Effect of current density on COD removal efficiency for wastewater using the electrocoagulation process

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ABSTRACT

The electrocoagulation process is an electrochemical technique that has demonstrated wide potential because it can be applied to wastewater from different industrial sectors considering the principles of electrolysis and coagulation. It is a low-cost method and of easy installation. Effluents with high concentrations of oils grease and chemical oxygen demand (COD) can be treated successfully obtaining high removal efficiencies. The purpose of this study was to analyze the effect of current density on the performance of oil/grease removal (OG), COD, and turbidity (NTU), as well as the pH variation and electrical conductivity during the process in batch mode. The effluent from the dairy industry with 172.6 mg OG/L, removed 70.30% OG, 75.38% COD with 76.92 A/m^2 in 25 min of electrolysis. Two combinations of 2Fe-1Al and 1Fe-2Al anodes were analyzed at 68.38 A/m^2, reaching removals of 74.74% and 70% of OG, respectively. Regarding the domestic kitchen effluent, the COD initial was 1,766 mg/L, and working with two Fe-anodes, the OG removal efficiencies in 15 min of electrolysis were 94.9%, 96.75%, and 96.53% with current densities of 37, 56, and 74 A/m^2, respectively. The OG removal efficiencies were similar with Al-anodes. The COD removal efficiencies with Fe-anodes were 66.7%, 76.9%, and 68.7%, and with Al-anodes were 76.3%, 77.4%, and 77.5%, respectively. Both effluents were studied with an inter-electrode distance of 3 cm, and the pH in both cases varied from 7 to 8, which favors the elimination of contaminants by adsorption. The electrical conductivity did not undergo major changes, favoring the economy of the electrocoagulation process. Electrocoagulation is a low-cost electrochemical process in order to remove organic pollutants.

Keywords: Electrocoagulation; Dairy industrial and domestic wastewater; Fe- and Al-anodes

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1. Introduction

Water is complex because it is linked to almost everything in the world. But complexity should not hinder understanding: Water is a precondition for human existence and for the sustainability of the planet [1]. Water pollution is mainly associated with inorganic, organic, biological, and radiological contamination [2]. Manufacturing and other industries use water during the production process for either creating their products or cooling equipment used in creating their products. According to the United States Geological Survey, industrial water is used for fabricating, processing, washing, diluting, cooling, or transporting a product. Water is also used by smelting facilities, petroleum refineries, and industries producing chemical products, food, and paper products [3]. Domestic water use is the water used for indoor and outdoor household purposes—all the things done at home: drinking, preparing food [3], bathing, washing clothes and dishes, brushing the teeth, watering the yard and garden, even washing the dog, etc. [4]. We are genetically programmed to consume fat as we can [5]. So the preparation of food requires oils and fats and many of them have it due to its natural composition (i.e., meats) or because they are industrially manufactured foods (i.e., pizzas). A wide variety of products containing oils and fats are used to prepare food in households, these include meat, deep-fried food, baked goods, different kinds of cheese, butter, etc. All this waste generated in the preparation of our food are considered fats, oil, and grease (FOG) [6]. FOG is usually produced at food service establishments or other food preparation facilities (FSE) [6,7]. During the washing of the utensils (dishes, etc.), FOG is incorporated into the water [8]. The U.S. Environmental Protection Agency has identified fats, oil, and grease as a pollutant of concern. They are listed alongside other pollutants such as: fertilizers, pesticides, and herbicides [9,10]. FOG can be solid or viscous liquid depending on the saturation of the carbon chain [6], FOG if left unchecked, may undergo reactions with other constituents in the wastewater to produce insoluble solids or FOG [7]. These properties confer the capacity of depositing on the internal walls of the sewer pipes [6,8]; which could lead to the blockage of pipes and consequently sanitary sewer overflows (SSOs) [7]. Its continuous elimination into the sewer system generates the decrease in the capacity of the pipes to transport the waste stream [6]. In the literature, we find several wastewater treatment technologies that can be used to remove contaminants. In each case, there are advantages and disadvantages. The electrocoagulation process (EC) has been suggested as a technique that can be applied to a wide variety of wastewater [11], for example, municipal sewage [12], oil–water emulsion [13], oily wastewater [14], petroleum refinery [15–17], organic material [18–21]; metals [22–24], oil and grease [25,26], clay suspension [27], etc. EC involves the generation of coagulants in situ [28,29] by dissolving electrically either Al- or Fe-ions from Al- or Fe-electrodes, respectively [29,30], that act as destabilizing agents and lead to neutralization of electric charge for removing pollutants [11,31]. The conductive metal plates are commonly known as 'sacrificial electrodes' and may be made of the same or different materials such as Fe [32] and Al/Fe-electrodes [33]. The metal ions generation takes place at the anode. Hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water. This process sometimes is called electroflocculation (EF) [29]. For the pollutant removal mechanism based on Fe/Al electrodes, you can consult the work done by [28,34–36]. EC has the advantages such as low sludge production, high process efficiency, no further effluent treatment [25], and potential for scalability [37,38]. Other additional factors that support its use are: minimal infrastructure requirements, no chemical handling requirement [39]; and, compared with the traditional coagulation/flocculation method, it is easy to operate and automate, and requires low capital and operating costs [35,40]. The objective of this study was to analyze the effect of current density on the removal efficiency of the different parameters of the electrocoagulation process, with two effluents containing oils and grease, using Fe- and Al-anodes.

2. Materials and methods

The two samples of wastewater were studied in reactors of the same characteristics. The electrolytic cell for the batch system (Fig. 1) was designed to treat 5 L of water, in the form of a parallelepiped whose internal dimensions are 0.13 m × 0.20 m × 0.25 m of transparent plexiglass material, whose thickness was 0.009 m. The separation between the anode and cathode was 0.03 m. The dimensions of the electrodes were 0.15 m × 0.133 m × 1/16″. The reactor has a ball valve for ½″ discharge.

2.1. Case one: Removal of oils and fats from an effluent from the dairy industry

Grupo Gloria is an industrial conglomerate of Peruvian investments with commercial presence throughout Peru. It has an important presence in Latin America. Its evaporated milk processing plant generates a high volume of wastewater, so other wastewater treatment options are of great interest in order to protect the environment. Fig. 2 shows the homogenizer tank from which the wastewater was obtained, and Table 1 shows the physicochemical characteristics of wastewater. The experiments were performed with commercial steel electrodes (Fe). A first set of experiments were performed with 3Fe (anodes, 0.117 m^2) and 2Fe (cathodes, 0.078 m^2). In the second group of experiments, an Fe-anode was removed and replaced by one of Al and in the third group of experiments two Fe-anodes were removed and replaced by two Al, in both cases of the same dimensions. The experiments were carried out without adjusting pH of raw wastewater at pH 7. The instruments used to measure the different parameters were a Hach HQ11D Portable pH Meter, a Hach HQ14D conductivity meter, a Hach 2100P portable turbidimeters, a Lovibond MD 200 COD photometer, a Boeco R300 1/8 HP Vacuum Pump, and a 0.5 HP CPM 600 centrifugal pump, Pedrollo brand to recirculate the effluent between 250 L tanks. The experiments were carried out in batch and continuous flow mode. But, in this case only the results in batch mode are presented.
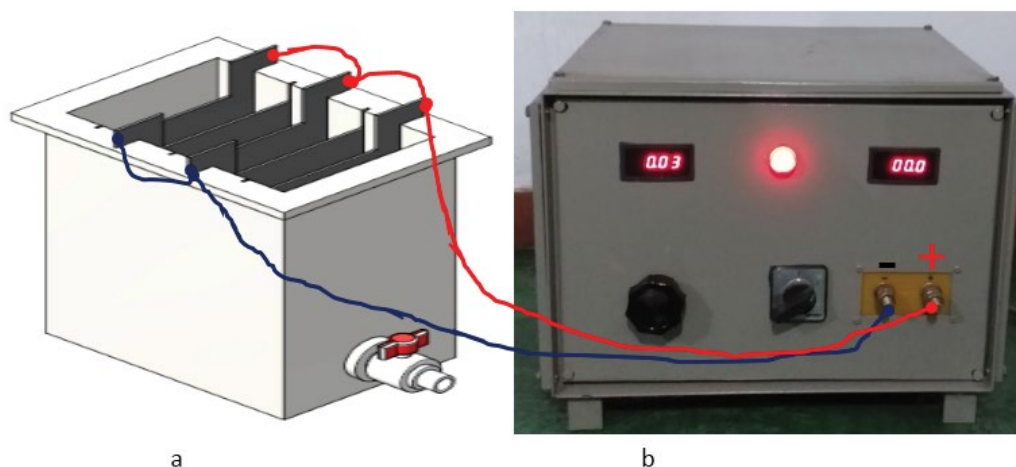
Fig. 1. (a) Reactor with three anodes and two cathodes and (b) power source, 100 A, 0–15 V.

