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Influence of electrodes connection mode and type of current in electrocoagulation process on the removal of a textile dye

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ABSTRACT

The main objective of this study was to investigate the operation of a continuous electrochemical reactor comprising an electrocoagulation part provided with aluminium electrodes and a settling zone, for the removal of a red nylosan dye (Acid Red 336). More precisely the effect of the operating parameters has been followed, e.g. initial pH, initial concentration (C_0), residence time (τ), current density (j), inlet flow rate (Q), direct/ alternating current (DC/AC) and electrode connection mode. Abatement yields between 87% and 96% for color and turbidity were observed with direct current, for dye concentration ranging from 100 mg L^{-1} to 1000 mg L^{-1}, at 300 A/m², for inlet flow rate Q = 15 L h^{-1} and 26 L/h. The specific electrical energy consumption was found at 9.5 kWh kg^{-1} removed dye for direct current and monopolar connection. At the same current density, bipolar connection with four electrodes immersed is more effective than monopolar connection in terms of color and turbidity abatement, but with far larger energy consumption. The performance difference between the two modes at comparable energy consumption is discussed.

Keywords: Electrocoagulation; Color; Turbidity; Red nylosan dye; Elecrode connection modes; Alternating/direct current

1. Introduction

Textile and dyestuff industries are the primary sources of dye emission into the environment. The dyeing step in textile production has the largest risk for the environment due to high concentrations of organic dyes, additives and salts used [1,2]. In most cases, this process represents the major part of the water consumption and generates wastewaters characterized by high chemical oxygen demand (COD), high dissolved and suspended solids, and high color contents [3,4].

Dyes represent a major part of the pollution problem as it is estimated that 50 % of their amount is not fixed on fibers and remain free in the wastewater generated. Due

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to the large presence of aryl groups in the dyes and their chemical stability, treatment methods for their removal e.g. adsorption, chemical coagulation, activated carbon, electrochemical oxidation, microfiltration, nanofiltration and biological treatment have been developed [5–10]. Most of these methods can be insufficient and often consist of simple contaminant transfer from one phase to another in the overall treatment process [11]. Although ensuring only the separation of pollutant waste, electrocoagulation (EC) process can be of large interest, because of the moderate energy and cost requirements [12–14].

In EC, the action of the electrical current between two electrodes allows the formation of metal ions by oxidation of the sacrificial anode. In a suitable pH range, insoluble metal hydroxides are generated from the ions and can then remove pollutants. The treatment can be achieved with aluminium or iron electrodes without addition of chemical flocculants or coagulants.

With aluminium anodes, the reactions involved are as follows:

$$\text{At the anode: } Al_{(s)} \rightarrow Al^{3+} + 3e^- \quad (1)$$

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

$$\text{In the solution: } Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad (3)$$

Highly charged cations destabilize the colloidal particles by the formation of polyvalent polyhydroxide complexes. These complexes have high adsorption properties, forming aggregates with pollutants. Once the floc is generated, the electrogenerated hydrogen bubbles attach the flocs and allows their flotation toward the liquid surface. The size distribution of hydrogen bubbles depends on pH, current density, electrode material, and cell arrangement [15–18].

EC technique has successfully been used for the treatment of waters containing suspended solids [19], various organic matters [20–22], heavy metals [16,23], arsenic [24,25], fluoride [26,28], phosphate [29,30] and wastewaters from tannery [31], dairy [32,33] and textile industry [12,13,15,34–38]. Most of these studies focus on the elimination of a pollutant in a conventional EC system (batch process, with monopolar electrodes, low concentrations and direct current). However few papers focused on continuous EC [17,30,39–42], in most cases with low inlet concentrations and flow rates, other electrical connection systems as with bipolar electrodes [43–46] and nature of the current applied [47–48].

The objective of this paper is to investigate the above operating conditions in view to improving the efficiency of the continuous treatment. In this study, wide ranges of nylosan dye initial concentrations (100–1000 mg L^{-1}) and inlet flow rates (15–60 L h^{-1}, corresponding to residence times ranging from 9 to 35 min) have been investigated. Moreover, alternating current (AC) mode has been tested for the treatment, in comparison with conventional direct current (DC) method. Besides, monopolar (MP) and bipolar (BP) electrode connation was examined, with discussion of both Al and energy consumption and treatment efficiency.

2. Materials and methods

2.1. Synthetic wastewater

The dye (red nylosan) was supplied by Tindal Company (Algeria). Its chemical formula is shown in Fig. 1. The main characteristics of this dye are given in Table 1. Experiments were carried out using a red nylosan dye N-2RBL (sodium 6-amino-5-[[4-chloro-3-[[(2,4-dimethylphenyl) amino] sulphonyl] phenyl]azo]-4-hydroxynaphthalene-2-sulphonate). The synthetic wastewater has been prepared by dissolving the dye in distilled water in the wastewater tank of Fig. 2, and has been diluted according to the desired concentration (100–1000 mg L^{-1}). The pH and the conductivity were adjusted to their desired levels with NaOH or HCl and NaCl respectively: the inlet conductivity was fixed at 2.5 mS cm^{-1} in all cases. These values were chosen to emulate the properties of wastewater issued from a textile industry in Algeria [39].

