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Evaluating the efficiency of electrochemical process in removing COD and NH_4-N from landfill leachate

M. Pirsaheb[a], E. Azizi[a,]*, A. Almasi[b], M. Soltanian[b], T. Khosravi[c], M. Ghayebzadeh[a], K. Sharafi[c,d]

[a]*Environmental Health Engineering Department, Public Health School, Research Center for Environmental Determinants of Health, Kermanshah University of Medical Sciences, Kermanshah, Iran, Tel. +98 9123446880; email: mpirsaheb@yahoo.com (M. Pirsaheb), Tel. +98 9143896906; Fax: +98 8138263048; email: azizi.esmaeel@gmail.com (E. Azizi), Tel. +98 9141586994; email: m.ghayebzadeh@gmail.com (M. Ghayebzadeh)*
[b]*Environmental Health Engineering Department, Public Health School, Health Research Center, Kermanshah University of Medical Sciences, Kermanshah, Iran, Tel. +98 9181317314; email: alialmasi@yahoo.com (A. Almasi), Tel. +98 9181312531; email: msoltanian_ehe@yahoo.com (M. Soltanian)*
[c]*Environmental Health Engineering Department, Kermanshah University of Medical Sciences, Kermanshah, Iran, Tel. +98 9189318647; emails: touba_khosravi@yahoo.com (T. Khosravi), kio.sharafi@yahoo.com (K. Sharafi)*
[d]*Department of Environmental Health Engineering, School of Public Health, Tehran University of Medical Science, Tehran, Iran*

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ABSTRACT

Landfilling leachate contains a high concentration of organic and inorganic pollutants. The present study was aimed to evaluate the efficiency of the electrocoagulation system in removing chemical oxygen demand (COD) and ammonia-nitrogen (NH_4-N) from leachate. The paper investigated the factors affecting removal efficiency such as current density (187.5, 375, and 562.5 A/m^2), electrode material (Al and Fe), and electrolysis time and pH of the solution (6.5 ± 0.2). The results indicated that, in optimum conditions, the highest COD and NH_4-N removal efficiencies were 66 and 63%, respectively. According to the results, performance of the Al electrode was better than that of the Fe electrode in terms of COD and NH_4-N removal. Therefore, the results showed that electrocoagulation can be applied for the leachate pretreatment as a feasible and reliable technique.

Keywords: Electrocoagulation; Landfill leachate; COD; NH_4-N

1. Introduction

Leachate is one of the main problems in the sanitary landfill, which contains large amounts of chemical oxygen demand (COD) and ammonia-nitrogen (NH_4-N) with strong color and odor, along with hazardous compounds such as aromatics and halogenated compounds, phenols, pesticides, and heavy metals [1–3]. Most of the time, concentration of COD and ammonium in leachate gets up to several 1,000 mg/L [4,5]. Therefore, discharging leachate without any suitable pretreatment causes a series of potential pollutant sources which threaten the environment and human life [6]. It should be noted that the concentration of ammonia in the landfilling leachate has been reported to be about 500–2,000 mg/L which is several times higher than the municipal wastewater.

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*Corresponding author.

However, decomposition of proteins in waste structures is the main source of ammonia in leachate [7]. It can be said that leachate is the most difficult effluent to treat among all the effluents due to high changes of leachate characteristics, complex composition, and quality fluctuation. Many studies have been carried out to find out suitable methods for treating the leachate [8,9]. Also, many treatment methods have been applied with various successful processes to treat leachate, such as aerobic biological [10] and anaerobic [11] biological processes, physico-chemical treatment [12,13], advanced oxidation process [14], and combined processes [6].

In the last two decades, electrocoagulation (EC) methods were used for the treatment of water and wastewater as a simple and efficient technique [15]. EC is a well-organized method which has some advantages such as less chemical requirement, cost-effectiveness, simple equipment, low retention time, intensity in varying reaction conditions, rapid sedimentation of electrogenerated flocculants, less sludge production, and high performance for the destruction of refractory pollutants and color removal [16–18]. It should be noted that EC–oxidation method has been successfully used for the treatment of textile dyeing wastewater [19], tannery wastewater [15], landfill leachate pretreatment [20,21], arsenic [22], olive oil mill wastewater [23], and petrochemical wastewater [24]. This technique involves *in situ* production of coagulants through the electrocatalytic oxidation of sacrificial anodes. Generally, two Al and Fe electrode materials have been used in EC process [25]. Destabilization and removal of particulate matters through the EC process occurs at the intersection of three fundamental stages: (a) coagulant: electrocatalytic oxidation of sacrificial electrode, (b) coagulation: destabilization of emulsion system and neutralization of surface electrical charge of colloids, and (c) flocculation: aggregation of suspended solids and flock generation [26]. According to the dissolution of electrode anode, generation of metallic hydroxide is an effective process in the destabilization of colloidal pollutants. In this technology, three main processes are used to remove pollutants: (a) electrical production of coagulants (EC), (b) electrical oxidation of pollutants (electro-oxidation), and (c) electroflotation [16].

