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Treatment of wastewater from the cleaning circuits of the margarine industry by electrocoagulation/flotation process

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ABSTRACT

This work aims to evaluate the applicability of electrocoagulation/flotation for the treatment of wastewater from the cleaning processes of the production line of the margarine industry. The electrochemical experiments were performed using plate aluminum electrodes (99% Al). The effects of pH, electrolysis time and current density were studied and discussed. Metal and energy consumption were also evaluated. Obtained results show that the removal of turbidity was up to 99.6% at pH 5.5. The reduction of chemical oxygen demand and total Kjeldahl nitrogen levels can reach 82%–86% and about 78%, respectively, at current densities greater than or equal to 62.5 A/m². The specific consumption of aluminum and energy was about 20.98 g/m³ and 0.156 kWh/m³, respectively when 0.05 A was applied for 15 min. Generally, the effluent treated by electrocoagulation/flotation becomes clear, clean and practically free from any visible pollutant especially when a current intensity of 0.05 A (62.5 A/m²) is applied. Treatment of margarine wastewater by electrocoagulation seems to be a plausible and promising solution because the process is fast, easily operated and leads to a final treated effluent of satisfactory quality.

Keywords: Margarine industry; Wastewater treatment; Electrocoagulation/flotation; Aluminum electrodes

1. Introduction

A wide variety of food oil products is available to the consuming public such as margarine, butter, mayonnaise, salad and cooking oils, ice cream fats, etc. Margarine is an inexpensive alternate to butter, made from oil or a combination of oils through the process of hydrogenation. This produces fats with great keeping qualities as well as higher melting points. The raw materials required for the manufacture of margarine are fat, blend, aqueous phase and additives. However, butter is mainly made from dairy milk and salt.

The characteristics and volume of wastewater discharged from food processing factories vary with the products and production procedures. Similar to the other food industry, margarine production consumes large quantities of water [1]. Specific water consumption in some companies can reach values between 0.7 and 2.71 m³/t of produced margarine [2]. Water is mainly used as raw material, for heating and cooling processes and for cleaning. For margarine production, wastewater results mainly from the cleaning circuits of the clean-in-place (CIP) plants [3]. The CIP system is a part of the modern margarine production facility that prevents bacteria growth and assures sanitary

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equipment conditions. Effluents from the margarine industry must be treated before being discharged into the natural environment or in the urban wastewater treatment plants.

Oily wastewater treatment was discussed by many authors. The methods employed include ultrafiltration [4], reverse osmosis [5], precipitation, coagulation, flocculation and flotation [6–8], physicochemical treatments followed by biological processes [9–11], biological methods [12], thermochemical treatment [13], photocatalysis [14], etc. The application of the fat trap for the wastewater treatment in margarine production was investigated by Aleksić et al. [1]. This physical process can give satisfactory results that are consistent with the regulation. Wastewater can be successfully treated with a fat trap only if effluent flow, oil and fat concentration, proper and regular cleaning and maintenance of the equipment are taken into account [1]. Microfiltration was used in the pre-treatment of margarine manufacturing wastewater because the effluent can cause problems in further treatment such as high costs for sludge disposal, coating in treatment plants, and saponification of fats in equalization tanks [15]. Furthermore, wastewater from margarine production can also be turned into high-quality water by using membrane bioreactor technology [16].

