Chemical Engineering Journal 149 (2009) 207–214

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

# Studies on the decolorization of textile dye wastewater by continuous electrocoagulation process

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ARTICLE INFO

*Article history:*
Received 30 June 2008
Received in revised form 8 October 2008
Accepted 17 October 2008

*Keywords:*
Continuous electrocoagulation
COD
Aluminum electrodes
Red dye
Decolorization

ABSTRACT

The performance of a continuous electrocoagulation (EC) process was investigated for decolorization and chemical oxygen demand (COD) abatement of a synthetic textile wastewater using aluminum electrodes. The effects of the relevant wastewater characteristics such as conductivity, influent pH and inlet dye concentration, but also of the key operating conditions such as current density and residence time were studied in order to optimize process performance. The results showed that color induced by a red dye was effectively removed (with a removal yield higher than 85%) for wastewater with a COD of 2500 mg/L and a dye concentration lower than 200 mg/L when pH ranged from 6 to 9, residence time was 14 min, current density was 31.25 mA/cm$^2$ and water conductivity was 2.4 mS/cm for an inter-electrode distance of 1 cm. Under these conditions, the COD abatement was also higher than 80%.

## 1. Introduction

Textile industries are among the most polluting industries in terms of the volume and the complexity of treatment of its effluents discharge. Wastewaters generated by textile industries are known to contain large amounts of toxic aromatic compounds, especially azo dyes. It is well known that some azo dyes and their degradation products, such as aromatic amines, are highly carcinogenic. Textile mill effluents are also characterized by high levels of color caused by residual dyes. The colored wastewater released into the ecosystem is also a dramatic source of aesthetic pollution and perturbation in the aquatic life. Moreover, textile wastewaters are known to exhibit various pH (either alkaline or acidic, depending on the process used), hot temperature, high biological and chemical (COD) oxygen demands and high concentrations of suspended solids. Conventional methods for dealing with textile wastewater consist of various combinations of biological, physical and chemical methods [1]. Adsorption and precipitation processes are very time-consuming and costly with low efficiency. Chemical degradation by oxidative agents such as chlorine constitutes the most relevant and effective method, but it may produce some very toxic secondary products, such as organochlorine compounds [2]. Photooxidation by UV/$H_2O_2$ or UV/$TiO_2$ needs additional chemicals and, therefore, may also cause a secondary pollution. Although biodegradation process is cheaper than other methods, it is less effective because of the toxicity of the dyes that induces an inhibiting effect on bacterial development [3].

In the recent years, investigations have been focused on the treatment of wastewaters using electrocoagulation (EC) because of the increase in environmental restrictions on effluent wastewater. Indeed, EC is a simple and efficient method for the treatment of most drinking waters and wastewaters. For a long time, it has been considered too expensive because of its high energy requirements and the high initial equipment costs, as compared to other technologies, especially biological treatments. However, EC has been shown recently to be competitive when biological treatments fail, while it avoids the formation of secondary pollutants [4]. The conventional EC technique uses a direct current source between metal electrodes immersed in polluted water. The electrical current causes the dissolution of metal plates of iron or aluminum into wastewater. The metal ions, at an appropriate pH, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate suspended particles or precipitate and adsorb dissolved contaminants [5].

The most widely used electrode materials in EC process are aluminum and iron. In the case of aluminum, the main reactions are

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doi:10.1016/j.cej.2008.10.018

**Nomenclature**

| | |
|---|---|
| $A_{450}$ | absorbance measured at 450 nm (AU) |
| $C_i$ | dye concentration in the inlet stream (kg/L) |
| $C_f$ | dye concentration in the exit stream (kg/L) |
| COD | chemical oxygen demand (mg/L) |
| $E_{dye}$ | specific energy consumption (kWh/kg dye removed) |
| EC | electrocoagulation |
| $F$ | Faraday's constant (96,487 C/mol $e^-$) |
| $I$ | current (A) |
| $j$ | current density (mA/cm$^2$) |
| $M_{Al}$ | molar mass of aluminum (0.02698 kg/mol) |
| $Q$ | inlet flow rate (L/h) |
| $R^2$ | correlation coefficient |
| $S$ | electrode surface area (m$^2$) |
| $t$ | time (h) |
| $U$ | cell potential (V) |
| $V$ | reactor volume (L) |
| $Y_{COD}$ | COD removal efficiency (%) |
| $Y_{COL}$ | color removal efficiency (%) |

*Greek letters*

| | |
|---|---|
| $\Delta m_{exp}$ | experimental weight loss of Al electrode (kg) |
| $\Delta m_{th}$ | theoretical weight loss of Al electrode (kg) |
| $\kappa$ | wastewater conductivity (S/m) |
| $\lambda_{max}$ | maximum wavelength of the absorption spectrum (nm) |
| $\mu_{Al}$ | specific mass consumption of the anode (kg Al/kg dye removed) |
| $\tau$ | residence time in the EC cell (s) |
| $\phi_{Al}$ | faradic yield of Al dissolution |

as

$$\text{Anode}: \mathrm{Al_{(s)}} \rightarrow \mathrm{Al^{3+}} + 3e^- \quad (1)$$

$$\text{Cathode}: 3\mathrm{H_2O} + 3e^- \rightarrow \frac{3}{2}\mathrm{H_{2(g)}} + 3\mathrm{OH^-} \quad (2)$$

$Al^{3+}$ and $OH^-$ ions generated by electrode reactions (1) and (2) react to form various monomeric species, which finally transform into $Al(OH)_{3(s)}$ according to complex precipitation kinetics:

$$\mathrm{Al^{3+}} + 3\mathrm{H_2O} \rightarrow \mathrm{Al(OH)_3} + 3\mathrm{H^+} \quad (3)$$

Freshly formed amorphous $Al(OH)_{3(s)}$ "sweep flocs" exhibit large surface areas which are beneficial for a rapid adsorption of soluble organic compounds and for trapping colloidal particles. Finally, these flocs are removed easily from aqueous medium by sedimentation or by flotation induced by the $H_2$ bubbles generated at the cathode [6–8], which is referred to as electroflotation.