1.1. EC mechanisms

Various electrochemical reactions simultaneously occur at the anode and cathode electrodes in the EC system.

1.1.1. Cathodic reactions

At the cathode electrode, H_2 gas is produced for the electrochemical reduction of H_2O in medium-alkaline condition (Eq. (1)) or reduction of the protons in the medium-acidic condition (Eq. (2)) [20]. Whenever the anode potential is adequately high, the second reaction might occur and oxygen gas is released (Eq. (3)). According to the high pH values of the solution, the metal cathode (Me) is chemically attacked by OH^- and H_2 gas is produced (Eq. (4)) [27].

$$2H_2O_{(l)} + 2e^- \rightarrow 2OH^-{}_{(aq)} + H_{2(g)} \tag{1}$$

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \tag{2}$$

$$2H_2O_{(l)} \rightarrow O_{2(g)} + 4H^+ + 4e^- \tag{3}$$

$$2Me + 6H_2O + 2OH^- \rightarrow 2Me\,(OH)_4^- + 3H_2 \tag{4}$$

1.1.2. Anodic reactions

Due to the electrocatalytic dissolution of sacrificial anodes, metal ions (Me^{n+}) are dissolved and distributed in electrolyte media (Eq. (5)) [28]. However, the generating metal ions (Fe^{2+} or Al^{3+}) combine with H_2O and hydroxyl ions to form the corresponding hydroxides (Eqs. (6)–(7)) [29,30].

$$Me_{(s)} \rightarrow Me^{n+}_{(aq)} + ne^- \tag{5}$$

$$Me^{n+} + H_2O \rightarrow Me(OH)^{n+} + H^+ \tag{6}$$

$$Me(OH)^{n+} + H_2O \rightarrow Me(OH)^{(n+)-1} + H^+ \tag{7}$$

According to (Eqs. (5)–(7)), whenever Al is used as the electrode material in appropriate conditions, the reaction occurs as follows.

$$Al_{(s)} \rightarrow Al^{3+} + 3e^- \tag{8}$$

When iron anodes are used, two mechanisms for the metal hydrolysis are reported as follows: [31]

$$\text{Mechanism 1: } Fe_{(s)} \rightarrow 4Fe^{2+}_{(aq)} + 8e^- \tag{9}$$

$$\text{Mechanism 2: } Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e^- \tag{10}$$

1.2. COD removal

In the electrochemical method, organic and inorganic materials are removed from aqueous solutions using various complex chemical reactions. Removal of organic materials through the EC system might be due to direct electrochemical oxidation on the anode surface and indirect electrochemical oxidation mediated by hydroxyl radicals, peroxodisulfates (in the presence of SO_4^{2-}), or active chlorine (in the presence of Cl^-). Totally, two main mechanisms in the EC process remove COD from aqueous medium as follows: (I) coagulation of particulate and colloidal organics due to the neutralization of surface electrical charge on colloids and making the suspension instable, and (II) oxidation of organic materials through the production of hydroxyl radicals and other oxidation agents [3]. During the EC process, hydroxides of metal ions (Al/Fe) will be formed. Therefore, these ions are hydrolyzed to form various Al or Fe polymeric species. These metal hydroxides have large surface area, high adsorption properties, and high capability to aggregate suspended solids and convert them into flock, as a result of binding to the pollutant, rapid adsorption of soluble organic compounds, and occurrence of trapping colloidal particles. These flakes are removed easily from the aqueous solution by sedimentation or flotation due to the settlement of enough density under gravity [27,32]. A second mechanism in the removal of COD by electrochemical method is by oxidizing organic matter, and producing oxidant agents such as hydroxyl radicals ($^\bullet OH$) or hypochlorite (HOCl) (if Cl^- is present) [33]. However, these organic pollutants are mainly degraded by the hydroxyl radicals by a reaction during the electrochemical oxidation process.