Table 1
Physicochemical characteristics of wastewater

Parameter	Value
pH	7.07
Chemical oxygen demand (COD), mg/L	2,860
Total suspended solids (TSS), mg/L	383.3
Oils and grease (O/G), mg/L	172.7
Conductivity, µs/cm	1,384
Turbidity, NTU	2,274

Fig. 2. Homogenizer tank: Grupo Gloria plant.

2.2. Case two: Removal of oils and fats from a domestic kitchen effluent

Households' characteristics: At the urban level, 86.5% of Peruvian homes use drinking water to drink from the public network, whether inside or outside the house or public tap pylon [41]. The study sample is an effluent of domestic cooking, water that has been used mainly for washing meat, vegetables, and washing utensils. Since the food served for breakfast, lunch, and dinner are different, the characteristics of the wastewater vary during the day. The samples were collected from the drain trap of the washer on different days and times. In total 500 L of residual domestic cooking water was collected and this presented a gray coloration, with the presence of coarse solids, which were separated with a strainer. The waste water was placed in two plastic cylinders with an individual capacity of 250 L, and in order to homogenize the wastewater, a pump was used to circulate it between them (Fig. 3).

Table 2 shows the physicochemical characteristics of domestic kitchen wastewater. The first set of experiments was performed with commercial steel electrodes. Two (Fe-Fe) anodes and three (Fe-Fe-Fe) cathodes were used. The areas of each set of electrodes were 0.078 and 0.117 m^2, respectively. A second group of experiments were carried out by removing an Fe-anode and replacing it with an Al-anode, maintaining the three Fe-cathodes. The sludge generated in the process was removed from the solution following the
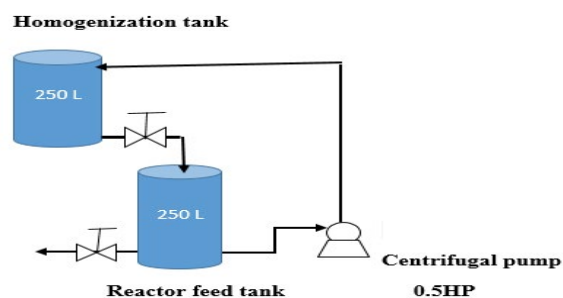

Fig. 3. Effluent homogenization system.

Table 2
Physicochemical characteristics from domestic kitchen wastewater

Parameter	Value
pH	6.8
Chemical oxygen demand (COD), mg/L	1,766
Total suspended solids (TSS), mg/L	141
Oil and grease (O and G), mg/L	43.1
Conductivity, µS/cm	1,286
Turbidity, NTU	386

procedure explained in the previous case. The instruments used for the measurements of the different parameters were a Eurolab Germany pH-meter 01, a Hach HQ14D conductivity meter, Orion AQ4500 turbidimeter, standard test method for oils and fats, and hydrocarbons in water (ASTMD3921), and a Pfeiffer vacuum model DUO 3 rotary vane pump.

2.3. Mathematical analysis

The removal efficiency (RE%) was calculated as follows:

$$RE(\%) = \frac{C_0 - C_t}{C_0} \times 100 \tag{1}$$

where C_0 is the initial concentration of the influent (before the EC process) and C_t is the concentration of the effluent (after the EC process). Theoretical mass (Δm_t, $\varnothing_c = 1$) of dissolved electrode was calculated as follows:

$$\Delta m_t = \frac{M}{zF} I \cdot t \tag{2}$$

where M is the molar mass of the substance in grams (g/mol), I is the amperage (A), z are electrons transferred per ion, F is the Faraday's constant (96,500 C/mol), and t is the total time the constant current was applied. The experimental mass (Δm_e), was calculated as follows:

$$\Delta m_e = m_i - m_f \tag{3}$$

where m_i and m_f are the masses of the electrode before and after the EC process. The theoretical (DR_t) and experimental (DR_e) dissolution rate of the electrodes in mg/C can be calculated as follows.

$$DR_t = \frac{M}{zF} \tag{4}$$

$$DR_e = \frac{1,000 \Delta m}{It} \tag{5}$$

where $M_{Fe} = 55.847$ g/mol and $M_{Al} = 26.981$ g/mol, and Δm is Fe or Al dissolved during the EC process in grams. The efficiency of the current $\varnothing_c$ (%) was calculated as follows.

$$\varnothing_c = \frac{\Delta m_e}{\Delta m_t} \times 10 \tag{6}$$

The energy consumption (CE) in kWh was evaluated as follows [39,42,43]:

$$CE = \frac{V \cdot I \cdot t}{3.6 \times 10^6} \tag{7}$$

where V is the potential difference (V). The energy consumption in kWh/m³ of treated water was evaluated as follows:

$$CEV = \frac{CE}{V_0} \tag{8}$$

where V_0 is the volume of solution (m³).

3. Results and discussion

3.1. Case one: Dairy industrial wastewater

In each figure, the three groups of experiments are shown. In solid black lines the removal profiles of the different parameters of the first group of experiments are shown at 59.83, 68.38, and 76.92 A/m². The second and third group of experiments were only performed at the average current density of 68.38 A/m², because there is not a large difference in removal with the maximum current density used, and they are shown in dotted and discontinuous lines in pink and red, respectively. Fig. 4 shows the effect of current density on the removal performance of oils and fats with three current densities. During the first 15 min using only Fe-anodes, the oils and greases are removed at constant speeds of 0.92%/min (59.83 A/m²), 1.31%/min (68.38 A/m²), and 1.61%/min (76.92 A/m²). After this time, the removal process is accelerated reaching values of 58.25%, 65.68%, and 70.30%, respectively, in 25 min. Other studies report 94.5% removal with Fe-electrodes at pH 7 with 30 V and with pH greater than 9 the removal efficiency decreases [44]. The electrochemically generated metallic ions are good coagulants [45], and the current density is the most sensitive operating parameter of electrocoagulation process [46].

