The Taguchi Approach in Studying and Optimizing the Electro-Fenton Oxidation to Reduce Organic Contaminants in Refinery Wastewater Using Novel Electrodes

Rowaida N. Abbas
Chemical Engineering Department
University of Baghdad
Baghdad, Iraq
rowida.abbas1307d@coeng.uobaghdad.edu.iq

Ammar S. Abbas
Chemical Engineering Department
University of Baghdad
Baghdad, Iraq
ammarabbas@coeng.uobaghdad.edu.iq

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Abstract-This study investigated the degradation of organic pollutants using an advanced electrochemical oxidation technique in a batch reactor cell consisting of a graphite anode, modified by electrodeposition of PbO_2, and a graphene-modified carbon fiber cathode. The experiment was designed by the Taguchi design approach with an orthogonal array of L_{18} to study and optimize the degradation of Chemical Oxygen Demand (COD) by the electro-Fenton oxidation process. Four process parameters, Current Density (CD), Temperature, Fe^{2+} concentration, and time were measured at different levels. The impact of each factor was analyzed by analysis of variance (ANOVA). Furthermore, a linear model analysis was applied for the Signal-to-Noise (S/N) ratio and mean values, obtaining the optimal conditions. The most significant parameter of the COD removal efficiency was time, and the least one was temperature.

Keywords-organic pollutants; Taguchi; electro-Fenton; COD removal

I. INTRODUCTION

Wastewater from the oil industry is frequently produced and discharged into the main water bodies, causing severe environmental issues. The amount and characteristics of pollutants in oil refinery wastewater depend on the type of oil being processed, the plant configuration, the operation procedures, and the type of processing units [1]. Pollutants in refinery wastewater often consist of free hydrocarbons, suspended solids, and inorganic matter, with a high concentration of salts, sulfides, ammonia, organic carbon, phenol, benzene, heavy metals, nutrient grease, and chemical additives [2, 3]. Phenol is the most produced contaminant in the refinery industry, which pollutes surface and ground water and causes harmful effects on human health [4-7]. Phenol and its derivatives are highly soluble in water, so they can be present in a wide range of water concentrations, from a few milligrams to high concentrations that reach 7000mg/L, whereas refineries contribute approximately 6-500mg/L. Phenolic compounds persist in water or transform various reactions such as chlorination and methylation into more harmful and toxic materials than phenol itself, such as chlorophenols and cresols [8]. Phenol is classified as one of the most harmful pollutants by the Environmental Protection Agency (EPA) and the European Agency (EA) and is toxic to humans and the environment [9]. Phenol can poison humans when ingested, inhaled, or in contact with the skin. Phenol vapors cause severe irritation to the eyes and mucous. Other severe symptoms caused by exposure to phenol include fatigue, headache, loss of consciousness, nausea, and vomiting. Exposure to phenol for a long time can change blood pressure and damage the liver, kidneys, and the nervous system [10, 11].

Phenol and phenolic compounds can be treated using several methods such as adsorption [12], biological treatments [13], coagulation/flocculation [14], chemical oxidation [15], ion exchange [16], and electrochemical oxidation processes [17]. Adsorption is limited by the adsorbers, as they cannot be easily renewable and need regeneration after a period of use, and the pollutant is not converted to less hazardous materials as it is concentrated and transferred to a vapor or an organic phase [18]. Biological treatment was related to many problems that concern the operating conditions, including providing a suitable environment for microorganisms, the low settle capacity of the sludge due to the low ratio of food to microorganisms, the formation of polymers consisting of lipids, proteins, and carbohydrates that negatively affect the settlement of the sludge, and the poisoning of microorganisms due to toxic compounds that require very long times for acclimatization and start-up [19, 20]. Coagulation requires the junction of non-reusable chemicals such as coagulants, flocculants, and aid chemicals and requires physical and chemical monitoring of the affluent. The volume of sludge generated increases, as well as the need for management and the cost of treated contaminates [14, 20-21]. Chemical oxidation, such as simple oxidation, ozonation, and hypochlorite treatment, requires completed physical and chemical treatments. Still, these

