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Phenol Reduction of Petroleum Refinery Wastewater Using Electro-Coagulation/Electro-Oxidation Technique

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ABSTRACT

Water pollution from industrial waste, toxic biological waste, and crude oil refining wastewater: All of these pollutants are released into the environment and pose a major problem today due to their toxic organic and inorganic contaminants. The study found that two electrical methods, electro-coagulation (EC) followed by electro-oxidation (EO) and electro-oxidation alone, effectively reduced organic phenol (C_6H_5OH) levels in oil-refining wastewater from Najaf refineries in Iraq. Both methods achieved remarkable success, though with differences in the factors affecting dissolved phenol removal. The work was done using aluminum and graphite electrodes as the cover of the electric cell and steel (SS) electrodes as the cathode of the cell, made of resistant plastic, in the first method, and graphite electrodes as the anode of the cell with steel (SS) electrodes in the other method. The initial concentration of phenol in the treated water was 50 ppm under the following conditions for both methods: electric current density of 10, 15, and 20 mA/cm^2, sodium chloride (NaCl) with concentration of 0, 1.5, and 3 g/l, and acidity of 3, 7, and 10 pH, with a fixed time of 1 hour for the EC process and 2.5 hours for the EO process for the first method, while in the other method, the time was varied from 2-4 hours. The results showed that the removal rate was directly proportional to the high current density and NaCl concentration under mild acidic conditions for the first method, with the optimum conditions for the removal process being a current density of 20 mA/cm^2, pH 7, and a NaCl concentration of 3 g/L. A removal rate of 95.05% was achieved for the first method under the aforementioned conditions. The results for the removal rate in the second method were obtained under the following conditions: a current density of 15 mA/cm^2, a pH of 3, a NaCl concentration of 3 g/L, and a time period of 3 hours. A removal rate of 96.3% was achieved under the mentioned conditions. Optimization tests were performed using the response surface methodology with Box-Behnken design to identify key operational factors influencing phenol removal from wastewater.

Introduction

The aquatic ecosystem is severely contaminated, causing significant damage to natural waterways and posing a significant threat to Earth's life [1]. Oil refinery wastewater is a significant source of pollution, containing harmful chemicals like oxygen demand, phenol, BTEX, and fats, oils, and grease [2]. Oil refineries produce large amounts of wastewater that contains many organic contaminants, is highly toxic, and doesn't break down easily. This wastewater is an important part of the world's energy supply [3] . Consequently, it is essential to develop efficient, cost-effective techniques for treating petroleum refinery effluent. Various

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methodologies, such as ion exchange, biosorption, membrane filtration, and coagulation-flocculation, have been used in this domain [4-7]. Electrochemical procedures such as electrocoagulation and electrooxidation are more effective due to their lower chemical requirements and greater control [8, 43].

EC coagulants are made by electrolyzing anodic electrodes, typically iron or aluminum, which release ions and form hydrogen gas at the cathode, forming metal ions that encapsulate contaminants [8].

Phenol and its derivatives are among the most common organic contaminants present in effluent from the oil refining sector. These substances are very harmful due to their poor biodegradability, high toxicity, and environmental consequences [9].

Electrochemical treatment technologies like electrooxidation, electrocoagulation, and Electroflotation (EF) have gained popularity for remediating various types of wastewater [10]. The EO procedure is a prevalent main treatment method within electrochemical treatment techniques [11, 44]. A multitude of investigations on the electro-oxidation of phenol have been conducted on a modest scale using various electrode materials [12].

A study on using Fe–Co–O–$_4$ thin film electrodes to electrolyze phenol in an alkaline environment found that 100% breakdown was achieved after 54 hours of electrolysis [13]. Many methods can conduct electrooxidation in wastewater treatment. For instance, when chloride ions are present in the solution, an indirect electrochemical oxidation (EO) process occurs, whereby organic pollutants such as phenol, dyes, glucose, and aniline may be degraded by chlorine and hypochlorite produced anodically during the EO process, via various mechanisms [14, 15].

$$\textit{Anode reaction:}\quad 2Cl^{-}\rightarrow Cl_2+2e^{-} \qquad (1)$$

$$\textit{Cathode reaction:}\quad 2H_2O+2e^{-}\rightarrow 2OH^{-}+H_2 \qquad (2)$$

$$\textit{Bulk solution:}\quad Cl_2 + H_2O\rightarrow HOCl + H^{+} + Cl^{-} \leftrightarrow OCl^{-} + Cl^{-} + 2H^{+} \qquad (3)$$

Pollutants on anodes are broken down by "active oxygen" that physically adheres to them during direct anodic oxidation.

Oxidative oxidation requires neither the addition of chemicals nor oxygen, nor the production of secondary pollutants, and it does not require intricate equipment. In the anodic oxidation process, the anode material is the principal component. The main types of anodes used in anodic oxidation are graphite, glassy carbon, carbon fiber, stainless steel, Ti/Pt–Ir, Ti/RuO_2, and boron-doped conductive diamond (BDD) [16]. Despite its considerable expense, BDD has shown superior overpotentials for oxygen and hydrogen evolution, along with stability and efficiency throughout the electrooxidation process [8].

These features enable their effective use in the oxidation of organic contaminants, electrosynthesis, decolorization of solutions, surfactant degradation, and oxidation of benzoic and carboxylic acids [16-20]. In the presence of chloride ions, graphite is favored for chlorine evolution owing to its higher overpotential for chlorine release than other anodes [8].

The EO technique offers benefits such as easy equipment, organic pollutant breakdown, low maintenance costs, and antimicrobial agent production, but faces challenges such as suspended particle removal and resistive film buildup [21].

Researchers frequently utilize EO due to its simplicity, clean reagents, and flexibility in eliminating various pollutants without the need for additional chemicals [22].

EC for potable water was first implemented in 1946 by Fred E. Stuart [23]. A multitude of research studies were undertaken in the latter part of the 20th century owing to the increasing interest [24].

EC is a cost-effective, eco-friendly method for treating industrial wastewater, offering expedited treatment, reduced sludge, and improved effluent transparency [25].

Electrolysis at the electrodes produces metal ions at the anode and hydrogen gas at the cathode, which are where the coagulants come from. The metal ions generated at the anode neutralize the charged particles in the wastewater, resulting in the formation of neutral particles [26, 27].

The primary reactions in the electrocoagulation process utilizing aluminum electrodes are as follows [28-30]:

anodic reaction:

$$Al \rightarrow Al^{3+} + 3e^- \tag{4}$$

$$2H_20 \rightarrow 4H^+ + O_2 + 4e^- \tag{5}$$

$$x\,Al^{+3} + y\,OH^- \rightarrow Al_x + \left(OH_y^{z+}\right) \tag{6}$$

cathode reaction:

$$2H_2O + 2e^- \rightarrow H_{2\ (g)} + 2OH^- \tag{7}$$

The present research aims to evaluate the practical application of using EC, EO, and EO techniques to treat actual wastewater obtained from Najaf refinery by reducing the phenol value using the mentioned methods and to study the effect of factors (NaCl concentration, CD, pH, and time) on the effectiveness of performing both methods.

Experimental Work for Two Methods: EC-EO and EO

80 liters of effluent from the crude oil refining process were extracted from the Najaf refinery and preserved in a laboratory refrigerator to maintain their characteristics, as shown in Table 1.

Table 1. Qualities of wastewater

Properties	Values
COD (ppm)	1340
TDS (ppm)	3400
Turbidity (NTU)	40.4
POD (ppm)	122.6
Phenol (ppm)	50
Oil (ppm)	30.6
Cl^-(ppm)	2340
PO_4(ppm)	0.15

Chemicals Used

HCl, conc. (36%), Thomas Indian Bakers' brand has been used to create a (1M) solution for the purpose of modifying the initial pH of wastewater and also to prepare 5% (v/v) solution for the purpose of cleaning and reactivating the electrodes.

NaOH, purity ≥ 97.0 %, pellets, Thomas Indian Bakers in brands, has been used to create a (5M) solution for the purpose of modifying the initial pH of wastewater.