2.2. Reactor design

The continuous EC cell shown in Fig. 2 consists of a rectangular tank for both metal dissolution and solid settling. The reactor machined out of methyl polymethacrylate comprises a 3 L electrochemical chamber and a larger settling/flotation tank being 5.5 L in volume. The waste liquid (1) is pumped to the electrochemical chamber (4) provided with aluminium (98 % pure Al) plate anodes and cathodes (dimensions 240 × 20 × 10 mm) used as vertical Al electrodes. The area of one electrode side was 48 cm^2. In the cell, the fluid leaves the chamber by continuous overflow and enters gently the settling section (5). The electrodes were

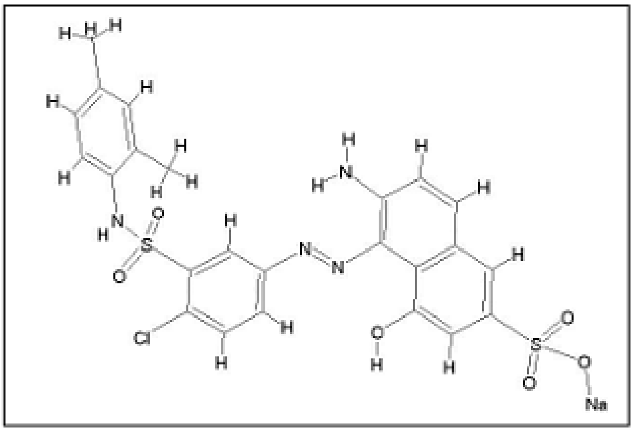


Fig. 1. The chemical structure of red nylosan dye.

Table 1
Main characteristics of Red nylosan dye

Color index number	Acid Red 336
CAS registry number	71873-39-7
Molecular formula	$C_{24}H_{21}ClN_4O_6S_2$, Na
Molecular weight	587.97 g mol^{-1}
UV absorption	λ_{max} 502 nm

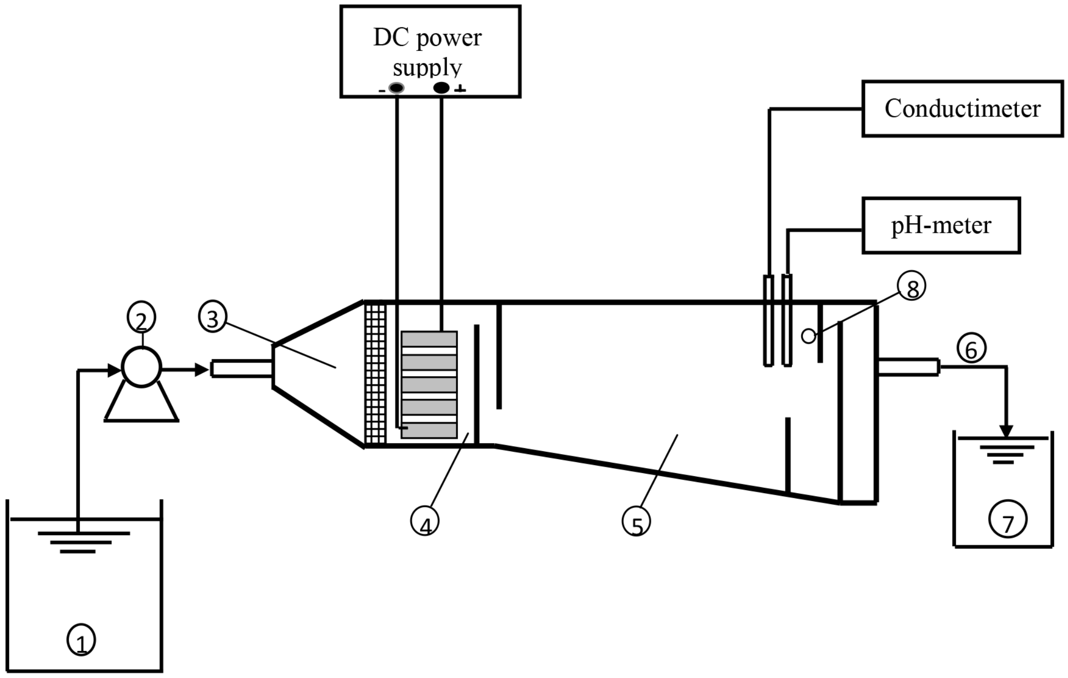


Fig. 2. Experimental setup of continuous EC cell. (1) wastewater tank, (2) peristaltic pump, (3) inlet section, (4) EC cell, (5) sedimentation-flotation chamber, (6) outlet section, (7) treated water tank, (8) sludge evacuation.

