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Optimization of Continuous Electro-Fenton and Photo electro-Fenton Processes to Treat Iraqi Oilfield Produced Water Using Surface Response Methodology

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Abstract. Considerable amounts of produced water (PW) is usually accompanied with the production of oil. Most countries with oilfields are generally water stressed countries. This study proposed electro-Fenton (EF) as alternative for the degradation of organic pollutants in PW. Continuous electro-Fenton processes was investigated using dimensionally stable anode Ti-RuO₂/IrO₂ and activated carbon fiber felt (ACFF) cathode. The effect of crucial process variables, namely, initial ferrous ions concentration (0.1-0.5 mM), current intensity (100-500 mA), and residence time (22-81 minutes) on the removal efficiency of COD was studied using contour and response surface plots. The experimental results were analyzed by analysis of variance (ANOVA). Multiple response optimization for continuous EF experiments reveals that at optimum conditions (initial ferrous ion concentration of 0.306 mM, current intensity of 156.6 mA, and residence time of 81.0 min) the COD removal efficiency was 73.33% and electrical energy consumption was 0.901 kWh/kg COD. Improvement of continuous EF process was investigated using UVA irradiation (Photoelectron-Fenton). It was found that COD removal efficiency for continuous EF was increased to 81.1% and 86.0% when using one and two UVA lamps (3 Watt each) respectively. It is concluded that EF is an effective process for treating produced water and further improvement can be achieved by photo assisting the process.

Keywords: Electro-Fenton; Photoelectron-Fenton; Produced water; Response surface methodology; COD removal.

1. Introduction

Oil is produced with large volume of wastewater, it is estimated that three barrels of water are produced for every barrel of crude oil [1]. Many countries have implemented more strict regulations for discharging PW [2].

Produced water (PW) is a complex mixture of dissolved and particulate organic and inorganic chemicals in water (mostly oils, salts, and minerals) [3]. Some factors such as geological location of the field, lifetime of its reservoirs affect the physical and chemical properties of produced water.

The oil content in produced water is frequently classified into four groups according to its nature of physical phase, which are: free oil (larger than 150 μm), dispersed oil (20-150 μm), emulsified oil (less than 20 μm), and dissolved oil.

Treatment methods of produced water can be classified into three main categories namely, primary to separate free oil such as gravity separators, secondary to removal dispersed oil such as coagulation and



flotation processes, and tertiary treatment to eliminate emulsified and soluble oil such as advanced oxidation processes (AOPs).

AOPs defined as the oxidation methods of aqueous solutions in the presence of highly active materials which can destroy the pollutants. Hydroxyl radical is a powerful oxidant which is able to non-selectively destroy most organic contaminants until their complete mineralization into CO₂, water and inorganic ions [4].

The conventional Fenton method, which achieved by the addition of Fe(II) salt to hydrogen peroxide (H₂O₂) in aqueous media has been found since the end of the 19th century. This Fenton reaction generates hydroxyl radicals (•OH) under acidic conditions that can oxidize organics and convert it to non-toxic products. However, this Fenton process produces large amounts of Fe(III) oxyhydroxide solid byproduct that inhibiting the catalytic role of Fe(II) in generating •OH [5]. Electro-Fenton (EF) is one approach to resolve these issues in conventional Fenton. In EF the Fe(III) reduced to Fe(II) at the cathode. Also, hydrogen peroxide in-situ generated at the cathode. [5]

New AOPs based on the electrochemical technology have been investigated in recent years, i.e., the so-called Electrochemical Advanced Oxidation Processes (EAOPs), have been developed. The EAOPs provide several advantages for the prevention and remediation of pollution problems because electron is a clean reagent. Other advantages include high energy efficiency, amenability to automation, easy handling because of the simple equipment required and safety because they operate under mild conditions (room temperature and pressure) [4].

In the EF process, hydroxyl radicals are produced by the reaction between hydrogen peroxide and ferrous ions, which can destroy organic compounds. The reduction of ferric ion to ferrous ion, which can reduce iron sludge production is one advantage of the EF process over the conventional Fenton process [6].

