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Treatment of the OUED SMAR town landfill leachate by an electrochemical reactor

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ABSTRACT

The electrocoagulation process is an effective, fast and economic technique for the treatment of water and wastewater. In this paper, electrocoagulation (EC) has been used for the removal of COD, total nitrogen, color, turbidity and bacteria from the Oued Smar (north of Algeria) town landfill leachate, characterized by high chemical oxygen demand, high concentration of nitrogen and black color. Firstly, the effects of process variables such as inter-electrode distance, magnetic stirring speed, current density and electrode material on the treatment efficiency, sludge volume production, pH and temperature evolution during the EC process were studied. Secondly, energy consumption and operating costs were calculated with aluminium and iron electrodes under the same experimental conditions.

The findings, in this study show that an increase in current density (125–500 A/m^2) enhanced the speed of treatment significantly, the inter-electrode distance was 2.8 cm and the stirring speed was 150 rpm for the studied leachate. The removal efficiencies of COD, total nitrogen, color and turbidity were respectively 70%, 24%, 56%, and 60% with Al electrodes and 68%, 15%, 28%, and 16% with Fe electrodes. Electrical energy consumption and operating cost with Al electrodes were 0.022 (kWh/L), 0.54 (US\$/m$^3_{\text{leachate treated}}$), respectively, and 0.019 (kWh/L), 0.47 (US\$/m^3) with Fe electrodes.

1. Introduction

One of the most important problems with designing and maintaining a landfill is managing the leachate that is generated when water passes through the waste. The leachate consists of various organic and inorganic compounds which may be either dissolved or suspended. These compounds are a potential pollution problem for ground and surface waters. The removal of these compounds from leachate is the usual prerequisite before discharging the leachate into natural waters.

Conventional landfill leachate treatments can be classified into four major groups: (a) leachate transfer: recycling and combined treatment with domestic sewage [1,2], (b) biodegradation: aerobic and anaerobic processes [3,4], (c) chemical and physical methods: chemical oxidation [5], Fenton's oxidative treatment [6] adsorption [7], chemical precipitation [8], coagulation/flocculation [7,9] sedimentation/flotation and air stripping [9,10], (d) membrane processes: nanofiltration [11–13], microfiltration [13], ultrafiltration [14] and reverse osmosis, [15,16].

Electrocoagulation is a simple and efficient method where the coagulating agent is generated in situ by dissolving electrochemically either aluminium or iron ions from respectively aluminium or iron electrodes. In this process the treatment is done without adding any chemical reagent, thus reducing the amount of sludge which must be disposed [17]. Its advantages include high particulate removal efficiency, compact treatment facility, relatively low cost and possibility of complete automation [18].

In this study, a pretreatment method involving electrocoagulation process is proposed and investigated. Electrocoagulation is the process of destabilizing suspended, emulsified, or dissolved contaminants in an aqueous medium by introducing an electric current into the medium. In its simplest form, an electrocoagulation reactor may be made up of an electrolytic cell with one anode and one cathode. The conductive metal plates are commonly known as 'sacrificial electrodes' and may be made of the same or different materials (anode and cathode) [19].

Electrocoagulation has been successfully used for the treatment of wastewaters such as electroplating wastewater [17], chemical mechanical polishing wastewater [19,20], laundry wastewater [21], pulp paper mill industry [22–24] and wastewater textile [25–27] and water purification [28] and urban wastewater [29], water defluoridation [31] and industrial water defluoridation [32], olive mill wastewater [33–35], and tannery wastewater [36], and baker's yeast wastewater [37] and slaughterhouse wastewater [38]. It has also been used to treat the leachate by some researchers [39–41].

In the present work, the efficiency of electrocoagulation in removing COD, turbidity, color, total nitrogen and bacteria from leachate of OUED SMAR town landfill was reported. The effect of operational parameters such as current density, inter-electrode distance, magnetic stirring speed and electrode material on the process efficiency, sludge production, pH and temperature evolution

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was explored and discussed to determine the optimum conditions. At last, a comparative operating cost analysis in term of electrical energy and electrode was presented.

