Treatment of textile Wastewater by electrocoagulation

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Abstract—The aim of this study is the treatment of solutions based on synthetic dyes used in the textile complex of Draa Ben Khedda / Tizi Ouzou, Algeria by electrocoagulation. The studied dyes are Red Solophényl 4GE, Blue Thio Bezathren R, Cibacron Blue Turquoise P-GR and Black Dianix RXN belonging to four classes widely used in textile industry: Direct dyes, vat, reactive and disperse. Using aluminum electrodes in Electrocoagulation in a batch mode has proven its efficiency for the treatment of these dyes, for a concentration of electrolyte (NaCl) added of 3 g / l, a current density of 33.54 mA/cm² and an electrolysis time of 1 h. We notice an abatement of coloring ranging from 87 to 97 % for different dyes treated, with a energy consumption ranging from 55.76 to 86.74 kWh and an amount of the dissolved aluminum ranging from 1.5 to 2.5 kg to remove 1 kg of dye. *Keywords*— Textile dye, Electrocoagulation, Aluminum

I. INTRODUCTION

The textile industry consumes large quantities of water and produces liquid wastes with high pollutant load of various types. Colorants and additives used by the industry can be a serious threat to the environment because their presence in the water, even in very small quantities, is highly visible and undesirable; therefore, their presence in aquatic systems reduces light penetration and thus retards the photosynthesis activity. They also have a tendency to complex metal ions, producing a micro-toxicity to wildlife and other organisms.

Therefore, the depollution of water contaminated by chemical compounds is necessary to protect the environment for future use of this non-conventional water and to minimize water losses due to this recycling. In this context, conventional treatment techniques have already been implemented such as carbon adsorption, chemical oxidation (H₂O₂, O₃, ClO₂ ...), flocculation / coagulation, membrane filtration and biodegradation. However, due to the highly variable composition of these waters, most of these conventional methods are beginning to be insufficient for a simple and effective treatment requiring minimal reagent. Electrocoagulation can be an alternative.

The EC value is an electrochemical method used for the purification of many types of wastewater [1]. This process

generates at suitable pH insoluble metal hydroxide that can remove pollutants. The treatment is done without the addition of chemical flocculants or coagulants, thereby reducing the amount of sludge that should be removed. This technique is easy and requires simple equipment and a small amount of sludge is produced [2].

The objective of this work is to apply the method of electrocoagulation in batch mode for the treatment of four solutions based on synthetic dyes used in the textile complex of Draa Ben Khedda / Tizi Ouzou, Algeria. The dyes studied are: Red Solophényl 4GE Blue Thio Bezathren R, Cibacron Blue Turquoise P-GR and Black Dianix RXN belonging to four classes widely used in textile industry: Direct dyes, vat, reactive and disperse.

II. BIBLIOGRAPHY

Electrocoagulation (EC) is a technology that results from the interaction of three keys technologies that are: electrochemistry, coagulation and flotation [3]. It is based on the fact that the stability of colloids, emulsions and suspensions, is influenced by the electric charges [4]. It refers to the electrochemical production of destabilizing agents that causes charge neutralization for the reduction of pollutants. It has been developed to minimize the disadvantages of conventional technologies in textile wastewater treatment [5-6]. The destabilization of the colloids occurs under the action of the electric field induced between the electrodes and the coagulating action of products generated by oxidation of the anode. These compounds are electrically charged; they destabilize the colloidal system in which solid particles are initially present and form flocs of particles that can be readily separated by flotation. The flotation is improved by the formation of gas bubbles during electrolysis of the solvent (water) [7].

When the electrode material used is aluminum, the reactions are as follows:

- At the anode: $Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3\acute{e}$ (1)
- At the cathode: $3H_2O + 3\acute{e} \rightarrow (3/2) H_{2(g)} + 3OH_{(aq)}^-$ (2)
- In the solution: $Al_{3}^{+}_{(aq)} + 3H_2O \rightarrow Al(OH)_3 + 3H^{+}_{(aq)}$ (3)

The metal cations (Al^{3+}) form mono complexes with hydroxides ions (OH) such as $Al(OH)^{2+}$, $Al(OH)_2^{+}$, $Al_2(OH)_2^{4+}$ and $Al(OH)_4^{-}$ and poly complex $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{-7+}$, and $Al_{13}(OH)_{34}^{5+}$, which are finally converted into Al $(OH)_3$ in a complex precipitation kinetics [8].

