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# Optimization of electrocoagulation process for treatment of rice mill effluent using response surface methodology

A. Ronaldo Anuf[a,*], K. Ramaraj[a], Vishnu Sankar Sivasankarapillai[b], Ragupathy Dhanusuraman[b], J. Prakash Maran[c], G. Rajeshkumar[d], Abbas Rahdar[e,*], Ana M. Díez-Pascual[f,*]

[a] *Department of Biotechnology, Centre for Research, Kamaraj College of Engineering and Technology, S.P.G.C. Nagar, Virudhunagar, Tamil Nadu, India*
[b] *Nano Electrochemistry Lab (NEL), Department of Chemistry, National Institute of Technology Puducherry, Karaikal 609609, India*
[c] *Department of Food Science and Nutrition, Periyar University, Salem 636 011, Tamil Nadu, India*
[d] *Department of Mechanical Engineering, PSG Institute of Technology and Applied Research, Coimbatore, Tamil Nadu, India*
[e] *Department of Physics, University of Zabol, Zabol, Iran*
[f] *Universidad de Alcalá, Facultad de Ciencias, Departamento de Química Analítica, Química Física e Ingeniería Química, Ctra. Madrid-Barcelona, Km. 33.6, 28805 Alcalá de Henares, Madrid, Spain*

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ABSTRACT

The present work explores the impact of electro coagulation (EC) method on the treatment of waste from rice mill industries using two different electrode materials (Iron (Fe) and Aluminum (Al)). The influence of different parameters such as inter-electrode distance (4–7 cm), effluent pH (6–8), current density (10–30 mA/cm$^2$) and treatment time (20–40 min) on the reduction of chemical oxygen demand (COD), total dissolved solids (TDS) and total soluble solids (TSS) of rice mill effluent (RME) was evaluated through batch experimental runs using Box-Behnken design. Results reveal that the percentage removal of COD, TDS and TSS increased up to an inter-electrode distance of 6 cm, pH of 7, current density of 20 mA/cm$^2$ and treatment time of 30 min and then decreased for both electrodes. In addition, mathematical models were developed for both electrodes in order to predict the experimental data. A numerical optimization method was applied to find out the optimal operating parameters to treat RME, and the percentage removal of COD, TDS and TSS was found to be 94.79, 96.62 and 88.76 %, using the Al electrode, as well as 76.63, 78.51 and 72.03 %, for the Fe electrode, respectively. The comparison of the results attained demonstrate that the Al electrode is more suitable to treat RME than Fe using EC method.

## 1. Introduction

The tremendous progress in industrialization has greatly accelerated the release of pollutants into the environment. This could cause negative effects to the society and has now turned to be a major threat to the environmental sustainability [1]. The release of industrial effluent into the nearby water bodies could lead to severe effects like eutrophication and eventually contribute to mortality of the aquatic life [2]. Rice serves as the major feeding crop throughout the world. In India, rice is considered as the staple food and is the integral part of the diet. Production of edible rice involves a process called milling to remove husk and rice bran from paddy rice. Prior to milling, parboiling is performed to reduce grain breakage and to avoid nutritional loss [3], which is a water consuming process, and soaking of paddy requires a huge quantity of water. Approximately six hundred billion liters of nutrient rich effluent was released from rice mill in every year to produce nearly 500 MMT of paddy [4]. Release of this effluent directly into the nearby aquatic bodies or land without appropriate treatment strategy is a most common practice which was a severe concern for the last two decades. This leads to deterioration of ground water quality and can have many adverse effects on the environment [5]. The water discharged into irrigated fields after soaking and cleaning can lead to health risk and can also impose threat to the quality of the crop plants due to the presence of high organic and inorganic content [6]. Due to the alkaline nature of the effluent with higher content of chemical oxygen demand (COD), total soluble solids (TSS) and organic matter, it cannot be used for any

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* Corresponding authors.
*E-mail addresses:* ronaldoanuf@gmail.com (A.R. Anuf), a.rahdar@uoz.ac.ir (A. Rahdar), am.diez@uah.es (A.M. Díez-Pascual).

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purposes without treating it with the proper method [7].

Treatment of industrial effluents can be carried out by different techniques such as physical, chemical and biological methods. Treatment using chemical methodologies like use of coagulants is ineffective due to the high cost of the reagent and the low COD removal rate. In addition, the possibility of generation of secondary pollutants is high using chemical coagulants [8]. Treatment by biological method requires a long-time span for the complete treatment and post treatment process. Electrocoagulation (EC) is a widely used method in wastewater treatment due to its advantages such as lower set up cost, cheaper electrode material and shorter reaction time than the other mentioned methods, combined with the simultaneous removal of organic and inorganic pollutants as well as suspended solids, versatility and easiness of automation [9–11]. Moreover, it offers the benefits of coagulation, flocculation and electrochemistry in a single process [12,13].

EC unit consists of an anode and cathode material that are used to carry out the oxidation and reduction reaction simultaneously. When the anode is subjected to a suitable current, dissolution of the electrodes take place and leads to the generation of metal ions, which act as effective coagulants and facilitate charge neutralization over the colloids in an effluent [14]. The metal and hydroxyl ions generated react together to form metal hydroxide precipitates that promote colloidal adsorption and the removal of contaminants from the aqueous phase through flocs [15]. Different process parameters like electrode material, pH, current intensity, electrolyte concentration and process time could influence the EC process in order to affect/improve the efficiency of COD, total dissolved solids (TDS) and TSS removal. Therefore, optimization of these parameters would likely improve the efficiency of the EC process. In the present study, Response surface methodology (RSM) has been applied to optimize the process variables that can influence the treatment process. In particular, the data obtained from the experiments were analyzed for optimization of processing parameters with respect to the responses [16]. The use of RSM aids to find out the interaction between independent variables, to predict the responses of the system to any new condition by generating an appropriate mathematical model and to reduce costs by decreasing the number of experiments. Box–Behnken Design (BBD) is a commonly used RSM design that helps to avoid experiments performed under process conditions that could yield unsatisfied experimental results [17,18]. Hence, the present study focuses on the design and development of an EC unit for the effective removal of COD, TDS and TSS from RME using two different electrode materials (Iron (Fe) and Aluminum (Al)), evaluates and optimizes the impact of processing parameters (inter-electrode distance (4–7 cm), effluent pH (6–8), current density (10–30 mA/cm$^2$) and treatment time (20–40 min)). To the best of our knowledge, this is the first research work on the development of an EC unit that can use two electrodes (Fe and Al) without any modification of the experimental set up.

## 2. Materials and methods

### 2.1. Materials

The RME used in this work was trapped before being drained out from parboiling process from a paddy processing industry located in Madurai District, Tamilnadu, India. The effluent samples were collected in an air-tight container and stored at 4 °C to avoid any degradation before the analysis. The effluent characteristics were analyzed using APHA standard methods and found to be as follows: turbid, yellowish color, pH of 6.1, TDS of 8130 mg/L, electrical conductivity of 11,614 micro-mho/cm, initial COD of 450 mg/L respectively.