1.3. Electro-oxidation mechanism in NH_4-N removal

Electrodegradation of NH_4-N could occur due to the direct (Eq. (11)) [33] and indirect (Eqs. (12) and (13)) oxidation in the presence of Cl^-. However, byproducts of ammonia and nitrite would be oxidized and form nitrogen gas and nitrate (Eq. (14)) [34].

Direct oxidation:

$$2NH_4^+ + 6OH^- \rightarrow N_2 + 6H_2O + 2H^+ + 6e^- \tag{11}$$

Indirect oxidation:

$$2NH_4^+ + 3HClO \rightarrow N_2 + 3H_2O + 5H^+ + 3Cl^- \tag{12}$$

In the presence of Cl^-:

$$2NH_4^+ + 3ClO^- \rightarrow N_2 + 3H_2O + 2H^+ + 3Cl^- \tag{13}$$

$$NO_2^- + HClO \rightarrow NO_3^- + H_2O + Cl^- \tag{14}$$

Obviously, these three reactions are disfavored in the acidic solution. Furthermore, hypochloric acid (HClO) is formed through the oxidation of chloride in the wastewater (Eq. (15)) and disproportion chlorine (Eq. (16)) [35]. In the basic solution, hypochloric acid is separated to form hypochloric ion (ClO^-) (Eq. (17)). On the other hand, hypochloric acid has low stability in acidic conditions and forms chlorine gas [36]. So, acidic solution is not appropriate to remove NH_4-N [34].

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{15}$$

$$Cl_2 + H_2O \rightarrow HClO + H^+ + Cl^- \tag{16}$$

$$HClO \rightarrow H^+ + OCl^- \tag{17}$$

The present study was aimed to evaluate the efficiency of the electrochemical coagulation–oxidation techniques in removing COD and ammonia-nitrogen (NH_4-N) from leachate in different operating conditions.

2. Materials and methods

2.1. Experimental setup

A container of the batch electrochemical reactor with the operational capacity of 800 mL was made (Fig. 1). The aluminum and iron electrodes with the dimensions of 40 × 100 mm were used as cathode and anode, respectively. Two electrodes (cathode and anode) were installed in the parallel form and the space between them was fixed as 30 mm in all the experiments. Multi-output 5 A and 30 V digital DC power was supplied (MICRO, PW-4053S) to establish optimum conditions. The current DC was kept constant at 0.75, 1.50, and 3.0 A. The current density on the surface of the electrodes was 187.5, 375, and 562.5 A/m^2. To keep homogeneous mixing of the reactor content, magnetic stirring element at the speed of 200 rpm was used. In each run of the experiment, the electrode material, current density, and electrolysis time were changed. The EC experiments were performed for 60 min and settling time of 60 min. After 60 min of settling, the COD and NH_4-N of the effluent were measured.

2.2. Leachate sources and experimental procedure

In this study, the leachate samples were collected from Kermanshah MSW site during spring and

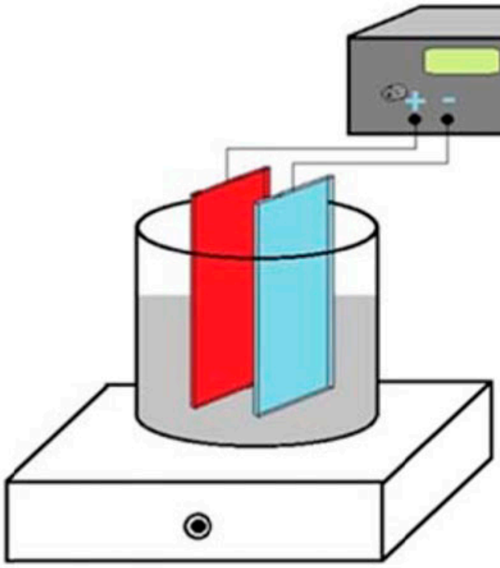

Fig. 1. Batch cell setup of EC system.

summer. The main characteristics of the leachate are represented in Table 1.

Relative removal of COD or NH_4-N was calculated using the following equation:

$$R(\%) = C_0 - C_t / C_0 \times 100 \tag{18}$$

where, C_0 is the initial COD or NH_4-N of the raw leachate and C_t is the COD or NH_4-N at the end of a predetermined settling time (*t*) after each running EC.

The amount of dissolving metals was theoretically calculated using Faraday's Law. This law could be written simply between current density and amount of dissolving substances.