Electrocoagulation technology, which is one of the most efficient methods used to remove pollutants from industrial wastewaters [17,18], allows destabilization of suspended, emulsified or dissolved contaminants in an aqueous medium by applying an electric current. It is an alternative method to classic chemical coagulation. Electrocoagulation is a simple, efficient and a low sludge producing process [19]. It is based on the in situ formation of coagulant through the electro-dissolution of sacrificial anodes such as iron and aluminum electrodes, thus avoiding usage of chemical coagulants. Electrocoagulation was applied to remove organic pollutants, oil and grease from canola oil wastewater using aluminum and iron electrodes [20]. The treatment leads to 80% removal of organic carbon and nearly 100% removal of suspended solids. It could remove between 52%–59% of oil and grease from canola oil wastewater. Palm oil mill effluent was also treated using electrocoagulation [21]. The oil removal efficiency of 72% could be achieved with a current density of 20 A/m^2 for 5 min at 40°C and pH 5 [21]. Nasution et al. [22] have also studied the effectiveness of electrocoagulation to treat palm oil mill effluent with oil and grease concentration of 4,000–6,000 mg/L. It was revealed that chemical oxygen demand (COD) in effluent pre-treated by anaerobic pond dropped 62.35% and 59.41% using Al and Fe electrodes, respectively. The electrocoagulation process was also applied for the treatment of dairy wastewater. According to Bensadok et al. [23], the efficiency of COD removal by this process using aluminum electrodes could reach 80% under optimal conditions. Other researchers have applied electrocoagulation for the treatment of palm oil mill effluent with the presence of hydrogen peroxide (H_2O_2) as an oxidizing agent and polyaluminum chloride (PAC) as coagulant-aid [24]. Their results demonstrate that electrocoagulation is able to achieve more than 70% COD removal in 180 min at current density 30–80 mA/cm^2 reliant upon the concentration of H_2O_2 and PAC.

The purpose of this work is to investigate the efficiency of the electrocoagulation process in treating effluents from the cleaning circuits of the margarine industry. The effects of pH, electrolysis time and current density were studied and discussed. Metal and energy consumption were also evaluated.

2. Materials and methods

2.1. *Effluent*

The wastewater treated in this work is from the cleaning processes of the production line of a margarine industry located in the industrial zone of Settat (Morocco). The cleaning of equipment and packaging machines by the use of hot water and detergents is work performed periodically to improve the production quality of the finished product. It is based on three main steps: (i) degreasing by hot water at 75°C–80°C, (ii) cleaning and disinfection of containers with hot water 75°C–80°C containing detergents, and (iii) rinsing again by hot water 75°C–80°C. The mixture of wastewater from these processes generates a composite effluent.

2.2. *Electrocoagulation process*

The electrochemical treatment was performed on a laboratory scale with an electrolytic cell. Two aluminum electrodes installed in parallel, with flat and rectangular shapes (18 cm × 2 cm × 2 mm), were used as anode and cathode. The active surface area of each one was 8 cm^2. The quantitative compositional mapping of aluminum electrodes was carried out using a Cameca SX100 electron probe microanalysis. Back-scattered electron (BSE) image was taken with 15 kV, 15 nA. Obtained results show that aluminum plates consist of about 99% Al (Fig. 1). In order to give a regulated electricity current to the electrochemical cell, a DC power supply (Advance Electronics PP10A, 0–50 V, 0–1 A) was used. Electrodes were connected to the positive and negative terminals of the DC power supply.

Electrocoagulation experiments were carried out with 200 mL of wastewater at 25°C ± 2°C. A magnetic stirrer (200 rpm) was used to maintain the homogeneous mixing

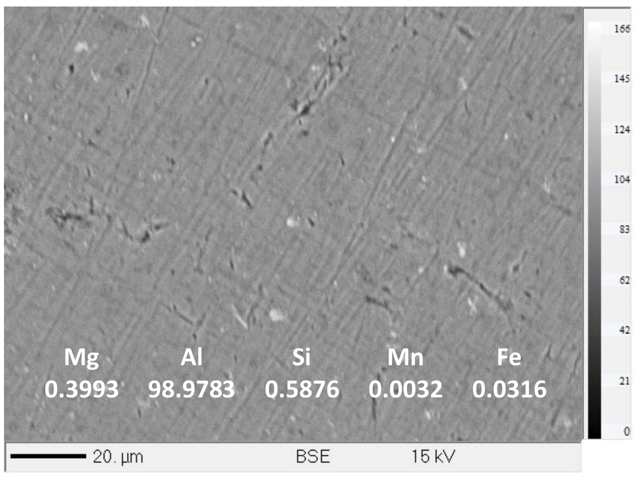

Fig. 1. Electron probe microanalysis of aluminum electrodes.

of the solution in the reactor. At the end of the treatment, obtained suspensions were allowed to stand for about 30 min to reach a spontaneous separation of formed flocs. The liquid phase was then separated in order to be analyzed.