### 2.2. Electro coagulation unit

A glass reactor with capacity of two liters that enables to place the electrode inside was used to carry out the EC process. Two different electrode (Fe and Al) materials with a plate thickness of 5 mm, dimensions of 50 mm × 60 mm and an effective surface area of 25 cm$^2$ were used individually in order to compare the influence of the electrode material on the RME treatment. Proper inter-electrode distance was kept to minimize the electrical resistance of the electrodes. Current density was maintained at a particular level with the aid of a DC power supply (0–30 V, 0–2 A). The cathode and anode ends were connected to the power supply and the experiments were performed at room temperature. A schematic representation of the experimental set up is provided in Fig. 1.

### 2.3. Experimental procedure

The collected RME was poured into the reactor and the electrodes were placed in the proper position according to the experimental design. In order to attain a uniform mixing, a magnetic stirrer was used that kept a constant stirring speed of 2000 rpm. Experiments were performed at different inter-electrode distance (4–7 cm), effluent pH (6–8), current density (10–30 mA/cm$^2$) and treatment time (20–40 min). After each batch of experiments, impurities present on the electrode surface were removed by treating with hydrochloric acid solution (15 %) for 2 min followed by distilled water. Settling of flocs was carried out by transferring the effluent to another beaker and kept undisturbed for 20 min. The flocs were removed and the treated effluent samples were analyzed to determine the TSS, TDS and COD content.

### 2.4. Analytical procedure

The efficacy of the EC process using two different electrodes was evaluated by examining the TDS, TSS and COD content following the Standard protocol for Examination of Waste Water (APHA, AWWA 2012) after each experimental run.

### 2.5. Mathematical model development

The optimization of the experimental process parameters including inter-electrode distance (4–7 cm), effluent pH (6–8), current density (10–30 mA/cm$^2$) and treatment time (20–40 min) on EC of RME was carried out by applying a four factor, three level Box Behnken design (BBD). The influence of the process variables on the selected responses (COD, TDS and TSS removal) and their interaction effect were examined. The total number of experimental runs was calculated by applying the following eq. [19].

$$N = 2F(F-1) + C_0 \qquad (1)$$

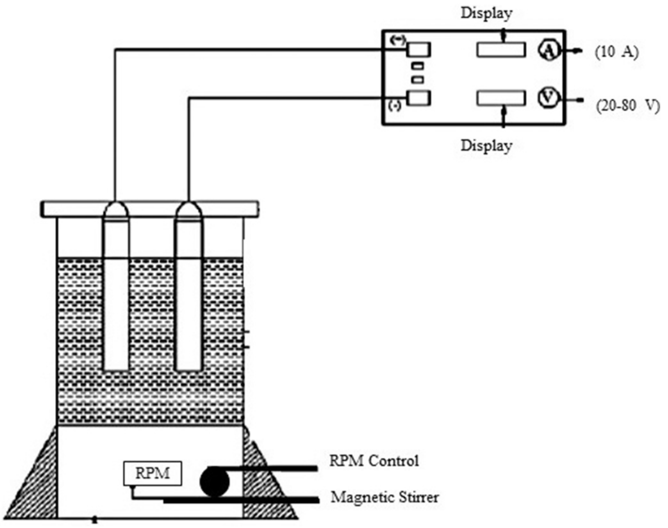

**Fig. 1.** Schematic representation of the electrocoagulation experimental setup.

where F denotes the total number of factors and $C_0$ the total number of central points. The developed design consists of 29 experimental runs and the experiments were performed randomly. The functional relation between the selected responses and the independent variables was evaluated by a non-linear regression method using a second order polynomial equation as shown below:

$$Z = \beta_0 + \sum_{b=1}^{k} \beta_b x_b + \sum_{b=1}^{k} \beta_{bb} x_b^2 + \sum_{a}^{k} \sum_{<j=2}^{k} \beta_{ab} x_a x_b + e \qquad (2)$$

where Z is the selected response; $x_a$ and $x_b$ are variables (a and b range from 1 to k); $\beta_0$ is the model intercept coefficient; $\beta_a$, $\beta_b$ and $\beta_{ab}$ are the interaction coefficients of linear, quadratic and the second-order terms; k is the number of independent parameters and $e_i$ the error.

The efficacy of the developed model was investigated using ANOVA. Fisher F-test was used to determine the statistical significance of the developed model. The Probability value was used to evaluate model terms with a 95 % confidence level. The entire statistical analysis was done using Design Expert Statistical Software package 13.0.0 (Stat Ease Inc., Minneapolis, USA).

## 3. Results and discussions

The interaction effect between four independent experimental parameters (detailed in Section 2.3) on the EC of the RME was studied using two different electrodes (Fe and Al) applying BBD. Twenty-nine statistically designed experiments were performed (Table 1) to determine the optimum parameters for the EC process.

### *3.1. Effect of process parameters on the EC of the RME*

### *3.1.1. Effect of initial pH*

To evaluate the influence of the effluent initial pH on COD, TDS and TSS removal efficiency, this parameter was adjusted prior to each experimental run using sodium hydroxide (NaOH) or hydrochloric acid (HCl) solutions. The generation of metal hydroxides and their stability strongly depends on the solution pH. Fig. 2 exhibits the relationship between the treatment efficiency and effluent pH. It is evident that the percentage removal of COD, TDS and TSS removal increased linearly with increasing initial pH and reached a maximum at a pH of 7. This is attributed to the increase in the formation of $Al(OH)_3$/$Fe(OH)_3$ ions at such pH. The percentage removal of COD, TDS and TSS improved from 48.13 to 74.11 %, 54.14 to 78.29 % and 46.60 to 72.75 %, respectively, using the Fe electrode. Similar results were observed using the Al electrode: the percentage removal increased significantly, from 68.50 to 94.72 % for COD, 68.75 to 96.11 % for TDS and 88.33 to 89.24 % for TSS. Thus, the electro coagulation performed at a suitable pH promotes effective and immediate adsorption of all soluble organic content in the effluent and their entrapment as colloidal material, which in turn facilitates the reduction of COD, TDS and TSS. A further increase in the pH results in the formation of $Al(OH)_4^-$/$Fe(OH)_4^-$ that dissolves rapidly in the effluent and prevents the formation of flocs [20,14], thereby reducing the percentage removal of COD, TDS and TSS.

