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Modelling COD Removal from Slaughterhouse Wastewater by Electrocoagulation Using Response Surface Methodology

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Abstract: Modeling is an indispensable tool for a better wastewater treatment strategy. However, the modelling of slaughterhouse wastewater treatment by electrocoagulation can be difficult to achieve because of the various physico-chemical mechanisms involved. It is in this context that the objective of this study was to model and optimize COD removal and electrical energy consumption by response surface methodology (RSM) during the treatment of slaughterhouse wastewater by electrocoagulation (EC). For this purpose, a full factorial design (FD) was first used to observe the effect of experimental parameters (stirring speed, pH, time and current intensity) on COD removal and energy consumption. Then, a central composite design (CCD) was performed to optimize COD removal and electrical energy consumption. The optimum conditions are obtained at the stirring speed of 871 rpm, pH = 6.83; time of 80 min and current intensity of 1.85 A. By applying these optimal conditions for the treatment, reductions of 84 ± 1.08% of COD; 93.86 ± 0.91% of BOD; 97.80 ± 0.86% of turbidity and 99.62 ± 0.12% of PO_4^{3-} and an energy consumption of 9 $KWh.m^{-3}$ were obtained. Thus, this study reveals that RSM is an effective tool for the modeling and optimization of electrocoagulation.

Keywords: Slaughterhouse Wastewater, Response Surface Methodology, Electrocoagulation

1. Introduction

Food processing industries (slaughterhouses) are known to produce large volumes of wastewater from animal slaughter, production and plant cleaning units [1]. Slaughterhouse wastewater contains high loads of biodegradable organic compounds, nitrogen, phosphorus [2]. It also contains oils and fats, colloidal matter, blood and cellulose [3]; as well as residues of antibiotics, vaccines and a high load of pathogenic micro-organisms [4]. Therefore, their discharge without adequate treatment into the environment can reduce the level of dissolved oxygen in surface waters, cause eutrophication of aquatic environments, negatively affect biological life and human health [5].

To reduce the pollution load of these waters, several treatment methods exist. There are physico-chemical, electrochemical and biological methods. However, despite the strong reduction of organic matter by biological methods, they require a long processing time and give low yields of nutrient removal (N and P). Among the physico-chemical methods, coagulation-flocculation is limited by a high use of chemical reagents, which results in a high treatment cost. Finally, the successful application of electrochemical methods such as electroflotation, electrooxidation and electrocoagulation has been observed in the treatment of slaughterhouse wastewater [6].

Electrocoagulation is an interesting treatment method because it is simple to use, does not cause secondary pollution of the treated water and has good purification efficiency [7]. It is described as the electrochemical version of chemical coagulation-flocculation because it uses the same principles [8]. During electrocoagulation, the passage of electric current through the anodes generates metal cations by oxidation (equation (1)), hydroxyl ions (OH^-) and dihydrogen (H_2) by reduction of water at the cathode (equation (2)). In the reaction medium, OH^- react with metal cations to form metal hydroxides (equations (3)-(5)). The latter participate in the removal of pollutants by (i) precipitation, (ii) adsorption and (iii) co-precipitation.

Anode (Oxidation):

$$Fe \rightarrow Fe^{2+} + 2\,e^- \quad (1)$$

Cathode (Reduction):

$$2\,H_2O + 2\,e^- \rightarrow H_2 + 2\,OH^- \quad (2)$$

Reaction medium:

