Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2020.24719

173 (2020) 65–76
January

Landfill leachate treatment with electrocoagulation: optimization by using Taguchi method

Ömer Apaydin[a],*, Ebru Özkan[b]

[a]*Environmental Engineering Department, Civil Engineering Faculty, Yıldız Technical University, İstanbul, Turkey, Tel. +90 212383 5387; email: apaydin@yildiz.edu.tr*
[b]*Environmental Engineering Department, Engineering Faculty, Pamukkale University, Denizli, Turkey, Tel. +90 258296 3187; email: eozkan@pau.edu.tr*

Received 7 December 2018; Accepted 25 July 2019

ABSTRACT

In this study, the electrocoagulation (EC) method was used to treat landfill leachate (LL). pH, reaction time (electrolysis time) and current density were selected as independent parameters for an experimental design employing Taguchi orthogonal arrays. These independent parameters were varied at five different levels (pH: 3, 5, 7, 9 and 11; reaction time: 5, 10, 15, 20 and 25 min; current density: 10, 20, 30, 40 and 50 mA/cm^2) to determine their effects on the removal efficiencies of selected dependent parameters. COD, soluble COD (sCOD) and color were selected as the dependent parameters and analyzed at the beginning and end of each batch experiment. Aluminum and iron electrodes were used for separate batch studies. The maximum removal efficiencies of COD, sCOD and color with Al electrode pairs were found to be 62% (for 3-5-2 levels), 51% (for 4-4-2 levels) and 88% (for 1-5-5 levels), respectively. Moreover, the removal efficiencies of COD, sCOD and color with Fe electrodes pairs were found to be 39% (for 4-3-1 levels), 39% (for 1-1-1 levels) and 81% (for 1-5-5 levels), respectively. The operational costs of LL treatment with Al electrodes pairs were calculated to be between 3.64 and 4.53 USD$/m^3 LL. According to the Taguchi method, the contributions of pH, current density and reaction time to COD removal efficiency were found to be 79.35%, 6.45% and 4.46%, respectively.

Keywords: Aluminum electrode; COD; Electrocoagulation; Iron electrode; Landfill leachate; Taguchi method

1. Introduction

Solid waste management is an important issue in all communities worldwide. When considered within the scope of the waste hierarchy, the final solid waste disposal method is landfill. Landfill is an engineered method for solid waste disposal and important to protect the environment. Physical, chemical and biological processes occur in the landfill depending on time, and they result in the production of gases such as methane, carbon dioxide, etc., and waste water (landfill leachate). LL contains lots of organic and inorganic pollutants [1]. LL has a complex structure and a large pollutant load, and it is moderately difficult to treat it to ensure that it meets discharge standards. LL contains large numbers of organic or inorganic pollutants such as BOD, COD, ammonia and heavy metals in excessive concentrations. Therefore, many treatment methods such as biological, chemical, physical, wetland and advanced oxidation processes have been applied to treat the LL [2]. In order to treat the LL, lots of methods have been examined in the published literature, including physical [3–12], chemical [13–22] and biological [23–32] approaches. In addition to the classical treatment methods mentioned above, in recent years, electrochemical methods (electrocoagulation, electro-Fenton, electro-dialysis, etc.) have been used as a pre-treatment stage in the LL treatment process. One of the electrochemical

---

* Corresponding author.

1944-3994/1944-3986

methods is electrocoagulation (EC), and in this process, flotation and precipitation occurs simultaneously. The EC process has many benefits, such as a simple equipment requirement, ease of operation, a larger floc structure, a fast and separable sludge structure, a lower treatment volume requirement and less excessive use of chemicals. In the EC process, sacrificial electrodes such as aluminum, iron, zinc, etc. are used to generate the release of active coagulant precursors into the solution. In an EC process, an electrical current is passed through a metal electrode; the anode material undergoes oxidation, while the cathode will be subjected to reduction or reductive deposition of elemental metals. Both Al^{3+} and Fe^{3+} react with OH^- to form $Al(OH)_{3(s)}$ or $Fe(OH)_{2(s)}$ according to complex precipitation kinetics [33]. The mechanism of EC is extremely dependent on the chemistry of the aqueous medium, especially its conductivity. The mechanism of generating ions by EC can be explained with the examples of iron and aluminum, which were used as the anode and cathode in this study. In an electrolytic system, iron produces iron hydroxide. In the case of iron or steel and aluminum anodes, two mechanisms for the production of the metal hydroxide have been proposed and are presented in Eqs. (1)–(11) as follows [34–36].

1.1. Mechanism 1

Anode

$$4Fe_{(s)} \rightarrow 4Fe^{2+} + 8e^- \tag{1}$$

$$Al_{(s)} \rightarrow Al^{3+} + 3e^- \tag{2}$$

Chemical

$$4Fe^{2+} + 10H_2O_{(l)} + O_2 \rightarrow 4Fe(OH)_3 + 8H^+ \tag{3}$$

$$Al^{3+} + 3H_2O_{(l)} \rightarrow Al(OH)_3 + 3H^+ \tag{4}$$

Cathode

$$8H^+ + 8e^- \rightarrow 4H_2 \tag{5}$$

1.2. Mechanism 2

Anode

$$Fe_{(s)} \rightarrow Fe^{2+} + 2e^- \tag{6}$$

Chemical

$$Fe^{2+} + 2(OH)^- \rightarrow Fe(OH)_2 \tag{7}$$

$$Al^{3+} + 3(OH)^- \rightarrow Al(OH)_3 \tag{8}$$

Cathode

$$2H_2O_{(l)} + 2e^- \rightarrow H_2 + 2(OH)^- \tag{9}$$

Overall

$$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_2 + H_2 \tag{10}$$

$$Al^{3+} + 3H_2O_{(l)} \rightarrow Al(OH)_3 + 3H^+ \tag{11}$$

The removal efficiency of the pollutants and the energy consumption of the process are vital parameters of the treatment systems and, in turn, depend on operational parameters such as current density, pH, electrolysis (or reaction or operation) time, electrode type and conductivity. So, process parameters need to be optimized to increase the treatment efficiency and decrease the energy consumption. Moreover, in classical optimization of system-affecting factors, the level of one factor is changed while the other factor levels are kept constant, then one by one, all factors are optimized. So, the classical optimization method requires more experimental study and is time consuming [37]. There are a lot of statistical experimental design techniques, such as response surface methodology and complete or partial factorial designs that are widely used in many scientific studies [38–44]. One of these techniques is the Taguchi method and includes the design of an experimental process that uses orthogonal arrays (OA) to allow independent evaluation of factors within the least number of attempts. This technique involves data conversion to a signal-to-noise (S/N) ratio which is a measure of the presented changes [45]. In addition to other statistical experimental design methods, the Taguchi method allows the control of parameters affecting an experiment in a controlled and non-controlled manner, and the method can be applied to an experimental design with a large number of design factors [46].

In this study, raw LL from Kemerburgaz Sanitary Landfill, Istanbul, was subjected to the electrocoagulation process with aluminum (Al–Al) and iron (Fe–Fe) electrodes for batch studies in the laboratory. Aluminum and iron electrode pairs were used as a monopolar array in separate working sets.

The main objective of the study was to evaluate the treatability of LL and optimize pollutant removal efficiencies by using the Taguchi method. Hence, a batch studies matrix was planned using Minitab software for optimized EC studies using the Taguchi method. During batch studies, removal efficiencies of selected dependent variables (COD, soluble COD and color) were analyzed both at the beginning and at the end of each batch study based on independent variables (current density, reaction time and pH). This study is important because it is the first study using the Taguchi optimization method and the L_{25} orthogonal arrays in investigating LL treatment.