**Table 1**
BBD design for the EC treatment of the RME.

| Std. order | pH ($X_1$) | TT (min, $X_2$) | CD (mA/cm$^2$, $X_3$) | IED (cm, $X_4$) |
|---|---|---|---|---|
| 1 | 6 | 20 | 20 | 5.5 |
| 2 | 8 | 20 | 20 | 5.5 |
| 3 | 6 | 40 | 20 | 5.5 |
| 4 | 8 | 40 | 20 | 5.5 |
| 5 | 7 | 30 | 10 | 4 |
| 6 | 7 | 30 | 30 | 4 |
| 7 | 7 | 30 | 10 | 7 |
| 8 | 7 | 30 | 30 | 7 |
| 9 | 6 | 30 | 20 | 4 |
| 10 | 8 | 30 | 20 | 4 |
| 11 | 6 | 30 | 20 | 7 |
| 12 | 8 | 30 | 20 | 7 |
| 13 | 7 | 20 | 10 | 5.5 |
| 14 | 7 | 40 | 10 | 5.5 |
| 15 | 7 | 20 | 30 | 5.5 |
| 16 | 7 | 40 | 30 | 5.5 |
| 17 | 6 | 30 | 10 | 5.5 |
| 18 | 8 | 30 | 10 | 5.5 |
| 19 | 6 | 30 | 30 | 5.5 |
| 20 | 8 | 30 | 30 | 5.5 |
| 21 | 7 | 20 | 20 | 4 |
| 22 | 7 | 40 | 20 | 4 |
| 23 | 7 | 20 | 20 | 7 |
| 24 | 7 | 40 | 20 | 7 |
| 25 | 7 | 30 | 20 | 5.5 |
| 26 | 7 | 30 | 20 | 5.5 |
| 27 | 7 | 30 | 20 | 5.5 |
| 28 | 7 | 30 | 20 | 5.5 |
| 29 | 7 | 30 | 20 | 5.5 |

### *3.1.2. Effect of treatment time*

Treatment time plays a crucial effect on the effluent treatment using EC process since it determines the cost effectiveness of the entire process [21]. Electrolysis facilitates the release of coagulants due to electro-dissolution of anodes. The process removal efficiency depends on the metal ion concentration generated from the electrodes. Upon increasing reaction time, the metal ion concentration and hydroxide flocs increase. In the current study, the reaction time was ranged from 20 to 40 min. The influence of reaction time on electro coagulation is displayed in Fig. 3. Results demonstrate that on increasing reaction time from 20 to 32 min, the percentage removal of COD, TDS and TSS improved from 62.33 to 74.11 %, 66.95 to 77.12 % and 60.72 to 71.59 % respectively for the Fe electrode. Similar response was observed using the Al electrode: the percentage removal of COD, TDS and TSS increased from 82.97 to 94.33 %, 82.06 to 96.11 % and 76.18 to 88.53 %, respectively, which is mainly attributed to the increase in metal ion formation at the initial stage of the electrolysis.

### *3.1.3. Effect of current density*

Current density determines the efficiency of the EC process, which influences the coagulant dosage and bubble generation rates, and aids to increase rapidly pollutant removal [22,23]. The ratio of the electrical current to the electrode surface area is defined as current density. The effect of the applied current density on the percentage removal of COD, TDS and TSS was investigated, and the results are included in Fig.4. Current density directly influences the effluent mixing property and the mass transfer rate at electrode surface. For both Fe and Al electrodes, it was found that the treatment efficiency improved as the initial current density was increased from 10 to 21 mA/cm$^2$. Further rise in the current density decreased significantly the percentage removal of TDS, TSS and COD using both electrodes. This behavior might be attributed to secondary reactions that occur at high current density, which lead to colloid charge reversal and thus cause re-dispersion of the colloids. Moreover, higher current density could also result in a reduction of the electrode lifetime [24].

### *3.1.4. Effect of inter-electrode distance*

Inter-electrode distance is a key process variable that controls the efficiency of the EC process [25]. In the current study, the effect of inter-electrode distance on EC was studied by modifying the distance from 4 to 7 cm at an interval of 1.5 cm, and the responses are depicted in Fig. 5. An effective response in treatment efficiency was observed for the shortest electrode distances. As the inter-electrode distance was increased linearly from 4 to 5.9 cm, the percentage removal of COD, TDS and TSS improved from 45.11 to 75.72 %, 55.75 to 78.71 % and 47.09 to 71.59 %, respectively, using the Fe electrode. Similar response was observed using the Al electrode: the percentage removal of COD, TDS and TSS improved from 67.90 to 94.33 %, 71.49 to 96.11 % and 62.86 to