The chemical reactions that take place at the anode in an alkaline medium [29,45,47,48] (Fig. 8) are as follows:
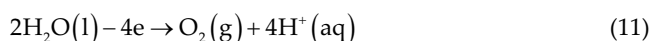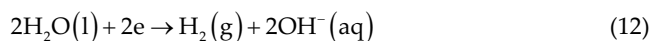
For the Fe–anode:

$$Fe(s) - 2e \rightarrow Fe^{2+}(aq) \tag{9}$$

At alkaline conditions:

$$Fe^{2+}(aq) + 2OH^-(aq) \rightarrow Fe(OH)_2(s) \tag{10}$$

In addition, there is an oxygen evolution reaction:

$$2H_2O(l) - 4e \rightarrow O_2(g) + 4H^+(aq) \tag{11}$$

The reaction at the cathode is:

$$2H_2O(l) + 2e \rightarrow H_2(g) + 2OH^-(aq) \tag{12}$$

The Fe^{2+} cation can hydrolyze near the anodes to produce a series of activated intermediates that are able to destabilize the finely dispersed particles present in the wastewater. The destabilized particles then aggregate to form flocs. At the same time, hydrogen bubbles produced at the cathode can float most of the flocs. Thus, the aggregates formed can be removed by decantation or flotation from wastewater [45,49]. Different complexes of Fe-hydroxides can be formed by polymerization ($Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^+$, $Fe_2(H_2O)_8(OH)_2^{4+}$, $Fe_2(H_2O)_6(OH)_2^{4+}$, $Fe(OH)_4^-$, etc.), giving rise to a gelatinous suspension that has the ability to remove pollutants from wastewater by complexation or by electrostatic attraction [25,28,50]. EC treatment is particularly effective for the destabilization of oil-in-water emulsions [51]. Oil droplets in oil-in-water emulsion exhibit net charge at the droplet surface. The Fe-ions produced
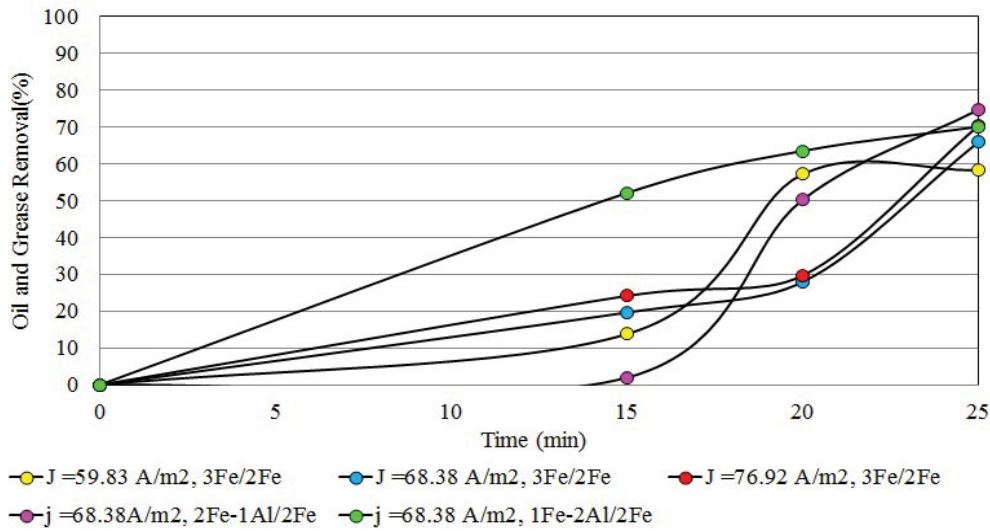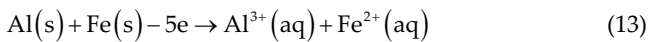
Fig. 4. Effect of current density on the removal of oils and fats.

by electrooxidation of the anode can form monomeric and polymeric hydroxides with a positive charge, which have great affinity for counter ions (water contaminants) and can cause coagulation [15,52,53]. Regarding the experiments with combined Fe-Al anodes at 68.38 A/m², it was observed that the removal efficiency is very low with (2Fe-1Al) anodes in the first 15 min. Finally, with this combination, a 74.64% of oil and grease removal was reached in 25 min, which is greater by 8.75% to the process with only Fe-anodes at the same current density, and higher by 4.34% with respect to the maximum applied current density (76.92 A/m²) using only Fe-anodes. With the combination of (1Fe-2Al) anodes, the speed of removal is better than with the other anode combinations. This is probably due to a greater amount of coagulant species generated in-situ. The removal speed in 25 min was 70.30%, equal to that reached at 76.92 A/m² with only Fe-anodes. The reactions that are taking place [45] are shown below:
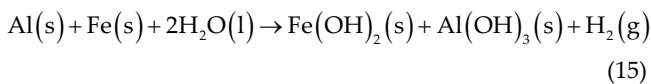
Anodic reactions: Eq. (11)

$$Al(s) + Fe(s) - 5e \rightarrow Al^{3+}(aq) + Fe^{2+}(aq) \tag{13}$$

Cathodic reaction: Eq. (12)
Solution reactions: (10)

$$Al^{3+}(aq) + 3OH^-(aq) \rightarrow Al(OH)_3(s) \tag{14}$$

Overall reaction:

$$Al(s) + Fe(s) + 2H_2O(l) \rightarrow Fe(OH)_2(s) + Al(OH)_3(s) + H_2(g) \tag{15}$$

According to the pH of the medium, Al^{3+} and OH^- give rise to various hydroxylated cations forms. In acidic medium, the predominance of the free Al^{3+} cation is observed until pH 3.5. As the pH increases, different hydroxo-complexes including monomers and polymers take part in the distribution such as: $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al_2(OH)_2^{4+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$,

$Al_8(OH)_{20}^{7+}$, $Al_{13}O_4(OH)_{24}^{7+}$, and $Al_{13}(OH)_{34}^{5+}$ [36,54,55]. The processes of coagulation with AL achieve significant removal of organic matter [55].

Fig. 5 shows the efficiency of removal of COD. The dairy industry is one of the largest sources of industrial effluents, characterized by high COD and biological oxygen demand (BOD) content [54,56].

The current density controls the speed of dissolution of the anode (anodic process) as well as the speed of hydrogen production (cathodic process). The influence of the variation of this parameter (between 59.83 and 76.92 A/m²) was examined on the efficient removal of COD. It is important to bear in mind that the higher the density of the current, the smaller the bubble size [57]. Therefore, there is an increase in the contact area between the gas (H_2) and the pollutants and the speed of removal of the contaminants is favored, and the efficiency of the flotation increases [58]. With Fe-electrodes, the maximum removal efficiency reached in 25 min was 75.38% with 76.92 A/m². At the same time, there is only a difference of 3.18% removal efficiency with 68.38 A/m². The removal efficiency with combined (2 Fe-1Al) anodes was 70.80% and with (1Fe-2Al) anodes of 65.87%, in both cases at 68 A/m². The removal efficiency with the same current density using only Fe-anodes was 72.20%. Other studies report a removal of 86.91% COD working with Al-electrodes at pH 9, and 5.04% removal at pH 3 [56], and 60%–80% COD removal [59]. A COD removal of 69.4%, 78.9%, and 80.4% was obtained with Fe-electrodes [44], 45.14% and 90.12% COD removal [60], and 67.27% COD removal [61]. The physicochemical processes have the disadvantage of using an expensive reagent while the soluble COD removal is low [56,62].