2. Materials and methods

2.1. Experimental setup

A 0.5 L plexiglass reactor was used for the investigation. Batch tests were conducted using 0.25 L of raw LL for each experimental set. LL was taken from the Odayeri Sanitary Landfill site located on the European side of Istanbul. The electrochemical cell consisted of a submerged anode and cathode, both of the same material (i.e., either aluminum or iron) in the form of flat sheets, in the 0.5 L plexiglass reactor. The electrode dimensions were 4.5 cm wide by 10 cm deep, resulting in a submerged surface area of 45 cm^2. In the study, electrolysis is the most important mechanism, and a digital GW Instek GPS 3030 DD power supply was used to power the EC operation. Both anode and cathode were placed parallel and vertical to each other with an inter-electrode distance of 4 cm. Electrode pairs were placed into the reactor in a monopolar configuration (Al–Al or Fe–Fe). The electrode pairs were cleaned by hand in distilled water before every run. For pH adjustment, 6 M of NaOH and/or 37% HCl of analytical grade were used. The pH and conductivity were measured by a WTW pH/Cond 340i SET 2. COD and sCOD were measured by a closed reflux method and color was measured by the platinum–cobalt (Pt–Co) scale [47]. All analyses were carried out in triplicate (deviations were lower than 5% in all cases). The experimental set-up is given Fig. 1.

LL was kept in the refrigerator at 4°C in the Environmental Engineering laboratory of Yildiz Technical University. Before each experimental study, LL was removed from the refrigerator and the test was not started until its temperature had reached the ambient temperature in the laboratory. The characteristics of the landfill leachate are given in Table 1. Moreover, no mixing was carried out in the EC reactor during experimental work processes. After each batch work, purified LL was left for 1 h and the supernatant was subjected to analysis. All wastewater analyses were performed in accordance with the Standard Methods for Examination of Water and Wastewater [47].

2.2. Experimental design based on the Taguchi method

The Taguchi method was used to create a set of experiments designed using Minitab software (Minitab 17.0 trial version). The Taguchi method includes the design of an experimental process that uses OAs to reduce the number of experiments required. OA refers to an experimental matrix designed with L_i stages, where i is the number of trials in the experimental matrix or the total degrees of freedom and includes a set of experiments where the settings of process parameters are varied. OAs allow evaluation of the effects of several process parameters to be determined efficiently. The selection of a suitable OA depends on the number of control factors and their levels [42,48]. In the study, the Taguchi method was used to decide the ideal conditions for the EC process. For this purpose, current density, pH and reaction time were selected as independent variables during this Taguchi experimental design. Each factor, which consisted of five levels and L_{25} orthogonal arrays, was taken to establish the ideal conditions for the aluminum and iron electrodes with the least number of trials. Batch studies were performed with both aluminum (Al–Al) and iron (Fe–Fe) electrodes pairs, separately. The factors and their levels in the batch studies are presented in Table 2.

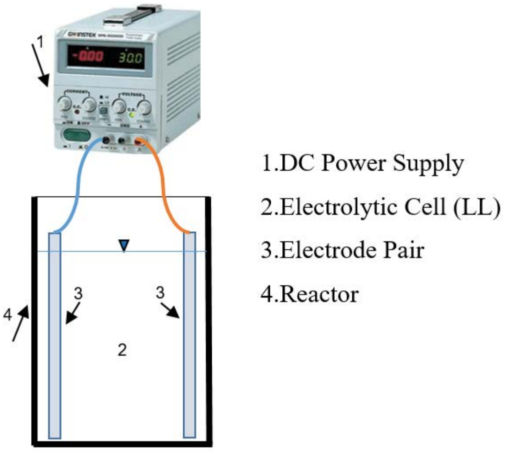

Fig. 1. Experimental set-up.

3. Results and discussion

In the study, Minitab software was used to analyze the collected data and to determine the effect of each parameter on the optimization criteria. Moreover, all levels of variables in Table 1 were used. During this work, COD, soluble COD and color results were expressed by percentage of removal through the following equation:

$$\text{Pollutant removal efficiency}(\%) = \left[\frac{C_i - C_e}{C_i}\right] \times 100 \tag{12}$$

where C_i is the initial concentration and C_e is the final concentration of the pollutant (mg/L). The numerical value of the maximum point in each graph clarifies the best value of that particular parameter shown in Table 3, and indicates the optimum conditions within the range of experimental conditions. Results related to the removal efficiency of COD, sCOD and color for 25 experimental batch studies are given in Table 3.

Table 1
Landfill leachate characteristics

Parameters	Values
pH (20 degrees centigrade)	8.1–8.8
Conductivity, mS/cm (20 degrees centigrade)	38.5–39.5
COD, mg/L	7,560–8,892
sCOD, mg/L	7,392–8,496
Color, Pt-Co	1,800–4,000

Table 2
Factors and their values corresponding to the levels to be studied in the EC experiments

Experimental factors	Experimental levels				
	1	2	3	4	5
A: Current density (mA/cm^2)	10	20	30	40	50
B: Reaction time (min)	5	10	15	20	25
C: pH	3	5	7	9	11

Table 3
Experimental variables, their levels, and results of conducted experiments corresponding to the L_{25} experimental plan

Batch experiment No	Variables and their levels			Removal of COD, %		Removal of sCOD, %		Removal of color, %	
	A	B	C	I	II	I	II	I	II
1	1	1	1	22	26	25	39	33	36
2	1	2	2	43	20	28	28	37	39
3	1	3	3	14	12	35	15	52	40
4	1	4	4	6	4	15	5	77	56
5	1	5	5	20	8	11	5	88	81
6	2	1	2	42	15	23	22	14	38
7	2	2	3	18	27	23	26	51	53
8	2	3	4	16	6	8	8	78	69
9	2	4	5	22	14	13	10	87	88
10	2	5	1	23	33	33	37	6	14
11	3	1	3	13	23	18	17	38	40
12	3	2	4	9	4	19	6	75	57
13	3	3	5	21	9	17	6	84	75
14	3	4	1	39	38	36	37	15	13
15	3	5	2	62	29	32	32	71	42
16	4	1	4	11	3	4	3	57	27
17	4	2	5	24	7	19	6	85	39
18	4	3	1	25	39	31	37	22	25
19	4	4	2	58	30	51	35	71	32
20	4	5	3	21	27	33	24	73	38
21	5	1	5	12	6	8	7	82	42
22	5	2	1	41	30	22	30	18	13
23	5	3	2	53	31	50	37	64	41
24	5	4	3	37	28	29	17	78	33
25	5	5	4	12	15	9	5	83	33

The results of the experimental studies obtained by using the most significant independent variables performed in the study are summarized in the following sub sections for both aluminum and iron electrode pairs based on Figs. 2–7 and Table 3.

3.1. Effect of current density on the process

One important parameter for efficient removal of pollutants in the EC process is current density. Current density is responsible for the metal hydroxide concentrations, the reaction rate of the process, the coagulant dosage, the bubble production and the effects on growth of flocs. In this study, current density was used in a range of 10–50 mA/cm^2 to assess the effect on LL treatment efficiencies of the electrochemical process. In the case of aluminum electrodes (Figs. 2–4), the highest removal efficiency for COD, sCOD and color was at level five (50 mA/cm^2), at level three (30 mA/cm^2) and at level five (50 mA/cm^2), respectively. On the other hand, in the case of the iron electrodes (Figs. 5–7), the highest removal efficiency for COD, sCOD and color was at level two (20 mA/cm^2), at level 2 (20 mA/cm^2) and at the first level (10 mA/cm^2), respectively.

