAOPs), such as Fenton (F), electro-Fenton (EF) and photo-Fenton (PF), have received increasing attention due to the efficient mineralization of toxic and persistent organic pollutants. It has been emphasized that these methods are more effective in removing the pollution parameters in various synthetic and real wastewater, especially in landfill leachate (Tejera et al. 2019; Mohajeri et al. 2019; Dolatabadi et al. 2021). In recent years, the most studied advanced EAOPs for the treatment of pollutants from leachate containing hard to decompose degradable substances are anodic oxidation-based anodic reactions (AO) and electro-Fenton (EF)-based cathodic reactions (Santos et al. 2019; Nidheesh et al. 2019). EF is one of the economical and environmentally friendly technologies that can be used in the treatment of domestic and industrial wastewater containing toxic and/or persistent organic pollutants. It takes place in three stages, namely Fenton process, oxidation reaction and coagulation (Zhang 2020; Ribeiro et al. 2020b; Li et al. 2021). In the Fenton process, the reaction between the iron ion and hydrogen peroxide leads to the generation of hydroxyl radicals under acidic conditions (Eq. 1). Hydroxyl radicals have the ability to react non-selectively with most persistent organics to yield dehydrogenated or hydroxylated derivatives until total mineralization is reached (Oturan et al. 2012). Ferric ion produced through Fenton reaction reacts more with H_2O_2 and regenerates iron ions (Sruthi et al. 2018). Although the Fenton process is efficient, processing of hydrogen peroxide, low regeneration of iron ions from ferric ions and the requirement for higher iron ion concentrations are some of the disadvantages of the Fenton process (Brillas et al. 2009; Nidheesh and Gandhimathi 2012; Nidheesh et al. 2013). The EF process is an extended Fenton process in which H_2O_2 is produced electrolytically through two-electron cathodic reduction of oxygen in acidic medium. In addition to that, the EF process eliminates other disadvantages of Fenton processes such as slow iron regeneration rate, sludge formation and increased reaction time and solution pH (Nidheesh et al. 2014). The EF process overrides these disadvantages of the Fenton process by electro-generation of ferrous ions at the cathode as in Eq. (2) by in situ generation of hydrogen peroxide at the cathode in the presence of dissolved oxygen as in Eq. (3), and less Fe ions are required for the degradation of pollutants than the Fenton process (Hammami et al. 2007; Nidheesh et al. 2014). Usually, the term EF is used to describe the link between the Fenton process and electrochemical oxidation, and this link can be of different types depending on the mechanism of attachment or formation of the Fenton reagent (Ganiyu et al. 2018; Klidi et al. 2019; Basturk et al. 2021). In the EF process, hydroxyl radicals (OH·) are produced with Fe^{2+} ions that dissolve into the system from the Fe electrode and mix with the solution. Hydroxyl radicals, which can easily degrade organic substances, show very good oxidant properties (Martinez-Huitle and Panizza 2018; Meng et al. 2019). Generally, carbon-based materials are used as cathodes in the EF process to produce H_2O_2 via the oxygen reduction reaction (ORR). In the presence of the electrically assisted Fenton process, a significant increase in the oxidizing power of H_2O_2 occurs (Sires et al. 2014; Nidheesh et al. 2018a). In this process, H_2O_2 can be continuously electrogenerated at a suitable cathode through electron reduction of O_2. Fe^{2+} ions are added as a catalyst in an acidic environment to obtain Fe^{3+} ions and OH^- from the Fenton reaction and to increase the low H_2O_2 oxidation ability (Eqs. 2,3) (Rueda-Marquez et al. 2020; Hussain et al. 2022).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + HO\cdot \qquad (1)$$

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

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \qquad (3)$$

Here, H_2O_2 is formed by simultaneous reduction of O_2 on the cathode surface, and Fe^{2+} is formed as a result of the reduction of Fe^{3+}. In consequence of the reaction of H_2O_2 and Fe^{2+}, hydroxyl radicals ($OH^.$) are produced homogeneously (Liu et al. 2007). Faster Fe^{2+} regeneration accelerates the formation of OH· radicals by the Fenton reaction. This cycle is sustained throughout the electrolysis, and approximately 75% of the spent O_2 is recovered by the O_2 formed as a result of the oxidation of water on the anode surface during the electrolysis (El-Ghenymy et al. 2012; Fernandes et al. 2017). In undivided cells, although the process proceeds in a complex manner over reactive oxygen species, the main oxidizing agents are OH· radicals. Pollutant-based organics in the same environment are decomposed by reacting with $OH^.$ radicals, and if the electrolysis time is sufficient, they become mineralized throughout the process (Nurhayati 2012; Javaid and Qazi 2019). Since the reactions are carried out at optimum pH 3.0 value, the Fe^{3+} species are predominantly in the form of $Fe(OH)^{2+}$. It is reduced electrochemically to Fe^{2+} for subsequent reactions (Eqs. 4, 5) and is formed in other reactions as shown in equations (6,7) (Sires and Brillas 2021; Divyapriya et al. 2021). This formation accelerates the removal of organic pollutants according to the anodic oxidation and Fenton type reactions applied separately (Divyapriya et al. 2018; Karatas et al. 2022).

$$Fe^{3+} + H_2O \rightarrow Fe(OH)^{2+} + H^+ \tag{4}$$

$$Fe(OH)^{2+} + e^- \rightarrow Fe^{2+} + OH^- \tag{5}$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2 + H^+ \tag{6}$$

$$Fe^{3+} + HO_2 \rightarrow Fe^{2+} + O_2 + H^+ \tag{7}$$

In this study, the applicability of the electro-Fenton process for the treatment of regular landfill leachate was investigated in detail. In order for this process to be applied effectively, the effect of operating parameters such as pH, current density, electrolysis time and H_2O_2 concentration has been investigated and optimized. In addition, oxidation reduction potential (ORP) analysis was performed. The removal efficiency of the method was evaluated with COD, TOC and AOX removals. Aluminium sheet was used as electrode in the study. In addition, the effect of operating conditions on the settling characteristics of the waste sludge produced from the method was also observed. At the same time, technical and economic evaluation was also carried out.

Materials and methods

Study area

In the study, 5 duplicate landfill leachate samples were taken from the entrance of the Sivas (Turkey) landfill leachate collection pool and experimental studies were carried out on these leachate samples (Fig. 1 a, b). In addition, samples were taken from the collection pool outlet and characterization studies of the leachate were carried out. Characteristics values of leachate samples taken from the collection pool outlet and pool inlet are shown in Table 2. Average values were taken as basis in wastewater samples taken from the outlet of the collection pool where leachate accumulated.

The difference in the AOX concentrations at the inlet and outlet values of the collection pond indicates that this is due to the AOX concentrations at the wastewater inlet and that the AOX produced independently of the treatment efficiency is mostly formed after the wastewater mixture. When the COD and TOC values in the table are examined, it has been observed that these values are lower than the normal leachate characteristics when compared with the normal solid waste landfill leachate characterization mentioned in other studies. Therefore, it is thought that the leachate coming into the collection pond is formed by the accumulation of water that is formed with precipitation and passes into the surface flow. As can be seen, the effluent values of the collection pool are above the discharge standards with the average concentration of COD (2200 mg/L) and TOC (425 mg/L). Furthermore, landfill leachate has a high electrical conductivity value due to its high chloride (anion) concentration, which allows electrochemical oxidation without the addition of more electrolyte.