$$O_{2(g)} + 4Fe^{2+} + 2H_2O \rightarrow 4Fe^{3+} + 4OH^- \quad (3)$$

$$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2 \quad (4)$$

$$Fe^{3+} + 3OH^- \rightarrow Fe(OH)_3 \quad (5)$$

Moreover, during electrocoagulation, many chemicals (complexation) and physical (electro-flotation, electrostatic attraction) reactions occur simultaneously [8]. Under these conditions, the modelling of electrocoagulation by the classical method may be biased [6]. Indeed, the classical method consists of fixing one operating parameter and varying the others. Thus, this method does not take into account the interactions between the experimental parameters, which results in an approximate modelling of the responses studied. The response surface methodology (RSM) can therefore be used to overcome this problem. Indeed, it maps the response surface, and optimizes the statistical models by specific algorithms such as the desirability function [9]. Furthermore, this method has been successfully used for modelling and treating wastewater by electrocoagulation [10, 11]. However, few works on the treatment of slaughterhouse wastewater by RSM have been observed. Thus, the main objective of this study is to treat slaughterhouse wastewater by electrocoagulation using RSM. The aim is to (a) model and optimize the removal of COD and energy consumption and (b) evaluate purification performance of electrocoagulation on slaughterhouse wastewater.

2. Materials and Methods

2.1. Slaughterhouse Wastewater Collection

The wastewater was collected from the cattle slaughterhouse in the city of Yamoussoukro (Côte d'Ivoire). The slaughterhouse wastewater was first filtered through a 2 mm diameter filter in order to remove pieces of flesh and blood clots. It was then characterized and stored at 4°C before treatment.

2.2. Electrochemical Treatment

The electrocoagulation treatment was carried out in a 1.8 L batch reactor (Figure 1). Fe electrodes (anodes and cathodes) with a surface area of 110 cm^2 each were used. Stirring of the wastewater inside the reactor was carried out with a magnet bar. For all tests, a volume of 1.7 L of wastewater was used. The electrical current was supplied by a DC generator (elc AL781D France) with a maximum current and voltage of 5 A and 60 V. During the treatment, the current intensity and the treatment time were kept constant. After treatment, the treated water was left to settle for 2 hours and the supernatant was collected and characterized.

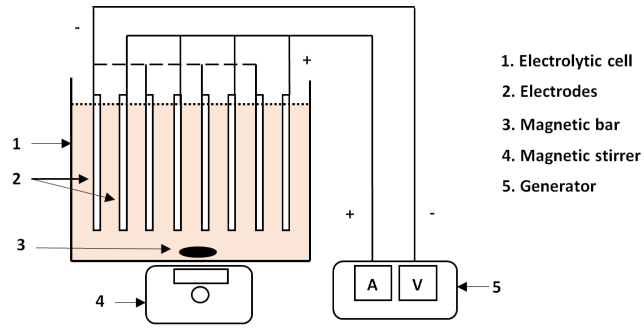

Figure 1. *Experimental electrocoagulation device.*

2.3. Experimental Design Methodology

EDM develops a statistical model that describes a complex phenomenon with a minimum number of experiments [12]. EDM studies the effect of the interactions of the parameters involved by varying them simultaneously. In this study, a full factorial design (FD) was first used to evaluate the effects of the main factors and their interactions on COD removal and energy consumption (assays 1-16). Then, a central composite design (CCD) was used to optimize the treatment (assays 17-31). The variables studied were agitation speed (X_1), pH (X_2), treatment time (X_3) and current intensity (X_4) (Table 1). The matrix, experimental range and responses are presented in Table 2. The variables and their variation limits were previously determined by exploratory tests. The NEMROD-W software (Version 9901 French, LPRAI-Marseille Inc., France) was used to calculate the model coefficients and for the modelling. Finally, ANOVA was used to determine the significant effects of the variables and to deduce the robustness of the models.

The FD and CCD models are given by equations (6) and (7):

$$Y\,(\%) = b_0 + \sum b_i X_i + \sum\sum b_{ij} X_i X_j + \varepsilon;\ i \neq j \quad (6)$$

Where *Xi* and *Xj*, the coded variables (-1 or +1): b0 the mean of the responses obtained, bi the main effect of factor *i* for the response *Y*, *bij* the interaction effect between factors *i* and *j* for the response and represents the error on the response.

$$Y\,(\%) = b_0 + \sum b_i X_i + \sum\sum b_{ij} X_i X_j + \sum b_{ii} X_i^2\ \varepsilon;\ i \neq j \quad (7)$$

Where, *Y*, *b0*, *bi*, *bii*, *bij*, *Xi* and *Xj* represent the predicted response, the constant coefficient, the linear coefficient, the interaction coefficient, the quadratic coefficient, and the coded values of the factors, respectively.