Electro-Fenton mainly relies on in situ and catalytic electro generation of Fenton's reagent – a mixture of Fe(II) ions and hydrogen peroxide (H₂O₂) to produce hydroxyl radicals (•OH) and react with organic pollutants in aqueous media, leading to their destruction (as equations 1-4) [7].



The optimum pH for COD removal is 3. A pH greater than 3 lower the COD removal efficiency. At a higher pH, the oxidation efficiency of EF process decreases due to the formation of low active Fe(OH)₃, which has a lower tendency to react with hydrogen peroxide [6]. pH lower than optimum affects the pollutant removal by producing less hydroxyl radicals, increased scavenging effects of H⁺ and hydroxyl radicals [8].

Electro-Fenton method has been applied successfully for the treatment of various wastewater such as paper mill wastewater [9], fertilizer manufacturing wastewater [10], Dairy industry wastewater [11], synthetic dye wastewater [12], photographic processing wastewater [13], and petroleum refinery wastewater [14].

The efficiency of electro-Fenton process can be further improved in the presence of UV irradiation by a process called photoelectro-Fenton (PEF). The catalytic effect of Fe²⁺ can be enhanced by assisting electro-Fenton process with UV irradiation. The photoelectro-Fenton process can increase the regeneration rate of Fe²⁺ in the presence of UV. An increased concentration of OH increases the oxidative capability of the process. In addition, H₂O₂ produces two OH by photocatalytic effect of UV irradiation (equation 5) [7].



The PEF process involves the solution irradiation with UVA light, whose action is quite complex. Photons can prevent the large accumulation of Fe(III) species, responsible for a gradual deceleration of decontamination, thanks to the reductive photolysis of [Fe(OH)]²⁺ via reaction (8). Moreover, this enhances the Fe²⁺ regeneration and the production of additional amounts of •OH. UVA photons can also photolyze organic intermediates like Fe(III)-carboxylate complexes, originated from the destruction of aromatic pollutants, via the general reaction (9) [15].



The traditional technique of experimental design, in which one process variable is changed, while the other variables are fixed, does not recognize the interaction between the process variables. Response surface methodology (RSM) is able to assess this interaction [6]. Response surface methodology finds the optimum values of process variables for a desirable response by using a statistical-based technique to evaluate the simultaneous effects between these variables [16, 17].

The aim of this work is to examine the effectiveness of Electro-Fenton process for treating Iraqi oilfield produced water. The response surface methodology has been employed to optimize the process conditions for EF for maximizing COD removal efficiency while minimizing electrical energy consumption. Moreover, this study aimed to further improve EF efficiency by UVA irradiation (photo-EF).

2. Material and Methods

2.1. Produced water sample

The produced water sample was collected from oilfield, midland oil company, Iraq. First, the sample was treated by Electrocoagulation unit to eliminate suspended and dissolved solids and to reduce COD. Then the sample was filtered, and analyzed. The characteristics of produces water used in this study was as follow: COD=457 mg/l, pH=7.2, oil and grease= 86 mg/l, NTU= 1.7.

2.2. Electrochemical reactor

The laboratory scale electro-Fenton system consists mainly of electrochemical reactor, feed tank, effluent tank, feed pump, magnetic stirrer, air pump, and DC power supply as shown in Figure (1).

The electrochemical reactor was a cylindrical shape made of Perspex with one liter working volume. The reactor has two openings, one near the bottom serve for feed inlet from feed pump and the other at the top, serve for effluent outlet by gravity.

Two electrodes were fixed inside the reactor. The anode was DSA Ti-RuO₂/IrO₂ mesh placed in the centre of the reactor. The cathode was a cylindrical activated carbon fiber felt (ACFF) fixed on the reactor inside wall.

The feed tank was a cylindrical Perspex tank with 2 L working volume, while the effluent tank was a cylindrical glass beaker. The electrochemical reactor placed above a magnetic stirrer (Type Jenway 1000) which keep homogeneity of the electrolyte solution. Compressed air was fed to the cathode by an air pump. Two UVA lamps (3 Watt each) was fixed in a glass jacket inside the reactor. Figure (2) is a photograph of the continuous experimental system used in this work.