Fig. 1 **a** Leachate inlet to the collection pond; **b** leachate raw sample

(a)　　　　(b)

Table 2 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
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

Design of experiments

Experiments were carried out in a double-walled reactor made of Plexiglas, measuring $100 \times 100 \times 130$ mm. Current and voltage control was provided by a digital power supply. Mixing was carried out with a magnetic stirrer. 1000 mL wastewater was used for each experiment in the reactor. In the reactor, 4 electrodes were spaced 20 mm apart, connected in monopolar parallel, used and totally immersed in the electrolyte. Iron (Fe) plates with the dimensions of $50 \times 70 \times 2$ mm and an active surface area of 210 cm^2 were used as the electrode material. After the desired current and voltage adjustment on the power supply was made, the electrochemical treatment process was started. The mixing speed has been set to 250 rpm. Samples taken from the reactor at certain time intervals were prepared for analysis by filtering (0.45 µm filter) and centrifuging. At the end of the experiment period, the electrodes were washed with distilled water, dried, weighed and recorded. The pH was increased to around 10–11 in order to limit the effect of the hydrogen peroxide used in the EF experiment and to convert the hydrogen peroxide into water and oxygen. The average voltage value was used in the energy consumption calculations. At the end of the experiment, 50 mL samples were taken from the reactor, and suspended solids analysis was carried out to measure the amount of SVI. pH, conductivity, oxidation reduction potential (ORP), temperature, COD, TOC and AOX measurements were taken on the filtered and centrifuged samples taken from the reactor at certain time intervals.

Analytical methods

pH, conductivity (CPC-505), oxidation reduction potential (ORP) (Hanna 2211), COD, TOC (TApollo 9000) and AOX measurements of leachate samples before and after the experiment were taken according to the experimental methods determined in Standard Methods for Water and Wastewater analysis (APHA/AWWA/WEF 2017). A standard curve has been prepared from potassium hydrogen phthalate ($C_8H_5KO_4$) standard solution (50–1000 mg O$_2$/L COD) for colorimetric analysis of COD. TOC measurements were taken according to the "high-temperature combustion" method with a TOC device using 680 °C oven temperature and dry air. A calibration curve was created with potassium hydrogen phthalate standard solution to be used in TOC analysis. The method applied for the measurement of AOX is based on the principle of washing and decomposing the bound inorganic halogens after adsorbing the organic halogen compounds to the activated carbon. The AOX measurement range is between 0.05 and 2.5 mg/L. Therefore, while AOX measurements were taken, real wastewater samples were diluted by about 20 times.

The removal efficiency of COD, TOC, AOX (%R) was calculated according to Eq. 8.

$$R(\%) = \frac{C_i - C_e}{C_i} \times 100 \tag{8}$$

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

Results and discussion

Effect of pH

The pH of the environment affects chemical processes such as dissolution, precipitation, redox and retention reactions between the effluent and the leachate. It is an expected situation that the electrode potential will increase linearly in the negative direction as the pH value of the solution increases. Electrical conductivity and pH are important parameters in electrochemical processes. The lower these values, the lower the current efficiency, and higher applied potentials

are needed to prevent the electrode from passivation. Lowering the pH is an important step in the EF process because the degradation of organic matter is most effective at pH ~ 3 (Umar et al. 2010). With the increase in the ambient pH value, the passive zone both narrows and increases in the passive corrosion current density. It is known that EF reactions are highly dependent on solution pH compared to other oxidation processes and generally occur in an acidic environment (pH 3). There are many studies on the effect of solution pH on the efficiency of oxidation (Wang et al. 2008; Jung et al. 2009; Ghoneim et al. 2011). Acidic environment is the suitable condition for H_2O_2 production. 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, TOC and AOX, was investigated in the electro-Fenton method. For this purpose, experiments were carried out by changing the initial pH value of the leachate in the range of 2–7. During the experiments, the current density of the sample was taken 150 A/m^2, the H_2O_2 concentration was 500 mg/L, and the electrolysis time was 30 min. As a result of the experiments, the changes of COD, TOC and AOX removals with pH are presented in Fig. 2. High removal efficiencies were obtained in the removal of pollutants at low pH values. The highest COD removal efficiency was observed at pH 3 with 71.7% at the 10th min (Fig. 2 a). In cases where the initial

pH was > 3, reductions in yield were observed. However, a low pH also encourages hydrogen formation by reducing the number of active sites to produce hydrogen peroxide (Wang et al. 2010).

Considering the AOX and TOC removals, the highest removal efficiency for these parameters was obtained at pH 3, as 97.4 and 92.19%, respectively (Fig. 2 b,c). In the study, pH 3 value with the highest removal efficiency was accepted as the optimum value. This is an expected pH in the EF process. In acidic conditions below pH: 3, the hydroxyl radical (OH·) can be interpreted as reducing the production of Fe^{+2} ions. At lower pH values, it forms stable complexes with H_2O_2, leading to the deactivation of the catalysts. Hydroxyl radicals can be scavenged by an overdose of Fe^{2+} in the electrolyte (Eq. 9), which impairs the effect of hydroxyl radicals to oxidize organic matter (Wang et al. 2012). As a result, it is an important step to work at the most suitable pH in terms of positively affecting the process efficiency. The reason for the high yields, especially at low pH (acidic conditions), can be thought as the iron electrode used as a result of Fenton reactions, being in a soluble form in acidic conditions and decomposing H_2O_2 to form OH· radicals. In other words, OH· radicals with organic compounds react rapidly in acidic medium and degradation. Also, at pHs below 3 the hydrogen peroxide will remain stable with respect to the
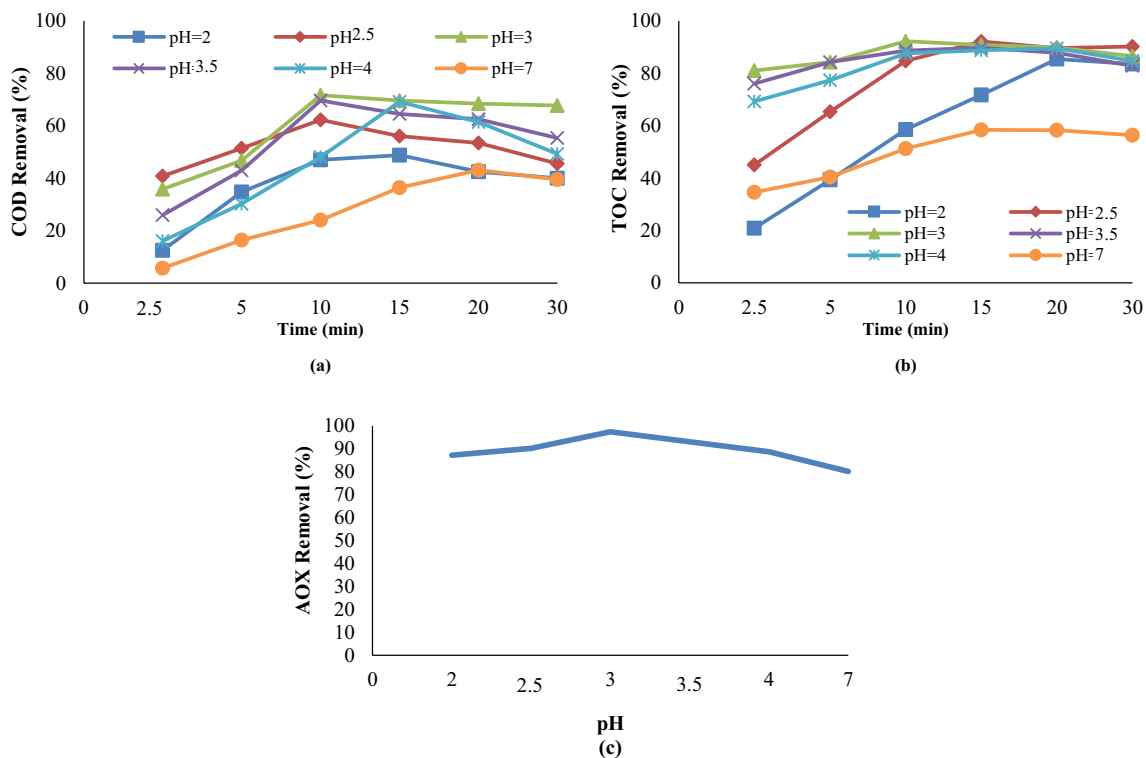

