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Assessment and modeling of COD removal efficiency in textile wastewater using electrocoagulation

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Abstract: In electrocoagulation (EC), coagulants are generated in situ through the corrosion of sacrificial anodes under the application of a direct current (DC) voltage. Simultaneously, electrolytic hydrogen gas (H_2) is produced at the cathode. Aluminum and iron are commonly used as anode materials; their dissolution leads to the formation of hydroxides and polymeric hydroxides. These species act as coagulants capable of destabilizing colloidal suspensions and emulsions, as well as adsorbing, neutralizing, or precipitating dissolved pollutants. Ultimately, they form flocs that can be removed by either sedimentation or flotation.

In this study, the influence of key operating parameters of EC, such as current density (j), solution pH, and conductivity (κ), was investigated for the removal of a red textile dye. The results show that current density has a significant effect on the removal of turbidity, color, and chemical oxygen demand (COD). Removal efficiencies of 77.25% for turbidity, 86.55% for color, and 78.5% for COD were obtained under the following conditions: initial dye concentration C_0 = 100 mg/L, j = 150 A/m², κ = 3.56 mS/cm, inlet flow rate Q = 0.15 L/min, treatment time t = 40 min, and initial pH = 7.65. Finally, a model relating COD removal to aluminum consumption was proposed, which satisfactorily described the experimental results.

Keywords: Electrocoagulation, Turbidity, COD, Dye, Textile wastewater, Modeling

1. Introduction

Wastewater from textile dyeing and finishing factories is a significant source of environmental pollution [1]. Textile wastewater is characterized by high COD, high concentrations of suspended solids (SS), low biodegradability, high-salt content and is the source of aesthetic pollution related to color. The colored wastewater released into the ecosystem is also a dramatic source of aesthetic pollution and perturbation in the aquatic life. Conventional methods for dealing with textile wastewater consist of various combinations of

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biological, physical and chemical methods [2]. 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 [3]. 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 [4].

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. The technique relies upon the electrochemical dissolution of sacrificial Al or Fe electrodes. The generated cations contribute to reduce the stability of suspended entities contained by reduction of their zeta potential. Also, upon formation of hydroxides ions at the cathode, metal ions complex to iron or aluminum hydroxides, which are known to be efficient coagulants. The hydrogen bubbles formed at the cathode adsorb the flocs formed by the process, and ensure their flotation, which simplifies their separation from the treated water. In the case of aluminum, the main reactions are as

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

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

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:

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

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 [5,6], which is referred to as electroflotation.

Electrocoagulation has been applied to the treatment of a wide variety of wastewaters differing significantly in nature and composition [7-11], including oil-containing suspensions [12–15], textile industry effluents [16–18], tannery wastewaters [19,20], and food-processing wastes [21,22]. A review of the available literature indicates that the mechanisms governing pollutant removal by electrocoagulation strongly depend on the characteristics of the wastewater being treated [23,24]. To a lesser extent, treatment performance is also influenced by the design of the electrocoagulation reactor and the prevailing hydrodynamic conditions. Furthermore, the effectiveness of the process should be assessed using multiple performance indicators, such as total organic carbon (TOC), COD, turbidity, color, absorbance at a specific wavelength, and the concentration of toxic contaminants, such as arsenic and phosphate.

The present study aimed to investigate the effectiveness of electrocoagulation for the treatment of textile wastewater in a batch system with liquid recirculation, with particular emphasis on the comparative evolution of color, turbidity, and COD. The treatment performance was also interpreted in relation to aluminum dissolution during the electrocoagulation process. For this purpose, a red dye belonging to the disperse dye class was selected as the model pollutant.

2. Experimental

2.1 Chemicals and analytical techniques

Two aluminum electrodes of rectangular shape (150 mm × 70 mm × 10 mm), have been used as anode and cathode, which corresponds to $S = 105$ cm^2 electrode surface area. The distance between the two electrodes was $e = 20$ mm. These were treated with an HCl aqueous solution for cleaning prior use to avoid passivation.

Experiments were carried out using a red dye solution consisting of a mixture of 2-naphthoic acid and 2-naphtol (Figure 1) with a total concentration (C_0) of 100 mg/L; the synthetic wastewater was noticeably dyed. Dye concentration was estimated from its absorbance characteristics in the UV–vis range (250 - 800 nm), using the wavelength that provided the maximum intensity ($\lambda_{max} = 503$ nm) and a UV–vis spectrophotometer (Secomam). Solution conductivity and pH were measured using a CDM210 conductimeter and a Consort C832 pHmeter.

