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Modelling current efficiency and ohmic potential drop in an innovated electrocoagulation reactor

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ABSTRACT

The objective of the present work is to study the impact of the electrodes configuration utilized in an innovated electrocoagulation reactor on the behavior of current efficiency and the ohmic potential drop via the treatment of real oily wastewater released from crude oil wells location (West Qurna 1 /Basra-Iraq). A monopolar mode of triple aluminum tubes was employed in a concentric arrangement to investigate the purpose of this work under the influences of the operational variables; the electrolysis time (10, 14, 25, 36, 40 min) and electric current density (1.75, 2.53, 4.39, 6.25, 7.02 mA/cm^2). The initial pH equaling 6.5 and the mixing speed was taken as 200 rpm. The results reveal a significant effect of the invented electrodes configuration on the studied responses values where they had decreased from 150 to 125% and 67.85 to 66.19 ohm respectively as the electrolysis time increases from 10 to 40 min whereas the ohmic drop raises from 28.02 to 107.64 ohm as the current density increased from 1.75 to 7.02 mA/cm^2 but the current efficiency had minimized from 157 to 114% for the same increment in current density. The lower values of the operational variables, the higher value of the current efficiency which is the most significant response. This study proved a notable efficiency of the present design of the electrocoagulation reactor.

Keywords: Electrocoagulation; Electrodes configuration; Current efficiency; Ohmic drop; Real oily wastewater

1. Introduction

Electrochemical reactors, among the treatment methods, are using the electrical power as the main parameter in their operation in order to remove the undesirable pollutants from several types of wastewater [1,2]. These types of reactors are consuming different values of electric current where this consumption is depending on several aspects such as the voltage applied, the configuration and arrangement of electrodes, etc. [3–6].

The electrocoagulation reactor is one of the electrochemical reactors that depends extremely on the current density in order to accomplish the removal of contaminants through the adsorption technique such as the removal of pigment [2], heavy metals [7–11], detergent [12] and oil content [13].

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In order to distinguish which reactor is the most practical among other kinds of electrochemical reactors, the most efficient electrocoagulation reactor the most effective of the electric current [5,14,15].

There are several kinds of electrocoagulation reactors that differ in their electrodes design according to configuration [16], arrangement [17], type of metal [18], and shape [19] or in the type of the electric current supplied which is AC or DC [20], shape of the reactor, and the operation mode of the reactor [21,22].

The performance of such type of reactors, used in wastewater treatment, is characterized according to several aspects such as the efficient removal of pollutants, energy efficiency and electrodes consumption [23]. For that purpose, several studies have been done in order to investigate the effect of reactor design on the performance such as the variation of electrodes shapes, i.e. plane plate, mesh, or cylindrical [23,24–27], and the arrangement of electrodes, i.e. bipolar [7,8] or monopolar [9,28].

The current efficiency, i.e. Faradaic efficiency or faradaic yield, is almost always an essential aspect from the energetic view in any type of electrochemical reactors especially the electrocoagulation reactor because the combined process of adsorption occurred in such reactor depends extremely on this important parameter [11]. The estimation of current efficiency value is obtained by the measurement of the theoretical and actual consumption values of electrodes as illustrated in Eqs. (1) and (2) [9,29]:

$$\text{Current efficiency} = (m_{exp.} / m_{theo.})*100 \quad (1)$$

$$m_{exp.}(g) = w_1 - w_2 \quad (2)$$

where $m_{exp.}$ is the experimental value of electrodes consumption, w_1 is the weight of electrodes before the experiment, and w_2 is the weight of electrodes after the experiment, and $m_{theo.}$ is the theoretical value of electrodes consumption that could be measured from Eq. (3) as follows:

$$m_{theo}(g) = CD \cdot t \cdot M / Z \cdot F \quad (3)$$

where CD is the current density in (mA/cm^2), t is the electrolysis time in (second), M is the molecular weight of electrodes metal in (g/mol), Z is the number of electrons presented in the reaction (for Al is 3 and Fe is 2), and F is Faraday's constant which equals (96485.34 Columb/mol).

The ohmic potential drop among the electrodes is an impalpable factor that may affect the performance of the current efficiency as well as the energy consumption. It is practically depending on several parameters such as the distance between electrodes, the current density, and the conductivity of the polluted solution. Eq. (4) represents the correlation of ohmic drop related to its parameters:

$$ODP = (CD*d)/k \quad (4)$$

where ODP is the ohmic drop (ohm), CD is the current density in (mA/cm^2), d is the distance between electrodes (cm), and k is the solution conductivity (mS/cm).