Table 1. *Experimental domains.*

Variables coded (Xi)	Factors (Ui)	Experimental domain				
		α_{min} = -2.00	-1	X_0	+1	α_{max} = +2.00
X_1	Stirring speed	700	800	900	1000	1100
X_2	pH	3	4	5	6	7
X_3	Time	60	75	90	105	120
X_4	Current intensity	1	1,375	1,75	2,125	2,5

Table 2. *Experimental matrix, experimentation design and responses of RSM.*

Essay	Experimental design				Experimentation plan				Responses	
	X_1	X_2	X_3	X_4	U_1: Stirring speed	U_2: pH	U_3: Time	U_4: Current intensity	COD removal efficiency: Y_1 (%)	Energy consumption: Y_2 (kW.h.m^{-3})
1	-1	-1	-1	-1	800	4.00	75	1.375	54.65	3.14
2	1	-1	-1	-1	1000	4.00	75	1.375	39.23	3.14
3	-1	1	-1	-1	800	6.00	75	1.375	71.00	4.05
4	1	1	-1	-1	1000	6.00	75	1.375	66.22	3.95
5	-1	-1	1	-1	800	4.00	105	1.375	82.60	4.89
6	1	-1	1	-1	1000	4.00	105	1.375	79.77	4.04
7	-1	1	1	-1	800	6.00	105	1.375	89.30	6.23
8	1	1	1	-1	1000	6.00	105	1.375	75.43	7.08
9	-1	-1	-1	1	800	4.00	75	2.125	66.66	5.79
10	1	-1	-1	1	1000	4.00	75	2.125	56.54	6.25
11	-1	1	-1	1	800	6.00	75	2.125	76.38	7.90
12	1	1	-1	1	1000	6.00	75	2.125	76.53	7.74
13	-1	-1	1	1	800	4.00	105	2.125	92.86	9.85
14	1	-1	1	1	1000	4.00	105	2.125	80.67	8.75
15	-1	1	1	1	800	6.00	105	2.125	83.18	15.09
16	1	1	1	1	1000	6.00	105	2.125	79.82	14.77
17	-2	0	0	0	700	5.00	90	1.750	80.24	4.01
18	2	0	0	0	1100	5.00	90	1.750	77.51	6.65
19	0	-2	0	0	900	3.00	90	1.750	82.36	5.95
20	0	2	0	0	900	7.00	90	1.750	79.57	8.89
21	0	0	-2	0	900	5.00	60	1.750	57.64	3.50
22	0	0	2	0	900	5.00	120	1.750	88.76	8.45
23	0	0	0	-2	900	5.00	90	1.000	67.62	2.39
24	0	0	0	2	900	5.00	90	2.500	86.30	7.95
25	0	0	0	0	900	5.00	90	1.750	82.20	6.80
26	0	0	0	0	900	5.00	90	1.750	79.62	3.86
27	0	0	0	0	900	5.00	90	1.750	75.66	5.79
28	0	0	0	0	900	5.00	90	1.750	73.13	4.79
29	0	0	0	0	900	5.00	90	1.750	80.08	9.12
30	0	0	0	0	900	5.00	90	1.750	84.18	8.57
31	0	0	0	0	900	5.00	90	1.750	84.31	5.79

2.4. Analytical

The pH, COD, BOD and PO_4^{3-} of each sample were determined according to the methods described by Rodier et al. [13].

COD removal and energy consumption were calculated from the following equations (8) and (9) respectively:

$$\text{COD}\ (\%) = \frac{(\text{Initial COD} - \text{Residual COD})}{\text{Initial COD}} * 100 \tag{8}$$

$$E = \frac{U * I * t}{V} \tag{9}$$

With *E* the energy consumption (kWh.m^{-3}), *U* the current voltage (V), *I* the current intensity (A), *t* the time (h) and *V* the treated volume (m^3).