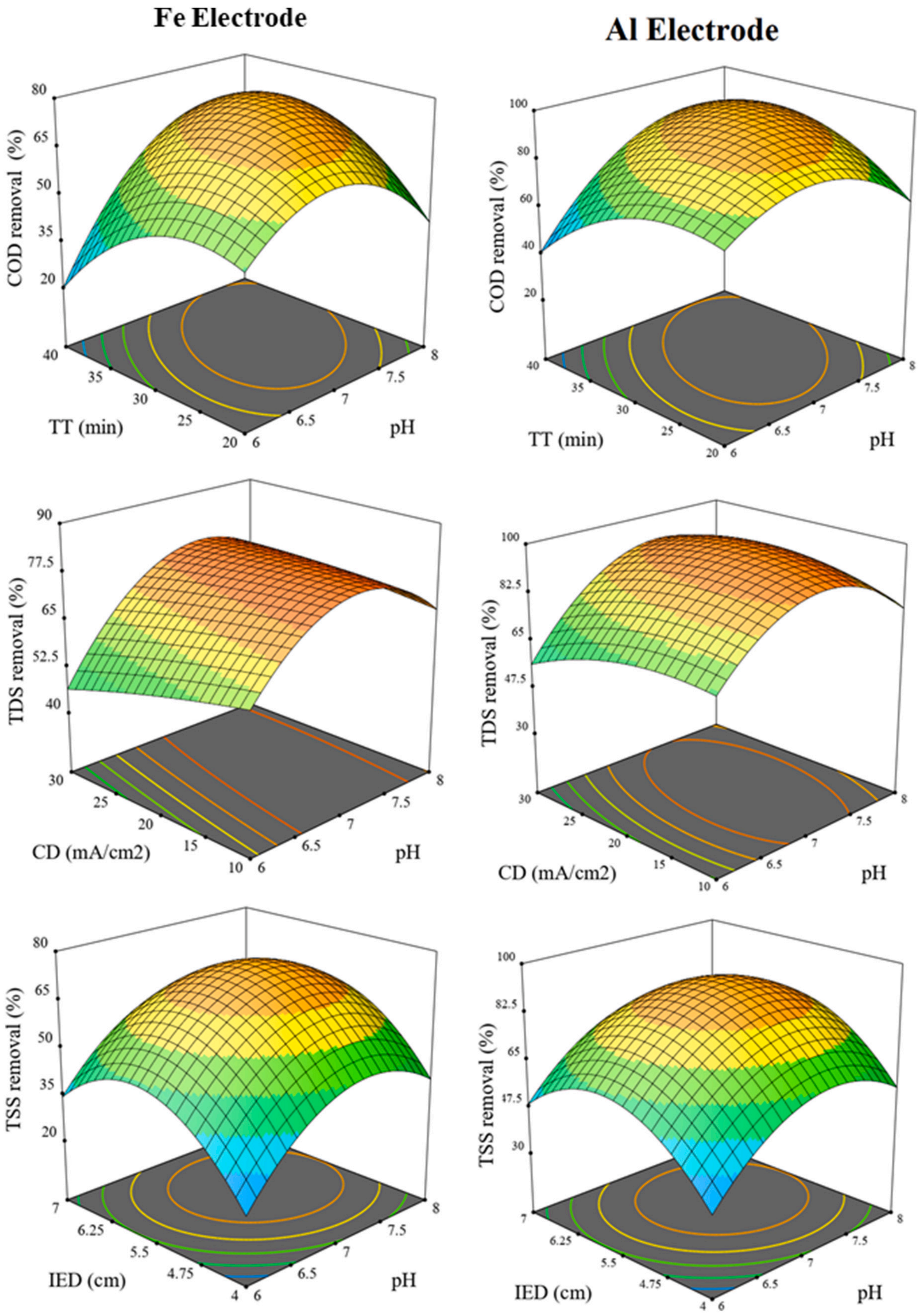

**Fig. 2.** Effect of pH on COD, TDS and TSS removal using Fe and Al electrodes.

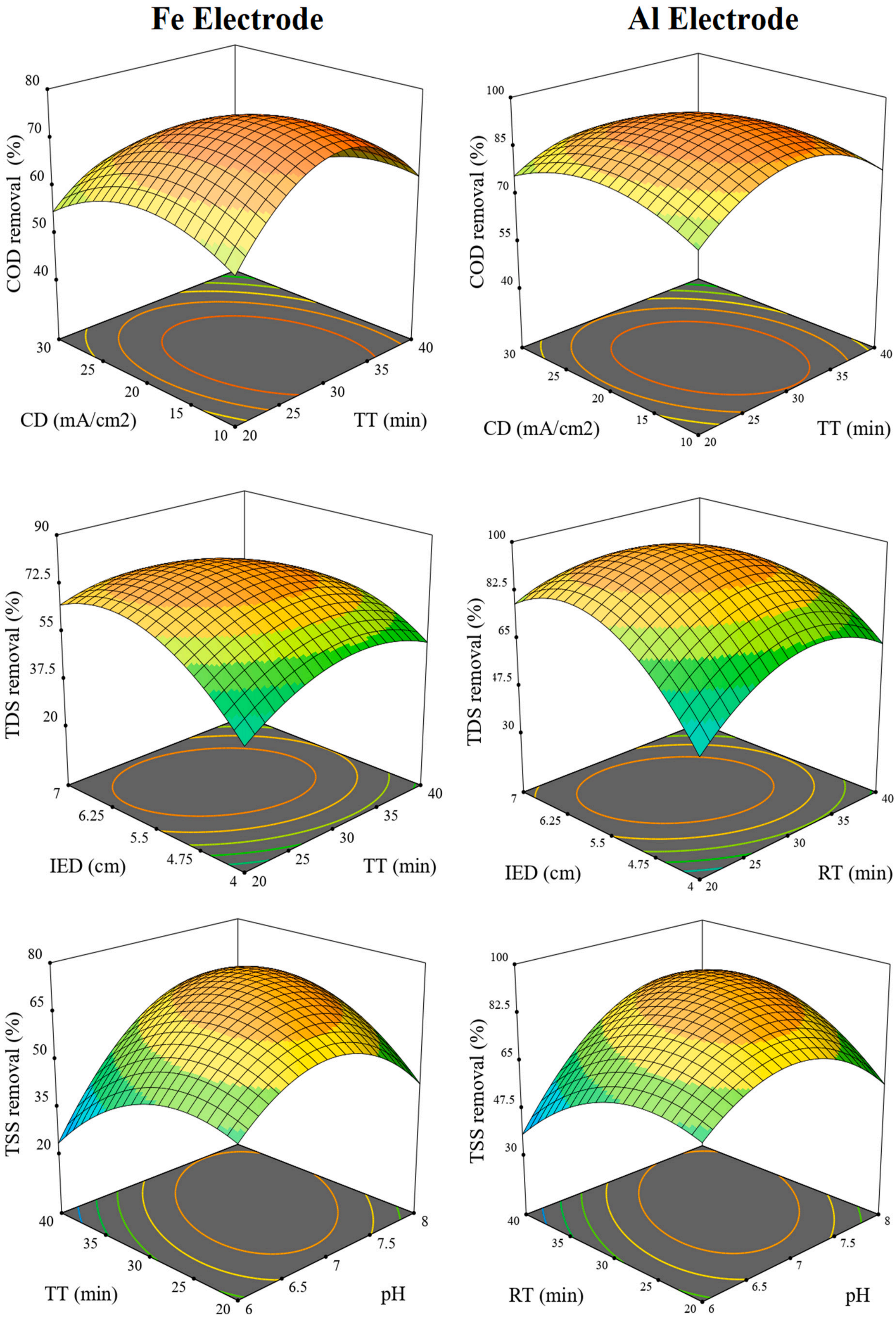

**Fig. 3.** Effect of treatment time on COD, TDS and TSS removal using Fe and Al electrodes.

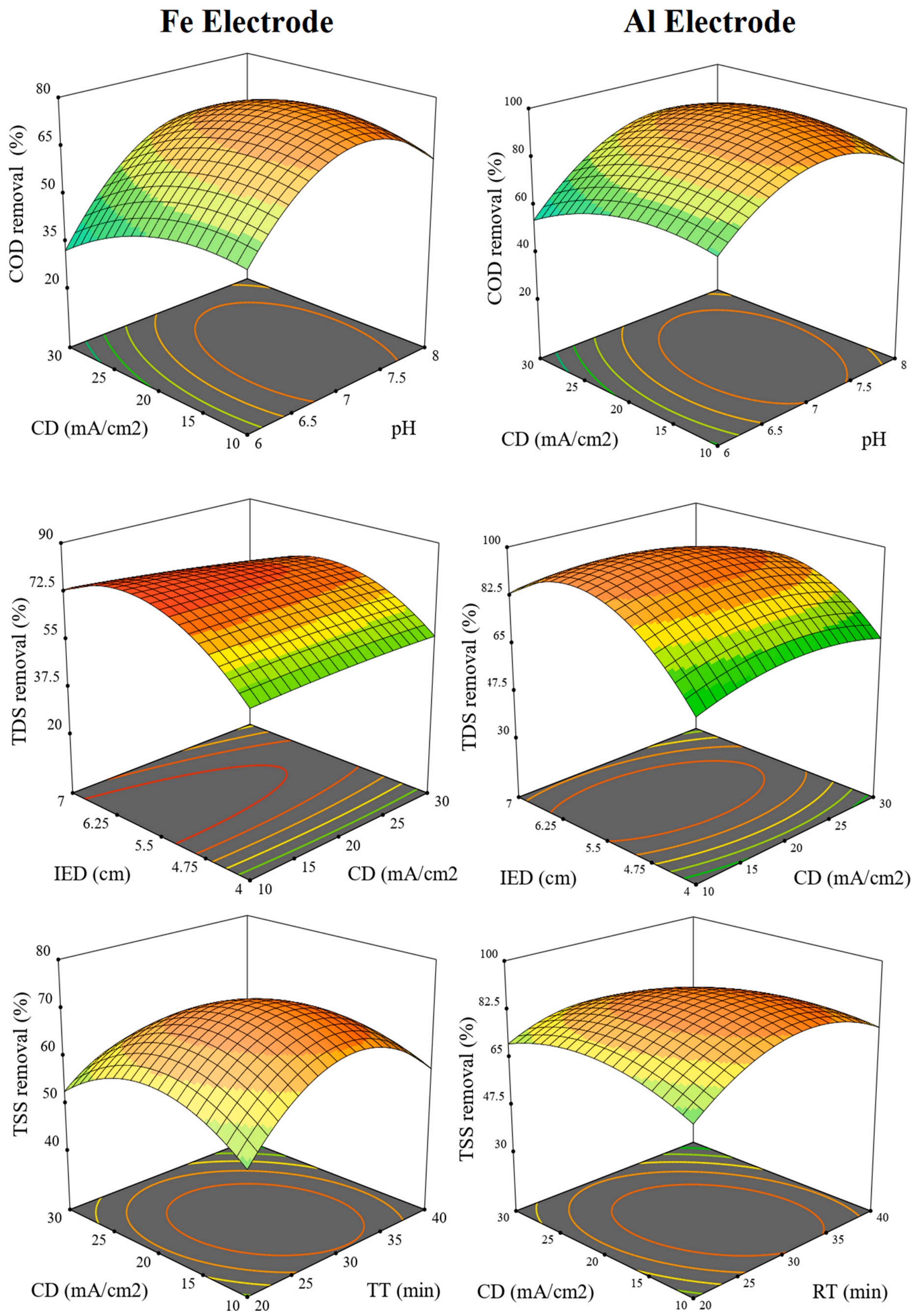

**Fig. 4.** Effect of current density on COD, TDS and TSS removal using Fe and Al electrodes.

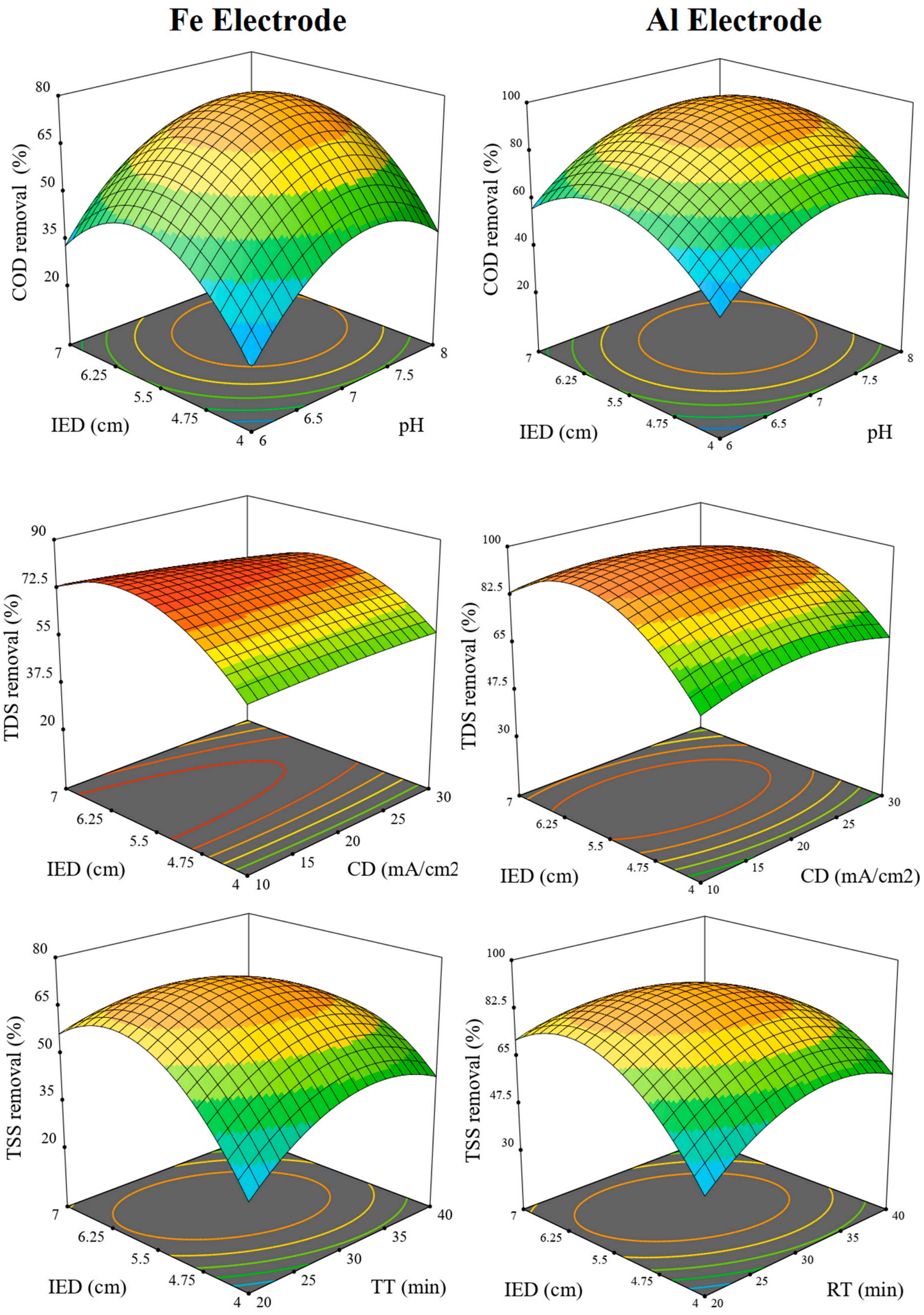

**Fig. 5.** Effect of inter-electrode distance on COD, TDS and TSS removal using Fe and Al electrodes.

88.54 %, respectively, as the electrode distance increased from 4 to 5.8 cm. This behavior could be attributed to a rapid rise in the anion discharge at the anode surface and an increased oxidation rate [26]. At higher inter-electrode distances (5.9 cm for the Fe electrode and 5.8 cm for the Al one), a decrease in treatment efficiency was observed for both electrodes. This could be ascribed to the fact that the floc distance growths with increasing inter-electrode distance and could eventually lead to a decrease in COD, TDS and TSS removal [27]. At higher distances the effective interaction between the oxidants and coagulants could become weaker, hence leading to a drop in the process efficiency [28].