The removal efficiencies for COD, sCOD and color based on changes in current density are summarized in Table 3:

For aluminum electrodes, the highest COD removal efficiency was obtained at 30 mA/cm^2. Compared with the value at 30 mA/cm^2, the COD removal efficiency at 20 mA/cm^2 decreased by at least 32%. Compared with the value at 30 mA/cm^2, the COD removal efficiency at 40 mA/cm^2 decreased by at least 6%. The highest efficiency of sCOD removal was at 40 mA/cm^2. Compared with the value at 40 mA/cm^2, the sCOD removal efficiency at 30 mA/cm^2 decreased by at least 30%. Compared with the value at 40 mA/cm^2, the removal efficiency at 50 mA/cm^2 decreased by at least 1%. The highest color removal efficiency was obtained at a current density of 10 mA/cm^2. Compared with the value at 10 mA/cm^2, the color removal efficiency at 20 mA/cm^2 decreased by at least 1%. Compared with the value at 10 mA/cm^2, the color removal efficiency at 30 mA/cm^2 decreased by at least 4%.

For iron electrodes, the highest COD removal efficiency was obtained at 40 mA/cm^2. Compared with the value at 40 mA/cm^2, the COD removal efficiency at 30 mA/cm^2 decreased by at least 2%. Compared with the value at 40 mA/cm^2, the COD removal efficiency at 50 mA/cm^2 decreased by at least 21%. The highest sCOD removal efficiency was obtained at 10 mA/cm^2. Compared with its value at 10 mA/cm^2, sCOD removal efficiency decreased by at least 2% at 20 mA/cm^2. The highest color removal efficiency

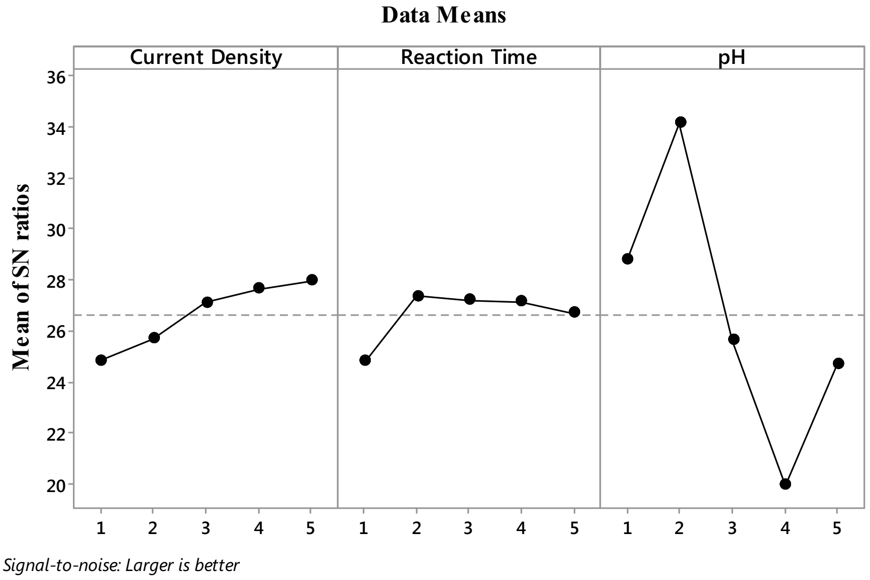

Fig. 2. Effect of the parameters on COD removal efficiency for Al electrodes. On the left-hand side, it shows the current density (10 to 50 mA/cm^2) based on levels (1 to 5). In the middle, it shows hydrolysis time (5 to 25 min) based on levels (1 to 5). On the right-hand side, it shows different initial pH (3 to 11) based on levels (1 to 5).

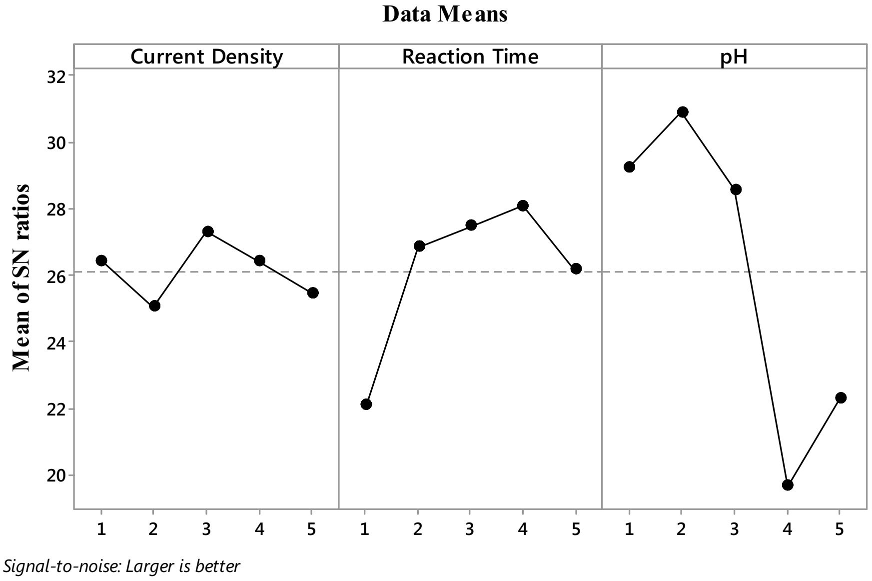

Fig. 3. Effect of each parameter on sCOD removal efficiency for Al electrodes. On the left-hand side, it shows the current density (10 to 50 mA/cm^2) based on levels (1 to 5). In the middle, it shows hydrolysis time (5 to 25 min) based on levels (1 to 5). On the right-hand side, it shows different initial pH (3 to 11) based on levels (1 to 5).

was obtained at a current density of 20 mA/cm^2. Compared with the value at 20 mA/cm^2, the color removal efficiency at 10 mA/cm^2 decreased by at least 7%. Compared with the value at 20 mA/cm^2, the color removal efficiency at 30 mA/cm^2 decreased by at least 14%.

A study performed by Kalyani et al. [49] observed that the color removal and total organic carbon (TOC) removal were increased as the current density increased up to the optimum value. The increase in current density resulted in the production of a large amount of Al^{3+} ions via anodic metal dissolution and more H_2 bubbles forming at the cathode, which are profitable for the separation or flotation process [50]. According to Heidmann and Calmano [51], a proportional relationship was established between the current density and power consumption, and it is necessary to identify the optimum value of current density to reduce the power consumption. In addition, Faraday's law also states that the amount of oxidized metal decreased as the gap between the electrodes is increased. The power consumption increased as the inter-electrode distance increased. This was due to the fact that there is more resistance offered when the electrodes gap increases and power consumption is directly proportional to the cell voltage [52].

3.2. Effect of reaction time on the process

It is known that there is a strong relationship between reaction time and the formation of the metal hydroxide necessary to remove the pollutant. In this study, the reaction time used was in the range of 5–25 min. The two examples with aluminum and iron electrodes showed the effect of reaction time on the efficiency of pollutant removal. In the case of the aluminum electrode (Figs. 2–4), the highest removal efficiency for COD, sCOD and color were at the second level (10 min), at the fourth level (20 min) and at the fourth level (20 min), respectively. On the other hand, for the iron electrode (Figs. 5–7), the highest removal efficiency for COD, sCOD and color were at the fourth level (20 min), at the fourth level (20 min) and at the third level (15 min), respectively.