Fig. 2 Effect of pH in the removal of a COD, b TOC and c AOX (current density: 150 A/m^2, H_2O_2: 500 mg/L, electrolysis time: 30 min)

formation of the oxonium ion (Nidheesh and Gandhimathi [2014a, b]). The EF process becomes less effective at pH < 3 due to Fe^{2+} regeneration, the reaction between Fe^{3+} and H_2O_2 (Ramirez et al. [2009]). As the electrolysis time increases, there is an increase in the removal efficiency of all pollutants. While there was rapid removal efficiency for COD and TOC, especially in the 0–15 min range, no significant change was observed even though there was a partial increase after 15 min. This situation can be explained by the rapidity of the reactions, especially in the electrolysis environment, and the dependence of the reaction on pH. Prevention of reaction recycling of the released OH^- ion with H^+ can be considered as the increase in removal efficiency and realizing the degradation of organic substances with OH· radicals formed. The decreases observed in the efficiency for COD and TOC parameters after 10 min in the study show that these parameters are significantly affected by the applied voltage and electrolytic time. Li et al. ([2022a]) evaluated the effectiveness of electro-Fenton on the degradation of waste landfill leachate in their study. In this study, optimum operating parameters and the importance of factors were evaluated by orthogonal experiment. Voltage, electrolytic time and H_2O_2/Fe^{2+} molar ratio which is used in COD removal are reported, and the order of importance of the factors is as follows: H_2O_2/Fe^{2+} > voltage > electrolyte time > H_2O_2/COD_0 > pH. It was concluded that H_2O_2/COD_0 and pH had less significant effect on COD removal. These results showed that H_2O_2/Fe^{2+}, voltage and electrolytic time play an important role in COD removal, while H_2O_2/COD_0 and pH have less important effects.

When Fe^{+2} ions rise above pH: 5–6, they form hydroxyl complexes instead of hydroxyl radicals (OH·), and H_2O_2 decomposes under basic conditions and loses its oxidation ability (Arslan-Alaton and Erdinc [2006]; Wang et al. [2016]). In other words, the increase in pH causes the electrocoagulation process in which the pollutants are removed by the complexation of the reactions due to electrostatic attraction and/or conversion of Fe^{2+} and Fe^{3+} to $Fe(OH)_n$ type structures (Mollah et al. [2001]). At higher pH values, iron species begin to precipitate as ferric hydroxides. Self-decomposition of H_2O_2 also an increase in a more alkaline environment, which brings up the fact that all H_2O_2 added to the electrolyte does not play a role (Li et al. [2022a]). Since $Fe(OH)_3$ will precipitate at higher pH values, it causes the decomposition of H_2O_2 to H_2O and O_2 (Eq. [10]). With the increase in pH, the iron ions in the medium turn into Fe^{3+} hydroxide form, which is formed under basic conditions and has a precipitating feature and causes the efficiency of the system to decrease. At pH values close to neutral, iron ions are mostly found in the form of hydroxyl complexes such as $Fe(OH)_2$, $Fe(OH)^-$. Therefore, the fact that Fe^{2+} oxidation depends on the ambient pH can be attributed to the oxidation of not only Fe^{2+} but also $Fe(OH)^-$ (Shemer and Linden [2006]).

$$Fe^{2+} +^{\bullet} OH \rightarrow Fe^{3+} + OH^- \tag{9}$$

$$H_2O_2 \rightarrow 2H_2O + O_2 \tag{10}$$

Effect of current density

Another working parameter that is effective in the EF method is the magnitude of the current applied to the electrons in the system and it is the most important parameter that affects the reaction rate of electrochemical processes. The applied current is the driving force for the reduction of oxygen leading to the formation of hydrogen peroxide at the cathode. In the EF process, the rate of OH formation during radical point electrolysis is controlled by the applied current. Increasing the current density increases the removal efficiencies up to an optimum value. Higher current application increases the amount of hydrogen peroxide produced. Accordingly, the number of hydroxyl radicals, which are highly reactive in the electrolyte medium and responsible for degradation, also increases (Ghanbari and Moradi [2015]; Kubo and Kawase [2018]; Marlina and Purwanto [2019]). In addition to increasing the amount of OH· in the solution, the use of high currents increases the regeneration of iron ions, which increases the effectiveness of the EF process. The degradation of organics by the electro-Fenton process is dependent on the appropriate Fe^{2+} concentration as well as the H_2O_2 concentration (Xia et al. [2015]). Higher electro-regeneration of Fe^{+2} ion than Fe^{+3} ion with increasing current increases the efficiency of EF chain reactions (Zhang et al. [2007]). Conversely, when a low current density is applied, it shows inhibition of the degradation rate due to the low concentration of oxidants produced; thus, Eq. ([1]) gives a smaller concentration of OH· radical point (Mohajeri et al. [2010]). For this reason, the current value to be used in the system should be well determined in order not to increase the cost arising from energy.

In order to examine the effect of current density on the COD, AOX and TOC removal efficiencies in the EF process, the current density was applied by changing it in the range of 50–300 A/m² and is presented in Fig. [3]. In the experimental study on the effect of current density, pH 3, H_2O_2 concentration was taken as 500 mg/L and electrolysis time was taken as 30 min and all experiments were carried out at room temperature. As shown in Fig. [3], there is an increase in the COD, TOC and AOX removal efficiencies when the current density is increased from 50 to 150 A/m² in the first 10 min of the electrolysis period. This increasing trend can be attributed to more OH· production at the anode surface as

well as higher electro-regeneration (Eq. 11) of ferrous ions from ferric ions at high current density, which increases the efficiency of Fenton chain reactions. The continuous conversion of Fe^{3+} to Fe^{2+} is a major advantage over chemical Fenton systems.

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \tag{11}$$
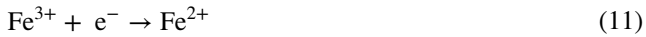
The highest removal efficiencies for all pollutant parameters were obtained at 150 A/m² current density. The graphs show that the optimum operating conditions are approximately pH 3, H_2O_2 500 mg/L, current density 150 A/m² and reaction time of 15 min. Under these conditions, COD, TOC and AOX removals were obtained as 71.7, 91.72 and 97.4%, respectively. Moving away from these points indicates decreased removal efficiency. This means that no increase or decrease is desired in any of the variables tested. The low removal efficiencies at low currents may be due to an inhibiting effect of the degradation rate, possibly due to the low concentration of oxidants produced. Figure 3 shows that the electrolysis time has a positive effect on the mineralization of the leachate and the removal of pollutant parameters. It is noted that the maximum COD, TOC and AOX removals are achieved in about half an electrolysis time of the studied range. After that, the removal efficiency did not change significantly. Obtaining the highest removal efficiencies at this current density means that the degree of anodic dissolution

of the iron increases. This situation explains the greater precipitate formation to remove contaminants. On the other hand, as the bubble size gets smaller with increasing current density, the bubble formation rate increases, and this results in higher pollutant removal efficiency with H_2 flotation tools (Orkun and Kuleyin 2012).