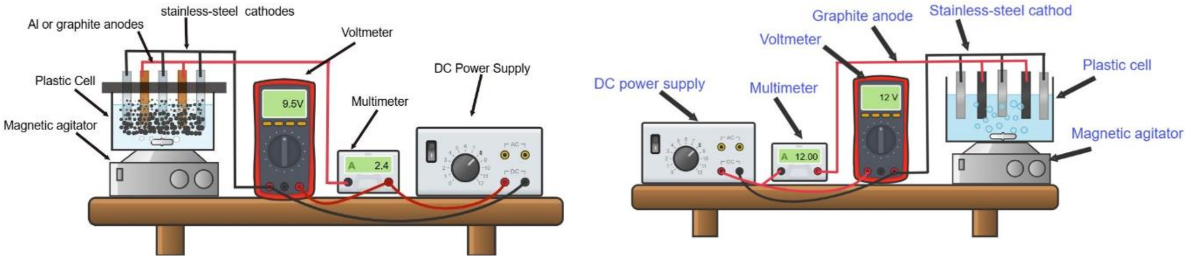
NaCl (purity 99.9%), Barcelona, Spain, has been employed to prepare concentrated solutions with appropriate electrical conductivities.

Distilled water for cleaning, Chemical Engineering Department Laboratories, University of Babylon.

Tools and Equipment

As shown in Fig. 1, we conducted the experiments in a single-electrode electrochemical cell for EC and EO in batch mode at ambient temperature. The cell was made from a plastic container measuring 20 cm in length, 6 cm in width, and 15 cm in height. The cell was then placed on a magnetic stirrer functioning at 200 rpm (DAIHAN LABTECH CO, 0-450 rpm).

The cathode electrodes in the electrochemical process included three sheets of 316-AISI stainless steel, whereas the anode electrodes consisted of two aluminum plates. In the electrochemical oxidation process, the anode electrodes were two graphite plates, and the cathode electrodes were three identical stainless-steel sheets. The electrodes measured uniformly at 18×5×0.3 cm. The submerged dimensions of each anode side were 8×5 cm, yielding an effective surface area of 160 cm^2. Each anode is positioned between two cathodes, separated by 2 cm, and the effective areas of the aluminum and graphite electrodes have been calculated. The anode electrode, potentially composed of aluminum during the process or graphite during electrolysis, is connected to the positive terminal of a DC power supply (MCH-305 D-II, 0–30V, 0–5A dual output), while the stainless-steel cathodes are attached to the negative terminal. An RMS multimeter (UNI-T, UT803) is then connected in series with the anode. We conducted several exploratory tests to assess the range of objects under examination and to gain a comprehensive understanding of the proposed method. The investigations revealed that the EC procedure lasted around 1 hour, but the EO process required 2.5 hours. During the first hour of each run in the EC and EO tests, aluminum electrodes function as anodes in the electrochemical process. Subsequently, we substitute the aluminum electrodes with graphite electrodes and use the electrooxidation procedure for the remainder of the period. Samples were collected and filtered using Whatman filter paper, and the filtrates were thereafter evaluated to establish the therapeutic effectiveness of phenol.



a b

Fig. 1. (a) Diagram of the sequential EC &EO process, and (b) diagram of the sequential EO process

Measurement and Analysis Method

A phenol solution with a known concentration of 100 ppm was prepared. The material was examined using an ultraviolet (UV) light spectrometer to determine the wavelength of phenol. The results were then compared with those of a previous reference solution. Phenol was detected in a wastewater sample from the Najaf refinery. Several measurements of known concentrations were performed to establish calibration curves for both processes, as shown in Figs. 2 & 3, using Eqs. 8 & 9 for phenol measurements. Three tests on the samples showed S values less than 0.05.

For EC-EO:

$$\begin{gathered} Y = 0.01\,x - 0.079 \\ R^2 = 99.35\ \% \end{gathered} \tag{8}$$

For (EO):

$$\begin{gathered} Y = 0.01\,x - 0.079 \\ R^2 = 98.83\ \% \end{gathered} \tag{9}$$

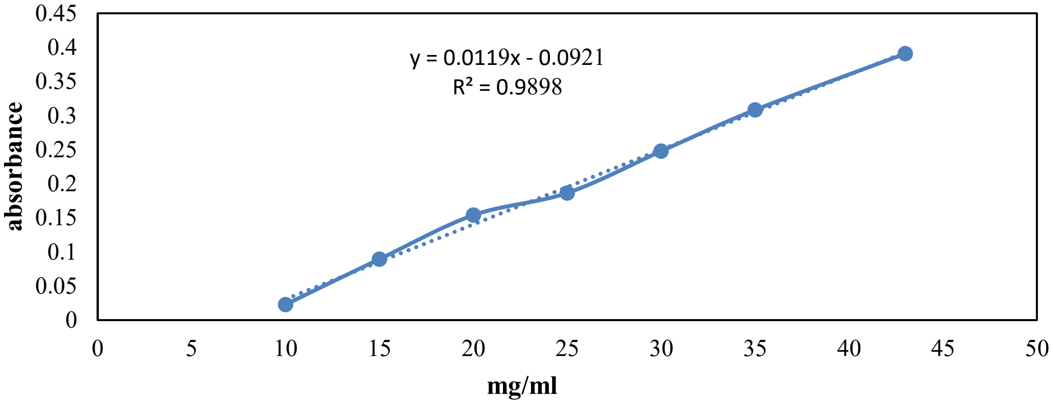


Fig. 2. Calibration curve for absorbance of phenol for EC-EO

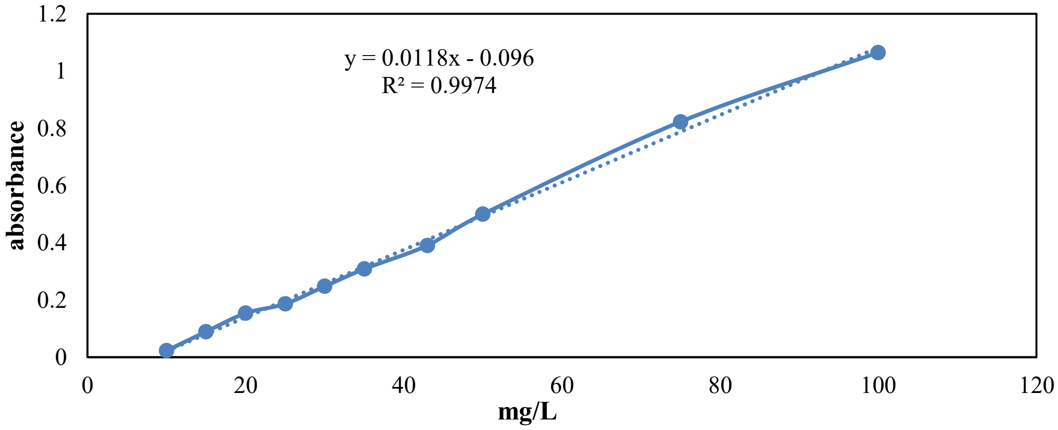


Fig. 3. Calibration curve for the absorbance of phenol for EO

Results and Discussion

For EC-EO Method

This study examines three controllable variables: Sodium chloride (NaCl) content, current density (CD), and pH of the solution, maintaining a consistent duration of 3.5 hours, including 1 hour for the EC process and 2.5 hours for the EO process. Table 2 illustrates three tiers of these characteristics. NaCl was used to enhance wastewater treatment. These criteria were selected for their significance in wastewater treatment.

Table 2. Customized levels for the experimental parameter levels

Parameters	Symbols	Level 1	Level 2	Level 3
CD (mA/cm^2)	B	10	15	20
pH	A	3	7	10
NaCl content (g/l)	C	0	1.5	3

As shown in Table 3, optimization experiments are conducted using response surface methodology (RSM) with a Box-Behnken design (BBD) to identify the statistically significant operational factors affecting the removal of phenol from wastewater [31]. The results were fitting to a second-order polynomial equation as follows [32]:

$$Y = \beta o + \beta_1 A + \beta_2 B + \beta_3 C + \beta_4 AB + \beta_5 AC + \beta_6 BC + \beta_7 A^2 + \beta_8 B^2 + \beta_9 C^2 \quad (10)$$

where Y is the predicted value for phenol concentration, β0 is the y-axis intercept parameter, β1, β2, and β3 are the linear part of the input variable xi, β4, β5, and β6 are the interaction impact of two input variables xi, and β7, β8, and β9 are the quadratic effect.