Electrocoagulation and electroflotation have successfully been used for the treatment of wastewaters such as electroplating wastewater [9], laundry wastewater [10], latex particles [11], restaurant wastewater [12], dairy wastewater [13] and slaughterhouse wastewater [14]. The applicability of EC process has also been demonstrated for the removal of arsenic [15], phosphate [16], sulfide, sulfate and sulfite [17], boron [18], fluoride [19], nitrate [20] and chromate [21] ions. Treatments of wastewaters containing textile dyes by EC have also been studied. The results of these works show that COD, color, turbidity and even dissolved solids can be efficiently removed using EC [2,10–12,22–26]. The continuous mode of the EC process has however been less investigated, except in a few studies [2,8,12,19,25,27,28], especially the conditions of floc removal.

The prime objective behind this study was to discuss the applicability of EC in the continuous mode to decolorize a typical wastewater from a Moroccan textile factory in terms of dye type and concentration. A red dye from the dispersive dye class was used in this purpose. Continuous EC process was carried out in an EC cell characterized by a simpler electrode geometry than in the literature (see, e.g., [2,8]), but that included a region dedicated to floc separation. Experiments were conducted to examine the effects of wastewater properties and operating parameters, such as current density ($j$), residence time ($\tau$), water pH and conductivity ($\kappa$), and influent dye concentration ($C_i$), both on color removal efficiency and COD abatement in the effluent stream in order to optimize the operating conditions of EC process simultaneously in terms of color removal, energy consumption and electrode consumption.

## 2. Materials and methods

### *2.1. Reactor design*

The electrocoagulation–electroflotation cell shown in Fig. 1 consists of a rectangular tank with a total volume of 8.6 L. This is divided into two compartments. The first compartment of volume $V_1 = 3.1$ L is provided with two aluminum electrodes of rectangular shape (240 mm × 20 mm × 1 mm), used as anode and cathode, which corresponds to $S = 48$ cm$^2$ electrode surface area. The distance between the two electrodes was $e = 10$ mm, which is a typical value in EC cells (see, for example, [19,29]). These were treated with an HCl aqueous solution for cleaning prior use to avoid passivation. The electrodes were placed at the bottom in the middle of the first compartment, as shown in Fig. 1. The length of the electrodes was maximized to reduce their height, taking into account both the volume of the first compartment and the gap between the electrodes. This design was retained to enhance mixing in the first compartment, using both the liquid flow and $H_2$ microbubbles in this purpose. Indeed, the microbubbles formed at electrodes, the liquid phase and the solid particles flowed co-currently in this compartment. Large electrodes presented therefore the advantage to maximize the formation of $H_2$ microbubbles, which promoted a homogenous dispersion of the gas phase above the electrodes and also favored floc removal by electroflotation in the second compartment: the EC cell was fed continuously with a peristaltic pump using the effluents from a wastewater tank. Residence time ($\tau$) could be varied by adjusting the liquid flow rate ($Q$) between 25 and 78 L/h. The two compartments were connected by a triangular groove (Fig. 1) and the wastewater passed from the first compartment to the second one by overflow. The volume $V_2 = 5.5$ L of the second compartment and the position of outlet tube were designed to avoid the presence of solid particles in the effluent stream. The sludge was essentially recovered by flotation. The floating materials were removed continuously from the second compartment by overflow, whereas the few more dense materials could settle. EC was always conducted in the intensiostat mode, using a digital DC power supply (Didalab, France) and recording cell potential ($U$) during the experiments. Current density ($j$) was varied between 20.8 and 62.5 mA/cm$^2$, which corresponded to current values ($I$) between 1 and 3 A. All the runs were performed at room temperature.

### *2.2. Chemicals and methods*

Experiments were carried out using a red dye solution consisting of a mixture of 2-naphthoic acid and 2-naphtol (Fig. 2) with a total concentration ($C_i$) that could be varied between 25 and