After electrocoagulation experiments, energy consumption and Al metal consumption can be calculated as follows:

$$\text{Energy consumption (kWh/m}^3\text{)} = \frac{U \cdot I \cdot t}{V} \quad (1)$$

$$\text{Al consumption (g/m}^3\text{)} = \frac{I \cdot t \cdot M}{Z \cdot F \cdot V} \quad (2)$$

where U is the cell voltage; I is the current; t is the electrolysis time; V is the volume of the solution to be treated; F is the Faraday's constant; M is the molecular weight and Z is the number of electron transfer.

2.3. Analysis

The main analyzed parameters were turbidity, pH, electrical conductivity, chemical oxygen demand (COD), biological oxygen demand (BOD_5), total Kjeldahl nitrogen (TKN) and total suspended matter (TSM). Turbidity was measured using a device (Waterproof TN-100/T-100, Eutech Instruments). The pH and conductivity were measured directly by the use of a pH meter (WTW series, Inolabo pH 730) and a conductivity meter model (HANNA Instruments EC 214 conductivity meter), respectively. The COD and BOD_5 measurements were performed according to the operating conditions described in the standardized experimental norms NF T90-101 and NF T90-103, respectively. The TKN analysis was carried out according to standard NF T90-110. TSM was analyzed according to the NF T90-105 standard.

All chemical reagents used were of analytical grade and all solutions were prepared in deionized water. All laboratory material was carefully cleaned before use by first washing with a neutral detergent solution and then with distilled water.

3. Results and discussion

3.1. Characterization of the effluent

The analytical characteristics of the margarine effluent are presented in Table 1. The average values of pH and electrical conductivity are of the order of 5.53 and 2.97 mS/cm, respectively. The values of COD and BOD_5 exceed the limit allowed by the Moroccan standard (500 mg O_2/L for COD and 100 mg O_2/L for BOD). The COD/BOD_5 ratio (2.13) which is less than 3 shows that the effluent pollution is biodegradable. The high turbidity (495 NTU) is mainly due to the high level of suspended matter (718 mg/L). This is generally responsible for the reduction of the penetration of light into the water and consequently the decrease in photosynthetic activity. This can lead to a drop in phytoplankton productivity and a decrease in dissolved oxygen concentration. The TKN content which is about 33.6 mg/L comes mainly from residual raw materials that are rich in oils and proteins.

Table 1
Characteristics of margarine industry wastewater

Parameters	Value
pH	5.53
Conductivity (mS/cm)	2.97
Turbidity (NTU)	495
COD (mg O_2/L)	2,688
BOD_5 (mg O_2/L)	902
COD/BOD	2.13
TSM (mg/L)	718
TKN (mg/L)	33.6

3.2. Treatment by electrocoagulation

The effect of pH on the reduction of turbidity was evaluated by varying its value from 2 to 10 (Fig. 2). Experiments were carried out by applying a current intensity of 0.05 A (62.5 A/m²). The results show a decrease in turbidity of about 99.6% when the margarine effluent is treated at its original pH without correction (pH~5.5). For this reason, we have chosen to optimize the other process parameters without changing the initial pH of the wastewater. We also see a decrease in removal efficiency of turbidity at pH too acidic and basic. This decrease is attributed to the amphoteric behavior of $Al(OH)_3$. In fact, aluminum is normally found as soluble Al^{3+} cations when the initial pH is low and as monomeric anions $Al(OH)_4^-$ at high pH values. These soluble species are useless for treating wastewater because it is the formation of polymerized species and $Al(OH)_3$ which leads to effective treatments.