1.1. Brief description of EC process

Electrocoagulation process involves three stages; coagulant formation through dissolution of metal ions of anode electrode, destabilization of pollutants, suspended particles and de-emulsification, and aggregation of instable phases and floc-forming [17,18,20]. Destabilization of pollutants, suspended particles and de-emulsification mechanism can be established through dispersed double layer compression, ion neutralization species existing in water and wastewaters, and flocs and sludge forming [17,42].

The electrocoagulation mechanism can be evaluated from the following equations:

The electrolytic dissolution of Al anodes by oxidation produces aqueous Al^{3+} species [42] and the electrode reactions are outlined below:

$$Al_{(s)} \rightarrow Al^{3+} + 3e^{-}. \quad (1)$$

At the aluminium cathodes reduction takes place which results in hydrogen bubbles being produced by the following reaction:

$$2H_2O + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}. \quad (2)$$

The H_2 bubbles float in EC reactor. The Al^{3+} ions further react as shown in Eq. (1) to a solid $Al(OH)_3$ precipitate. Those precipitates form flocs that combine water contaminants as well as a range of coagulant species and metal hydroxides formed by hydrolysis as shown below:at acidic conditions

$$Al^{3+} + 3H_2O \leftrightarrow Al(OH)_{3(S)} + 3H^{+} \quad (3)$$

at alkaline conditions

$$Al^{3+} + 3OH^{-} \rightarrow Al(OH)_3. \quad (4)$$

These coagulants destabilize suspended particles or precipitate and adsorb dissolved contaminants.

In addition, there is an oxygen evolution reaction

$$2H_2O + 4e^{-} \rightarrow O_2 + 4H^{+}. \quad (5)$$

The reaction at the cathode is

$$2H_2O + 2e^{-} \rightarrow H_2 + 2OH^{-}. \quad (6)$$

A mechanism for the production of the metal hydroxides using iron anodes in the range of pH 4 to pH 9 has been proposed [42]:

$$\text{Anode}: 2Fe_{(s)} + 12H_2O_{(l)} \rightarrow 2Fe(H_2O)_3(OH)_{3(aq)} + 6H^{+} + 6e^{-} \quad (7)$$

$$\text{Bulksolution}: 2Fe(H_2O)_3(OH)_{3(aq)} \rightarrow 2Fe(H_2O)_3(OH)_{3(s)} \quad (8)$$

$$\text{Bulksolution}: 2Fe(H_2O)_3(OH)_{3(s)} \rightarrow Fe_2O_3(H_2O)_{6(s)} + 3H_2O_{(l)} \quad (9)$$

$$\text{Cathode}: 6H^{+} + 6e^{-} \rightarrow 3H_{2(g)} \quad (10)$$

$$\text{Overall}: 2Fe_{(s)} + 9H_2O_{(l)} \rightarrow Fe_2O_3(H_2O)_{6(s)} + 3H_{2(g)}. \quad (11)$$

The Fe $(OH)_2$ and Fe $(OH)_3$ formed remains in the aqueous solution as a gelatinous suspension, which can remove pollutants from wastewater either by electrostatic attraction or by complexation followed by coagulation.

Ferric ions generated by the electrochemical oxidation of the iron electrode may form monomeric ions, Fe $(OH)_3$, and hydroxyl complexes with hydroxide ions and polymeric species, namely, $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^{+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$, and $Fe_2(H_2O)_6(OH)_4^{4+}$. The formation of these complexes depends strongly on the solution pH [42].

Formation of rust (dehydrated hydroxides) occurs as shown in the following [43]:

$$2Fe(OH)_3 \rightarrow Fe_2O_3 + 3H_2O \quad (\text{hematite, maghemite}) \quad (12)$$

$$Fe(OH)_2 \rightarrow FeO + H_2O \quad (13)$$

$$2Fe(OH)_3 \rightarrow Fe(OH)_2 + Fe_3O_4 + 4H_2O \quad (\text{magnetite}) \quad (14)$$

$$Fe(OH)_3 \rightarrow FeO(OH) + H_2O \quad (\text{goethite, lepidocrocite}). \quad (15)$$

2. Materials and methods

2.1. Landfill site description and leachate characteristics

The study was carried out in Algiers (North of Algeria) Domestic Solid Waste Landfill, located in the village of OUED SMAR, 13 km away from the east of Algiers. The landfill is constructed on a total area of 375,000 m^2. Annual range temperature is 2–45 °C, and the rainfall is 700–800 mm. The daily solid waste entering the plant is approximately 1600–4000 t.