Fig. 6 shows the effect of current density on the removal of turbidity (NTU). The removal levels in 25 min with Fe-electrodes at 68.38 and 76.92 A/m² were 82.95% and 85.66%, respectively. The removal levels with combined (2Fe-1Al) and (1Fe-2Al) electrodes at 68.38 A/m² in 25 min were 83.64% and 81.31%, respectively. With the second anode combination, the removal rate is higher up to 20 min compared with the other anode arrangements. This is due to the higher production
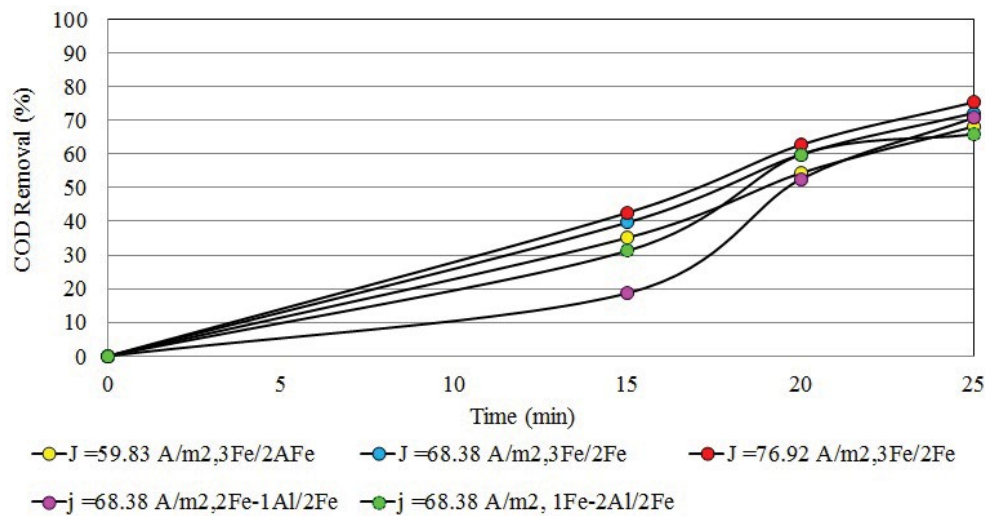
Fig. 5. Effect of current density on the removal of chemical oxygen demand (COD).
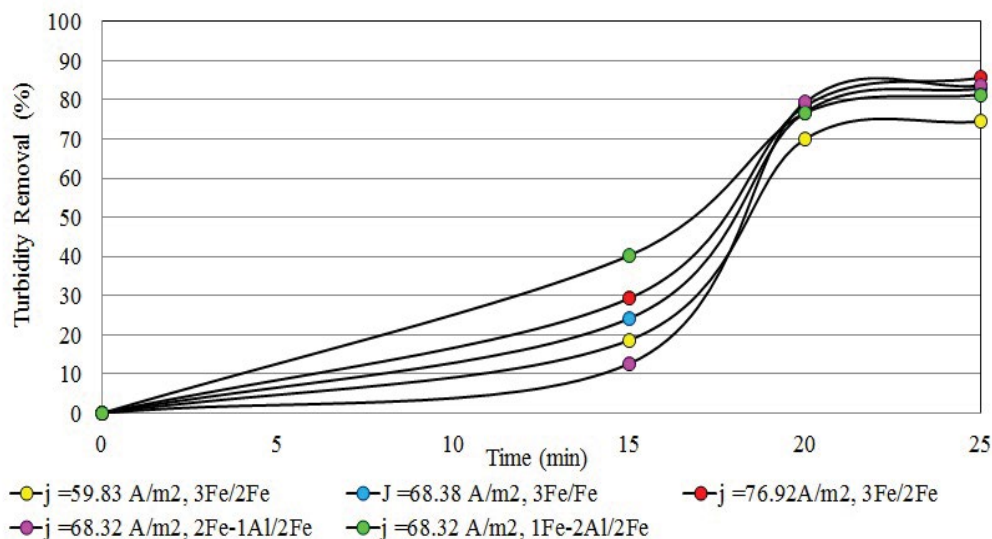

Fig. 6. Effect of current density on turbidity removal (NTU).

of coagulant species due to Fe and Al hydrolysis explained above. Other studies report NTU removal levels of 93.46%, 94%, and 86.7% with Al-anodes at pH of 5, 6, and 7 with 3 V for 10 min of electrolysis [63], and 95% with Fe-anodes at 50 A/m² at a pH 7 [64]. It is important to keep in mind that the pH has a significant effect on the performance of the EC process [53]. However, very poor removals are found either at low pH (<2.0) or high pH (>10). This behavior was attributed to the amphoteric character of $Al(OH)_3$ that does not precipitate at pH less than 2.0. However, high pH will increase $Al(OH)_3$ solubility and lead to the formation of soluble $Al(OH)_4^-$ which is useless for water treatment [65].

Fig. 7 shows the effect of current density on the removal of total suspended solids (TSS). With only Fe-electrodes in 25 min of electrolysis, the TSS removal levels with 76.92 and 68.32 A/m² were 80.76% and 76.88%, respectively. The removal

profiles with combined (1Fe-2Al) anodes at 68.32 A/m² and only Fe-anodes at 76.92 A/m² are very similar. At the end of the process, there is only a difference of 1.92% in favor of the Fe-electrodes. The presence of Al-anodes reduces energy consumption due to the greater formation of coagulant species due to the presence of the Al-anode. The combination of (2Fe-1 Al) anodes in 25 min reached 81.47% removal, surpassing by 4.59% the process with Fe-anodes with the same current density. Other studies show that the removal levels achieved with Al-anodes were 44.18%, 54.23%, 58.08%, and 63.5% with voltages of 5, 10, 15, and 20 V, pH 8 in 50 min of electrolysis [66]. It is well known that electrical potential not only determines the coagulant dosage rate but also the bubble production rate and size, and the flocs growth which can influence the treatment efficiency of the electrocoagulation process [54,67,68].

Fig. 8 shows the effect of the current density on the pH. The pH changes experienced by the solution are 0.48, 0.73, and 0.94 with 59.83, 68.38, and 76.92 A/m^2, respectively. The EC process starts in a slightly alkaline medium and reaches a maximum value of 8.01 with 76.92 A/m^2. The increase in pH can be explained by taking into account the production of the hydroxyl ion at the cathode (Eq. (12)) which finally forms Fe-hydroxide (Eq. (10)). The pH is influenced by the cathodic process. If the current density increases, the pH also increases. Other studies also indicate this trend [69,70], and affect the performance of the process [71,72] as discussed above.

The pH–time profiles are not presented with combined anodes, because the profiles overlap and the pH is kept below 7.80 with the two combinations of anodes. Other studies show that the pH stabilized below 9 starting with a value close to 8.5 with Al-anodes [59]. The linear pH–time trends for the three current densities are (pH = 7.1289 + 0.0389t,

$R^2 = 0.9941$, 76.92 A/m^2; pH = 7.08 + 0.029t, $R^2 = 0.9941$, 68.38 A/m^2; pH = 7.0536 + 0.0194t, $R^2 = 0.9693$, 59.83 A/m^2). The best linear trend is given to the average current density. The effects of pH of water or wastewater on electrocoagulation are reflected by the current efficiency as well as the solubility of metal hydroxides [48]. The pH affects the solubility of Fe (OH)$_3$ and Al (OH)$_3$. The range of pH 6 to 7 is ideal for both types of hydroxides [44,73].