Table 3. Box-Behnken design experiment with operational parameters and effectiveness of removal for the EC-EO system [31]

Run No.	pH	CD (mA/cm^2)	NaCl (g/l)	Phenol (ppm)	E_{cons} (KWh/m^3)
1	3	10	1.5	59.94	39.2
2	10	10	1.5	35.47	38.08
3	3	20	1.5	73.63	94.08
4	10	20	1.5	85.52	89.6
5	3	15	0	64.16	77.28
6	10	15	0	59.08	58.8
7	3	15	3	76.96	80.4
8	10	15	3	63.14	63
9	7	10	0	47.06	33.6
10	7	20	0	74.93	89.9
11	7	10	3	52.5	36.4
12	7	20	3	95.05	137.76
13	7	15	1.5	67.85	64.68
14	7	15	1.5	67.85	64.68

Multivariate Regression Model for EC-EO

The final equation expressed in coded factors is expressed as follows:

$$\text{Phenol} = 68.296875 - 2.813125\,A + 15.63375\,B + 5.575625\,C + 7.95375\,AB - 1.911875\,AC + 3.67\,BC - 2.205\,A^2 - 1.33\,B^2 + 0.865\,C^2 \tag{11}$$

The equation expressed in coded factors may facilitate predictions on the response at specified values of each element, as seen in Fig. 4. Typically, elevated component values are designated +1, while diminished levels are designated -1. The encoded equation is important in determining the relative influence of the variables by analyzing the factor coefficients.

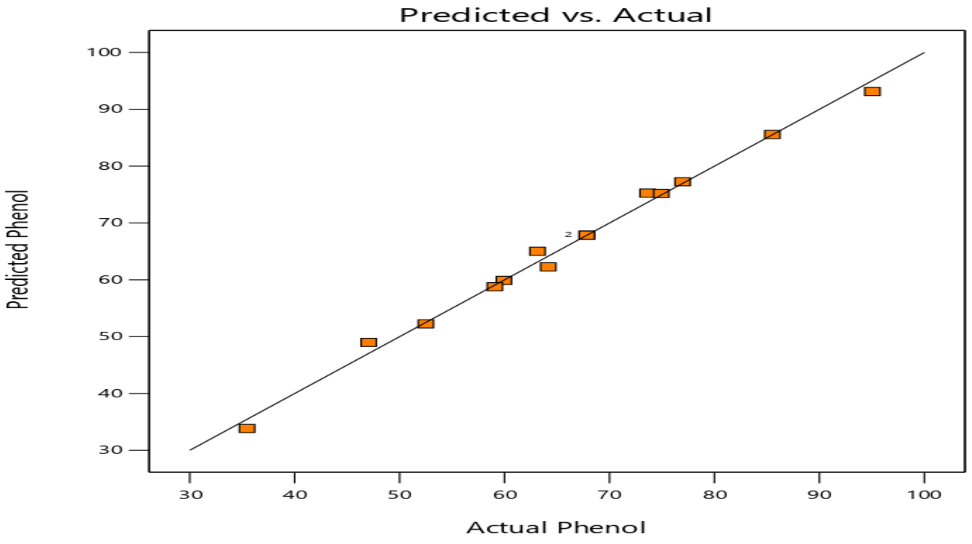


Fig. 4. Phenol behavior against Phenol actual

Analysis of ANOVA Optimization

We perform an ANOVA to evaluate the impact of each controllable factor on the process response, specifically the efficiency of phenol removal. The ANOVA evaluation facilitates a deeper comprehension of the acceptability of the results and the extent to which the experiments are conducted under controlled settings [33].

The data indicate that a substantial proportion of writers hail from the United States, India, and China. Additionally, several writers are affiliated with esteemed higher education institutions, such as the University of São Paulo, the University Estadual Paulista "Júlio de Mesquita Filho," and Tehran University of Medical Sciences. This report offers a summary of the volume and allocation of contributors to the progress of ANOVA research [34].

As seen in Table 4, the model F-value of 67.43 indicates that the model is statistically significant [38]. The probability of an F-value of this magnitude arising from random noise is about 0.05%.

P-values below 0.0500 indicate that model terms are statistically significant. A, B, C, AB, and BC are pertinent model words in this instance. Values greater than 0.1000 indicate that the model terms are not significant. Model reduction may improve your model if several inconsequential terms are present, except those necessary for hierarchical support.

Table 4. ANOVA Analysis data [38]

Source	Sum of squares	df	Mean of square	F-value	P-value	
Model	3039.01	9	337.67	67.43	0.0005	Significant
A-PH	71.52	1	71.52	14.28	0.0195	
B-CD	1896.06	1	1896.06	378.61	<0.0001	
C-NaCl	241.16	1	241.16	48.16	0.0023	
AB	330.51	1	330.51	66.00	0.0012	
AC	19.10	1	19.10	3.81	0.1226	
BC	53.88	1	53.88	10.76	0.0305	
A^2	26.54	1	26.54	5.30	0.0827	
B^2	5.66	1	5.66	1.13	3476	
C^2	2.39	1	2.39	0.4781	0.5273	
Residual	20.03	4	5.01			
Lack of Fit	20.03	3	6.68			
Pure Error	0.0000	1	0.0000			
Cor Total	3059.04	13				

For EO Method

Four adjustable factors—sodium chloride (NaCl) concentration (g/l), current density (CD) (mA/cm²), solution pH, and time—are investigated in this paper. The four tiers of these qualities are shown in Table 5. Wastewater treatment was improved by NaCl. These criteria were chosen for their relevance in affecting wastewater treatment.

Table 5. Levels that are specific to the experimental parameter levels

Parameters	**Symbols**	**Level 1**	**Level 2**	**Level 3**
CD (mA/cm^2)	A	10	15	20
pH	B	3	7	10
Time (h)	C	2	3	4
NaCl content (g/l)	D	0	1.5	3

According to Table 6, six optimization tests are conducted using response surface methodology (RSM) with Box-Behnken design (BBD) to find the important operational factors that influence phenol removal from wastewater [31].

Table 6. Box-Behnken design experiment with operational parameters and effectiveness of removal for the EO system [31]

Run no.	**pH**	**CD (mA/cm^2)**	**NaCl (g/l)**	**Time (hour)**	**Phenol (ppm)**	**E$_{cons}$ (KWh/m^3)**
1	3	10	1.5	3	76.48	26.4
2	3	20	1.5	3	83.6	78.72
3	10	10	1.5	3	52.22	31.68
4	10	20	1.5	3	44.8	97.92
5	7	15	0	2	39.8	45.2
6	7	15	0	4	50.76	90.24
7	7	15	3	2	82.52	33.6
8	7	15	3	4	78.9	68.16
9	7	10	0	3	52.53	37.44
10	7	20	0	3	36.45	96.96
11	7	10	3	3	76.44	29.28
12	7	20	3	3	83.62	78.72
13	3	15	1.5	2	71.55	33.6
14	10	15	1.5	2	56.34	33.6
15	3	15	1.5	4	90.49	70.08
16	10	15	1.5	4	55.34	79.68
17	7	10	1.5	2	77.23	20.48
18	7	20	1.5	2	60.34	58.24
19	7	10	1.5	4	70.32	41.6
20	7	20	1.5	4	75.92	112.64
21	3	15	0	3	47.44	59.04
22	10	15	0	3	39.24	47.52
23	3	15	3	3	96.3	46.08
24	10	10	3	3	56.73	41.76

Multivariate Regression Analysis

The conclusive equation in coded variables is represented as follows:

$$\text{Phenol} = 57.45 - 1.34*13.43*B + 3.23*C + 18.08*D3.82*AB + 5.62*AC + 5.82*AD-5.15*BC - 7.63*BD3.65*CD + 6.15*A^2 + 1.84*B^2 + 8.60*C^2 + 0.0000*D^2 \tag{12}$$

The equation in terms of coded factors can be used to predict the response for given levels of each factor, as seen in Fig. 5. By default, the high levels of the factors are coded as +1, and the low levels are coded as -1. The coded equation is useful for identifying the relative impact of the factors by comparing the factor coefficients.