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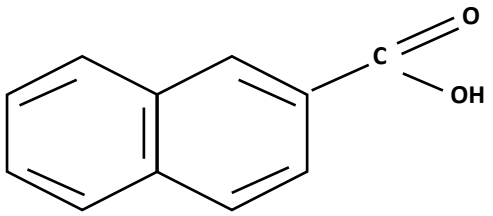
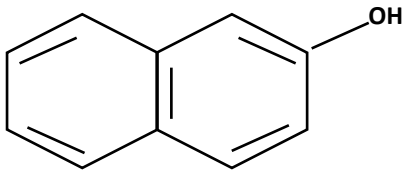
The COD levels were determined using the standardized colorimetric method with excess of hexavalent chromium and subsequent measurement of the optical density. Turbidity of the waters was measured using a Hanna Ins. LP 2000 spectrophotometer. Data were given in Nephelometric Turbidity Unit (NTU).

Concentrations of Al were determined by atomic absorption (Varian AA-20), after dilution and acidification of the solution samples with nitric acid for total dissolution of the metal species. In most cases, the liquid fractions had to be filtered using conventional 0.45µm filters to remove the suspended solids, prior to injection into the atomic absorption apparatus.



2- naphtoic acid



2-naphtol

Figure 1. Molecular structure of the constituents of the red dye

2.2 Set-up and protocol

The batch experimental set up is shown in figure 2. The EC cell was a parallel plate electrochemical cell out of methyl polymethacrylate provided with two facing electrodes machined in aluminum. Experiments were conducted batch wise with recirculation of the liquid in the circuit, which consists of a peristaltic pump, the cell and a double- walled tank for temperature control at approx. 20 °C and separation of the gas formed. Two liters of wastewater were introduced in the tank and a gentle agitation was ensured along the run. The operating parameters were set as follows: initial dye concentration (C_0) = 100 mg/L, initial pH = 7.65, conductivity (κ) = 3.65 mS/cm, and liquid flow rate (Q) = 0.15 L/min. The current density ranged from 40 to 200 A/m² using an AFX 2930 SB DC power supply and the cell voltage was continuously recorded. Twenty-milliliter samples were collected during the 1-hour runs to monitor the treatment progress, as follows. The pH was observed to increase regularly from its initial value, depending on the current density applied. The pH of the samples was adjusted to 7 within 0.5 for optimal precipitation of Al hydroxides.

For all experiments 2 ml of the two-phase samples was taken for analysis of the metal content, the rest was allowed to settle for 24 h. Determination of the COD levels, absorbance at 503 nm, and turbidity was made from the clear liquid in the sample vial. All concentrations were corrected from the change in volume due to the regular sampling.

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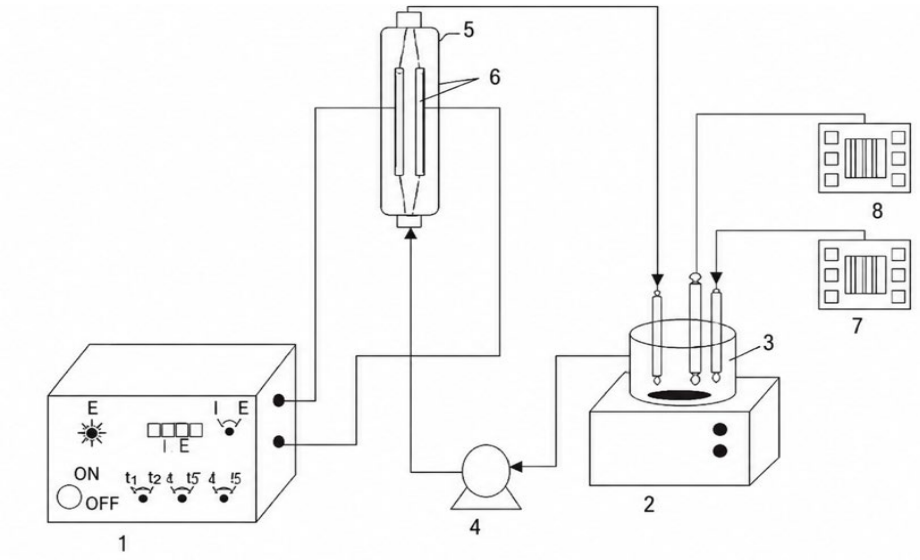

Figure 2. Schematic diagram of experimental set-up. (1) DC power supply; (2) magnetic stirrer; (3) plexiglass reactor; (4) peristaltic pump; (5) electrochemical cell; (6) aluminum electrodes; (7) conductimeter; (8) pH meter

Color, turbidity, and COD removal efficiencies (Y_{COL}, Y_{TR}, Y_{COD}) were expressed as a percentage and defined as

$$\mathbf{Y(\%) = \frac{C_0 - C_f}{C_0} . 100} \quad (4)$$

The subscripts, "0" and "f", are used to distinguish concentration in the inlet and the outlet streams of the electrocoagulation cell.