This study aims to investigate the effect of the electrodes configuration on the behavior of the current efficiency and the potential ohmic drop among the electrodes via the treatment of real oily wastewater discharged from crude oil wells location (West Qurna 1 /Basra-Iraq) under the influences of the electrolysis time and the current density. The present reactor, which has been invented by the author [30], consists of triple concentric aluminum tubes which had been arranged in a monopolar mode. According to the literature and my experience, there is no previous study that is concerned about studying these responses for this kind of electrodes configuration.

2. Experimental work

2.1. Design of experiment

In the present study, a central composite rotatable design (CCRD) and a statistical program (Minitab-17) were carried out to design the experiments and to estimate the effect of the innovated configuration of electrodes used in the electrocoagulation reactor on the behavior of current efficiency and the ohmic drop under specific operational variables listed in Table 1.

A total of 11 experiments were performed as cube points: 4, center points in the cube: 3, axial points: 4, and the rotatability α is +1.414. All of the runs were done at room temperature and a constant mixing speed of 200 rpm. In each experiment, 500 ml of the oily wastewater was placed in the electrocoagulation reactor. The current density was adjusted according to the designed value shown in Table 1, then the experiment was started. At the end of each experiment, the electrodes were washed using 0.1 N HCl and 0.1 N NaOH solutions and washed again by distilled water then dried to be weighted. After that, they are cleaned using sandpaper and washed by distilled water to be ready for the next run.

2.2. Chemicals

The oily wastewater was collected from the effluent of the wet oil's unit, a petroleum station located in West Qurna 1/Basra-Iraq. The characterization of this wastewater is presented in Table 2. Samples were collected in polypropylene containers and preserved at 4°C to be treated by the electrocoagulation reactor.

2.3. Instruments

A conductivity meter (ATC-China) was employed to evaluate the value of conductivity. The electric current was supplied to the electrocoagulation reactor containing the simulated wastewater by using a digital type (model 305D, 0–5 A, 0–30 V) provided by SYADGONG company. Other tools as follow:

1. Digital balance (500g × 0.01g) (PROF company).
2. Magnetic Stirrer (ALFA company: HS-860) ; 0–1000 rpm.

Table 1
Natural and coded operational variables

Natural variable (Xi)	Coded variables				
	–1.414	–1	0	1	1.414
X_1: Electrolysis time (min.)	10	14	25	36	40
X_2: Current density (mA/cm^2)	1.75	2.53	4.39	6.25	7.02

Table 2
Characterization of oily wastewater discharged from West Qurna 1/Basra-Iraq

Parameters	Value
Oil content (ppm)	523.11
pH	6.5
Turbidity (NTU)	168
TS (mg/l)	179023
TSS (mg/l)	65623
TDS (mg/l)	113400
Conductivity (mS/cm)	126

3. Digital timer (SEWAN company).
4. Aluminum tubes with different diameters and thicknesses.

2.4. Apparatus

A batch system of electrocoagulation reactor was made of PVC, with 1 liter of total volume, where the experiments had carried out in a batch mode at room temperature. A concentric configuration of triple aluminum tubes had been utilized where the inner and the outer tubes were selected as the anode while the intermediate tube was selected as the cathode. They were connected to a power source of direct current (DC-power supply) in a monopolar manner.

The X-ray diffractometer (XRD 6000/Shimadzu-Japan) apparatus and the energy dispersive spectrometric (EDS) test used (Oxford instrument-X-act) that was manufactured by United Kingdom had been performed to investigate the elemental composition of aluminum electrodes as is shown its results in Table 3 and Figs. 1 and 2.

The individual active area for each pair of tubes and the distances among these tubular electrodes are listed in Table 4. The schematic of the electrocoagulation reactor and its electrodes is illustrated in Fig. 3.

3. Results and discussion

According to the experimental design of factorial method, the impact of each one of the operational variables on the electric current efficiency, the ohmic drop in both zones and the total ohmic drop are shown in the following explanations which focuses on the effect of the electrodes configuration as well as the electric connection manner on the studied responses.

3.1. Effect of electrolysis time

Table 5 shows the influence of the contact time on the current efficiency when the current density was taken at its mean value (4.39 mA/cm^2). Moreover, the ohmic drop was measured for the two zones, the first one located between the inner tube and the mid tube and the second zone was located between the mid tube and the outer tube, as well as the total value of the ohmic drop had estimated also.

As seen in Table 5, the electric conductivity raises as the electrolysis time increased due to the abundance of different compounds released and interacted along the duration of the experiment as found by AlJaberi, 2018 [15]. Numerous ions are presented in the oily wastewater discharged from West Qurna crude oil location due to the nature of the ground of Basra as noted its characterization in Table 1. During the electrolysis process, the following reactions could be occurred, such as in the present of (Cl) ions, as investigated by Spasojevic et al., 2015 [31] (Eqs. (5) to (10)); therefore, and due to the potential values of these reactions, the conductivity will be increased.

