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Optimization of the electrocoagulation process by central composite design for the elimination of poly black dye using recycled aluminum electrodes

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ABSTRACT

In the present study, electrocoagulation process employing recycled aluminum electrode was applied for the removal of a "poly black" (pb) dye and for the treatment of a real textile effluents. Effect of major operating parameters such as pH, concentration, electrodes dissolution, and voltage were investigated for color removal efficiency. Central composite design modeling using response surface methodology was carried out, the parameters considered are the pH (5–10), the distance between the electrodes (2–6 cm), the intensity of the current (0.20–0.40 A). The obtained correlation coefficient R^2 were found equal to 94.84% at t = 30 min, 96.29% at t = 45 min and 96.26% at t = 60 min for color removal, which indicated that the actual results fit relatively good with the predicted results by the application of the quadratic models. According to the models, the most influential interactions were pH-current intensity and pH-distance between the electrodes. Moreover, 3D response surface graphs were presented to investigate the optimum condition of the studied variables. For the response optimizer, the most economical operating parameters for a decolorization efficiency of 99.27% are: a pH of 7.46, a current intensity of 0.28 A, an electrolysis time of 60 min and a distance between the 1.2 cm electrodes. The operating cost in the optimal conditions was calculated as 1.06×10^{-3} DZD (6.80×10^{-6} $)/L of treated water, which corresponds to energy consumption of 2.66×10^{-4} kWh/L.

Keywords: Aluminum; Dye; Electrocoagulation; Recycling; Response surface methodology

1. Introduction

Textile industry effluents are one of the major sources of environmental contamination and public well-beings, mostly in many urban regions. These effluents are severely polluted with the complex organic and inorganic chemicals which are used during various steps of textile processing [1]. Therefore, to protect the environment, textile wastewater must be treated up to the safe discharge limits as recommended by legal bodies of different countries [2,3].

The conventional methods, generally used for the removal of dyes from industrial polluted waters, are mainly biological and physicochemical treatments as well as their various combinations. Nevertheless, biological treatments are better than other methods, but the toxicity of the dyes usually prevents bacterial growth thus limiting the effectiveness of

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bleaching [4]. Physico-chemical methods are generally based on adsorption [5], coagulation–flocculation [6], chemical oxidation and photodegradation [7]. However, these treatment methods usually consume a significant amount of chemicals, which sometimes leads to secondary pollution and a huge volume of sludge [8].

The electrochemical techniques offer the advantage of providing electrons which is a clean, versatile and effective reagent for the elimination of contaminants. More importantly, it is simple to operate and does not produce any toxic compounds. Numerous electrochemical techniques including electrochemical oxidation, electroreduction, electrodialysis, electrofloatation, electrocoagulation has proved its efficiency in contaminant removal from wastewater [9]. Electrocoagulation (EC) is an attractive method for the treatment of various kinds of wastewater, by virtue of various benefits Including environmental compatibility, versatility, energy efficiency, safety, selectivity, amenability to automation, and cost effectiveness. This process is characterized by simple equipment, easy operation, a shortened reactive retention period, a reduction or absence of equipment for adding chemicals and decreased amount of precipitate or sludge which sediments rapidly [10]. The electrocoagulation process creates in the water to be purified complexes of the type $Al_2(OH)^{5+}$, $Al_2(OH)^{4+}$ or hydroxides $Al(OH)_3$, $Fe(OH)_2$, $Fe(OH)_3$, which play the role of coagulant by forming aggregates of particles which are decanted afterwards to have clear water [11]. The electric field prevailing in the electrolysis cell causes the migration of colloidal particles towards the anode, which has the effect of increasing their probability of encounter, thus promoting coagulation–flocculation. The electrolysis of water also leads to the formation of small bubbles of oxygen and hydrogen (whose average size is less than 100 micrometers) respectively at the anode and at the cathode. These bubbles are mainly made up of hydrogen because the formation of oxygen is a secondary reaction and often of negligible importance at the anode. These microbubbles are then adsorbed on the flocculated materials and can cause them to rise. This phenomenon is sometimes favored to recover sludge by flotation. The foam formed may be of mediocre stability and the oxidizable materials fall to the bottom of the tank. Despite this fact, many light and heavy particles remain in suspension and it is necessary to resort to their separation by decantation or filtration [12]. Electrocoagulation (EC) is an effective method for the treatment of textile wastewater. Research has mainly focused on the technical performance of this process, while its economic aspect has been most often neglected.

The response surface methodology (RSM) is usually used in chemical engineering, bioengineering, and pharmaceutical engineering. Multiple regression equations can be used for fitting values of variables and responses, estimating the coefficients of regression equations, selecting optimum condition for the process to achieve the best response [13].

In this work, the electrocoagulation process was used for the removal of a dye from the textile industry (poly black). Aluminum anodes produced from recycled cans were used as electrodes. Additionally, to optimize the operating conditions of (EC) by reducing the number of experiments required, central composite design (CCD) modeling in response surface methodology was performed to study the effects of many operating parameters, these findings we obtain could pave the way for a significant reduction in the number of experiments required for further research and make the recycled aluminum-based electrode a promising candidate for the development and scale-up of a low-cost EC electrode. This paper also discusses a simplified operating cost analysis of EC textile wastewater treatment using recycled aluminum electrodes taking into account the energy consumed and dissolved aluminum concentration.

2. Materials and methods

2.1. Electrocoagulation process

Electrocoagulation is the process of destabilizing suspended, emulsified, or dissolved contaminants in an aqueous medium by Introducing an electric current into the medium. In its simplest form, an electrocoagulation reactor may be made up of an electrolytic cell with one anode and one cathode. The conductive metal plates are commonly known as 'sacrificial electrodes' and may be made of the same or different materials (anode and cathode) [14]. Electrocoagulation is the electrochemical production of destabilization agents (such as Al, Fe) that brings about neutralization of electric charge for removing pollutant. Once charged, the particles bond together like small magnets to form a mass. This process has proven very effective in removing contaminants from water and is characterized by reduced sludge production, no requirement for chemical use, and ease of operation [15]. Colloid – destabilizing agents that effect on-charge neutralization is produced by electrolysis in the EC process. For example, aluminum anodes are used to produce Aluminum cations which have the same effect as the addition of Al-based coagulants in conventional treatment systems [15].

Electrocoagulation consists of chemical and physicochemical reactions that are caused by an electric current. It takes place in three successive stages [16]:

- Formation of coagulants by electrolytic oxidation of the consumable electrode;
- Destabilization of colloids;
- Aggregation of destabilized particles.

The colloid destabilization mechanism can be summarized as follows [16]:

- Diffuse double layer compression;
- Neutralization of the charges of ionic species present in the water;
- Floc formation.

During the electrocoagulation process, the electrochemical reactions that occur by electrodes subjected to direct current are the reduction of water at the cathode (main reaction), as well as the oxidation reaction of the metal at the anode [17]. Under appropriate pH conditions, all of the M^{3+} ions generated in solution hydrate and then react with water to form mainly metal hydroxides $M(OH)_3$. In fact, the metal ions produced can undergo other spontaneous reactions to give corresponding hydroxides and/or polyhydroxides

[18]. In the case of an aluminum electrode, various monomeric and polymeric species are formed and then ultimately turn into $Al(OH)_3$ (solid). These amorphous $M(OH)_3$ species, also called "flocculation fields", have a large surface area suitable for the rapid adsorption of soluble organic compounds and the trapping of colloidal particles, via electrostatic forces of the Van der Waals type, this leads to the formation of flocs which will then be easily separated from the aqueous medium either by sedimentation, or by flotation favored by the generation of bubbles of hydrogen (H_2) and dioxygen (O_2) [17].

- Anodic reaction:

$$Al \rightarrow Al^{3+}_{(aq)} + 3e \quad (1)$$

- Cathodic reaction:

$$2H_2O + 2e \rightarrow H_{2(g)} + 2OH^- \quad (2)$$

- Reaction in solution:

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

The mass '*m*' of metal dissolved at the anode per second is given by Faraday's law [18]:

$$m(\text{theorectical}) = \frac{I \cdot t \cdot M}{Z \cdot F} \quad (4)$$

where *I* is the imposed intensity (A). *M* is the molar mass of the metal (g/mol), *F* is the Faraday constant (C/mol), *Z* is the valence of the ion formed, and *t* is the electrolysis time (s).

The value of the energy consumption as a function of the treated water is calculated according to the following expression [18]:

$$C_{\text{energy}} = \frac{U \cdot I \cdot t}{V} \quad (5)$$

where *U* is the applied voltage (V), *I* is the current (A), *t* is the electrolysis time (h) and *V* is the volume (m^3) of water treated at the optimal electrolysis time.

The value of the aluminum consumption at the electrodes is calculated according to the following Faraday's law [18]:

$$C_{Al^{3+}(\text{electrode})} = \frac{I \cdot t \cdot M}{Z \cdot F \cdot V} \quad (6)$$

where *V* is the volume of the reactor solution.

2.2. Synthetic wastewater

Synthetic wastewater was prepared from powder of the poly black dye with a concentration of 10 mg/L (offered by an Algerian Company, which uses it to color polyester). An amount of 1 g/L of NaCl is added to the solution as an electrolyte. The initial pH of the solution was found to be 6.45, and the current was initially adjusted to the desired value between 0.20 and 0.40 A.

The characteristics of the synthetic colored solution before the treatment by electrocoagulation are grouped in Table 1.

2.3. Experimental set-up

The experiments were carried out in an electrocoagulation device containing two recycled aluminum electrodes; these monopolar electrodes have the dimensions of 130 mm × 50 mm × 50 mm); synthetic water is introduced into the EC tank (glass reactor) with a volume of 3 L; and after adding the NaCl; the two electrodes each have a nominal submerged surface of 36 cm^2 and placed at different distances (from 2 to 6 cm), the system is submitted to magnetic stirring. The reactor is fitted with a stopcock, at a certain height from its base, allowing samples to be taken for spectroscopic analysis of the dye after determining the calibration curve. The experimental setup has been shown in Fig. 1.