Table 1
Composition and physico-chemical characteristics of landfill leachate considered

Parameter	Mean	Unit
COD	11,170 ± 266	mg/L
BOD	6,245 ± 163	mg/L
TSS	3,450 ± 412	mg/L
TDS	10,865 ± 476	mg/L
Conductivity	21.73 ± 0.12	mS/cm^2
SO_4^{2-}	1,436 ± 183	mg/L
Cl^-	1,549 ± 128	mg/L
NH_4-N	3,000 ± 261	mg/L
Turbidity	786 ± 58	NTU
Alkalinity	2,295 ± 221	mg/L $CaCO_3$
pH	6.5 ± 0.2	–

$$W = \text{item}/nF \tag{19}$$

where, W is dissolved metal (g Me/m^2), i is current density (A/m^2), t is time (s), M is molecular weight of Me ($M_{Al} = 27$, $M_{Fe} = 55$ g/mol), n is the number of electrons involved in the oxidation reaction ($n_{Al} = 3$ and $n_{Fe} = 2$), and F is Faraday's constant, 96,485 C/mol (or As/mol).

3. Result and discussion

3.1. Effect of electrode materials

Two electrodes of aluminum and iron were used in the monopolar EC process. The obtained results of both electrode materials are shown in Fig. 2. The results indicated that the relative removal of COD and NH_4-N by the Al electrode had a better outcome than the Fe electrode. After 60 min of time and current density of 562.5 A/m^2, removal performance of COD with the Fe and Al electrodes was 58.17 and 65.57%, respectively. Also, the relative removal of NH_4-N by the Fe and Al electrodes with the time of 60 min and current density of 187.5 A/m^2 was 48.63 and 62.8%, respectively. Results of Ilhan et al. [37] study showed that the COD removal efficiency with Fe electrode and Al electrode was 35 and 56%, respectively (after 30 min contact time and 631 A/m^2 current density). Also, ammonia-nitrogen removal efficiency after 30 min contact time and 631 A/m^2 current density was 11 and 14% using Fe electrode and Al electrode, respectively.

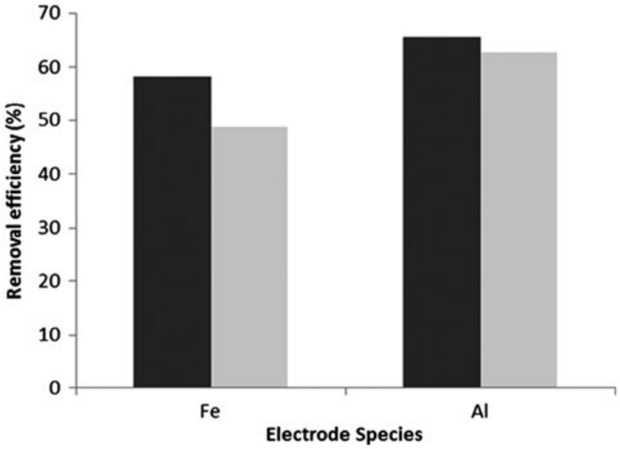

Fig. 2. Comparing two electrode materials in COD (■) and NH_4-N (▨) removal from leachate in the current density of 562.5 A/m^2 and operation time of 60 min.

3.2. Effect of current density

According to some studies, current density is a significant operating factor which measures the dosage of the coagulant in the EC process [16,24,38]. It should be noted that the variation of current density could increase the electrode dissolution and result in removing pollutants using coagulant and bubble generation [27]. Overall, by decreasing the current density EC process occurs. Although the amount of hydroxide flakes for the removal of pollutants decreases at lower current density, settling of organic and inorganic compounds on the electrodes causes efficiency reduction of the process [39]. By increasing current density, the extent of anodic dissolution increases; as a result, the highest amount of sludge is produced. Furthermore, energy consumption and operating costs increase with the excessive amounts of current density.

The examined current density which was dispersed to the surface of electrodes were 187.5, 375, and 562.5 A/m^2. The results showed that, with increasing current density, the removal efficiency of the system was slightly increased owing to the occurrence of electrochemical oxidation of pollutants at the anode electrode surface. Fig. 3 shows the efficiency of each electrode material in the removal of COD and NH_4-N. By increasing current density from 187.5 to 562.5 A/m^2, removal efficiency of COD was increased from 26.86 to 65.57% with Al electrode and 23.28 to 57.17% with Fe electrode after the time of 60 min, respectively. Moreover, the removal efficiency of NH_4-N was increased from 34.23 to 62.80% (Al electrode) and 24.56 to 48.63% (Fe electrode), with increasing current density from 187.5 to 562.5 A/m^2 and time of 60 min. According to Ilhan et al.'s work, efficient removal of COD and NH_4-N was increased from 45.5 to 59.1% and 8.8 to 14.3% after the time of 30 min due to increasing current density from 348 to 631 A/m^2 [37]; the result of removal efficiency was less than that of the present work due to higher contact time in this study.