At the anode:

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (5)$$

$$Cl_2 + H_2O \rightarrow HClO + Cl^- + H^+ \quad (6)$$

$$6ClO^- + 3H_2O \rightarrow 2ClO_3^- + 4Cl^- + 6H^+ + (3/2)\,O_2 + 6e^- \quad E_0 = 1.14\ V \quad (7)$$

At the cathode:

$$ClO^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^- \quad E_0 = 0.89V \quad (8)$$

$$2HClO + ClO^- \rightarrow ClO_3^- + 2Cl^- + 2H^+ \quad (9)$$

$$2ClO^- \rightarrow O_2 + 2Cl^- \quad (10)$$

Moreover, the electrical conductivity was varied with the contact time according to the precipitation and con-

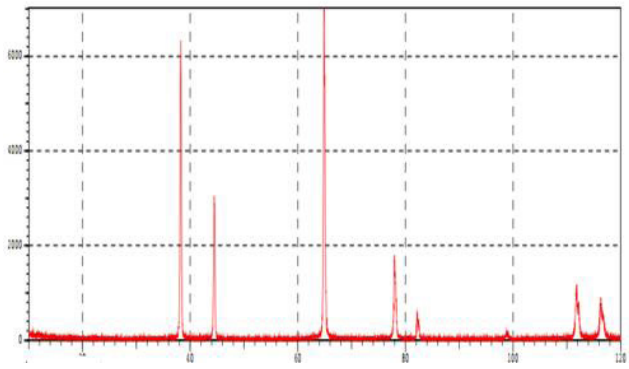


Fig. 1. X-ray -test for aluminum tube.

Table 3
The elemental composition of aluminum tubes as weight percentage.

Metal	Weight %
Al	82.06
Mg	0.97
Fe	0.06
Si	0.76
O	3.82
C	12.33

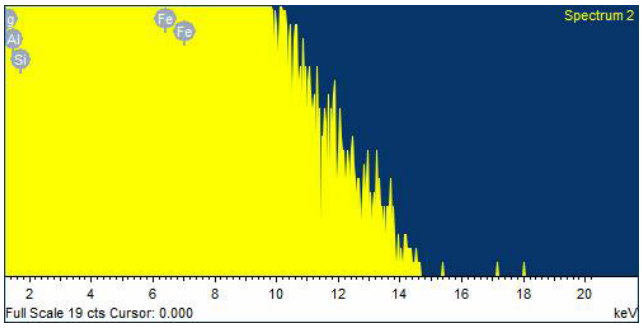


Fig. 2. Elemental composition of aluminum tube (EDS-test).

Table 4
Dimensions of the aluminum tubes electrodes

	Tube wet height (cm)	Outer diameter (cm)	Inner diameter (cm)	Distance in between (cm)	Active surface area (cm^2)
Outer tube (anode)	4.00	7.70	7.30	1.60	165
Mid. tube (cathode)	4.00	5.85	5.55		
Inner tube (anode)	4.00	4.30	3.70	1.55	120

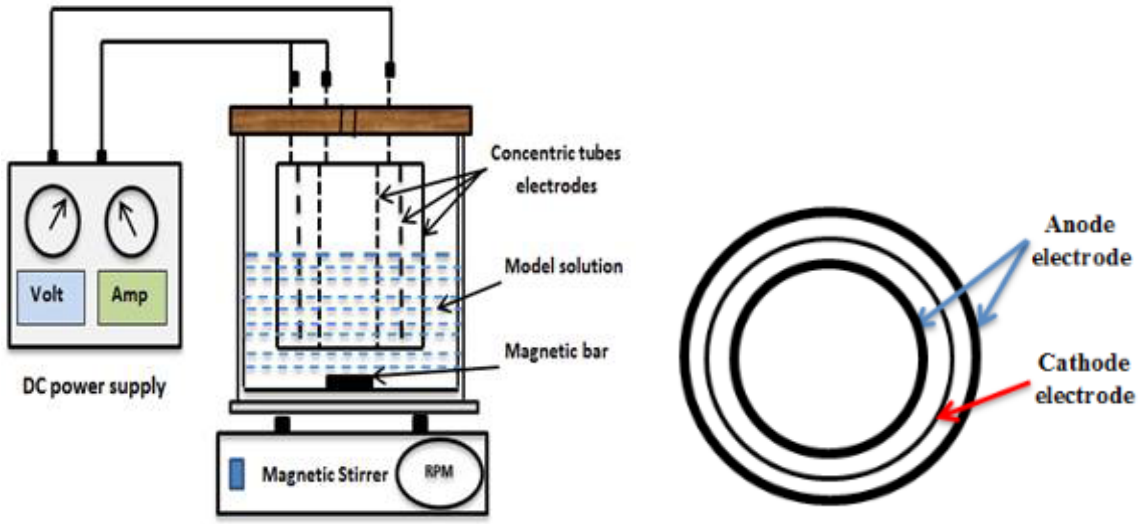


Fig. 3. Schematic of the electrocoagulation reactor and its electrodes.

