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Simultaneous removal of natural organic matter and turbidity from Oued El Harrach River by electrocoagulation using an experimental design approach

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ABSTRACT

In this study, the application of full factorial design and a surface response methodology was used to model the two-factor influence. Time and current intensity on bacterial biomass, color, turbidity, and natural organic matter (NOM) removal from Oued El Harrach River were discussed. A factorial experimental design was used to investigate the bacterial biomass and color removal from Oued El Harrach River situated in northern Algeria which is treated by electrocoagulation using iron plate electrodes. However, a response surface methodology (RSM) was used to reduce the turbidity and NOM. The bacteria removal efficiency was determined after 30 min of treatment. The combined effects of operating parameters on removal turbidity and NOM were also analyzed. A regression model was found to fit the experimental data very well. Besides, the results were statistically analyzed using the student's *t*-test, analysis of variance, *F*-test, and lack of fit in order to define the most important process variables affecting the removal of bacteria biomass, color turbidity, and NOM from Oued El Harrach River. The predicted results using factorial regression model showed high regression coefficient values (i.e. $R^2_{bact} = 0.9687$ and $R^2_{colo} = 0.9983$) which are in substantial agreement with experimental data. On the other hand, predicted values of turbidity and NOM obtained from central composite model were in close agreement with the experimental values (i.e. $R^2_{turb} = 0.9442$ and $R^2_{NOM} = 0.9432$). This study proved that factorial design and RSM could efficiently be applied for the wastewater treatment modeling by electrocoagulation process.

Keywords: Bacteria; Electrocoagulation; Factorial design; Response surface; Water treatment

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1. Introduction

The Oued El Harrach River is located in northern Algeria. It consists of various organic and inorganic compounds which may be either dissolved or suspended. These compounds are commonly classified as potential pollution problem for surface waters and the ecosystem. Bacterial biomass is among the various forms of pollution problem. Its removal is considered as the usual prerequisite. There are several conventional techniques which are used to remove bacterial biomass from wastewater, such as ozonation, adsorption, oxidation, biosorption [1], membrane process chemical oxidation [2], Fenton's oxidative treatment [3] adsorption [4], chemical precipitation [5,6], coagulation/flocculation [4,7], sedimentation/flotation and air stripping [7,8], membrane processes as nanofiltration [9,10], microfiltration [10], and ultrafiltration [11].

The electrocoagulation (EC) has been tested on several waters, such as synthetic natural water [12], laundry wastewaters [13], pasta and cookie processing wastewaters [14], dairy wastewaters [15], alcohol distillery wastewaters [16], and distillery effluents [17]. This technique has many advantages compared to conventional processes in terms of its simple equipment, e.g. easy operation, less retention time, chemicals adding reduction or absence, electro-generated flocks rapid sedimentation, and less sludge production [18,19]. In addition, the experimental design technique application in the electrocoagulation process can result in improved product yield and reduced process variability, and it is also possible to reduce the experiment number [20,21]. EC is an evolving technology that has been effectively applied in the industrial wastewater treatment containing various pollutants, such as textile wastewaters, olive mill wastewaters [22,23], and polluted water containing several toxic substances [24].

Electrocoagulation is a simple and efficient method where the coagulating agent is generated *in situ* by dissolving electrochemically either aluminum or iron ions from, respectively, aluminum or iron electrodes [25,26]. Coagulants dissolved from the anodes during the electrocoagulation process cause the destabilization of pollutants. Iron anodes can produce ferrous or ferric ions according to Eq. (1). Ferrous and ferric hydroxides are produced according to Eq. (2), where m depends on the pH [27]. Hydrogen formation on the cathode (Eq. (3)) is usually the main reduction reaction occurring on the iron electrodes during EC treatment [28]. According to Eq. (4), Fe^{2+} after oxidation become Fe^{3+} in dissolved oxygen presence and in a neutral or high pH (above 7.0) [29].

$$\mathrm{Fe_{(s)}} \rightarrow \mathrm{Fe^{n+}_{(eq)}} + n\mathrm{e^-} \tag{1}$$

$$\mathrm{Fe^{n+}_{(eq)}} + \mathrm{OH^-} \leftrightarrow \mathrm{Fe(OH)}_m^{(n-m)+} \tag{2}$$

$$\mathrm{2H_2O + 2e^-} \rightarrow \mathrm{H_{2(g)} + 2OH} \tag{3}$$

$$\mathrm{4Fe^{2+}_{(aq)} + 10H_2O + O_{2(aq)}} \rightarrow \mathrm{4Fe(OH)_{3(s)} + 8H^+} \tag{4}$$

Hydrogen evolution can induce particles flotation [28]. This electroflotation technology can be used for efficient precipitated, coagulated, or flocculated material separation [29].

This study aimed to establish the functional relationships between some operating parameters, namely time and current intensity. The experimental work is carried out using experiment design in order to examine the main factors affecting the different responses and their interactions. The electrocoagulation efficiency to remove bacteria biomass, color, turbidity, and natural organic matter (NOM) from Oued El Harrach River was also reported. It can be noted that the NOM is composed by complex mixture of different organic materials such as bacteria, viruses, humic acids, fluvic acids, polysaccharides, and proteins [30].

2. Material and methods

2.1. Electrocoagulation process description

Electrocoagulation process involves three stages: coagulant formation through dissolution of anode electrode metal ions, pollutants destabilization, suspended particles, and de-emulsification, and instable phases and flock-forming aggregation [18,31,32]. Pollutant destabilization, suspended particles, and de-emulsification mechanism can be established through dispersed double-layer compression, ion neutralization species existing in water and wastewaters, and flocks and sludge forming [18,33].