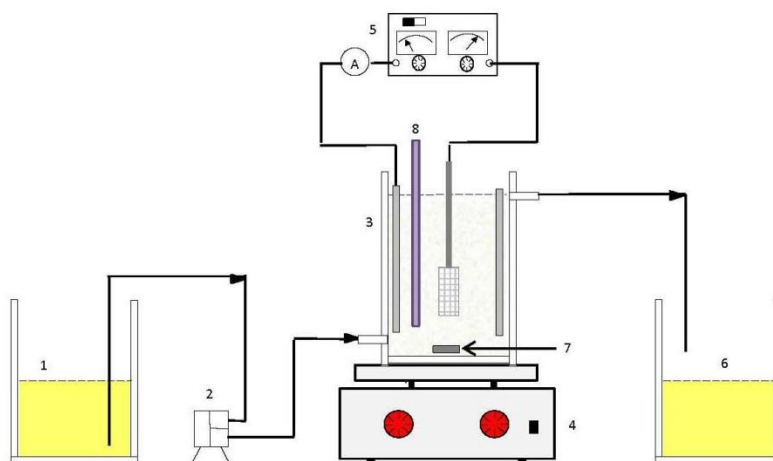


Figure 1. Schematic diagram of continuous photo-electro-Fenton experimental system. (1) Feed tank (2) Pump, (3) EC reactor, (4). Magnetic stirrer, (5) DC power supply, (6) Product tank, (7) Teflon bar, (8) UVA lamp.



Figure 2. Photograph of continuous photoelectron-Fenton experimental system.

2.3. Analytical procedure

All samples were filtered through Whatman filter paper with a pore size of 11 μm . COD was analyzed using a COD thermoreactor (RD125, Lovibond) and a direct reading spectrophotometer (MD200, Lovibond)

The equation used to calculate the percentage of COD removal (R%) was:

$$R\% = \frac{\text{COD}_0 - \text{COD}}{\text{COD}_0} \times 100 \quad (10)$$

Where COD_0 and COD are the initial and final chemical oxygen demand respectively.

2.4. Experimental Design

A total of 20 experiments were performed to optimize and determine the relationship between the removal efficiency of COD with respect to crucial operating parameters, i.e., initial Fe(II) ions concentration (0.1-0.5 mM), current intensity (100-500 mA), and residence time (22-81 minutes). Response surface Methodology (RSM), the central composite design (CCD) was performed using MINITAB software (version 17). Experimental data were fitted to a quadratic equation:

$$Y_i = b_o + \sum b_i x_i + \sum b_{ii} x_i^2 + \sum b_{ij} x_i x_j \quad (11)$$

Where b_o , b_i , and b_{ij} are the regression coefficients for the equation terms. Y_i is the percentage removal of COD (R%), and electrical energy consumption (EEC). The regression coefficients were analyzed by the F-test and P-value. The statistical significance of the model was tested by the analysis of the variance (ANOVA). The relationship between the response and the variables was used to construct a three dimensional surface plots to study the effect of variables on the response. Multiple response optimization of the EF process was done to determine the optimum parameters for maximum COD removal efficiency and for minimum power consumptions.

2.5. Experimental Procedure

All electro-Fenton experiments were conducted in a continuous mode under galvanostatic conditions. Before starting-up the process, compressed air was fed to the cathode by an air pump with 2.5 L/min for 15 minutes to saturate the solution with oxygen and was maintained during the process of electrolysis. In each run, produced water introduced into the feed tank and the electrochemical reactor and all runs were performed under stirring at 500 rpm. Initial pH values were adjusted to 3 with 0.1 M solution of H_2SO_4 . The feed pump was adjusted to the desired flow rate and switched on.

The average of voltage from the start to the end of experiment was used for the determination of energy consumption, at the end of each run, the power supply was switched off and a sample of the effluent was taken from the reactor exit, and filtrate then analysed by COD thermoreactor and direct reading spectrophotometer.