Table 5
Effect of electrolysis time on the studied responses

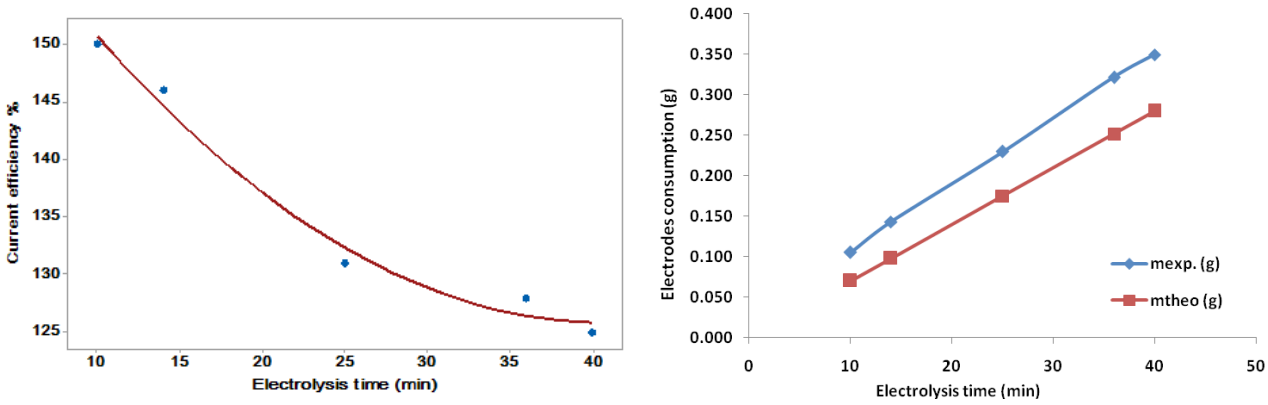
Run No.	Electrolysis time (min)	Final conductivity (mS/cm)	$m_{exp.}$ (g)	m_{theo} (g)	Current efficiency %	Ohmic drop (zone:1) (ohm)	Ohmic drop (zone:2) (ohm)	Total ohmic drop (ohm)
1	10	118.49	0.105	0.070	150	38.51	29.33	67.85
2	14	118.99	0.143	0.098	146	38.35	29.21	67.56
3	25	120.20	0.230	0.175	131	37.97	28.92	66.88
4	36	121.17	0.322	0.252	128	37.66	28.68	66.35
5	40	121.46	0.350	0.280	125	37.57	28.62	66.19

sumption of salts, such as $CaCO_3$ and $MgCO_3$, and total suspended solids, among the concentric electrodes

Fig. 4a shows the inverse relationship between the current efficiency and the electrolysis time in spite of the direct relations between the electrodes consumption (experimental and theoretical values) with the electrolysis time as shown in Fig. 4(b) because the theoretical consumption of the electrodes is estimated according to Eq. (3) which is depended on a fixed values of its parameters. While the experimental values are measured before and after each experiment of the designed time and at the mean value of the current density. Therefore, the irregular difference in the experimental consumption occurred due to the formation of the oxide layer and the accumulation of gases bubbles above the electrodes that will minimize the experimental consumption then lead to the disparity ratios of ($m_{exp.}/m_{theo.}$).

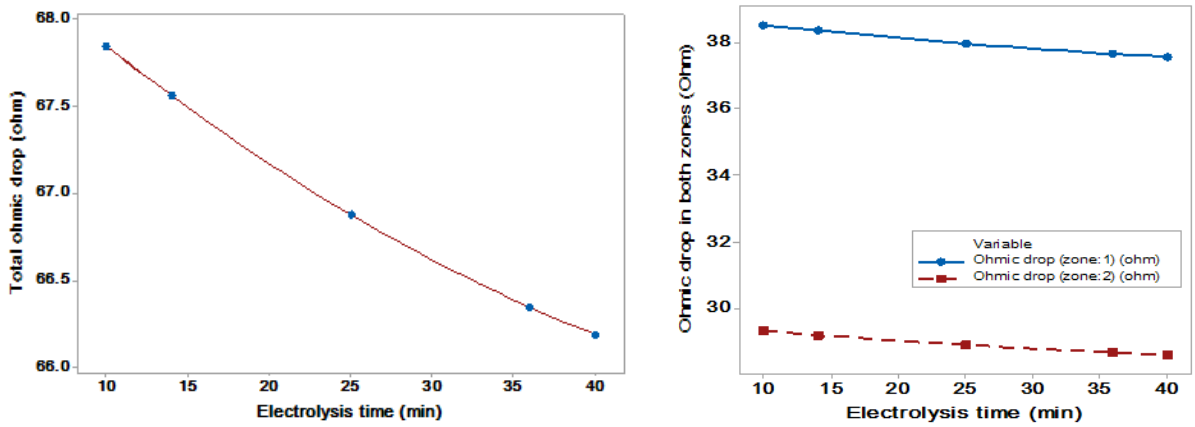
The mathematical correlations of the current density and the experimental and theoretical consumption of the electrodes related to the electrolysis time had been explained in Eqs. (11)–(13). But, in general, the obtained values of the current density had exceeded the 100% which indicates the efficient design of the present electrocoagulation reactor as proved previously by Brahmi et al. [9] and Beyazit [29].