Fig. 9 shows the profiles of how the temperature increases during the electrolysis process. Temperature is always considered an important parameter in any chemical or electrochemical separation process. It is expected that the higher the current density, the greater the change. In 25 min, the temperature changes were 3.7°C, 5.8°C, and 7.7°C with 59.83, 68.38, and 76.92 A/m^2, respectively. After 15 min, the greatest changes in temperature occurred, probably due to the adherence of floccules on the electrodes. The temperature variation profiles are not presented with the combined electrodes.
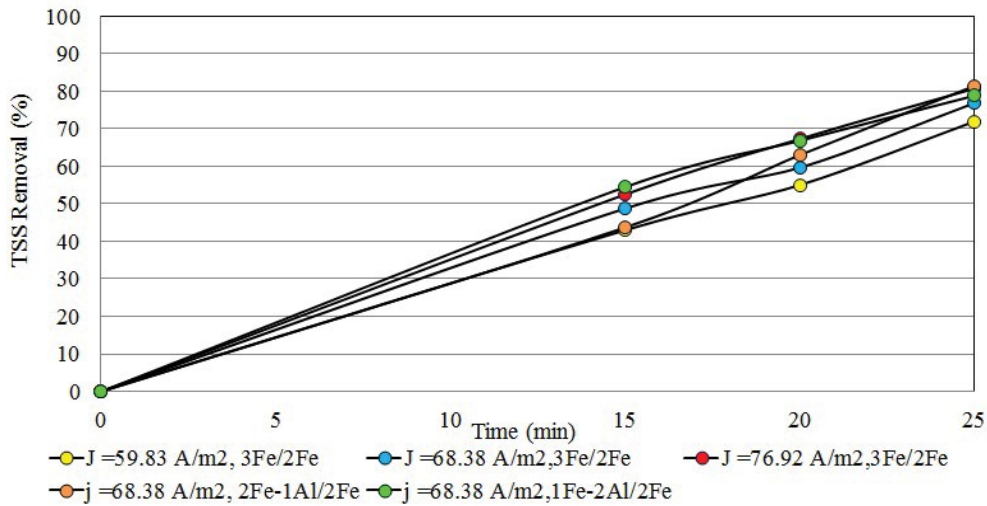
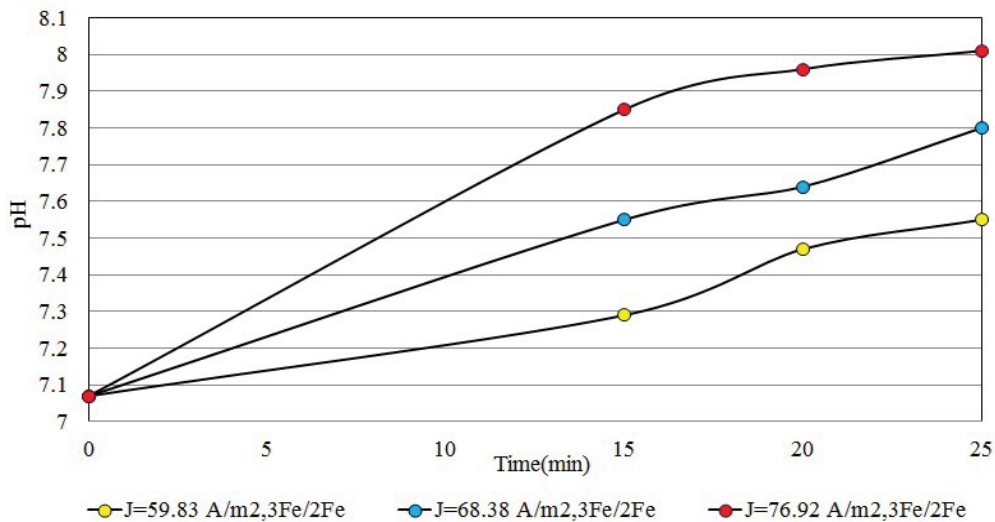Fig. 7. Effect of current density on the removal of total suspended solids (TSS).

Fig. 8. Effect of current density on pH.

The profiles overlap. Other studies show a linear variation of the temperature with electrolysis time at low current density (99 A/m^2) [74]. At higher temperature, the increase in solubility of precipitates of Al(OH)$_3$ and generation of unstable flocs can be observed. Consequently these may have adverse effects on the efficiency of the process [74]. Therefore, the Al precipitation is enhanced at lower temperatures, which results in a better removal [75].

Fig. 10 shows the effect of current density on the electrical conductivity. With Fe-electrodes, the percentages of decrease in conductivity in 25 min were 10.26%, 10.48%, and 12.36% with 59.83, 68.32, and 76.92 A/m^2, respectively. With the combined (2Fe-1Al) and (1Fe-2Al) anodes for the same time and average current density, the percentages of decrease were 10.98% and 13.37%, respectively. The electrical conductivity determines the conduction capacity of electrical current in an electrochemical process, and is related to the presence of ions in the solution. High ion concentration generates high conductivity and low resistance to the passage of electric current, which favors electrolysis. When the electrolytic conductivity is low, the current efficiency will decrease. And, high-applied bias potential is needed which will lead to the passivation of electrode and increase treatment cost [76]. Generally, NaCl is added in order to increase the electrolytic conductivity [76,77]. In our case it was not necessary to add NaCl.

Table 3 shows the energy consumption for each cubic meter of treated wastewater, as well as for each kg O-G and kg COD removed. The best levels of oil-grease and COD removal were obtained at 68.32 A/m^2.

3.2. Case two: Removal of oils and fats from a kitchen domestic effluent

Next we present the first group of graphs of the different parameters of the EC process with Fe-anodes. Fig. 11 shows
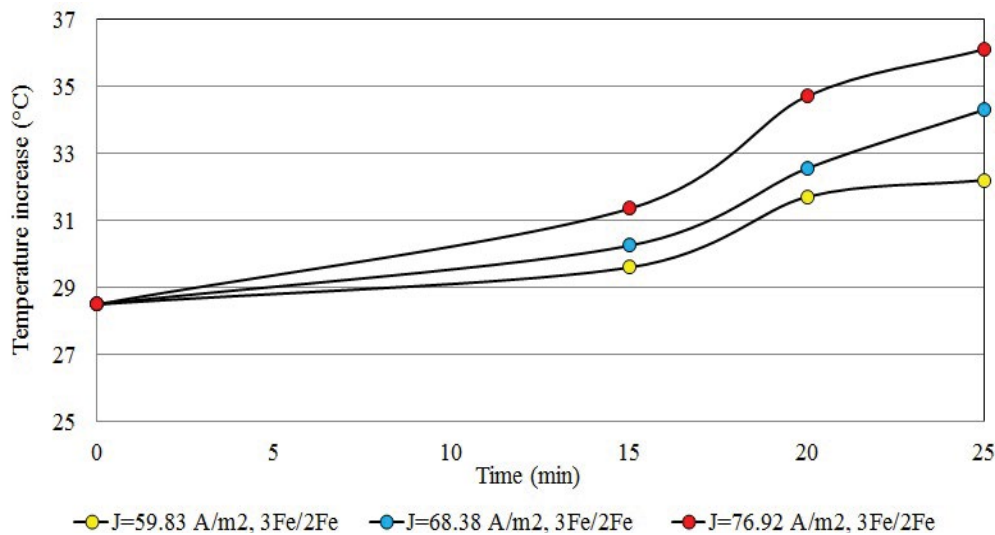
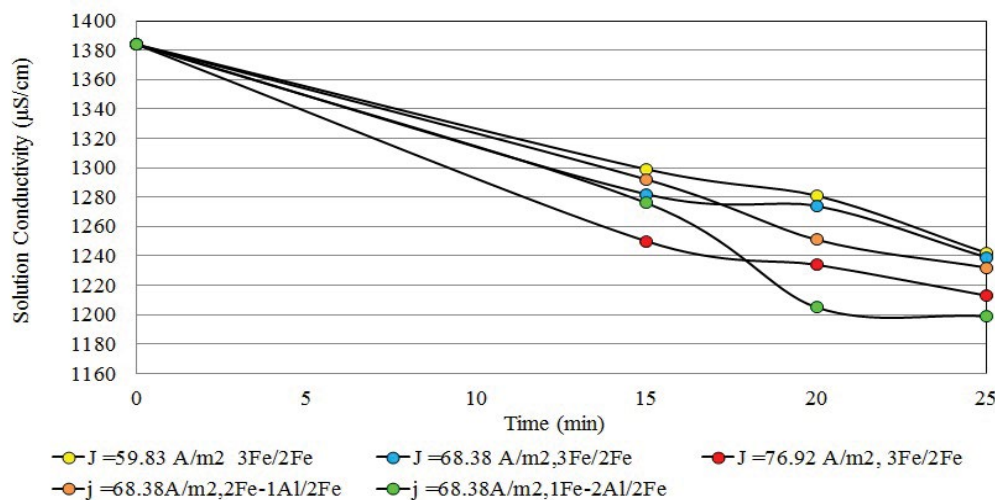Fig. 9. Effect of current density on the temperature increase of the solution.