Leachate utilized in this study was collected from the OUED SMAR town landfill in a closed container and stored in obscurity at T = 4 °C. It was characterized using the standard methods [44,45]. The pH, conductivity and turbidity were measured using HANNA pH-meter (pH211), HANNA conductivity-meter (EC 214) and HANNA turbidmeter (2100N, HACH, USA) respectively. COD was measured by using Thermoreactor Closed Reflux Colorimetric Method (5220D). Suspended solids were obtained by centrifugation then drying at 105 °C [50], BOD_5 analysis was carried out using OXYTOP WTW (T90103) and the color intensity was determined by measuring the sample absorbance at 450 nm [50] using the UV–Vis spectrophotometer, mini1240SHIMADZU. Total Coliforms and *Clostridium* were counted using the BCPL medium (NF T 90-413) and liver-meat Agar (NF T 90-415) respectively. The main characteristics of this effluent before treatment are presented in Table 1.

The ratio BOD_5/COD value (0.12) indicates the less biodegradability of leachate compared to untreated wastewaters in which ratios encountered are in the range of 0.4 to 0.8. This result authorizes the utilization of the electrochemical process to remove the organic compounds from leachate before discharging the leachate into natural waters.

Table 1
Characteristics of leachate.

Parameter	Data ranges
pH	7.6–8.9
Conductivity (ms/cm)	12.15–13.55
COD (mg/l)	28,200–34,200
Suspended solids (mg/l)	490–520
BOD5/COD	0.09–0.12
Turbidity (NTU)	125–141
Total nitrogen (mg/l)	1599–1631
Total Coliforms	$2.5 \cdot 10^4$/100 ml
Clostridium	150/100 ml

2.2. Reactor design

The experimental setup is shown in Fig. 1. The electrocoagulator was made of Plexiglas cell, with a special cover supporting a series of parallel aluminium or iron sheets used as sacrificial electrodes. Both aluminium cathodes and anodes were made from plates with dimensions of 50×40×1 mm (L×W×T).

The experiments were realised with 500 cm^3 of the leachate placed into the electrolytic cell, the electrodes were connected to a DC power supply, voltage and intensities supplied was respectively 10 V, 0.5 A, 1 A and 2 A. All the experiments were performed in batch mode at room temperature. At the end of each EC treatment study, a solution with flocks was allowed to settle for 1 h in the container before chemical analysis, the amount of sludge produced was expressed as the ratio to whole solution. The samples for chemical analysis were taken from limpid phase. Neither centrifuging nor filtration was performed in this study.

In preliminary tests, the relative standard deviation was estimated and experiments were repeated to determine the experimental error.

3. Results and discussion

In this section, results obtained during the study are given and discussed.

3.1. Repeatability tests

To estimate the relative standard deviation (RSD%) [50] during the treatment of leachate by electrocoagulation, a series of four experiments was carried out with aluminium electrodes under the same experimental conditions (0.5 L of leachate, current density = 175 A/m^2, inter-electrode distance = 2.8 cm, magnetic stirring speed = 150 rpm, treatment time = 30 min).

At the end of each test and after 1 h settling period, the removal efficiency of COD, total-N, color and turbidity was calculated, sludge volume was determined and the quantity produced was expressed as the ratio to whole solution.

From the results obtained, presented in Table 2, we can conclude that electrocoagulation makes it possible to carry out an electrochemical treatment of leachate with an interesting repeatability (R.S.D. <3%).