2.4. Electrode cleaning

The parallel and rectangular recycled aluminum plates (electrodes) have been previously treated with an abrasive paper and HCl solution at their surface, in order to homogenize them and remove impurities and deposits that can contaminate them. The electrodes are taken out of the reactor, after each electrocoagulation manipulation for the purpose of examining and cleaning them, since the decomposition residues of pollutants can deposit on their surface.

2.5. Experimental design and analytical method

2.5.1. Response surface methodology

In the optimization process, we seek to improve the behavior of a system or a process, in a well-defined experimental field, influenced by one or more variables. Response surfaces methodology is a collection of experimental strategies from mathematical and statistical methods that allows an experimenter to choose the best combination of parameter levels that optimizes a process [19].

It makes it possible to find in an empirical but economic way the link that exists between the parameters

Table 1
Characteristics of the synthetic colored solution before the treatment by electrocoagulation

Parameters	Values
pH	6.45
Turbidity (NTU)	42.5
Concentration (mg/L)	10
COD (mgO_2/L)	45
MES (mg/L)	42.30

(factors) of a process or system and the quality characteristics (the responses) sought by the experimenter [19].

2.5.2. Mathematical modeling of the response

In the absence of any information on the function which links the response to the factors, we give ourselves a priori a law of evolution whose most general formulation is as follows:

$$y = f\left(x_1, x_2, x_3, \ldots, x_n\right) \tag{7}$$

This function is too general and it is customary to take a limited Taylor–Mac Laurin expansion, is an approximation. If the derivatives can be considered as constants, the previous expansion takes the form of a polynomial of greater or lesser degree [20]:

$$y = a_0 + \sum a_i x_i + \sum a_{ij} x_i x_j + \sum a_{ij} x_i^2 + \cdots \tag{8}$$

where y: is the magnitude in which the experimenter is interested; this is the response or the magnitude of interest, $x_{i,j}$: represents a level of factors i or j, and a_0, a_i, a_{ij}, a_{ii} are the coefficients of the polynomial.

This model is called the a priori model or the postulated model. The established models are valid prediction models in the field of study, which must always be specified. They are not theoretical models based on physicochemical or mechanical laws. In a few rare cases, it is possible to use known theoretical physical laws [21].

Among the three types of response surface planes commonly used, namely Box–Behnken plans, Doehlert plans and centered composite planes, we opted of the third type. Composite shots lend themselves well to the sequential flow of a study. Indeed, these plans give the possibility of beginning the study with a minimal number of experiments. Then, if the model is validated the study usually ends otherwise, we have the possibility to add more experiments without losing the results of the tests carried out previously. Additional tests allow a quadratic model to be established [21].

The additional tests are represented by experimental points located on the coordinate axes and by new center points. The points on the coordinate axes are called the star points. Composite designs therefore have three parts:

- Factorial plan: it is a complete or fractional factorial plan at two levels by factors. The experimental points are at the top of the field of study;
- Star plane: the points of the star plane are on the axes and they are, in general, all located at the same distance from the center of the study area;
- The points in the center of the study area. It is always expected experimental points located at the center of the study domain, and this for both factorial and star planes [22].

Fig. 2 shows the points of the centered composite design for two and three factors.

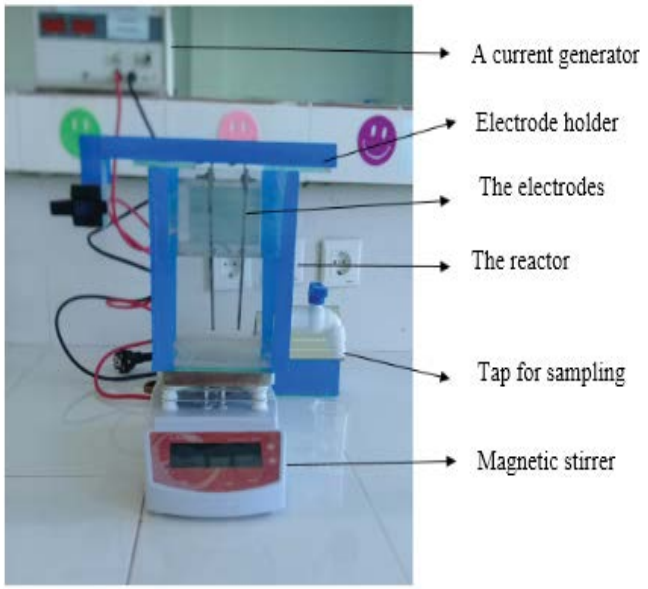

Fig. 1. Schematic of the electrocoagulation reactor.

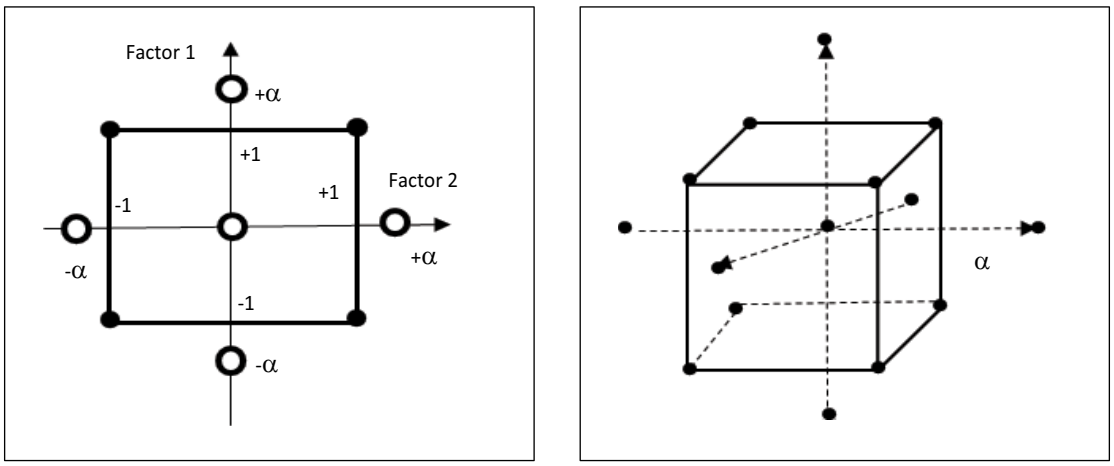

Fig. 2. Composite two- and three-factor designs.

The arrangement of the experimental points in space conditions the variance of the prediction of the response as well as its spatial arrangement, in this case we are looking for the quality of iso-variance by rotation: a distribution of variance which is independent of a rotation: depends only the distance to the origin. Rifi et al. [22] demonstrated that the condition of iso-variance by rotation is satisfied for a value of the distance from the points in star to the central point defined as:

$$\alpha = \sqrt[4]{2^k} \tag{9}$$

where k is the number of factors.

The condition of iso-variance by rotation is therefore expressed in a simple manner; it is independent of the number n_0 of repetitions at the center of the experimental [20].

The CCD, which is the standard RSM, was selected in order to study and find the relationship between the response and studied factors, and for optimization. Knowing the constraint of the variables owing to their difference in units and/or difference in limits of variation, the variables were coded according to the following Eq. (10):

$$X_i = \frac{X_i - X_{cp}}{\Delta X_i} \tag{10}$$

where X corresponds to code level; X_{cp} is the uncoded value at the central point; X_i is the uncoded value, and ΔX_i is the change value between levels.

3. Results and discussions

3.1. Experimental results

3.1.1. Effect of initial pH

In wastewater treatment using electrocoagulation, pH plays a very important role in determining treatment efficiency. Therefore, experiments were designed to determine the optimum pH of synthetic textile wastewater that allowed for maximum discoloration rate. The effect of pH on the treatment efficiency was examined by altering the initial pH from 5 to 10 (current intensity = 0.28 A, distance inter electrodes = 1.2 cm, reaction time = 60 min) and keeping all other parameters constant.

From Fig. 3 it can be noticed that the initial pH of the solution varies during the treatment by the electrocoagulation process, the pH increases when it is an acid medium and it decreases for an alkaline solution. It is also noted that all the pH values studied stabilize towards a value between 7 and 8. These curves also confirm that basic pH are the most favorable for the treatment of a colored solution by EC, thus giving the highest yields.

- For pH = 5 and 6:

As we have just said, the pH increases as a function of time, which is reflected by a lowering of the acidity of the medium, this is perhaps explained by a reduction of H^+ ions at the level of the cathode with a clear observation release of hydrogen following the reaction:

$$2H^+ + 2e \rightarrow H_2^{\uparrow} \tag{11}$$

- For basic pH 8, 9 and 10:

The observed decrease in pH with time is probably explained by the reaction of Al^{3+} ions with hydroxide ions according to Eq. (1).

3.1.2. Effect of the initial dye concentration

Given the synthetic nature of our affluent, it was considered interesting to study the effect of the initial concentration of poly black on the elimination of the latter by the electrocoagulation process. For this, we carried out tests at initial concentrations of 10–100 mg/L. The results obtained are shown in Fig. 4.

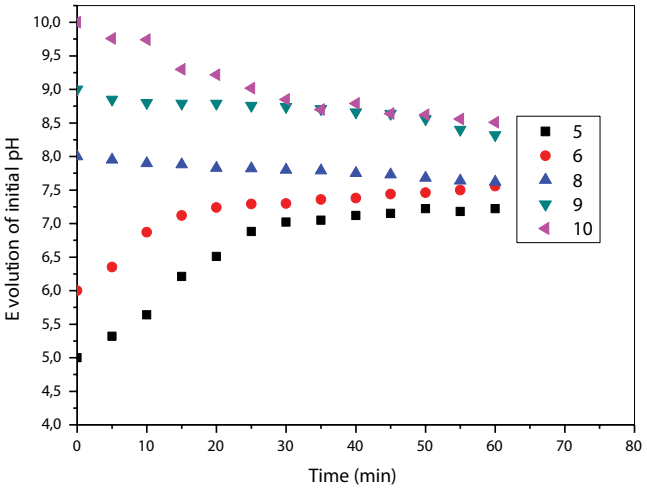

Fig. 3. Evolution of pH during electrocoagulation, for different initial pH values as a function of time (Int = 0.28 A, Dis = 1.2 cm, [NaCl] = 1 g/L).

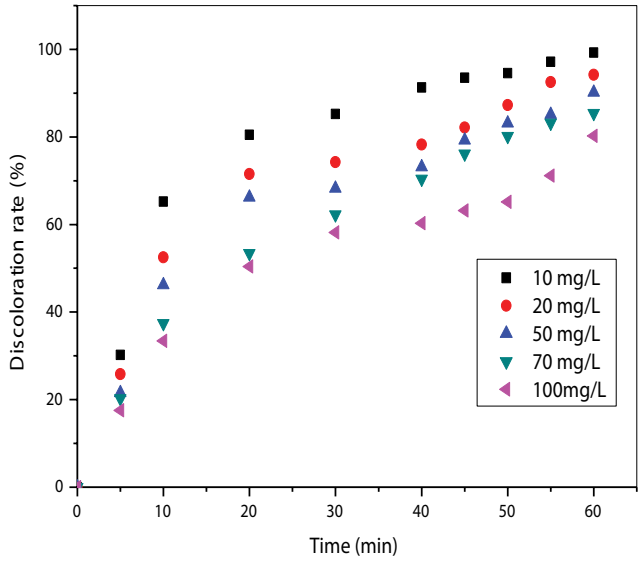

Fig. 4. Elimination of the dye under the effect of its initial concentration (mg/L) as a function of time (Int = 0.28 A, Dis = 1.2 cm, [NaCl] = 1 g/L, pH = 7.46).

On reading these results, it is observed that the removal efficiency of poly black decreases when its initial concentration increases. In addition, it is observed that the times required to reach dye removal percentages of 80%–85% are higher for the high concentrations.

3.1.3. Effect of electrodes dissolution

Fig. 5 shows metal dissolution as changes in dissolved metal concentration during effluent treatment. As predicted by Faraday's law, these figures clearly show that the rate of dissolution of the metal is proportional to the intensity of the current applied. Higher intensities dissolve more metals.

3.1.4. Effect of applied voltage

The cell voltage represents the reading of the voltage on the stabilized generator. This voltage value is a function of the intensity of the current, the electrode material and the effluent. For an effluent with high conductivity, the voltage is low if there is no deposit on the surface of the electrodes which would prevent the passage of current. Fig. 6 shows that the voltage, for each current density, evolves independently of all other densities. The highest densities evolve on lines of higher tensions.

The cell voltage formula can be simplified to (neglecting the over voltages at the electrodes and the no-load voltage U_0):

$$U = R \cdot I \tag{12}$$

where R, the cell resistance, depends on the conductivity of the effluent and I is the current intensity. We can therefore understand why the highest current densities correspond to higher cell voltages given that the intensity increases and the effluent remains the same, the ohmic resistance does not vary either.

3.2. Modeling results

3.2.1. Experimental design

To optimize the electrocoagulation process, each parameter in the design (RSM) was studied at five different levels (–a, –1, 0, +1, + a). The choice of five levels for each variable is required by this design in order to explore the region of the response surface near the optimum. In this work, three independent variables are studied; therefore, for k = 3, it would take 8 cubic points, 6 axial points and 6 central bridges, which makes a total of 20 experiments for a second-order polynomial model. Using three factors, the value of a equals 1.68.

The parameters chosen, within the framework of this study are: the initial pH, the distance between the electrodes and the intensity of the current, the ranges and the levels of these parameters are represented in Table 1, with the discoloration rate (Y) constitutes the response variable.

The results of the experiments of the composite design are shown in Table 2 which shows the statistical combinations of the independent variables X_1 (initial pH), X_2 (distance between electrodes), X_3 (current intensity), with the discoloration yields (Y) for different treatment times: 30, 45 and 60 min.

The results of the experimental design were investigated using Minitab Software (version 17) to estimate the response of the dependent variable of all experiments. The central composite design consists of three independent variables (pH, distance between electrodes, current intensity). The experiment was done with the values of the

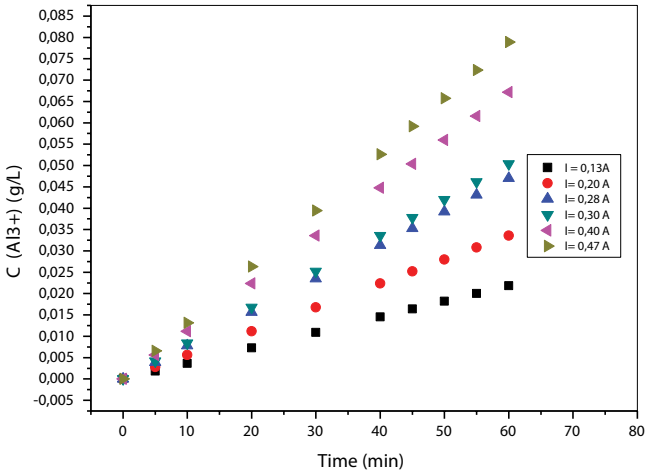

Fig. 5. Evolution of dissolved aluminum concentration during treatment by electrocoagulation (Int = 0.28 A, Dis = 1.2 cm, [NaCl] = 1 g/L, pH = 7.46).

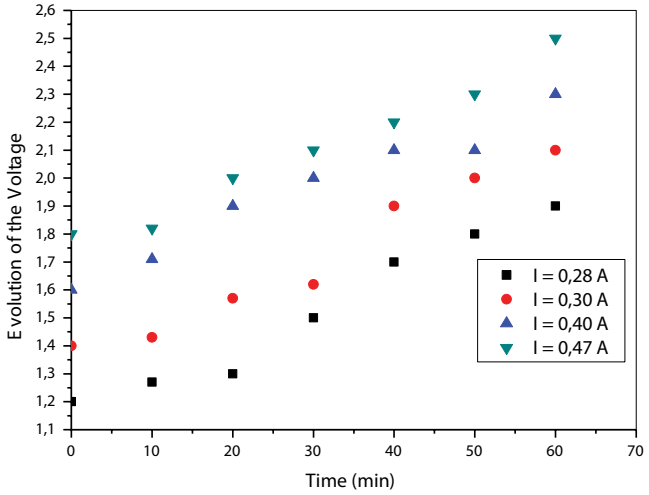

Fig. 6. Evolution of voltage as a function of time during treatment for different current intensities (pH = 7.46, Dis = 1.2 cm, [NaCl] = 1 g/L).

Table 2
Experimental ranges and levels of independent test variables

Factors \ Domain	–a	–1	0	1	+a
X_1: Initial pH	3.30	5	7.5	10	11.70
X_2: Distance (cm)	0.64	2	4	6	7.36
X_3: Current intensity (int) (A)	0.13	0.20	0.3	0.4	0.47

design information that has been combined by MINITAB (Table 3). The discoloration rate responses (Y) for different electrocoagulation treatment times are represented by the following second-order models:

- For t = 30 min

$$Y_{30\min} = 90.160 - 0,885\text{pH} - 1.231\text{Dis} - 1.731\text{Int} - 5,420\text{pH}\times\text{pH} - 1.123\text{Dis}\times\text{Dis} - 3,053\text{Int}\times\text{Int} - 0.235\text{pH}\times\text{Dis} + 0.240\text{pH}\times\text{Int} + 0.520\text{Dis}\times\text{Int} \quad (13)$$

- For t = 45 min

$$Y_{45\min} = 95.486 - 1.032\text{pH} - 1.248\text{Dis} - 1.897\text{Int} - 5.285\text{pH}\times\text{pH} - 0.735\text{Dis}\times\text{Dis} - 2.400\text{Int}\times\text{Int} - 0.321\text{pH}\times\text{Dis} + 0.566\text{pH}\times\text{Int} + 0.179\text{Dis}\times\text{Int} \quad (14)$$

- For t = 60 min

$$Y_{60\min} = 98.355 - 1.192\text{pH} - 1.493\text{Dis} - 0.636\text{Int} - 3.598\text{pH}\times\text{pH} - 0.727\text{Dis}\times\text{Dis} - 1.765\text{Int}\times\text{Int} - 0.321\text{pH}\times\text{Dis} + 0.510\text{pH}\times\text{Int} + 0.116\text{Dis}\times\text{Int} \quad (15)$$

The values of the coefficients of determination of the discoloration rate for t = 30, 45 and 60 min are R^2 = 94.84%, 96.29% and 96.26%, respectively, suggesting a high significance of the obtained models, which are in good agreement with the experimental data, and all obtained values met good adequacy and satisfactory level. It is also observed that the polynomial coefficients of the models obtained for different treatment times are a little different indicating that the reaction behavior differs with time.

3.2.2. Contours and response surfaces

The study of contour plots and response surfaces provides a simple method of optimizing the processing rate and identifying interactions between variables. Each curve represents, in our case, an infinity of combinations between two variables when the third one is maintained at a constant level. From the consumption point of view it is preferable to use a current of –1, and according to the literature the optimal distance for electrocoagulation (1 cm) has been recommended by several authors [23,24], for this in this case the distance is fixed at –1, and the initial pH at the centered value 0.

Figs. 7–9 show that we can have a decolorization greater than 90% (dark green band) at t = 30 min, 95% at t = 45 min, 98% at t = 60 min, when the values of the parameters to be controlled are minimal. By increasing these values, the decolorization gradually decreases. If we set two parameters as minimum values, the decolorization exceeds 99% and only if the initial pH is in the center.

Figs. 10–12 show 3D plots that describe the effect of initial pH, applied current and electrodes distance on removals efficiency of dye. It can be concluded (at t = 60 min) that the efficiency will be optimum when the current intensity is at a level between –1.0 to 0.0 (between 20 to 30 A), distance between electrodes at a level between –a to –1.0 (0.64 to 2 cm), and pH at a level between –1 to 0 (5 to 7.5).

3.2.3. Interaction effect: pH-applied current and pH-distance of electrodes

The initial pH has most effect on treatment efficiency, it's the responsible of formation of the different aluminum species in solution. It should be noted that in this case, the distance between the electrodes is set to the minimum value (2 cm). When the initial pH is set at 5, for an increase in current intensity of 0.2–0.4 A; it can be seen that the rate of elimination of the dye remains almost constant (80%–85%), the same remark was observed for an initial pH of 10; however, for a pH of 7.5 (middle of the study interval) a maximum of dye elimination is observed specially for a current intensity of 0.3 A (>90%), these observations concern all the treatment periods (30, 45 and 60 min); the optimum is obtained in the middle of the two study intervals (pH and current intensity). Working at high current intensities can cause other side reactions near the anode. For example, direct oxidation of the pollutant and formation of oxygen may occur. The production of oxygen in the vicinity of the anode illustrated in the following reaction Eq. (16) plays a negative role in the efficiency of electrocoagulation.

$$4OH^- \rightarrow O_2 + H_2O + 2e \quad (16)$$

The pH and the electrodes distance can have an important effect on elimination of dye; in this part of study, the current intensity is fixed at 0.2 A. The optimum is obtained in the middle of the pH study interval (7.5) and for a distance varying from 2 to 6 cm; for a treatment time of 30 min it can exceed 85%; and for the same study intervals, a dye removal efficiency of 90% to 95% is obtained for a treatment of 45 min, and more than 95% for a distance of less than 4 cm and for the same pH value (7.5).

In all the figures concerning the effect of pH, the maximum dye elimination is obtained around 7, which has been observed by several authors [25]. Thus, it is also found that amorphous $Al(OH)_3$ has minimum solubility in the pH range of 6.5–7.8, which represents an optimum interval for better coagulation [26]. When the pH is between 4 and 9, the Al^{3+} and OH^- produced by the electrodes react to form different oligomeric species such as $Al_6(OH)_{15}{}^{3+}$, $Al_7(OH)_{17}{}^{4+}$, $Al_{13}(OH)^{5+}{}_{34}$, to finally transform into an amorphous compound insoluble in water: Al $(OH)_{3(S)}$, via complex polymerization/precipitation kinetics [27]. Adsorption of Al^{3+} in solution at high pH, producing either the solid $Al(OH)_3$ or the monomeric anionic species $Al(OH)_4^-$, depending on the pH of the solution which strongly depends on the chemical structure of the dye. The $Al(OH)_3$ formation process is therefore active for a pH range from 4 to 9.

This pH range encompasses the initial pH optimum found in our case (pH = 7.5). However, pH strongly affects the size of hydrogen bubbles. The typical size of the bubbles produced during electrocoagulation by aluminum electrodes varies between 20 and 70 μm [28].

3.2.4. Effect of applied current and distance of electrodes

In any electrochemical process, current applied is the most significant parameter. The supplied current determines the dissipation of metal ions at the anode; thus,

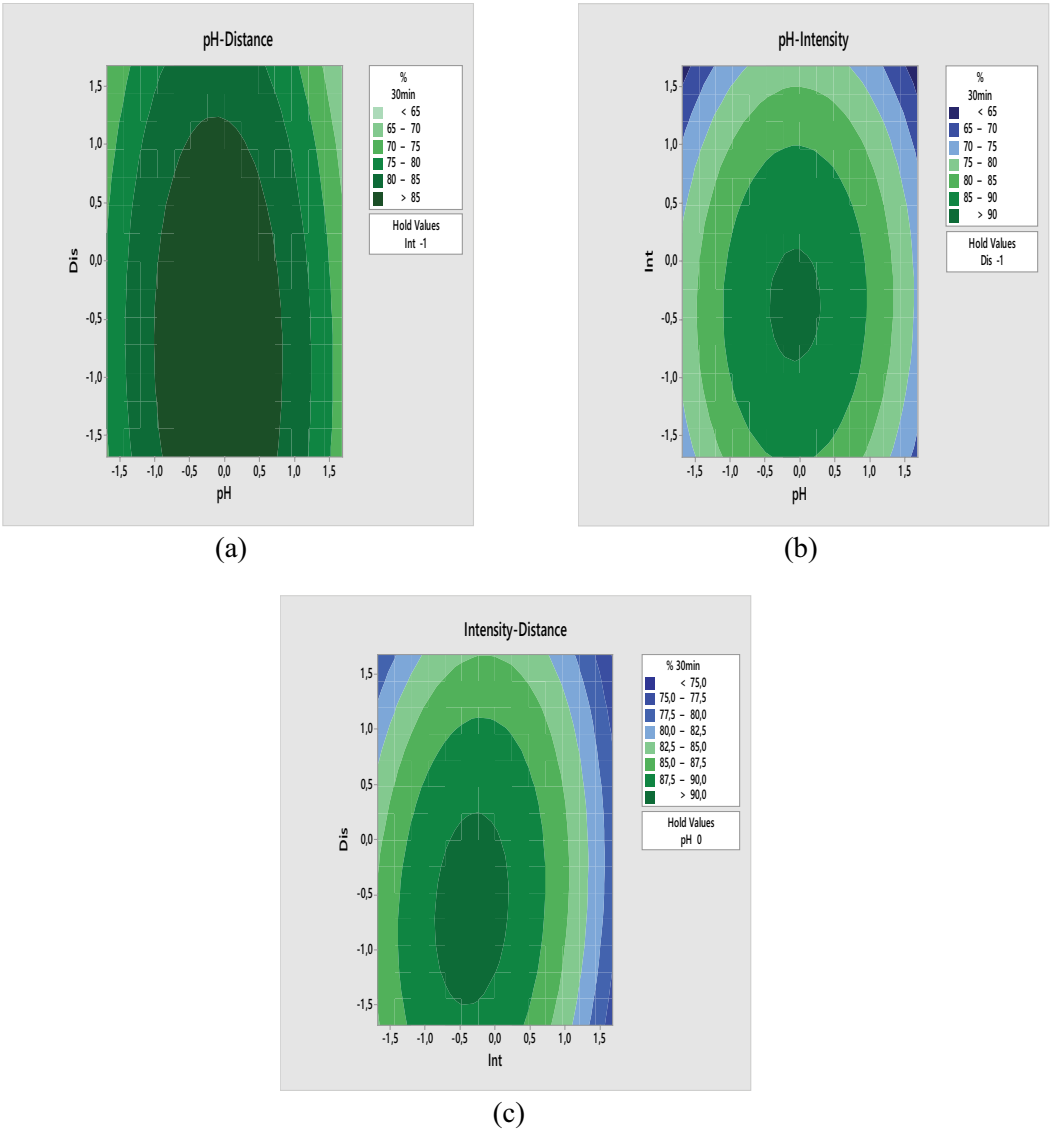

Fig. 7. Contours plots of the discoloration rates for each pair of variables in coded values when the third is set for distance (–1) and intensity (–1) or mean for initial pH, at t = 30 min (pH-Dis (a), pH-Int (b) and Dis-Int (c)).

the EC process gets considerably affected [29]. To study the effect of applied current and distance of electrodes on elimination of dye, the pH is maintained at the middle of interval (7.5). The distance between the electrodes directly influences the ohmic drop. Indeed, for a constant current intensity (I), the reduction in the distance between the two electrodes favors a reduction in the resistance (R) and consequently a reduction in the voltage (U) and in the energy consumption [30]. According to Faraday's law, the applied intensity is directly proportional to the quantity of coagulants produced at the anode. An increase in electric current favors the reaction described by Eq. (4), inducing better elimination of pollutants [31].

An increase in intensity is also favorable to the formation of dihydrogen at the cathode (Reaction 2), thus allowing the flotation of metal hydroxides towards the surface of the reactor. According to Figs. 3 and 4, a remarkable effect of the two parameters (current intensity and distance between electrodes) on the dye elimination rate was observed. Maximum dye removal is obtained (>90%) for distances between the electrodes varying from –1 (2 cm) to 0 (4 cm) for current intensities from –0.5 (0.25 A) to 0 (0.3 A) and this for 30 min of treatment; and >96% for –1.5 (1 cm) up to almost 0 (4 cm) distance for a current of –0.5 (0.25 A) for 45 min of reaction, finally for a treatment of 60 min, a yield >98% is obtained for an intensity varying from –1 (0.2 A) to 0.5 (0.35 A) for the same distances.

To explain the role of current intensity in the removal of pollutants in general, Kobya et al. [32] showed that at high current densities, the anodic dissolution of aluminum increases, resulting in a greater rate of precipitates necessary for the elimination of pollutants. Likewise, the level of production of gas bubbles increases, their sizes decrease with the increase of the intensity of the current applied,

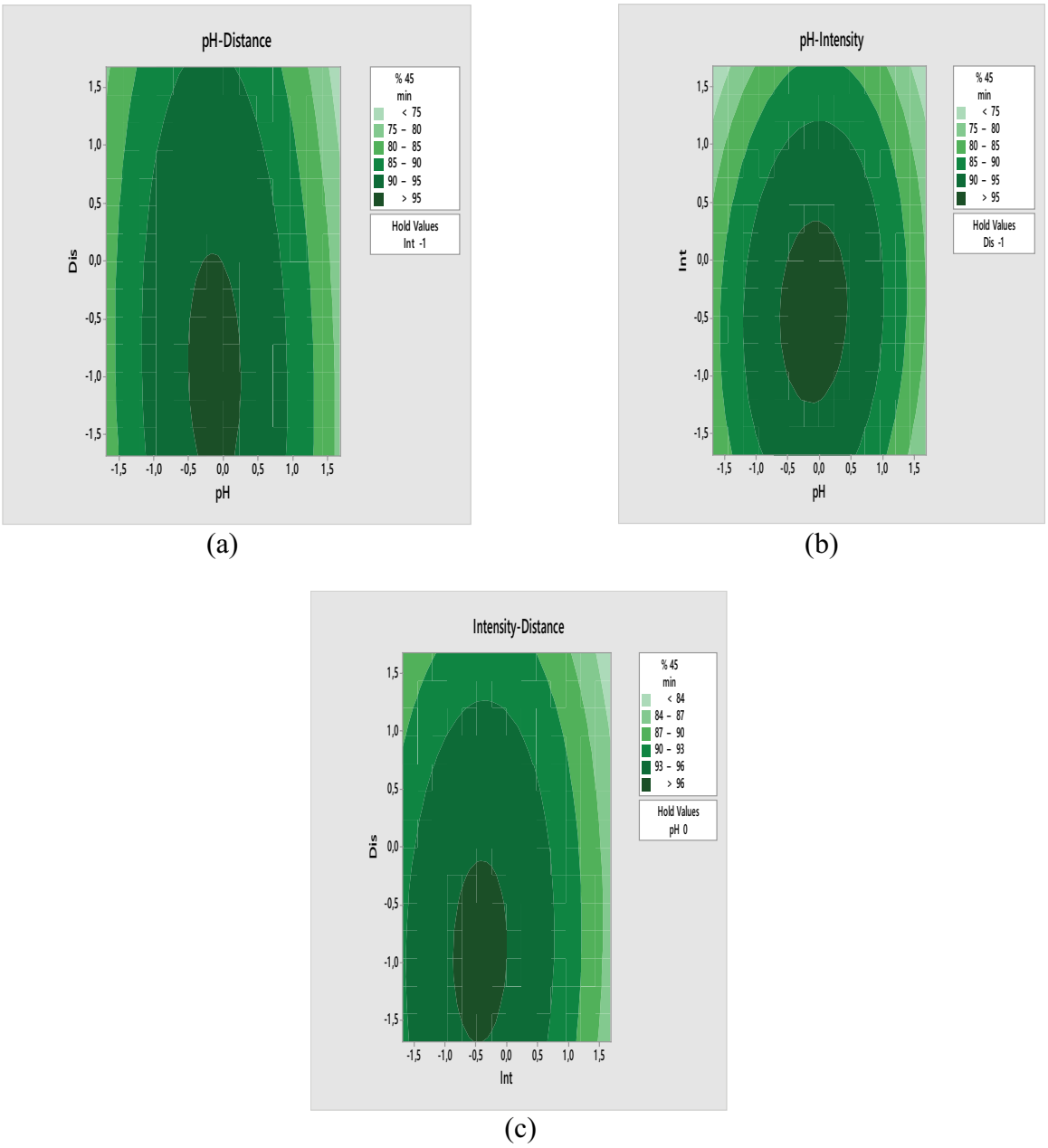

Fig. 8. Contours plots of the discoloration rates for each pair of variables in coded values when the third is set for distance (–1) and intensity (–1) or mean for initial pH, at t = 45 min (pH-Dis (a), pH-Int (b) and Int-Dis (c)).

which can be beneficial for a great performance of pollutant removal by hydrogen flotation [33].

3.2.5. Factor effects and interactions

The individual, quadratic and interaction effects of the different factors have been presented.

From Fig. 13 it can be seen that for the graph of the main effects (at t = 30, 45 and 60 min): the initial pH, the distance and the intensity have a negative effect on the discoloration rate, and for the effects interactions: strong interactions between (pH-Distance), negligible interactions between (pH-Intensity) and (Distance-Intensity) at t = 30 and 45 min, but it was observed a negligible interaction between (Distance-Intensity) and strong interaction (pH-Intensity), (pH-Distance) at t = 60 min.

3.2.6. Optimization diagram

It concerns a response optimization tool from Minitab that illustrates the effects of different experimental parameters on the predicted responses for a stored model. The optimization of electrocoagulation parameters is shown in the Fig. 18.

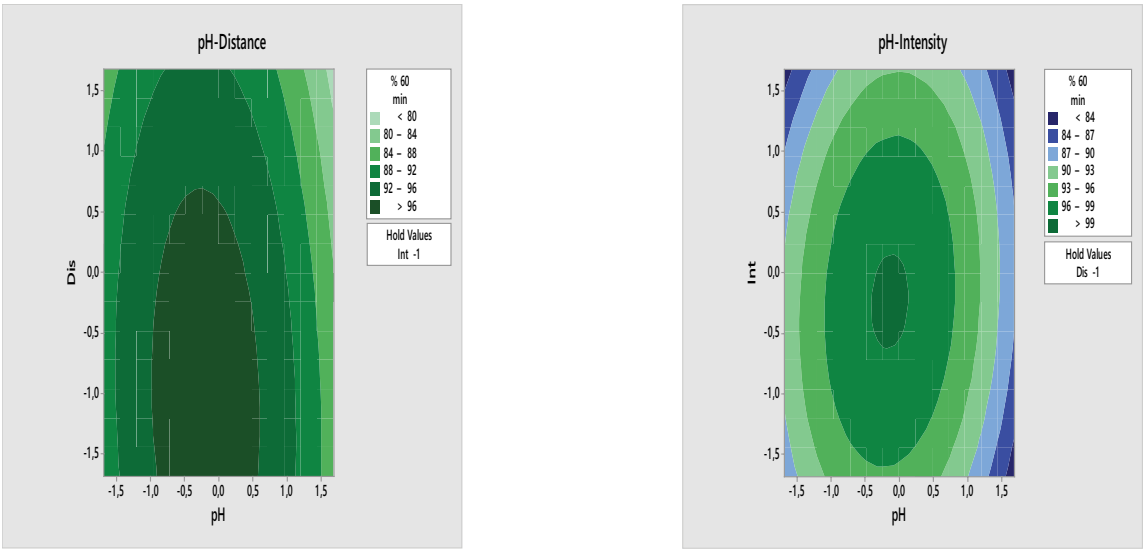

Fig. 9. Contours plots of the discoloration rates for each pair of variables in coded values when the third is set for distance (–1) and intensity (–1) or mean for initial pH, at t = 60 min (pH-Dis (a), pH-Int (b) and Dis-Int (c)).

Fig. 10 shows the optimum discoloration rate by the electrocoagulation treatment for different studied times.

- At t = 30 min, the maximum of discoloration rate is 90.8663%, corresponding to the coded initial pH value '–0.0849' (7.28), the distance between the electrodes '–0.6285' (2.74 cm), and the current intensity '–0.3567' (0.26A).
- At t = 45 min, the maximum of discoloration rate obtained is 96.505% corresponding to the coded values of the initial pH '–0.0849' (7.29), the distance between electrodes '–0.8664' (2.27 cm) and the current intensity '–0.4247' (3.15A).
- At t = 60 min the maximum of discoloration rate reached 99.2675% corresponding to the coded initial pH value '–0.1529' (7.46), the distance between the electrodes '–1.0023' (1.2 cm), and the current intensity '–0.2208' (0.28 A).

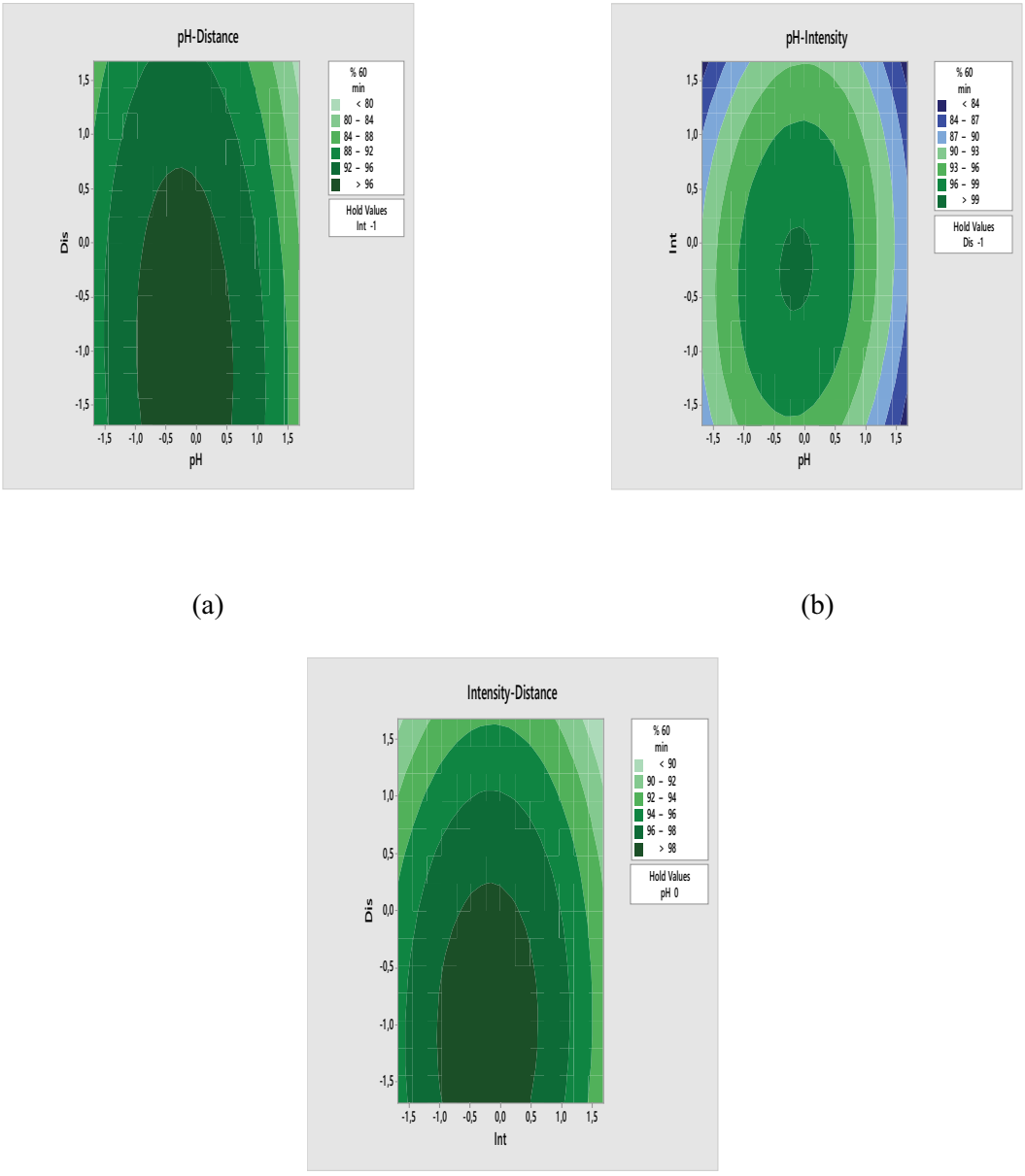

Fig. 10. Response area of the discoloration rate for each pair of variables under coded values when the third variable is set for distance (–1) and intensity (–1), or mean for initial pH, at 30 min (pH-Dis (a), pH-Int (b) and Dis-Int (c)).