While the ohmic drop is minimizing as the electrolysis time is increased and the electric conductivity increases according to Eq. (4) when the current density was fixed at mean value (Fig. 5). The behavior of this response in both zones is similar but the values of ohmic drop in these zones are different due to the difference in the distance between the electrodes and the increase or decreasing of ions interaction and gases bubbles releasing that could be affecting the value of the conductivity in each zone. Eqs. (14)–(16) relate



(a) (b)

Fig. 4. (a) Current efficiency vs. electrolysis time (current density = 4.39 mA/cm^2); (b) Experimental and theoretical electrodes consumption vs. electrolysis time (current density = 4.39 mA/cm^2).



(a) (b)

Fig. 5. (a) Total ohmic drop vs. electrolysis time (current density = 4.39 mA/cm^2); (b) Ohmic drop in both zones vs. electrolysis time (current density = 4.39 mA/cm^2).

the ohmic drop to the electrolysis time totally and individually.

$$m_{exp.}\ (g) = 0.0081\ t + 0.0262 \quad (R^2 = 0.9995) \tag{11}$$

$$m_{theo.}\ (g) = 0.007\ t + 1x10^{-16} \quad (R^2 = 1.0000) \tag{12}$$

$$\text{Current efficiency (CE\%)} = 169.5 - 2.147\ t + 0.026\ t^2 \quad (R^2 = 0.9850) \tag{13}$$

$$\text{Ohmic drop (zone 1) (ohm)} = 38.95 + 0.04703\ t + 0.000314\ t^2\ (R^2 = 1.00) \tag{14}$$

$$\text{Ohmic drop (zone 1) (ohm)} = 29.69 + 0.03627\ t + 0.000249\ t^2\ (R^2 = 1.00) \tag{15}$$

$$\text{Total ohmic drop (ohm)} = 68.65 + 0.08634\ t + 0.0.000623\ t^2\ (R^2 = 1.00) \tag{16}$$

The contour plot shown in Fig. 6 explains that the maximum value of the current efficiency is obtained at the lowest period of the time's range but the ohmic drop was clearly a high. Therefore, the current density should be increased due to the difficulty of controlling the electrical conductivity that is already possessed by the real oily wastewater. The increment in the conductivity value may have occurred due to the chemical interactions between different ions.

Fig. 7 summarizes the values of the current efficiency against the contact time and the other responses when the current density was fixed at mean value. It explains the decay of current efficiency with time as proved by Spasojevic et al. [31]. As noted, the current efficiency and the potential ohmic drop had decreased from 150 to 125% and 67.85 to 66.19 ohm respectively as the electrolysis time increases from 10 to 40 min in case of mean value of the current density (4.39 mA/cm^2) due to the notable increment of the theoretical consumption of electrodes (m_{theo}) as a result of increasing the electrolysis time then more ions will be released form electrodes due to the continuous passing of the electric current

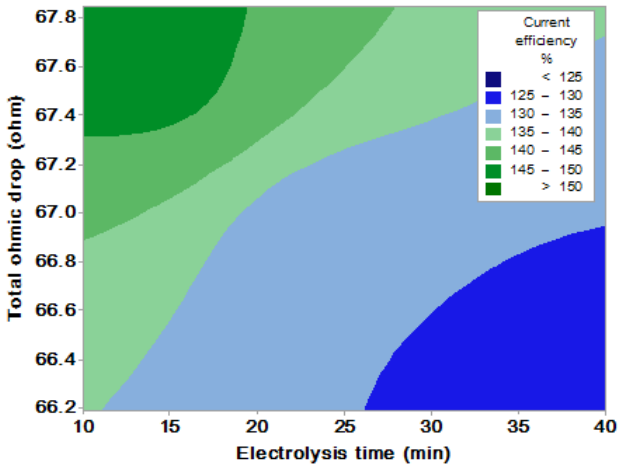


Fig. 6. Contour plot of current efficiency vs. total ohmic drop and electrolysis time.