3.3. Effect of electrolysis time

Optimum time of EC is a significant factor for saving electrical energy consumption. According to the results, the inorganic and organic forms of nitrogen were removed during the EC process. After the time of 30 min, significant changes of NH_4-N removal were observed. By increasing time to 60 min, the removal efficiency of ammonia-nitrogen increased. Accordingly, the effect of electrolysis time was explored in the range of 60 min. Removal efficiency of COD and NH_4-N was increased with increase in time (Figs. 4 and 5).

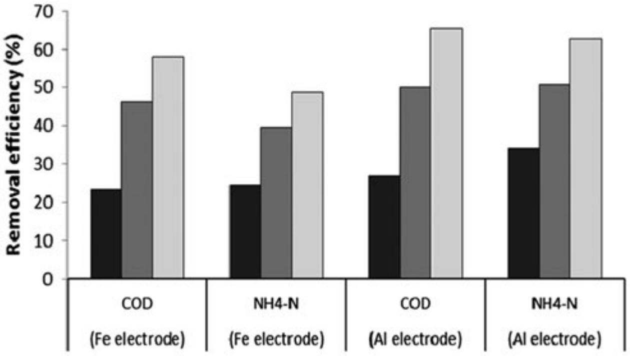

Fig. 3. Comparing efficiency of two electrode materials (Al and Fe) in COD and NH_4-N removal at different current density (187.5 ■, 375 ■, and 562.5 A/m^2 ■) and 60 min contact time.

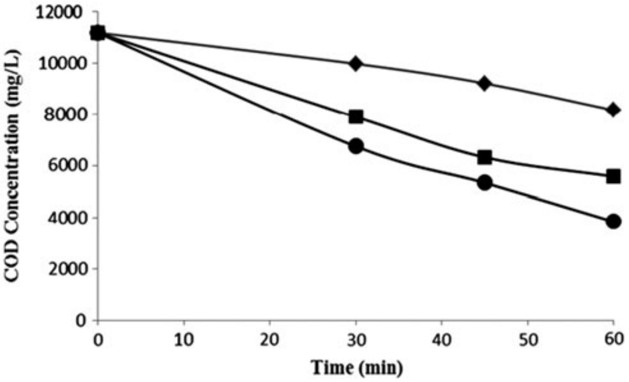

Fig. 4. Variation of COD removal within different time and current density (187.5 ♦, 375 ■, and 562.5 A/m^2 ●) using Al electrode.

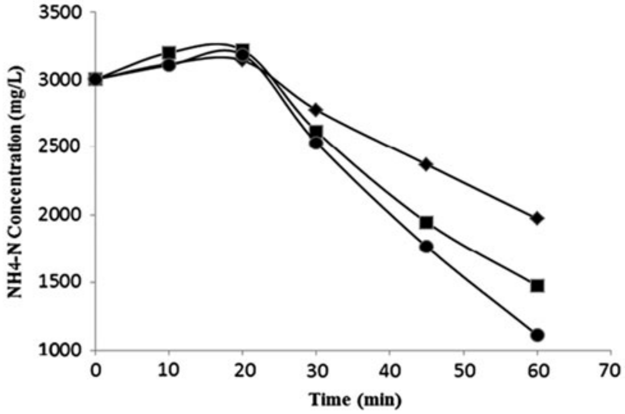

Fig. 5. Variation of NH_4-N removal within different time and current density (187.5 ♦, 375 ■, and 562.5 A/m^2 ●) using Al electrode.