Corresponding author: Ammar S. Abbas

processes require chemicals, thereby, production, transportation, and management of the necessary oxidants for the pre-treatment step. The type of oxidant directly affects the efficiency of the process. The creation of intermediates that may be more toxic than the treated substance has a limited effect on chemical oxygen demand, aromatic compounds, and volatile substances, especially when using hypochlorite, and the formation of sludge [15, 22]. Ion exchange technology needs a high initial cost of the selective resin, maintenance cost, time-consuming regeneration, and large equipment if there is a large volume of wastewater to be treated. Beads are easily fouled by organic matter that requires a physicochemical pre-treatment to remove these pollutants, which is not effective for organic target pollutants [16].

At this time, electrochemical treatment is one of the cleanest and most eco-friendly methods used as an alternative to other treatment processes for environmental protection. Electrochemical technology is a most promising one for the treatment of organic pollutants in refinery wastewater. It includes electrocoagulation-electroflotation, anodic oxidation, electrochemical reduction, and advanced electrochemical oxidation [23-24]. Electrochemical methods do not need to add a lot of chemical reagents because the leading reagent used is the electron, which is considered a clean reagent. These processes can also remove contaminants from even a massive quantity of different phases of polluted streams and require lower temperature and pressure than other non-electrochemical processes. Many electrochemical processes have high selectivity, which prevents the formation of unwanted byproducts because the applied potential can be controlled, which helps attacking specific bonds. The chemical substances added in electrochemical processes are small and safe. Electrochemical methods operate flexibly and their control processes, and equipment, are simple and inexpensive [17, 24-25]. Electrochemical oxidation processes are popular for treating organic pollutants in refinery wastewater and are classified into direct and indirect. Direct oxidation occurs on the surface of the anode electrode, and indirect oxidation uses an oxidizing mediator that forces organic matter to contribute to the oxidation reaction, such as hypochlorite [26], chlorine [27], and hydrogen peroxide (H_2O_2) [28]. The electro-Fenton oxidation process is an indirect advanced oxidation process that uses the electrogenerated hydroxyl radical (•OH) on the surface of electrodes, which has a high oxidation capacity in the oxidation of organic substances [29]. In the electro-Fenton oxidation process, ferrous ion (Fe^{2+}) is added externally, and H_2O_2 is generated electrochemically in-situ on specific electrodes that have appropriate cathodic potential by a two-electron reduction of oxygen [30]:

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (1)$$

The electricity used in electro-Fenton oxidation processes is clean and contamination-free, so these processes are eco-friendly and do not generate secondary pollutants. •OH is generated in the solution by using Fe^{2+} as a catalyst which attacks the organic contaminants [30]:

$$Fe^{2+} + H_2O_2 + H^+ \rightarrow Fe^{3+} + {}^{\bullet}OH + H_2O \quad (2)$$

$$\bullet OH + RH \rightarrow R^{\bullet} + H_2O \quad (3)$$

The Fe^{3+} ion produced in (2) can transform to Fe^{2+} by a one-electron reduction reaction [30]:

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (4)$$

The electrode material is an important factor that affects the performance of the electro-Fenton oxidation because it affects selectivity, efficiency, mechanism, and the products of the oxidation process [2, 31]. So, when choosing the electrode, material-specific properties must consider its electrical conductivity, selectivity, catalytic activity, cost, lifetime, and oxygen overvoltage [31]. The effect of the cathodic reaction on the efficiency of the overall oxidation process is also taken into account [31, 32]. Carbon electrodes are widely used as anodes and cathodes in electro-Fenton processes. They have high surface, high porosity, corrosion resistance, and offer low-pressure drops compared to other materials because they have a large space. Examples of these electrodes are graphite [33-35], reticulated vitreous carbon [36], activated carbon fibers [37], and carbon nanotubes [38]. This study uses the Taguchi approach to investigate the electro-Fenton oxidation process of organic pollutants in refinery wastewater. The process was carried out in a batch reactor cell consisting of a graphite anode modified by electrodeposition of a PbO_2 film on its surface and a carbon-fiber cathode modified by graphene. Different operating conditions were used in the oxidation process, and the effect of these parameters on the COD removal was also studied.