It is well known that current intensity determines not only the dose of the coagulant but also the rate of bubble production and the growth of flocs [25] that can influence the effectiveness of electrocoagulation treatment. Therefore the current intensity is considered as an essential parameter in electrocoagulation experiments. It is very useful in determining optimal treatment conditions in terms of cost and efficiency.

To study the influence of current intensity on the treatment, we have performed a series of tests by applying current intensities ranging from 5 mA to 0.08 A. The study was carried out at pH = 5.5 for 15 min as electrolysis time. Fig. 3 shows the evolution of turbidity as a function of the current intensity. The obtained results highlight that the treatment rate increases with increasing intensity or density of the applied electric current. Indeed, the current density is proportional to the intensity of the electric current, through a surface S, according to the following relation:

$$J = \frac{I}{S} \quad (3)$$

We note that the turbidity varies inversely with the current intensity. This could be explained by the fact that increasing I (A) or J (A/m²) causes the increase of the quantity of coagulant formed by dissolution of the sacrificial anode (faraday law) and promotes the release of the gas bubbles at the cathode. The size of the hydrogen gas bubbles evolved at the cathode decreases with the increase in the intensity of the applied current. Increasing the density of microbubbles and decreasing their size is beneficial for having an upward flow, that is, removal of flocs formed by flotation at the surface.

The effect of current density on the reduction of COD was also evaluated. The results obtained and illustrated in Fig. 3 highlight a remarkable decrease in COD during the treatment of the effluent. As we can see, the COD reduction can reach 82%–86% for current densities greater than or equal to 62.5 A/m² (0.05 A).

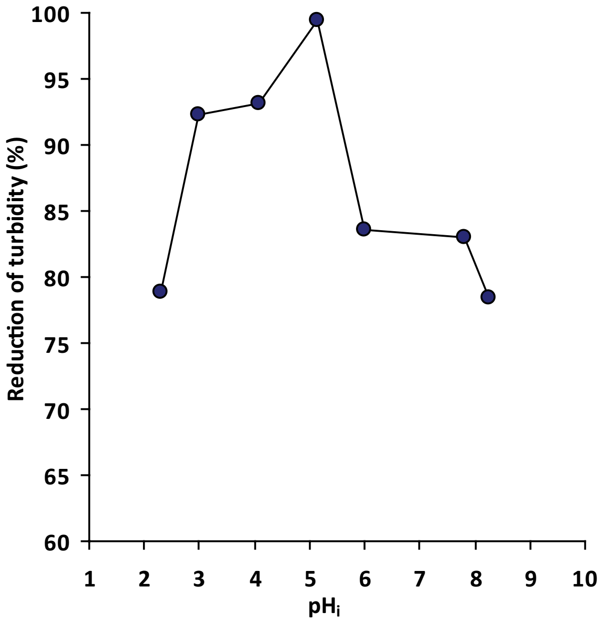

Fig. 2. Effect of pH on the treatment of margarine effluent by the electrocoagulation process.

On the other hand, we performed a kinetic study at pH 5.5 (without correction) by imposing current intensities of 0.01 A (12.5 A/m²); 0.03 A (37.5 A/m²) and 0.05 A (62.5 A/m²). The experiments were conducted over a time interval ranging from 0 to 30 min. The results obtained show that the treatment time plays an important role during the electrocoagulation process. From Fig. 4, we observe a significant decrease in the turbidity of the effluent during the electrolysis time for the different current densities studied. This decrease is even faster than the current density is high. A minimum value of the turbidity is observed after 15 min of treatment for a current density of 62.5 A/m² which corresponds to a current intensity equal to 0.05 A. Furthermore, we find that when the current density increases, the treatment time decreases due to the strong dissolution of the aluminum sacrificial electrode. It turns out that the electrolysis time plays a major role and has a remarkable effect on the effectiveness of electrocoagulation treatment. The results highlight that not only electrolysis time limits the elimination kinetics; applied current density also plays a role. It turned out therefore that there is a synergistic effect of these two parameters on the performance of the electrocoagulation process. The electrolysis time required to achieve treatment can be reduced by increasing the applied current. As can be seen from Fig. 4, electrolysis time decreases from 25–30 to 15 min by increasing current density from 37.5 to 62.5 A/m².