2.2. Reactor design

The EC experiments were conducted in a 1-L Plexiglas reactor. The anode and cathode, fully immersed in water, with dimensions of 4.5 cm × 5.5 cm × 1 cm ($L \times W \times T$) made of plate iron were connected to a digital dc power supply in bipolar mode. The experiments were realized with 0.5 L of wastewater placed into the electrolytic cell. All the experiments were performed in batch mode at room temperature. At the end of each electrocoagulation treatment study, a solution with flocks was allowed to settle for 1 h in the container before bacteria analysis; the amount of sludge produced was expressed as a ratio. The samples for chemical analysis were taken from limpid phase. Neither centrifuging nor filtration was performed in this study.

2.3. Response determination

All variables are assumed to be measurable, the full factorial design and the response surface can be expressed as follows:

$$Y = f(X_1,\ X_2,\ X_3 \ldots X_i)$$

where Y is the system answer, and X_i is the action variable called factors.

The responses ($\%Y_\mu$) were determined by measuring the yield efficiency and defined as:

$$\%Y_\mu = ((Y_0 - Y_{EC})/\ Y_0) \times 100 \quad (5)$$

where Y_0 is the initial concentration and Y_{EC} is the final concentration after electrocoagulation process. The removal bacterial, color, turbidity, and NOM responses were measured under suitable conditions. Initial pH, conductivity, temperature of 25˚C, and distance between electrodes of 1.5 cm were used.

3. Results and discussion

The Oued El Harrach River consists of various pathogenic micro-organisms and parasites, which is one of the biggest pollution problems for local ground and surface waters. Initial wastewater quality before treatment process is presented in Table 1.

3.1. Experimental designs for removal tests

The parameters involved in the destruction of charge bacteria, color, turbidity, and NOM in the wastewater were carried out by experimental design using statistical software JMP (Version 9). The factors levels (−) and (+) in the destruction bacteria for current intensity (X_1) and time (X_2) are given in Table 2. All the experiments were carried out with iron electrode for coagulation treatment.

For better understanding of all effects, the number of experiments (run) is given by $a^K = 2^2$, where a is the number of levels and K is the number of factors. The two levels assigned to each variable are expressed in coded forms as (−) and (+) [34,35].

The model performance was evaluated by the variance analysis (ANOVA) which included the Fisher's F-test (overall model significance), its associated probability $p > F$, the determination coefficient R^2, and the lake of fit. The student's t-value for the estimated coefficients and the associated probabilities $p > |t|$ was also applied.

3.1.1. Full factorial design

An experiment two-level factorial design (2^2) was adopted for the quantification effects of the two factors on the bacteria biomass removal and color (Table 3).

Six possible combinations with the bacterial biomass and color inactivation yield were tabulated in order to investigate the coefficients effects, standard deviation coefficients, standardized effects, , and other statistical fitted model parameters. The Fisher's "F" and student's t-test were used to determine the parameter significance regression coefficients. The p-values were used as tools to check the significance of each of the interactions among the variables. In general, larger the t magnitude is and smaller the p value will be the more significant is the corresponding coefficient term [36].

Main factor, interaction effect, model coefficients, each coefficient standard deviation, and probability for the full factorial designs are presented in Table 4. As can be seen, all main factors and their interactions are significant at 5% of probability level ($p < 0.05$). The regression equation developed from different experiment sets shows the yield dependence on individual parameters as well as interactions for simultaneous parameters variation [37,38].

Results show also that the Fisher's "F" was very much higher compared to tabulated value for the full factorial design.

3.1.2. Analysis of variance

The variance analysis for the full factorial design is presented in Table 5. The p value is lower than 0.05 (95% confidence) indicates that the model is considered to be statistically significant for bacteria biomass and color. DF stands for the degrees of freedom

Table 1
Initial wastewater quality

Parameters	Concentration
Bacterial biomass (DO_{600})	0.839
Color (DO_{415})	0.415
NOM (mg/l)	540.78
Turbidity (NTU)	227

Table 2
Factors, levels, and responses used in experimental designs

Factors	Level (−)	Level (+)
X_1 current intensity "Int." (A)	2	3
X_2 time "*t*" (min)	10	30
Responses		
$Y_{bact.}$ (%)	Bacterial biomass	"Bact."
$Y_{colo.}$ (%)	Color	"Colo."
Y_{turb}. (NTU)	Turbidity	"Turb."
Y_{NOM} (mg/l)	Natural organic matter	"NOM"

Table 3
A two-level factorial design of both runs (bacteria biomass and color removal)

Run	Int. (A)	*t* (min)	Removal efficiency (%) Bact.	Removal efficiency (%) Colo.
1	−1	−1	70.00	49.42
2	+1	−1	93.00	54.53
3	0	0	93.00	58.31
4	0	0	92.77	64.00
5	−1	1	94.81	63.59
6	1	1	99.00	93.24

associated with each variation source. The squares sum indicates the associated squares sum for each variation source and the mean square is calculated by dividing the squares sum by DF. The Fisher's "*F*" ratio (model mean square divided by the error mean square) tests the hypothesis that all the regression parameters are zero. Prob. > *F* is the probability of obtaining a greater *F*-value by chance alone, if the specified model fits no better than the overall response mean.