One of most important parameters that affect the application of any method of wastewater treatment is the cost. The operation cost in EF process includes material, consuming of energy cost, labor, maintenance, and disposal and fixed cost. Consuming of energy cost is the major cost in EF process. The electrical energy consumption (EEC) for EF treatment was calculated using the following equation [15]:

$$EEC = \frac{U.I.t. 1000}{(COD_o - COD) V} \quad (12)$$

where:

EEC = electrical energy consumed (kWh/kg COD)

U= voltage (Volt)

I= current intensity (A)

t= time (h)

V= water volume (Liter)

COD_o and COD= initial and final COD (mg/l)

3. Results and Discussion

3.1. Experimental Design Analysis

The results of the total number of 20 experiments with six center points based on the response surface methodology (RSM) with central composite design (CCD) are shown in Table 1.

Table1. Experimental design and the obtained responses.

Run	Fe(II) concentration, C, (mM)	Current intensity, I, (mA)	Residence Time, RT, (min)	COD removal efficiency, R%	Electrical Energy Consumption, EEC (kWh/kg COD)
1	0.1	0.5	81.0	66.3	5.2894
2	0.5	0.3	51.5	63.4	1.7769
3	0.3	0.3	51.5	71.2	1.5825
4	0.3	0.3	51.5	72.3	1.5580
5	0.3	0.1	51.5	59.7	0.3935
6	0.3	0.3	51.5	71.7	1.5713
7	0.3	0.3	51.5	72.1	1.5621
8	0.5	0.1	81.0	62.5	0.5911
9	0.1	0.5	22.0	49.2	1.9373
10	0.3	0.3	51.5	70.9	1.5894
11	0.5	0.1	22.0	46.7	0.2148
12	0.3	0.3	22.0	58.3	0.8253
13	0.3	0.3	81.0	77.1	2.2986
14	0.3	0.3	51.5	71.7	1.5708
15	0.5	0.5	81.0	70.3	4.9907
16	0.3	0.5	51.5	65.6	3.3994
17	0.1	0.1	22.0	46.4	0.2161
18	0.1	0.3	51.5	61.0	1.8471
19	0.5	0.5	22.0	49.9	1.9101
20	0.1	0.1	81.0	64.4	0.5730

The relationship between COD removal efficiency (R%), electrical energy consumption (EEC), and the three process variables were fitted to a second order polynomial, Eqn. (13), (14) as given below:

$$\text{COD (R\%)} = 21.69 + 94.1 I + 95.3 C + 0.410 RT - 155.6 I^2 - 166.1 C^2 - 0.00131 RT^2 + 19.9 I^2C + 0.079 I^2RT + 0.022 C^2RT \quad (13)$$

$$\text{EEC} = 0.297 - 2.115 I - 2.313 C + 0.00280 RT + 6.64 I^2 + 4.53 C^2 - 0.000079 RT^2 - 1.071 I^2C + 0.12075 I^2RT - 0.00543 C^2RT \quad (14)$$

Table 2 show the ANOVA for the removal efficiency of COD (R%) response and variables selected to fit the model. The F-value of 52.34 for the model implying that the model is significant. A p-value lower than 0.05 indicates that the model is statistically high significant. Terms with p-values less 0.05 indicates that these terms are significant. The model was also tested using the determination coefficient (R^2). The closer R^2 values to 1, the stronger the model and better predict of response. The determination coefficient value of 0.9792 for COD removal efficiency illustrate that the data prediction ability of the response surface model was satisfactory.

Table 2. ANOVA for COD removal efficiency (R%).

Source	Sum of Squares	DF	Mean square	F-value	p-value	Remark*
Model	1625.97	9	180.663	52.34	0.000	S
I	46.66	1	46.656	13.52	0.004	S
C	2.89	1	2.894	0.84	0.381	NS
RT	812.34	1	812.342	235.33	0.000	S
I*I	106.55	1	106.549	30.17	0.000	S
C*C	121.41	1	121.412	35.17	0.000	S
RT*RT	3.57	1	3.571	1.03	0.333	NS
I*C	5.07	1	5.072	1.47	0.253	NS
I*RT	1.76	1	1.758	0.51	0.492	NS
C*RT	0.13	1	0.133	0.04	0.849	NS
Residual	34.52	10	3.452			
Lack-of-fit	33.05	5	6.610	22.51	0.002	
Pure error	1.47	5	0.294			
Cor total	1660.49	19				

* S=significant, NS=not significant

Table 3 show the ANOVA for the electrical energy consumption (EEC) response and variables selected to fit the model. The F-value of 581.12 for the model implying that the model is significant. A p-value lower than 0.05 indicates that the model is statistically high significant. Comparison between the actual and predicted values of EEC showed a good correlation between the observed and the predicted values with coefficient of determination (R^2) value of 0.9981.