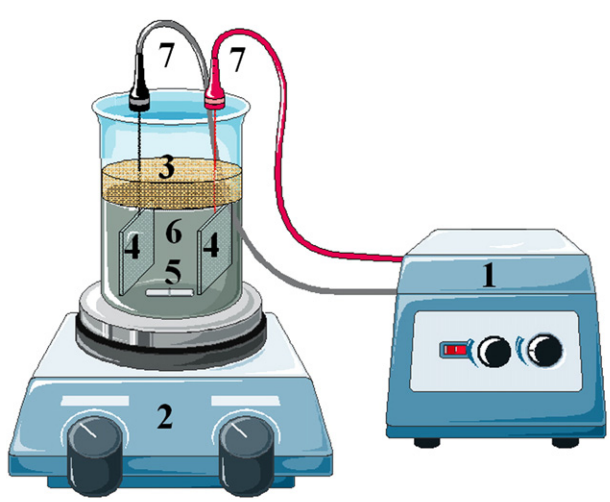

Fig. 1. The electrochemical reactor in the laboratory experiments. (1) DC power supply, (2) magnetic stirrer, (3) cover, (4) Al electrodes, (5) magnetic bar-stirrer, (6) leachate and (7) electric wire.

Table 2
Results of repeatability tests.

	COD (%)	Total-N (%)	Turbidity (%)	Color (%)	Sludge (%)
Test 1	54.37	13.95	41.35	24.45	14.98
Test 2	53.15	14.4	39.1	23.87	16.01
Test 3	51.46	15	40.6	23.41	15.68
Test 4	52.54	14.37	39.1	23.99	15.22
$\bar{x}$	52.88	14.43	40.04	23.93	15.47
S_r^2	1.47	0.18	1.26	0.18	0.21
R.S.D. (%)	2.29	2.9	2.8	1.75	2.9

$\bar{x} = \sum^{ni}_{i=1} x_i/n$: arithmetic mean.
$S_r^2 = \sum^{ni}_{i=1} (x_i - \bar{x})^2/n-1$: standard deviation of repeatability.
R.S.D. = $100(S_r/\bar{x})$: relative standard deviation.

3.2. Effect of the inter-electrode distance on the electrocoagulation efficiency

In a parallel-plate monopolar reactor, the electrical field can be controlled by varying the applied current, but once distance between the electrodes changes the electrical current change. In order to investigate the effect of inter-electrodes distance on the efficiency of process, the reactor was arranged such that the electrode was positioned at either 0.5 cm or 2.8 cm apart. Fig. 2 shows the COD, turbidity and color removal efficiency using various inter-electrode distance at the same experimental conditions.

The results indicate that when the inter-electrode distance was increased from 0.5 to 2.8 cm, the removal of COD increased by about 10% after 15 min of treatment time. However, for turbidity and color removal, no discernable differences were observed. The inter-electrode distance retained for the treatment of leachate was 2.8 cm. COD abatement ratio of about 70% was obtained and then allows rejecting directly this treated water to the sewage plant [1]. This result is more interesting than those reported by research teams utilizing coagulation–flocculation technique and chemical precipitation with abatement ratios ranging from 22% to 30% [1,39].

3.3. Effect of stirring speed on the process efficiency

The theory of EC has been discussed by a number of authors, it is generally accepted that the EC process involves three successive stages:(1) formation of coagulants by electrolytic oxidation of the sacrificial electrode; (2) destabilization of the contaminant and particulate suspension; (3) aggregation of the destabilized phases to form flocs. During the second stage, homogeneous solution medium will be much more efficient for the contact between the contaminant and coagulants.

To determine the effect of mixing on the efficiency of this electrochemical process, experiments were carried out using various stirring speed values. As shown in Fig. 3(a), the COD removal efficiency increase by about 12%, after 15 min of treatment time when the magnetic stirring speed was increased from 50 rpm to 150 rpm.

Sludge production is another important parameter in characterizing the EC process. So, the quantity of sludge produced was examined. It is seen from Fig. 3(b) that the increase in sludge production by increasing stirring speed from 50 to 150 rpm was 15% at the end of 30 min contact time. With increasing agitation, more interaction of coagulants with contaminant is expected. A value of about 150 rpm was retained.