In the EF process, Fe^{2+} given to the environment is supplied from the dissolving iron electrodes depending on the current density at the anode, and it appears with a certain current density in a sufficient amount of solution. In Fenton and similar oxidation processes, the production of hydroxyl radicals generally decreases with the rapid depletion of Fe^{2+}. The reason for this is explained as being by researchers in similar studies that the rate constant of the reaction between OH· and Fe^{2+} (3.2 108 M-s-) is approximately 10 times the rate constant of the reaction between OH· and H_2O_2 (Qiang et al. 2003; Choi et al. 2010; He and Zhou 2017). Therefore, rapid regeneration of Fe^{2+} is essential for the continuous production of OH. Although H_2O_2 and Fe^{2+} can be produced simultaneously, the applied electrical force primarily favours the formation of H_2O_2. Again, in this method, the OH· radical can be produced in a controlled manner in direct proportion to the electric current used. Based on this feature, it is easily possible to correlate the reactions of other substances with the OH· radical concentration, with the amount of electric current used. Higher applied current density means higher applied voltage in the electrochemical system (Wang et al. 2010; Nidheesh and Gandhimathi 2012). However,
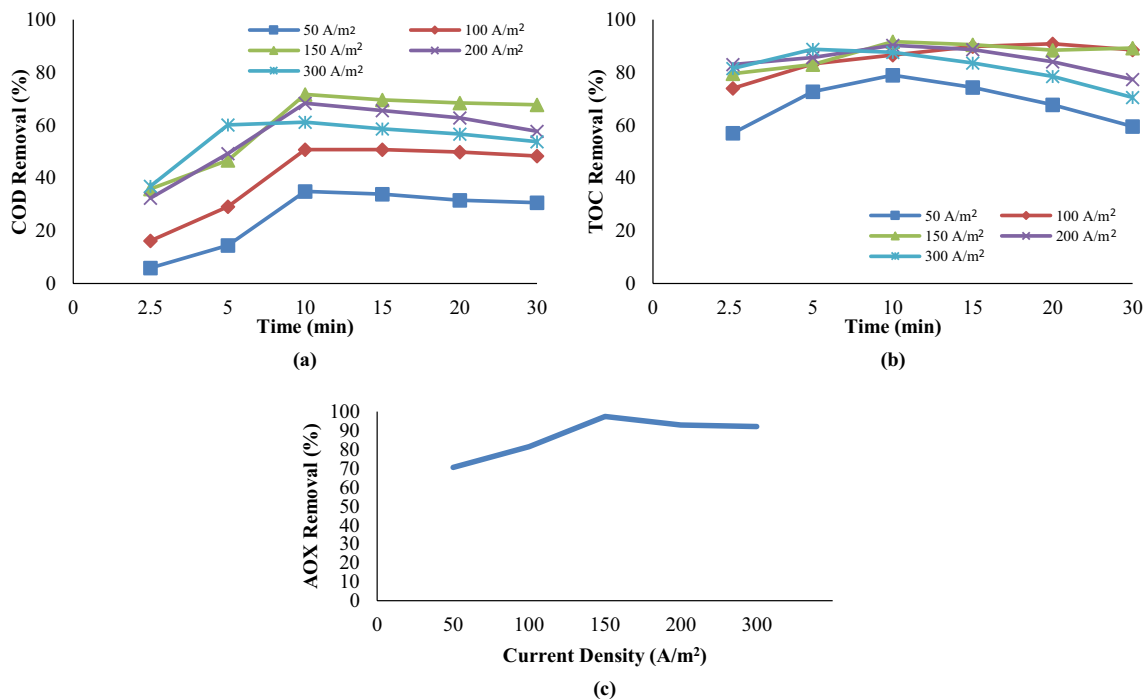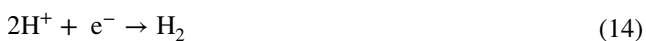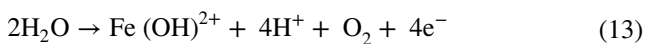

Fig. 3 Effect of current density on **a** COD, **b** TOC and **c** AOX removal (pH: 3, H_2O_2: 500 mg/L, reaction time: 30 min)

in high-current conditions, while the energy consumption increases, the efficiency does not show much change. The degradation rate of organic pollutants is constant after 300 mA. This is due to the H_2O formation as in Eq. (12) (Ozcan et al. 2008). The efficiency of the EF will be realized less at higher current density. The reason for this is due to competitive electrode reactions in the electrolytic cell. Further increase in current density will cause competitive electrode reactions such as oxygen discharge at the anode (Eq. 13) and hydrogen production at the cathode (Eq. 14). These reactions inhibit the main reactions (Eqs. 12–15) and cause a decrease in EF activity (Zhang et al. 2006; Korbahti and Tanyolaç 2008; Varank et al. 2020).

$$4H^+ + O_2 + 4e^- \rightarrow 2H_2O \tag{12}$$

$$2H_2O \rightarrow Fe(OH)^{2+} + 4H^+ + O_2 + 4e^- \tag{13}$$

$$2H^+ + e^- \rightarrow H_2 \tag{14}$$

$$2H_2O \rightarrow HO \bullet + H^+ + 4e^- \tag{15}$$

Oxidation reduction potential (ORP) is a measure of water's cleanliness and ability to break down contaminants and is measured in millivolts (mV). In short, the ORP measurement determines the quality of the water. If the result of the measurement is positive, it indicates that the oxidation, that is, rusting and spoiling/corrosive effects, of this water is dominant; if a negative value is found, this water has reducing, that is, antioxidant power. ORP has a close relationship with the specific electrical charge during the electro-oxidation process and can affect the COD removal efficiency. 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 the electro-oxidation process (Wang et al. 2018; Mei et al. 2019). The change of ORP values over time at different current density values in the EF process is shown in Fig. 4. ORP decreases with increasing current density and electrolysis time. This

decrease value accelerated, especially with the increase in the current density. This can be explained by the inability of the Fe electrode used as the anode in the EF process to maintain its oxidation potential in the environment. During the electrolysis, the iron dissolved in the anode passes into the environment as Fe^{+2}, and the OH^- ions formed, especially at the cathode, during the electrolysis of water rapidly degrade the H_2O_2 in the environment and the oxidation potential decreases.

Effect of H_2O_2 concentration

The main source of OH· radicals that will form in the system in EF oxidation is H_2O_2. H_2O_2 consumption in F and EF reactions is one of the most important factors limiting the effectiveness of the method (Teymori et al. 2020). In order to determine the optimum value of H_2O_2 concentration, which is one of the components of the EF process, different doses of H_2O_2 between 0.25 and 2.0 g/L were added to the leachate ($H_2O_2/Fe^{+2} \cong 1$). In the study, the treatment efficiencies obtained as a result of the experiments carried out to determine the relationship between different H_2O_2 concentration and the treatment efficiency at constant pH(3) and current density (150 A/m^2) are presented in Fig. 5. With the increase in the H_2O_2 concentration, the removal efficiency of pollutants increases (Ting et al. 2009). As the initial H_2O_2 concentration added to the system increases, the amount of OH· radical formed increases, resulting in an increase in COD removal (Fig. 5 a). Since hydroxyl radicals are very active and fast from the moment they are formed in the environment, they react in a short time with the organic substances in the environment and increase the removal efficiency. It has been observed that COD removal increased up to 71.7% at 0.5 g/L H_2O_2 concentration in 10 min oxidation time. In similar studies, it has been reported that reduced feeding time increases the rate of COD removal at baseline (Lopez et al. 2004; Zhang et al. 2006; Umar et al. 2010; Nidheesh and Gandhimathi 2012). A rapid COD reduction in the first 10 min of the process may be due to oxidation of easily