Table 1 cites some application of electrocoagulation process for the treatment of textile industry.

TABLE 1 EXAMPLES OF APPLICATION OF THE EC IN THE TEXTILE

References	Composition	Performance	Electrodes
Kobya et al. (2003) [9]	textile effluents	98% reduction of turbidity, 77% COD	Fe and Al
Raju et al.(2008) [10]	synthetic textile effluents	99% reduction in TSS, 62% of COD	Fe and Al
Daneshvar et al. (2006) [11]	Basic dye solution	20 to 100% abatement of color, 75 to 99% COD abatement	Anode Fe, steel cathode
Arslan- Alaton (2008) [12]	Reactive real effluent bath	Steel> Al, 99 to 100% abatement of color, Al> steel for COD The steel consumes 9 kWh / m3 while Al is consumed 5kWh / m3	Al, steel, stainless steel
Can et al. (2006) [13]	textile effluent	Alun CAP more effective in CEC, CEC provides 80% COD reduction against 23% for EC	Al
Muthukuma r <i>et al.</i> (2007) [14]	CI acide orange 10	60% reduction of color	Fe
Zaroual <i>et</i> <i>al.</i> (2006) [15]	Basic textile effluent	100% reduction of color and 84% de COD.	Fe

Daneshvar et al. ((2003) [16]	Color Orange II in aqueous solution	Over 84% of COD removal, more than 98% Color removal	Fe
Alinsafi <i>et</i> <i>al.</i> (2005)[17]	Reactive dye textile and textile effluent	Color eliminated 90-95% and 30- 36% COD	Al

If we consider that the only chemical reactions occurring in the reactor electrocoagulation are metal oxidation at the anode and reduction of water at the cathode. It is possible to determine the mass of dissolved metal and hydrogen gas formed during a time t a current electrolysis I, using Faraday's law:

$$m_{\text{theo}} = \frac{\text{I.t.M}}{\text{n.F}} \tag{4}$$

With:

m: mass of dissolved metal or gas formed (g).

I : current intensity (A).

M: molar mass of the studied metal $(g.mol^{-1})$.

F : the Faraday constant (96500 $Cb.mol^{-1}$).

N : number of electrons involved in the reaction.

T : electrolysis time (s).

The energy consumed (in kWh.m⁻³) is given by the following equation [18]:

$$E = \frac{U.Lt}{V}$$
(5)

With:

U : is cell voltage (V).

 I_{-} : is the intensity (A).

T : is the time (h).

V : is the volume (m^3) .

The equation (5) includes the energy of the anode dissolution and energy gas evolution.

III. EXPERIMENTAL

The effluents are prepared in the laboratory by dissolving in distilled water. The treated volume is 200 ml. The experimental equipment (Figure 1) consists of an installation comprising a reactor which includes two aluminum electrodes connected to a continuous current generator Dc power supply PS-305D. Both electrodes are rectangular plates arranged in parallel and separated from each other at a fixed distance of 2 cm, with a 5.3 cm width and a 4.5 cm height (active area = 23.85 cm^2). Agitation is provided by a magnetic stirrer type N0690 / 1. The electrolysis time is 1 h.



Fig.1 Electrolysis cell

We treated solutions based on these dyes whose recipes used at the complex are as follows (Table 2):

	TABLE 2 DYES PREPARE			
Red Solophenyl 4GE	Blue Thio Bezathren R	Blue Turquoise Cibacron P-GR	Black Dianix RXN	
20 g of dye powder 20 g NaCl; 1 L of fresh water at 100°C;	20 g Dye powder; 4 g Sodium carbonate; 20 g of Ml caustic	5 g Dye powder 80 g Urea 20g Sodium Carbonate	20 g Dye 10 Drops of concentrat e acetic acid	
1ml Fixer (flo-fixed RD). Concentrati on is 20g/l.	soda; 1 L Freshwater 100 ° C; 12 g Hydro sodium sulfite; 4ml Oxygenated water H ₂ O ₂ . Concentrati	1 L Fresh water at 60 °C. The Concentra tion is 5 g/l.	CH ₃ COO H (84%) 1 L Cold freshwater The Concentra tion is 20 g/l.	

For all the treated samples, we performed before and after treatment the following physico-chemical analysis:

- * pH meter (HANNA pH210).
- *Conductivity meter (HANNA / EC214).
- *Turbidity (turbe 550 type) .