Therefore, 60 min represents an optimal treatment time; a neutral pH of the solution, a small distance between the electrodes which can avoid the ohmic drop and a relatively low current intensity have obtained the best treatment performance by electrocoagulation process.

3.2.7. Comparison between experimental and calculated results

In order to validate the obtained models, a comparative study between the experimental and calculated results was carried out, concerning the discoloration rates for the three times studied.

All the curves presented in Figs. 15–17 show good convergence between the experimental and calculated results, and that for different times; the majority of the points are superimposed, which shows that the models obtained can have a good presentation of the EC process.

3.3. Effectiveness of optimized electrocoagulation parameters for treatment of real textile effluents

Efficacy of the optimized electrocoagulation parameters (pH = 7.46; intensity = 0.28 A; interelectrode distance = 1.2 cm) were assessed for chemical oxygen demand (COD), color, turbidity of real textile effluents. Fig. 14 shows

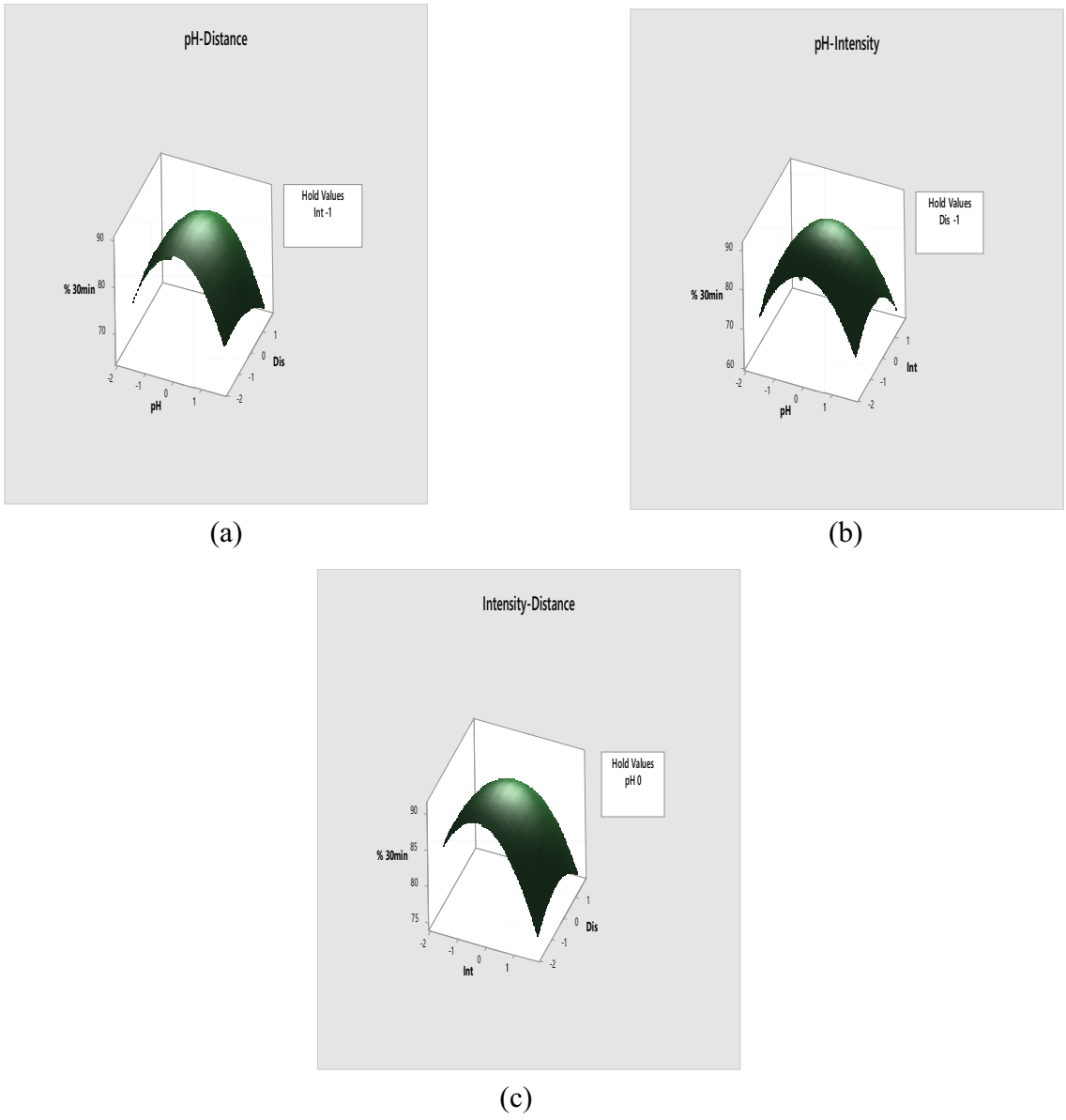

Fig. 11. Response area of the discoloration rate for each pair of variables under coded values when the third variable is set for distance (–1) and intensity (–1), or mean for initial pH, at 45 min (pH-Dis (a), pH-Int (b) and Dis-Int (c)).

the results after and before treatment of the textile effluent by electrocoagulation.

The average value of λ_{max} was found to be 571 nm. It was investigated during the course of treatment that almost all the color of real textile effluents get vanished producing a virtually color less solution (color removal efficiency ~ 97%). COD and turbidity removal efficiency were also appeared to be promising (90.31%), (91.18%), respectively, under the mentioned operating conditions.

3.4. Characterization of recycled electrodes before and after treatment by electrocoagulation

Tables 4 and 5 represent the elemental composition of the metal of the electrodes produced by XRF (X-ray fluorescence).

During the electrocoagulation process, the amount of aluminum reduces dramatically compared to the other components, which verifies the presence of the electrocoagulation mechanism (which is based on the reactions of anodic dissolution).

4. Energy consumption and treatment cost of EC

In this part, the estimated cost of the treatment by electrocoagulation process and consumption of energy are carried out, after 60 min of electrolysis with recycled aluminum electrodes; the power consumption and the cost of treatment are calculated for different applied current intensities.

The operational cost is given by the following formula [17]:

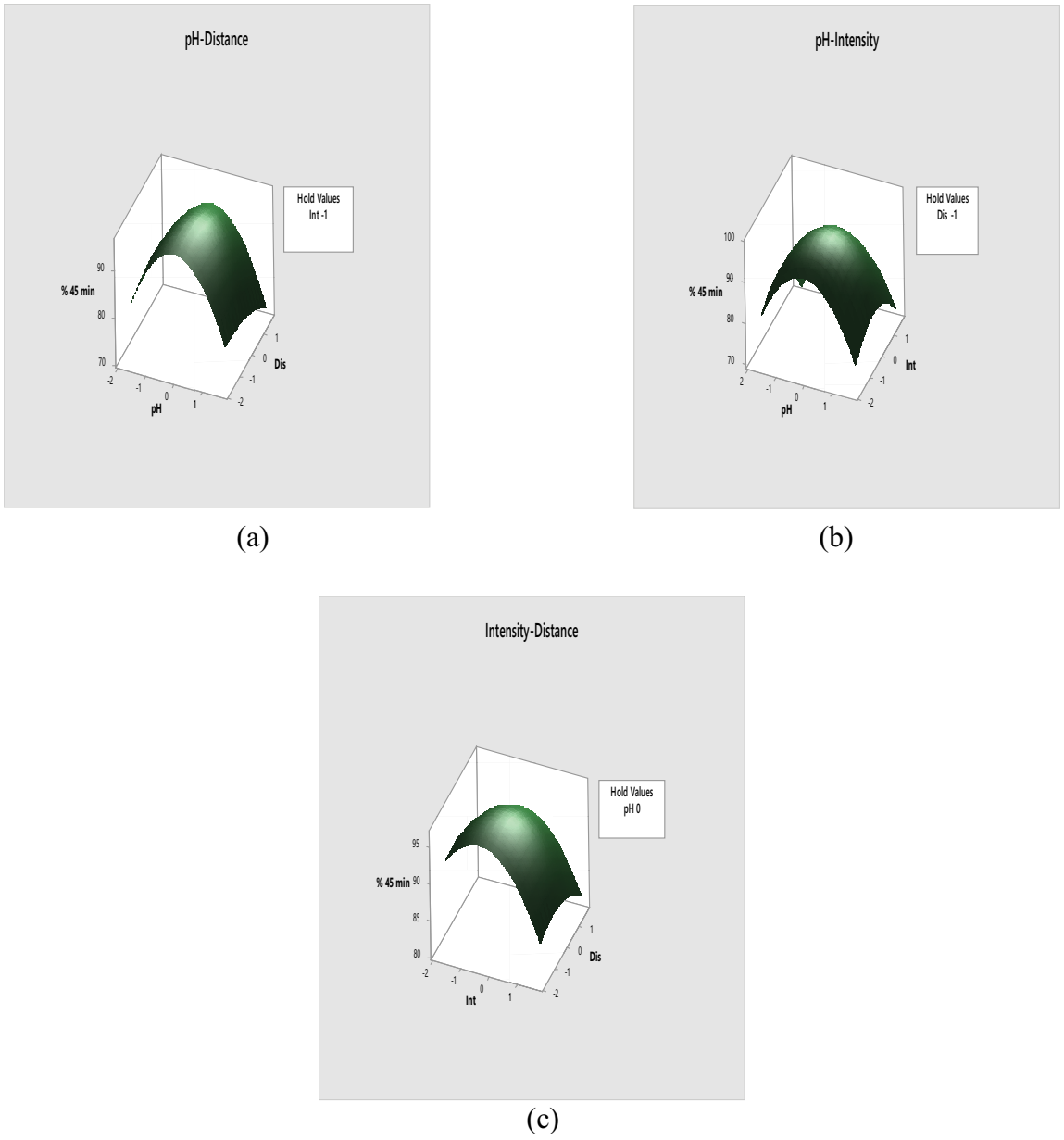

Fig. 12. Response area of the discoloration rate for each pair of variables under coded values when the third variable is set for distance (–1) and intensity (–1), or mean for initial pH, at 60 min (pH-Dis (a), pH-Int (b) and Dis-Int (c)).

$$\text{Cost} = aC_{\text{energy}} + bC_{\text{Al}^{3+}\text{electrode}} \quad (17)$$

with a and b are ratios concerning the price of energy on the Algerian Market; C_{energy}: consumed energy; $C_{\text{Al3+}}$ electrode: aluminum consumption electrode

They are of the order of:

a = 1,904 DZD/kWh (DZD: Algerian dinar) [34].
And b = 250 DZD/kg of recycled aluminum.