Table 4
Regression analysis for bacteria biomass and color

Term	Coefficient	Standard error	*t*-value	*p*-value
Bact.				
Constant	90.53	1.17	76.88	0.0002*
X_1	6.95	1.14	4.82	0.0405*
X_2	7.55	1.14	5.23	0.0346*
X_1X_2	−4.85	1.14	−3.37	0.0781
Colo.				
Constant	64.79	0.39	162.67	<.0001*
X_1	8.69	0.48	17.81	0.0031*
X_2	13.22	0.48	27.10	0.0014*
X_1X_2	6.13	0.48	12.58	0.0063*

*It means "interation effect" where $p < 0.05$.

Table 5
ANOVA for the regression model representing bacteria biomass (*Bact.*) and color (*Colo.*)

Term	DF	SS	MS	*F*	*p* > *F*
Bact.					
Model	3	0.0034	0.0001	22.78	0.0243
Residual	2	0.0001	0.0000		
Total	5	0.0035			
Colo.					
Model	3	1,151.69	383.89	402.24	0.0025*
Residual	2	1.90	0.952		
Total	5	1,153.59			

*It means "interation effect" where $p < 0.05$.

To confirm the model correlation and significance, a new experiment was developed (Fig. 1(a) and (b)), the experimental values agreement with the prediction model considering both the correlation between theoretical and experimental values. The obtained model coefficients for bacteria biomass removal (Fig. 1(a)) was evaluated as $R^2 = 0.9687$ indicates that the factorial model accuracy was satisfactory. The coefficient R^2_{adj} (0.9218) is more suitable for comparing models with independent variables of different numbers [39]. The color experimental response is given in Parity plot (Fig. 1(b)). The determination coefficient (R^2) was evaluated as 0.9983, and the adjusted *R*-square value (R^2_{adj}) was 0.9958. The two values were both close to 1, which indicated a relatively high correlation degree between the actual and the predicted responses [40].

The standardized effects of Pareto chart at $p = 0.005$ is presented in Fig. 2. All the values presented an absolute value higher than 2.3 ($p = 0.005$), which were located at the dash line right, were significant. A *p*-value less than 0.005 (95% confidence) indicates that the model is considered to be statistically significant.

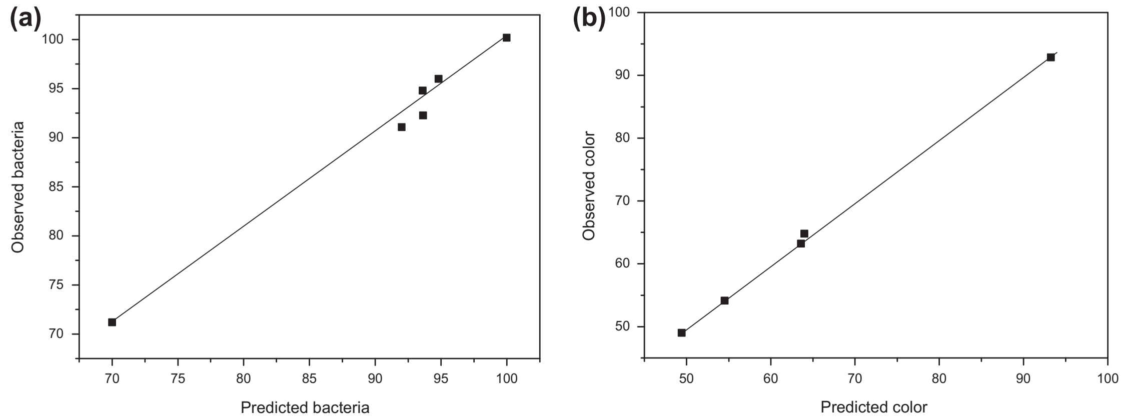

Fig. 1. Parity plot comparing the bacteria biomass (a) and color (b) uptake data with the Oued El Harrach River predictions model by electrocoagulation process.

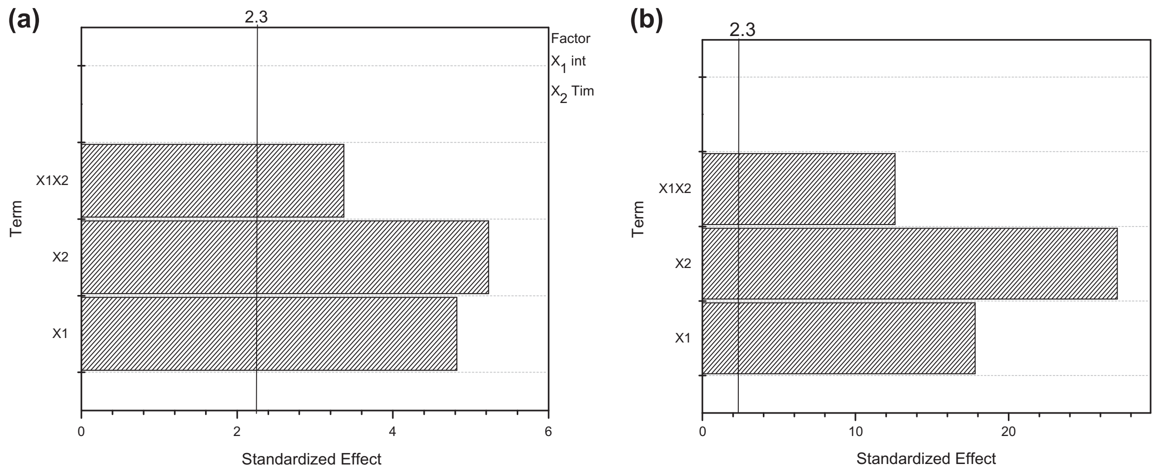

Fig. 2. Standardized effects of Pareto chart on the removal efficiency for (a) bacteria biomass and (b) Oued El Harrach River color by electrocoagulation process.