The cost of the electrocoagulation process is determined through the consumption of the electrodes and the electrical energy. The kinetics of the dissolution of the aluminum electrode and the electrical energy consumed were monitored for three applied current densities 12.5, 37.5 and 62.5 A/m² (Fig. 5). The results obtained show that the amount of aluminum as well as the energy consumed increases steadily with the treatment time. For example, when applying a current of 0.05 A (i.e. 62.5 A/m²), aluminum and energy consumption increases from 2.8 to 41.97 g Al/m³ and from 0.02 to 0.312 kWh/m³ when the electrolysis

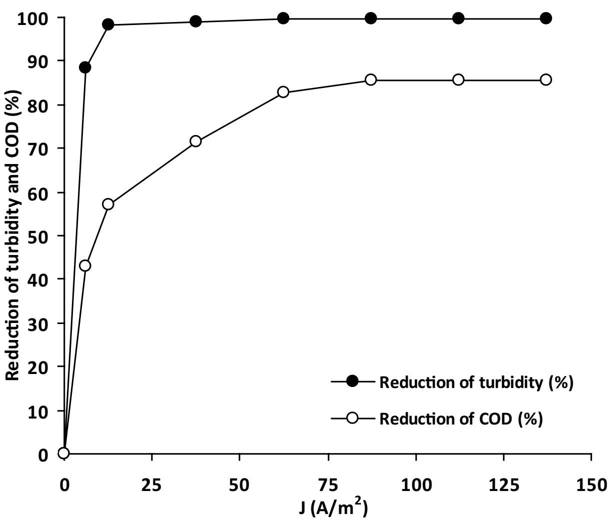

Fig. 3. Effect of current density on the reduction of turbidity and COD of margarine effluent.

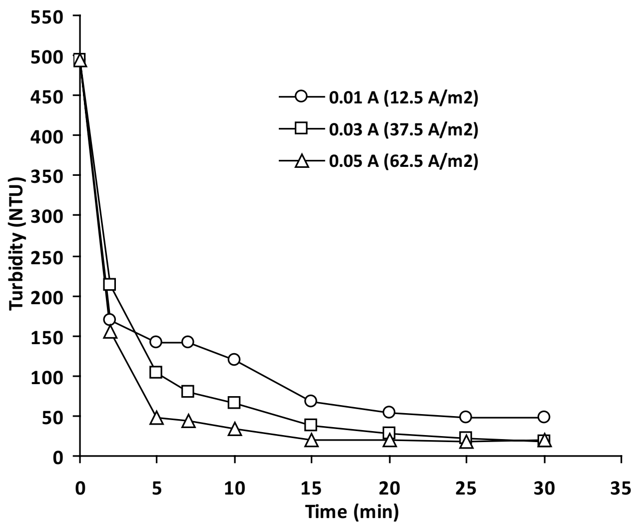

Fig. 4. Effect of electrolysis time on the turbidity of margarine effluent.

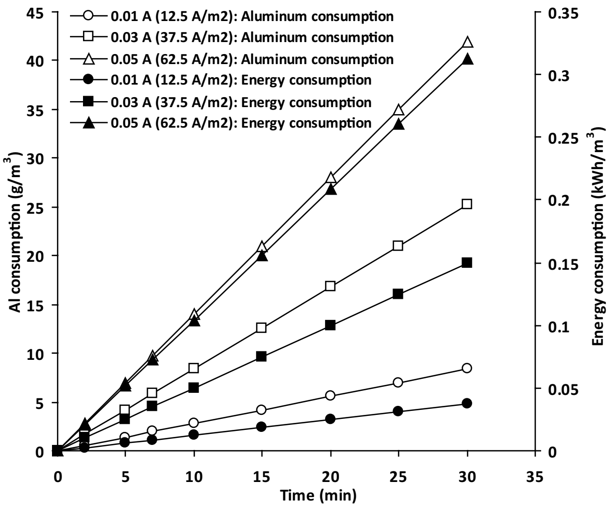

Fig. 5. Aluminum and energy consumption as a function of electrolysis time for different applied current densities.