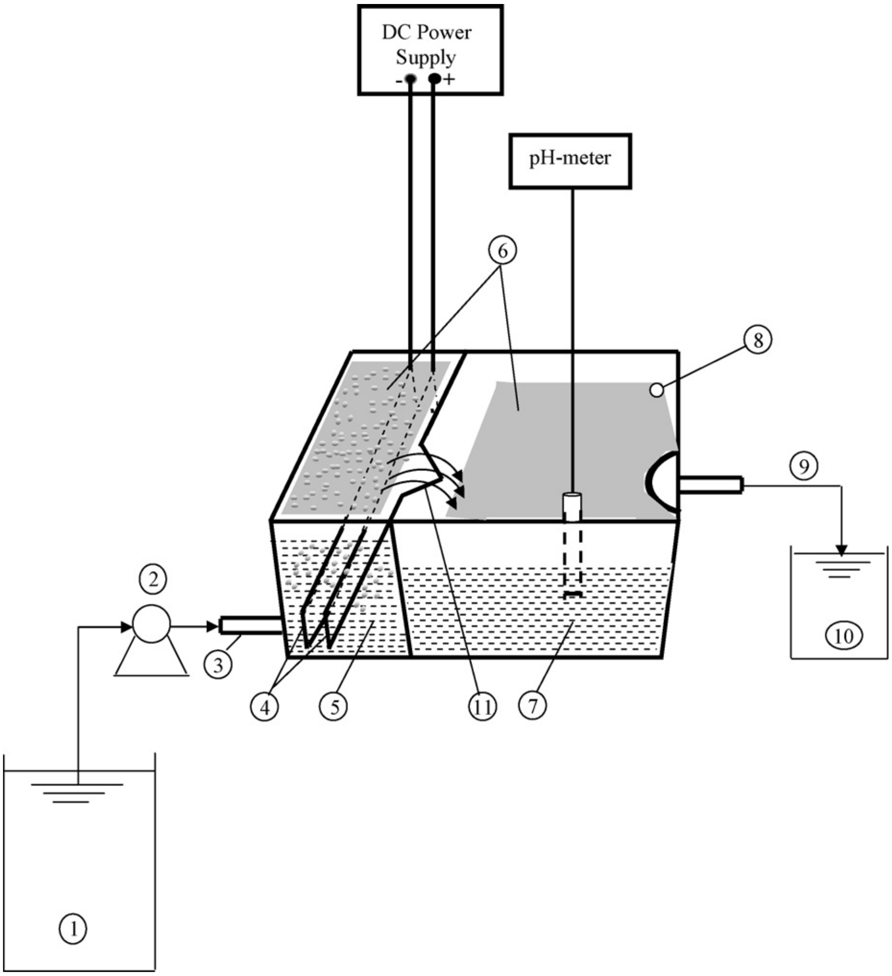

**Fig. 1.** Experimental setup of continuous EC cell (1: wastewater tank; 2: peristaltic pump; 3: inlet of the first compartment; 4: electrodes; 5: first compartment; 6: sludge; 7: second compartment; 8: sludge exit (overflow); 9: treated effluent outlet; 10: treated water tank; 11: outfall).

200 mg/L. This mixture belongs to the dispersive dye class [2]. This class groups nearly insoluble organic and non-ionic compounds that are however applied in aqueous solution using the simple immersion technique [2]. This dye and its concentration are typical of wastewater from a Moroccan textile factory. Synthetic solutions were prepared by mixing the dye in tap water in the wastewater tank of Fig. 1. Solution conductivity and pH were measured using a CD810 conductimeter (Radiometer Analytical, France) and a ProfilLine pH197i pHmeter (WTW, Germany). Dye concentration was estimated from its absorbance characteristics in the UV–vis range (200–800 nm), using the wavelength that provided the maximum intensity ($\lambda_{max} = 450$ nm) and a UV–vis spectrophotometer (Pye Unicam, SP 8–400, UK). COD was measured using the standard closed reflux colorimetric method. COD in the inlet stream was about 2500 mg/L for $C_i = 100$ mg/L. Influent pH was varied between 3.1 and 9.1 using the minute addition of 0.1 M $H_2SO_4$ or NaOH solutions [2,7,30,31]. The conductivity $\kappa$ (i.e. the ionic strength) of wastewater was adjusted using the addition of sodium chloride in the range 1.5–5 mg/L, which covers the range usually explored in the literature (see, e.g., [8,19,29]). NaCl is a salt exhibiting low toxicity at moderate level, reasonable cost, high conductivity and high solubility, but it plays also the role of supporting electrolyte. This addition had always a negligible effect on the influent pH of the solutions.

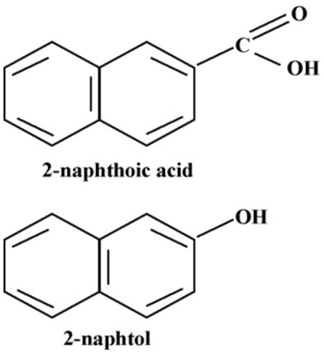

**Fig. 2.** Molecular structure of the constituents of the red dye.

During EC process, absorbance was measured over time on samples recovered from the exit stream of the reactor. Absorbance at 450 nm ($A_{450}$) was measured every 2 min in order to determine the time required for the establishment of steady-state conditions. COD was measured only when steady-state conditions were established. All the analyses were conducted at room temperature. COD and color removal efficiencies ($Y_{COD}$, $Y_{COL}$) were expressed as a percentage and defined as

$$Y_{COL}(\%) = \frac{C_i - C_f}{C_i} \times 100 \qquad (4)$$

$$Y_{\mathrm{COD}}(\%) = \frac{\mathrm{COD_i} - \mathrm{COD_f}}{\mathrm{COD_i}} \times 100 \tag{5}$$

In Eq. (4), $C_i$ and $C_f$ are the dye concentrations expressed in kg/L in the inlet and the exit streams of the electrocoagulation cell, respectively. The same subscripts, “i” and “f”, are used to distinguish COD in the inlet and the outlet streams in Eq. (5).