fully immersed in an aqueous solution in the transparent reactor. The gap between the two neighboring electrode plates was kept constant at 10 mm for all the experiments as suggested before [39]. Regulated direct current (DC) was supplied from a rectifier (10 A, 0–30 V; GPS3303$_{3CH}$) whereas for alternating current (AC) a source (0–5 A, 0–270V, 50 Hz; EC1000S) was used. Time *t* was counted from the switch on of the current generator. Two connection modes of the electrodes were tested as illustrated in Fig. 3. A monopolar configuration with two electrodes (MP) and a bipolar configuration with four electrodes (BP) were used as shown in Fig. 3a and 3b respectively. This configuration is more representative of continuous industrial reactors in which the numerous bipolar electrodes inserted form a stack of individual cells in series. For MP configuration, Al dissolution occurs in a single-cell system, whereas electrochemical reactions occur in three cells formed by the four electrodes in BP configuration.

The sludge was essentially recovered by flotation. The floating materials were removed continuously from the second compartment by overflow (8), whereas the denser materials could settle. The solution to be treated was continuously circulated in the flow circuit by means of a peristaltic pump (Heidolph PD 5006, Pump drive) allowing flow rates up to 60 L h^{-1}.

2.3. Analytical methods

Samples (40 mL) were collected from the reservoir (7) every 5 min until steady state was attained. In practice, the reactor reaches steady state conditions within at least one residence time [39]. Dye concentration was estimated from its absorbance characteristics in the UV–vis range (250–800 nm), using the wavelength providing the maximum intensity (l_{max} = 502 nm) and a UV–vis spectrophotometer (Shimadzu-1240 S). Solution conductivity and pH were measured using a Hanna EC214 conductimeter and a Hanna pH210 pH-meter respectively. The UV–visible spectra of the filtered samples were recorded with an Anthelie Light (Secomam, Domont, France) spectrophotometer in the range 200–600 nm using 1 cm-path quartz cells. Turbidity of the waters was measured using a Hanna HI88703 spectrophotometer. Data were given in nephelometric turbidity unit (NTU).

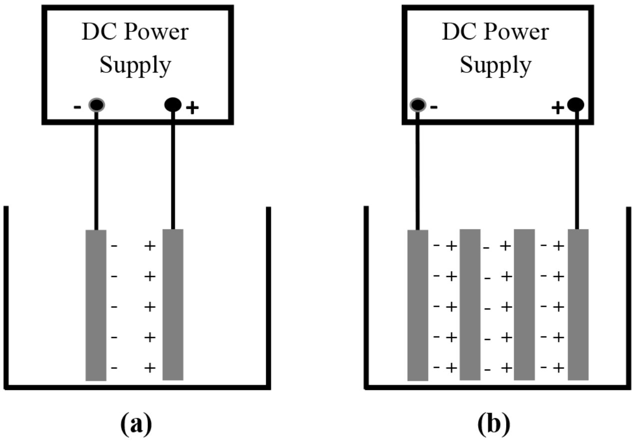


Fig. 3. Electrodes connection configurations.

Absorbance and turbidity removal efficiencies (Y_{Col}, Y_{Tr}) expressing the visual aspects of the solution were defined in percents as:

$$Y(\%) = \frac{C_0 - C_f}{C_0} \times 100 \tag{4}$$

Subscripts "0" and "*f*" are respectively for the concentrations in the inlet and the outlet streams of the electrocoagulation cell. The specific electrical energy consumption per kg dye removed and per m^3 wastewater treated (E_{dye} and E_{water} respectively) together with the specific electrode consumption per kg dye (μ_{Al}) were calculated from the liquid absorbance as follows:

$$E_{dye}\left(\frac{kWh}{kg\ dye}\right) = \frac{UI}{1000Q\left(C_0\, Y_{COL}\right)} \tag{5}$$

$$E_{water}\left(\frac{kWh}{m^3}\right) = \frac{UI\tau}{V} \tag{6}$$

$$\mu_{Al}\left(\frac{\text{kg Al}}{\text{kg dye}}\right) = \frac{3600\, M_{Al}\, I\, \varphi_{Al}}{3F}\frac{1}{Q\left(C_0\, Y_{COL}\right)} \tag{7}$$

with initial dye concentration C_0 (kg m^{-3}), current intensity *I* (A), cell voltage *U* (V), residence time *t* (h), liquid volume *V* (m^3), molar weight of aluminium M_{Al} = 0.027 kg mol^{-1}, Faraday's constant F (96487 C mol^{-1}) and the faradic yield f_{Al} of Al dissolution. f_{Al} was defined as the ratio of the weight loss of the aluminium electrodes during the experiments Dm_{exp} over the amount of aluminium consumed calculated according to Faraday's law, Dm_{th} [39,41]:

$$F_{Al} = \frac{\Delta m_{exp}}{\Delta m_{th}} = \frac{3F}{3600\ M_{Al} I\tau}\Delta m_{exp} \tag{8}$$

This feature depends on pH and the amount of other species present in the solution e.g. co-existing anions [49].