II. EXPERIMENTAL WORK

A. Taguchi Experimental Design

The Taguchi experimental design was used to examine the effect of several factors on the response characteristics with a minimum number of experiments using the orthogonal array technique. The studied parameters were three operating temperatures (15, 25, and 35°C), three current densities (2, 5, and 8mA/cm^2), three concentrations of Fe^{2+} (0.1, 0.2, and 0.4mM), and 6h of electrolysis. Three levels were investigated for each current density, temperature, and Fe^{2+} concentration, and six levels of time, as shown in Table I.

TABLE I. CONTROL PARAMETERS AND LEVELS

Control parameters	Levels
Temperature (°C)	15, 25, 35
Current density (mA/cm^2)	2, 5, 8
Fe^{2+} concentration(mM)	0.1, 0.2, 0.4
Time (h)	1, 2, 3, 4, 5, 6

Table II presents the orthogonal array layout of L_{18} ($3^3 \times 6^1$) used in this study. The notation 3^3 denotes 3 parameters studied on 3 different levels, and 6^1 represents one of the parameters investigated on 6 levels. Taguchi parameter design identifies the best-operating levels that lead to high COD removal. It was investigated which factors had the most and least significant effects on the efficiency of the process. A mathematical relation between the process parameters and the output was employed using regression analysis.

TABLE II. LAYOUT OF L_{18} ($3^3\times6^1$) EXPERIMENTAL DESIGN

No	Time (h)	CD (mA/cm^2)	T, (°C)	Fe^{2+} (mM)
1	1	2	15	0.1
2	1	5	25	0.2
3	1	8	35	0.4
4	2	2	15	0.2
5	2	5	25	0.4
6	2	8	35	0.1
7	3	2	25	0.1
8	3	5	35	0.2
9	3	8	15	0.4
10	4	2	35	0.4
11	4	5	15	0.1
12	4	8	25	0.2
13	5	2	25	0.4
14	5	5	35	0.1
15	5	8	15	0.2
16	6	2	35	0.2
17	6	5	15	0.4
18	6	8	25	0.1

B. Electro-Fenton Oxidation Process

The aqueous solution was prepared to simulate the organic pollutants of the al-Dora refinery wastewater (350ppm COD). The electro-Fenton oxidation process was used to treat the phenolic compound, carried out in an open undivided cell equipped with an anode and cathode having a distance of 3cm between them. The volume of the treated solution was 0.4L. The process used PbO_2 deposit on graphite as an anode and carbon fiber modified by graphene as a cathode. A rotation of 200rpm was achieved in the treated solution using a magnetic stirrer with a heating plate (LABINCO, model L-81). Compressed air was bubbled into the treated solution at 1L/min flow rate using an electromagnetic air pump (ACO-001). The electrolysis process for the treated solution was carried out at three different operating temperatures (15, 25, 35°C ± 1°C). The pH of the treated solution was adjusted to 3 by adding 1M H_2SO_4 to maximize H_2O_2 production. The experiments were carried out by applying different CD 2, 5, and 8mA/cm^2 using a DC power supply (KORAD KA3005D). $FeSO_4{\cdot}7H_2O$ was added externally as a catalyst with different concentrations of 0.1, 0.2, and 0.4mM. Sodium sulfate (Na_2SO_4) was used as a supporting electrolyte with a concentration of 0.05M to improve the conductivity of the solution. Electrolysis was carried out for up to 6h, and samples were collected after each 1h to check the degradation efficiency by COD analysis. COD removal (%) was calculated with:

$$\text{COD removal},\% = \frac{COD_o - COD_t}{COD_o} \times 100 \quad (1)$$

COD_o and COD_t were the initial and the COD concentration at time t in (mg/L) respectively.