The removal efficiencies of COD, sCOD and color, based on the changes in reaction time, are summarized in Table 3:

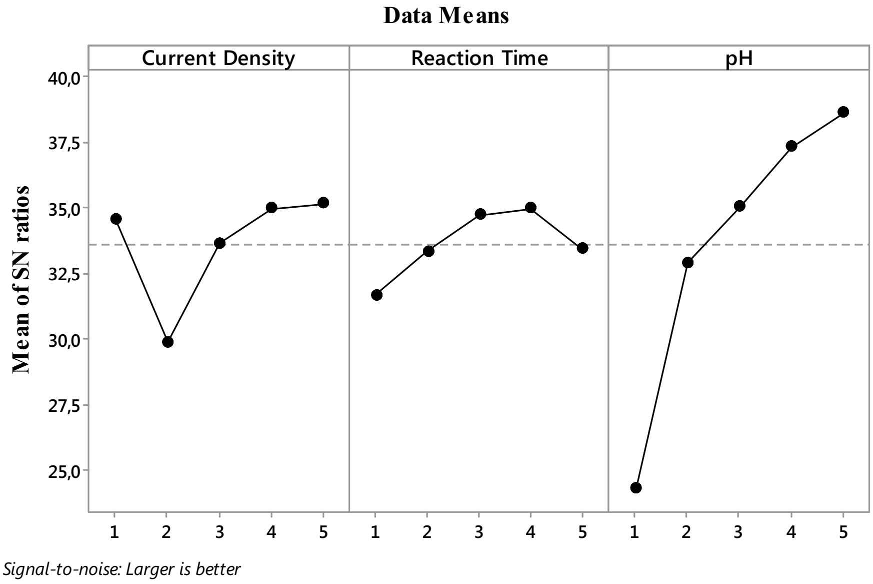

Fig. 4. Effect of each parameter on color removal efficiency for Al electrodes. On the left-hand side, it shows the current density (10 to 50 mA/cm^2) based on levels (1 to 5). In the middle, it shows hydrolysis time (5 to 25 min) based on levels (1 to 5). On the right-hand side, it shows different initial pH (3 to 11) based on levels (1 to 5).

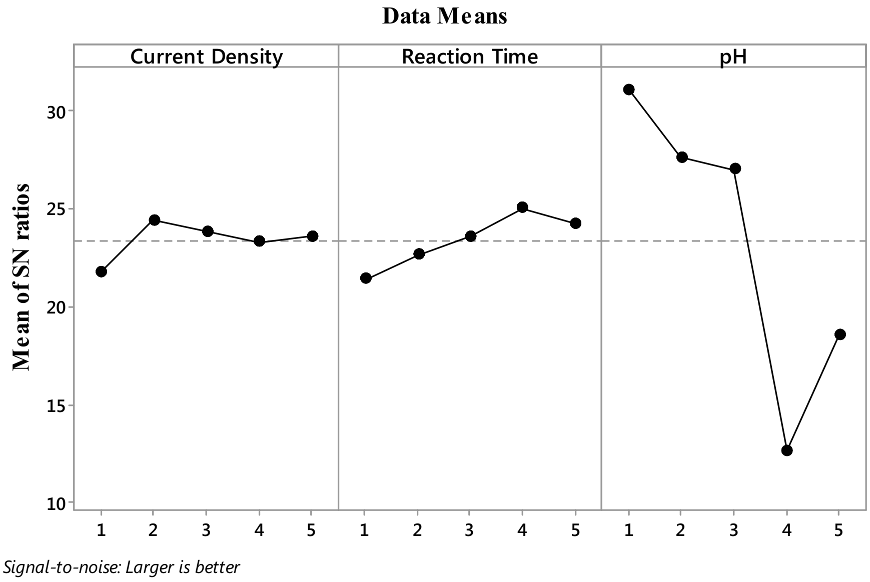

Fig. 5. Effect of each parameter on COD removal efficiency for Fe electrodes. On the left-hand side, it shows the current density (10 to 50 mA/cm^2) based on levels (1 to 5). In the middle, it shows hydrolysis time (5 to 25 min) based on levels (1 to 5). On the right-hand side, it shows different initial pH (3 to 11) based on levels (1 to 5).

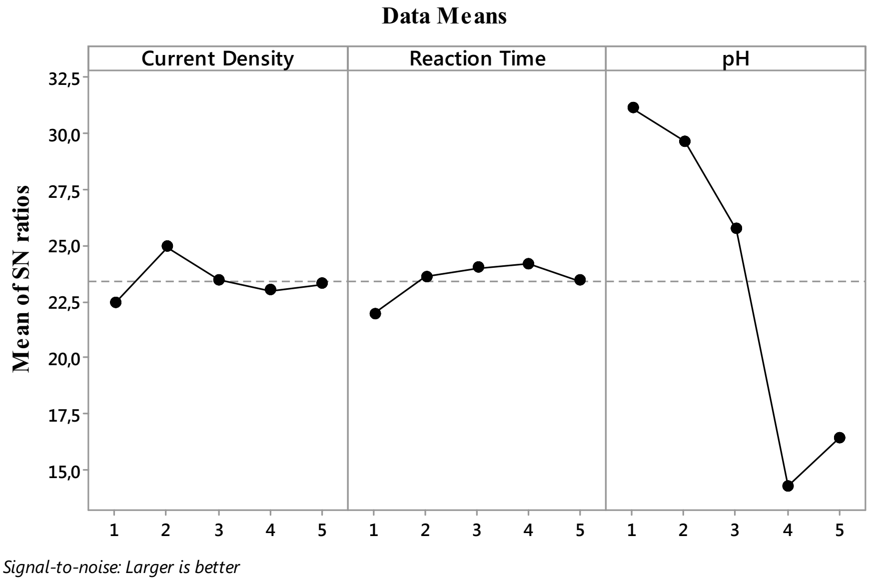

Fig. 6. Effect of each parameter on sCOD removal efficiency for Fe electrodes. On the left-hand side, it shows the current density (10 to 50 mA/cm^2) based on levels (1 to 5). In the middle, it shows hydrolysis time (5 to 25 min) based on levels (1 to 5). On the right-hand side, it shows different initial pH (3 to 11) based on levels (1 to 5).

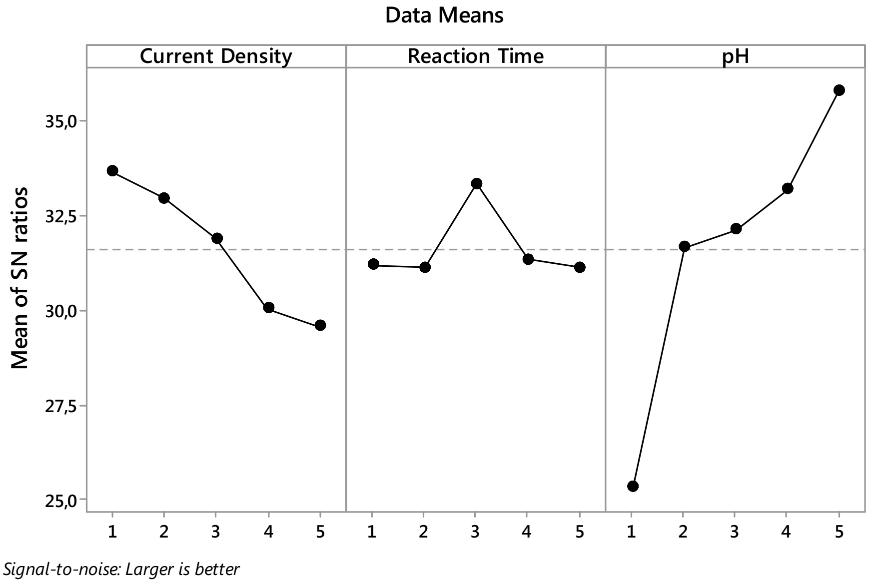

Fig. 7. Effect of each parameter on color removal efficiency for Fe electrodes. On the left-hand side, it shows the current density (10 to 50 mA/cm^2) based on levels (1 to 5). In the middle, it shows hydrolysis time (5 to 25 min) based on levels (1 to 5). On the right-hand side, it shows different initial pH (3 to 11) based on levels (1 to 5).