The specific electrical energy consumption per kg dye removed ($E_{\mathrm{dye}}$) and the specific electrode consumption per kg dye ($\mu_{\mathrm{Al}}$) under steady-state conditions were calculated as follows

$$E_{\mathrm{dye}}\left(\frac{\mathrm{kWh}}{\mathrm{kg\,dye}}\right) = \frac{\mathrm{UI}}{1000\,Q\,(C_i Y_{\mathrm{COL}})} \tag{6}$$

$$\mu_{\mathrm{Al}}\left(\frac{\mathrm{kg\,Al}}{\mathrm{kg\,dye}}\right) = \frac{3600\,\mathrm{M_{Al}}I\,\phi_{\mathrm{Al}}}{3F}\,\frac{1}{Q\,(C_i Y_{\mathrm{COL}})} \tag{7}$$

using the dye concentration in the inlet stream $C_i$ (kg/m$^3$), current intensity $I$ (A), cell voltage $U$ (V), liquid flow rate $Q$ (L/h), molar weight of aluminum $M_{\mathrm{Al}} = 0.02698$ kg/mol, Faraday's constant $F$ (96,487 C/mol e$^-$) and the faradic yield $\phi_{\mathrm{Al}}$ of Al dissolution. $\phi_{\mathrm{Al}}$ was estimated as the ratio of the weight loss of the aluminum electrodes during the experiments $\Delta m_{\mathrm{exp}}$ and the amount of aluminum consumed theoretically at the anode $\Delta m_{\mathrm{th}}$:

$$\phi_{\mathrm{Al}} = \frac{\Delta m_{\mathrm{exp}}}{\Delta m_{\mathrm{th}}} \tag{8}$$

This parameter depends upon the pH and the amount of other soluble species, for example co-existing anions [32].

## 3. Results and discussion

### 3.1. Effect of current density on the color removal efficiency

In all electrochemical processes, current density is the most important parameter for controlling the reaction rate within the electrochemical reactor [33]. It is well known that current density determines the production rate of coagulant, adjusts also bubble production, and hence affects the growth of flocs [4,5]. To investigate the effect of current density $j$ on the efficiency of color removal, continuous electrocoagulation process was carried out using various current densities at fixed inlet concentration $C_i = 100$ mg/L with pH values between 7.7 and 8.0 and an inlet flow rate $Q = 25.2$ L/h. The data monitored for 32 min operation is presented in Fig. 3.

Fig. 3 shows that the time required to achieve steady-state conditions, i.e. the values on the plateau region, decreased when $j$ increased from 20.83 to 31.25 mA/cm$^2$ and then became nearly constant at about 12 min. An increase in current density from 20.83 to 62.5 mA/cm$^2$ yielded an increase in the efficiency of color removal from 78 to 93%. This could be expected: when the current density increases, the amount of Al$^{3+}$ cations released by the anode and therefore of Al(OH)$_3$ particles also increases. However, the difference between the curves corresponding to different current densities became small when $j$ was higher than 31.25 mA/cm$^2$. An optimum current density can therefore be defined. Indeed, when the increase of $Y_{\mathrm{COL}}$ with $j$ is slight, the disadvantages of working at high current densities prevail: first, the amount of sludge increases typically as $j$; then, the same stands for electrode material consumption, while energy consumption rises as $j^2$ and induces heating by Joule effect. These trends were confirmed experimentally in Figs. 4 and 5 for $E_{\mathrm{dye}}$ and $\mu_{\mathrm{Al}}$, respectively, and they agree qualitatively with data already reported in the literature for textile dye wastewaters [4,7,29,34–38]. In particular, the electrode consumption per kg dye should be minimized because it can constitute up to 80% of the total operating costs for aluminum electrodes, according to Bayramoglu et al. [38].

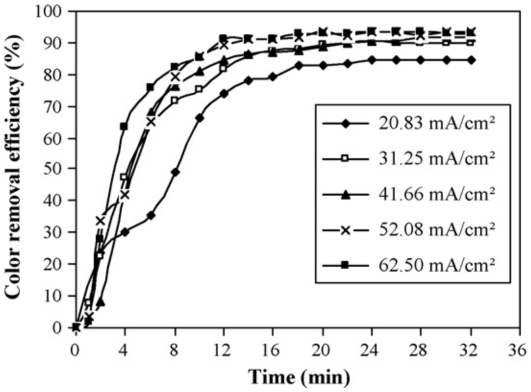

**Fig. 3.** Effect of current density $j$ on the color removal efficiency: $C_i = 100$ mg/L, influent pH 7.74, $Q = 25.2$ L/h, $\kappa = 2.4$ mS/cm.

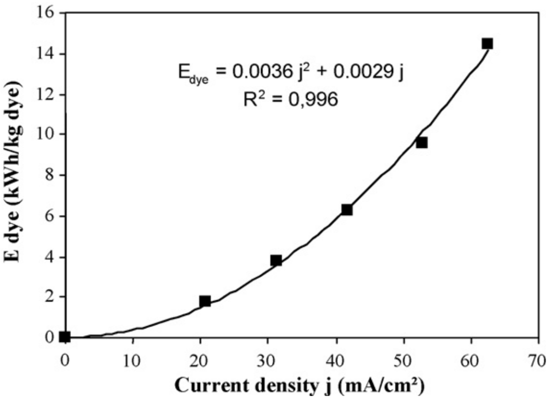

**Fig. 4.** Effect of current density $j$ on energy consumption ($E_{\mathrm{dye}}$) under steady-state conditions: $C_i = 100$ mg/L, influent pH 6.15, $Q = 37.2$ L/h, $\kappa = 2.4$ mS/cm.

For the experimental conditions of Fig. 3, optimum current density seemed therefore to be between 30 and 40 mA/cm$^2$, as it can be considered that $Y_{\mathrm{COL}} = 80\%$ corresponds to the minimum acceptable value for EC process. Consequently, $j = 31.25$ mA/cm$^2$ was retained for further experiments, which corresponded to a current value of 1.5 A, because it minimized energy consumption. This value ensured indeed color and COD removal efficiencies higher than 80%, while $E_{\mathrm{dye}}$ and $\mu_{\mathrm{Al}}$ values were similar or even lower than those reported by several authors in the literature [34–36].