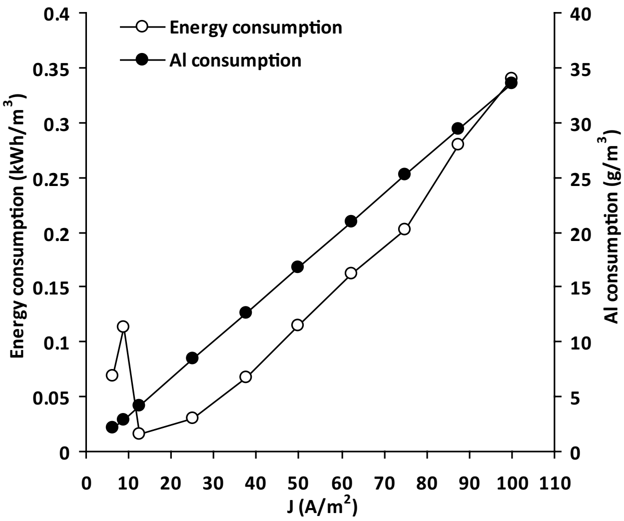

Fig. 6. Aluminum and energy consumption as a function of current density applied during an electrolysis period of 15 min.

time goes from 2 to 30 min. This increase can be explained by the fact that, at higher currents, the formation of aluminum ions is produced rapidly.

On the other hand, the evolution of electric energy and aluminum consumption as a function of current density (Fig. 6) shows that the dissolution of aluminum increases linearly as the current density increases. As can be seen, Al consumption increases from 2.1 to 33.57 g/m^3 by increasing the current from 0.005 A (i.e. 6.25 A/m^2) to 0.08 A (i.e. 100 A/m^2). We also observe an increase in the energy consumed when the intensity of current increases. For example, the energy consumption increases from 0.016 to 0.34 kWh/m^3 when current density goes from 12.5 to 100 A/m^2. This can be explained by Eq. (4) which shows that the energy consumed is proportional to the voltage and current intensity for a given duration t of the experiment.

$$E = U \times I \times t \tag{4}$$

Taking into account the optimal operating conditions obtained in the first step, the consumption of aluminum and energy was about 20.98 g/m^3 and 0.156 kWh/m^3, respectively, when 0.05 A was applied for 15 min.

It should be noted that it is possible to reduce energy consumption by increasing the electrical conductivity of the reaction medium. The addition of sodium chloride NaCl can reduce the voltage between the electrodes, at a constant current intensity, due to the decrease in resistance. NaCl salt is generally chosen because it is non-toxic and its cost is moderate [26]. The energy consumption is proportional to the voltage applied between the electrodes, so the energy consumption should decrease with the increase of the conductivity.

3.3. *Treatment efficiency*

The analysis of the effluent after its electrocoagulation treatment at different current intensities shows an almost complete elimination of turbidity (99%–99.6%). On the other hand, the COD of the effluent decreased from 2,688 mg O_2/L to 960 mg O_2/L, 576 mg O_2/L and 476 mg O_2/L when applying current densities 12.5, 37.5 and 62.5 A/m^2, respectively. The COD reduction rates are in the order of 64%, 79% and 82% when current intensities of 0.01 A, 0.03 A and 0.05 A are applied, respectively. Some studies have shown that the rate of COD abatement reaches a maximum limit that cannot be exceeded even if the electrical intensity and/or contact time continue to be increased [27]. Chemical analysis of the treated effluent revealed TKN removal efficiencies of about 78% (Fig. 7).