The effect of absolute standardized value of each factor and its interaction appeared at each bar right. Fig. 2(a) and (b) gives the relative importance of the individual and interaction effects with time and current intensity.

Operating current density is critical in batch electrocoagulation process as it is the only operating parameter that can be controlled directly [41,42]. In this system, electrode spacing is fixed and current is continuously supplied. The graphical interpretation of models (bacteria biomass and color removal) is shown in Fig. 3. The bacteria biomass uptake was found to decrease with increasing current intensity indicating that bacteria biomass cell cannot resist the electrical field increases. Consequently, the EC treatment effect on color values may be considered as an electric charge function.

The final predicted model in terms of significant actual factor for abatement of bacteria biomass (Eq. (6)) and color removal (Eq. (7)) by EC are determined by the factorial design model and given below:

$$Y_{\text{bact.}} = 90.53 + 6.95X_1 + 7.55X_2 - 4.85X_1X_2 \tag{6}$$

$$Y_{\text{colo.}} = 64.79 + 8.69X_1 + 13.22X_2 + 6.13X_1X_2 \tag{7}$$

3.1.3. Interaction effects

According to Eqs. (6) and (7), the main observed effects of current intensity (X_1) and time (X_2) had a significant positive effect on bacteria biomass and color removal, respectively. Besides, the interaction

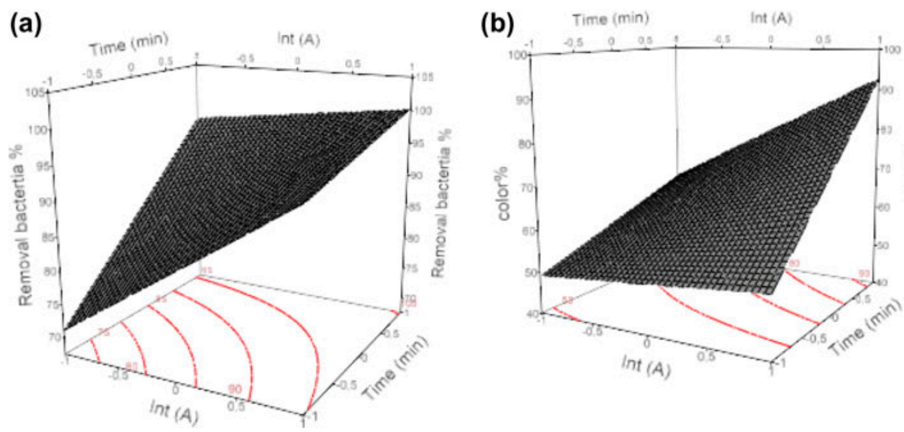

Fig. 3. The graphical interpretation of models showing the current intensity effect (X_1) and time (X_2) on removal (a) bacterial biomass and (b) Oued El Harrach River color.

effect of both factors, the current intensity (X_1) and the time (X_2) on color removal is presented in Fig. 4. It is noted that the time interaction and current intensity were not statistically significant on the bacterial biomass response. Electrocoagulation removed color with high efficiency (Fig. 3(b)), and it is noticeable that the color removal increases vs. time as well as the electric current. The model predicts total color removal at intermediate current density, high treatment time, and high initial pH. Vepsalainen et al. [44] found the same results explaining that the EC treatment effectively removes toxic pollutants from pulp mill effluents; also, Khoufi et al. and Hanafi et al. [23,43] removed toxicity which is mainly caused by polyphenols (tannins) from olive mill wastewaters.

3.1.4. Central composite design

The full factorial design is not statistically significant for the other responses as turbidity and NOM. Consequently, the response surface methodology (RSM) was applied in order to modeling the (EC) efficiency on both previous responses. Table 6 shows all variables with response of turbidity and NOM.

Regression analysis for both responses turbidity and NOM is presented in Table 7. The effects of variables and their interactions can be estimated showing that the current intensity and time have a negative effect on both responses, i.e. turbidity and NOM. Also, the results show that the *F*-value was very much higher compared to tabulated value for central composite model fitting in the ANOVA which is given in Table 8.

Fig. 4(a) shows the actual and the predicted removal turbidity plot. The actual values were the measured response data for a particular run and the predicted values were determined by approximating function employed for the models. The determination coefficient (R^2) was evaluated as 0.9442, and the adjusted R-square value (R^2_{adj}) was 0.9044. The two values were both close

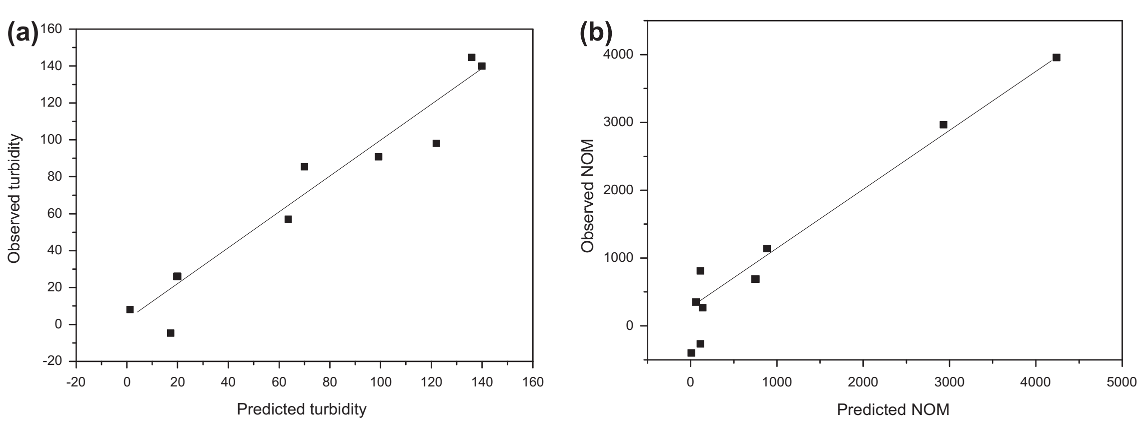

Fig. 4. Parity plot comparing the turbidity (a) and NOM (b) uptake data with the model prediction.