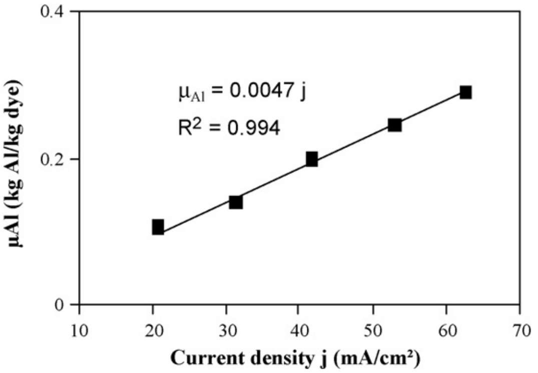

**Fig. 5.** Effect of current density $j$ on electrode consumption ($\mu_{\mathrm{Al}}$) under steady-state conditions: $C_i = 100$ mg/L, influent pH 6.15, $Q = 37.2$ L/h, $\kappa = 2.4$ mS/cm.

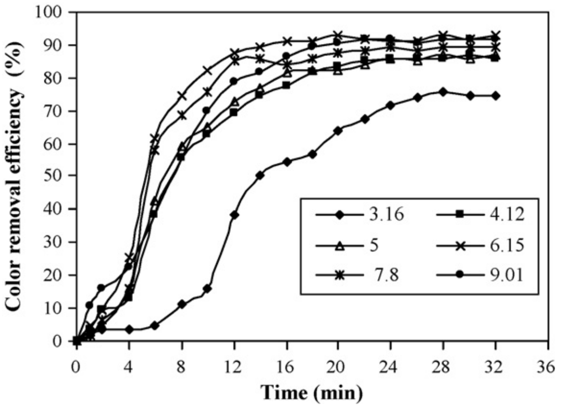

**Fig. 6.** Effect of the influent pH on the color removal efficiency: $C_i = 100$ mg/L, $j = 31.25$ mA/cm$^2$, $Q = 25.2$ L/h, $\kappa = 2.4$ mS/cm.

### 3.2. Effect of influent pH on the color removal efficiency

It has already been established that pH is a key operating factor influencing the performance of electrocoagulation process [4,7,29,34,37,38]. To examine its effect, the pH of the wastewater was adjusted to the desired value for each experiment by adding either sodium hydroxide or sulfuric acid. Fig. 6 demonstrates that color removal was poor at pH 3.16, but remained between 80 and 90% when pH was varied between 4.1 and 9.0. The maximum efficiency of color removal was observed both at pH 6.15 and 9.0. However, steady-state conditions were achieved more rapidly and $Y_{COL}$ was higher in the initial transient period when pH was between 6 and 8. This difference could be explained by monitoring the pH of the outlet stream (Fig. 7): pH in the outlet stream was similar to the influent pH when this was 6.15, whereas it was lower than the influent pH when this was 9.01. Indeed, pH is known to change during EC under batch conditions, as already described in many already cited papers [4,7,29,34,36–38]. EC process exhibits some buffering capacity because of the balance between the production and the consumption of OH$^-$ and the need for charge neutralization before the final transformation of soluble Al compounds into Al(OH)$_3$ particles [5]. These prevent high change in pH from neutrality, as in Fig. 7. Indeed, the buffering pH is usually about 7, which is the case in this work (about 6.3), although it may depend on other soluble species in the aqueous medium.

The effect of pH can be explained as follows. The main reactions during EC are:

$$\text{Anode}: \ \mathrm{Al}^{0}_{(S)} \rightarrow \mathrm{Al}^{3+} + 3e^- \tag{9}$$

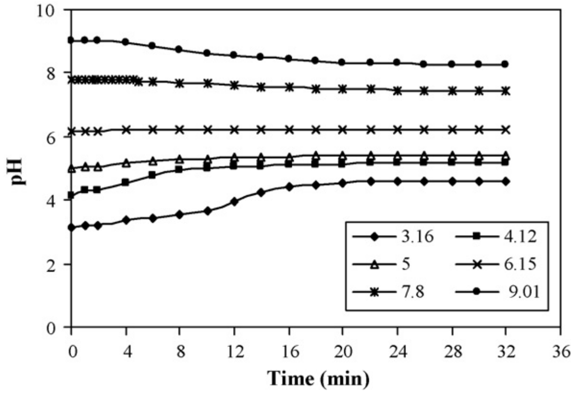

**Fig. 7.** Evolution of pH values during EC for different values of influent pH: $C_i = 100$ mg/L, $j = 31.25$ mA/cm$^2$, $Q = 25.2$ L/h, $\kappa = 2.4$ mS/cm.

$$\text{Cathode}: \ 2\mathrm{H_2O} + 2e^- \rightarrow \mathrm{H}_{2(g)} + 2\mathrm{OH}^- \tag{10}$$