III. RESULTS AND DISCUSSION

A. Taguchi Design Analysis for COD Removal

The effect of the most influencing parameters on the removal of organic pollutants was investigated by applying the Taguchi Experimental Design (TED). The process operating parameters of the L_{18} ($3^3\times6^1$) orthogonal array were specified. The phenol and COD removal by the electro-Fenton oxidation process calculated from (5) are shown in Table III.

TABLE III. TED OF L_{18} ($3^3\times6^1$) ORTHOGONAL ARRAY

No.	Time (h)	CD (mA/cm^2)	Temp. (°C)	Fe (mM)	COD removal (%)	S/N
1	1	2	15	0.1	33.52	30.51
2	1	5	25	0.2	43.62	32.79
3	1	8	35	0.4	51.78	34.28
4	2	2	15	0.2	48.32	33.68
5	2	5	25	0.4	57.32	35.17
6	2	8	35	0.1	52.34	34.38
7	3	2	25	0.1	50.87	34.13
8	3	5	35	0.2	67.81	36.63
9	3	8	15	0.4	68.75	36.75
10	4	2	35	0.4	73.64	37.34
11	4	5	15	0.1	60.87	35.69
12	4	8	25	0.2	69.78	36.87
13	5	2	25	0.4	75.41	37.55
14	5	5	35	0.1	73.65	37.34
15	5	8	15	0.2	79.36	38.00
16	6	2	35	0.2	82.32	38.31
17	6	5	15	0.4	82.89	38.37
18	6	8	25	0.1	84.72	38.56

The experimental data were analyzed using MINITAB 19. The Taguchi method uses a signal-to-noise ratio (S/N) to evaluate the optimum removal conditions and determine the response variation of the mean value that causes experimental noise. It uses it to recognize the levels of controlling factors that minimize the response variance from noise factors. The quantitative measure S/N refers to the mean of the output value (desired value) to the standard deviation of the output value (undesired value) [39-40]. S/N can be divided into three categories: Smaller The Better (STB), Nominal The Better (NTB), and Larger The Better (LTB). The type of S/N was chosen depending on the goal of the experimental process and the desired process quality characteristics. The LTB S/N was selected to maximize the phenol and COD removal efficiency. The LTB response can be evaluated by the standard formula for this type of characteristic [41- 44]:

$$\left(\frac{S}{N}\right)_i = -10\log\frac{1}{n}\sum_{i=1}^{n}\frac{1}{Y_{ij}^2} \quad (2)$$

where i is trail number, Y_{ij} represents the response of the measured value for the i-th trial and the j-th run, and n is the number of replications for the experimental combination. In general, the highest S/N value is preferable.

Figure 1 shows a plot of S/N against CD, T, Fe^{2+}, and t. Figure 2 shows the main effects graph for the means and represents the relationship between the operating parameter under study and the response obtained. The results plotted in Figures 1 and 2 are shown in Tables IV and V respectively, showing the effect of each operating parameter on COD removal. The ranks in these tables are based on delta statistics. The delta statistic equals to the difference between each factor's highest and lowest average. The highest delta value was assigned to Rank 1, followed by Rank 2 for the second-highest delta value, and so on [45]. The delta values in Tables IV and V indicate that the time of the experiment had the most significant effect on the COD removal. The second parameter that influenced COD removal was Fe^{2+} concentration, followed by current density, while temperature had the lowest impact on COD removal.

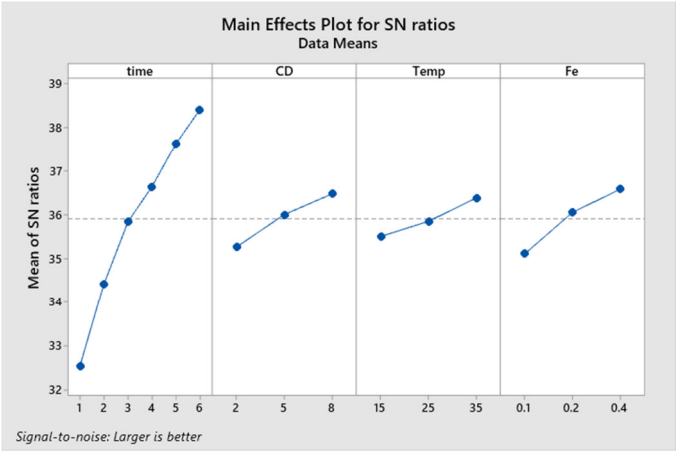

Fig. 1. Main effects plot for S/N.