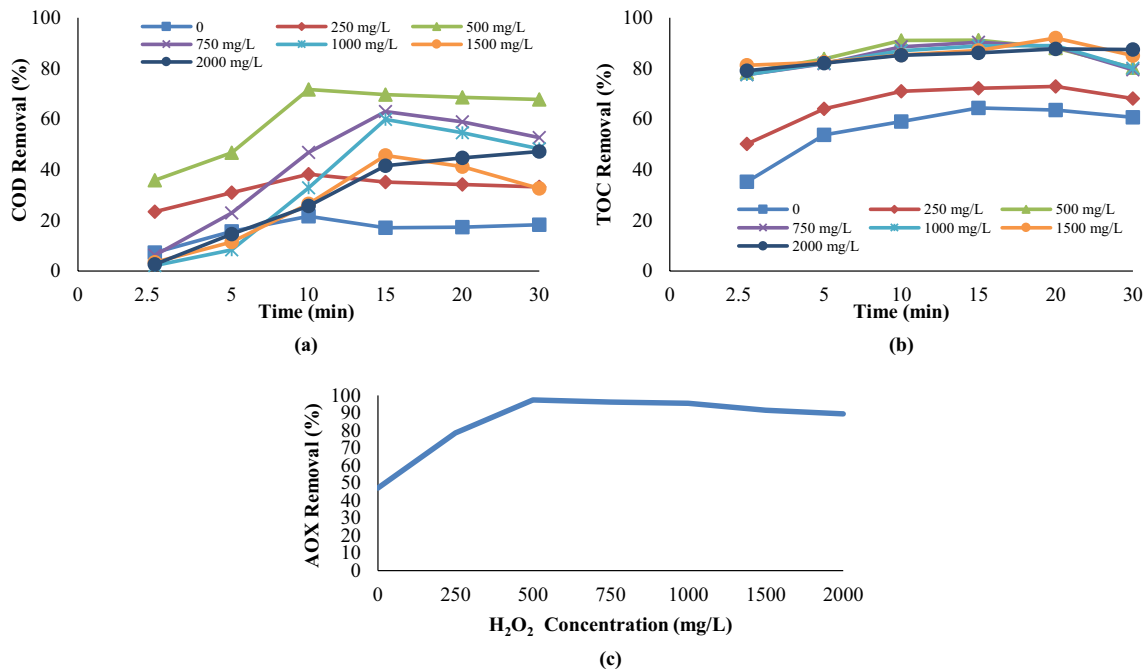**Fig. 4** ORP change over time in the EF process
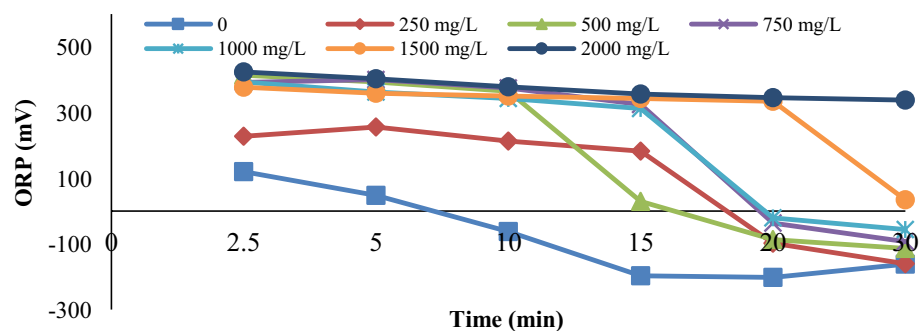
Fig. 5 Effect of H$_2$O$_2$ concentration on **a** COD, **b** TOC and **c** AOX removal (pH: 3, current density: 150 A/m^2, electrolysis time: 30 min)

degradable organic compounds. However, the reduction in removal rates at other reaction times (10-30 min) is probably due to a gradual slowing of the rate of degradation due to the formation of short-chain organic acids and hardly oxidizable by-products (Khajouei et al. 2019). The purpose of chemical oxidation is to remove the organic pollutant from the environment without creating a different pollutant or toxic product as an intermediate product while oxidizing. Otherwise, toxic substances may form as intermediate products and cause low yield. In the oxidation time (10 min.) where the highest removal efficiency was obtained, the removals of COD, TOC and AOX were found to be 97.4, 91.03 and 97.4%, respectively (Figs. 5, 6). It is seen that the removal efficiencies remain almost constant at values above the H$_2$O$_2$ concentration, where sufficient oxidant concentration is provided (0.5 g/L). In trials where higher concentration values (0.75–2.0 g/L H$_2$O$_2$) were applied, no significant increase in COD removal was achieved. Further increase in H$_2$O$_2$ from 0.5 to 2 g/L decreased the COD removal efficiency by 4%. At higher hydrogen peroxide concentrations, a decrease in treatment efficiency was observed due to increased reactions as in Eq. (16) and recombination of hydroxyl radicals and other scavenging reaction Eq. (18), which retard the efficiency of the Fenton process. Similar results were also reported by Daud et al. (2013), Xavier et al. (2015), Nidheesh and Rajan (2016), Hassan et al. (2017) and Güvenç et al. (2019). Another reason for this may be the decrease
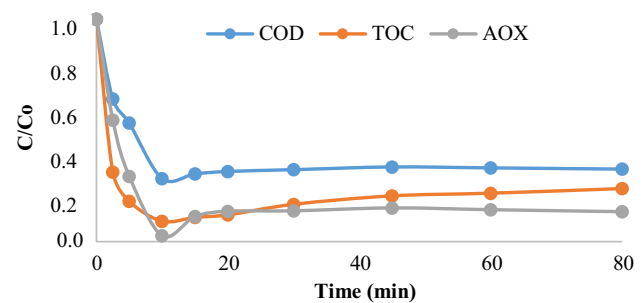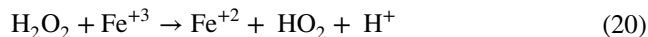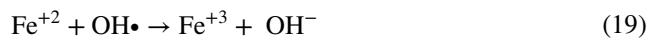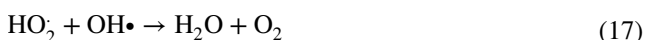

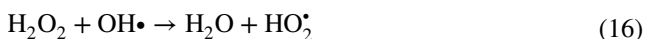**Fig. 6** Effect of reaction time on COD, AOX and TOC removal (pH: 3, current density: 150 A/m^2, H$_2$O$_2$ concentration: 500 mg/L)

in the H$_2$O$_2$/Fe(II) ratio as more of the iron plates dissolve and go into solution as explained below.

To maximize the efficiency of the process, it is crucial to determine the optimum operational H$_2$O$_2$/Fe^{2+} molar ratio. The increase in H$_2$O$_2$/Fe(II) molar concentration in electro-Fenton oxidation causes parasitic reactions in the system to dominate. As the dose of H$_2$O$_2$ used as an oxidant increases, oxidation of organic substances takes place, but after a certain time there is little or no change in the environment. This process continues until the H$_2$O$_2$ threshold is reached. A rapid reduction in aquatic toxicity occurs with continued addition of the oxidant after the threshold point. For this reason, a certain amount of H$_2$O$_2$ must be given in the EF process (Khatri et al. 2018; Kerboua et al. 2021). A decrease in the purification efficiency has been observed at higher hydrogen peroxide

concentrations, due to the increased reactions between hydrogen peroxide and hydroxyl radicals. The reduction in removal efficiency at a high dose of H_2O_2 is due to the scavenging effect of H_2O_2 hydroxyl radicals (Eqs. 16, 17) and recombination of hydroxyl radicals (Eq. 18) (Xavier et al. 2015; Wang et al. 2022). On the other hand, the formation of OOH·, which is significantly less reactive than hydroxyl radicals, also reduces the reaction rate (Babuponnusami and Muthukumar 2012; Rahmani et al. 2015). The concentration of the hydroxyl radical, which is the main oxidizing agent in the EF process, increases with the increase in the Fe^{2+} concentration. However, when there is an excess of iron ions in the electrolyte solution, it can consume hydroxyl radicals and affect the degree of degradation (Babuponnusami and Muthukumar 2014). Mohajeri et al., (2010, 2019) emphasized that the removal efficiency decreases with any increase in the H_2O_2/Fe^{2+} molar ratio above 1 in their study on landfill leachate treatment by electro-Fenton oxidation. They interpreted that the reason for this may be the change in the reaction mechanisms of Fenton and the emergence of some side reactions, and that excess hydrogen peroxide has a suppressive effect (Eq. 16). This reaction leads to the production of the hydroperoxyl radical, a species with much weaker oxidizing power compared to the hydroxyl radical, and at the same time, an excess of hydrogen peroxide can cause spontaneous decomposition of H_2O_2 into oxygen and water and recombination of OH radicals. This phenomenon of dissociation and recombination reduces the concentration of hydroxyl radicals and the efficiency of compound elimination (Eqs. 17, 18) (Ting et al. 2009; Lodha and Chaudhari 2007; Bautista et al. 2007; Ay et al. 2008). Moreover, at higher iron catalyst dosages, excess Fe^{2+} will act as hydroxyl radical scavenger and oxidized to Fe^{3+} as shown in Eq. (19). Thus, the hydroxyl radical available for the oxidation of organic matter will decrease and the COD removal efficiency will decrease. At lower doses, the amount of Fe^{2+} leaching will be insufficient for sufficient hydroxyl radical production and COD removal efficiency will decrease (Baiju et al. 2018). On the other hand, when the molar ratio (H_2O_2/Fe^{2+}) is low, COD removal decreases due to the scavenging effect of excess Fe^{2+} (Eq. 19). There is a competition between Fe^{2+} and organic compounds for hydroxyl radicals, resulting in reduced COD and TOC removal efficiencies. Hydroxyl radicals generated via Fenton reactions react with excess Fe^{+2} ions present and get converted into its ionic form. Similar results were also reported by Laiju et al (2014), Sruthi et al (2018). Furthermore, the Fe^{3+} formed can react with H_2O_2 (Eq. 20) to form Fe^{2+} and hydroperoxyl radicals (HO_2) in solution (Meriç et al. 2004; Badawy and Ali 2006; Deng 2007; Lee and Shoda 2008).