### 3.2. Mathematical model development

The results obtained from BBD were evaluated using a multiple regression analysis method. The relationship between the independent process parameters and the responses were expressed using three different empirical models derived from the experimental data in order to assess the correlation between the responses and experimental parameters. The final model, in terms of coded factors, is shown below:

For the Fe electrode

$$\begin{aligned} Y_1 = {} & 74.59+(9.61*X_1)-(1.17^{*}\ X_2)-(4.92*X_3)+(7.37*X_4)+(12.76*X_1\ X_2)+(3.25*X_1\ X_3)+(0.32*X_1\ X_4)-(4.10*X_2\ X_3)-(5.60*X_2\ X_4) \\ & -(2.62*X_3\ X_4)-\left(17.80*X_1{}^2\right)-13.43*X_2{}^2\big)-\left(6.81*X_3{}^2\right)-\left(21.24*X_4{}^2\right) \end{aligned} \tag{3}$$

$$\begin{aligned} Y_2 = {} & 77.13+(8.31*X_1)-(0.86*X_2)-(2.41*X_3)+(5.94*X_4)+(13.91*X_1\ X_2)+(4.40*X_1\ X_3)-(1.22*X_1\ X_4)-(3.08*X_2\ X_3)-(6.37*X_2\ X_4) \\ & -(2.93*X_3\ X_4)-\left(14.67*X_1{}^2\right)-\left(11.03*X_2{}^2\right)-\left(0.86*X_3{}^2\right)-\left(14.25*X_4{}^2\right) \end{aligned} \tag{4}$$

$$\begin{aligned} Y_3 = {} & 71.59+(8.65*X_1)-(0.93*X_2)-(3.45*X_3)+(5.76*X_4)+(10.43*X_1X_2)+(5.07*X_1X_3)-(0.73*X_1X_4)-(3.93*X_2X_3)-(8.68*X_2X_4) \\ & -(1.59*X_3X_4)-\left(16.36*X_1{}^2\right)-\left(11.80*X_2{}^2\right)-\left(8.40*X_3{}^2\right)-\left(18.76*X_4{}^2\right) \end{aligned} \tag{5}$$