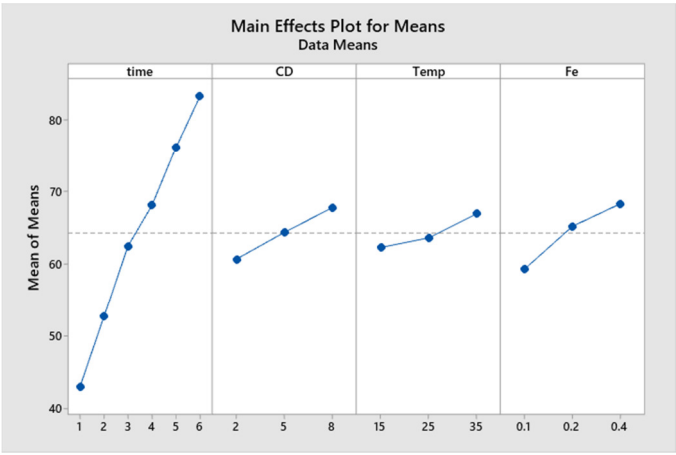

Fig. 2. Main effects plot for means.

TABLE IV. RESPONSE OF S/N FOR COD REMOVAL

Level	Time (h)	CD (mA/cm^2)	Temp. ($^\circ$C)	Fe (mM)
1	32.53	35.25	35.50	35.10
2	34.41	36.00	35.85	36.05
3	35.83	36.47	36.38	36.58
4	36.63	-	-	-
5	37.63	-	-	-
6	38.41	-	-	-
Delta	5.89	1.22	0.88	1.48
Rank	1	3	4	2

TABLE V. RESPONSE OF MEANS FOR COD REMOVAL

Level	Time (h)	CD (mA/cm^2)	Temp. ($^\circ$C)	Fe (mM)
1	42.97	60.68	62.28	59.33
2	52.66	64.36	63.62	65.20
3	62.48	67.79	66.92	68.30
4	68.10	-	-	-
5	76.14	-	-	-
6	83.31	-	-	-
Delta	40.34	7.11	4.64	8.97
Rank	1	3	4	2

B. Analysis of Variance for Means and S/N Ratio

The orthogonal array in the Taguchi experiments contributes to each process parameter in the process response. ANOVA is used to show which parameters have the most significant effect on the process and find the percentage contribution of each parameter to the change in the value of the dependent parameter [46]. The order of significance of the process variables can be easily identified by ANOVA because it is possible to calculate the source of the variation during the process [47]. The ANOVA table involves the degree of freedom (DF), the sequential sum of squares (Seq SS), the adjusted sum of squares (Adj SS), the adjusted mean of squares (Adj MS), F-value, and p-value. These terms are defined in many handbooks as statistical expressions and are studied and analyzed with experimental designs. These factors showed the importance of each parameter on the process performance [48]. A small variation in any process variable with an excellent percent contribution will greatly influence the process performance. A portion of the total variance detected in the experiment for each effecting parameter is the percent contribution. So, the factor of the greater value of percentage contribution contributes more than other factors to the process's final results [49].

Tables VI and VII show that the highest sum of squares (SS) value went to time t, which had the most significant effect on COD removal. These tables also showed that the statistical significance of the mentioned factors is as follows: Fe^{2+}, CD, and T based on p-values. Time is the most significant factor in phenol and COD removal, having 5 degrees of freedom. The least important factor in the processes is T, and the degree of freedom for the CD, T, and Fe^{2+} variables is 2. In general, when F is greater than 4, the change in the parameter studied significantly affects the process performance [40]. It is clear that all factors, time, current density, ferrous ion concentration, and temperature, significantly affect the removal of COD by the electro-Fenton oxidation process since the F values are greater than 4.