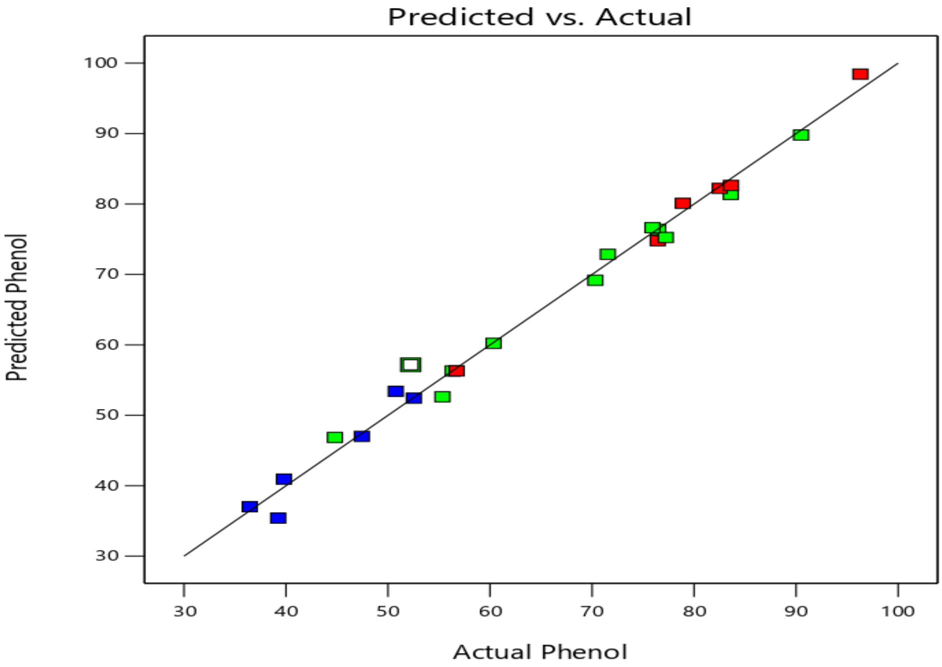


Fig. 5. Phenol behavior against the actual phenol

Optimization Analysis of ANOVA

To evaluate the acceptability of the results and the quality of the tests performed under controlled conditions, we use ANOVA to examine how each controllable factor affects the process response, specifically phenol removal success [34].

The data indicate that a substantial proportion of writers hail from the United States, India, and China. Furthermore, several writers are associated with esteemed higher education institutions, like the University of São Paulo, São Paulo State University "Júlio de Mesquita Filho," and Tehran University of Medical Sciences. This report offers a comprehensive analysis of the quantity and distribution of contributions to the progression of ANOVA research [35].

As seen in Table 7, the model's F-value of 64.73 indicates statistical significance. The likelihood of an F-value of this size resulting from random noise is around 0.01%.

P-values less than 0.0500 indicate that model terms are statistically significant. In this case, B, C, D, AB, AC, AD, BC, BD, CD, A^2, and C^2 are relevant model terms. Values beyond 0.1000 indicate that the model terms are insignificant. Model reduction may improve your model if several insignificant terms are present, provided they are not essential for hierarchical support.

Table 7. ANOVA analysis data

Source	Sum of squares	df	Mean of square	F-value	P-value	
Model	6951.58	13	534.74	64.73	<0.0001	Significant
A-CD	21.11	1	21.11	2.56	0.1410	
B-PH	2165.18	1	2165.18	262.11	<0.0001	
C-Time	128.79	1	128.79	15.59	0.0027	
D-Salt	3821.79	1	3821.79	462.66	<0.0001	
AB	59..05	1	59.05	7.15	0.0233	
AC	126.45	1	126.45	15.31	0.0029	
AD	135.26	1	135.26	16.37	0.0023	
BC	107.59	1	107.59	13.03	0.0048	
BD	235.96	1	235.96	28.56	0.0003	
CD	53.14	1	53.14	6.43	0.0295	
A^2	151.41	1	151.41	18.33	0.0016	
B^2	12.96	1	12.96	1.57	0.2389	
C^2	295.67	1	295.67	35.79	0.0001	
D^2	0.0000	0				
Residual	82.60	8.26				
Cor Total	7034.19	23				

Effect of Operational Parameters on Phenol Removal

Impact of PH

For EC-EO

The findings indicated that the optimal, most expensive phenol elimination rate occurs at pH 7, as shown in Figs. 6 & 7. Experiments conducted under these circumstances demonstrated the generation of sodium hydroxide radicals and active chlorine species, including Cl_2, HOCl, and ClO^-, as detailed in the preceding paper [35]. In this medium, $Al(OH)_3$ is not very soluble and settles as thick "flocs," which are powerful and big. The surfaces of these precipitates carry positive charges that neutralize the phenol molecules' charge. At pH 7, the phenol molecules are not bonded to anything, so they may be more readily absorbed.

After coagulation, some of the phenol remains dissolved, but less. This makes it easier for free radicals (•OH) and oxidizing species generated at the anode to fully oxidize the remaining phenol. A neutral medium (pH ≈ 7) strikes a beneficial compromise between generating free radicals and keeping them stable, as they don't break down as quickly as they do in a very alkaline medium, as demonstrated in the previous study [45]. The findings indicated that the phenol removal process in this approach (EC-EO) is more effective in acidic or neutral environments, depending on operational factors.

For EO

The results demonstrated that pH 3 yielded the greatest and most cost-effective phenol removal rate, as seen in Figs. 8 & 9. Experiments conducted under these conditions demonstrated the formation of sodium hydroxide radicals and active chlorine species, such as Cl_2, HOCl, and ClO, which are potent oxidants that efficiently decompose phenol while simultaneously increasing the concentration of H+, thereby facilitating oxidation reactions at the electrode surface. However, several studies have reported that the optimal pH for the highest clearance rate is 7, as noted in the preceding publication [35, 45].

We conclude from both methods that the first (EC-EO) requires a neutral, acidic medium, while the second (EO) requires an acidic medium.

Impact of CD

For EC-EO

Electric current is a crucial factor in both EC and EO processes for phenol removal from wastewater, and is influenced by other parameters such as temperature, pollutant concentration, and electrode type. Experiments were performed using several current densities (10, 15, and 20 mA/cm²), revealing that optimal phenol elimination occurred at a current density of 20 mA/cm². This finding aligns with prior investigations [15, 36].

In the EO process, elevating the current results in a heightened production of HCLO, while in the EC process, raising the current leads to an augmentation in mass due to the substantial release of Al ions by anodic dissolution, as dictated by Faraday's law in Eq. 13 [38].

$$C_{Al} = \frac{M.I.t}{n.F.V} \tag{13}$$

where C_{Al} = theoretical concentration of Al (g/l)
M = the molecular weight of Al (g/mol)
I = the applied current (A)
n = number of electrons (n = 3 for Al)
F = Faraday constant (96500 C/mol)
V = volume of liquid in the reactor (l)

It is clear that high currents accelerate electrode (Al) decomposition, increasing consumption costs. To reduce these costs, we increase the process time at lower currents, as shown in Figs. 7, 10 & 11.

For EO

While experiments conducted with this method at different current levels (10, 15, and 20 mA/cm²) showed the best phenol removal at 15 mA/cm². Other factors, such as time and sodium chloride concentration, depend on the current. This result is consistent with previous studies [39].

In electrolysis, increasing the current increases the production of hypochlorous acid (HOCl) or sodium hypochlorite (NaClO) when chloride ions (Cl^-) are present in the solution, thereby speeding up the reactions at the electrodes (anode and cathode) [40].

The clearance rate is superior at a high current of 20 mA/cm² for 4 hours. The increased current produces a large amount of reactive free radicals, enhancing the speed and efficiency of removal. However, the increased current also depletes some oxidizing agents through side reactions or the formation of unwanted byproducts. Therefore, due to these side reactions, a longer duration is required to achieve sufficient phenol removal, as shown in Fig. 8.

Also, a low current of 10 mA/cm² and a short duration of 2 hours were shown to yield optimal removal. The low current produces sufficient free radicals for effective removal. This is attributed to the low current's inability to induce loss of oxidizing agents, due to the absence of side reactions common at high currents. Consequently, the response time of the removal process is reduced, as shown in Figs. 12 & 13. We find that effective removal efficiencies can be achieved in both scenarios; however, the difference lies in the consumption of the oxidizing agents and their reaction with phenol.