Table 6
Central composite design fit for the yield turbidity and NOM

				Removal efficiency	
Run	Config.	Int. (A)	t (min)	Turbidity	NOM
1	00	0	0	19.8	753.33
2	A0	1	0	60.2	63.6
3	00	0	0	19.8	753.33
4	00	0	0	20	752.32
5	−+	−1	1	70	4,240
6	00	0	0	19.8	750
7	++	1	1	1.5	11.25
8	00	0	0	20	750
9	–	−1	−1	136	886.66
10	0A	0	1	17	113.33
11	+−	1	−1	14	140
12	0a	0	−1	99.2	113.33
13	a0	−1	0	122	2,933.33

Table 7
Regression analysis for turbidity and NOM

Term	Coefficient	Standard error	t-value	p-value
Turbidity				
Constant	25.99	6.513	3.99	0.0052*
X_1	−20.525	7.8762	−3.20	0.0150*
X_2	−47.775	6.40	−7.46	0.0001*
X_1X_2	−18.1875	7.84	−2.32	0.0535
X_1X_1	51.50	9.43	5.46	0.0009*
X_2X_2	16.95	9.43	1.80	0.1155
NOM				
Constant	688.59	161.64	4.26	0.0037*
X_1	−1,307.52	158.93	−8.23	<.0001*
X_2	537.43	158.93	3.38	0.0117*
X_1X_2	−870.52	194.65	−4.47	0.0029*
X_1X_1	968.38	234.25	4.13	0.0044*
X_2X_2	−416.75	234.25	−1.78	0.1185

*It means "interation effect" where $p < 0.05$.

Table 8
ANOVA for the regression model representation

Term	DF	SS	MS	F	$p > F$
Turbidity					
Model	5	29,194.93	5,838.99	23.7286	0.0003*
Residual	7	1,722.514	246.07		
Total	12	30,917.45			
NOM					
Model	5	17,619,347	3,523,869	23.2514	0.0003*
Residual	7	1,060,888	151,555		
Total	12	18,680,234			

*It means "interation effect" where $p < 0.05$.

to 1, which indicated a relatively high correlation degree between the actual and the predicted responses. In the present study, the NOM removal quantity (NOM mg/l) shows that the model fits well with the experiment (Fig. 4(b)). The predicted R^2 of 0.9432 is in reasonable agreement with the adjusted R^2 of 0.9026 which is in reasonable agreement with experimental results.

In order to determine the influence of most important parameters, a standardized Pareto chart was employed (Fig. 5). It consists of bars with a length proportional to the estimated effects of absolute value, divided by the standard error. The bars are displayed in effects of size order, with the largest effects on top. In this study, the chart includes a vertical line at the critical t-value for a "α" of 3.9, and its bar effect (positive or negative) is smaller than the critical t-value that is considered neither significant nor affecting the response variable.

The removal turbidity effect on time was significant as shown in Fig. 5(a). Moreover, for the NOM response, the interaction between current intensity and time was negatively significant (Fig. 5(b)).

Effect of both current intensity and time on turbidity and NOM during the EC treatment was elucidated in Fig. 6(a) and (b), respectively. The NOM was decreased with the current intensity. However, the current intensity and time interaction effect (X_1X_2) had negative significant impact on NOM removal which confirms the previous result (ANOVA), i.e. more the NOM decreased the time and the current intensity increased. It is noticed that the NOM starts to decrease after 10 min of treatment process. On the other hand, the lowest increase in NOM has been observed after 30 min. This result may be explained by the bacteria destruction by electric field, than the bacteria cell polysaccharide and proteins liberalization. According to Sharp and al. [45], the hydrophobic NOM has a higher charge density than hydrophilic NOM and is therefore more easily removed during electrocoagulation treatment. However, the turbidity increase was affected by residual iron after 30 min of treatment process. Although residual iron concentration was correlated with initial pH and electric charge, the model terms could not predict the residual iron in sufficient accuracy. Bubbles and normal flocculation caused by EC treatment may be a source of high turbidities. Moreover, residual metals can cause problems such as deposition and process chemical decomposition where water is consumed. The concentration of residual metals (Fe) and the formation of Fe(III) hydrolysis products were elevated in EC treatment because they are not very soluble [46]. This sludge production can be removed by sedimentation or filtration [47].