At low pH, such as 2–3, cationic monomeric species Al$^{3+}$ and Al(OH)$_2^+$ predominate. 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 us Al$_6$(OH)$_{15}^{3+}$, Al$_7$(OH)$_{17}^{4+}$, Al$_{13}$(OH)$_{34}^{5+}$ that finally transform into insoluble amorphous Al(OH)$_{3(s)}$ through complex polymerization/precipitation kinetics [38]. When pH is higher than 10, the monomeric Al(OH)$_4^-$ anion concentration increases at the expense of Al(OH)$_{3(s)}$. In addition, the cathode may be chemically attacked by OH$^-$ ions generated together with H$_2$ at high pH values [30]:

$$2\mathrm{Al} + 6\mathrm{H_2O} + 2\mathrm{OH}^- \rightarrow 2\mathrm{Al(OH)}_4^- + 3\mathrm{H}_2 \tag{11}$$

Two main mechanisms are generally considered to explain pollution removal: *precipitation* for pH lower than 4 and *adsorption* for higher pH. Adsorption may proceed on Al(OH)$_3$ or on the monomeric Al(OH)$_4^-$ anion, depending on the dye chemical structure. The formation of Al(OH)$_{3(s)}$ is therefore optimal in the 4–9 pH range. However, pH affects also bubble size [39]. Typical bubble sizes in electrocoagulation always fall in the range of 20–70 $\mu$m [9]. They are far smaller than those observed in conventional air-assisted flotation, which provides both sufficient surface area for gas–liquid–solid interfaces and mixing efficiency to favor the aggregation of tiny destabilized particles. Hydrogen bubbles, which usually obey to a lognormal size distribution, are also known to be the smallest about neutral pH [40].

Figs. 6 and 7 confirm the above-mentioned trends. As expected, the evolution of $Y_{COL}$ depended on the influent pH, but the discrepancies between different influent pH values were smaller than in batch conditions, as compared to Essadki et al. [36]. Although the key mechanism of pollution removal was undoubtedly adsorption, the presence of two maxima for $Y_{COL}$ values at pH 6.1 and 9.0, respectively, shows that dye removal by EC could result in fact from several complex mechanisms. Under continuous flow conditions at constant residence time and $j$, the main effect of working with an influent pH far from the buffering pH value appeared to be an additional delay for the establishment of steady-state conditions.

As a conclusion, pH can be adjusted in the optimum range to favor color removal by continuous EC process. The optimum must be chosen in order to achieve a compromise between best coagulation and best flotation conditions, while minimizing soluble Al content in the effluents and maintaining simultaneously pH close to neutrality in order to avoid a downstream treatment for pH adjustment. For these reasons, pH 6.1 was preferred and has been used in the next sections.

### 3.3. Effect of inlet flow rate on the color removal efficiency

In order to investigate the effect of the residence time on the color removal efficiency, the inlet flow rate $Q$ was progressively increased from 25.2 L/h to 30.6, 37.2, 48, 60 and 78 L/h, respectively, for $j = 31.25$ mA/cm$^2$ and influent pH 6.1. As shown in Fig. 8, the continuous EC process provided a color removal efficiency higher than 80% for all the conditions studied, except for $Q = 78$ L/h. Fig. 8 shows also that $Y_{COL}$ passed through a maximum about 37.2 L/h. Indeed, increasing $Q$ presented the advantage to enhance mixing in the first compartment. Beyond 37.2 L/h, the effect of mixing could no more compensate the influence of the decrease in residence time on the kinetics of color removal. The optimal inlet flow rate could therefore be considered to be 37.2 L/h. This flow rate corresponds to a residence time of about 14 min in the EC cell, divided into two parts: 5 min in the first compartment and 9 min in the second one. On the basis of the literature [2,8,12,19,25,27,28,41], it must also be

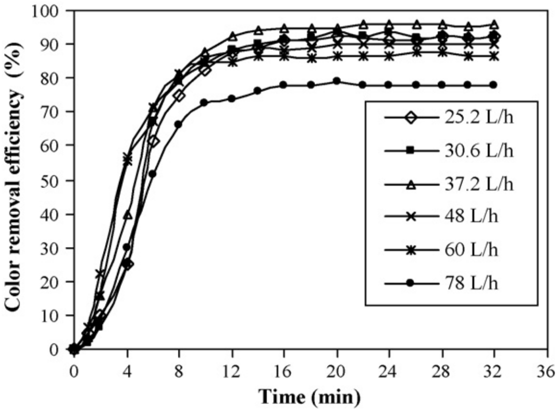

**Fig. 8.** Effect of inlet flow rate on the color removal efficiency: $C_i = 100$ mg/L, $j = 31.25$ mA/cm$^2$, influent pH 6.15, $\kappa = 2.4$ mS/cm.

mentioned that the residence time in the first compartment of this paper (i.e. the only one containing the electrodes) is usually lower than the values usually reported in EC cells used for decolorization.

### 3.4. *Effect of influent concentration on the color removal efficiency*

Fig. 9 illustrates the evolution of removal efficiency of color removal as a function of the influent dye concentration, using the optimum conditions obtained previously for current, residence time and pH. In this figure, dye solutions with different concentrations in the range 25–200 mg/L were treated by continuous EC process. The data shows that the continuous EC process gave satisfactory results for all the concentrations studied, as $Y_{COL}$ was always higher than 80% in the exit stream of the EC cell. As the dye concentration was increased from 25 to 100 mg/L, not only the dye removal yield increased, but the dye concentration in the effluents $C_f$ also decreased from about 11 to 4 mg/L. This behavior may stem both from a thermodynamic limitation resulting from the shape of the adsorption isotherm and from a kinetic limitation at low $C_i$ values. In this work, a kinetic limitation seems however the most probable because of the dye concentrations used: when $C_i$ decreases, longer residence times are required to achieve dye removal. Conversely, $Y_{COL}$ decreased from 96 to 89% when $C_i$ was increased from 100 to 200 mg/L (Fig. 9) and $C_f$ doubled approximately. This shows clearly that the adsorption capacity of the flocs became exhausted when $C_i$ was higher than 100 mg/L, as the total amount of sludge was constant for all concentrations at constant current density. Similar results have already been reported in the literature [2,26,37]. As a conclusion, the EC process is robust because it can achieve more than 80% color removal for $C_i$ between 25 and 200 mg/L, but the $j$ and $Q$ values defined previously are optimum for $C_i$ between 80 and 100 mg/L.