3. Results and Discussion

3.1. Characteristics of Slaughterhouse Wastewater

The slaughterhouse wastewater is characterized by high organic matter with a BOD of 5700 mgO_2/L and a COD of 6594 mgO_2/L COD. This wastewater also contains high TSS (3600 mg/L), PO_4^{3-} (55 mg/L) and high turbidity (726 NTU) as shown in Table 3. In addition, it has a high conductivity (2242 µS/cm) favorable to electrocoagulation treatment. Indeed, a high conductivity increases the performance of electrocoagulation by lowering the ohmic resistance of the wastewater. Finally, the characteristics of this wastewater, largely superior to the Ivorian regulations, show that its treatment is imperative.

Table 3. Characteristics of slaughterhouse wastewater.

Parameters	Values	Standards*
pH	6.88	5.5 - 9.5
Conductivity (µS/cm)	2242	200 - 1000
Turbidity (NTU)	726	-
COD (mg/L)	6594	< 300
BOD (mg/L)	5700	< 100
PO_4^{3-} (mg/L)	55	15
TSS (mg/L)	3600	30
NH_4^+ (mg/L)	135	-

* Standards of ivorian ministry in charge of environment, waters and forests protection.

Table 4. Analysis of variance results for response surface quadratic model for COD removal and energy consumption.

Source	Degree of freedom	Mean square	F-value	Pr > F
COD removal: Y_1				
Model	10	2.78663×10^2	15.8395	0.352 **
Residual	5	1.75929×10^1		
Total	15			
Energy consumption: Y_2				
Model	10	19.9431	39.1176	0.0405 ***
Residual	5	0.5098		
Total	15			

< 1%; *< 0.1%
$R^2 = 0.874$; $R^2_{adj} = 0.763$ for COD removal
$R^2 = 0.841$; $R^2_{adj} = 0.702$ for energy consumption.

3.2. Effects of Experimental Parameters on COD Removal and Energy Consumption

Table 4 presents the ANOVA of the FD regression parameters.

The probability Pr < 5% indicates that the model is significant for COD removal. Furthermore, the coefficients $R^2 = 0.969$ and R^2adj = 0.908 show that the model fit is good. Indeed, according to Joglekar et al. [14], the model fit is good when $R^2 > 0.80$. Furthermore, the value of $R^2 = 0.969$ shows that only 3.1% of the total variation cannot be explained by the empirical model.

The ANOVA of the energy consumption model shows that the model is significant (Pr < 0.1%). Indeed, a model is considered significant if its probability is less than 5%. The value of the correlation coefficient ($R^2 = 0.987$; R^2adj = 0.962) indicates that only 1.3% of the total variation is not taken into account by the model.

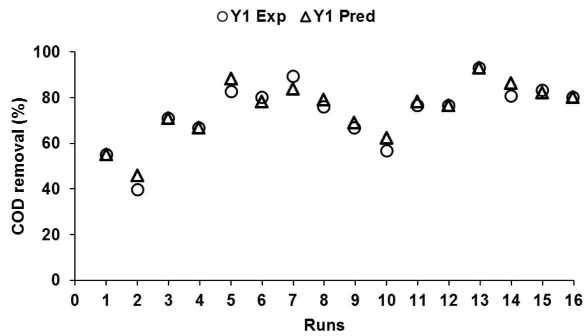

Figure 2. Comparison of calculated and predicted values for COD removal by RSM.

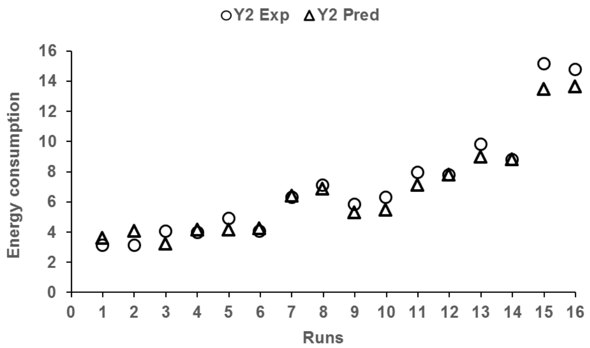

Figure 3. Comparison of calculated and predicted values for energy consumption by RSM.