3.4. Effect of current density on the process efficiency

Operating current density is critical in electrocoagulation process, as it is the only operational parameter that can be controlled directly. In this system electrode spacing is fixed and current is supplied continuously. It is well-known that the current density determines

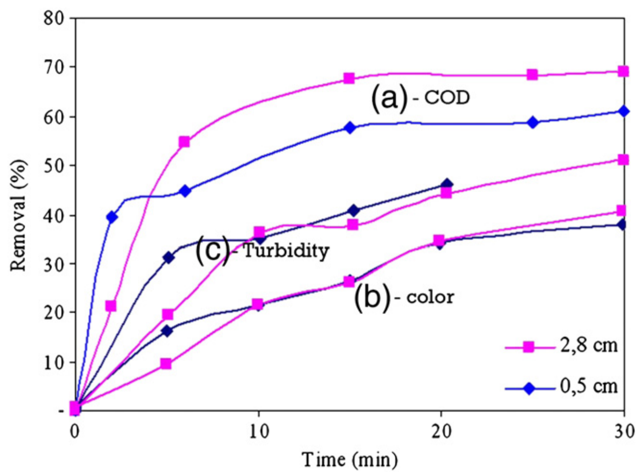

Fig. 2. Effect of the inter-electrode distance on the electrocoagulation efficiency, (a) COD removal, (b) color removal, and (c) turbidity removal. (Current density = 250 A/m^2, stirring speed = 150 rpm).

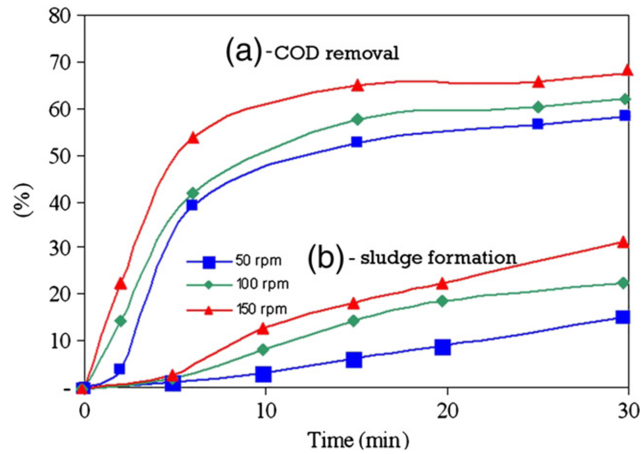

Fig. 3. Effect of stirring speed on the treatment efficiency, (a) COD removal and (b) sludge formation. (Current density = 250 A/m^2, inter-electrode distance = 2.8 cm).

the coagulant dosage rate, and adjusts the rate and size of the bubble production, and hence affects the growth of flocs [30,31,42], which can influence the treatment efficiency of electrocoagulation.

Another experimental test was performed to determine the effects of current density on COD, turbidity, total nitrogen and color removal efficiency by increasing its value from 125 to 500 A/m^2. It would also be of interest to examine its effects on the quantity of sludge, change of temperature and pH during electrocoagulation. The results are shown in Fig. 4.

When current density was increased from 125 to 500 A/m^2, the removal efficiency of color turbidity COD, total nitrogen and the quantity of sludge generated were respectively increased from 3% to 21%, 4% to 36%, 41% to 58%, 2% to 9%, and 2% to 13% in the first 5 min and from 17% to 56%, 23% to 60%, 56% to 71%, 14% to 23%, and 15% to 60% after 30 min of treatment time.

In EC process, the amount of metal dissolved is dependent on the quantity of electricity passed through the solution. A simple relationship between current and the amount of metal dissolved can be derived from Faraday's law [30]:

$$m = I.M.t/n.F.V \tag{16}$$

where m is the metal concentration in the electrolytic cell, I the current intensity (A), t the time (s), M the molecular weight of metal, n the number of electron moles, F the Faraday's constant (96,485 C/mol) and V the volume of electrolytic cell. According to the present operating conditions, the concentration of aluminium released to the aqueous solution was calculated and presented in Fig. 4(f).

The highest current produced the quickest treatment; at high current the amount of aluminium oxidized increases, resulting in a greater amount of precipitate for the removal of pollutants. In addition, it was demonstrated that bubbles density increases and their size decreases with increasing current density [48,49], resulting in a greater upwards flux and faster removal of color and pollutants.

The temperature and pH are continuously observed during the treatment. Temperature in the reactor increases with increasing current density and contact time, when current density was 125 A/m^2, temperature changed from 17.9 to 19.6 °C and from 17.9 to 34.6 °C when current density was 500 A/m^2. The temperature tends to increase as a result of electrolytic reactions. pH gradually increased due to dominant activities of the cathode, when current density was 125 A/m^2, pH changed from 8.25 to 8.48 and from 8.25 to 8.69 when current density was 500 A/m^2.