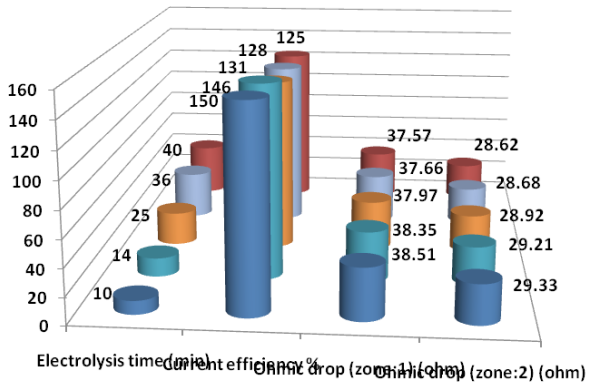


Fig. 7. Current efficiency vs. both zones of ohmic drop and electrolysis time.

through the electrodes that will lead to increase the conductivity then the ohmic drop will be minimized consequently.

3.2. Effect of current density (CD)

The details listed in Table 6 show the same behavior of the final conductivity with the increase of current density because of the continuous releasing of different ions which lead to several reactions among these ions as explained in Eqs. (5)–(10) that are mentioned before in this study. But the increase of the conductivity with the increment of the current density is higher than those noted in case of varying the electrolysis time with a fixed value of current density.

Fig. 8 reveals the inverse relation of the current efficiency with the current density when the electrolysis time was taken at mean value. As explained in the previous interpretation of this response with the varying of the duration of the treatment, the ratio of (m_{exp}/m_{theo}) is irregular also due to the non-linearity behavior of the experimental consumption of the electrodes in comparison to the theoretical value that was evaluated directly from Eq. (4).

The generation of the oxide layer above the electrodes and the continuous releasing of gases bubbles which then accumulate and raise to the surface of the solution will affect the behavior of the experimental consumption; therefore, the current efficiency will minimize but all the values are larger than 100%, so the reactor is still effective under the restrictions of the operational variables in the present study. The experimental and theoretical consumption of the electrodes as well as the current efficiency are practically related to the current density according to the mathematical correlations shown in Eqs. (17), and (18).

In contrast, the ohmic drop is raised as the current density and the electric conductivity increased according to Eq. (4) in the case of a fixed value of the electrolysis time (Fig. 9). The individual ohmic drop seems to be larger in zone 1 due to the wider distance between its electrodes, but it increases more than the other zone as the current density increased which indicates that the electrical conductivity in the wider zone is minimizing in case of a higher current density. The two zones and the total ohmic drop are related to the current density according to the mathematical correlations [Eqs. (20)–(22)].

$$m_{exp.}\ (g) = 0.0398\ CD + 0.0262\ Mm \qquad (R^2 = 1.0000) \qquad (17)$$

$$m_{theo.}\ (g) = 0.0398\ CD + 8x10^{-05} \qquad (R^2 = 1.0000) \qquad (18)$$

$$\text{Current efficiency (CE\%)} = 169.4 - 11.95\ CD + 0.8153\ CD^2 \quad (R^2 = 0.983) \qquad (19)$$

$$\text{Ohmic drop (zone 1) (ohm)} = 0.2289 + 8.967CD + 0.05073CD^2\ (R^2 = 1.00) \qquad (20)$$

$$\text{Ohmic drop (zone 1) (ohm)} = 0.1617 + 6.834CD + 0.03893CD^2\ (R^2 = 1.00) \qquad (21)$$

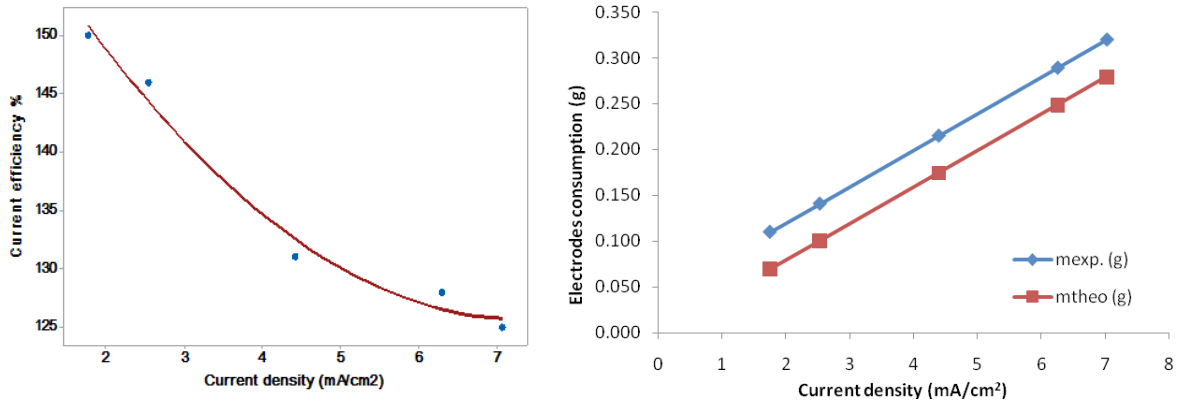
$$\text{Total ohmic drop (ohm)} = 0.3934 + 15.80CD - 0.08986CD^2 (R^2 = 1.00) \qquad (22)$$