To achieve maximum efficiency, an appropriate balance between current and time must be found to avoid unwanted side reactions, as described previously [46, 47].

It was found that the current density in both methods is directly proportional to the rate of phenol removal, depending on other influencing factors.

Impact of NaCl

For EC-EO

The addition of sodium chloride significantly influences the phenol removal process. Experiments indicate that the addition of NaCl positively influences both processes, by enhancing conductivity and reducing voltage. During this process, several forms of chlorine, including hypochlorous acid, are generated at the anode electrodes. These facilitate the decomposition of organic molecules by a mechanism known as indirect oxidation, as shown in Eqs. 11 & 12 [15].

$$2Cl^- + 2e^- \rightarrow Cl_2 \quad (14)$$

$$Cl_2 + H_2O \rightarrow HCl + HClO \quad (15)$$

Figs. 11 & 14 show the effect of adding NaCl on the phenol removal process. Laboratory experiments were conducted without adding salt and with adding salt in quantities of 0, 0.15, and 3 g/l. It was found that salt has a positive effect on the removal process at all current densities, as reported in previous studies [35, 36].

For EO

The incorporation of sodium chloride substantially influences the phenol elimination process. Experiments demonstrate that incorporating sodium chloride favorably influences the electrooxidation process by improving conductivity and lowering the potential. The anode electrodes produce many chlorine species, including hypochlorous acid, throughout this process. These forms promote the breakdown of organic molecules by a process termed indirect oxidation.

Figs. 13 & 15 illustrate the impact of NaCl supplementation on the process of phenol removal. We performed laboratory experiments with and without salt, at concentrations of 0, 1.5, and 3 g/L. Research demonstrates that salt enhances the removal process at all current densities, as supported by previous studies [41].

We conclude that, in both methods, sodium chloride is directly proportional to the current density and the removal rate.

Impact of Time

For EC-EO

The duration was established at three and a half hours, with the electrocoagulation process requiring one hour and the electrooxidation procedure necessitating two and a half hours for all tests.

For EO

The results showed that the duration of the electrooxidation process greatly affects the rate of phenol removal, depending on other factors such as current density and sodium chloride concentration. The optimal removal rate was achieved in three hours with a current density of 15 mA/cm², a sodium chloride concentration of 3 g, and a pH of 3. The values fluctuate throughout the procedure. The study results indicate that time significantly influences CD density and the efficacy of phenol removal. This is evident in Figs. 16 & 17.

The likelihood that phenol decomposes into less hazardous chemicals, such as carbon dioxide and water, increases over time. Excessive processing time may result in heightened energy consumption without substantial gains in efficiency and the generation of unwanted oxidation byproducts, as shown in a previous study [42].

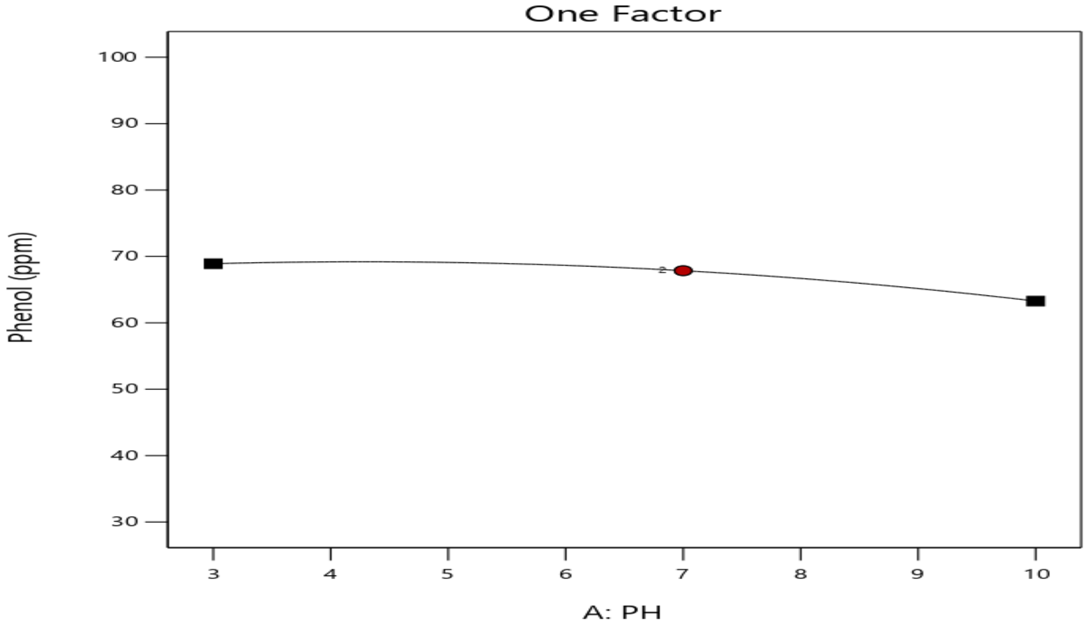


Fig. 6. Effect of pH on phenol removal efficiencies at pH = 7, CD = 15 mA/cm^2, and NaCl = 1.5 g/l

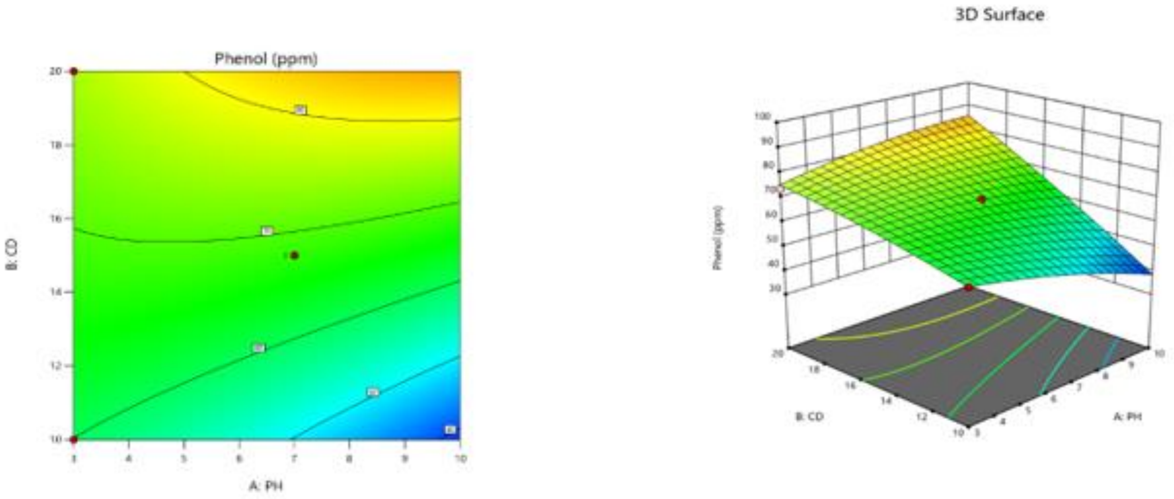


Fig. 7. Contour and 3D surface for phenol % vs pH and CD

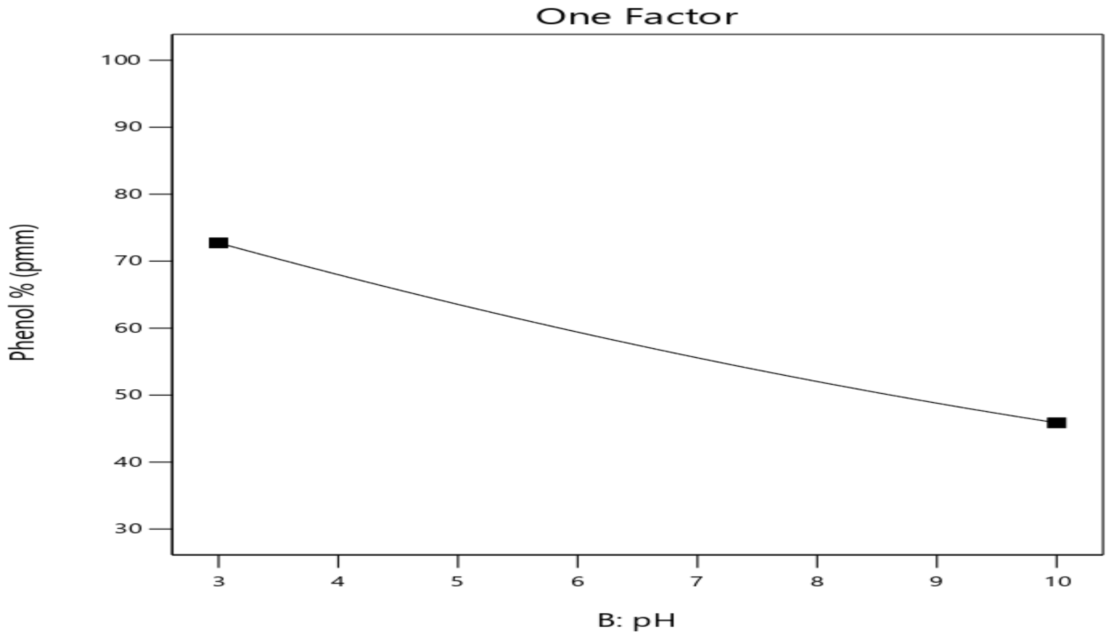


Fig. 8. Effect of pH on phenol removal efficiencies at t = 3 h, CD = 15 mA/cm^2, and NaCl = 1.5 g/l