The specific electrical energy consumption per kg dye removed (E_{dye}) and the specific electrode consumption per kg dye (μ_{Al}) were calculated as follows

$$\mathbf{E_{dye}\left(\frac{kWh}{kg\,dye}\right) = \frac{U\,I\,t}{1000\,V\,(C_0\,Y_{COL})}} \quad (5)$$

$$\mathbf{\mu_{Al}\left(\frac{kg\,Al}{kg\,dye}\right) = \frac{3600\,M_{Al}\,I\,t\,\Phi_{Al}}{3F}\,\frac{1}{V\,(C_0\,Y_{COL})}} \quad (6)$$

using the dye concentration in the inlet stream C_0 (kg/m^3), current intensity I (A), cell voltage U (V), electrolysis time t (h), liquid volume V (m^3), molar weight of aluminum M_{Al} = 0.02698 kg/mol, Faraday's constant F (96487 C/mol) and the faradic yield Φ_{Al} of Al dissolution. Φ_{Al} was estimated as the ratio of the weight loss of the aluminum electrodes during the experiments Δm_{exp} and the amount of aluminium consumed theoretically at the anode Δm_{th}:

$$\mathbf{\Phi_{Al} = \frac{\Delta m_{exp}}{\Delta m_{th}}} \quad (7)$$

This parameter depends upon the pH and the amount of other soluble species, for example co-existing anions [25].

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3. Experimental results

3.1 Color, turbidity, and COD removal efficiency

Four current densities 40, 100, 150 and 200 A/m^2 were tested for the treatment of the synthetic wastewater at fixed inlet concentration C_0 = 100 mg/L with pH values between 7 and 7.5. The results, expressed in the form of color, turbidity and COD reduction versus time, are shown in figures (3-5). Both final reduction rate and kinetics of all parameters was observed to increase with the current density. This can be explained by the fact that the amount of Al^{3+} species formed by dissolution of the anode, increases with the current density according to Faraday's law and previous results [7,16,24].

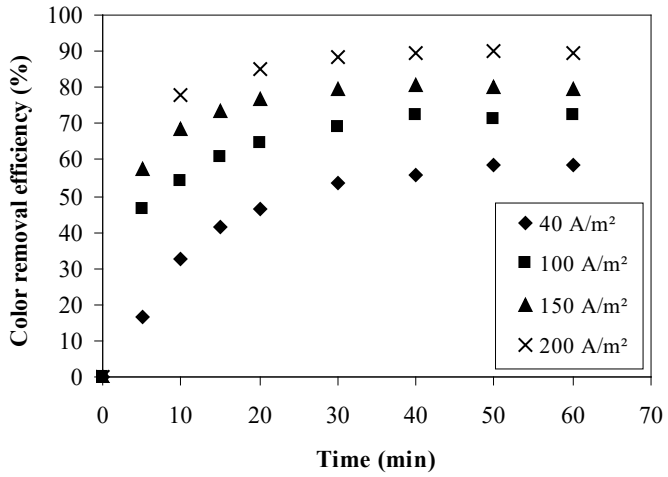

Figure 3. Effect of current density j on the color removal efficiency: C_0 = 100 mg/L, initial pH 7.65, conductivity κ = 3.56 mS/cm

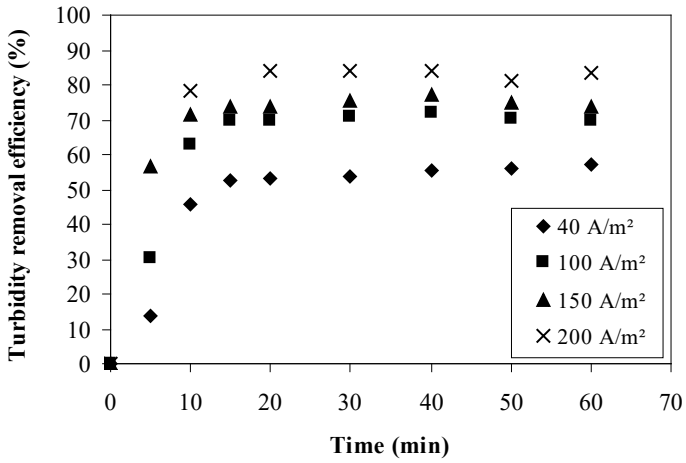

Figure 4. Effect of current density j on the turbidity removal efficiency: C_0 = 100 mg/L, initial pH 7.65, conductivity κ = 3.56 mS/cm