The comparison of the actual values (experimental values) and the values predicted by the model is presented in Figures 2 and 3. These show that the theoretical and experimental values are very close to each other for COD removal and energy consumption. This proximity reflects the robustness of the statistical models obtained.

The first order functions (Equations (10) and (11)) indicate the relationship between the independent variables and the responses.

$$Y_1\ (\%) = 73.177 - 3.901\ X_1 + 4.055\ X_2 + 9.776\ X_3 + 3.402\ X_4 + 1.169\ X_1X_2 - 0.130\ X_1X_3 + 0.711\ X_1X_4 - 5.076\ X_2X_3 - 1.658\ X_2X_4 - 2.224\ X_3X_4 \quad (10)$$

$$Y_2\ (kW.h.m^{-3}) = 7.041 - 0.076\ X_1 + 1.310\ X_2 + 1.796\ X_3 + 2.476\ X_4 + 0.110\ X_1X_2 - 0.101\ X_1X_3 - 0.064\ X_1X_4 + 0.645\ X_2X_3 + 0.547\ X_2X_4 + 0.801\ X_3X_4 \quad (11)$$

Equation (10) shows that COD removal is strongly influenced by treatment time ($b_3 = 9.776$). The positive value of b_3 shows that by increasing the treatment time, the COD removal is better. When the time is increased from 75 to 105 min, the COD removal rate improves on average by 2*9.776 or 19.55%. The pH was the second most important factor with a positive effect ($b_2 = 4.055$). The percentage of COD removal increases on average by 8.10% when the pH is increased from 4 to 6. The third most important factor is the stirring speed with a negative effect ($b_1 = - 3.901$). The negative value of the coefficient shows that the increase in the stirring speed from 800 to 1000 rpm reduces the COD removal rate by an average of 7.80%. The last parameter that influences the studied response is the current intensity. Its value ($b_4 = 3.402$) means that the increase in current intensity enhances COD removal. The average COD removal rate increases by 6.80% when the current intensity increases from 1.375 to 2.125 A. The interactions X_2X_3 (pH and time) presented by figure 4, is the most significant interaction with b_{23} = - 5.076. The left side of figure 4 shows the evolution of COD removal at pH = 4 while on the right side, COD removal at pH = 6 is observed. The value of 83.97% is obtained for a time of 105 min and for pH = 4. The negative value of the interaction b_{23} shows that the simultaneous increase in pH and time results in a 10.15% decrease in COD removal.

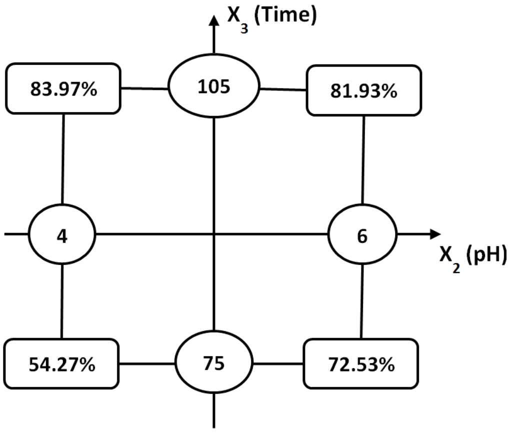

Figure 4. *Interaction b_{23} between pH and electrolysis time for COD removal.*

Equation (12) shows that the energy consumption is related to the current intensity (b_4 =2.476) and the treatment time (b_3 = 1.796). The positive values of b_3 and b_4 prove that the increase in treatment time and current intensity leads to an increase in the energy consumption. At the interaction level, X_3X_4 (time and current intensity) represents the most significant interaction with a negative effect (b_{34} = 0.801). Figure 5 shows the effect of the X_3X_4 interaction on energy consumption. It can be seen that when the treatment time and current intensity increase simultaneously, the energy consumption increases strongly.