$$H_2O_2 + OH\bullet \rightarrow H_2O + HO_2^{\bullet} \quad (16)$$

$$HO_2^{\bullet} + OH\bullet \rightarrow H_2O + O_2 \quad (17)$$

$$HO \bullet + OH\bullet \rightarrow H_2O_2 \quad (18)$$

$$Fe^{+2} + OH\bullet \rightarrow Fe^{+3} + OH^- \quad (19)$$

$$H_2O_2 + Fe^{+3} \rightarrow Fe^{+2} + HO_2 + H^+ \quad (20)$$

In terms of removal efficiency, it is seen that TOC and AOX parameters are not much affected by the H_2O_2 feeding mode (Fig. 5 b-c). Del Moro et al. (2016) they have emphasized in their study on the treatment of landfill leachate that the AOX profile largely depends on the applied operating conditions (current density, reaction time, etc.) and the type of organic compounds (aliphatic or aromatic compounds) in the leachate. In addition to these findings in other studies, it was concluded that working at higher current densities is more efficient in terms of the formation and cascading of HOCs. When the other studies on AOX removal were examined, different AOX formation and removal profiles were obtained under variable experimental working conditions. Study results show that AOX formation generally increases during the initial stages of reaction times, followed by removal of AOX (Zhang et al. 2018a, b; Iskurt et al. 2022).

In Fig. 7, the variation of ORP at different H_2O_2 concentrations with time is given. As the oxidant concentration increases, the oxidation potential of the medium increases and it becomes more stable. This situation can be explained by the rise in the concentration of H_2O_2, as already explained.

In addition, a spectrum scan was carried out in the wavelength range (WR) of 190–1100 nm, of the output samples obtained under optimum conditions with the highest treatment efficiency (Fig. 8). In the spectrum scanning of the samples taken at certain time intervals (2–80 min), the conversion of organic substances into different compounds as a result of the reaction with OH· radicals was examined, and an evaluation was made in terms of optimum pH value. According to the results obtained from the analyses, it was verified from the spectrum scanning that the optimum electrolysis time (10 min) and pH (3) values were determined as optimum values.

Sludge sedimentability and economic analysis

Sludge formation is influenced by many factors, such as the characteristics of the treated wastewater and the applied treatment parameters such as current density and reaction time. In some cases, COD removal and the volume of sludge produced should be evaluated together to determine the optimum process parameters (Guven 2021). One of the most important advantages of electrochemical methods over other treatment methods is that less sludge is generated as a result of treatment. However, it still requires the addition of iron
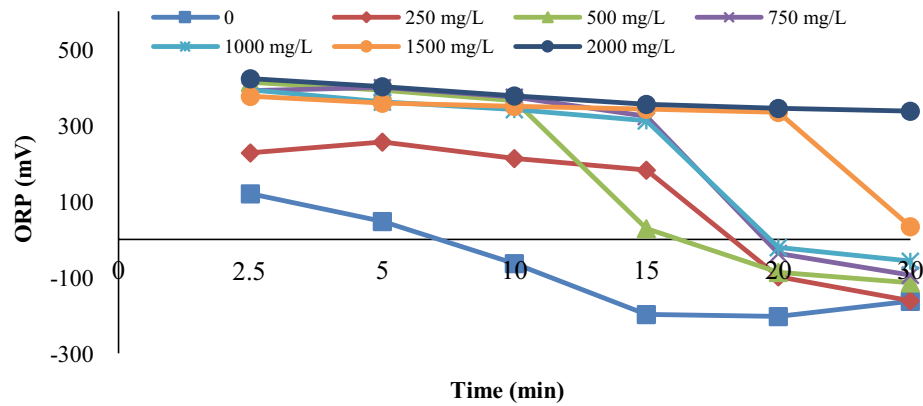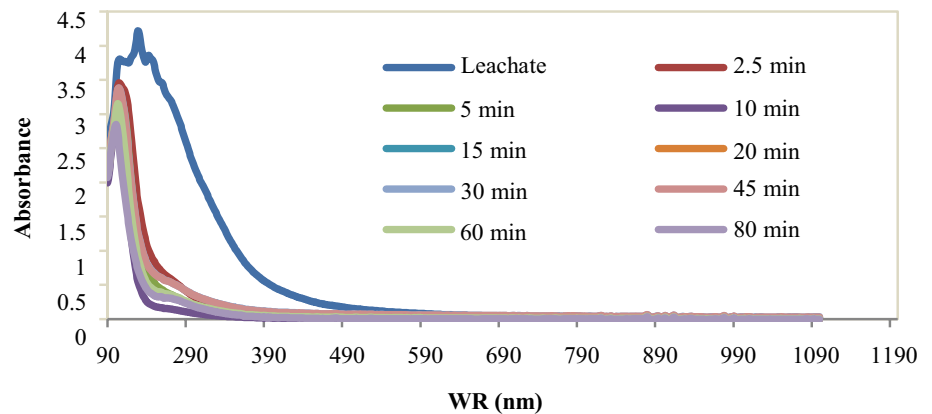
Fig. 7 Time-dependent ORP change in EF process

Fig. 8 Spectrum scanning at optimum conditions based on time in the EF process

or H_2O_2, which can easily cause iron sludge precipitation (Zhang et al. 2018a, b; Lu 2021; Meng et al. 2022). With the increase in the electrolysis time, iron ions accumulate in the aqueous medium and lead to the precipitation of $Fe(OH)_3$; however, it can reduce the amount of electricity use. Thus, these types of pollutants are decomposed by oxidation and coagulation, but the sludge is not treated. On the other hand, sludge production can be reduced by the EF process, in which H_2O_2 is added from the outside and Fe^{2+} is produced in situ (Nidheesh et al. 2018b; Casado 2019; Zhang et al. 2019). The electro-Fenton system can continuously regenerate iron ions from ferric ions and further reduce ferric sludge compared with traditional Fenton. EF technology requires a smaller dosage and produces less iron sludge (Casado 2019; Li et al. 2022b). Due to the large flocs formed in the unit, the settling characteristics of such sludge are quite good. However, gas bubbles occurring at the anode and cathode can increase the sedimentation time of the sludge formed (Xu et al. 2020). Generally, the settling characteristics of the formed sludge are determined by the sludge volume index (SVI). SVI is generally used to control the settling properties of biological suspensions and is obtained by dividing the volume of precipitating sludge (mL/L) by the ratio of suspended solids (mg/L). Sludge with an SVI value of < 100 is considered to have good settling characteristics. Table 3

Table 3 SVI values of sludge obtained in different operating conditions in the EF process

pH	SVI (mL/g)	Current density (A/m²)	SVI (mL/g)	H_2O_2 (mg/L)	SVI (mL/g)
2	102.94	50	125	0	–
2.5	82.67	100	109.75	250	199.07
3	**67.92**	**150**	**69.23**	**500**	**73.39**
3.5	77.25	200	80.97	750	65.93
4	106.19	300	105.46	1000	69.95
7	109.37			1500	72.36
				2000	90.63

Bold indicates the pH results (pH 3) which is best condition

shows the SVI values of the sludge obtained in the EF process under different operating conditions. As can be seen from the table, when the effect of initial pH on the sludge settleability was examined, it was observed that the SVI value decreased at pH 3. This event suggests that $Fe(OH)_n$ flocs may occur as larger and stable flocs under slightly acidic conditions. It was detected that SVI values increased with increasing current density and H_2O_2 concentration. The reason for the increase observed in high H_2O_2 concentrations can be considered as the deterioration of the flock structure

of H_2 and O_2 in the reactions occurring at the anode and cathode and decreasing the sedimentation ability of the sludge. However, an increase in anode dissolution increases sludge formation during the electrolysis period, resulting in an additional secondary treatment cost (Ling et al. 2022).