For the Al electrode

$$\begin{aligned} Y_1 = {} & -1028.46+(201.79*X_1)+(1.89*X_2)+(2.51*X_3)+(118.15*X_4)+(1.25*X_1X_2)+(0.37*X_1X_3)-(0.30*X_1X_4)-(0.038*X_2X_3)-(0.387*X_2X_4) \\ & -(0.19*X_3X_4)-\left(16.87*X_1{}^2\right)-\left(0.13*X_2{}^2\right)-\left(0.07*X_3{}^2\right)-\left(8.72*X_4{}^2\right) \end{aligned} \tag{6}$$

$$\begin{aligned} Y_2 = {} & -1082.16+(206.20*X_1)+(3.85*X_2)+(1.71*X_3)+\big(121.78*X_{4)}+(1.14*X_1X_2)+(0.50*X_1X_3)-(0.14*X_1X_4)*(0.053*X_1X_3*0.58*X_2X_4) \\ & -(0.14*X_3X_4)-\left(17.18*X_1{}^2\right)-\left(0.12*X_2{}^2\right)-\left(0.07*X_3{}^2\right)-\left(8.75*X_4{}^2\right) \end{aligned} \tag{7}$$

$$\begin{aligned} Y_3 = {} & +96.12+(8.39*X_1)-(1.13*X_2)-(1.99*X_3)+(6.10*X_4)+(12.96*X_1X_2)+(4.67*X_1X_3)-(1.90*X_1X_4)-(3.41*X_2X_3)-(8.27*X_2X_4) \\ & -(3.61*X_3X_4)-\left(19.01*X_1{}^2\right)-\left(15.18*X_2{}^2\right)-\left(5.79*X_3{}^2\right)-\left(18.51*X_4{}^2\right) \end{aligned} \tag{8}$$

where, $Y_1$, $Y_2$, $Y_{3,}$ denote the COD (%), TDS (%), TSS (%) removal, and $X_1$, $X_2$, $X_3$ and $X_4$ denote the pH, reaction time, current density and electrode distance, respectively.

Pareto analysis of variance (ANOVA) was performed in order to evaluate the experimental derived results. The higher model F-value (150.73, 303.79 and 52.49 for the percentage removal of COD, TDS and TSS, respectively, for the Fe electrode, and 970.07, 114.37 and 59.92 for the Al electrode, respectively,) and its associated lower *p*-values ($p < 0.0001$) demonstrate the validity of the developed mathematical models (Table 2).

The statistical analysis yielded the following values for the coefficient of determination ($R^2$): 0.993, 0.996 and 0.981 for the percentage removal of COD, TDS and TSS using the Fe electrode, as well as 0.999, 0.991 and 0.983 for the Al electrode. Analogously, the values of the adjusted coefficient of determination (adj-$R^2$) were: 0.986, 0.993 and 0.962 for the percentage removal of COD, TDS and TSS using the Fe electrode, as well as 0.997, 0.983 and 0.967 for the Al electrode. Regarding the predicted coefficient of determination (pre-$R^2$), the values obtained were: 0.964, 0.983 and 0.893 for the percentage removal of COD, TDS and TSS using the Fe electrode, as well as 0.995, 0.951 and 0.906 for the Al electrode. These coefficients measure how well a statistical model predicts an outcome. The better a model is at making predictions, the closer these coefficients will be to 1. Therefore, the very high values obtained herein corroborate that the selected model is the most appropriate to depict the relationship between the process parameters and the responses. The percentage coefficient of variance (CV) values were also calculated: 3.78, 1.82 and 5.89 for the percentage removal of COD, TDS and TSS for the Fe electrode, as well as 0.992, 2.89 and 4.36 for the Al electrode. The low values obtained indicate a high reliability in the experimental results. The value of adequate precision was higher than 26 for all the responses, which demonstrates the accuracy of the developed models.

**Table 2**
ANOVA for the responses.

| | Fe electrode | | | | | | Al electrode | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | COD (%) | | TDS (%) | | TSS (%) | | COD (%) | | TDS (%) | | TSS (%) | |
| Source | F value | p-value | F value | p-value | F value | p-value | F value | p-value | F value | p-value | F value | p-value |
| Model | 150.73 | <0.0001 | 303.79 | <0.0001 | 52.49 | <0.0001 | 970.07 | <0.0001 | 114.37 | <0.0001 | 59.92 | <0.0001 |
| $X_1$ | 310.13 | <0.0001 | 686.25 | <0.0001 | 109.33 | <0.0001 | 1970.92 | <0.0001 | 194.73 | <0.0001 | 125.45 | <0.0001 |
| $X_2$ | 4.6 | 0.05 | 7.29 | 0.0173 | 1.39 | 0.2577 | 91.22 | <0.0001 | 3.51 | 0.0818 | 0.1516 | 0.7029 |
| $X_3$ | 81.36 | <0.0001 | 57.47 | <0.0001 | 16.93 | 0.0011 | 200.76 | <0.0001 | 11.01 | 0.0051 | 16.67 | 0.0011 |
| $X_4$ | 182.45 | <0.0001 | 350.25 | <0.0001 | 48.54 | <0.0001 | 1137.64 | <0.0001 | 103.15 | <0.0001 | 53.93 | <0.0001 |
| $X_{12}$ | 182.17 | <0.0001 | 640.65 | <0.0001 | 53 | <0.0001 | 1288.04 | <0.0001 | 154.97 | <0.0001 | 65.59 | <0.0001 |
| $X_{13}$ | 11.83 | 0.004 | 64.08 | <0.0001 | 12.54 | 0.0033 | 113.65 | <0.0001 | 20.11 | 0.0005 | 12.64 | 0.0032 |
| $X_{14}$ | 0.1127 | 0.742 | 4.91 | 0.0438 | 0.2579 | 0.6195 | 1.75 | 0.2068 | 3.34 | 0.089 | 0.0232 | 0.8812 |
| $X_{23}$ | 18.75 | 0.0007 | 31.35 | <0.0001 | 7.06 | 0.0187 | 118.74 | <0.0001 | 10.73 | 0.0055 | 14.27 | 0.002 |
| $X_{24}$ | 35.07 | <0.0001 | 134.3 | <0.0001 | 36.71 | <0.0001 | 276.73 | <0.0001 | 63.09 | <0.0001 | 38.79 | <0.0001 |
| $X_{34}$ | 7.65 | 0.0152 | 28.32 | 0.0001 | 1.23 | 0.2858 | 69.48 | <0.0001 | 12.03 | 0.0038 | 2.43 | 0.1417 |
| $X_1^2$ | 574.66 | <0.0001 | 1155.24 | <0.0001 | 210.94 | <0.0001 | 3781.29 | <0.0001 | 540.63 | <0.0001 | 240.13 | <0.0001 |
| $X_2^2$ | 327.01 | <0.0001 | 652.81 | <0.0001 | 110.78 | <0.0001 | 2344.68 | <0.0001 | 344.74 | <0.0001 | 130.5 | <0.0001 |
| $X_3^2$ | 84.18 | <0.0001 | 3.97 | 0.0663 | 56.24 | <0.0001 | 846.62 | <0.0001 | 50.18 | <0.0001 | 50.55 | <0.0001 |
| $X_4^2$ | 818.04 | <0.0001 | 1089.47 | <0.0001 | 277.28 | <0.0001 | 5116.95 | <0.0001 | 513.04 | <0.0001 | 315.06 | <0.0001 |
| $R^2$ | 0.9934 | | 0.9967 | | 0.9813 | | 0.999 | | 0.9913 | | 0.9836 | |
| Adj- $R^2$ | 0.9868 | | 0.9934 | | 0.9626 | | 0.9979 | | 0.9827 | | 0.9672 | |
| Pre- $R^2$ | 0.9644 | | 0.9836 | | 0.8931 | | 0.9955 | | 0.9519 | | 0.9068 | |
| CV (%) | 3.78 | | 1.82 | | 5.89 | | 0.992 | | 2.89 | | 4.36 | |
| Adeq.Pre. | 40.95 | | 61.70 | | 24.37 | | 106.61 | | 37.84 | | 25.70 | |