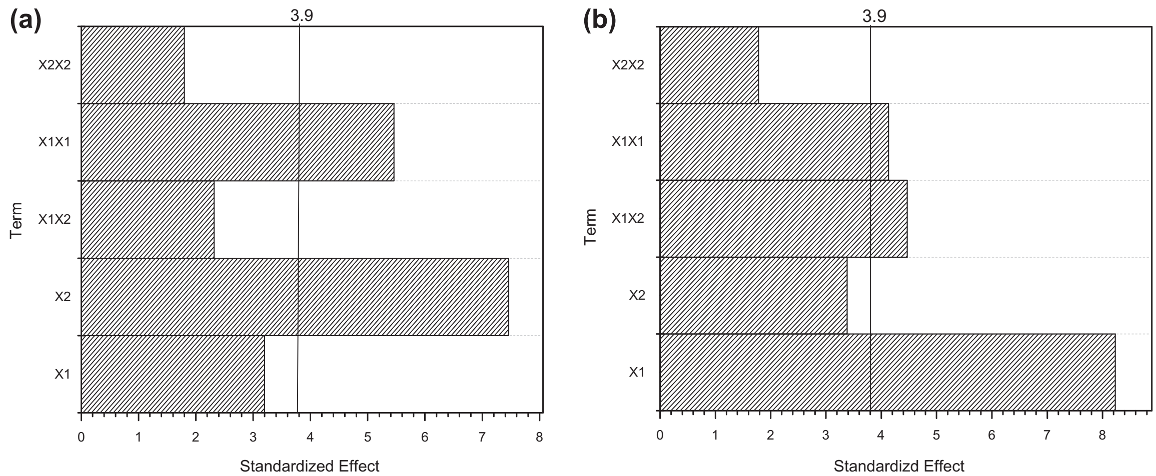

Fig. 5. Standardized effects of Pareto chart on the removal efficiency for turbidity (a) and NOM (b) of Oued El Harrach River by electrocoagulation process; (X_1) current intensity, (X_2) time.

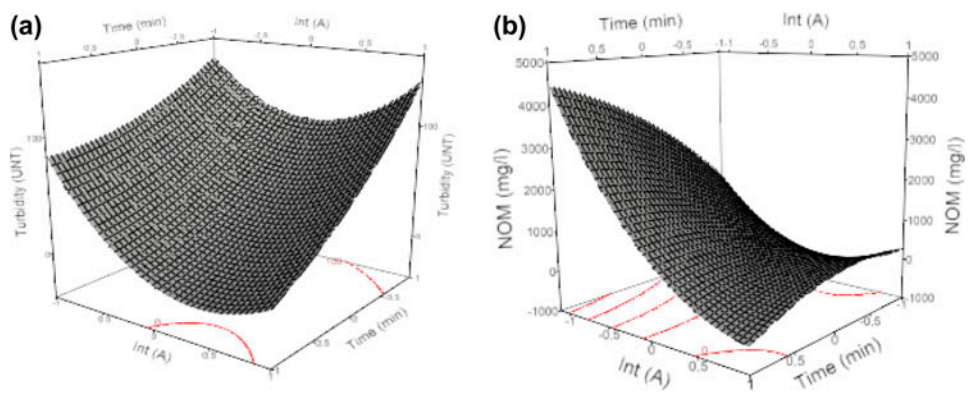

Fig. 6. Response surface plots showing the current intensity effect (X_1) and time (X_2) on removal turbidity (a) and NOM (b) of Oued El Harrach River.

The response central methodology application resulting in the following regression equation which is an empirical relationship between current density and time for removal turbidity and NOM is presented in Eqs. (8) and (9), respectively.

$$Y_{\text{turb.}} = 25.99 - 20.52X_1 - 47.75X_2 + 51.50X_1^2 \quad (8)$$

$$Y_{\text{NOM}} = 688.59 - 1,307.52X_1 + 537.43X_2 - 870.52X_1X_2 + 968.38X_1^2 \quad (9)$$

3.1.5. Interaction effects

The effect of each factor was statistically significant ($p < 0.05$). For turbidity and NOM, it was shown that the main effect X_1, X_2 and interaction X_1, X_2 are highly significant. The interaction plots for turbidity and NOM removal (Table 7) showed that time and current intensity interaction played a major role.

Dissolved iron hydroxides in solution and further coagulates impurities in wastewater forming large agglomerates as seen in previous Eq. (2). Bratby [27] showed that ferrous iron produced from anodes does not coagulate well with NOM. However, iron cations and hydrolyzed species can cause destabilization of particles or chemical aggregates in the solution [33,48]. Thus, ferrous and ferric irons form flocs which are easily separated during the filtration. Ferrous and ferric ions as well as hydroxides are more soluble at low pH; their high charge have a great influence on bacteria cell and neutralization of organic particles as well as other pollutants [44]. EC treatment removes impurities which are in colloidal form, as in chemical coagulation. The main particle removal mechanisms are the same for electrocoagulation and inorganic chemical coagulants: double-layer compression, adsorption destabilization, bridging, and precipitation [28]. In addition, ferrous hydroxide forms a dark-green gelatinous suspension which plays an important part on contaminants removal by complication or electrostatic attraction. This phenomenon is characterized by the electric field application on the colloidal particles charge. The smallest particles (bacteria and NOM) are the first to be removed by electrocoagulation.

4. Conclusions

In this study, the current intensity and experimental time factor effects on EC treatment efficiency were studied. Bacterial biomass removal, color, turbidity, and NOM were used as responses. Full factorial design and RSM were carried out to achieve a better understanding of the relationship between the factors and responses. Moreover, bacteria biomass removal and color were investigated using full factorial design. However, responses of turbidity and NOM were also investigated by applying RSM.

The results show that after a short treatment time with an electrical current increase induces total bacterial biomass elimination. The electrocoagulation process appeared to be an efficient technique to remove micro-organisms from Oued El Harrach River. Full factorial design and RSM can provide a further insight for electrocoagulation process of potential use.