*The UV visible spectrometer SHIMADZU / UV-1800 $% \left(1 + \frac{1}{2} \right) = 1000$.

After decantation the absorbance of treated solutions is measured between 400 and 800 nm. The percent of discoloration was calculated by the equation:

Abatement (%) =
$$\frac{C_0 - C_{ap}}{C_0} \times 100$$
 (6)

With:

C₀: initial solution concentration in mg/l.

Cap: Concentration after treatment in mg/l.

The Faraday efficiency is calculated as follows:

$$\varphi_{c} = \frac{\Delta m_{exp}}{\Delta m_{the}}$$
(7)
With:
$$\Delta m_{exp}: \text{Experimental Mass of anode dissolved (g).}$$
$$\Delta m_{the}: \text{ Theoretical Mass of aluminum released from the anode (g)}$$

The electrode consumption is calculated as followed:

$$\mu_{Al} = \frac{\text{Mass of aluminum}}{\text{Mass of dey}}$$
(8)

IV. RESULTS AND DISCUSSION

The wavelengths of maximum absorption are 514; 594; 646 and 530 nm respectively for the Red Solophenyl 4GE (RS), Blue Thio Bezathren R (BB), Cibacron Turquoise Blue P-GR (BT) and Black Dianix RXN (ND). Based on the absorption maxima of each dye we proceeded to establish calibration curves, to estimate their concentrations. The Calibration curves for the four colors are given by the straight lines (figure 2).



Fig 2 Calibration curves for the four dyes absorption maxima

A. Effect of dye concentration

In order to assess the role played by the initial concentration of the dye during treatment with electrocoagulation in batch, we performed experiments keeping all other parameters constant, namely the concentration of electrolyte ([NaCl] = 1 g/l) and the intensity of current (I = 0.6 A). The concentration of the dyes is varied from 3 to 640 mg/l. The results obtained are expressed as rate abatement for a given initial dye concentration (Figure 3). The ranges of variation of initial pH of colored solutions are: Red Solophényl 4GE: pH = 6.5 to 8.8; Blue Thio Bezathren R: pH = 7.5 to 9.7; Blue Turquoise Cibacron P-GR: pH = 6.9 to 7.9 and Black Dianix RXN: pH = 6.3 to 6.5.



Fig 3 Evolution of abatement rate of the concentration depending on the initial concentration of four textile dyes RS, BB, BT and ND. ($pH_{solutio}$; $J = 25.16 \text{ mA/cm}^2$; [$NaCl_{ladded} = 1 \text{ g/l}$, time of electrolysis= 1 hour).

From Figure 3, it is concluded that there is an increase in rate abatement as function of the increase of dye concentration for the RS, and ND BB, however for BT the best rate of reduction equal to 90 % at a concentration of 174.32 mg/l. For RS, BB and ND the rate of reduction rises from 12 to 99 %, 42 to 95 % and 83 to 97 % with the increase of dye concentration 3.7 to 219.6 mg/l, 28 to 400 mg/l and 115 to 640 mg/l respectively.

Differences in the rate of discoloration are observed on reading these graphs. It appears that the treatment of more colored solutions requires more reaction time to reach the maximum rate of elimination. This may be due to insufficient formation of complex metal hydroxide to coagulate the excess of dye molecules present at high concentrations [2].

Figure 4 shows the specific energy requirements and of electrode per kg of dye removed (E_{dye}) necessary for decolorization based on the initial concentration of dye for the four dyes (Red Solophenyl 4GE, Blue Thio Bezathren R, Blue Cibacron Turquoise P -GR and Black Dianix RXN).



Fig 4a: Evolution of the specific energy and electrodes consumption (E_{dye} and μ_{Al}) depending on the initial concentration of RS dye (J = 25. 16 mA /cm²; [NaCl]_{added} = 1g/l; t_{ele} = 1 hour).



Fig 4b Evolution of the specific energy and electrodes consumption (E_{dye} and μ_{AI}) depending on the initial concentration of BB dye (J= 25.16 mA/cm²; [NaCl]_{added}= 1g/l, time of electrolysis = 1 hour).



Fig 4c: Evolution of the specific energy and electrodes consumption (E_{dye} and μ_{Al}) depending on the initial concentration of BT dye. (pH_{solution}; $j = 25.16 \text{ mA/cm}^2$; [NaCl]_{added} = 1 g/l; t _{ele} = 1 hour).