Technoeconomic analysis of EF systems shows that raw materials have the highest share in total costs compared to electricity costs for highly efficient anodic materials and heterogeneous catalysts such as Fe, Ni, Pd, Pt and Ag, which can be found in all three phases of matter. Economic cost is a strong limiting parameter for large-scale implementation of EF systems. To overcome this limitation, the EF method should be highly competitive in terms of cost compared to conventional wastewater treatment methods currently available (Popat et al. 2019). Since EF has the ability to completely oxidize for wastewater treatment, it seems more economically logical to combine other low-cost alternative treatment systems such as biochemical treatment for leachate treatment. Economic costs in EF systems can be measured in terms of expenditure and operating costs. Because each type of EF system has a variety of reactor designs and optimal operating factors, the overall process cost can vary greatly. Close attention should be paid to the economic evaluation of energy consumption, which represents the largest share of the total operating cost of the EF system, despite the different cost-inducing parameters. Other costs such as electrode material, electrical energy and chemical consumption, maintenance and labour costs are considered to be a part of the total operating cost (Varank et al. 2018; Ismail et al. 2021; Shokri and Fard 2022). In the literature, chemical material has been measured according to electrode material ($/kg) and electrical energy ($/kWh) costs (Kobya et al. 2009).

The operating cost was determined using Eq. 21 (results not shown here).

$$\text{Operating Cost} \left(\$/m^3 kg\right) = 0,06C \text{ energy} + 0,6C \text{ electrode} + 1,0034\ CH_2O_2 \tag{21}$$

In this equation, C_{energy} is the energy consumption in EF processes (kWh/m^3), $C_{electrode}$ is the amount of solute aluminium or ferrous metal electrode in the EF reactor (kg electrode/m^3 of treated leachate water), and CH_2O_2 represents the amount of chemical consumed in the EF process.

When evaluated in terms of operating costs, it has been determined that in terms of treated leachate ($/$m^3$), in high-current-density conditions, energy consumption increases while the removal efficiency does not change much. In order not to increase the cost of energy, the current value to be used in the system should be well determined. Experimental studies examining the effect of current density on operating cost, stirring speed 250 rpm, current density 50–300 A/m^2, electrolysis time 30 min, pH 3 and H_2O_2 concentration 500 mg/L have been carried out under initial conditions.

When examined in terms of cost and removal efficiencies, it has been determined that 150 A/m^2 current density values are more economical than other current density values. In addition, as the current density increases, energy consumption and operating costs also increase (Wang et al. 2020). The lower value of current density requires longer time for pollutant removal, which requires larger facilities and operating costs. In contrast, higher value of current density causes energy wastage due to partial electrical energy heating and increases power consumption and operating costs (Atmaca 2009; Sahinkaya 2013). The application of significant current density can significantly increase process energy consumption with a negligible increase in system performance. Therefore, current density should be operated at an optimum level to maximize pollutant removal efficiency with minimum power consumption. As the addition of chemical substance increases, the operating cost increases in direct proportion. Similar results were confirmed by Alavi et al. (2019), Rouidi et al. (2020), Asaithambi et al. (2020) and Yu et al. (2023). Asaithambi et al. (2020), in their study on the removal of COD from landfill leachate wastewater using EAOPs and determination of power consumption, emphasized that the percentage of COD removal increased from 58.50 to 97% when the H_2O_2 concentration was increased from 75 to 300 mg/L. However, when the H_2O_2 concentration was increased to 450 mg/L, COD removal has fallen percentage from 97 to 80.50% and the power consumption linearly from 4 to 2.45 kWh/m^3. This is because at low H_2O_2 concentration, COD removal is very slow due to insufficient radical $\cdot OH$ production. The increase in H_2O_2 concentration increased the COD removal with the formation of a larger amount $\cdot OH$. Increasing the COD concentration further, it has reduced the %COD removal as well as the power consumption by half at the appropriate reaction times. However, it was reported that % COD removal was not observed significantly at high H_2O_2 concentrations. This is due to the recombination of hydroxyl radicals, the scavenging effect of H_2O_2 and the inhibition of iron corrosion by H_2O_2, as described previously. The hydroperoxyl radical ($\cdot HO_2$) produced from hydrogen peroxide decomposition is a weak oxidant compared to $\cdot OH$. In addition, excessive use of H_2O_2 will also interfere with COD measurement since the remaining amounts of H_2O_2 can deplete $K_2Cr_2O_7$ and significantly increase COD levels (Yazdanbakhsh et al. 2015; He and Zhou 2017). For this reason, considering the removal efficiency, 500 mg/L H_2O_2 concentration is seen as the most suitable value.

Reaction time plays an important role in electrocoagulation. Due to the increased energy consumption and operating cost, there must be sufficient electrolysis time for the reaction to occur. As the electrolysis time increases, so do the mixing process, current flow and anode dissolution. At the appropriate electrolysis time, more significant amount

of hydroxyl and metal ions, which act as coagulants, will be produced from the electrodes and thus the removal performance will be improved (Ling et al. 2022). This situation may explain the fact that in the longer reaction time, H_2O_2 produces a large amount of hydrogen peroxide, which causes spontaneous decomposition into oxygen and water and recombination of OH· radicals. Thus, the concentration of hydroxyl radicals and, accordingly, their removal efficiency decrease (Thirugnanasambandham et al. 2015). In the experimental study, in which the effect of the electrolysis time on the operating cost was evaluated, the experiments were carried out by changing the electrolysis time in the range of 2.5–80 min under optimum conditions (pH: 3, current density: 150 A/m^2, H_2O_2 concentration: 500 mg/L and 250 rpm mixing speed). Considering the removal efficiencies in the EF process, the highest removal values were reached in 10 min of electrolysis time.

Comparison of COD and AOX removal from leachate in different process

A brief summary of the studies on landfill leachate removal by advanced oxidation processes (EAOPs) such as Fenton (F) and electro-Fenton (EF) has been presented in Table 4. The current results in the literature on pollutant removals and energy consumption under different systems are presented in the table. As can be seen, all relevant processes show the result that it can efficiently remove organics from leachate with COD removal ranging from 57 to 89%. For example, it has been highlighted that successful results were obtained in the removal of organic materials from BorjChakir (Tunisia) landfill leachate with the EF process. Experimental design methodology has been used to determine the optimal operating conditions for efficient mineralization of organic matter in landfill leachate. In the most optimal conditions (I = 1000 mA, purification time = 8 h and pH = 3), 78% COD removal has been achieved. Based on the experimental results obtained, the authors emphasized that the EF process is a viable environmentally friendly technology for the efficient treatment of landfill leachate and can be generalized to wastewater treatment (Trabelsi et al. 2012). Khajouei et al. (2019) used response surface methodology in their study on the treatment of composite leachate and they have done optimized the EF process based on the D-optimal algorithm. Removal efficiencies of 68.4, 78.7 and 99.3%, respectively, were obtained for COD, BOD and PO$_4$-P under optimum conditions (pH = 3, [H_2O_2] = 0.25 M, electric current = 3A and electrolysis time = 100 min). A decrease in the biodegradability index of BOD$_5$/COD from 0.60 to 0.31 indicated that most of the organic matter in the compost leachate was biodegradable and oxidized by the EF process. The