The results obtained are collated in Table 6.

Table 6 shows that energy consumption increases with current intensity. These observations are in good agreement with data already reported in the literature for wastewater from textile industries. Therefore, the intensity of the electric current is an important parameter for reducing the energy input [35]. And from the results of calculating the cost of electrocoagulation treatment, it can be seen that the latter increases with the intensity of the current. On the other hand, it can be considered that for an intensity of 0.28 A (optimal intensity), the treatment cost is 1.06×10^{-3} DZD/L (6.80×10^{-6} $/L) of treated water for recycled aluminum electrodes.

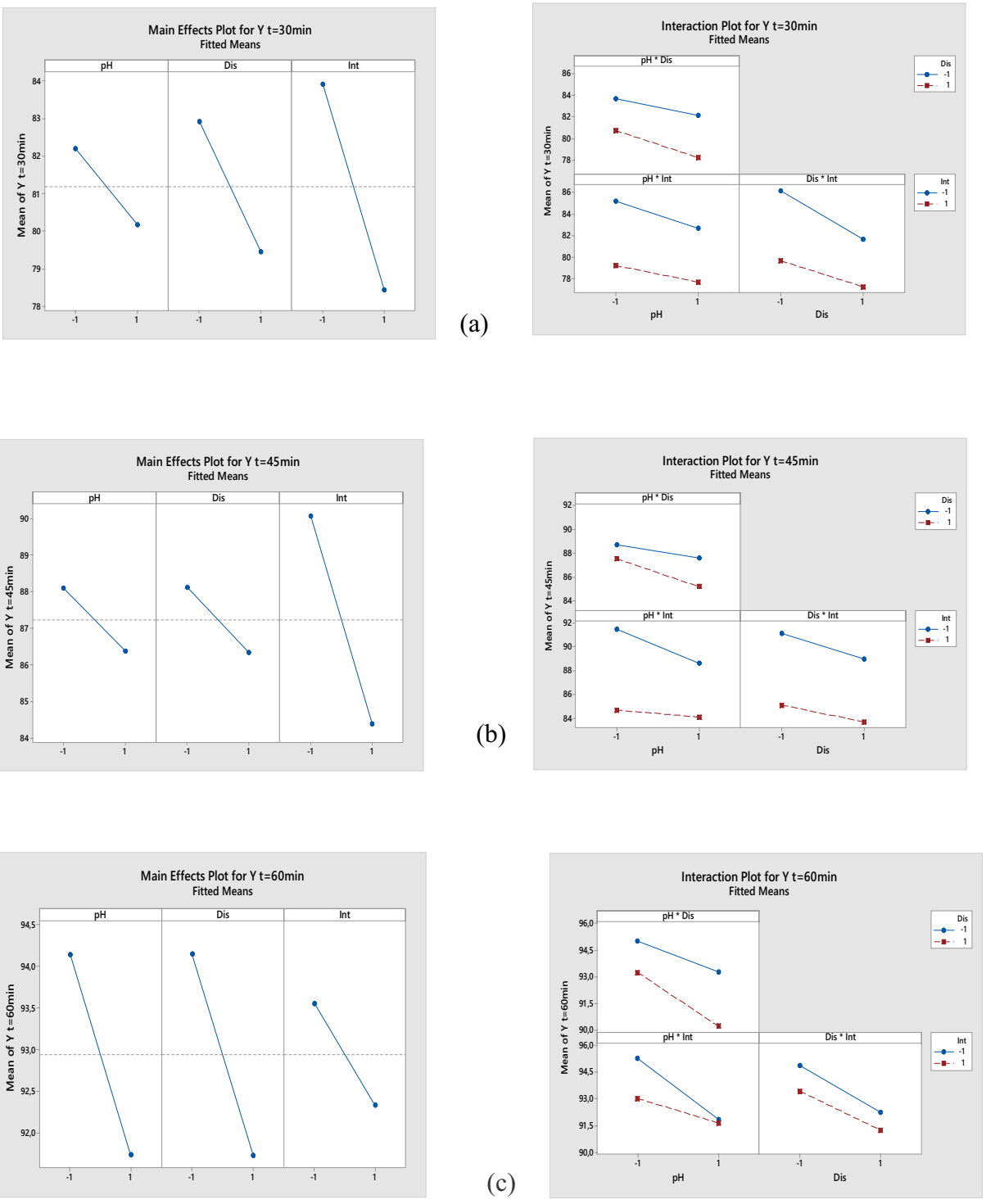

Fig. 13. Effects and interactions of factors, for 30 min (a), 45 min (b) and 60 min (c)) obtained by complete factorial design.

5. Conclusion

In this study, response surface methodology approach using central composite design was applied to develop a mathematical model and to optimize process parameters (pH, distance inter electrodes and current intensity) for the textile dye removal by electrocoagulation process using recycled aluminum electrodes.

During modeling, the values of the coefficients of determination of the discoloration rate for t = 30, 45 and 60 min are R^2 = 94.84%, 96.29% and 96.26%, respectively, suggesting a high significance of the obtained models, which are in

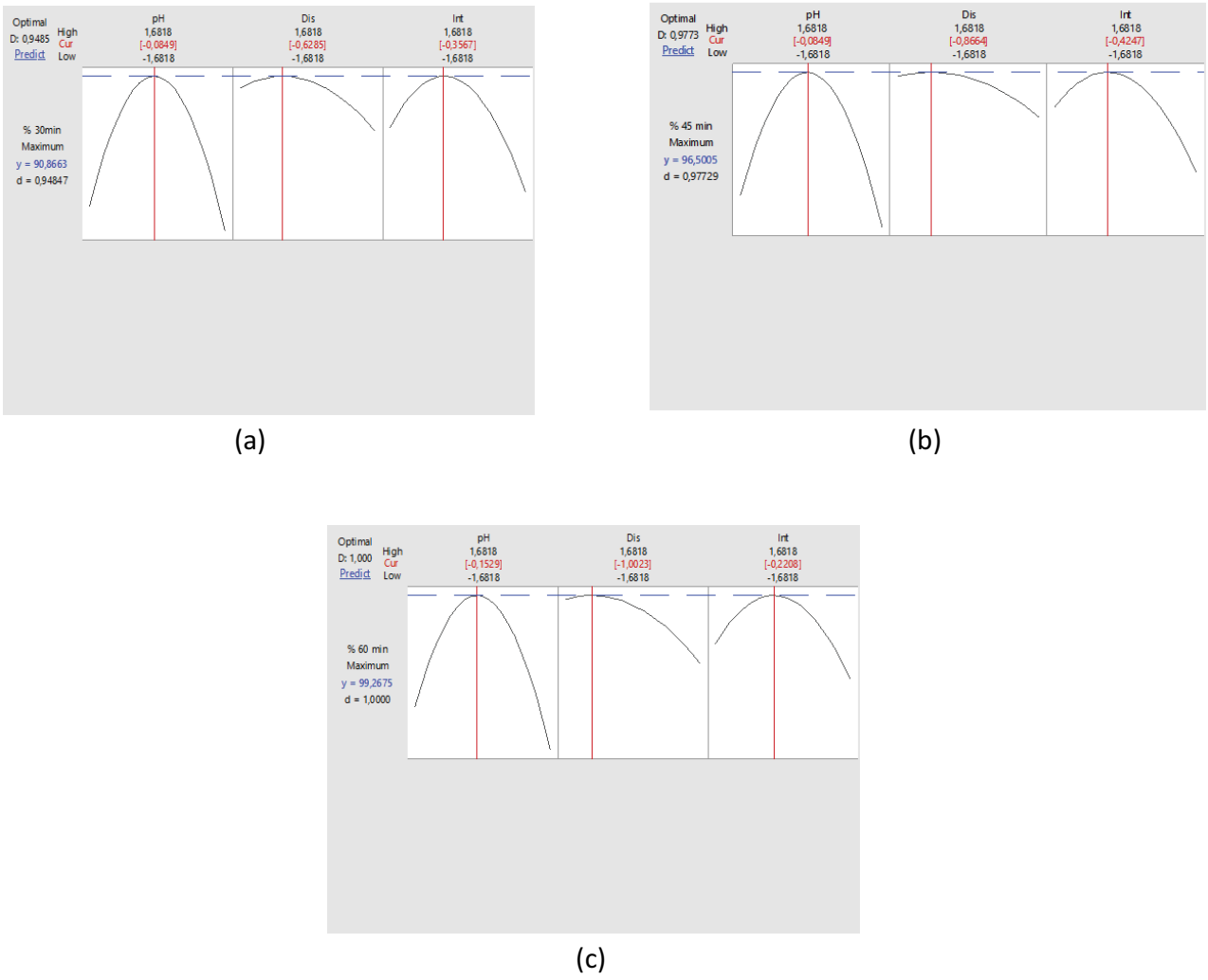

Fig. 14. Optimization of electrocoagulation parameters for different time of treatment (a) 30 min, (b) 45 min, and (c) 60 min).