For aluminum electrodes, the highest COD removal efficiency was obtained at level 5 (25 min). Compared with the value at level 5 (25 min), the COD removal efficiency at level 4 (20 min) decreased by at least 6%. The highest efficiency of sCOD removal was obtained at level 4 (20 min). Compared with the value at level 4 (20 min), the sCOD removal efficiency at level 3 (15 min) decreased by at least 2%. Compared with the value at level 4 (20 min), the removal efficiency at level 5 (25 min) decreased by at least 30%. The highest color removal efficiency was obtained at level 5 (25 min). Compared with the value at level 5 (25 min), the color removal efficiency at level 4 (20 min) decreased by at least 1%.

For iron electrodes, the highest COD removal was obtained at level 3 (15 min). Compared with the value at level 3 (15 min), the COD removal efficiency at level 4 (20 min) decreased by at least 1%. Compared with the value at level 3 (15 min), the COD removal efficiency at level 2 (10 min) decreased by at least 23%. The highest sCOD removal efficiency was obtained at level 1 (5 min). Compared with its value at level 1 (5 min), sCOD removal efficiency decreased by at least 23% at level 2 (10 min). The highest color removal efficiency was obtained at level 4 (20 min). Compared with the value at level 4 (20 min), the color removal efficiency at level 5 (25 min) decreased by at least 7%. Compared with the value at level 4 (20 min), the color removal efficiency at level 3 (15 min) decreased by at least 14%.

3.3. Effect of pH on the process

In this study, the most important parameter for removal efficiencies of pollutants in the electrochemical processes was found to be pH. It was also stated in previous studies that pH was the most important factor for pollutant removal via electrochemical processes [41,53].

In the case of the aluminum electrode (Figs. 2–4), the highest removal efficiency for COD, sCOD and color were at the second level (pH = 5), at the second level (pH = 5) and at the fifth level (pH = 11), respectively. On the other hand, a study conducted by Asaithambi et al. [54] using aluminum electrodes, color and TOC removal efficiency increased at an effluent pH value from 5 to 7.5, but decreased when the pH was from 7.5 to 11. When this current study is compared with the previous study [54] to determine the pH value for optimum color removal, it is understood that optimum removal efficiencies are obtained at different pH values. This can be explained theoretically in the absence of any pollutants for pure water under standard conditions: with Al electrodes in acidic medium, monomeric hydroxometallic cations $Al(OH)_3$ are formed (Eqs. (4) and (11)). For neutral media, both polymeric hydroxometallic cations and metal hydroxide precipitates coexist at higher pH. The net charge on the surface of the amorphous metal hydroxide precipitate changes from positive to negative and the polymeric cations remain in the solution. More OH^- can be formed in neutral conditions compared with acidic and alkaline media in the electrocoagulation process [55,56]. Nevertheless, LL has a lot of pollutants and it is difficult to develop these theoretically known electrochemical conditions in it.

In contrast, for iron electrodes (Figs. 5–7), the highest removal efficiency for COD, sCOD and color was at the first level (pH = 3), at the first level (pH = 3) and at the fifth level (pH = 11), respectively.

For aluminum electrodes, the removal efficiencies of COD, sCOD and color based on the changes of pH are summarized below using Table 3:

The highest COD removal efficiency was obtained for a pH = 5. The COD removal efficiency at pH = 7 was reduced by 65% compared with the value at pH = 5. The highest removal efficiency for sCOD was obtained when pH = 5. When compared with pH = 5, the removal efficiency at pH = 7 decreased by 31%. The highest color removal efficiency was realized at pH = 11. The color removal at pH = 9 was reduced by 12% compared with the value at pH = 11.

For iron electrodes, the highest COD removal efficiency was obtained for pH = 3. Compared with pH = 3, the COD removal efficiency at pH = 5 decreased by at least 21%. The highest removal efficiency for sCOD was obtained when pH = 3. Compared with the value at pH = 3, sCOD removal efficiency at pH = 5 decreased by at least 3%. The highest color removal efficiency was realized at pH = 11. The color removal efficiency at pH = 9 was reduced by at least 21% compared with the value at pH = 11.

The effects of performance criteria on COD, sCOD and color removal efficiency for aluminum electrodes are illustrated in Figs. 2–4, respectively. The experiments performed within the investigated range showed that optimal current density, reaction time and pH value were 50 mA/cm^2, 20 min and 5, respectively.

The effects of the performance criteria on COD, sCOD and color removal efficiency for iron electrodes are illustrated in Figs. 5–7, respectively. The experiments performed within the investigated range showed that optimal current density, reaction time and pH value were 20 mA/cm^2, 20 min and 3, respectively.

The Taguchi design experiment (L_{25}) was carried out to optimize the effective parameters to efficiently remove LL pollutants using an electrocoagulation process. The larger-the-better *S/N* ratio was used to analyze the results of the batch experimental studies. Analysis of variance (ANOVA) was performed to examine the effective parameters and their confidence levels on the COD, sCOD and color removal efficiencies. ANOVA is used to explore the process parameters that significantly affect the process responses. The resulting ANOVA values for COD removal, sCOD removal and color removal performance for aluminum and iron electrodes are given in Tables 4 and 5, respectively.

Table 4 shows the result of the ANOVA test for COD, sCOD and color removal performance with aluminum electrodes. According to the ANOVA analysis, the factors in the tables are in accordance with their significance. According to Table 4 (Al–Al electrodes pairs), the first important independent parameter for efficient removal of COD was the initial pH with a contribution value of 79.35%. The second important independent parameter for COD was the current density with a contribution value of 6.45%. The third important independent parameter for COD was the reaction time with a contribution value of 4.46%. By contrast, the first important independent parameter for efficient removal of sCOD was initial pH with a contribution value of 66.71%. The second important independent parameter for sCOD was the reaction time with a contribution value of 15.51%. The third important independent parameter for sCOD was the current density with a contribution value of 4.35%. In addition, the first important independent parameter for efficient removal of color was pH with a contribution value of 74.31%. The second important independent parameter on color was the reaction time with a contribution value of 8.56%. The third important independent parameter for color was the current density with a contribution value of 5.19%. Hence, in the case with aluminum electrodes, the sequence related to the importance of COD removal efficiency was found to be pH > current density > reaction time. The sequence related to the importance of sCOD and color removal efficiency was found to be pH > reaction time > current density.