TABLE VI. ANOVA FOR S/N OF COD REMOVAL

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Time	5	70.3327	70.332	14.066	104.65	0.000
CD	2	4.5298	4.5298	2.2649	16.85	0.003
Temp	2	2.3735	2.3735	1.1868	8.83	0.016
Fe^{2+}	2	6.7040	6.7040	3.3520	24.94	0.001
Residual error	6	0.8065	0.8065	0.1344	---	---
Total	17	84.746	---	---	---	---

TABLE VII. ANOVA FOR MEANS OF COD

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Time	5	3328.85	3328.85	665.771	84.30	0.000
CD	2	151.65	151.65	75.824	9.60	0.013
Temp	2	68.42	68.42	34.208	4.33	0.069
Fe^{2+}	2	249.09	249.09	124.546	15.77	0.004
Residual error	6	47.38	47.38	7.897	---	---
Total	17	47.38	----	----	----	---

Moreover, the significance of process factors using a p-value indicates that when the p-value is less than 0.05, the process parameter significantly affects the quality characteristic [40]. Therefore, time is the most significant parameter, followed by Fe^{2+} concentration and current density, while the temperature is the least important parameter of COD removal. The adequacy of the model results can be evaluated using

random and normally distributed residual plots, such as histogram of residuals, normal probability, residual fit, and residual order. Figure 3 depicts the residual plots for COD removal. The following should be noted:

- Histograms showed that data have no outliers and are not skewed.
- Normal probability plots showed that data are distributed normally, factors employ the response, and there are no outliers.
- Residuals versus order showed that there are methodical influences in the data because of the time or the order of collection data.
- Residuals versus fitted data showed that there is constant variance and the relationships are nonlinear. In the ideal case, the data points should randomly drop around the zero line.

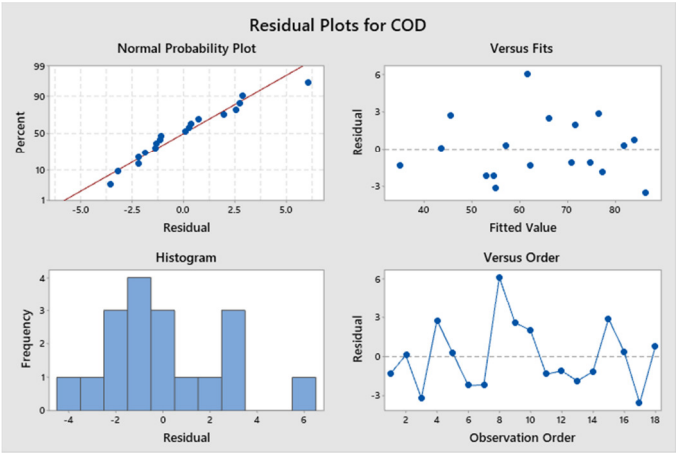

Fig. 3. Residual plots of COD removal.

C. Multiple Regression Model and Optimization

The equation that describes the multiple regression model for COD removal was:

$$COD = 11.08 + 7.936X1 + 1.185X2 + 0.2319X3 - 144.2X4^2 \quad (7)$$

where X1 is the time of experiment (h), X2 is CD (mA/cm^2), X3 is Temperature (^{o}C), and X4 is Fe^{2+} (mM). The squared value of the correlation coefficient (R^2) for the predicted equation was 98.07%. All four factors affected the efficiency of COD removal. The most influential parameter was time, followed by Fe^{2+}concentration and current density, while the temperature was the least effective parameter. Table VIII shows the highest possible COD removal obtained from the optimization of (7). The optimal COD values ranged between 78.34-94.02%. The highest optimal value of COD removal agrees with the findings obtained from the S/N and means analysis results. The selected operating parameters to examine their effects were temperature = 35 ^{o}C, Fe^{2+} = 0.4mM, CD = 8mA/cm^2, and t = 6h. The regression equation (7) was utilized to observe the effect of the process variables on COD removal by the electro-Fenton oxidation process.

