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Elimination of phenol by sonoelctrochemical process utilizing graphite, stainless steel, and titanium anodes: optimization by taguchi approach

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Abstract

Phenol is one of the worst-damaging organic pollutants, and it produces a variety of very poisonous organic intermediates, thus it is important to find efficient ways to eliminate it. One of the promising techniques is sonoelectrochemical processing. However, the type of electrodes, removal efficiency, and process cost are the biggest challenges. The main goal of the present study is to investigate the removal of phenol by a sonoelectrochemical process with different anodes, such as graphite, stainless steel, and titanium. The best anode performance was optimized by using the Taguchi approach with an L_{16} orthogonal array. the degradation of phenol sonoelectrochemically was investigated with three process parameters: current density (CD) (25, 50, 75, and 100 mA/cm^2), time (1, 2, 3, 4 h), and phenol concentration (100, and 200 mg/l). Signal-to-noise (S/N) ratio and analysis of variance (ANOVA) were utilized to examine the impact of each factor. The optimal conditions for phenol removal were 100 mA/cm^2, 100 mg/l of phenol, and 4 hours of electrolysis. Under optimal operating conditions, the phenol removal efficiency was 80.99%. The CD was the most influential factor on phenol elimination effectiveness, while the phenol concentration had the least impact.

Keywords: organic pollutant; indirect oxidation; wastewater; ultrasonic; removal.

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1- Introduction

Water is an essential component of life, and it is required for all living species. On the other side, the world is experiencing a catastrophic lack of drinking water so it's the highest essential resource that must be protected and reused whenever possible. Water pollution is today a big global concern due to a range of factors such as insufficient treatment of sewage, industrial wastes, and challenges with marine dumping, radioactive waste material, and numerous agricultural operations [1, 2].

The petroleum refinery industry converts crude oil into over 2500 refined products, such as gasoline, kerosene, aviation turbine fuels, diesel fuels, fuel oil, petroleum waxes, bitumen, and lubrication oils. A significant amount of water is needed to achieve these products; roughly, 80–90% of the water that is supplied ends up as wastewater [3].

Phenol is one of the most dangerous environmental toxins that exist in wastewater rejected by petroleum refineries. The Agency for Environmental Protection (EPA) lists phenol as one of 129 chemical substances that are considered significant pollutants and recommends reducing the concentration of phenol in wastewater by no more than 1 mg/l [4 - 6]. Depending on World Health Organization (WHO) recommendations, the maximum concentration of phenolic compounds in drinkable water is 0.001 mg/l. The EU set a limit of less than 0.0005 mg/l for phenol in all forms [7]. