

Continuous flow electrocoagulation in the treatment of wastewater from dairy industries

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ABSTRACT

Dairy industry wastewater contains high levels of organic matter, consisting mainly of fat, protein and products of their partial microbial decomposition. In the present study, the use of continuous electrocoagulation is proposed for the primary treatment of dairy wastewater. The electrochemical treatment was carried out in a continuous flow cell with aluminum electrodes. The influence of the voltage, the distance between the electrodes and the hydraulic residence time (HRT) on the process performance was assessed, by measuring the removal of color, turbidity, total organic carbon (TOC) and chemical oxygen demand (COD). The optimum voltage, distance between the electrodes and HRT were 10 V, 1 cm and 90 min, respectively, yielding a current density of 13.3 A.m^{-2} . Under these conditions, removal of color, turbidity, TOC and COD were 94%, 93%, 65% and 69%, respectively, after a steady state was reached in the continuous flow reactor.

Key words | aluminum electrodes, dairy wastewater, electrocoagulation, flow continuous reactor, hydraulic residence time

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INTRODUCTION

Water is used throughout all processes in dairy industries, for cleaning, sanitization, heating, cooling and floor washing, thus yielding large volumes of wastewater (Sarkar *et al.* 2006). Dairy wastewater mainly contains milk products and its derivatives (Baskaran *et al.* 2003) and is characterized by: high biological and chemical oxygen demand (BOD and COD, respectively); high levels of dissolved and suspended organic material including fats, oil and grease; and nutrients, such as ammonia, phosphates and other minerals. Therefore, proper attention is required before disposal of such wastewaters (Ayeche 2012; Andrade *et al.* 2014).

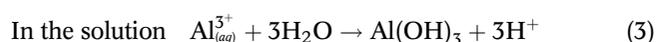
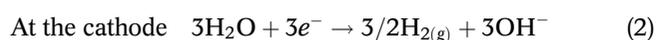
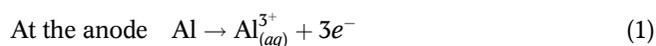
Dairy wastewaters are usually treated by coagulation and flocculation of suspended matter and emulsified fat, followed by biological treatment processes (TezcanUn & Ozel 2013; Andrade *et al.* 2014). Coagulation/flocculation is one of the most important physicochemical treatment steps in industrial wastewater treatment, focused on reducing the suspended and colloidal solids that cause high turbidity and also contribute to the BOD and COD of the wastewater (TezcanUn & Ozel 2013). Conventional coagulation

processes require large amounts of reactive chemicals and constant control of the process.

The use of electrocoagulation (EC) in wastewater treatment has been attracting interest, due to its flexibility and environmental compatibility (Harif *et al.* 2012). This technique has some advantages over conventional methods, such as its simple equipment, easy operation, low retention time, lack of chemicals needed, the rapid sedimentation of the electrogenerated flocs and a lower production of sludge. Therefore, it has been shown that EC can be used as an effective and reliable method for reducing or removing a large variety of pollutants in wastewater (Kobya *et al.* 2006).

Aluminum (Al) and iron are commonly used as electrodes, and their cations are generated by the dissolution of sacrificial anodes upon the application of a direct current. The metal ion generation takes place at the anode, while hydrogen gas is produced at the cathode as shown in reactions (1) and (2). The metal ions generated are hydrolyzed in the electrochemical cell to produce metal hydroxide ions, according to reaction (3), and the solubility of the metal

hydroxide complexes formed depends on the pH and ionic strength. Insoluble flocs are generated in a pH range between 6.0 and 7.0 as can be seen for the solubility of aluminum hydroxide $\text{Al}(\text{OH})_{3(s)}$ described by [Bensadok *et al.* \(2011\)](#) and [Cotillas *et al.* \(2013\)](#). Metal species react with negatively charged particles present in the water to form flocs. The *in situ* generation of coagulants means that EC processes do not require the addition of any chemicals. The gases produced at the cathode during the electrolysis of water cause the resulting flocs to float ([Daneshvar *et al.* 2006](#)).



EC has been applied successfully to remove phenolic compounds ([Adhoum & Monser 2004](#)), decolorize reactive dye solutions ([Daneshvar *et al.* 2006](#); [Pi *et al.* 2014](#); [Pirkarami & Olya 2014](#)), clarify suspended clay solutions ([Holt *et al.* 2004](#)), treat textile wastewater ([Khandegar & Saroha 2013](#)), and remove heavy metals ([Eiband *et al.* 2014](#); [Xu *et al.* 2014](#)) from aqueous solutions. The advantages of EC include its high particulate removal efficiency, its compact treatment facilities, and the relatively low cost of its complete automation ([C nizarez *et al.* 2006](#)).

EC has been very successfully employed in removing suspended oil/grease and organic material from a variety of industrial effluents, such as oil refinery wastes ([Chen *et al.* 2000](#)). Some studies on the EC treatment of dairy industry wastewater have been reported ([Tchamango *et al.* 2010](#)). [Tchamango *et al.* \(2010\)](#) used aluminum electrodes in batch reactors and reported COD, nitrogen and turbidity removal efficiencies of 61%, 81% and 100%, respectively. [Kushwaha *et al.* \(2010\)](#) used iron electrodes and reported significant COD removal of 70%. These studies were focused only on batch systems. Since dairy industries generate large volume of wastewater, usually 0.2 to 10 L of waste per liter of processed milk ([Balannec *et al.* 2005](#)), the practical use of batch reactors is limited. This situation motivates the study of continuous flow systems, in order to obtain the best possible layout of the electrodes configuration in the reactor, the effluent flow rate, hydraulic residence time (HRT), etc., that optimizes the removal efficiency of suspended organic material in the system.

The present work aimed to study the primary treatment of dairy wastewater by a continuous flow EC process using aluminum electrodes, investigating the influence of operation

variables distance between the electrodes (DE), voltage (V) and HRT in relation to simultaneous removal of color, turbidity, total organic carbon (TOC) and COD using factorial design. pH, current density and conductivity were monitored on the best operating conditions.

EXPERIMENTAL

Materials and methods

In the present study, a synthetic dairy wastewater (SDW) was prepared by dissolving 2 g of powdered whole milk in a suitable amount of distilled water to obtain 1 L of solution ([Leal *et al.* 2006](#)). SDW was used in the present study to minimize fluctuations in the composition of the effluent, ensuring standardization of parameters at time zero. Several investigators have used the same method for making SDW ([Kushwaha *et al.* 2010](#); [Tchamango *et al.* 2010](#)). The simulated wastewater was freshly prepared whenever required.

Analytical determinations

Apparent color was measured by spectrophotometry using a colorimeter DR870, Hach, USA ([APHA 1999](#)). Turbidity was measured by direct reading in a turbidity meter DR870, Hach, USA ([APHA 1999](#)). The determinations of TOC were carried out in a total organic carbon analyzer (TOC-5000A, Shimadzu, Japan). The sample (1 mL) was filtered through a membrane (0.45 μm) and diluted to 25 mL with distilled water. The content of TOC was determined by the difference between the concentrations of total carbon and inorganic carbon.

COD was measured by the standard microscale colorimetric method ([APHA 1999](#)). The digestion of samples was carried out in a dry block heater (MA 4004, Marconi, Piracicaba, SP, Brazil) at 150 °C for 2 h. After cooling the samples, the readings were obtained using a digital colorimeter (DR870, Hach) previously calibrated using potassium hydrogen phthalate. The pH of the wastewater was measured at room temperature (25 °C) in a digital pH meter (pH LAB 827, Metrohm, Switzerland). The electrical conductivity of the samples was measured using a portable digital conductivity meter CON 10, Oakton, USA ([APHA 1999](#)). The current density used in the experiments was calculated using Equation (4), where the area is in m^2 :

$$\text{CD} = \frac{\text{Amper}}{\text{Area}} \quad (4)$$

Electrocoagulation

The electrochemical system consisted of a glass reactor with a working volume 2.0 L. The cell was fed with a constant flow of SDW using a peristaltic pump (MIT DC Power Supply MS 3005), at flow rates suitable to maintain nominal residence times of 30, 60 and 90 min. The system had four aluminum electrodes (2 cathodes and 2 anodes), with dimensions of 150 mm × 70 mm × 2 mm with inter-electrode spacing of 1, 2 and 3 cm connected in parallel monopolar mode. The total submerged area of the electrodes in the SDW was 0.021 m². The distance between the electrodes was adjusted using glass plates attached to the electrode holder of the reactor. The current was maintained at a constant level by means of a direct current power supply (MIT DC POWER SUPPLY MS 3005) with digital precision (0–20 V, 0–5 A), equipped with an ampere meter and a voltmeter. Schematic representation of the system is shown in Figure 1.

Experiments were carried out following the conditions specified in a factorial design with three factors set at two levels (2³). Before each experiment, the pH of the SDW was adjusted to 6.0 (Cotillas *et al.* 2013) using 0.1 mol.L⁻¹ NaOH or 0.1 mol.L⁻¹ H₂SO₄ and adding aliquots of solution of 10% NaCl until a conductivity of 1,489 μS.cm⁻¹ was reached. Then, the SDW was poured into the EC reactor, to completely fill the reactor, covering the electrodes, and the pump and the DC power supply were started.

Samples from the system effluents were periodically collected for analysis of COD, TOC, color, turbidity, pH and conductivity. All of the samples were analyzed in duplicate to ensure data reproducibility, and additional measurements were carried out, if necessary. The sludge that floated to the surface was continuously manually removed from the reactor to prevent accumulation, preventing the sludge occupying the space of the liquid in

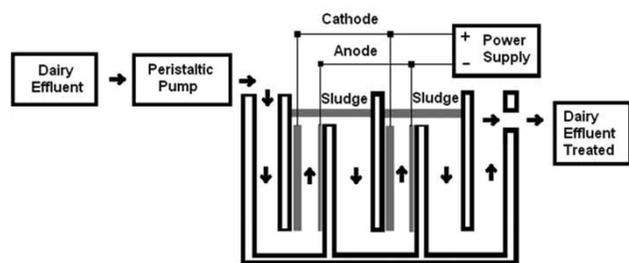


Figure 1 | Schematic diagram of the continuous EC system.

the reactor and changing the reaction volume in each experiment.

The data obtained from the experiments were analyzed using factorial design methodology. Three analytical steps: the adequacy of the various models run (sequential model sum of squares and model summary statistics); analysis of variance (ANOVA); and response surface plotting were performed to establish the optimum condition for the color, turbidity, TOC and COD removal.

Three variables were evaluated in this study: the distance between the electrodes (DE) = 1, 2 and 3 cm; hydraulic residence time HRT = 30, 60 and 90 min. and voltage (V) = 5, 7.5 and 10 V, and a total of 12 experiments were carried out, with real and coded values shown in Table 1. For each experimental condition a flow rate was calculated considering the effective volume of the reactor (distance between electrodes) and the HRT.

Energy consumption

The energy consumptions per m³ treated is shown in Equation (5) according to Kobya & Demirbas (2015).

$$C_{\text{energy}} = \frac{U \times i \times t_{\text{EC}}}{V_{\text{ef}}} \quad (5)$$

where U is the applied voltage, i is the current (A), t_{EC} is the operating time (s or h) and V_{ef} is volume of the treated effluent (m³) in the EC reactor.

Table 1 | EC treatment of SDW following a factorial design 2³ (real and coded values). The responses are expressed as percentage removal of each parameter

Run	V (V)	DE (cm)	HRT (min)	Color (%)	Turbidity (%)	TOC (%)	COD (%)
1	-1 (5)	-1 (1)	-1 (30)	5.5	2.0	27.0	10.1
2	-1 (5)	-1 (1)	1 (90)	46.2	44.8	26.4	6.2
3	-1 (5)	1 (3)	-1 (30)	0	0	0.58	5.3
4	-1 (5)	1 (3)	1 (90)	21.5	20.8	20.0	5.1
5	1 (10)	-1 (1)	-1 (30)	22.7	25.5	23.3	25.7
6	1 (10)	-1 (1)	1 (90)	94.1	93.4	65.0	69.5
7	1 (10)	1 (3)	-1 (30)	3.6	3.9	21.7	6.8
8	1 (10)	1 (3)	1 (90)	66.4	68.0	51.2	43.0
9	0 (7.5)	0 (2)	0 (60)	37.2	32.7	40.7	42.9
10	0 (7.5)	0 (2)	0 (60)	45.8	46.2	37.5	42.6
11	0 (7.5)	0 (2)	0 (60)	35.2	32.7	40.1	37.0
12	0 (7.5)	0 (2)	0 (60)	37.9	33.3	35.2	36.0

RESULTS AND DISCUSSION

The main parameters of the used SDW before EC were: color = $9,648 \pm 573 \text{ mg Pt.L}^{-1}$, turbidity = $1,296 \pm 51 \text{ NTU}$, TOC = $1,105 \pm 73 \text{ mg.L}^{-1}$, COD = $2,200 \pm 277 \text{ mg.L}^{-1}$, conductivity = $87 \pm 11 \text{ }\mu\text{S.cm}^{-1}$ and pH = 6.0 ± 0.2 . These characteristics were maintained uniform throughout the study.

Table 1 presents the real and coded values for the independent variables (distance between electrodes, HRT and voltage) and the responses in terms of color, turbidity, COD and TOC for each experiment.

From the results obtained in the factorial design (Table 1) it is possible to see that the best removal efficiency for each of the variables (color, turbidity, TOC and COD) was obtained in run 6 under the best operation conditions (DE = 1 cm, HRT = 90 min and V = 10 V). The energy consumption at the optimum condition (run 6) was 1.88 KWh/m^3 . Figure 2 shows the results of the efficiency of removal after the process of EC. After reaching a steady state at 180 min the efficiency removal were kept constant until time 360 min.

Run 8, which differed from run 6 only in the distance between the electrodes (DE = 3 to 1 cm) also yielded good results for removal of color (66 to 94%), turbidity (68 to 93%), COD (43 to 69%) and TOC (51 to 65%). The other run conditions produced removal efficiencies that were always lower than 50%.

It is possible to observe that there was a considerable reduction in color, turbidity, TOC and COD with a HRT of 90 min, a 1 cm gap between the electrodes and voltage of 10 V. These removal results are similar to those reported for Kushwaha et al. (2010), a batch system using iron electrodes, showed a removal of COD and turbidity of 70% and 100%, respectively, at a batch time of 90 min.

The statistical analysis of the experimental data reported in Table 1 permitted the validation of empirical models for

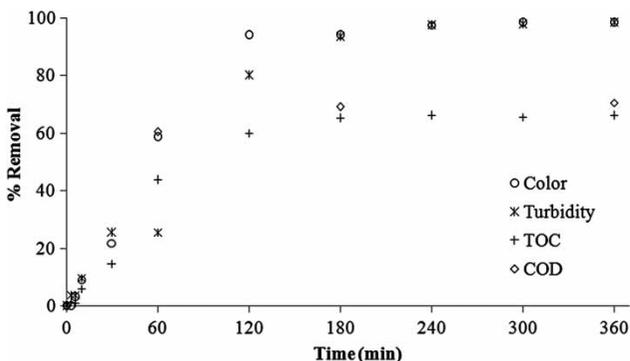


Figure 2 | Efficiency of removal of color, turbidity, TOC and COD for run 6 (10 V, 1 cm between electrodes, residence time 90 min).

efficiency of removal of color, turbidity, and for each experiment as a function of the distance between electrodes, the HRT and the voltage for each operational time evaluated. The models were validated by ANOVA that yielded correlation coefficients of 0.98, 0.98 and 0.87, respectively; and an *F*-test (calculated values 10.2, 9.5 and 2.2 higher than the tabulated ones, respectively), making the models valid at 95% confidence levels.

Through the statistical analysis (*F*-test) of the experimental data reported in Table 1, the models of color Equation (6), turbidity Equation (7) and TOC Equation (8) can be obtained as illustrated by the removal efficiency for color (5th column), turbidity (6th column) and TOC (7th column).

$$\text{Color} = 34.9 + 14.55 \times V - 9.98 \times \text{DE} + 24.91 \times \text{HRT} + 9.36 \times V \times \text{HRT} \quad (6)$$

$$\text{Turbidity} = 33.2 + 14.77 \times V - 8.51 \times \text{DE} + 25.08 \times \text{HRT} + 9.18 \times V \times \text{HRT} \quad (7)$$

$$\text{TOC} = 32.04 + 10.78 \times V - 6.18 \times \text{DE} + 11.12 \times \text{HRT} + 6.44 \times V \times \text{HRT} \quad (8)$$

where V, DE and HRT correspond to the coded values for voltage, distance between electrodes and HRT, respectively.

The models shows that the increase in voltage, and consequently in current density, and in HRT provides an increased removal of color, turbidity and TOC while the increase in the distance between the electrodes causes the opposite effect. According to Modirshahla et al. (2007), increasing the distance between the electrodes causes decrease of electrical current, and to achieve a certain current density, the voltage must be increased. On the other hand, conductivity decreases when the distance between the electrodes increases.

Figures 3–5 present the response surface and the contour plots obtained by Equations (6)–(8), respectively. It can be seen that the maximum efficiencies of removal of color, turbidity and TOC were obtained at a voltage, distance between electrodes and HRT of 10 V, 1 cm and 90 min, respectively. Further optimization of the process would require increasing the voltage and decreasing the HRT. Therefore these optimized conditions showed high removal efficiency of organic matter.

Comparing Figures 3 and 4, it is possible to observe a similar behavior of the effects of V, DE and HRT with efficiency of removal. This similarity is consistent with that reported in the literature (Richter & Neto 2003).

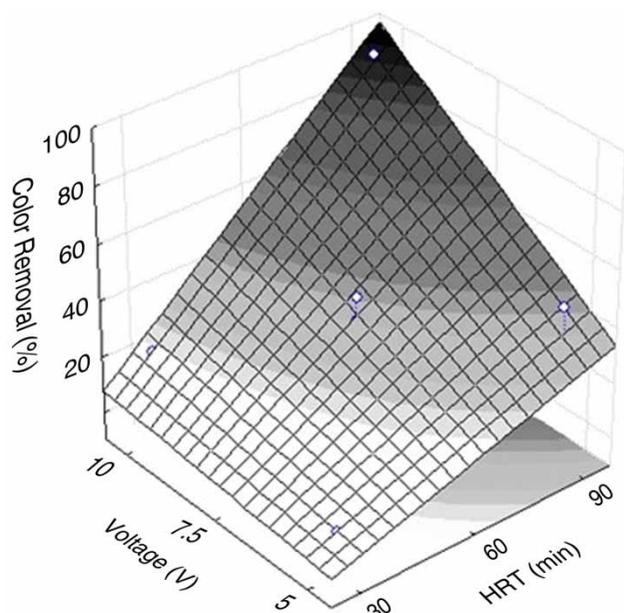


Figure 3 | Influence of voltage, distance between electrodes and hydraulic residence time on the efficiency of color removal.

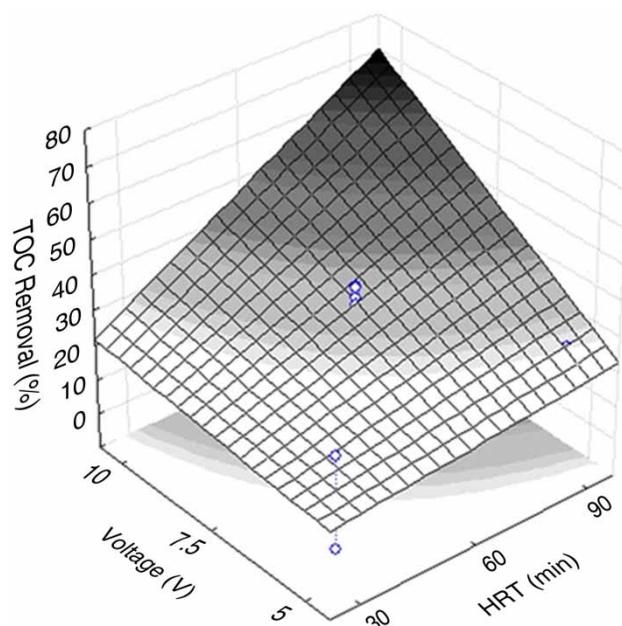


Figure 5 | Influence of voltage, distance between electrodes and hydraulic residence time on the efficiency of TOC removal.

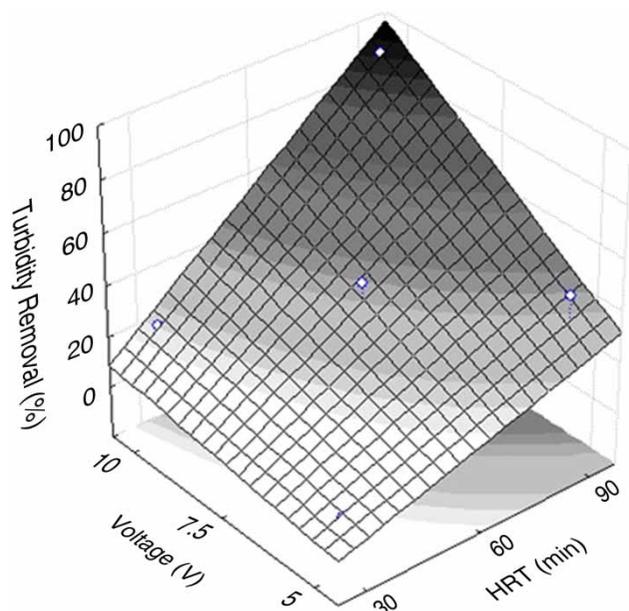


Figure 4 | Influence of voltage, distance between electrodes and hydraulic residence time on the efficiency of turbidity removal.

The negative effect of the distance between the electrodes is due to an increased resistance caused by the aqueous medium, which leads to a reduction in the electric current between the electrodes, slowing the leaching of aluminum, consequently hindering color, turbidity and TOC removal (Modirshahla *et al.* 2007).

For comparison of the results obtained, current densities were calculated for each voltage in the reactor, defined as

the current between the electrodes per unit area ($A.m^{-2}$). The positive effect of voltage in the treatment dairy wastewater was due to an increased current density, which leads to an increase in the electric current between the electrodes, and in consequence increased leaching of aluminum.

Bensadok *et al.* (2008, 2011) reported the experimental results of a batch reactor and showed that for high current densities ($\geq 25 A.m^{-2}$), abatement of the various pollution variables occurs in two phases as follows:

- (1) The first phase of decreasing pollution features, or the 'reactive stage' during which the COD and the turbidity removal yields increase with the current density. This can be explained by the fact that Al^{5+} production resulting from anode dissolution increases with current density.
- (2) The second, called here the 'stationary phase' is reached after 2 min of treatment: in this period, a further increase in aluminum concentration has no further effect on the treatment. Indeed, the suspended and dissolved organic matters were no longer limited in spite of the higher aluminum concentrations allowed by longer periods of Al anode dissolution. At this optimum time (2 min) and with $50 A.m^{-2}$, COD and turbidity removals attained, respectively, 81% and 96%.