Table 5 shows the result of the ANOVA test for COD, sCOD and color removal performance with iron electrodes. According to ANOVA analysis, the factors in the tables are in accordance with their significance. According to Table 5 (Fe–Fe electrodes pairs), the first important independent parameter for efficient removal of COD was the initial pH

Table 4
ANOVA values for COD, sCOD and color removal efficiency using aluminum electrodes

Source	DF	Seq SS	*C*, %	Adj SS	Adj MS	*F*-value	*p*-value
a. COD removal performance							
Current density	4	404.2	6.45	404.2	101.05	1.99	0.161
Reaction time	4	279.3	4.46	279. 3	69.83	1.37	0.301
pH	4	4,973.3	79.35	4,973.3	1,243.33	24.45	0.000
Error	12	610.3	9.74	610.3	50.86		
Total	24	3,590.4	100				
b. Soluble COD removal performance							
Current density	4	156.4	4.35	156.4	39.09	0.97	0.458
Reaction time	4	556.9	15.51	556.9	139.24	3.47	0.042
pH	4	2,395.2	66.71	2,395.2	598.81	14.91	0.000
Error	12	481.9	13.42	481.9	40.16		
Total	24	3,590.4	100.00				
c. Color removal performance							
Current density	4	899.8	5.19	899.8	224.90	1,30	0.323
Reaction time	4	1,482.6	8.56	1,482.6	370.60	2,15	0.13
pH	4	12,876.2	74.31	12,876.2	18.66	3,219	0.000
Error	12	2,069.7	11.94	2,069.7	172.50		
Total	24	17,328.3	100				

Notes: DF: degree of freedom, SS: sum of squares, *C*: contribution, MS: mean of square. %*C* = 100 (sum of squares/total sum of squares).

with a contribution value of 88.10%. The second important independent parameter for COD was the reaction time with a contribution value of 3.35%. The third important independent parameter for COD was the current density with a contribution value of 1.96%. By contrast, the first important independent parameter for efficient removal of sCOD was the initial pH with a contribution value of 92.14%. The second important independent parameter for sCOD was the reaction time with a contribution value of 0.98%. The third important independent parameter for sCOD was the current density with a contribution value of 0.42%. In addition, the first important independent parameter for efficient removal of color was pH with a contribution value of 57.27%. The second important independent parameter for color was the current density with a contribution value of 20.40%. The third important independent parameter for color was the reaction time with a contribution of 5.45%.

Hence, in the case with iron electrodes, the sequence related to the importance of the COD and sCOD removal efficiency was found to be pH > reaction time > current density. On the other hand, the sequence related to the importance of color removal efficiency was found to be pH > current density > reaction time.

Predicted removal efficiency values for optimum experimental conditions obtained using Minitab software for aluminum and iron electrodes are given in Table 6.

Table 6 shows the optimum working conditions and predicts the response under these conditions. For aluminum electrodes, the best COD removal conditions were found to be 50 mA/cm^2, 20 min and pH = 5; under these conditions the predicted COD removal efficiency was 60.65%. The predicted sCOD and color removal efficiency for aluminum electrodes were found to be 45.82% and 100%, respectively. For iron electrodes under the best removal conditions, predicted COD and dissolved COD removal efficiency were 41.04% and 38.10%, respectively.

3.4. Costs analysis

In the EC process, cost is considered a vital parameter that affects the application of any method of wastewater treatment. This study looked at the ideal conditions for the EC process. The operating cost includes material cost (electrode cost) and electrical energy cost (energy cost). The operating cost was calculated per m^3 of treated wastewater for both Al and Fe electrodes. The energy cost and electrode cost in the EC process can be calculated using Eqs. (13) and (14), respectively. The sum of these costs was calculated as the operational cost in the study.

$$\text{Energy cost} = \frac{E\,T\,If_e}{1{,}000\text{V}} \tag{13}$$

where the energy cost is in kWh/m^3, E is the voltage (V), I is the electrical current (A), T is the reaction time (h), f_e is a unit of electrical cost (0.1 USD$/kWh), 1,000 is a multiplier for kilowatt conversion, and V is the cubic volume of the treated LL (m^3).

$$\text{Electrode cost} = \frac{ITf_m M_W}{zFV} \tag{14}$$

where the electrode consumption is in kg/m^3, f_m is unit electrode cost (1.82 USD$/kg Al and 0.36 USD$/kg Fe), M_W is the molecular weight of the metal electrode (Al: 0.027 kg/mole;

Table 5
ANOVA values for COD, sCOD and color removal efficiency using iron electrodes

Source	DF	Seq SS	*C*, %	Adj SS	Adj MS	*F*-value	*p*-value
			a. COD removal performance				
Current density	4	74.31	1.96	74.31	18.58	0.89	0.497
Reaction time	4	126.89	3.35	126.89	31.72	1.53	0.256
pH	4	3,333.58	88.10	3,333.58	833.40	40.14	0.000
Error	12	249.13	6.58	249.13	20.76		
Total	24	3,783.92	100				
			b. Soluble COD removal performance				
Current density	4	17.39	0.42	17.39	4.349	0.20	0.935
Reaction time	4	40.06	0.98	40.06	10.016	0.45	0.768
pH	4	3,784.07	92.14	3,784.07	946.016	42.82	0.000
Error	12	265.12	6.46	265.12	22.094		
Total	24	4,106.65	100.00				
			c. Color removal performance				
Current density	4	1,884.6	20.4	1,884.6	471.1	3.43	0.043
Reaction time	4	503.8	5.45	503.8	125.9	0.92	0.485
pH	4	5,290.2	57.27	5,290.2	1,322.5	9.63	0.001
Error	12	1,647.7	17.84	1,647.7	137.3		
Total	24	9,236.2	100				

Notes: DF: degree of freedom, SS: sum of squares, *C*: contribution, MS: mean of square.

Table 6
Predicted pollutant removal efficiency values for optimum experimental conditions (aluminum and iron electrodes)

Source	Current density	Reaction time	pH	Predicted, %	C.L.,%
		For aluminum electrodes			
COD	5	4	2	60.65	49.44–71.85
Soluble COD	4	4	2	45.82	35.86–55.78
Color	5	4	5	100	80.04–121.31
		For iron electrodes			
COD	4	4	1	41.04	33.87–48.19
Soluble COD	4	4	1	38.10	30.71–45.48
Color	1	3	5	80.28	48.80–111.75

Notes: C.L: confidence limit.

Fe: 0.056 kg/mole), F is Faraday constant (96,485 coulomb/mole or ampere.second/mole), T is the reaction time (s) and z is the electron transfer number (Al: 3 and Fe: 2).

It is known that hydrogen is formed at the cathode by electrolysis during the process. The amount of hydrogen formed in the process can be theoretically calculated with Eq. (15).

$$n_{H_2} = \frac{IT}{F} H \tag{15}$$

where n_{H_2} is the amount of hydrogen obtained at cathode (mole), T is the reaction time (second) and H is an independent variable with a value of 0.5.

The operating costs and total costs for the optimum removal efficiencies of COD, sCOD and color using Al and Fe electrodes were calculated by Eqs. (2) and (3). The life stage of LL used in the current study can be characterized as being mid-life. The energy costs for COD, sCOD and color for aluminum electrodes were found to be 3.64, 3.64 and 4.53 USD\$/m^{3}LL, respectively. In a preliminary study [57], the COD removal cost for young leachate was found to be 6.91 USD\$/m^{3}LL. In another study using mid-life LL from the same landfill [22], the COD removal cost was found to be 4.37 USD\$/m^{3}LL (with aluminum electrodes). In a study conducted by Asaithambi et al. [54] using aluminum electrodes, the optimum energy cost of removal of color and TOC for LL was found to be 0.39 USD\$/m^{3}LL for old LL. The cost of pollutant removal can be explained in relation to the age of the LL. It was said that treatment of mid-life or older LL with EC is more economical than for young leachate. The energy costs for COD, sCOD and color for Fe electrodes were found to be

4.53, 0.59 and 0.1 USD\$/m^{3}LL, respectively. Operational costs for COD, sCOD and color using Al electrodes were found to be 5.01 , 5.01 and 6.24 USD\$/m^{3}LL, respectively. The operational costs for COD, sCOD and color for Fe electrodes were found to be 5.58, 0.93 and 0.28 USD\$/m^{3}LL, respectively. Moreover, the theoretical hydrogen reuse at the cathode was calculated based on Eq. (4) and found to be between 0.003 mole and 0.01 moles for both Al electrodes and Fe electrodes.