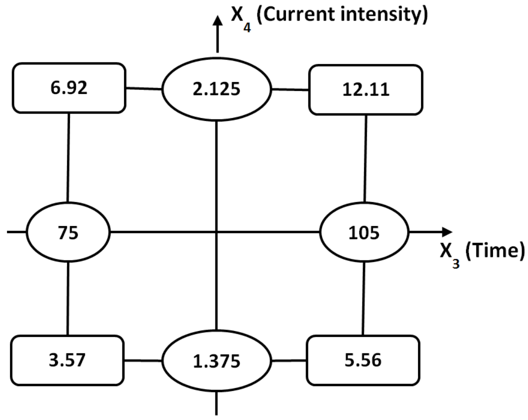

Figure 5. *Interaction b_{34} between current intensity and electrolysis time (min) for energy consumption.*

The FD methodology identified factors that have a significant effect on COD removal and energy consumption. However, this methodology does not allow the optimization of the treatment. Therefore, treatment optimization is discussed in section 3.3.

3.3. Optimization of COD Removal and Electrical Energy Consumption

Equations (12) and (13) show the relationship between the independent variables and the responses.

$$Y_1 (\%) = 79.753 - 2.828 X_1 + 2.471 X_2 + 9.111 X_3 + 3.825 X_4 + 1.169 X_1X_2 - 0.130 X_1X_3 + 0.711 X_1X_4 - 5.076 X_2X_3 - 1.658 X_2X_4 - 2.224 X_3X_4 - 0.940 X_1^2 - 0.417 X_2^2 - 2.359 X_3^2 - 1.419 X_4^2 \quad (12)$$

$$Y_2 (kW.h.m^{-3}) = 6.389 + 0.169 X_1 + 1.118 X_2 + 1.610 X_3 + 2.114 X_4 + 0.110 X_1X_2 - 0.101 X_1X_3 - 0.064 X_1X_4 + 0.645 X_2X_3 + 0.547 X_2X_4 + 0.801 X_3X_4 - 0.087 X_1^2 + 0.436 X_2^2 + 0.075 X_3^2 - 0.127 X_4^2 \quad (13)$$

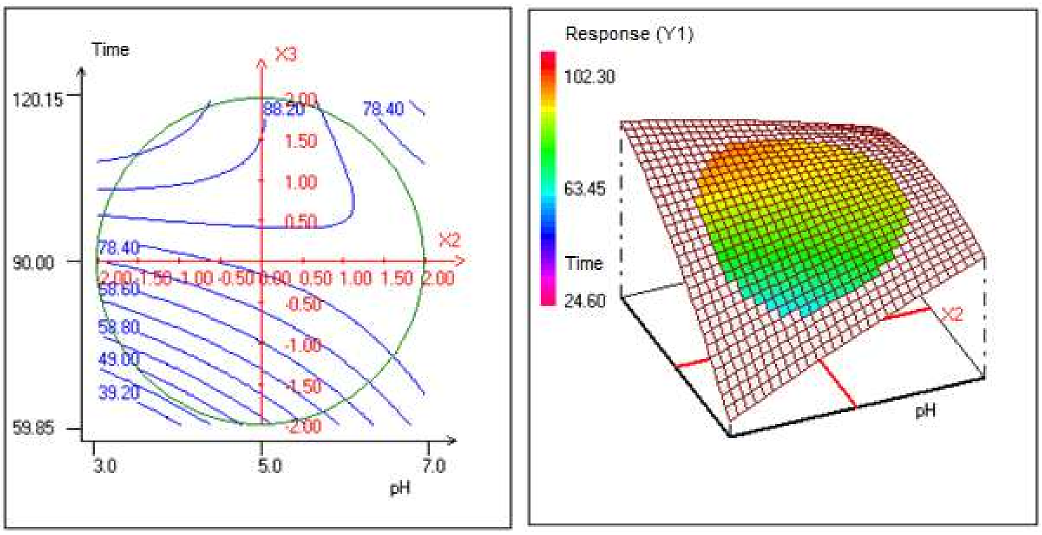

Figure 6. *Contour plots of COD removal obtained from RSM using Nemrod-W Software.*