3. Results and discussion

3.1. Effect of current density and connection mode

The applied current density is the major operating parameter governing the coagulant production rate, oxygen evolution, heat generation, and the bubble production rate, and hence it rules the EC process performance and operating costs [12,14,39,50]. To determine the effect of current density *j* on the efficiency of color and turbidity removal and energy consumption, continuous electrocoagulation runs with a direct current (DC) were carried out at various current density levels ranging from 100 to 400 A m^{-2}, while maintaining the rest of the investigated parameters constant: initial concentration C_0 = 100 mg L^{-1}, inlet solution pH 7.0 and inlet flow rate *Q* = 15 L h^{-1}. The data monitored for 60 min operation time are presented in Fig. 4. The negative values of absorbance and turbidity removal yields at short times can be explained by the formation of solid polymeric aluminum hydroxide, yet without efficient adsorption of the dye, thus resulting in transient increased medium turbidity [12,16].

As shown in Fig. 4, steady operation of the continuous cell can be attained within less than one hour. The transient period is as much shorter as the current density is high and is reduced in BP mode: as a matter of fact, with this connection mode the three cells are actually operated in series, resulting in a threefold higher aluminium dissolution rate in the cell than in MP configuration for the same current density. At steady state, higher performances of the treatment are allowed by higher current density as expected, with absorbance abatement yield passing from 90 to 97% for current density at 100 and 400 A/m^2 respectively with monopolar configuration. Use of bipolar electrodes improved the treatment efficiency, with absorbance abatement at 91 and 98% with the above current densities. As explained above, this gain in abatement is linked to the higher Al consumption in BP configuration. The turbidity removal yield follows comparable variations with the current density and the electrode connection mode. The above observations are in agreement with the results obtained by Ghosh et al. [45] for the treatment of water containing fluorides, Attour et al. [30] for the treatment of phosphate-containing water, and those of Yu et al. [46] obtained for phosphorus removal.

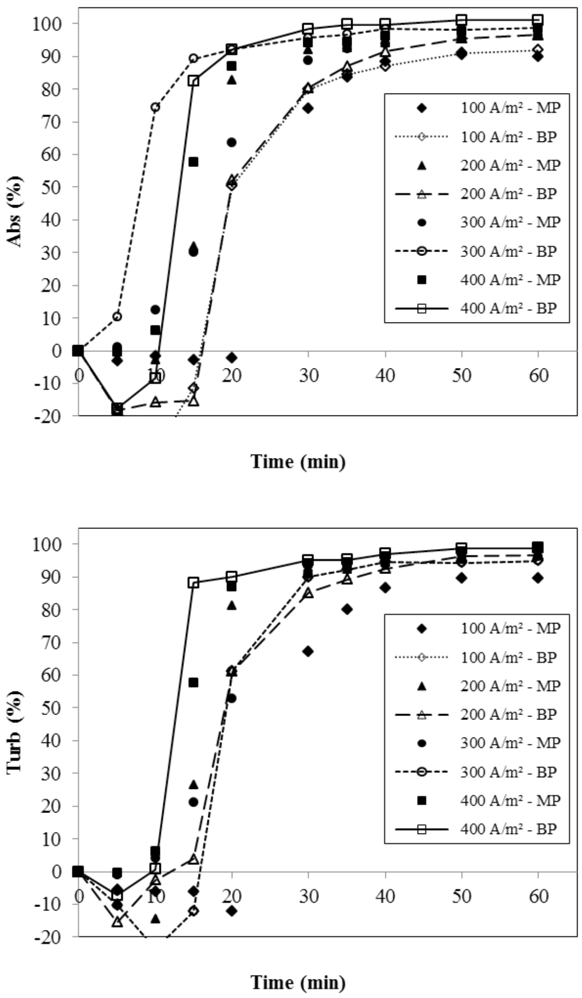


Fig. 4. Transients of absorbance and turbidity removal yields during the treatment at different current densities *j* and electrical connections: C_0 = 100 mg L^{-1}, inlet solution pH 7.0, Q = 15 L h^{-1}.

The cell voltage varied from 4.5 to 25 V depending on the current density and the electrode configuration (data not shown). Although steady operation of the EC reactor was attained only after 20–60 min, the cell voltage varied quite little from start-up of the run. Voltage in BP mode ranging from 7.5 to 25 V was not perfectly equal to three times that at MP, because of current by-pass phenomenon for which the current does not flow only to the facing electrode but can also pass directly through the surrounding liquid to a more distant electrode. This effect is to be more pronounced in the present case of sufficient space around the electrode stack in the cell, in spite of the moderate conductivity of the solution.