Fig. 10 shows that the maximum value of the current efficiency is obtained at the lowest values of the current density and the total ohmic drop as a consequence according to the direct relation between the ohmic drop and the electric current density. The increment in the current density has increased the release of different ions from both electrodes which leads to increase the impact of raising the conductivity on the increment of the zone's ohmic drop. Meanwhile, the largest zone's surface area the higher ohmic drop due to the difference of the current distribution according to the value of the active surface area. The influence of the current density, in the present configuration, on the studied responses is clearly summarized in Fig. 11. As observed, the current efficiency has decreased from 157% at the lowest limit of current density, i.e. 2.53 mA/cm^2 to 114% at 7.02 mA/cm^2 of the current density along the duration of the experiment (25 min) due to the increase of the ohmic drop value as a result of increasing the electric conductivity from 114.77 to 119.50 mS/cm for the same period.

The experimentally results established a relationship between the current efficiency and current density and

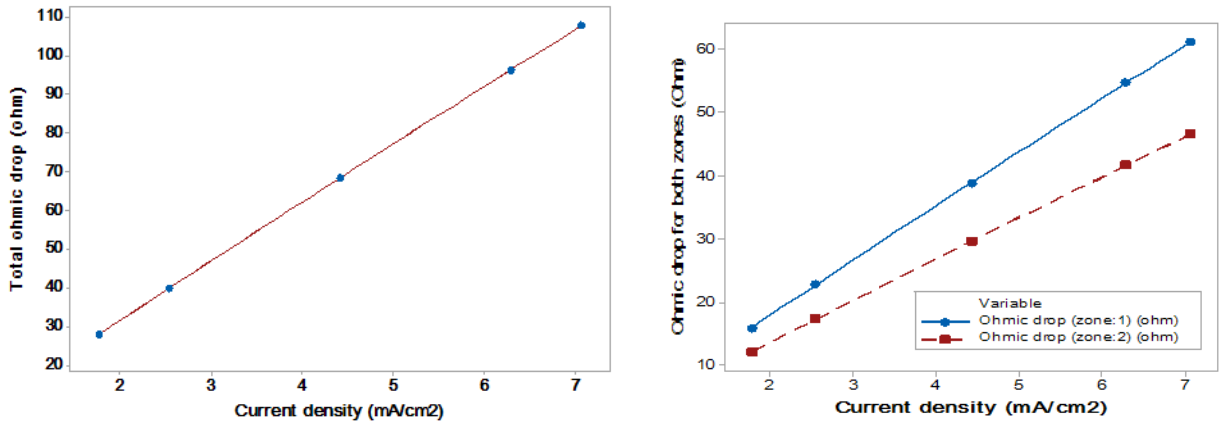
Table 6
Effect of current density on the studied responses

Run No.	Current density (mA/cm^2)	Final conductivity (mS/cm)	$m_{exp.}$ (g)	m_{theo} (g)	Current efficiency %	Ohmic drop (zone:1) (ohm)	Ohmic drop (zone:2) (ohm)	Total ohmic drop (ohm)
1	1.75	114.77	0.110	0.070	157	15.91	12.11	28.02
2	2.53	115.62	0.141	0.101	140	22.73	17.31	40.05
3	4.39	117.46	0.215	0.175	123	38.85	29.59	68.44
4	6.25	118.97	0.289	0.249	116	54.62	41.60	96.22
5	7.02	119.50	0.320	0.280	114	61.10	46.54	107.64



(a) (b)

Fig. 8. (a) Current efficiency vs. current density (electrolysis time = 25 min); (b) Experimental and theoretical electrodes consumption vs. current density (electrolysis time = 25 min).



(a) (b)

Fig. 9. (a) Total ohmic drop vs. current density (electrolysis time = 25 min); (b) Ohmic drop in both zones vs. current density (electrolysis time = 25 min).

have proved the inverse relation between the ohmic drop of both zones and the current density because the ohmic drop is increased as the current is raised. According to the previous results, the current efficiency is extremely affected by the configuration of the electrodes employed in the electrocoagulation reactor as well as the chemical reactions occurring via the treatment of the wastewater. Vogt [32] proved that the current efficiency is influenced by the anodic oxidation and the autoxidation of hypochlorite whereas linear axial increase of the hypochlorite bulk concentration in the electrochemical reactor was estimated according to the obtained mathematical model for the anodic current efficiency in chlorate electrosynthesis.