authors concluded that the EF process can be considered a promising and cost-effective alternative to by the Fenton process in treating compost leachate. Ling et al. (2022), in their study, compared the efficiency of electrocoagulation (EC) and electro-Fenton (EF) processes for treatment of landfill leachate. The influence of operating parameters including initial pH, contact time and a COD/H_2O_2 mass ratio for COD and colour removal from leachate has investigated. Under optimum pH (6) and reaction time value (17.5 min) in EC process, 66.2 and 94.4% COD and colour removal were obtained in EC treatment, respectively. In the EF process, 69.8% colour and 88.1% COD removal (COD:H_2O_2 9.5) were obtained with optimum pH (9) value and electrolysis time (6.7 min). According to the results obtained, higher treatment efficiency of the EF process than the EC process was obtained. In addition, it was determined that 46.7% less sludge was produced in the EF process under optimal conditions compared to the EC process. The results of the study showed that the treatment operating cost of the EF process is 35.8% lower than the EC process. It has been emphasized that the EF system is an energy efficient process for the removal of waste leachate, due to its many advantages. In Crispim et al. (2022), they have researched the feasibility of electro-Fenton (EF) and photoelectro-Fenton (PEF) processes as alternatives for the treatment of local landfill leachate with high organic content (COD = 2684.7 mg/L). For this purpose, a continuous flow reactor was designed using boron doped diamond anode (Nb/BDD) and carbon felt cathode (FC) electrodes. The effects of current density (30, 60 and 90 mA/cm^2) and UV radiation wavelength (UVA and UVC) have been investigated to evaluate the energy consumption and treatment efficiency between the two processes. The best efficiency for removing organic matter (COD) has been found to be 66, 68 and 89% for EF, PEF UVA and PEF UVC, respectively, at 4-h electrolysis time and a current density of 90 mA/cm^2. In the same conditions, the energy consumption was obtained as 19.41, 17.61 and 17.59 kWh kg/COD. According to the results of the study, it has been reported that the percentage of COD removal occurred in the order of EF < PEF UVA < PEF UVC.

Considering the response relationship between treatment cost and treatment effect, including reducing the pollution load and toxicity of leachate, it is clearly understood from studies that some electrochemical AOP techniques need to be combined to further improve the mineralization rate of landfill leachate and reduce unnecessary reaction time. In addition, the optimization of stable operating parameters and its combination with other technologies to achieve the desired improvement effect requires further evaluation.

Table 4 Comparison of pollution parameter removals from waste landfill leachate by advanced oxidation processes

Process	Electrodes	Current density (mA/cm^2)	H$_2$O$_2$ or Fe^{2+}	COD$_0$ (or AOX, BOD) (mg/L)	Volume treated (mL)	Time (min)	COD (or AOX) Removal (%)	kWh/kg COD	References
EF	Anode: Pt Cathode: carbon felt	1000	Fe^{+2} = 0.135 mM	10,200	400	480	78	–	Trabelsi et al. (2012)
EF	Anode: Fe Cathode: Fe	–	H2O2=7.5,15,100,200 mg/L	COD=70,000–90000	1000	180	COD=80%	–	Aval et al. (2017)
EF	Anode: Fe Cathode: Fe	15	Fe=560 mg/L	COD=3100±400	1800	360	COD=71±.6%	207±20	Hu et al. (2018)
EF	Anode: Fe Cathode: Fe	2.0 A	Fe=15 mM	COD=1625	800	120	COD=74.7%	5.76	Li et al. (2022b)
EF	Anode: Fe Cathode: Fe	3.0 A	H$_2$O$_2$=0.25 M	COD=12,560 BOD=7540 PO$_4$-P=4.5	1000	100	COD=63.4% BOD=78.7% PO$_4$-P=99.3%	–	Khajouei et al. (2019)
EC/EF	Anode: IrO$_2$–RuO$_2$/TiOx Cathode: CF	30	Fe^{2+} = 1 mM	300–1600	1000	–	COD=57%	67.8	Ding et al. (2021)
EF	Anode:Ti/RuO$_2$–IrO$_2$ Cathode=CF	50.42	Fe^{2+} = 0.11 M H$_2$O$_2$ = 0.34 M	5000	200	120	COD=57%	–	Zhang et al. (2006)
EC + EF	Anode: Fe Cathode=Fe	–	COD:H$_2$O$_2$=9.5	COD=10,800 Colour=14,750	500	17.5 6.7	COD=66.2% Colour=94.4% COD=69.8% Colour=88.1%	RM 3.13/m^3 RM 2.01/m^3	Ling et al. (2022)
EF PEF–UVA PEF–UVC	Anode: (Nb/BDD) Cathode=FC	90		COD=2684 TOC=1220	1000	240	COD=66% COD=68% COD=89.1% TOC=89.9%	19,41 17,61 17,59	Crispim et al. (2022)
EF	Anode: Fe Cathode=Fe	2.5 A	H$_2$O$_2$/COD=0.6	COD=28,000	600	60	COD=85%	-	Alavi et al. (2019)
EF	**Anode: Fe Cathode = Fe**	**150**	**H$_2$O$_2$=500 mg/L (H$_2$O$_2$:COD=4.4)**	**COD=2200 TOC=425 AOX=50**	**1000**	**30**	**COD=71.7% TOC=90.87% AOX=97.4%**	**(10 min) 1.771($)/m^3 (30 min) 2.234($)/m^3**	**This work**

Conclusion

One of the main problems in solid waste landfills is the remediation and management of leachate, which produces large quantities of organic matter. If leachate is discharged directly into the receiving environment or treated unsystematically, it can cause significant risks to health and the environment. There are many factors affecting the treatment efficiency of the EF technique. Therefore, experimental variables such as reaction time, pH, current density, H_2O_2 concentration and feed mode should be correctly selected. In this study, the applicability of EF system for the treatment of landfill leachate was investigated and the effect of operating parameters such as pH, current density, hydrogen peroxide concentration, COD, TOC and AOX removal efficiencies was evaluated in relation to operating cost and sludge amount. The results showed that the EF technique performs well in the treatment of landfill leachate. Based on the findings obtained in this study, the optimal variables of EF technique for treatment of landfill leachate were determined as pH 3, current density 150 A/m^2, H_2O_2 concentration 500 mg/L (H_2O_2:COD 4.4) and electrolysis time 10 min. Under these conditions, the COD, TOC and AOX removal efficiencies 71.7, 90.87 and 97.4%, respectively, have been obtained. The results obtained show that the leachate meets the discharge standards for Turkey in terms of the investigated parameters. In addition, within the scope of this study, the collapsibility and operating costs of the sludge formed as a result of the EF process were also investigated and it was determined that the sludge formed as a result of the EF process had good settling properties. The sedimentation properties of the sludge formed after treatment (sludge volumes after 30 min. of settling: V69.23) are quite good. However, it is possible to further improve the settling volume. Thus, the disposal costs of the waste sludge produced by the EF technique will be significantly reduced. Electro-Fenton process has many advantages such as less sludge production, shorter treatment time, lower operating costs and continuous Fe^{2+} regeneration. Considering these advantages, it appears to be an economically and environmentally viable, which can be considered as promising technology when compared to other advanced oxidation processes. However, these data alone are not sufficient for an effective evaluation of the economic feasibility of the process. Therefore, future research should focus on the economic cost of the electro-Fenton process, such as reactor type, electrode used, energy and chemical expenditure costs, depending largely on suitable processes through which higher removal efficiencies will be achieved.

Declarations

Conflict of interest The authors declare that they have no competing financial interests or personal relationships that appear to affect the work reported in this article.

Consent for publication Not applicable.

Ethical approval Not applicable.

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