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References

[1] S.H. Hasan, P. Srivastava, M. Talat, Biosorption of lead using immobilized *Aeromonas hydrophila* biomass in up flow column system: Factorial design for process optimization, J. Hazard. Mater. 177 (2010) 312–322.
[2] Á. Anglada, A.U. Urtiaga, I. Ortiz, Dionissios Mantzavinos, Treatment of municipal landfill leachate by catalytic wet air oxidation: Assessment of the role of operating parameters by factorial design, Waste Manage. 31 (2011) 1833–1840.
[3] A. Žgajnar, J. Gotvajn, M. Zagorc-Končan, Cotman Fenton's oxidative treatment of municipal landfill leachate as an alternative to biological process, Desalination 275 (2011) 269–275.
[4] W. Li, T. Hua, Q. Zhou, S. Zhang, L.F. Xiang, Treatment of stabilized landfill leachate by the combined process of coagulation/flocculation and powder activated carbon adsorption, Desalination 264 (2010) 56–62.
[5] E. Neczaj, E. Okoniewska, M. Kacprzak, Treatment of landfill leachate by sequencing batch reactor, Desalination 185 (2005) 357–362.
[6] S.H. Hasan, P. Srivastava, M. Talat, Biosorption of Pb (II) from water using biomass of *Aeromonas hydrophila:* Central composite design for optimization of process variables, J. Hazard. Mater. 168 (2009) 1155–1162.
[7] H. Hasar, S.A. Unsal, U. Ipek, S. Karatas, O. Cınar, C. Yaman, C. Kınacı, Stripping/flocculation/membrane bioreactor/reverse osmosis treatment of municipal landfill leachate, J. Hazard. Mater. 171 (2009) 309–317.
[8] P. Palaniandy, M.N. Adlan, H.A. Aziz, M.F. Murshed, Application of dissolved air flotation (DAF) in semi-aerobic leachate treatment, Chem. Eng. J. 157 (2010) 316–322.
[9] T. Mariam, L.D. Nghiem, Landfill leachate treatment using hybrid coagulation–nanofiltration processes, Desalination 250 (2010) 677–681.
[10] M. Ince, E. Senturk, G. Onkal Engin, B. Keskinler, Further treatment of landfill leachate by nanofiltration and microfiltration—PAC hybrid process, Desalination 255 (2010) 52–60.
[11] X. Yu-Dong, Y. Dong-Bei, Z. Yi, N. Yong-Feng, Fractionation of dissolved organic matter in mature landfill leachate and its recycling by ultrafiltration and evaporation combined processes, Chemosphere 64 (2006) 903–911.
[12] Y.Ş. Yildiz, A.S. Koparal, Ş. İrdemez, B. Keskinler, Electrocoagulation of synthetically prepared waters containing high concentration of NOM using iron cast electrodes, J. Hazard. Mater. 139(2) (2007) 373–380.
[13] J. Ge, J. Qu, P. Lei, H. Liu, New bipolar electrocoagulation–electroflotation process for the treatment of laundry wastewater, Sep. Purif. Technol. 36 (2004) 33–39.
[14] G. Roa-Morales, E. Campos-Medina, J. Aguilera-Cotero, B. Bilyeu, C. Barrera-Díaz, Aluminum electrocoagulation with peroxide applied to wastewater from pasta and cookie processing, Sep. Purif. Technol. 54 (2007) 124–129.
[15] İ.A. Şengil, M. özacar, Treatment of dairy wastewaters by electrocoagulation using mild steel electrodes, J. Hazard. Mater. 137 (2006) 1197–1205.
[16] Y. Yavuz, EC and EF processes for the treatment of alcohol distillery wastewater, Sep. Purif. Technol. 53 (1) (2007) 35–140.
[17] N. Kannan, G. Karthikeyan, N. Tamilselvan, Comparison of treatment potential of electrocoagulation of distillery effluent with and without activated *Areca catechu* nut carbon, J. Hazard. Mater. 137 (2006) 803–1809.
[18] N. Drouiche, S. Aoudj, H. Lounici, M. Drouiche, T. Ouslimane, N. Ghaffour, Fluoride removal from pretreated photovoltaic wastewater by electrocoagulation: An investigation of the effect of operational parameters, Proc. Eng. 33 (2012) 385–391.
[19] N. Drouiche, S. Aoudj, H. Lounici, H. Mahmoudi, N. Ghaffour, Development of an empirical model for fluoride removal from photovoltaic wastewater by electrocoagulation process. Desalin. Water Treat. 29 (2011) 96–102.