3.5. Effect of the electrode material

The most widely used materials are aluminium and iron. For comparative purpose, electrocoagulation was carried out with both materials under similar experimental conditions. The compared results of COD, color, total nitrogen, turbidity, temperature, pH and sludge production, obtained with the same current density (500 A/m^2) and operating time of 30 min are presented in Fig. 5.

In Fig. 5(a), it appears clearly that both materials are almost equally effective in reducing COD. However, aluminium was found to be more effective in removing turbidity and color from leachate than iron. The removal efficiency of total nitrogen was increased from 15 to 24% when Fe electrodes were changed by Al electrodes. The pH of leachate changes during the process from 8.25 to 8.69 with Al electrodes and from 8.25 to 9.24 with Fe electrodes.

The reduction of COD is in the same magnitude for both metals used; this is the training for both hydroxides of iron and Al which polymerize in much the same way [47,48].

For color and turbidity, the resulting effluent treated with aluminium was found clear and stable, whereas that treated with iron electrodes appeared greenish first, and then turned yellow and turbid. The green and yellow colors could have resulted from Fe(II) and Fe(III) species generated during the electrolysis and characterized by their yellow–brown color [50].

According to Fig.5(b), the least volumetric sludge production was determined to be in the process with Fe electrodes, it was increased from 30% to 60% when Fe electrodes were changed by Al electrodes.

3.6. Total Coliform and Clostridium removal

The leachate consists of various pathogenic microorganisms and parasites, they pose a potential pollution problem for local ground and surface waters. The OUED SMAR town landfill leachate was found to have a total Coliform concentration in the range of $2.5 \cdot 10^4$ TC/100 ml and *Clostridium* in the range of 150 C/100 ml.

Another experimental test was performed to determine the effect of electrocoagulation on the removal efficiency of bacteria. The electrocoagulation process appeared to be an efficient means to remove the microorganisms from leachate. Indeed, total Coliform and *Clostridium* removal of about 99% and 96% was obtained with the same current density (500 A/m^2) and operating time of 30 min.

3.7. Operating cost

One of the most important parameters that must be determined to evaluate a method of wastewater treatment is the cost. The operating