### 3.3. *Diagnostics of model adequacy*

In general, the model needs to be validated in order to confirm that it provides an accurate approximation to the actual (experimental) values. Investigating and optimizing the parameter without evaluating the satisfactory fitness of the model could lead to misleading and poor results. In this regard, diagnostic plots like normalized plot and the parity plot between predicted and actual values aid to validate the satisfactory fitness of the developed model and to analyze the relationship between the actual and predicted values. The parity plots for the selected responses and the two electrodes are provided in Fig. 6. As can be observed, for both electrodes, the values lie close to a straight line, which corroborates that the experimental derived results are in accordance with the predicted ones. Overall, results suggest that the developed models for the Fe and Al electrodes can be applied to identify the optimal experimental conditions that provide the best percentage reduction of COD, TDS and TSS.

Additionally, the normality of the residuals was examined. The normal distribution of the residuals for the different responses selected was assessed using the normal probability plot, as depicted in Fig. 7 for both electrodes. As can be observed, the values lie in a straight line and the residuals are small, which corroborates that the data were normally distributed, hence that the developed models are able to predict the data with high precision.

### 3.4. *Optimization and authentication of process parameters and responses*

Derringer's desired function methodology was applied to identify the optimum experimental conditions to attain the highest percentage removal of COD, TDS, and TSS for both electrodes. The best conditions for the Fe electrode were found to be a pH of 7.29, a current density of 18.64 mA/cm$^2$, an inter-electrode distance of 5.8 cm, and an electrolysis time of 31.33 min, which led to percentage removals of 77.04, 79.18 and 73.13 for COD, TDS and TSS, respectively. Analogously, using the Al electrode, a pH of 7.29, a current density of 20.89 mA/cm$^2$, an inter-electrode distance of 5.6 cm, and an electrolysis time of 30.92 min were the optimal conditions, that led to the highest percentages of removal: 95.46, 97.11 and 89.83 for COD, TDS and TSS. the Al electrode. Taking into account the practical feasibility, the optimal conditions were slightly modified, and experiments were carried out under these conditions: a pH of 7.3, a current density of 19 mA/cm$^2$, an inter-electrode distance of 5.8 cm, and an electrolysis time of 32 min, for the Fe electrode, as well as a pH of 7.3, a current density of 21 mA/cm$^2$, an inter-electrode distance of 5.6 cm, and an electrolysis time of 31 min for the Al electrode. Results revealed that the percentages of removal of COD, TDS and TSS (76.63, 78.56 and 72.03 for the Fe electrode, as well as 94.79, 96.62 and 88.76 for the Al electrode, respectively) were very close to the predicted values.

### 3.5. *Cost analysis (CA)*

The following data was used for estimating the operating cost: Rectifier installation cost (800 $), EC tank installation cost (100 $ for a capacity of 100 m$^3$/day), maintenance cost (0.003 $/m$^3$), electricity cost (0.085 $/h), labor cost (0.005 $/m$^3$), transportation and disposal of sludge (0.01$/kg), Fe and Al electrode cost (3.41$/kg and 2.23$/kg, respectively). Cost calculation was derived from the equation described by Sridhar et al. [29] taking into account the modified optimal conditions for both electrodes detailed in the previous section. Overall, the operating cost for the Fe electrode was 4.27 $/m$^3$ of effluent, while for the Al one was 2.36$/m$^3$.

## 4. Conclusion

Environmental concerns are the major driving force accounting for the development of novel strategies for treatment of effluents. EC process was successfully used to treat the RME using Fe and Al electrodes. A four factor, three level BBD was used to investigate and optimize the process parameters (inter-electrode distance (4–7 cm), effluent pH (6–8), current density (10–30 mA/cm$^2$) and treatment time (20–40 min)). All the process parameters had a significant effect on the percentage removal of COD, TDS and TSS from RME using both electrodes. Experimental data were statistically analyzed, and mathematical models were developed for the responses (COD, TDS and TSS). The optimal conditions to treat the RME using EC process were derived and validated. Validation experiments carried out under the optimal condition