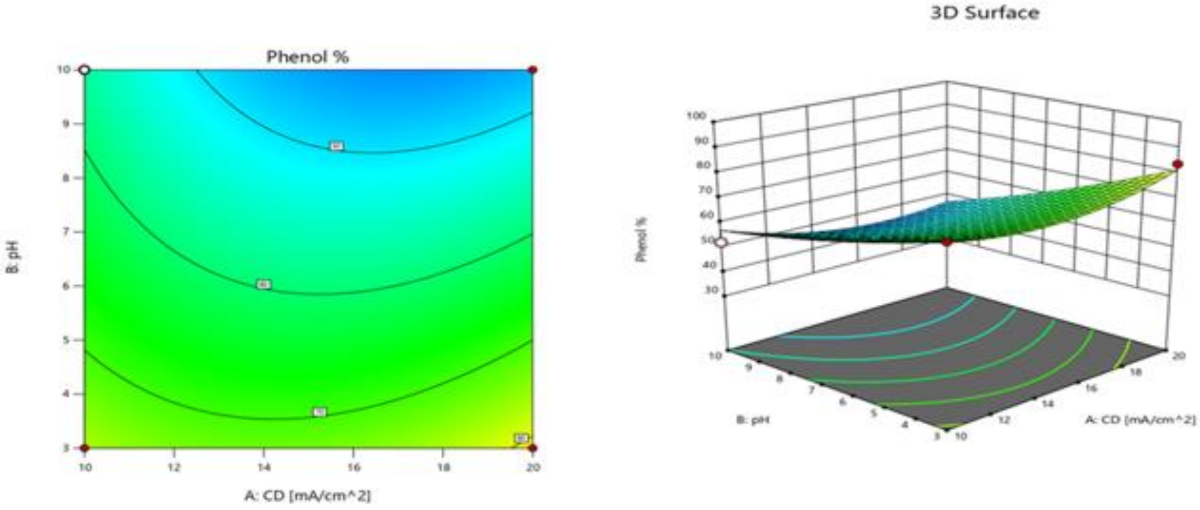


Fig. 9. Contour and 3D surface for pH and CD vs Phenol %

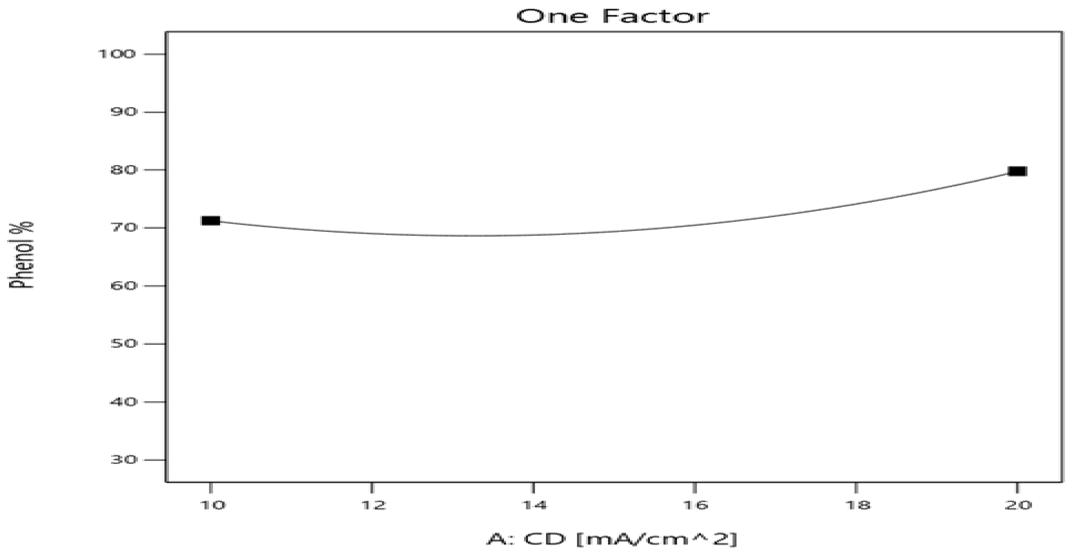


Fig. 10. Effect of CD on phenol removal efficiencies at CD = 20 mA/cm^2, t = 4h, NaCl = 1.5 g/l, and pH = 6.5