Fig 4d Evolution of the specific energy and electrodes consumption (E_{dye} and μ_{Al}) depending on the initial concentration of ND dye. (pH _{solution}, J = 25.16 mA/cm²; [NaCl] _{added} = 1g/l; t _{ele} = 1 hour).

Figure 4 shows a reduction in the specific energy (E_{dye}) and electrode (μ_{Al}) -consumption as function of the increase of the initial concentration of each dye. This is due to the increase of the conductivity with the increase of the initial concentration of the solution.



Fig 5a Evolution of the abatement of the concentration of RS dye depending on the salt content (J = $25 \text{ mA} / \text{cm}^2$, time electrolysis = 1 hour).



Fig 5b Evolution of the reduction of the concentration of dye BB depending on the salt content ($J=25 \text{ mA/cm}^2$, time electrolysis = 1 hour)



Fig 5c Evolution of the abatement of the concentration of ND dye depending on the salt content (J=25 mA/cm² rate, time electrolysis =1 hour).



Fig 5d Evolution of the abatement of the concentration of dye BT depending on the salt content ($J = 25 \text{ mA/cm}^2$, time electrolysis = 1 hour)

B. Effect of salt concentration

The increase of the conductivity by the addition of sodium chloride is known to reduce the voltage (U) between the electrodes at a constant current density, due to the decreased resistance of the contaminated water [19, 20, 21]. The energy consumption, which is proportional to the voltage applied between the electrodes should decrease. The choice of sodium chloride reflects the fact that the chloride ions significantly reduce undesirable effects of other anions such as SO_4^{2-} and HCO_3^{-} , for example by avoiding the precipitation of calcium carbonate contained in hard water, which could form an insulating layer on the electrode surface and thereby increase the resistance of the electrochemical cell [22].

Furthermore, it is important to know the effect of the presence of NaCl on the abatement efficiencies of coloration. The influence of salt is investigated by varying its concentration from 0.5 to 6 g/l for several values of the dye concentration (mg/l). Current density was fixed at 25 mA/cm². The results obtained (Figure 5) are expressed as the reduction rate (%) and the concentration of NaCl g/l.

From Figure 5 it is seen that:

For a concentration of 150 mg/l of RS, the maximum rate abatement reached 98 % observed at a dose of 0.75 g/L of salt. Beyond that an additional dose salt has no importance on the efficiency of the process.

The best reductions of 92 %, 90 % and 89 % at dye concentrations 200 mg / 1 for BB 174.32 mg / 1 for BT and 162.694 mg/l for ND are observed at a dose of 1 g salt/l.

Increased salinity does not bring significant efficiency gains for the three dyes.

During electrocoagulation process, we noticed a bubbling gas at the cathode. These are hydrogen bubbles produced due to the electrolysis of water which may also induce oxygen to the anode [23]. It was reported that these gas bubbles collect the pollutants on the surface of the solution which are removed by filtration or decantation [16]. Thus this method combines several clearance mechanisms, which makes it attractive; however, it consumes more electric power to dissolve the sacrificial anode and to electrolyze water.

Figure 6 illustrates the decrease in the consumption of energy $E (kWh/m^3)$ caused by the increase of the conductivity of the polluted water through the different dyes.



Fig 6a Effect of conductivity on energy consumption (RS dye) at a constant current density (J = 25 mA/cm^2 , time electrolysis = 1 hour).



Fig 6b Influence of conductivity on energy consumption (BB dye) at a constant current density ($J=25 \text{ mA/cm}^2$, time electrolysis = 1 hour).



Fig 6c Influence of conductivity on energy consumption (BT dye) at a constant current density ($J = 256 \text{ mA/cm}^2$, time electrolysis = 1 hour).



Fig 6d Influence of conductivity on energy consumption (ND dye) at a constant current density ($J = 256 \text{ mA} / \text{cm}^2$, time electrolysis = 1 hour).

It can be seen that energy consumption divided by a factor of 4 when the conductivity is increased by a factor 7 for 162.758 mg/l concentration of the BB, a factor of 3, 4 and 5 when the conductivity is increased by a factor 5.5 and 7 respectively for concentrations of 150 mg/l of RS, 162.694 mg/l of ND and 174.32 mg/l of BT.