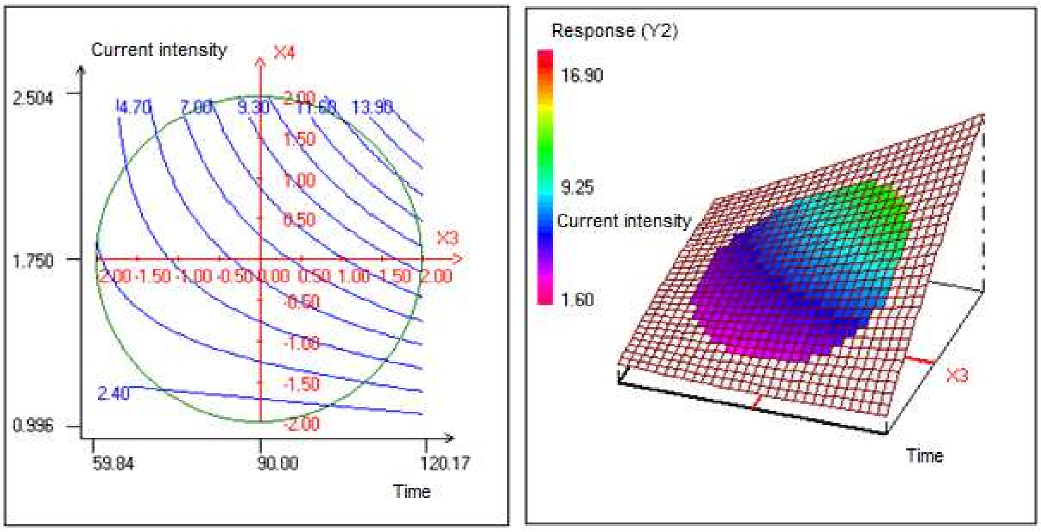

Figure 7. *Contour plots of energy consumption obtained from RSM using Nemrod-W Software.*

These equations show that the factors that strongly influence COD removal are treatment time (b_3 = 9.111) and current intensity (b_4 = 3.825) respectively. Stirring speed (b_1 = - 2.828) and pH (b_2 = 2.471) also have a significant effect on COD removal. The most significant X_2X_3 (pH and time) interaction is shown in Figure 6. When pH and time are maintained at values above 5 and 90 min, a decrease in COD removal is observed. Figure 7, which presents the interaction between time and current intensity, shows that as time and current intensity increase, there is a significant increase in electrical energy consumption.

Current intensity and time are very important factors for electrocoagulation. Indeed, these factors govern the amount of coagulant generated. Thus, the mass of coagulant produced is proportional to the current intensity and treatment time [15]. The iron coagulants generated (Fe^{2+}/Fe^{3+}), in the presence of hydroxyl ions (OH^-), are transformed into iron hydroxides $Fe(OH)_2$ and $Fe(OH)_3$, which promote the removal of COD by neutralization and flocculation of colloidal particles, co-precipitation and complexation or electrostatic attraction [16]. Furthermore, the current intensity determines the amount of H_2 governed by Faraday's law [17]. The H_2 bubbles cause the removal of pollutants by flotation due to the adsorption of pollutants on the surface of the H_2 bubbles [18]. However, high current intensities can negatively affect COD removal. Indeed, an overdose of coagulants can reverse the charge of colloidal particles and redisperse them in the medium leading to a decrease in COD removal. The improvement in COD removal with increasing pH (from 4 to 7) is justified by the nature of the coagulants generated. At neutral pH, the predominant iron species are $Fe(OH)_2$ and $Fe(OH)_3$, which are known to improve COD removal [19]. As for the stirring speed, an increase in its value is detrimental to the performance of electrocoagulation, because a very high stirring speed prevents the formation of flocs, thus reducing the elimination of pollutants by adsorption and coprecipitation.