TABLE VIII. OPTIMAL COD REMOVAL AND OPERATING PARAMETERS

Time (h)	CD (mA/cm^2)	Temp. (^{o}C)	Fe^{2+} (mM)	COD removal (%)
5	8	15	0.2	78.34
6	8	25	0.1	82.72
6	2	35	0.2	83.80
6	5	15	0.4	85.81
6	8	35	0.4	94.02

D. The Impact of Operating Parameters on COD Removal

The effect of current density on COD removal from simulated wastewater was studied using the electro-Fenton oxidation process at 35^{o}C temperature, 0.4mM Fe^{2+} concentration, supporting electrolyte consisting of 50mM sodium sulfate (Na_2SO_3), solution pH at 3, and applied CD ranging from 2 to 8mA/cm^2.

1) The Effect of Current Density

Figure 4 shows the effect of Current Density (CD) on the removal of COD. The removal of COD was 86.9, 89.28, 91.65, and 94.02% at 2, 4, 6, and 8mA/cm^2 respectively, after 6h of electrolysis. Increasing CD from 2 to 8mA/cm^2 improved the rate of degradation of organic pollutants. Organic degradation increased with CD due to an increase in the amount of H_2O_2 generated electrochemically by electrolysis. When the amount of hydrogen peroxide increases, •OH increases enough to react with the organic pollutants that existed in the treated solution, minimizing COD values [50, 42]. The obtained results agree with other works that treated organic wastewater using the electro-Fenton oxidation process [2, 51-53].

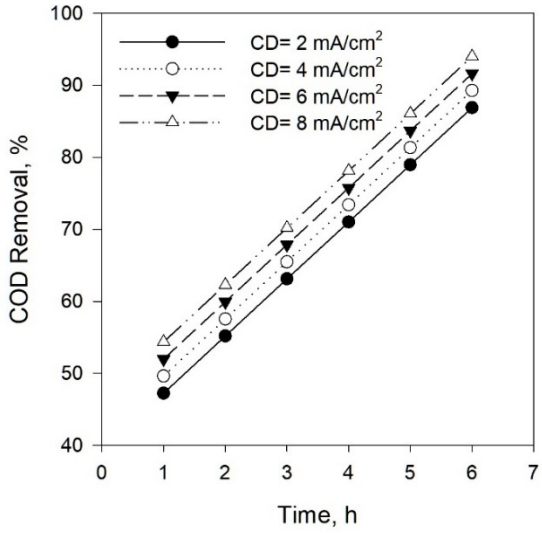

Fig. 4. Effect of applied current density on COD removal from phenolic wastewater, Temperature=35^{o}C, Fe^{2+}=0.4mM, Na_2SO_3=50 mM, pH=3.

2) The Effect of Fe^{2+} Concentration

Fe^{2+} added to the system was externally considered an important parameter that influences the performance of the electro-Fenton oxidation process. The reaction rate increased with increasing concentration of Fe^{2+} up to a limiting value. Increasing the concentration above this value decreased organic

degradation due to scavenging reactions [53, 54]. Figure 5 shows that the removal of COD was 81.03, 85.05, 90.92, and 94.02% at 0.05, 0.1, 0.2 and 0.4mM Fe^{2+} respectively. Increasing Fe^{2+} leads to increased organic degradation because of the increased amount of produced •OH radicals [55]. The results showed that ferrous ion concentration greatly affected the removal, especially on COD, which proved that the electro-Fenton oxidation process was largely influenced by the reaction conditions in the bulk solution between Fe^{2+} and H_2O_2, not by the electrochemical reactions on the electrode surface [56]. The results obtained agree with [56], who reached a COD removal efficiency of 95.9% using 0.7mM $FeSO_4$ concentration and 25mA/cm^2 CD.