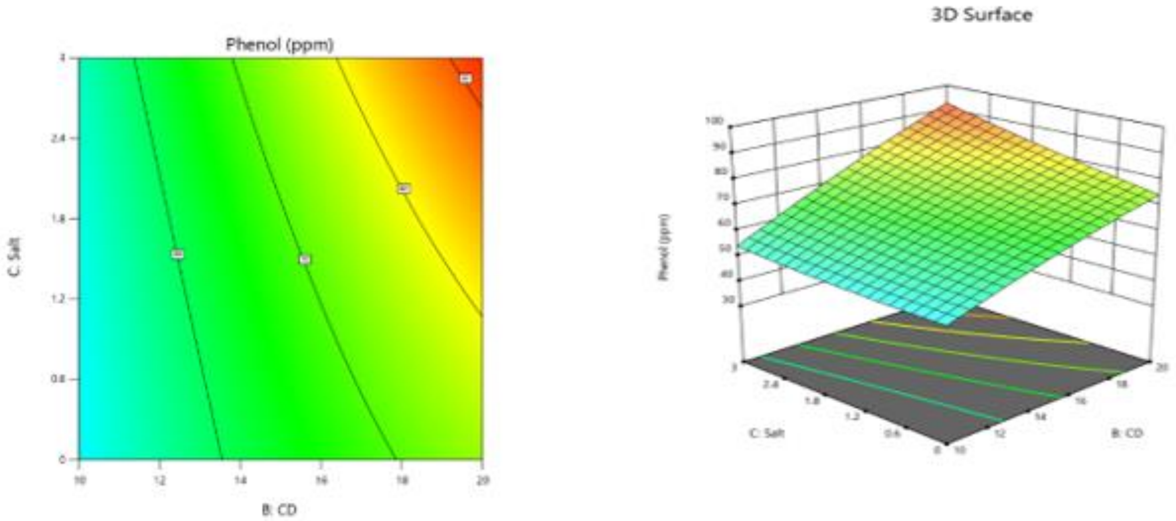


Fig. 11. Contour and 3D surface for phenol % vs CD and NaCl

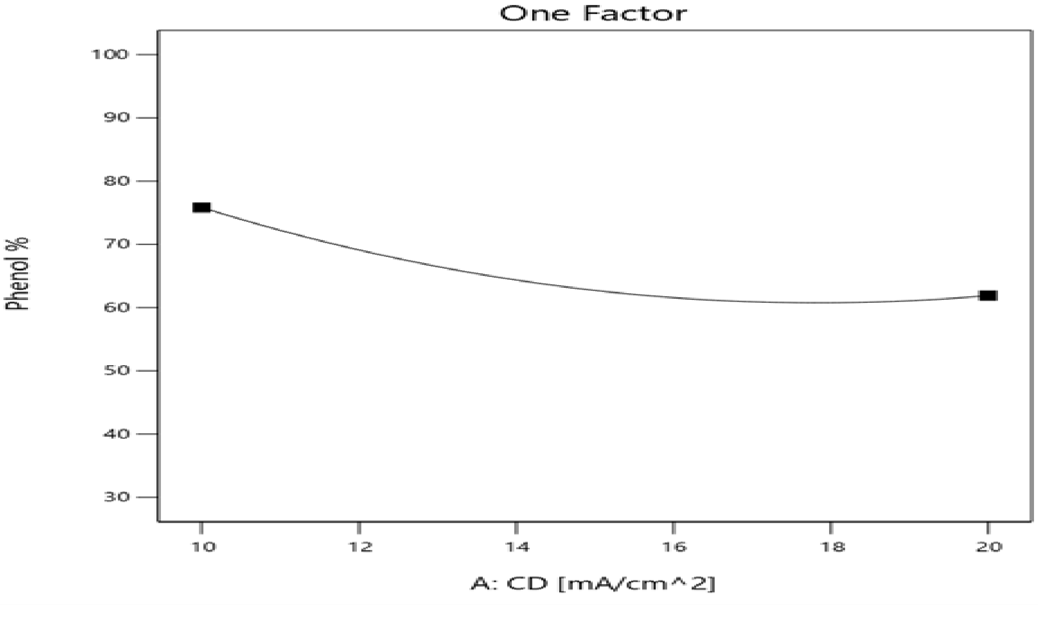


Fig. 12. Effect of CD on phenol removal efficiencies at CD = 10 mA/cm^2, t = 2 h, NaCl = 1.5 g/l, and pH = 6.5

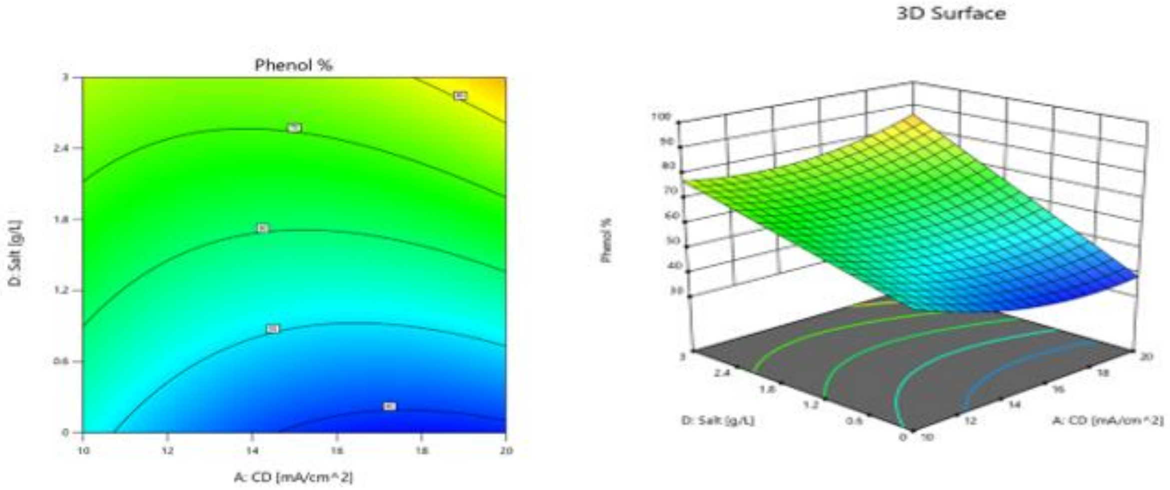


Fig. 13. Contour and 3D surface for salt and CD vs phenol %

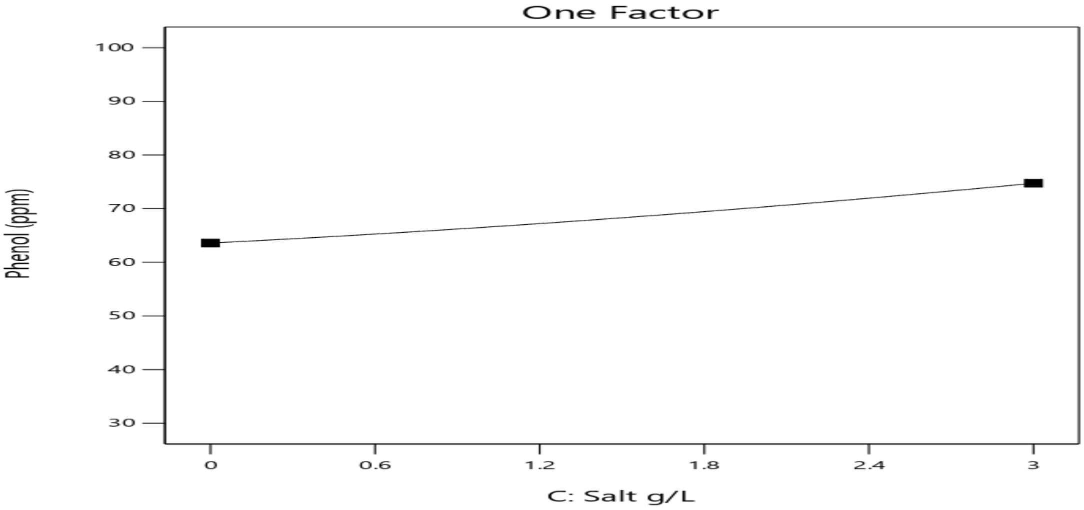


Fig. 14. Effect of NaCl on phenol removal efficiencies at pH = 7, NaCl = 3 g/l, and CD = 15 mA/cm^2

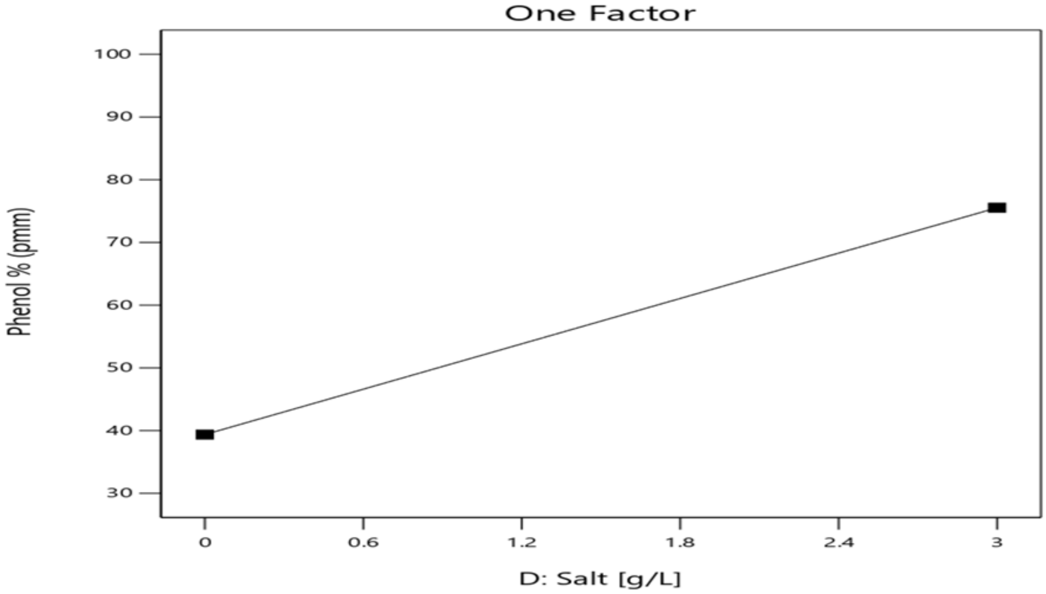


Fig. 15. Effect of NaCl on phenol removal efficiencies at pH = 7, CD = 15 mA/cm^2, and t = 3 h