Table 3. ANOVA for electrical energy consumption (EEC).

Source	Sum of Squares	DF	Mean square	F-value	p-value	Remark*
Model	36.3510	9	4.0390	581.12	0.000	S
I	24.1442	1	24.1442	3473.78	0.000	S
C	0.0144	1	0.0144	2.07	0.181	NS
RT	7.4653	1	7.4635	1073.82	0.000	S
I*I	0.1943	1	0.1943	27.95	0.000	S
C*C	0.0904	1	0.0904	13.01	0.005	S
RT*RT	0.0130	1	0.0130	1.87	0.201	NS
I*C	0.0147	1	0.0147	2.11	0.177	NS
I*RT	4.0603	1	4.0603	584.18	0.000	S
C*RT	0.0080	1	0.0080	1.14	0.310	NS
Residual	0.0695	10	0.0070			
Lack-of-fit	0.0688	5	0.0138	97.01	0.000	
Pure error	0.0007	5	0.0001			
Cor total	36.4206	19				

* S=significant, NS=not significant

3.2. Effect of Process Variables on COD Removal

Figures. 3-5 represents the three-dimensional (3D) response surface and two-dimensional (2D) contour plots of COD removal efficiency as a function of applied current intensity, initial Fe^{2+} concentration, and residence time.

In the range of residence time (RT) used in this study (22-81 min), it was found that the COD removal efficiency increase almost linearly with the RT of produced water inside the continuous EF reactor. No plateau was found when increasing RT on the efficiency of organics degradation. Increasing time lead to increase the degradation of organic compounds to simpler products and then further decomposed to simpler and lower molecules.

Also in the range of initial ferrous ion concentration (0.1-0.5 mM) it was observed that COD removal efficiency is proportional to the ferrous ion concentration in the range of 0.1-0.31 mM. This increase is due to the catalytic decomposition of H_2O_2 by ferrous ions as represented in the Fenton reaction (equation 2.14). At higher ferrous ion concentration (more than 0.31 mM) it was found that the COD removal efficiency starts to decrease. This decrease is due to the reaction of excess ferrous ions with the generated hydroxyl free radicals (as shown in equation 2.18) which leads to harm the process efficiency.

The COD removal efficiency increase with increasing the applied current intensity from 100 – 340 mA. Beyond 340 mA the COD removal efficiency starts to falls due to increase the reaction that scavenged the hydroxyl radicals.