4. Conclusions

According to the results obtained in the studies with Al electrodes, the highest COD and sCOD removal efficiencies were obtained at pH values of 5, 62% and 51%, respectively. The highest color removal efficiency was found to be 88% for both types of electrodes at a pH = 11. Due to the high cost of electricity, the reaction time for the EC process was planned to be no more than 25 min and the pollutant removal efficiencies achieved were not that high. EC treatment costs have been investigated in this study, and the results showed that operating costs were different for each electrode type. Consequently, for the study conducted with aluminum electrodes, the operational costs of the COD, sCOD and color removal were calculated to be between 5.01 and 6.24 USD\$/m^{3}LL. Moreover, in the study conducted with iron electrodes, the operational costs of the COD, sCOD and color removal were calculated to be between 0.28 and 5.58 USD\$/m^{3}LL. ANOVA results revealed that the most effective parameter was pH because it produced the highest contribution percentages for all three pollutants. Considering COD as the most important pollutant in the leachate, the contribution of independent variables to the process for Al and Fe electrodes can be shown to be pH > current density > reaction time and pH > reaction time > current density, respectively.

Acknowledgment

This study was carried out in the laboratories of the Department of Environmental Engineering at Yildiz Technical University.

References

[1] W.A. Worrell, P.A. Vesilind, Solid Waste Engineering, 2nd ed., SI, 2012.

[2] B. İnanç, B. Çalli, A.M. Saatçi, Characterization and anaerobic treatment of the sanitary landfill leachate in Istanbul, Water Sci. Technol., 41 (2000) 223–230.

[3] Y. Zhou, M. Huang, Q. Deng, T. Cai, Combination and performance of forward osmosis and membrane distillation (FO-MD) for treatment of high salinity landfill leachate, Desalination, 420 (2017) 99–105.

[4] S.M. Iskander, S. Zou, B. Brazil, J.T. Novak, Z. He, Energy consumption by forward osmosis treatment of landfill leachate for water recovery, Waste Manage., 63 (2017) 284–291.

[5] J. Li, A. Niu, C.J. Lu, C.J. Lu, J.H. Zhang, M. Junaid, P.R. Strauss, P. Xiao, X. Wang, Y.W. Ren, D.S. Pei, A novel forward osmosis system in landfill leachate treatment for removing polycyclic aromatic hydrocarbons and for direct fertigation, Chemosphere, 168 (2017) 112–121.

[6] K. Kosutic, D. Dolar, T. Strmecky, Treatment of landfill leachate by membrane processes of nanofiltration and reverse osmosis, Desal. Wat. Treat., 55 (2015) 2680–2689.

[7] M. Sir, Z. Honzajkova, Treatment of municipal landfill leachate by the process of reverse osmosis and evaporation, Fresenius Environ. Bull., 24 (2015) 2245–2250.

[8] Y. Dong, Z. Wang, C. Zhu, Q. Wang, J. Tang, Z. Wu, A forward osmosis membrane system for the post-treatment of MBR-treated landfill leachate, J. Membr. Sci., 471 (2014) 192–200.

[9] G. Wang, Z. Fan, D. Wu, L. Qin, G. Zhang, C. Gao, Q. Meng, Anoxic/aerobic granular active carbon assisted MBR integrated with nanofiltration and reverse osmosis for advanced treatment of municipal landfill leachate, Desalination, 349 (2014) 136–144.

[10] A.M. Schiopu, G.C. Piuleac, C. Cojocaru, I. Apostol, I. Mamaliga, M. Gavrilescu, Reducing environmental risk of landfills: leachate treatment by reverse osmosis, Environ. Eng. Manage. J., 11 (2012) 2319–2331.

[11] M. Sir, M. Podhola, T. Patocka, Z. Honzajkova, P. Kocurek, M. Kubal, M. Kuras, The effect of humic acids on the reverse osmosis treatment of hazardous landfill leachate, J. Hazard. Mater., 207 (2012) 86–90.

[12] W. Rukapan, B. Khananthai, C. Chiemchaisri, W. Chiemchaisri, T. Srisukphun, Short- and long-term fouling characteristics of reverse osmosis membrane at full scale leachate treatment plant, Water Sci. Technol., 65 (2012) 127–134.

[13] V. Oloibiri, S. De Coninck, M. Chys, K. Demeestere, S.W.H. Van Hulle, Characterisation of landfill leachate by EEM-PARAFAC-SOM during physical-chemical treatment by coagulation-flocculation, activated carbon adsorption and ion exchange, Chemosphere, 186 (2017) 873–883.

[14] A. Alighardashi, Z.K. Esfahani, S.A. Anbardan, Landfill leachate treatment in an integrated adsorption-chemical oxidation process including CNT and nZVI-H_2O_2, Desal. Wat. Treat., 82 (2017) 338–345.

[15] M. Orkun, A. Kuleyin, Treatment performance evaluation of chemical oxygen demand from landfill leachate by electro-coagulation and electro-Fenton technique, Environ. Prog. Sust. Energy, 31 (2012) 59–67.

[16] J. Ding, L. Wei, H. Huang, Q. Zhao, W. Hou, F.T. Kabutey, Y. Yuan, D. Dionysiou, Tertiary treatment of landfill leachate by an integrated electro-oxidation/electro-coagulation/electro-reduction process: performance and mechanism, J. Hazard. Mater., 351 (2018) 90–97.

[17] E.C. Rada, I.A. Istrate, M. Ragazzi, G. Andreottola, V. Torretta, Analysis of electro-oxidation suitability for landfill leachate treatment through an experimental study, Sustainability, 5 (2013) 3960–3975.

[18] Z. Li, S. Yuan, C. Qiu, Y. Wang, X. Pan, J. Wang, C. Wang, J. Zuo, Effective degradation of refractory organic pollutants in landfill leachate by electro-peroxone treatment, Electrochim. Acta, 102 (2013) 174–182.

[19] M.J.K. Bashir, H.A. Aziz, S.Q. Aziz, S.S. Abu Amr, An overview of electro-oxidation processes performance in stabilized landfill leachate treatment, Desal. Wat. Treat., 51 (2013) 2170–2184.

[20] H. Zhang, X. Ran, X. Wu, Electro-Fenton treatment of mature landfill leachate in a continuous flow reactor, J. Hazard. Mater., 241 (2012) 259–266.

[21] M. Umar, H.A. Aziz, M.S. Yusoff, Trends in the use of Fenton, electro-Fenton and photo-Fenton for the treatment of landfill leachate, Waste Manage., 30 (2010) 2113–2121.

[22] F. İlhan, U. Kurt, Ö. Apaydin, M.T. Gönüllü, Treatment of leachate by electrocoagulation using aluminum and iron electrodes, J. Hazard. Mater., 154 (2008) 381–389.

[23] M. Saleem, A. Spagni, L. Alibardi, A. Bertucco, M.C. Lavagnolo, Assessment of dynamic membrane filtration for biological treatment of old landfill leachate, J. Environ. Manage., 213 (2018) 27–35.