The energy consumption for the treatment of the synthetic waste E_{water} was then shown to vary from 0.15 to 1.6 kWh m^{-3} with monopolar configuration and from 0.26 to 3.2 kWh m^{-3} with bipolar electrodes. The E_{dye} values increased from 1.6 to 16 kWh kg^{-1} dye removed at steady state with the monopolar (MP) mode, and from 2.5 to 32 kWh kg^{-1} with the bipolar (BP) mode (Fig. 5). The above data made it possible to estimate the amount of Al required per kg dye removed (Fig. 5, right) for the case of MP connection: higher currents, although more efficient in the abatement, are to generate sludge with a higher Al content: this point in addition to the cost of Al plates, has to be accounted for when defining the optimal operating conditions for the treatment.

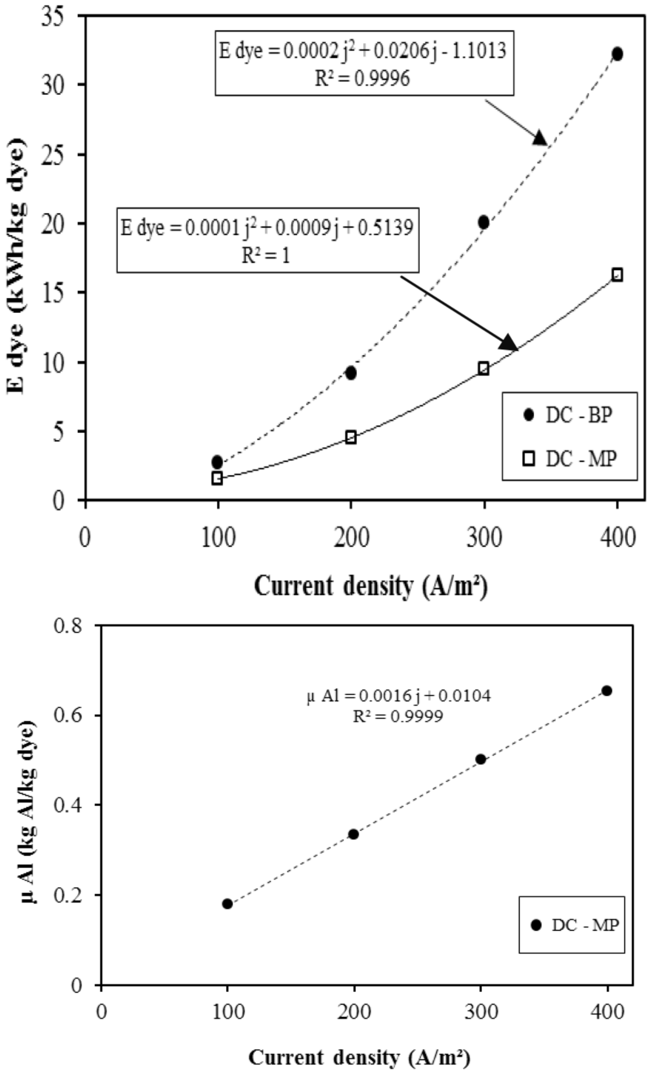


Fig. 5. Effect of current density *j* and electrical connections on energy consumption (E_{dye}) and electrode consumption (m_{Al}): C_0 = 100 mg L^{-1}, inlet solution pH 7.0, Q = 15 L h^{-1}.

The selection of current density and connection mode has to be made depending on the desired abatement yields of absorbance and turbidity. Because of the different electrical features, the two connection modes were compared at two current density levels for the sake of comparable energy consumption and Al dissolution rate (in the form of the outlet Al concentration), i.e. with lower current for BP mode, as shown in Table 2.

Monopolar configuration allows high efficient in the treatment, with energy consumption in the order of 1 kWh m^{-3} and moderate amount of Al in the sludge produced. With bipolar electrodes, treatment at 200 A m^{-2} exhibits comparable efficiency, with comparable energy consumption, however with higher Al dissolution rates – because of the three cells formed by the electrodes: in addition to the additional costs of the sacrificial metal plates, this mode generates richer Al-containing sludge. The above results show that attention has to be paid in defining the electrical connection. In spite of high relevancy in industrial practice, BP configuration was not considered in the following sections.

3.2. *Influence of the type of current*

In general, direct current (DC) is used in electrocoagulation processes. However, the alternating current (AC) has been rarely evaluated for electrocoagulation and only in discontinuous operation [43,47,48]. To study the effect of the type of current on color and turbidity removal by continuous electrocoagulation, experiments were conducted by varying current density from 100 to 400 A m^{-2} with direct current and alternating current. DC was shown to be far more efficient than AC in terms of color and turbidity removal (data not shown). For AC, the highest color removal yield – only at 52.5% – is reached only at 400 A m^{-2}. For turbidity, the removal yield peaked at 35% at 400 A m^{-2}. The poor efficiency of AC could not be interpreted up to now. It is likely that continuous polarization would be preferred for efficient polarization of the solid $Al(OH)_3$ interface, for efficient interaction with the dye molecules. Because of the poor performance of AC, DC has been applied in the runs presented below.