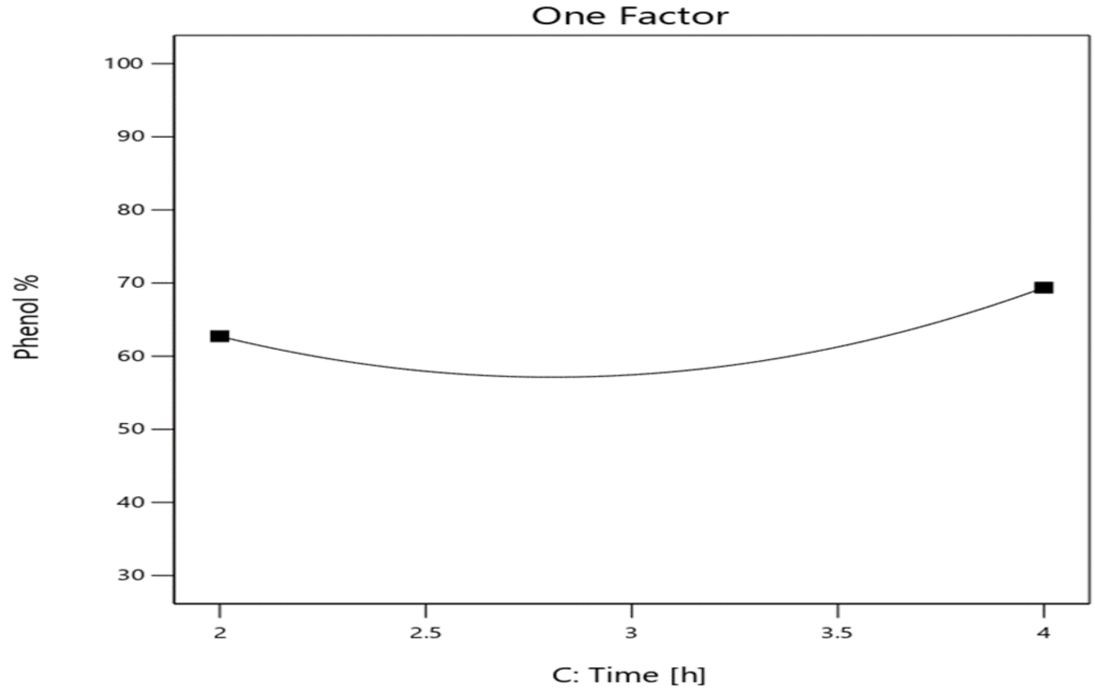


Fig. 16. Effect of time on phenol removal efficiencies at pH = 7, CD = 15 mA/cm^2, and NaCl = 1.5 g/l

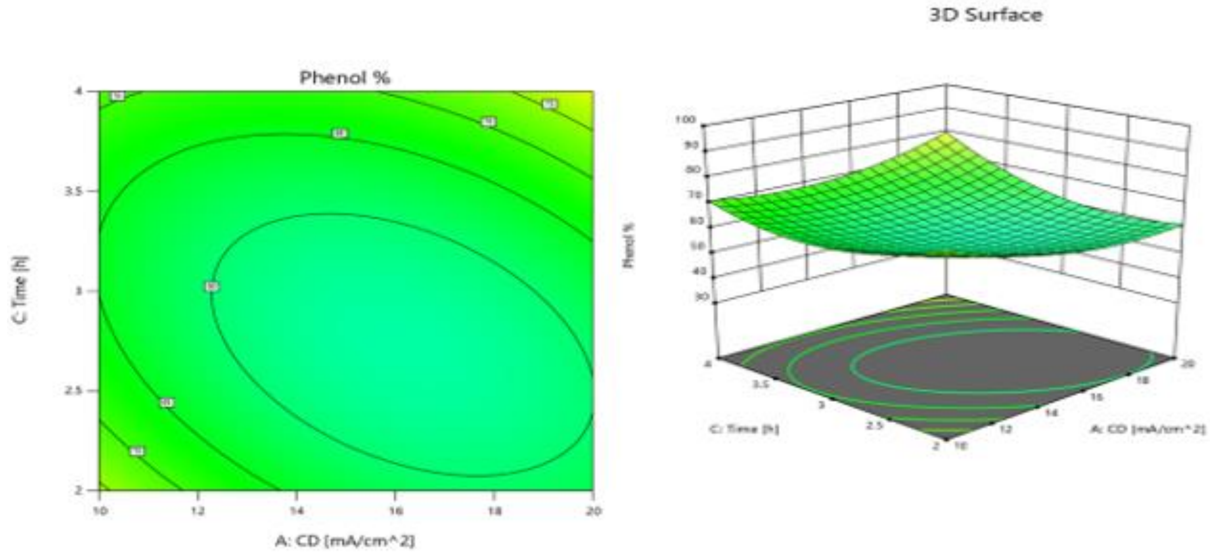


Fig. 17. Contour and 3D surface for time and CD vs phenol %

The results showed that the removal rate was directly proportional to the high current density and sodium chloride concentration under mild acidic conditions for the first method. The optimum conditions for the removal process were CD = 20 mA/cm^2, pH = 7, and NaCl = 3 g/l. A removal rate of 95.05% was achieved under the mentioned conditions for the first method. The results for the second method were: CD = 15 mA/cm2, pH = 3, NaCl = 3 g/L, and t = 3 h. A removal rate of 96.3% was achieved under the mentioned conditions. Compared to previous studies, as shown in Table 8.

Table 8. Comparison table with previous studies comparing the results of the manuscript

Wastewater source	The technology used	Anode/Cathode	Initial phenol (mg/l)	Operating condition	Removal efficiency	Best condition	Ref.
Wastewater from an olive millin Extremadura, Spain.	EC	Zn/S.S	740	pH = 3-9 CD = 120-500 A/m^2 NaCl = 0-1.5 g/l space = 0.5-2 cm	84.2%	pH = 3.2 CD = 250 A/m^2 NaCl = 1.5 g/l t = 180 min space = 1 cm	Fajardo et al. (2015)
Diwaniyah refinery	EC-EO	Al/S.S Graphite/S.t	0.17	CD = 5-15 mA/cm^2 pH = 7 NaCl = 2 g/l	96.2%	CD = 12 mA/cm^2 NaCl = 2 g/l t=60min	Abbar and Alkurdi (2021)
Synthetic	EC	Al/S.S	120	CD = 4, 6, and 8mA/cm^2 t = 1-120 min	60%	CD = 6 mA/cm^2 t = 120 min	Kadhim and Abdulredha (2021)
Synthetic	EO	BDD S.S	100	CD = 50-200 A/m^2 t = 30-240 min pH = 3.6-9.6 NaCl = 2-6 g/l	92%	t = 50 min pH = 7.6 CD = 200 A/m^2 NaCl = 4 g/l	Akhtar and Kobya (2025)

Energy Consumption

The wastewater treatment procedure needs electrical energy. Consequently, E_{cons} must be articulated and analyzed to circumvent costly endeavors.

The E_{cons} value was calculated using Eq. 16.

All calculations were conducted at a capacity of one liter. The EO process duration was established at 2-4 hours.

$$E_{cons} = \frac{E \times I \times t}{1000V} \tag{16}$$

E_{cons}: energy consumed
E: cell potential
I: the current
t: time
v: volume

Conclusion

As mentioned, industrial water, such as oil-refining water, contains many toxic organic and inorganic substances that are harmful to the environment and to life. One of the most important of these toxic and carcinogenic organic substances is phenol dissolved in oil-refining water taken from the Najaf Refinery. The amount of this substance was reduced using effective electrical methods, as in the first method (EC, EO) and the other (EO), and was effectively removed using operating conditions such as CD, pH, and NaCl. Both processes took 3.5 hours for the first method, while in the second method, an additional variable was added, which is the time factor 2-4 hours. Design-Expert software was used to calculate the time for each of the 13 laboratory experiments for the first method and 24 experiments for the other method. Experiments in the first method showed that high CD with medium to strong acidity removed phenol more effectively when the amount of NaCl increased (pH = 7, CD = 20 mA/cm^2, NaCl = 3 g/l), while the optimum removal conditions in the second method were CD = 15 mA/cm^2, pH = 3, NaCl = 3 g/l, and t = 3 h. The results of the analysis of variance (ANOVA) showed that the P and F values indicate that all process variables affect phenol removal, either directly or in relation to one another. We hope that this study has at least reduced the phenomenon of environmental pollution.

Nomenclature

Symbol	**Description**
Y	Absorbance of phenol
X	Concentration of phenol
A	CD (Current Density)
B	pH
C	NaCl concentration
D	Time
COD (ppm)	Chemical oxygen demand
TDS (ppm)	Total dissolved solids
BOD (ppm)	Biological oxygen demand
CD	Current density

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