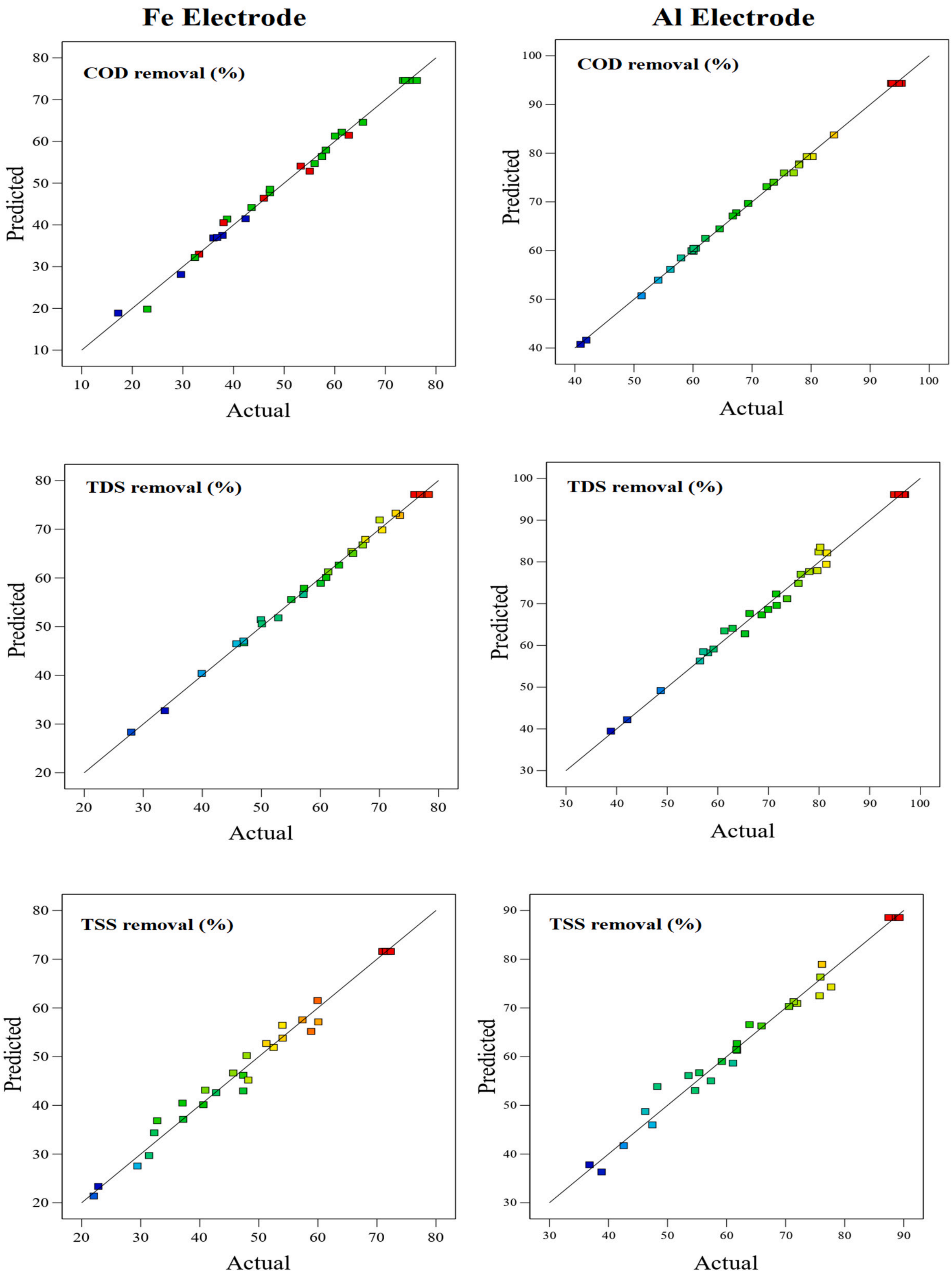

**Fig. 6.** Parity plots between predicted and actual values for the different responses using Fe and Al electrodes.

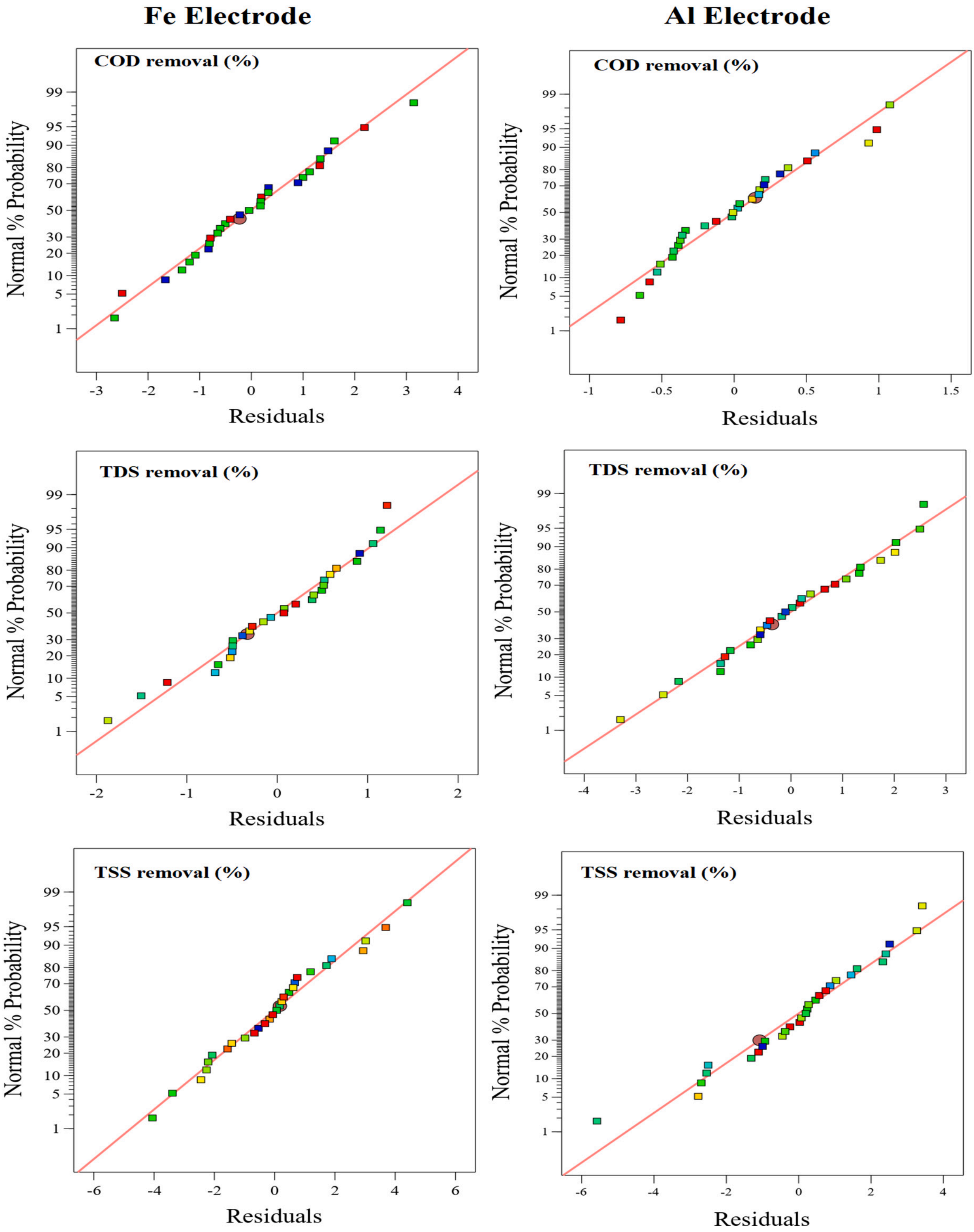

**Fig. 7.** Normal probability plots for the different responses using both electrodes.

revealed that the percentages of removal of COD, TDS and TSS were higher for the Al electrode (94.79, 96.62 and 88.76, respectively) than for the Fe electrode (76.63, 78.56 and 72.03). Similarly, the calculated operating cost was lower for the Al electrode when compared with the Fe electrode. Results clearly demonstrate that the EC process using the Al electrode can be applied to effectively reduce the COD, TDS and TSS content, and the treated water could also be used for agriculture purposes.

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## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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## Data availability

Data will be made available on request.

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