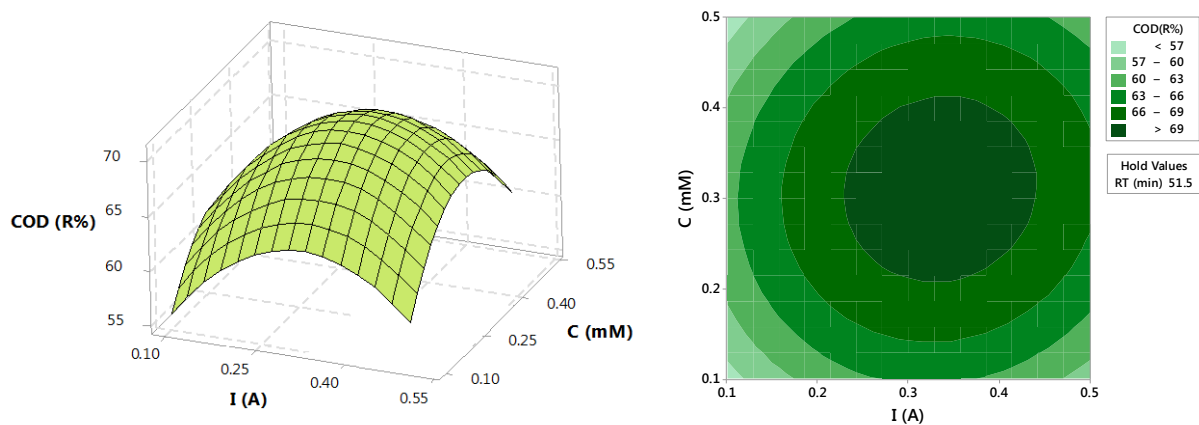


Figure 3. Surface plot and contour plot for COD removal efficiency (R%) vs C, I, at RT = 51.5 minutes.

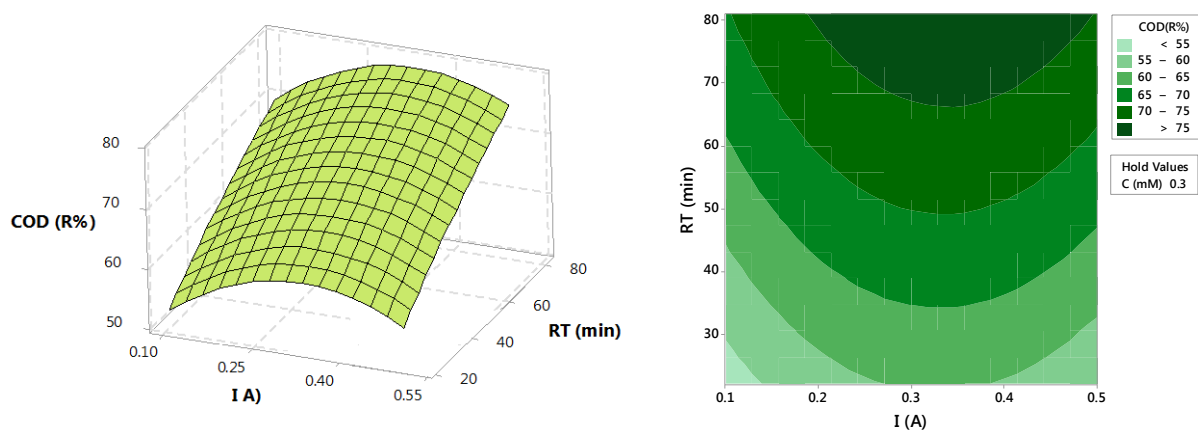


Figure 4. Surface plot and contour plot for COD removal efficiency (R%) vs RT, I, at C = 0.3 mM.

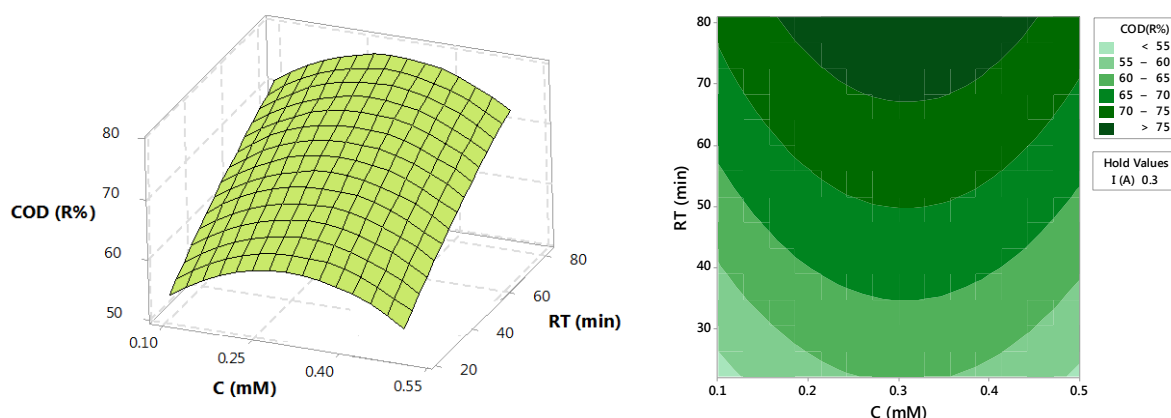


Figure 5. Surface plot and contour plot for COD removal efficiency (R%) vs RT, C at I=0.3A.