The positive effect of HRT on the treatment of dairy wastewater was due to an increase in the reaction time, increasing

the leaching of aluminum and aluminum hydroxide formation. Higher HRT also contributed to decrease turbulence in the tank, favoring floc formation and flotation.

The results in Table 1 also show that the highest efficiency of COD removal (69%) was also obtained in run 6 (10 V, 1 cm and 90 min). Tchamango *et al.* (2010) in their studies in a batch system obtained the highest COD removal (60%), using a pair of aluminum electrodes, and attributing the low removal to the presence of carbohydrates, along with some dissolved organic compounds.

Removal of 70% and 99% of COD and oil-grease, respectively, was obtained by Sengil & Ozacar (2006), on dairy wastewater treatment by EC using mild steel electrodes. Kushwaha *et al.* (2010), using iron electrodes, obtained for the parameters COD, total sulfur (TS), total nitrogen (TN), and turbidity, removal efficiencies of 70%, 48.2%, 92.75% and 99.8%, respectively.

Experimental data of COD removal were statistically treated and Figure 6 presents the standard Pareto chart, which shows the effects of the variables (voltage, distance between electrodes and HRT) on the efficiency of removal of COD. The voltage and HRT presented positive significant effects ($P < 0.05$) on the efficiency of COD removal. The distance between the electrodes did not affect this response to a statistically significant level.

Variations in pH, current density and conductivity of the aqueous medium

pH

For all experimental conditions the initial pH was 6.0 and an increase in the pH with the operation time was observed until the system reached steady state (180 min). Then the pH remained constant between 7.8 and 9.2 until the end of each run. This tendency was attributed to the buffering capacity of the system: $\text{Al}^{3+}/\text{Al}(\text{OH})_3$ (Kobyta *et al.* 2006). Generally, the effluent pH is increased during EC, due to the constant leaching of Al^{3+} ions to the solution. In pH values close to 7.0, Al^{3+} ions are converted to $\text{Al}(\text{OH})_3$ and act as a coagulant. Residual Al^{3+} ions occur when the pH is lower than 4.0 and greater than 10.0 (Adhoum & Monser 2004).

Current density

For all runs no variation in the current density with the operating time was observed as shown in Figure 7. The low variation in the current density is indicative of non-passivation of the electrodes.

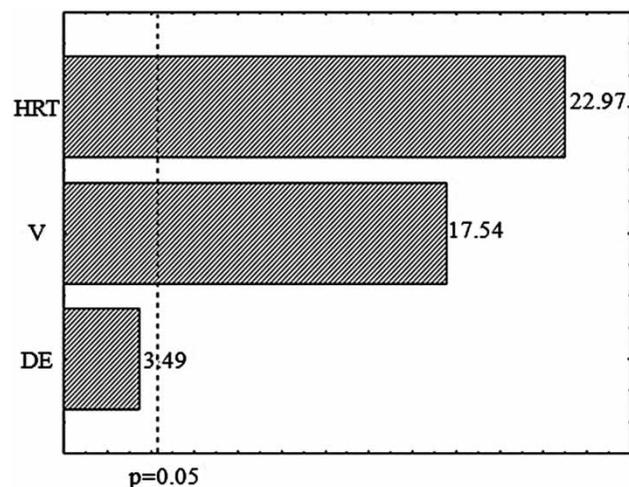


Figure 6 | Pareto chart of the effects of voltage, distance between electrodes and hydraulic residence time on the removal efficiency of COD ($P < 0.05$). Experimental data and conditions are shown in Table 1.