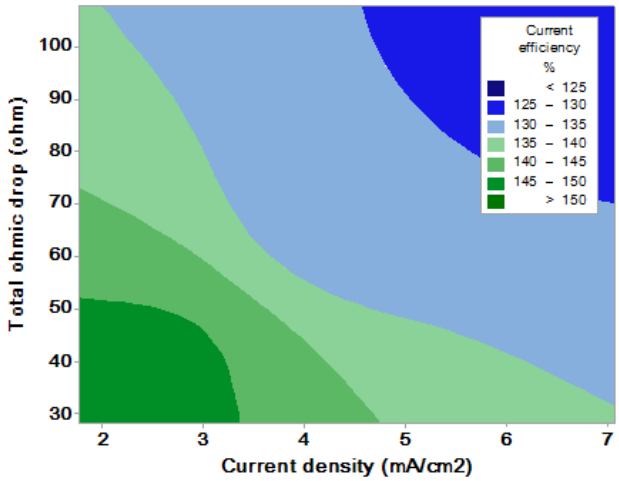


Fig. 10. Contour plot of current efficiency vs. total ohmic drop and current density.

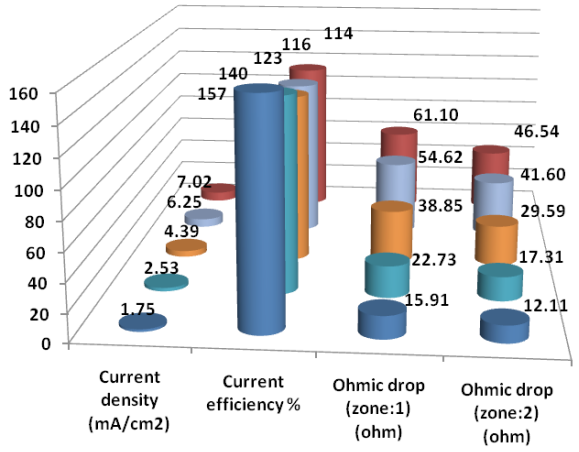


Fig. 11. Current efficiency vs. both zones of ohmic drop and current density.

Spasojevic et al. [31] developed mathematical models for anodic, cathodic and overall current efficiencies in an electrochemical hypochlorite production acting as a stirred reactor under the conditions of constant value of current density as 100 mA/cm^2 and at 8.3 < pH < 8.7 value. These models have developed depending on the release of hydrogen and oxygen from electrodes, i.e. the anodic oxidation and cathodic reduction of hypochlorite taking place as mass-transfer controlled reactions, where all findings of these efficiencies are less than 100% for all the values of the hypochlorite concentration. Weixing et al. [33] studied the current efficiency for the recovery of organics from aspartame effluents with electrolysis that was 69.9% only. While Mariagiorgia et al. [34] revealed that the current efficiency increases as the current density is raised until it reaches the limiting current density (LCD) that was 60% then tends to minimize. Mayanle and Masanori [35] have found that the current efficiency could be reached to 110% as the concentration of electrolyte is minimized by using pulse intervals technique. Volgin et al. [36], have proved the dependence of the electrodes design on the current efficiency.

The obtained results of the present configuration of the electrodes have provided an important addition for such types of electrochemical reactors because there was a lack of information about the relation between the current efficiency and the potential ohmic drop in one side and the electrocoagulation design on the other side. The present reactor had achieved high values of current efficiency for all values of contact time and current density regardless of their ranges.

4. Conclusions

The employment of an electrocoagulation reactor as a practical technique to treat wastewater is requiring a deep study in order to cover the idea of how it works in a complete manner. Therefore, the present study focused on the main effective parameter,i.e. the current efficiency, via the electrocoagulation of real oily wastewater using an innovated configuration of electrodes under the impact of the electrolysis time and the current density. The results reveal a significant effect of the invented electrodes configuration on the studied responses values. They had decreased from 150 to 125% and 67.85 to 66.19 ohm respectively as the electrolysis time increases from 10 to 40 min whereas the ohmic drop raises from 28.02 to 107.64 ohm as the current density increased from 1.75 to 7.02 mA/cm^2 but the current efficiency had minimized from 157 to 114% for the same increment in current density. The lower values of the operational variables the higher value of the current efficiency which is the most significant response. This study proved a notable efficiency of the present design of the electrocoagulation reactor.

Acknowledgments

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Symbols

CCRD — Central composite rotatable design
CD — Current density in (mA/cm^2)
d — Distance between electrodes (cm)
F — Faraday's constant (96485.34 columb/mol)
M — Molecular weight of electrodes metal in (g/mol)
$m_{exp.}$ — Experimental value of electrodes consumption
$m_{theo.}$ — Theoretical value of electrodes consumption
ODP — Ohmic drop (ohm)
t — Electrolysis time in (sec.)
w_1 — Weight of electrodes before the experiment
w_2 — Weight of electrodes after the experiment
Z — Number of electrons presented in the reaction
k — Solution conductivity (mS/cm)

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