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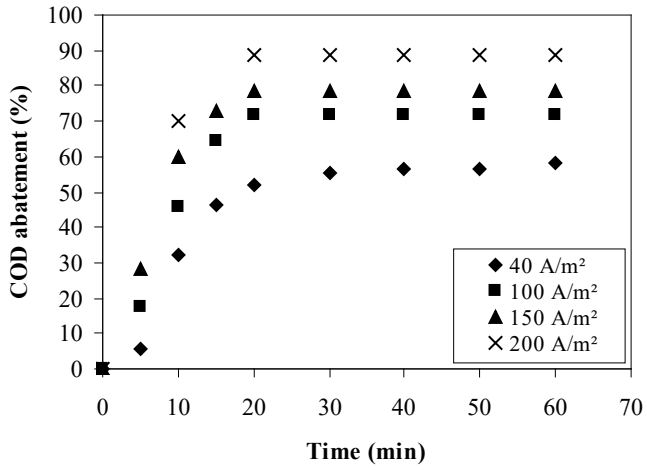

Figure 5. Effect of current density j on the COD abatement: C_0 = 100 mg/L, initial pH 7.65, conductivity κ = 3.56 mS/cm

4. Interpretation

4.1 Model for COD abatement

Interpretation of the experimental variations was carried out by using an overall mechanistic model based on the complexation of suspended matter by aluminum species was employed to describe COD removal from concentrated suspensions [20]. This approach was employed for evaluation of the pollutant complexation by the Al and Fe species. The assumptions of the model are briefly reminded below

- The COD level, [S], expressed in mgO_2 by liter (mg/L) varies from $[S]_0$ at initial time, to $[S]_{inf}$ upon very large amounts of coagulant. The two levels are determined by experiments, depending on the electrode nature and the waste considered.
- Electrocoagulation proceeds by complexation by the dissolved metal species, M

$$\mathbf{M + n\ S \rightarrow M\text{-}S} \qquad (8)$$

where coefficient n is expressed in mg dissolved metal per mgO_2.

- Coagulation equilibrium constant was defined by the overall expression [24]

$$\mathbf{K = \frac{[M-S]}{[M]_{free}[S]}} \qquad (9)$$

The expression of the COD level can be yielded from mass balances in the metal dissolved and in COD, taking into account rel. (9).

$$\mathbf{[S] = [S]_{inf} + \frac{-\left\{1 + K\left(\frac{[M]}{n} - [S]_R\right)\right\} + \sqrt{\left\{1 + K\left(\frac{[M]}{n} - [S]_R\right)\right\}^2 + 4K[S]_R}}{2K}} \qquad (10)$$

where $[S]_R$ refers to the COD level which can treated by the coagulation, i.e. the difference ($[S]_0 - [S]_{inf}$).

Fitting of the experimental data to rel. (10) was to yield parameters K and n. However, it was observed that very different couples (K, n) led to similar values of the objective function, calculated as the sum of the squared

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deviations between experimental and predicted COD levels; for all cases, ratio (K/n) attained comparable values, of the order of 10^{-2} L mg^{-1}. Contrary to the case of oil suspensions with very high COD levels investigated previously [14,24] the present wastewaters have a moderate COD level which can be neglected as compared to $[M]/n$ ratios in rel. (10). Using first-order polynomial expansions, rel. (10) could therefore be simplified to

$$[S] \approx [S]_{inf} + \frac{[S]_R}{1 + \frac{K}{n}[M]} \qquad (11)$$

The representation of the theoretical COD abatement (rel. 11) according to the aluminum concentration is then possible. Figure 6 shows that the model (rel. 11) allows a good fitting of the experimental data.