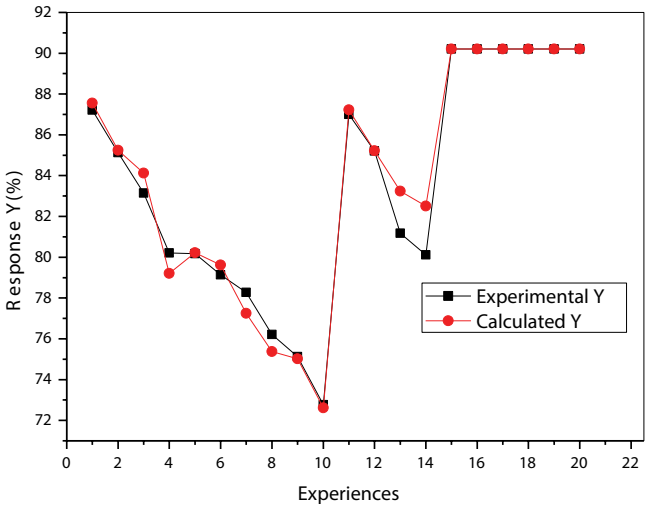

Fig. 15. Comparison of experimental and calculated results, at: t = 30 min.

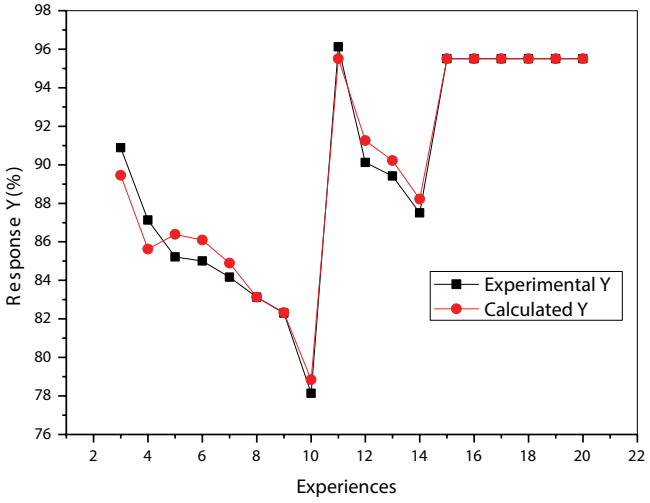

Fig. 16. Comparison of experimental and calculated results, at: t = 45 min.

good agreement with the experimental data, and all obtained values met good adequacy and satisfactory level. The interaction between the process variables was assessed using the obtained 2D and 3D plots.

The optimal values of the different parameters (pH, distance between electrodes and current intensity) give very satisfactory yields.

- For a treatment time of 30 min, the maximum discoloration rate reaches 90.8663% corresponding to an initial pH of 7.28, a distance between the electrodes of 2.74 cm and a current intensity of 0.26 A;
- For t = 45 min the maximum discoloration rate is 96.505% corresponding to an initial pH of 7.29, a distance of 2.27 cm and a current intensity of 3.15 A.
- For t = 60 min, the maximum rate of discoloration exceeds 99% for an initial pH of 7.46, a distance of 1.2 cm with an intensity of 0.28 A; which corresponds to the optimum parameters for the elimination of the poly black dye by the electrocoagulation process.

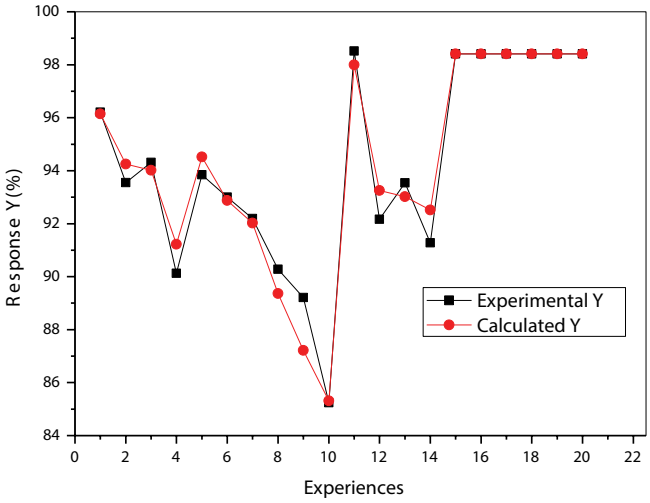

Fig. 17. Comparison of experimental and calculated results, at: t = 60 min.

Fig. 18. Real textile effluents before and after electrocoagulation treatment.

Table 3
Composite plan centered with 3 factors

Experiences	Coded values			Real values			Discoloration rate in percent (Y)		
	X_1	X_2	X_3	X_1	X_2	X_3	$Y_{30\ min}$	$Y_{45\ min}$	$Y_{60\ min}$
1	–1	–1	–1	5	2	0.2	87.21	92.11	96.21
2	+1	–1	–1	10	2	0.2	85.12	90.17	93.55
3	–1	+1	–1	5	6	0.2	83.14	90.88	94.31
4	+1	+1	–1	10	6	0.2	80.21	87.13	90.13
5	–1	–1	+1	5	2	0.4	80.17	85.21	93.85
6	+1	–1	+1	10	2	0.4	79.14	85.01	93.00
7	–1	+1	+1	5	6	0.4	78.28	84.17	92.19
8	+1	+1	+1	10	6	0.4	76.21	83.21	90.28
9	–a	0	0	3.3	4	0.3	75.13	82.45	89.21
10	+a	0	0	11.70	4	0.3	72.77	78.14	85.24
11	0	–a	0	7.5	0.64	0.3	87.00	96.12	98.52
12	0	+a	0	7.5	7.36	0.3	85.21	90.21	92.17
13	0	0	–a	7.5	4	0.13	81.17	89.41	93.54
14	0	0	+a	7.5	4	0.47	80.12	87.50	91.28
15	0	0	0	7.5	4	0.3	90.21	95.50	98.41
16	0	0	0	7.5	4	0.3	90.21	95.50	98.41
17	0	0	0	7.5	4	0.3	90.21	95.50	98.41
18	0	0	0	7.5	4	0.3	90.21	95.50	98.41
19	0	0	0	7.5	4	0.3	90.21	95.50	98.41
20	0	0	0	7.5	4	0.3	90.21	95.50	98.41

Table 4
Elemental composition of the metal of the electrodes produced by X-ray fluorescence before treatment by electrocoagulation

Al	Si	P	S	Ti	V	Cr
309.7 KCps 18.100%	8.9 KCps 0.508%	2.2 KCps 0.0234%	20.6 KCps 0.236%	0.1 KCps 0.002%	3.9 KCps 0.082%	 14.390%
Mn	Fe	Co	Ni	Cu	Zn	As
56.3 KCps 0.858%	 65.142%	5.0 KCps 0.023%	9.3 KCps 0.251%	6.8 KCps 0.139%	1.2 KCps 0.013%	0.0 KCps 0.006%
Zr	Nb	Mo	Sn	Sb	Ta	W
1.0 KCps 0.000%	2.2 KCps 0.000%	58.9 KCps 0.164%	0.1 KCps 0.0041%	–0.1 KCps 0.011%	–0.2 KCps 0.009%	–0.3 KCps 0.038%
Pb	Bi	Scale of inte				
0.2 KCps 0.000%	–0.1 KCps 0.000%	 0.8893				

Table 5
Elemental composition of the metal of the electrodes produced by X-ray fluorescence after treatment by electrocoagulation

Al	Si	P	S	Ti	V	Cr
237.9 KCps 14.595%	7.9 KCps 0.445%	2.2 KCps 0.0235%	17.7 KCps 0.205%	–0.1 KCps 0.001%	4.0 KCps 0.086%	 15.001%
Mn	Fe	Co	Ni	Cu	Zn	As
55.5 KCps 0.879%	 68.067%	5.0 KCps 0.023%	9.4 KCps 0.267%	6.8 KCps 0.147%	1.1 KCps 0.013%	0.1 KCps 0.007%
Zr	Nb	Mo	Sn	Sb	Ta	W
1.0 KCps 0.000%	2.1 KCps 0.000%	59.0 KCps 0.176%	0.1 KCps 0.0033%	–0.1 KCps 0.011%	–0.2 KCps 0.009%	–0.3 KCps 0.039%
Pb	Bi	Scale of inte				
0.2 KCps 0.000%	–0.0 KCps 0.000%	 0.9235				

Table 6
Energy consumption and cost of treatment by EC process

Experiences	Intensity (A)	Voltage (V)	C (Al^{3+}) (g/L)	b (DZD/g)	Consumed energy (kWh/L)	Cost (DZD/L)
Exp 1	0.13	1.5	0.0218	0.0055	9.75×10^{-5}	3.05×10^{-4}
Exp 2	0.2	1.7	0.0336	0.0084	1.70×10^{-4}	6.05×10^{-4}
Exp 3	0.28	1.9	0.0470	0.0117	2.66×10^{-4}	1.06×10^{-3}
Exp 4	0.3	2.1	0.0503	0.0126	3.15×10^{-4}	1.23×10^{-3}
Exp 5	0.4	2.3	0.0671	0.0168	4.60×10^{-4}	2.00×10^{-3}
Exp 6	0.47	2.5	0.0789	0.0197	5.88×10^{-4}	2.67×10^{-3}

- The optimized operational conditions of electrocoagulation were also applicable for assessing the treatment efficiency for real textile effluents, and for effectively eliminating of color.

The cost of treatment by this process (EC) with recycled aluminum is relatively low compared to other methods 1.06×10^{-3} DZD (6.80×10^{-6} $) /L of treated water, which corresponds to energy consumption of 2.66×10^{-4} kWh/L. Overall, the electrocoagulation process has proven to be effective, inexpensive, and a promising alternative to conventional treatment procedures for removing poly black dye.

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