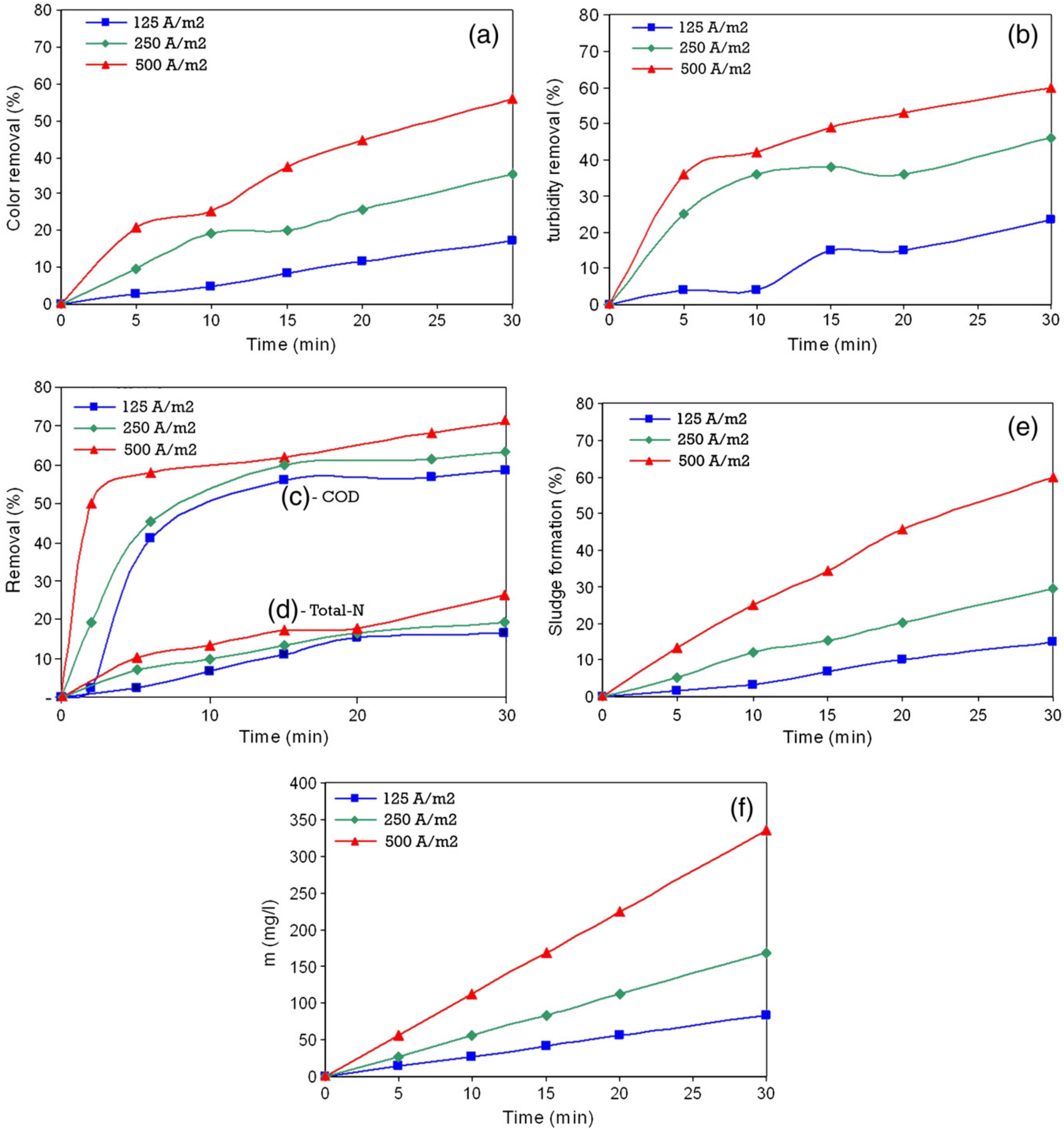

Fig. 4. Effect of current density on the process efficiency (a) color removal, (b) turbidity removal, (c) COD removal, (d) total-N removal, (e) sludge formation ratio, and (f) theoretical metal dissolution. (Stirring speed = 150 rpm, inter-electrode distance = 2.8 cm).

cost includes material (mainly electrodes) cost, utility (mainly electrical energy) and other cost items such as labor, maintenance, sludge dewatering and disposal. In this study, energy and electrodes material costs are taken into account as major cost items.

Electrical energy consumption was calculated using the commonly used Eq. (17)[26,46]:

$$E = Uit \tag{17}$$

where E is the electrical energy in KWh per kg of pollutant removal or per volume of leachate treated, U the cell voltage in volt (V), I the current in ampere (A) and t is the time of treatment.

The calculation of current efficiency (φ) (Eq. (18)) was based on the comparison of experimental weight loss of electrodes (ΔM_{exp}) during electrocoagulation with theoretical amount of metal dissolution (ΔM_{theo}) according to the Faraday's law (Eq. (19))

$$\varphi\ (\%) = 100\left(\Delta M_{Exp} / \Delta M_{Theo}\right) \tag{18}$$

$$\Delta M_{Theo} = MIt/nF \tag{19}$$

where M is the molecular weight of the metal (g/mol), n the number of electron moles and F is the Faraday constant ($F = 9.6485 \cdot 10^4\ C\ mol^{-1}$).