Moreover the cell voltage monitored in the treatment was usually larger with AC than with DC, varying from 10.5 to 18 V in the current density range considered, in comparison with voltages below 12 V with direct current.

The variation of the electrical energy consumption calculated in kWh/m^3 of treated effluent for the two connection modes and the current nature, is illustrated in Fig. 6. At 400 A m^{-2} and after 35 min transient regime, E_{dye} is 1.6 kWh m^{-3} for DC-MP, 2.5 kWh m^{-3} for AC-MP – with a far lower efficiency – and 3.2 kWh m^{-3} for DC-BP. Because of the poor efficiency of alternative current, direct current (DC) with MP electrodes connection has been considered.

3.3. *Effect of the inlet flow rate*

For this parameter of large significance in continuous operation, four values for the inlet flow rate have been tested (15, 26, 38 and 60 L h^{-1}), for *j* = 300 A m^{-2} and inlet

Table 2
Comparison of treatment parameters and performance depending on the connection mode and the current density

Connection mode	Current density ($A\ m^{-2}$)	Cell voltage (V)	Y_{abs} (%)	[Al] ($mg\ L^{-1}$)	E_{water} ($kWh\ m^{-3}$)	μ_{Al}/dye (kg Al/kg dye)
MP	300	9.2	96	54.8	0.88	0.57
MP	400	12.5	97	73.1	1.60	0.75
BP	100	8	91	54.8	0.26	0.62
BP	200	13	96	109.6	0.83	1.14

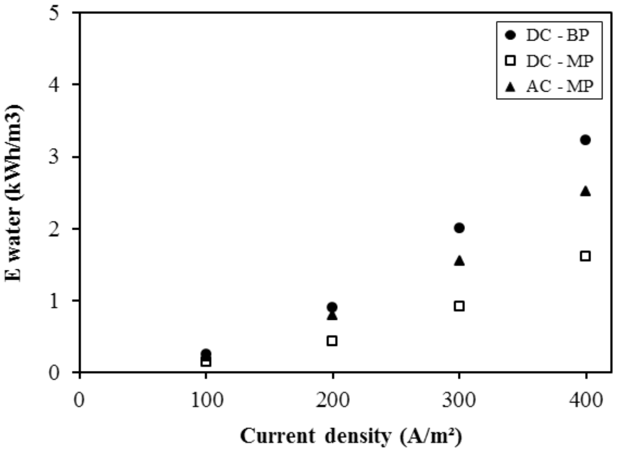


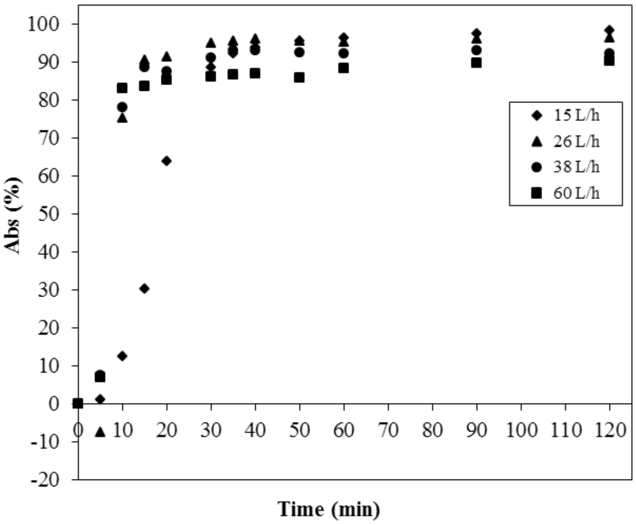
Fig. 6. Energy consumption (E_{water}) versus current density with AC or DC and for the two connection modes: C_0 = 100 mg L^{-1}, inlet solution pH 7.74, Q = 15 L h^{-1}.

solution pH at 6.8. The corresponding residence times were 35, 19, 14 and 9 min, respectively. As shown in Fig. 7, the continuous EC process is efficient for the various flow rates, with color removal yield larger than 90 % at steady state in all cases. These figures show that Y_{Col} is at its highest at 15 L h^{-1} at 98.5% after 120 min operation (steady state) and 92.5% after 35 min transient regime, which corresponds to the cell residence time. On the other hand for Q = 38 L h^{-1}, $Y_{Col(max)}$ is 93% with 35 min close to steady state values at 92%. For Q = 60 L h^{-1}, $Y_{Col(max)}$ is 86% after 35 min treatment and 90% at steady state conditions. The observed effect simply result from the larger amounts of dissolved Al at low flow rates. Comparing the results obtained in this study with published ones [17,33,41,42,43], the inlet flow rates do not exceed 20 L h^{-1} to obtain a color removal ratio higher than 80%.