Fig. 10. Effect of current density on the electrical conductivity.

Table 3
Unit energy consumption in batch mode: kWh/m^3, kWh/kg O-G and kWh/kg COD

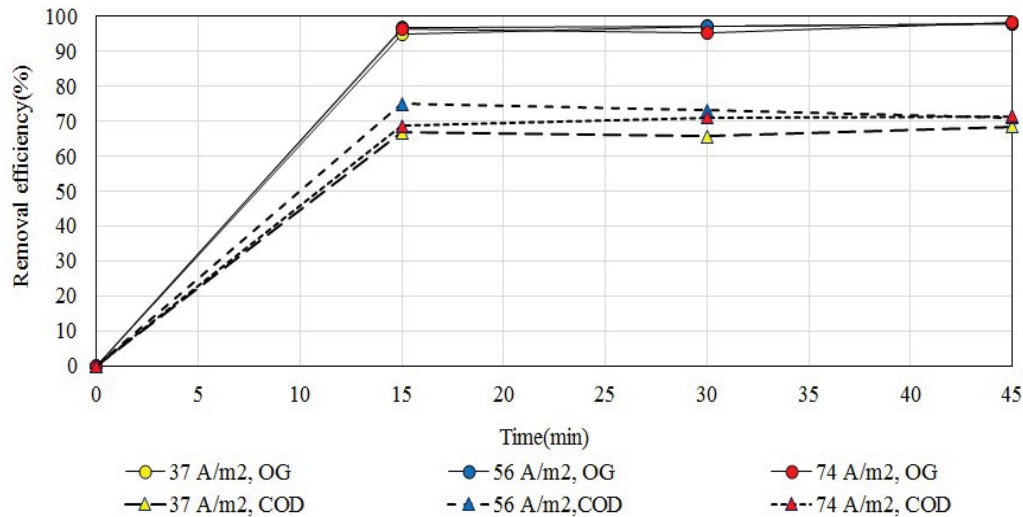| N run	I (A)	j (A/m^2)	t (min)	Voltage (V)	CE (kWh)	CEv (kWh /m^3)	Oil–grease (mg/L)	CE (kWh)/ (kg O-G)	COD (mg/L)	CE (kWh)/ (kg COD)
3	7	59.83	25	1.3	0.004	0.758	100.6	7.538	1,950	0.389
3	8	68.38	25	1.5	0.005	1.000	113.8	8.787	2,065	0.484
3	9	76.92	25	1.6	0.006	1.200	121.4	9.885	2,156	0.557

Fig. 11. Effect of current density on the removal of OG and COD.

the effect of current density on the removal of oils and grease (OG), as well as COD. The profiles of the removal efficiencies are given at three current densities. The removal efficiencies of OG are very high. At 15 and 45 min of electrolysis, they were (94.9% and 97.91%), (96.75% and 97.68%), and (96.52% and 98.14%), with 37, 56, and 74 A/m^2, respectively. The removal efficiencies of the COD in the first 15 and 40 min were (66.72% and 68.42%), (74.95% and 71.04%), and (68.69% and 71.31%), working with 37, 56, and 74 A/m^2, respectively. Both parameters are excellently removed in 15 min. Other studies show a removal efficiency range of 60% to 70% [78,79], and a 75% removal efficiency in 15 min [80].

Fig. 12 shows the effect of current density on the removal of turbidity (NTU) and total suspended solids (TSS). The removal efficiencies with 15 and 40 min of electrolysis were (70.23% and 91.71%), (75.34% and 98.68%), and (97.56% and 97.77%), with 37, 54, and 74 A/m^2, respectively. Using Fe-electrodes with voltages of 5, 10, and 15 V, the removal performance was maintained in a range of 82%–88% [81]. The removal efficiencies of TSS in 15 and 40 min of electrolysis were (78.01%, 95.74%); (68.79%, 94.33%), and (93.62%, 92.91%) with 37, 56, and 74 A/m^2, respectively. In the studies carried out by Phalakornkule et al. [82] and Kurt et al. [83], in both cases a removal of approximately 90% was achieved.

Fig. 13 shows the behavior of pH and electrical conductivity at different current densities. The pH of the solution increases continuously until 30 min, reaching values of (7.60, 37 A/m^2, 7.90, 56 A/m^2, and 8.2, 74 A/m^2), and at 45 min it is stabilized at 7.30, 7.90, and 7.90 with the densities of currents mentioned above. The net change in pH in all three cases is approximately 1.