3.3 Optimization and Validation

Multiple response optimization was performed for maximizing COD removal efficiency (R%) while minimizing electrical energy consumption (EEC). The optimization gave initial Fe^{2+} concentration of 0.306 mM, current intensity of 156.6 mA, and residence time of 81.0 minutes as optimal points. The optimization predicts removal efficiency of 73.33% and energy consumption of 0.901 kWh/kg COD at these optimal points. Figure.6 illustrate the response optimization of COD removal efficiency, and electrical energy consumption.

Validation experiment conducted under the optimal parameters gave 73.9% COD removal efficiency and 0.900 kWh/kg COD electrical energy consumption, which in agreement with the predicted values.

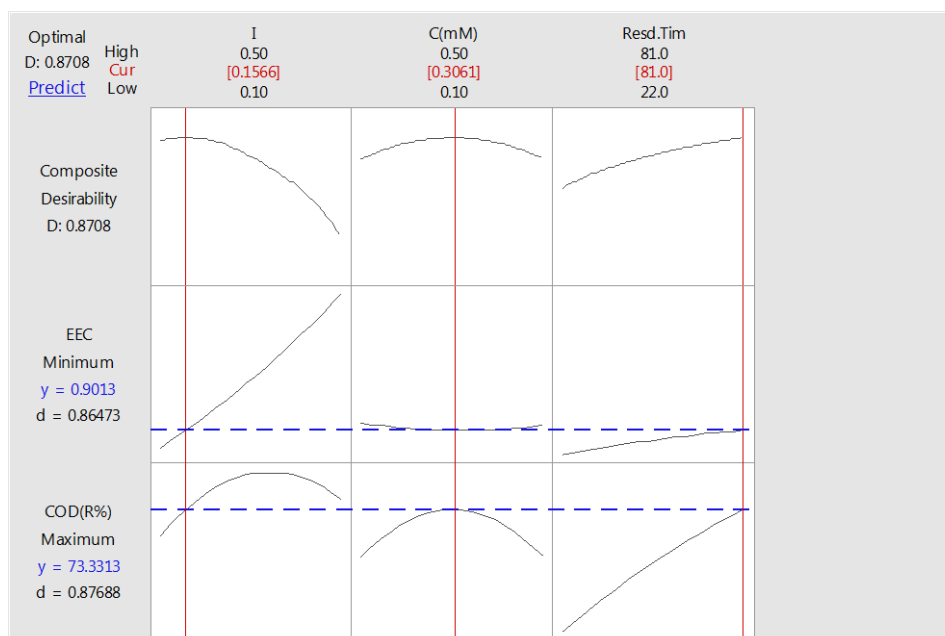


Figure 6. Multiple response optimization of COD removal efficiency (R%), and electrical energy consumption (EEC).

4. Photoelectro-Fenton

To investigate the effect of UVA irradiation on the COD removal efficiency, one and two UVA lamps (3 W each) was applied to the reaction media at optimum operating conditions (initial Fe^{2+} concentration of 0.306 mM, current intensity of 156.6 mA, and reaction time of 81.0 minutes). It was

found that COD removal efficiency was increased from 73.9% to 81.1% and 86.0% using one and two UVA lamps respectively.

5. Conclusions

Electro-Fenton (EF) process is an effective green method to oxidize and degrade organic pollutants in oilfield produced water. By using response surface methodology (RSM), multiple response optimization for maximizing COD removal efficiency while minimizing electrical energy consumption revealed that under optimum operating conditions (initial Fe^{2+} concentration of 0.306 mM, current intensity of 156.6 mA, and residence time of 81.0 minutes) the COD removal efficiency (R%) = 73.33% and EEC= 0.901 kWh/kg COD. It was found that assisting EF process by irradiation with UVA (6 Watt) increase R% about 10% when operating at optimum conditions found in this study.

Model equation that relate COD removal efficiency (R%) with the process variables have been built in a second-order polynomial form for continuous EF (Equation 13) process. The model equation well represent the experimental results, with determination coefficient (R^2) equal to 0.9792.

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