The optimization by CCD was done by maximizing COD removal and minimizing electrical energy consumption. To do this, the desirability function of the Nemrod software was used. To maximize COD removal and minimize energy consumption, weighting factors of 5/5 (high) and 3/5 (low) were used. Desirability is a value that varies from 0 to 100%. It provides information on the adequacy between the solution and the imposed criteria. If the value is close to 100%, it means that the solution is very close to the desired response. Conversely, if the desirability is close to zero, it means that it is very difficult to achieve the desired response [20].

Table 5. *Optimal conditions.*

Factors				**Weights**		**Responses**		
Stirring speed ($tr.min^{-1}$)	**pH**	**Time (min)**	**Current intensity (A)**	**Abatement DCO (%)**	**Energy consumption ($KWh.m^{-3}$)**	**Abatement DCO (%)**	**Energy consumption ($KWh.m^{-3}$)**	**Desirability (%)**
871	6.83	80	1.85	1	1	82.49	8.61	97.21

Table 6. *Characteristics of slaughterhouse wastewater after treatment.*

Parameters	**After treatment**	**Abatement (%)**
pH	8.71 ± 0.02	-
Conductivity (µS/cm)	2242 ± 84	-
Turbidity (NTU)	16 ± 0.2	97.80 ± 0.86
COD (mg/L)	1055 ± 46	84 ± 1.08
BOD (mg/L)	350 ± 25	93.86 ± 0.91
PO_4^{3-} (mg/L)	0.21 ± 0.06	99.62 ± 0.12
TSS (mg/L)	11 ± 05	99.69 ± 0.20

The optimization results are shown in Table 5. The value of the desirability function is 97.21%. This shows that the results predicted by the model are very close to the desired results. To confirm the validity of the predicted results, the optimal conditions were used in triplicate for the treatment of slaughterhouse wastewater. The treatment results are recorded in Table 6. The COD removal rate (84 ± 1.08%) is higher than the response predicted by the software (82.49%), while the energy consumption is 9 kWh m^{-3} compared to a value of 8.61 predicted by the model.

The turbidity of the treated water has decreased significantly after treatment. It went from 726 NTU before treatment to 16 NTU after treatment, corresponding to an abatement of 97.80 ± 0.86%. The concentration of PO_4^{3-} fell sharply with an abatement of 99.62 ± 0.12%. Respective reductions of 93.86 ± 0.91% and 99.69 ± 0.20% were obtained for BOD and SS. Our results are very similar to those of [21] which achieved 94%, 84%, 87% removal of turbidity, COD and BOD respectively, [22] with 86% COD and 100% Ptot and Psol and [23] with 97, 93, 84 and 81% removal of BOD, COD, TN and TSS, respectively. PO_4^{3-} removal occurred by adsorption and precipitation on the surface of $Fe(OH)_2$, $Fe(OH)_3$ and in the form of $FePO_4$ and $Fe_3(PO_4)_2$ [22]. The closeness of our results to those of the literature shows the efficiency of electrocoagulation to treat slaughterhouse wastewater. Furthermore, RSM leads to an accurate modelling of COD removal and energy consumption.

4. Conclusion

This study allowed the optimization of the electrocoagulation using the response surface methodology and the treatment of slaughterhouse wastewater by the optimal conditions obtained. The optimal conditions were obtained with a stirring speed of 871 rpm, an initial pH of 6.83 (close to the natural pH of the wastewater), a treatment time of 80 min and a current intensity of 1.85A. Under this optimal condition, respective reductions of 84 ± 1.08%, 93.86 ± 0.91%, 97.80 ± 0.86% and 99.62 ± 0.12% of COD, BOD, turbidity and PO_4^{3-} were obtained. The electrical energy consumption was 9 $kWh.m^{-3}$. This study therefore shows that electrocoagulation is an efficient method for the treatment of slaughterhouse wastewater. Furthermore, the response surface methodology is very useful for the modelling and optimization of electrocoagulation. The results obtained thus allow us to consider the application of RSM for the treatment of slaughterhouse wastewater on a pre-industrial and industrial scale.

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