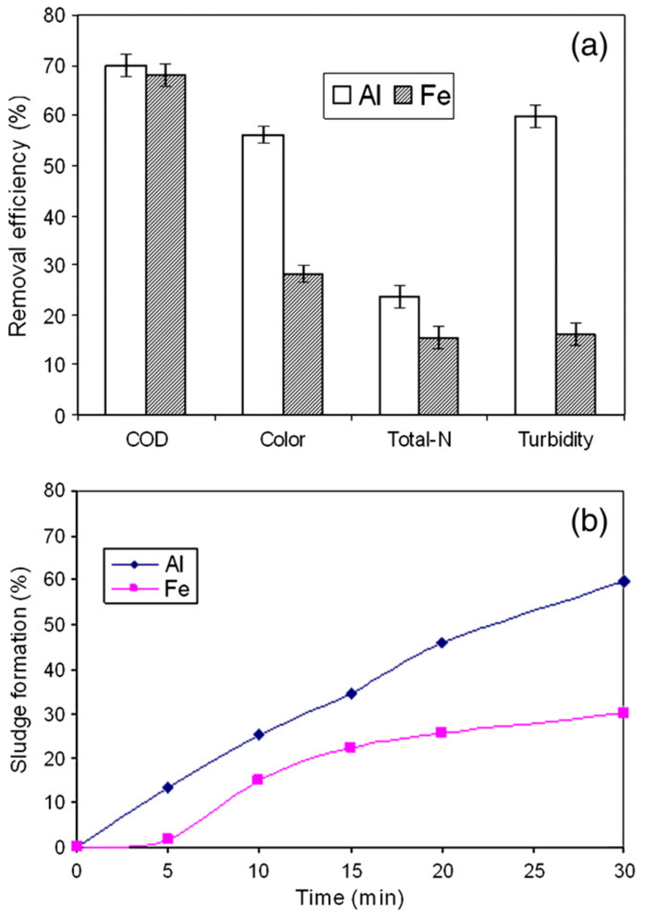

Fig. 5. Effect of electrode material on the electrocoagulation efficiency, (a) COD, color, turbidity and total-N removal and (b) sludge formation ratio. (Current density = 500 A/m^2, stirring speed = 150 rpm, inter-electrode distance = 2.8 cm).

The specific electrical energy consumption (Seec) was calculated as a function of Fe and Al electrodes weight consumption during treatment in kWh/kg $_{\text{electrode material}}$ by Eq. (20)[46]

$$\text{Seec} = 10^{-3}\ \text{nFU} / 3.6\ \ \text{M}\varphi. \tag{20}$$

Given Algerian market in January 2011, unit prices are as follows: electrical energy price 0.0247 US$/kWh and electrode material price 0.36 US$/kg Aland 0.3 US$/kg Fe. The calculated values are shown in Table 3.

As seen, it is clear that iron electrodes are energetically efficient than aluminium. The operating cost increases from 0.47 to 0.54 US$/m^3 when Fe electrodes were changed by Al electrodes.

4. Conclusion

The present study has shown the applicability of electrocoagulation method in the treatment of leachate from OUED SMAR town landfill. The influence of variables such as stirring speed, current density, inter-electrode distance and the type of electrode on the removal efficiency of COD, total nitrogen, color and turbidity has been studied. The sludge volume produced has been determined.

Table 3
Characteristics parameters calculated for EC process with Al and Fe electrodes.

Metal	E (kWh/L)	ΔM_{Exp}	ΔM_{Theo}	φ (%)	Seec (kWh/kg$_{met}$)	OC (US$/m^3)
Fe	0.0196	1.0842	1.041	104.15	0.0922	0.47
Al	0.022	0.391	0.336	116.37	0.2559	0.54

Concentration of the pollutants was significantly reduced during the treatment process. The removal percentage has a tendency to increase with the increase in current density and stirring speed. The results indicate that more than 70%, 60%, 56%, and 24% of COD, turbidity, color and total nitrogen were respectively reduced after 30 min of treatment time when current density was applied as 500 A/m^2.

When Al electrodes were changed by Fe electrodes: the removal efficiency of COD, total nitrogen, color and turbidity and the sludge production were decreased respectively from 70 to 68%, 24 to 15%, 56 to 28%, 60 to 16%, and 60 to 30% and the final pH of leachate was changed from 8.69 to 9.2.

The electrocoagulation process appeared to be an efficient means to remove the microorganisms from leachate. Indeed, total Coliform and *Clostridium* removal of about 99% and 96% was obtained.

Aluminium and iron electrodes are almost equally efficient in reducing COD but the iron electrodes are energetically efficient than aluminium. The operating cost increases from 0.47 to 0.54 US$/m^3 when Fe electrodes were changed by Al electrodes. Thus, iron which is less toxic and more acceptable in agriculture field may be retained as the most appropriate electrode material to treat the leachate by electrocoagulation process.

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