Examination of the appearance of the margarine effluent, after its treatment by electrocoagulation, shows a marked improvement (Fig. 8). As we can see, the effluent becomes clear, clean and practically free from any visible pollutant especially when a current intensity of 0.05 A (62.5 A/m^2) is applied.

4. Conclusion

This work shows that electrocoagulation is an effective process for the treatment of liquid wastes resulting from the washing of equipment used in the manufacture of margarine. This treatment makes it possible to obtain clear and non-turbid residual effluents. It turned out that the electrocoagulation technique is quick and simple to implement. Experiments allowed us to draw the following conclusions: the three factors pH, electrolysis time and current intensity greatly influence the effectiveness of electrocoagulation treatment. The rate of treatment increases with the increase in the intensity of the applied electric current which favors the dissolution of the sacrificial aluminum anode (faraday law) and the release of the hydrogen gas bubbles at the cathode. The rate of turbidity reduction of the effluent can reach its maximum of 99.6% at the original pH of the effluent (pH 5.5). The turbidity decreases significantly during the electrolysis time for the different current

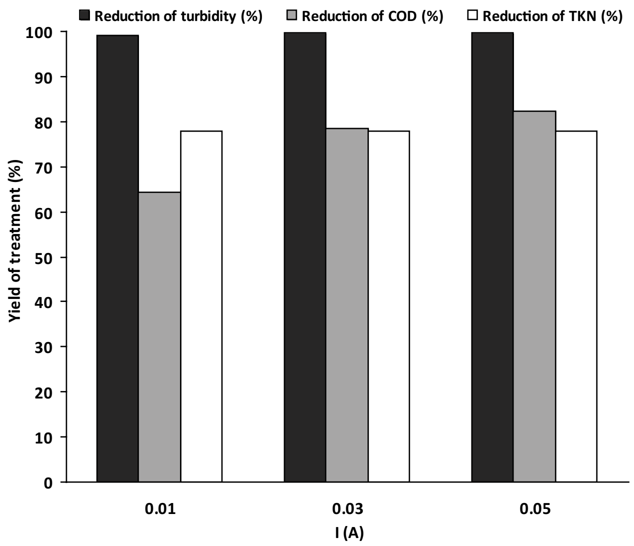

Fig. 7. Reduction of turbidity, COD and TKN of margarine effluent as a function of current intensity applied during an electrolysis period of 15 min.

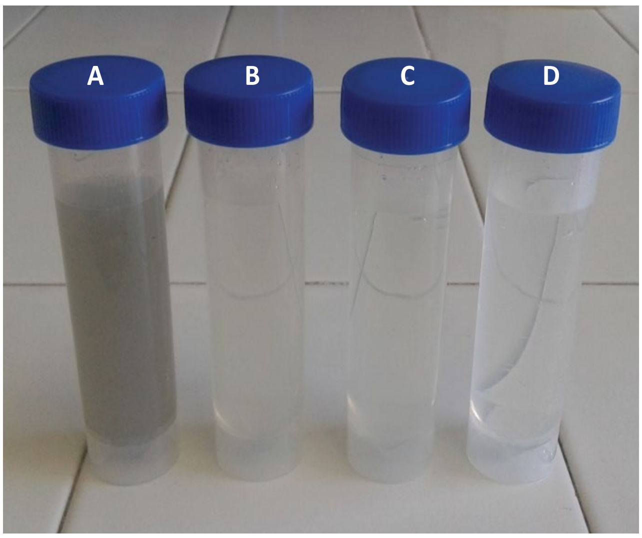

Fig. 8. Margarine effluent before (a) and after treatment by electrocoagulation [12.5 A/m² (b), 37.5 A/m² (c) and 62.5 A/m² (d)].

densities; this decrease is even faster than the current density is high. The reduction of COD and TKN levels can reach 82%–86% and about 78%, respectively, at current densities greater than or equal to 62.5 A/m². Furthermore, under optimal conditions of pH and electrolysis time, the specific consumption of aluminum and energy is in the order of 20.98 g/m³ and 0.156 kWh/m³.

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