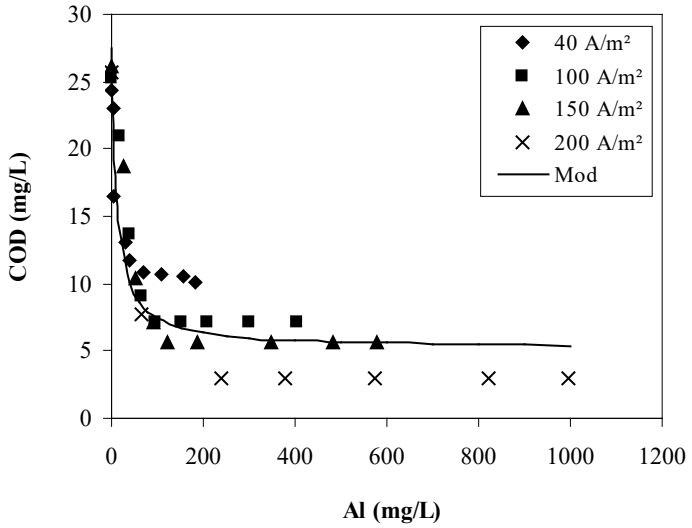

Figure 6. Variations of the COD with the amount of aluminum dissolved, for different current densities, C_0 = 100 mg/L, initial pH 7.65, conductivity κ = 3.56 mS/cm

It appears that the influence of the current density on the COD abatement suggested by the rough experimental data presented paragraph 3 is not so preponderant. In fact, increasing the density leads also to much more dissolved aluminium which seems to be the most significant parameter governing the abatment of the pollution. An aluminium concentration of 400 mg/L is sufficient for a reduction of the COD of approximately 79.5 %. The value of (K/n) is 0.089 L/mg. These results are of the same order of magnitude as those obtained for textile wastewater which varied from 0.0099 to 0.01472 [26].

These various curves show well the agreement between values of experimental and theoretical COD. The model is more regular and the light differences can be due to the simplicity of the model for simulate a complex process in a wide scale of operating parameters.

It should be noted that this model (rel. 11), can be applied to other types of pollution, such as turbidity (Figure 7).

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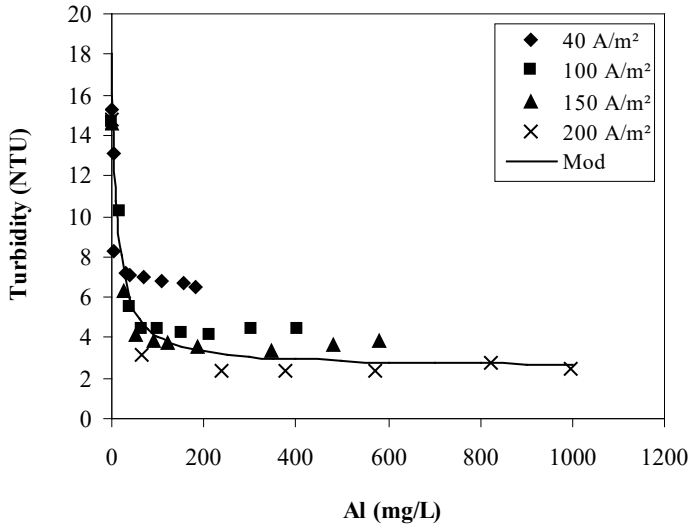

Figure 7. Variations of the turbidity with the amount of aluminum dissolved, for different current densities, C_0 = 100 mg/L, initial pH 7.65, inlet flow rate Q = 0.15 L L/min, conductivity κ = 3.56 mS/cm

5. Conclusion

Electrocoagulation using aluminum electrodes proved to be an effective treatment process for textile wastewater. Under the operating conditions investigated (C_0 = 100 mg/L, current density j = 150 A/m², retention time t = 40 min, initial pH = 7.65, inlet flow rate Q = 0.15 L/min, and conductivity κ = 3.56 mS/cm), high removal efficiencies were achieved for color, turbidity, and COD, exceeding 86.55%, 77.25%, and 78.5%, respectively.

The proposed model, which correlates COD removal with aluminum consumption, provided a satisfactory representation of the experimental data. Furthermore, the model was found to be applicable to turbidity removal, demonstrating its broader potential for predicting treatment performance. Future investigations should focus on a more comprehensive characterization of the residual pollutants present after treatment. The observed pollutant removal can be considered satisfactory provided that the remaining contaminants do not include highly toxic compounds, such as residual dyes or potentially hazardous degradation by-products generated from the initial pollutants.

Conflicts of interest

The authors declare no conflict of interest.

Author contributions

Conceptualization, B.M. and N.A.; methodology, B.M.; software, B.M., M.H. and F.M.; validation, B.M., M.H. and F.M; formal analysis, B.M and B.K.; investigation, B.M, B.K and N.A.; resources, B.M; data curation, B.M, M.H, F.M and N.A; writing-original draft preparation, B.M.; writing-review and editing, B.M and N.A.; visualization, B.M; supervision, B.M.; project administration, B.M.; funding acquisition, B.M.

All authors have read and agreed to the published version of the manuscript.

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