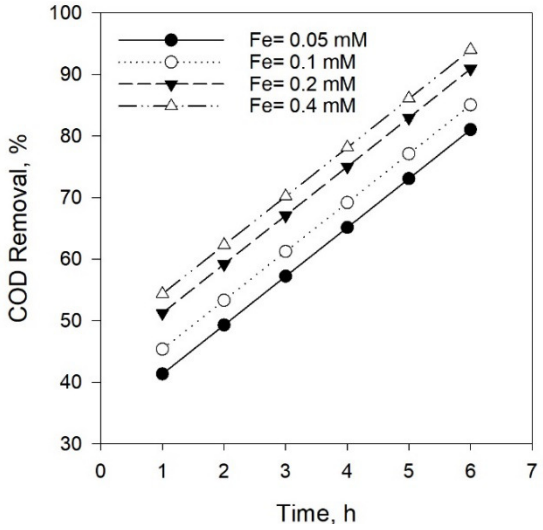

Fig. 5. Effect of Fe^{2+} concentration on COD removal from phenolic wastewater, temperature=35°C, CD=8mA/cm^2, Na_2SO_3=50mM, pH=3.

3) The Effect of Temperature

Sometimes, a temperature increase would lead to an increase in the degradation performance. Choosing an optimum temperature is essential because a further increase in temperature is not needed and depends on the system. Figure 6 presents the effect of temperature on the removal of COD and shows that it was 89.38, 91.70, and 94.02% at 15, 25, and 35°C. Increasing the process temperature leads to a slight increase in the degradation of the organic pollutants by the electro-Fenton oxidation process because raising the temperature promotes the generation of Fe^{2+} [57]. The obtained results agree with the temperature behavior reported in [58, 59], who found that the increase in temperature affects relatively low the organic degradation of the electro-Fenton oxidation process compared to other factors. Furthermore, many studies noticed that the best range of operating temperature is 25-35°C because lower or higher temperatures have a negative impact on removal efficiency. A lower temperature causes a slower initial kinetic process and reduces the degradation performance and reaction rate. Higher temperatures negatively influence the removal of organic contaminants because, at these temperatures, the electrogenerated H_2O_2 is dropped due to the decrease in the dissolved oxygen concentration. Furthermore, at high temperatures, H_2O_2 decays rapidly and decomposes into less reactive water and oxygen [58, 59].

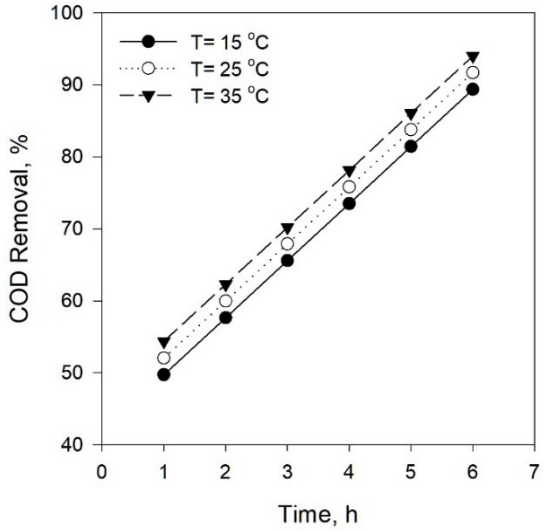

Fig. 6. Effect of operating temperature on COD removal from phenolic wastewater, CD=8mA/cm^2, Fe^{2+}=0.4mM, Na_2SO_3=50 mM, pH=3.

IV. CONCLUSION

A Taguchi experimental design was achieved using the L_{18} orthogonal array technique. The effect of the variable parameters on the removal of COD was studied by choosing four factors: CD, T, Fe^{2+}, and t. The optimum conditions was obtained from a linear model analysis for the S/N ratios and means. The optimum operating conditions for the removal of COD were temperature of 35°C, current density of 8mA/cm^2, Fe^{2+} concentration of 0.4mM, and 6h electrolysis duration. The removal of COD under optimal operating conditions was 94.02%. The time of process variable had the most significant effect on COD reduction efficiency, and the temperature had the lowest impact. The electro-Fenton oxidation process of organic pollutants was influenced mainly by the bulk conditions like the Fe^{2+} concentration, as the electro-Fenton process occurs in bulk between Fe^{2+} and H_2O_2, and not at the electrode surface. Increasing the operating temperature is crucial because it will improve Fe^{2+} generation, but a further increase is not essential.

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