To conclude, a NaCl concentration of 3 g/l seems to be quite reasonable because it offers moderate power consumption by Joule effect and prevents rapid degradation of the electrode surface [17, 24].

C. Effect of the current density

The current density is considered as a critical parameter in electrocoagulation [25], specifically for the kinetics of reduction of the COD and discoloration. When the current density increases, the processing time reduced due to the strong dissolution of the anode.

The effect of current density on the removal of dyes was evaluated by repeating the same test with the same operating conditions, namely the salt [NaCl] = 3 g/ l; pH_{solution}; t_{electolyse}; dye concentration, but with different intensities (and this for each dye). The applied current varies between 0.1 A and 0.8 A. Figure 7 shows experimental results of variation of the reduction rate in concentration as a function of current intensity.



Fig 7 Evolution of the reduction rate of concentration of the four RS textile dyes, BB, and BT according to the current intensity value (time electrolysis= 1 h, [NaCl] = 3 g/1).

From Figure 7, we find:

For the RS, the rate abatement increases from 97 % to 98% for current intensity of 0.1 A and 0.8 A respectively for a dye concentration of 150 mg/1.

For BB and ND, the variation of the current intensity from 0.1 to 0.8 A does not affect the abatement for dye concentrations of 162.58 and 200.18 mg/l of BB 162.694 mg/l of ND.

However, for BT, the abatement rate increases from 82% to 80% when I= 0.1A and from 89% to 91% when I=0.8 A for concentrations of dye 174.32 mg/l and 229.8 mg/l respectively.

Kobya et al. [3, 21], confirmed that at higher current densities, the anodic dissolution of aluminum increases, causing a greater rate of the precipitates necessary for pollutant removal.

It should be noted that the size of gas bubbles decreases (their number increases) with increasing of current intensity applied, which can be beneficial for high performance of removing the pollutant from the flotation hydrogen [21].

The Faraday efficiency is the ratio of the experimental amount of dissolved aluminum on the theoretical amount calculated according to faraday law.

Figure 8 shows the variation of Faraday efficiency based on the current density for the four colors.



Fig 8 Evolution of the coulombic efficiency as a function of current density for the four dye $(pH_{solution}; time electrolysis = 1h, [NaCl] = 3 g/ l)$.

Faraday efficiency values are comprised between 28 % and 132.204 % for the four dyes.

Figure 9 illustrates the energy requirements per kg of dye removed necessary for decolorization depending on the current density. Energy consumption increases in a continuous manner as a function of current density for various dyes RS, BB, and BT n. This is observed for discharges of textiles, contaminated by dyes studied by various authors [21, 26].

However, energy consumption represents only a fraction of the process cost, between 20 and 50 % according to Bayramoğlu [27]. According to this author, the main cost is due to the consumption of the electrode material. Electrode consumption μ_{Al} (kg of aluminum consumed per kg of dye removed) increases with increasing the current density



Fig 9a Influence of the current density on the specific energy and the electrodes consumption (RS dye). (Time electrolysis=1h, [NaCl] = 3 g/l).



Fig 9b Influence of the current density on the specific energy consumption and the electrodes (BB dye). (Time electrolysis =1 h, [NaCl] = 3 g/l).



Fig 9c Effect of current density on the specific energy and electrodes consumption (BT dye). (Time electrolysis =1 h, [NaCl] = 3 g/l).



Fig 9d. Influence of the current density on the specific energy and the electrodes consumption (ND dye). (time electrolysis = 1 h, [NaCl] = 3 g/l).

V. CONCLUSIONS

This work aimed to study the removal of synthetic dyes contained in the textile effluents by electrocoagulation.

We applied it for the removal of four dyes, belonging to four classes: direct dye (Red Solophenyl 4GE), vat dye (Blue Thio Bezathren R), reactive dyes (Blue Cibacron Turquoise P-GR) and dye dispersed (Black Dianix RXN). For a concentration of electrolyte (NaCl) added to solutions of around 3 g/l, a current density of 34 mA/cm² and an electrolysis time of hour, we observe a reduction in color from 87 to 97 % for the studied dyes, with a power consumption ranging from 55.76 to 86.74 kWh and an amount of aluminum dissolved from 1.5 to 1 kg 2.488 kg to remove dye. The Treated medium is basic. The electrocoagulation in a batch mode has proven to be effective for the treatment of textile dyes.

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