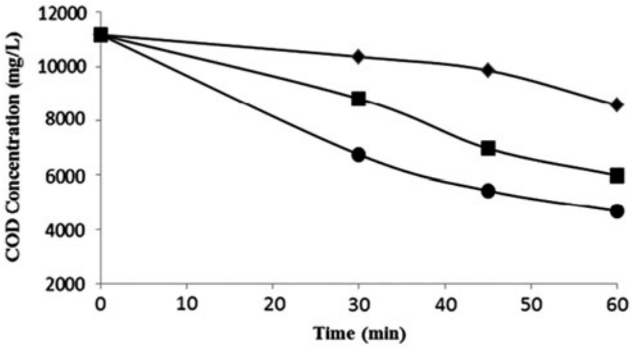

Fig. 6. Variation of COD removal within different time and current density (187.5 ♦, 375 ■, and 562.5 A/m^2 ●) using Fe electrode.

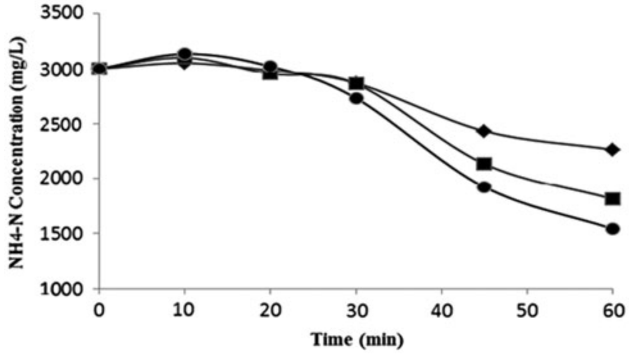

Fig. 7. Variation of NH_4-N removal within different time and current density (187.5 ♦, 375 ■, and 562.5 A/m^2 ●) using Fe electrode.

Particularly, NH_4-N removal was more dependent on electro-oxidation time. However, increasing the removal efficiency of NH_4-N after the time of 30 min might be due to the decrease in COD concentration, as well as occurrence of catalytic oxidation process at the electrode surface (Figs. 6 and 7).

As shown in Fig. 6, the COD concentration was decreased from 11,170 to 6,746 and 6,813 mg/l using Al and Fe electrodes, respectively, after the time of 30 min EC. Also, it decreased up to 3,845 and 4,672 mg/l after the time of 60 min. When the time of electrolysis raised, the concentration of metal ions and their hydroxide flakes raised; also, the amount of bubble generation was increased. Consequently, the pollutants of leachate were eliminated by the effect of coagulation and flotation.

The results showed that the C/N ratio was more at the beginning of the process and the COD removal rate was higher than that of the NH_4-N. Also, it indicated that the mineral composition was further removed than the organic compounds of NH_4-N. After nearly 30 min, the C/N ratio gradually decreased; as a result, the removal of NH_4-N occurred more quickly.

3.4. Dissolution rate of the sacrificial anode electrode

The results showed that dissolution of sacrificial anode electrode and anode electrode dimensions were reduced during the EC process. The dissolution rate of the anode electrode in the electrolyte solution was directly related to the electrolysis time, current density, and molecular weight of the electrode, and inversely related to the number of electrons in the oxidation reaction (2 electrons of Fe and 3 electrons of Al). Table 2 depicts the increasing dissolution of anode which yielded an increase in the removal efficiency of COD and NH_4-N. Also, the results indicated that, in the similar conditions of time and current density, the dissolution rate of Fe electrode was higher than Al electrode, which led to production of large quantities of sludge at iron rather than aluminum electrodes. It can be said that sludge produced in EC with Al electrode was more sedimented; as a result, a more clear supernatant than EC with Fe electrode was produced.

Table 2
Dissolution rate of anode electrode in different values of current density and removal efficiency of the system

Electrode species	Electrolysis time (min)	Current density (A/m^2)	W (g Me/m^2)	Dissolution rate per electrode surface (g Me/0.004 m^2)	COD removal (%)	NH_4-N removal (%)
Al	60	187.5	62.96	0.2518	26.86	34.23
	60	375	125.92	0.5037	50.03	50.76
	60	562.5	188.88	0.7555	65.57	62.80
Fe	60	187.5	192.38	0.7695	23.28	24.56
	60	375	384.77	1.5390	46.45	39.60
	60	562.5	577.16	2.3086	58.17	48.63

4. Conclusions

According to the experimental treatment of landfill leachate by EC, the results indicated that the EC process can be utilized to remove COD and NH_4-N from leachate under optimal conditions. The EC process was found as an appropriate and alternative method, combined with existing techniques or as a solitary unit for the effective treatment of high-toxic landfill leachate. Further, studies on the efficient electrode materials, electrolyte concentrations, and combination of oxidative agents can improve the EC performance for the removal of organic and inorganic pollutants in future.

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