The optimal rate of feed could thus be taken at a moderate level i.e. 15 and 38 L h^{-1} for the sake of higher removal efficiency. However, operating at low flow rates results in higher amount of Al dissolved, and higher Al contents in the sludge produced, with μ_{Al} criterion increasing from 0.15 to 0.57 kg Al per kg dye when the flow rate is decreased from 60 to 15 L h^{-1}. The final selection of the flow rate to be used must take into account the abatement yield targeted and the amount of Al dissolved per kg dye removed.

3.4. Effect of inlet dye concentration

For assessment of the role exerted by the inlet concentration of the dye during treatment with continuous EC,



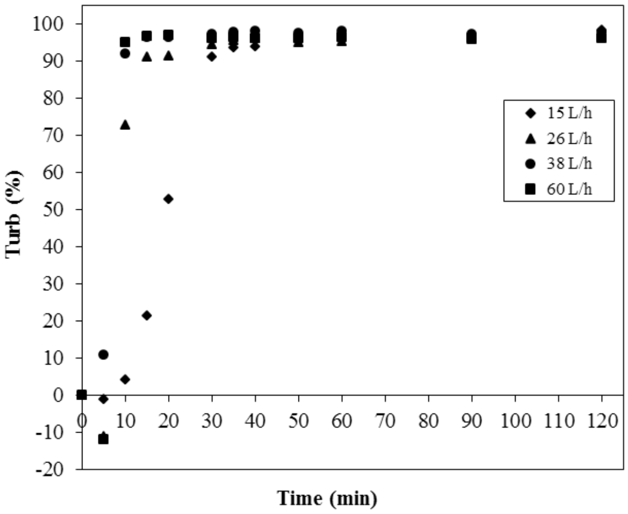


Fig. 7. Transient of absorbance and turbidity removal yields during the treatment at different inlet flow rates Q: C_0 = 100 mg L^{-1}, inlet solution pH 6.8, j = 300 A m^{-2}.

experiments were performed at 300 A m^{-2}, with the lowest flow rate and an inlet pH at 6.9. The transients of color and turbidity removal yields for six different inlet concentrations (100–1000 mg L^{-1}) are shown in Fig. 8. In all cases Y_{Tr} is higher than 93% and Y_{Col} higher than 87%. Moreover, as expected, higher removal yields could be obtained from

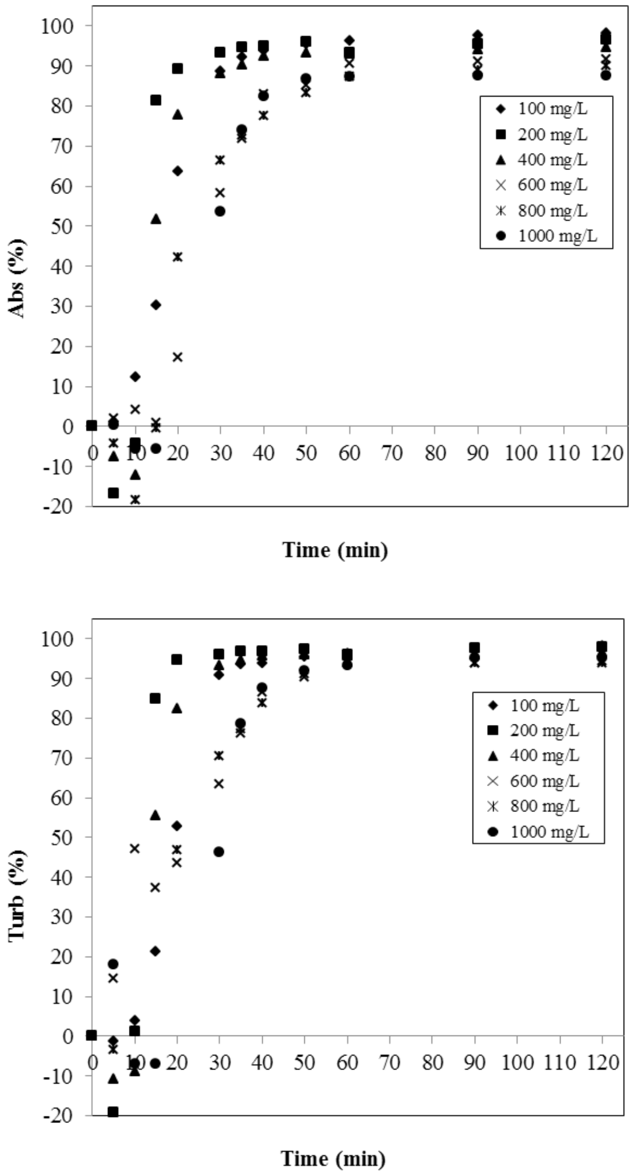


Fig. 8. Transients of absorbance and turbidity removal efficiencies during the treatment for different initial concentration C_0 : Q = 15 L h^{-1}, inlet pH 6.9, j = 300 A m^{-2}.

dilute wastewater than with higher inlet concentrations: the efficiency of dye removal is also to depend on the concentration ratio Al (III) hydroxide/Dye; probably because of sorption equilibria of the dye molecule on the Al (III) flocs.