[24] K. Mariam, E. Lobna, A. Haifa, C. Yassmine, H. Cyrine, N. Salma, F. Sami, T. Ismail, Use of dairy reject and fermented *Aleo* sp. leaf gel mixture in the biological pre-treatment of landfill leachate, Water Practice Technol., 13 (2018) 219–228.

[25] C. Yuan, Y. Wang, T. Zhu, X. Feng, G. Ma, F. Yu, Y. Xie, Multistage biological contact oxidation for landfill leachate treatment: optimization and bacteria community analysis, Int. Biodeterior. Biodegrad., 125 (2017) 200–207.

[26] L. Jurczykand, J. Koc-Jurczyk, Biological removal of ammonia from municipal landfill leachate (Kozodrza, Poland) at limited oxygen access and presence of kaldnes media, J. Ecol. Eng., 19 (2018) 131–139.

[27] L. Wu, L. Zhang, Y. Xu, C. Liang, H. Kong, X. Shi, Y. Peng, Advanced nitrogen removal using bio-refractory organics as carbon source for biological treatment of landfill leachate, Sep. Purif. Technol., 170 (2016) 306–313.

[28] F.A. El-Gohary, G. Kamel, Characterization and biological treatment of pre-treated landfill leachate, Ecol. Eng., 94 (2016) 268–274.

[29] Y. Xia, P.J. He, H.X. Pu, F. Lu, L.M. Shao, H. Zhang, Inhibition effects of high calcium concentration on anaerobic biological treatment of MSW leachate, Environ. Sci. Pollut. Res., 23 (2016) 7942–7948.

[30] B. Pieczykolan, I. Plonka, M. Kosel, Changes of the quality of activated sludge during the biological treatment of landfill leachate, Environ. Protect. Eng., 42 (2016) 33–42.

[31] Y. Cui, Q, Wu, M, Yang, F. Cui, Three-dimensional excitation-emission matrix fluorescence spectroscopy and fractions of dissolved organic matter change in landfill leachate by biological treatment, Environ. Sci. Pollut. Res., 23 (2016) 793–799.

[32] G.T. Muller, A. Giacobbo, E.A. dos Santos Chiaramonte, M.A.S. Rodrigues, A.M. Bernardes, The effect of sanitary landfill leachate aging on the biological treatment and assessment of photoelectrooxidation as a pre-treatment process, Waste Manage., 36 (2015) 177–183.

[33] N. Daneshvar, A. Oladegaragoze, N. Djafarzadeh, Decolorization of basic dye solutions by electrocoagulation: an investigation of the effect of operational parameters, J. Hazard. Mater., 129 (2006) 116–122.

[34] M.Y.A. Mollah, R. Schennach, J.R. Parga, D.L. Cocke, Electrocoagulation (EC)—science and application, J. Hazard. Mater., B84 (2001) 29–41.

[35] K. Rajeshwar, J. Ibanez, Environmental Electrochemistry Fundamentals and Applications in Pollution Abatement, Academic Press, London, 1997.

[36] G. Chen, Electrochemical technologies in wastewater treatment, Sep. Purif. Technol., 38 (2004) 11–41.

[37] K. Thirugnanasambandham, V. Sivakumar, J.P. Maran, Response surface modelling and optimization of treatment of meat industry wastewater using electrochemical treatment method. J. Taiwan Inst. Chem. Eng., 46 (2015) 160–167.

[38] N. Yusoff, M. Ramasamy, S. Yusup, Taguchi's parametric design approach for the selection of optimization variables in a refrigerated gas plant, Chem. Eng. Res. Des., 89 (2011) 665–675.

[39] J.M. Parks, On stochastic optimization: Taguchi Methods demystified; its limitations and fallacy clarified, Probab. Eng. Mech., 16 (2001) 87–101.

[40] K. Yang, E.C. Teo, F.K. Fuss, Application of Taguchi Method in optimization of cervical ring cage, J. Biomech., 40 (2007) 3251–3256.

[41] A. Deghles, U. Kurt, Treatment of raw tannery wastewater by electrocoagulation technique: optimization of effective parameters using Taguchi method, Desal. Wat. Treat., 57 (2016) 14798–14809.

[42] F. Özyonar, H. Muratcobanoglu, O. Gökkus, Taguchi approach for color removal using electrocoagulation with different electrode connection types, Fresenius Environ. Bull., 26 (2017) 7600–7607.

[43] H.Y. Yen, C.P. Lin, Adsorption of Cd(II) from wastewater using spent coffee grounds by Taguchi optimization, Desal. Wat. Treat., 57 (2016) 11154–11161.

[44] F. Özyonar, Optimization of operational parameters of electrocoagulation process for real textile wastewater treatment using Taguchi experimental design method, Desal. Wat. Treat., 57 (2016) 2389–2399.

[45] M.P. Elizalde-Gonzalez, L.E. Garcia-Diaz, Application of a Taguchi L16 orthogonal array for optimizing the removal of Acid Orange 8 using carbon with a low specific surface area, Chem. Eng. J., 163 (2010) 55–61.

[46] S. Irdemez, Y.S. Yildiz, V. Tosunoglu, Optimization of phosphate removal from wastewater by electrocoagulation with aluminum plate electrodes, Sep. Purif. Technol., 52 (2006) 394–401.

[47] APHA, AWWA, Standard Methods for the Examination of Water and Wastewater, 20th ed., American Public Health Association, Washington, DC, 1998.

[48] O. Gökkus, Y.S. Yıldız, B. Yavuz, Optimization of chemical coagulation of real textile wastewater using Taguchi experimental design method, Desal. Wat. Treat., 49 (2012) 263–271.

[49] K.S.P. Kalyani, N. Balasubramanian, C. Srinivasakannan, Decolorization and COD reduction of paper industrial effluent using electro-coagulation, Chem. Eng. J., 151 (2009) 97–104.

[50] F. Ozyonar, B. Karagozoglu, Treatment of pretreated coke wastewater by electrocoagulation and electrochemical peroxidation processes, Sep. Purif. Technol., 150 (2015) 268–277.

[51] I. Heidmann, W. Calmano, Removal of Cr(VI) from model wastewaters by electrocoagulation with Fe electrodes. Sep. Purif. Technol., 61 (2008) 15–21.

[52] C. Ricordel, H. Djelal, Treatment of landfll leachate with high proportion of refractory materials by electrocoagulation: system performances and sludge settling characteristics. J. Environ. Chem. Eng., 2 (2014) 1551–1557.

[53] B.G. Chen, X. Chen, P.L. Yue, Electrocoagulation and electroflotation of restaurant wastewater, J. Environ. Eng., 126 (2000) 858–863.

[54] P. Asaithambi, D. Beyene, A.R.A. Aziz, E. Alemayehu, Removal of pollutants with determination of power consumption from landfill leachate wastewater using an electrocoagulation process: optimization using response surface methodology (RSM), Appl. Water Sci., 8 (2018) 69.

[55] N. Modirshahla, M.A. Behnajady, S. Kooshaiian, Investigation of the effect of different electrode connections on the removal efficiency of Tartrazine from aqueous solutions by electrocoagulation, Dyes Pigm., 74 (2007) 249–257.

[56] M. Kobya, O.T. Can, M. Bayramoglu, Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes, J. Hazard Mater., 100 (2003) 163–178.

[57] Ö. Apaydin, E. Özkan, Leachate Treatment by Electrocoagulation, 15th International Conference on Environmental Science and Technology, Rhodes, Greece, August 31 to September 2, 2017.