### 3.5. *Effect of EC process on COD in the effluent stream*

The COD of the influent dye solution was 2500 mg/L for $C_i = 100$ mg/L. During the EC process under the following conditions (pH ranging from 4.5 to 8.5, 14 min residence time, $j = 31.25$ mA/cm$^2$, inter-electrode distance of 1 cm and conductivity $\kappa = 2.4$ mS/cm), the COD abatement was always higher than 80%, as in Essadki et al. [36]. Under steady-state conditions, the observed trends always followed those of $Y_{COL}$ and they will not be reported in detail.

### 3.6. *Effect of wastewater conductivity on the color removal efficiency*

Sodium chloride (NaCl) is usually employed to increase the conductivity of the water or wastewater to be treated. Wastewater conductivity affects faradic yield, cell voltage and therefore energy consumption in EC cells. In addition, increasing water conductivity using NaCl has other advantages: e.g., chloride anions could significantly reduce the adverse effects of other anions, such as $HCO_3^-$ and $SO_4^{2-}$ [2,42]. Indeed, the existence of carbonate anion would lead to the precipitation of $Ca^{2+}$ ion. These can form an insulating layer on the surface of the cathode (Eqs. (12) and (13)), which could increase the ohmic resistance of the electrochemical cell [9,32,39].

$$HCO_3^- + OH^- \rightarrow CO_3^{2-} + H_2O \qquad (12)$$

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \qquad (13)$$

Conversely, an excessive amount of NaCl induces an overconsumption of the aluminum electrodes due to “corrosion pitting”; Al dissolution may become irregular [42]. This is the reason why NaCl addition has been limited to 5 g/L. Additionally, it is important to monitor how the presence of NaCl affects the color removal efficiency. Indeed, $Y_{COL}$ usually increases with the addition of NaCl [39], but this result is not general [9,32,43].

In this work, the conductivity of wastewater was adjusted using NaCl in the range 1500–5000 mg/L for three current densities (10.4, 20.8 and the optimum value without NaCl addition: 31.25 mA/cm$^2$); the EC process was applied as before on the three wastewaters with different conductivities. The results are presented in Fig. 10: this illustrates the evolution of the color removal efficiency with

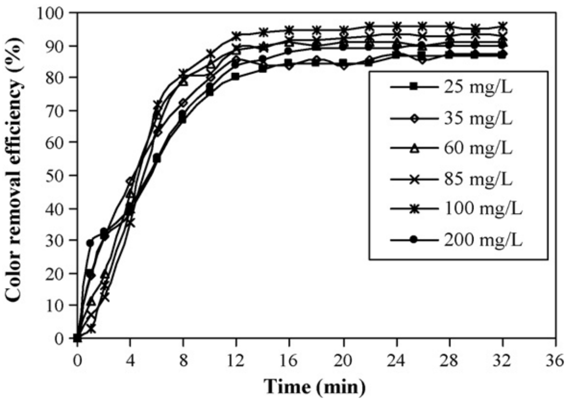

**Fig. 9.** Effect of influent concentration on the color removal efficiency: $C_i = 100$ mg/L, $j = 31.25$ mA/cm$^2$, influent pH 6.15, $Q = 37.2$ L/h, $\kappa = 2.4$ mS/cm.

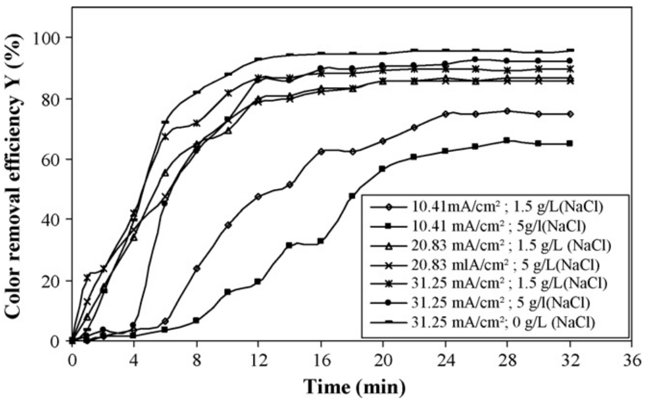

**Fig. 10.** Effect of NaCl addition on the color removal efficiency for different values of current density $j$: $C_i = 100$ mg/L, influent pH 6.15, $Q = 37.2$ L/h.