3.5. Effect of inlet solution pH

The inlet solution pH affects the speciation of trivalent aluminum in the solution, the surface charge of the particle and the performance of electrochemical process [16,18,30–32,36,42,43,51], in spite of the buffering capacity of Al hydroxides species. Tests have been made considering acidic, neutral or alkaline pH in the inlet solution.

The yield of color and turbidity removal is affected by the inlet pH (Fig. 9). At inlet pH of 3.46, minimum Y_{Col} of 64% and Y_{Tr} of 82% were obtained at steady state. Fig. 9 shows that Y_{Col} and Y_{Tr} exceed 92% for other inlet solution pH. The maximum efficiency of color removal was observed at inlet pH 6.9 with Y_{Col} = 92.5% and Y_{Tr} = 93.5% at steady state.

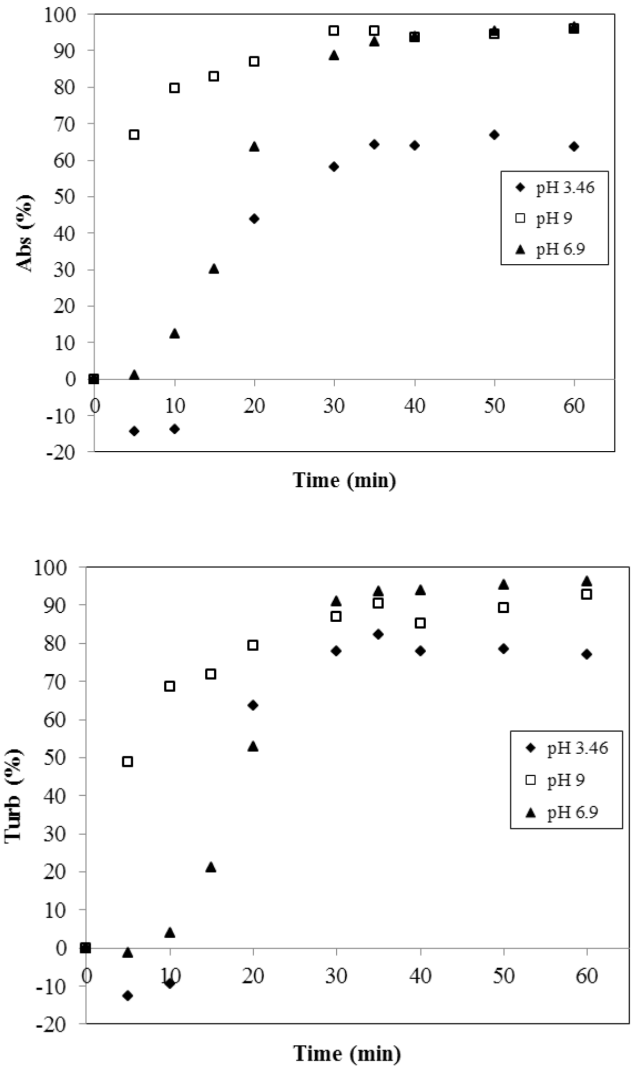


Fig. 9. Transients of absorbance and turbidity removal yields in the treatment for different inlet pH: C_0 = 100 mg L^{-1}, j = 300 A m^{-2}, Q = 15 L h^{-1}.

4. Conclusions

This study aimed to study the removal of a dye largely used in textile industry by continuous electrocoagulation. The abatement yield of the dye is affected by current density, inlet solution pH, inlet flow rate, initial concentration, connection mode and type of current. DC mode was found to be far more efficient than AC mode. At the same current density, the bipolar electrocoagulation mode (BP) was found to be more effective on turbidity and color removal than monopolar electrocoagulation (MP), mainly because of the multiplicity of EC cells in series. However the energy consumption at fixed current density using MP connection was far lower in the monopolar mode since only one cell is involved. For the case examples studied, BP connection although allowing comparable efficiency at comparable energy consumption was shown to result to larger dissolution rates of sacrificial aluminum.

For the DC mode and monopolar connection, abatement yields near 87% and 96% for color and turbidity were reached as the initial concentration of the dye ranged from 100 mg L^{-1} to 1000 mg L^{-1} , with a flow rate Q at 15 L h^{-1} (corresponding to residence time τ = 35 min) and the inlet solution pH was larger than 6.9. The maximum efficiency of color removal was observed at inlet pH 6.9 with Y_{Col} = 97.5% and Y_{Tr} = 98.5%, when the inlet dye concentration was lower than 400 mg/L. The specific electrical energy consumption (E_{dye}) was 9.5 kWh/kg removed dye. According to the results obtained, the continuous EC mode has proven to be effective for the treatment of appreciable flow rates of textile dyes.

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