[20] V. Ponnusami, V. Krithika, R. Madhuram, S.N. Srivastava, Biosorption of reactive dye using acid-treated rice husk: Factorial design analysis, J. Hazard. Mater. 142 (2007) 397–403.
[21] D. Kavak, Removal of boron from aqueous solutions by batch adsorption on calcined alunite using experimental design, J. Hazard. Mater. 163 (2009) 308–314.
[22] O. Yahiaoui, H. Lounici, N. Abdi, N. Drouiche, N. Ghaffour, A. Pauss, Treatment of olive mill wastewater by the combination of ultrafiltration and bipolar electrochemical reactor processes, Chem. Eng. Process. 50 (2011) 37–41.
[23] S. Khoufi, F. Feki, S. Sayadi, Detoxification of olive mill wastewater by electrocoagulation and sedimentation processes, J. Hazard. Mater. 142 (2007) 58–67.
[24] M. Vepsalainen, J. Selein, M. Pummiainen, M. Sillanpaa, Combined electrocoagulation and chemical coagulation of paper mill mechanically cleaned water, J. Pulp Pap. Sci. 33 (2007) 233–239.
[25] N. Drouiche, N. Ghaffour, S. Aoudj, M. Hecini, T. Ouslimane, Fluoride removal from photovoltaic wastewater by aluminium electrocoagulation and characteristics of products, Chem. Eng. Trans. 17 (2009) 1651–1656.
[26] M. Behloul, H. Grib, N. Drouiche, N. Abdi, H. Lounici, N. Mameri, Removal of malathion pesticide from polluted solutions by electrocoagulation: modeling of experimental results using response surface methodology, Sep. Sci. Technol. 48 (2013) 664–672.
[27] N. Boudjema, N. Drouiche, N. Abdi, H. Grib, H. Lounici, A. Pauss, N. Mameri, Treatment of Oued El Harrach River water by electrocoagulation noting the effect of the electric field on microorganisms, J. Taiwan Inst. Chem. Eng. 45 (2014) 1564–1570.
[28] G. Chen, Electrochemical technologies in wastewater treatment, Sep. Purif. Technol. 38 (2004) 11–41.
[29] M. Vepsäläinen, J. Selin, P. Rantala, M. Pulliainen, H. Särkkä, K. Kuhmonen Amit Bhatnagar, M. Sillanpää, Precipitation of dissolved sulphide in pulp and paper mill wastewater by electrocoagulation, Environ. Technol. 32 (2011) 1393–1400.
[30] M. Vepsäläinen, M. Ghiasvand, J. Selin, J. Pienimaa, E. Repo, M. Pulliainen, M. Sillanpää, Investigations of the effects of temperature and initial sample pH on natural organic matter (NOM) removal with electrocoagulation using response surface method (RSM), Sep. Purif. Technol. 69 (2009) 255–261.
[31] O. Yahiaoui, L. Aizel, H. Lounici, N. Drouiche, M.F.A. Goosen, A. Pauss, N. Mameri, Evaluating removal of metribuzin pesticide from contaminated groundwater using an electrochemical reactor combined with ultraviolet oxidation, Desalination 270 (2011) 84–89.
[32] N. Drouiche, N. Ghaffour, H. Lounici, M. Mameri, Electrocoagulation of chemical mechanical polishing wastewater, Desalination 214 (2007) 31–37.
[33] S. Aoudj, A. Khelifa, N. Drouiche, M. Hecini, HF wastewater remediation by electrocoagulation process, Desalin. Water Treat. 51 (2013) 1596–1602.
[34] R. Ragonese, M. Mulholland, J. Kalman, Full and fractionated experimental designs for robustness testing in the high-performance liquid chromatographic analysis of codeine phosphate, pseudoephedrine hydrochloride and chlorpheniramine maleate in a pharmaceutical preparation, J. Chromatogr. A 870 (2000) 45–51.
[35] A. Massumi, N.M. Najafi, H. Barzegari, Speciation of Cr(VI)/Cr(III) in environmental waters by fluorimetric method using central composite, full and fractional factorial design, J. Microchim. 72 (2002) 93–101.
[36] M. Azharul Islam, V. Sakkas, T.A. Albanis, Application of statistical design of experiment with desirability function for the removal of organophosphorus pesticide from aqueous solution by low-cost material, J. Hazard. Mater. 170 (2009) 230–238.
[37] S. Demim, N. Drouiche, A. Aouabed, S. Semsari, CCD study on the ecophysiological effects of heavy metals on *Lemna gibba*, Ecol. Eng. 57 (2013) 302–313.
[38] M.E.R. Carmona, M.A.P. da Silva, S.G. Ferreira Leite, Biosorption of chromium using factorial experimental design, Process Biochem. 40 (2005) 779–788.
[39] S. Demim, N. Drouiche, A. Aouabed, T. Benayad, O. Dendene-Badache, S. Semsari, Cadmium and nickel: Assessment of the physiological effects and heavy metal removal using a response surface approach by *L. gibba*, Ecol. Eng. 61 (2013) 426–435.
[40] S. Demim, N. Drouiche, A. Aouabed, T. Benayad, M. Couderchet, S. Semsari, Study of heavy metal removal from heavy metal mixture using the CCD method, J. Ind. Eng. Chem. 20 (2014) 512–520.
[41] N. Daneshvar, A. Oladegaragoze, N. Djafarzadeh, Decolorization of basic dye solutions by electrocoagulation: An investigation of the effect of operational parameters, J. Hazard. Mater. 129 (2006) 116–122.
[42] H. Ratnaweera, N. Hiller, U. Bunse, Comparison of the coagulation behavior of different Norwegian aquatic NOM sources, Environ. Int. 25(2–3) (1999) 347–355.
[43] F. Hanafi, O. Assobhei, M. Mountadar, Detoxification and discoloration of Moroccan olive mill wastewater by electrocoagulation, J. Hazard. Mater. 174 (2010) 807–812.
[44] M. Vepsäläinen, H. Kivisaari, M. Pulliainen, A. Oikari, M. Sillanpää, Removal of toxic pollutants from pulp mill effluents by electrocoagulation, Sep. purif. Technol. 81 (2011) 141–150.
[45] E.M. Sharp, S.A. Parsons, B. Jefferson, Seasonal variations in natural organic matter and its impact on coagulation in water treatment, Sci. Total Environ. 363 (2006) 183–194.
[46] J. Duan, J. Gregory, Coagulation by hydrolysing metal salts, Adv. Colloid Interface Sci. 100–102 (2003) 475–502.
[47] M. Vepsäläinen, M. Pulliainen, M. Sillanpää, Effect of electrochemical cell structure on natural organic matter (NOM) removal from surface water through electrocoagulation (EC), Sep. Purif. Technol. 99 (2012) 20–27.
[48] W. Lemlikchi, S. Khaldi, M.O. Mecherri, H. Lounici, N. Drouiche, Degradation of disperse red 167 azo dye by bipolar electrocoagulation, Sep. Sci. and Technol. 47 (2012) 1682–1688.