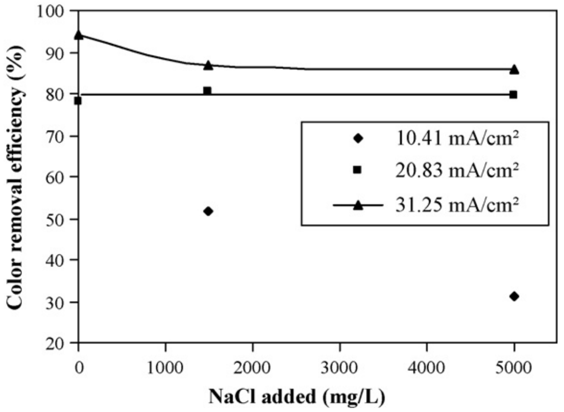

**Fig. 11.** Effect of NaCl addition on the color removal efficiency for different values of current density *j* under steady-state conditions $C_i$ = 100 mg/L, residence time $\tau$ = 14 min, influent pH 6.15, *Q* = 37.2 L/h.

time as a function of electrolyte concentration. This shows that $Y_{COL}$ decreased when NaCl concentration was increased. For example, for *j* = 10.41 mA/cm$^2$, $Y_{COL}$ was significantly reduced, from 53 to 33%, when NaCl content was increased from 1.5 to 5.0 g/L. Under steady-state conditions, this trend appeared more clearly in Fig. 11. At higher current, the discrepancies due to NaCl were slighter: in other words, increasing the conductivity of wastewaters had not a considerable effect on color removal efficiency, but the negative influence of conductivity on $Y_{COL}$ was however confirmed at high current density (Fig. 11). For example, it seemed insignificant for *j* = 20.83 mA/cm$^2$ and an increase of NaCl concentration from 0 to 5 g/L; in this case, $Y_{COL}$ remained around 79%, regardless of conductivity, but for *j* = 31.25 mA/cm$^2$, $Y_{COL}$ decreased slightly when salt concentration was increased from 0 to 1.5 g/L. However, a further addition of NaCl appeared again to have no significant effect on color removal.

As a conclusion, working without NaCl could be the best solution in order to maximize dye removal. One can first remark that this behavior differs from several results from the literature [2,37], which highlights that the influence of NaCl addition depends on the type of dye that has to be treated. However, it must be mentioned that the absence of supporting electrolyte is never the optimum situation from an energetic point of view. This is illustrated by Fig. 12 that reports the evolution of specific energy consumption as a function of NaCl addition. In terms of $E_{dye}$, it is obvious from Fig. 12 that electrolyte addition was beneficial for the continuous EC process, as in batch process, except when current density was too low (*j* = 10.8 mA/cm$^2$). For higher current density, $E_{dye}$ measured without NaCl addition was about twice higher than when 5 g/L of NaCl were added. For *j* values equal to or higher than 20.8 mA/cm$^2$, another advantage was that the increase of specific electrode consumption with NaCl addition remained slight, which limited the increase in operating costs.

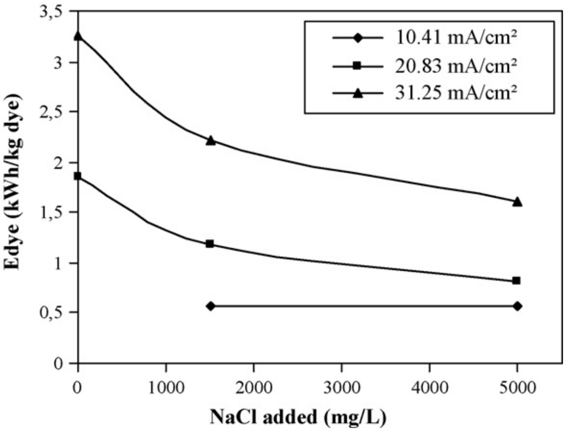

**Fig. 12.** Effect of NaCl addition on energy consumption $E_{dye}$ for different values of current density *j*: $C_i$ = 100 mg/L, residence time $\tau$ = 14 min, influent pH 6.15, *Q* = 37.2 L/h.

As a result, NaCl addition may be used as an adjustable parameter that depends on the objectives of the operator: when maximizing removal efficiency is the point, it must be avoided; when it is possible to accept $Y_{COL}$ values a bit smaller, it constitutes a good way for minimizing operating costs.

## 4. Conclusions

A series of experiments was performed in order to find the effects of operating parameters for COD abatement and decolorization of textile dye wastewater by electrocoagulation under steady-state conditions. Dye removal by continuous EC process was affected by current density, influent pH, influent dye concentration and electrolyte concentration, as in batch process, but also by residence time that played the role dedicated to operation time in batch operation. However, sludge separation by flotation was always achieved with the reactor geometry used as EC cell in this work. Our results emphasize the opportunity to apply continuous EC process for the efficient removal of color and organic pollutants from wastewater at low operating costs. The results showed that for an influent dye concentration up to 200 mg/L with an influent pH adjusted at about 6, the use of a current density of 31.25 mA/cm$^2$, a residence time of 14 min (but only 5 min in the first compartment including the electrodes), a distance between electrodes of 1 cm and a water conductivity of 2.4 mS/cm, gave access to a dye removal efficiency between 85 and 95%. Optimum conditions, i.e. $Y_{COL}$ = 95%, were achieved when $C_i$ was between 80 and 100 mg/L with a specific energy consumption about 3.2 kWh per kg of dye removed. This value could still be halved by rising water conductivity, using the addition of NaCl, but at the expense of a slight decrease in dye removal efficiency. With the above-mentioned conditions, the COD abatement was always higher than 80%. Finally, it must be mentioned that regardless of NaCl addition, the values of $E_{dye}$ reported in Section 3.6 for the optimum conditions (between 1.6 and 3.2 kWh/kg dye) are particularly low if one takes into account that no mechanical agitation is required. This highlights the high effectiveness of continuous EC process for the removal of the dye used in this work.

## Acknowledgements

The authors would like to express their gratitude to the High School Technology of Casablanca, Morocco, in which the experiments were conducted and to the University of M'sila, Algeria, for its financial support and assistance.

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