Other studies also indicate that the pH of the effluent changes slightly when the pH value is in the neutral range (around 6–8) [77]. The effluent pH after electrocoagulation treatment would increase for acidic influent but decrease for alkaline influent. The increase of pH at acidic condition was attributed to hydrogen evolution at cathodes, reaction (Eq. (12)) [48,84]. In fact, besides hydrogen evolution, the formation of (Fe(OH)$_3$ or Al(OH)$_3$) hydroxides near the anode would release H$^+$ leading to decrease of pH. When pH is between 4 and 9, the Al^{3+} and OH$^-$ ions generated by the electrodes react to form various monomeric species such as Al(OH)$_2^+$, Al(OH)$_2^{2+}$, and polymeric species such as Al$_6$(OH)$_{15}^{3+}$, Al$_7$(OH)$_{17}^{4+}$, and Al$_{13}$(OH)$_{34}^{5+}$, that finally transform into insoluble amorphous Al(OH)$_3$(s) through complex polymerization/ precipitation kinetics [85]. When the pH is less than 4, the main mechanism by which pollutants are removed is precipitation, and at pH between 4 and 9, the removal process is by adsorption. The initial pH in this case was 6.8 and it was always below 8. This pH range favors the formation of Al(OH)$_{3(s)}$ [86]. With Fe-electrodes, EC efficiency is effective in alkaline conditions, this is due to the formation of Fe(OH)$_3$(s) [87,88] (Eq. (15)), which is more effective in the process sweep flocculation than Fe(OH)$_2$(s) formed in acid medium (Eq. (14)) [89,90]. In addition, there is also oxygen evolution reaction leading to pH decrease (Eq. (11)). The hydrogen bubbles produced at the cathode are smallest and
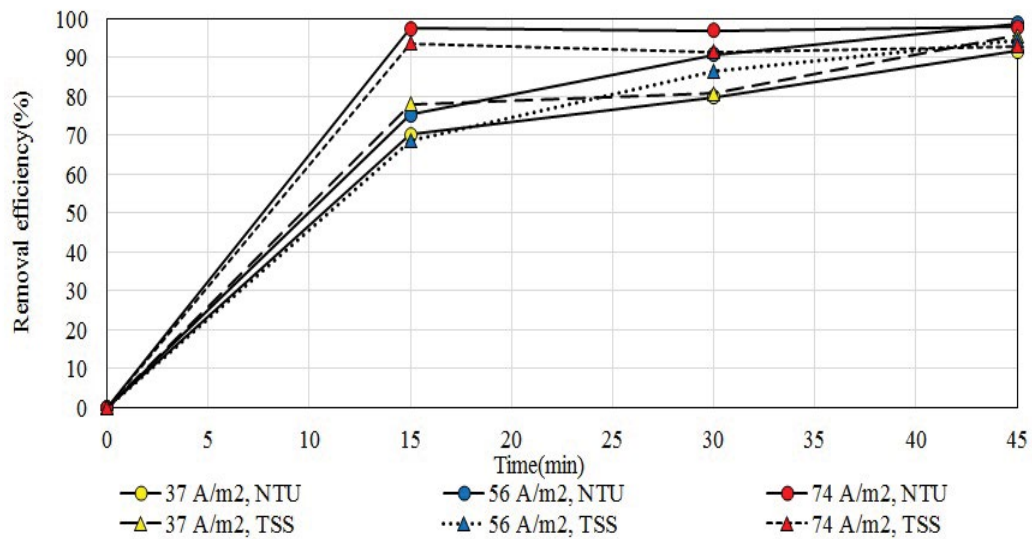
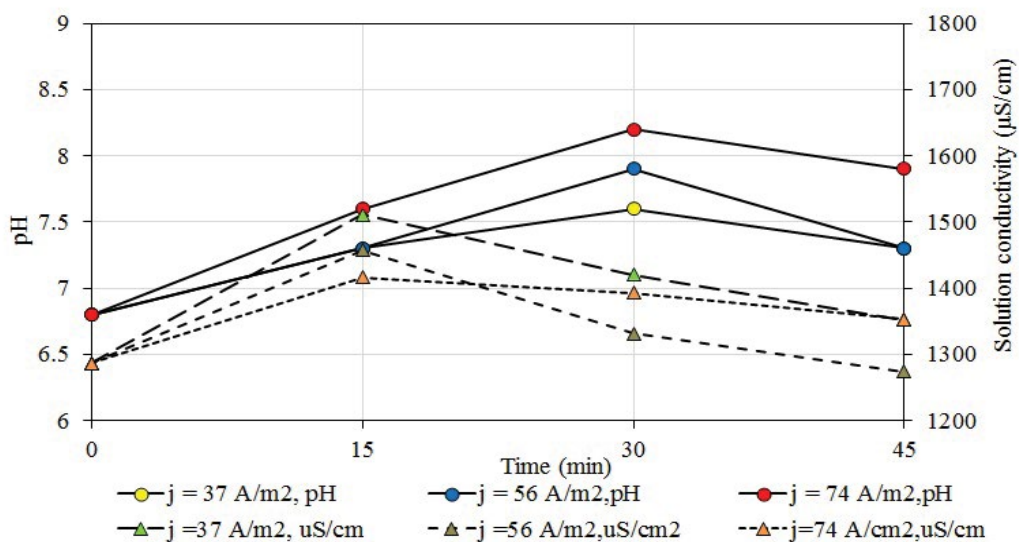Fig. 12. Effect of current density on the removal of NTU and TSS.

Fig. 13. Effect of current density on pH and electrical conductivity.

finest at neutral pH, providing sufficient surface area for gas–liquid–solid interfaces and mixing efficiency to favor the aggregation of tiny destabilized particles and colloids [77]. When the medium does not have adequate electrical conductivity, part of the electrical energy is transformed into heat making the process inefficient and increasing the cost of the process. It is well known that pollutant removal efficiency is improved when conductivity is higher [46]. In this case, it was not necessary to use NaCl to improve the electrical conductivity. During the first 15 min, the conductivity increased (+225 µS/cm, 37 A/m², +171 µS/cm, 56 A/m² and +200 µS/cm, 74 A/m²) probably due to the different Fe-species formed that have not had the possibility of interacting and forming clots. When it takes more time from 15 to 45 min, the conductivity probably decreases due to a greater interaction of the coagulating particles. The initial conductivity (1,286 µS/cm) has

not undergone abrupt changes, which translates into a good level of removal of contaminants. The net change in conductivity for each current density was (+66, –12, and +67 µS/cm).

Fig. 14 shows the effect of the current density on the temperature of the solution. The increase in temperature depends on the applied current density and the electrolysis time.

The increase in temperature over time has a linear dependence for each current density. Linear relationships are also reported in studies conducted by Reátegui et al. [15] and Olanipekun et al. [74]. The electrochemical reaction rate similar to any other chemical reaction rates increases when the temperature of the solution increases. The reason could be due to increase in mobility and collision of ions with the hydroxide polymer [91]. In these processes, the increase in temperature does not favor the removal of contaminants.
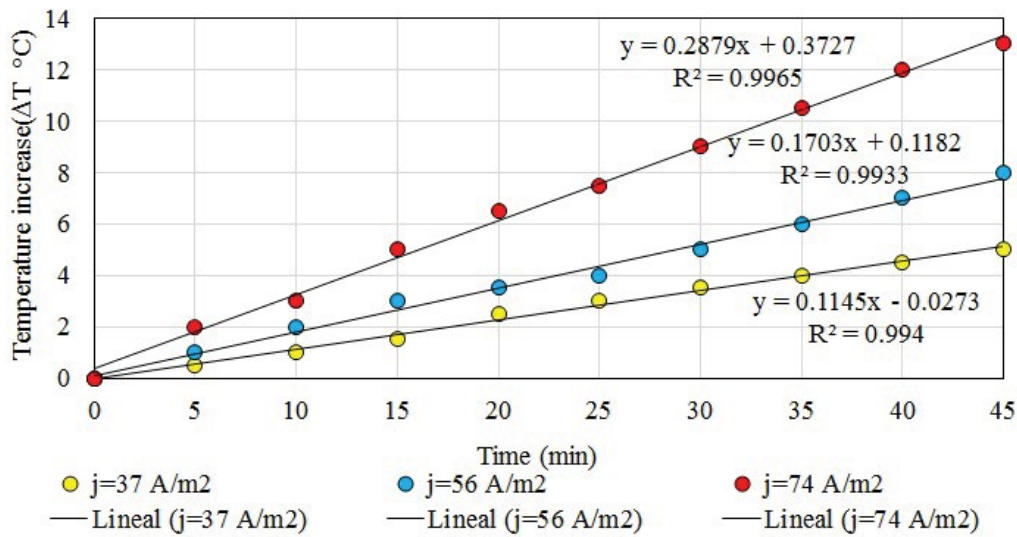
Fig. 14. Effect of current density on the increase of temperature in the solution.

The solubility of the Fe- or Al-compounds is increased making the process less efficient.

Table 4 is a summary of the EC process in batch mode. It shows the theoretical and experimental consumption of the Fe-anodes, as well as the experimental dissolution rate of the anodes and current efficiency that in some cases is greater than 100% by chemical reactions that additionally affect the anodes. Table 5 summarizes the energy consumption of the EC process and also shows the energy consumption per kg of dissolved material, calculated by

$CE_m = 1,000\ CE/\Delta m_e$, kWh/kg, and the operating cost (OC) in US\$/m^3 of water treated using Fe-electrodes. The results with Fe-electrodes show that in 15 min of electrolysis, the best responses are obtained for each parameter analyzed above. Taking this as a basis, a set of experiments with Al-electrodes was performed considering this time at the same current densities. The comparative figures in batch mode with Fe- and Al-anodes are shown below.