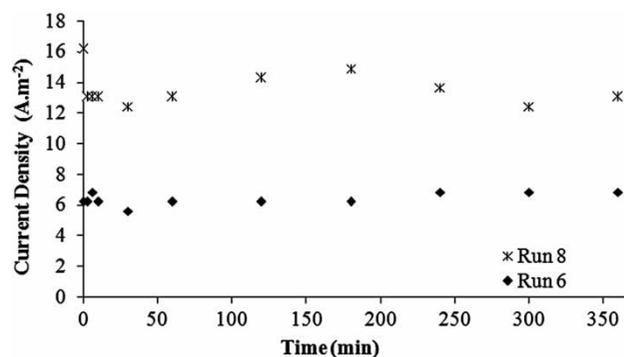


Figure 7 | Variation in the current density with operating time for runs 6 and 8.

Adhoum & Monser (2004) established that the current density in EC cell is one of most important parameters for controlling the rate of reaction in electrochemical processes. This parameter determines the rate of production of coagulants and size of the hydrogen bubbles. With an increase in the current density (increasing voltage), the amount of metal leachate is higher, resulting in a greater amount of hydroxide for removing pollutants.

Chen *et al.* (2000) observed a decrease in the time necessary for the treatment of an oil-water emulsion, when the current density was increased. The authors attributed this effect to an increase in the amount of oxidized aluminum, resulting in a greater amount of precipitation and, consequently, a greater removal of colloidal particles. Thus, an increase in current density implies in an increase in the amount of coagulant (Al^{3+}) produced by dissolving electrochemical aluminum anode (Chen *et al.* 2002).

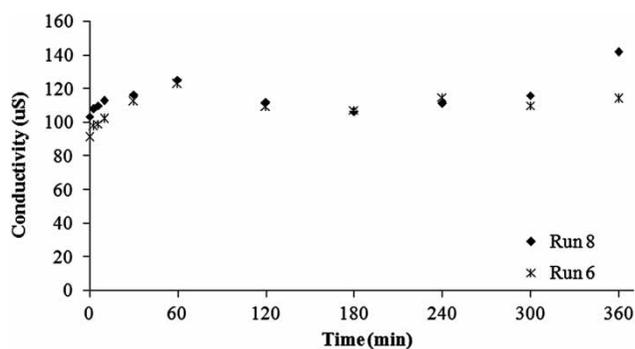


Figure 8 | Variation in the conductivity with operating time for runs 6 and 8.

Conductivity

Figure 8 shows the results for the conductivity of runs 6 and 8 (Table 1). There is a slight variation in the conductivity of the solution in the transient phase of the process. After the steady state is reached, conductivity remains constant until the end of the experiment. The increase in conductivity is related to an increase in the quantity of aluminum and hydroxide ions due to the electrolytic treatment. Wong et al. (2002), Donini et al. (1994) and Jia-Qian (1988) describe that increased conductivity is of fundamental importance for the development of EC/electroflotation processes.

The use of an electrolyte such as NaCl is common for the investigation of the effect of conductivity on EC processes. The higher conductivity accelerates the production of coagulant, increasing the efficiency of removal of organic matter, reducing energy consumption due to the use of lower voltages (Jia-Qian 1988; Wong et al. 2002; Adhoum & Monser 2004).

CONCLUSIONS

For EC in a continuous flow cell with aluminum electrodes for SDW, the optimum voltage, distance between the electrodes, and HRT were 10 V, 1 cm and 90 min, respectively, yielding a current density of 13.3 A.m⁻². After reaching a steady state in the continuous flow reactor removal rates obtained were: color (94%), turbidity (93%), TOC (65%) and COD (70%). The residual aluminum was 2.8 mg.L⁻¹. The voltage and the HRT have a positive influence on the system performance, while the distance between the electrodes negatively influenced the removal of color, turbidity, TOC and COD. The EC continuous process could be applied as a primary treatment of dairy effluents, which are characterized as having a high pollution charge.

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