Fig. 15 shows the influence of the current density on the removal efficiency of OG and COD. In both cases, the removal

Table 4
Mass consumption of Fe-anodes in batch mode

I (A)	j (A/m²)	t (min)	Anodes	Anodes mass (g)			Δm_e	Δm_t	DR_e (mg/C)	$\varnothing_c$ (%)
				After EC	Before EC	Δm				
3		15	Anode 1	202.49	202.1	0.39	0.77	0.78	0.285	98.5
			Anode 2	199.35	198.97	0.38				
	37	30	Anode 1	168.15	167.37	0.78	1.46	1.56	0.270	93.4
			Anode 2	172.56	171.88	0.68				
		45	Anode 1	171.5	170.29	1.21	2.40	2.34	0.296	102
			Anode 2	175.75	174.56	1.19				
4.5		15	Anode 1	167.29	166.74	0.55	1.10	1.17	0.272	93.8
			Anode 2	171.78	171.23	0.55				
	56	30	Anode 1	171.07	169.96	1.11	2.28	2.34	0.281	97.3
			Anode 2	166.54	165.37	1.17				
		45	Anode 1	174.49	172.63	1.86	3.66	3.51	0.301	104.1
			Anode 2	169.29	167.49	1.80				
6		15	Anode 1	220.98	220.2	0.78	1.53	1.56	0.283	97.9
			Anode 2	201.92	201.17	0.75				
	74	30	Anode 1	220.11	218.36	1.75	3.25	3.12	0.301	104.0
			Anode 2	201.09	199.59	1.50				
		45	Anode 1	201.97	199.71	2.26	4.58	4.68	0.283	97.7
			Anode 2	205.2	202.88	2.32				

Table 5
Energy consumption in batch mode

N° run	I (A)	j (A/m²)	t (min)	Voltage (V)	CE (kWh)	CE$_v$ (kWh/m³)	CE$_m$ (kWh/kg)	OC (US$/m³)
1			15	8.8	0.007	1.3	8.5	0.339
2	3	37	30	9.0	0.013	2.7	9.2	0.670
3			45	9.1	0.020	4.1	8.5	1.054
4			15	12.7	0.014	2.9	13.0	0.624
5	4.5	56	30	12.9	0.029	5.8	12.7	1.278
6			45	12.7	0.043	8.6	11.7	1.947
7			15	16.3	0.024	4.9	16.0	0.997
8	6	74	30	16.2	0.049	9.7	15.0	2.025
9			45	16.2	0.073	14.6	15.9	2.972

efficiency is higher with Al-electrodes. The removal of OG is indifferent to the electrode material. In both cases, the removal is greater than 94% with pH that varies in a range of 6.8–8, with the three current densities experienced. The pH is a very important parameter that influences the performance of the EC process [86,92]. The removal of OG with Al was superior with respect to Fe in (+1.85%, 37 A/m², +0.23%, 56 A/m², and 0.93%, 74 A/m²). Both electrodes with the current densities excellently worked to remove oils and greases. With respect to Fe-electrodes, COD removal was better with Al-electrodes in (+9.56%, +0.45%, and +8.87%) at the same current densities. The initial temperature of the solution was 22°C, and in 15 min it increased by +1.5°C, +3°C, and +4.5°C, respectively. The initial temperature of the solution was 22°C, and in 15 min of electrolysis with the current densities applied, it increased by +1.5°C, +3°C, and +4.5°C, respectively. The solubility of aluminum sulfates decrease with temperature, and, therefore, the precipitation of the aluminum sulfate is enhanced at lower temperatures [46]. Current density is the most important parameter in all electro-chemical processes. Applied current density determines the rate of the coagulant release, bubble production and growth of flocs [93,94].

Fig. 16 shows that in 15 min the level of turbidity removal, working with 36 and 56 A/m, the process with Al slightly exceeded the process with Fe at +6.05% and +2.06%, respectively. However, Fe has a better turbidity removal performance (97.56%, 74 A/m), exceeding by +20.01% the removal process with Al-electrodes at the same current density.

The level of removal of TSS with Al-electrodes with 36 and 56 A/m was superior to the process with Fe in +1.42% and +23.51%, respectively. With a current density of 74 A/m, the Fe-process turned out to have a better removal level at +7.1% than the Al-process. The current density is expected to exhibit a strong effect on EC [85,86], especially on the kinetics of turbidity removal: the higher the current, the shorter the treatment. This is ascribed to the fact that at high current density, the extent of anodic dissolution of Al increases, resulting in a greater amount of precipitate for the removal of pollutants. Moreover, the bubble generation rate increases and the bubble size decreases with increasing current density. These effects are both beneficial for high pollutant removal by H$_2$ flotation [86].

4. Conclusions

The removal of COD was investigated by applying the EC process. High levels of oil–grease (>65%) and COD (>72%) were achieved in a range of pH 7–8. Electrocoagulation is an
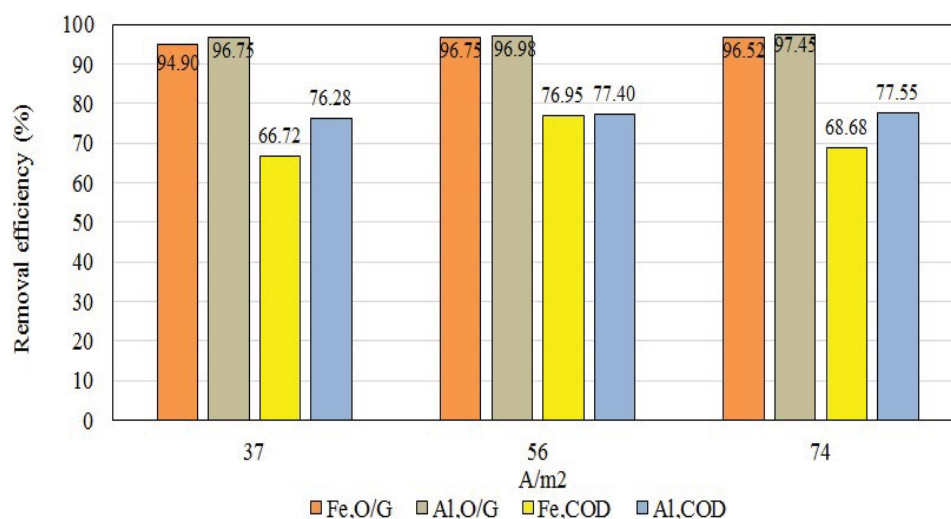

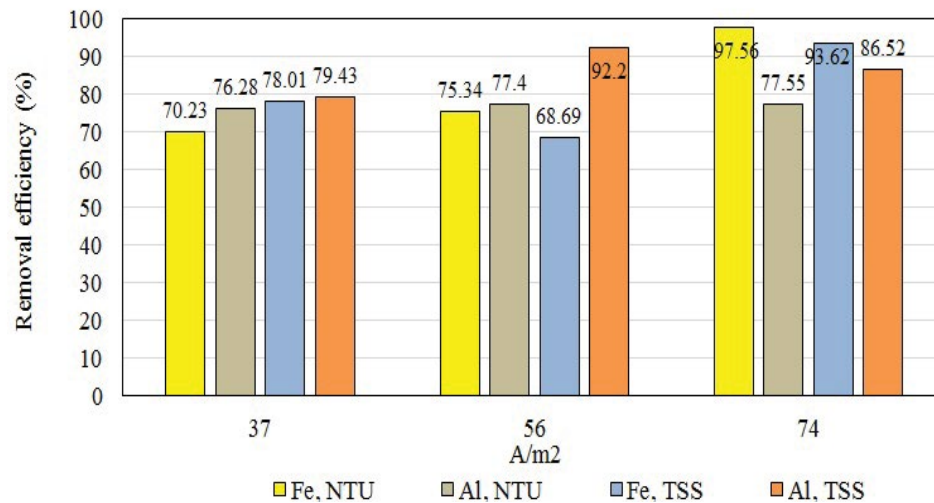Fig. 15. Effect of current density on the removal of O/G and COD.

Fig. 16. Effect of current density on the removal of NTU and TSS.

attractive low-cost technique to remove organic pollutants and is an easy process to control.

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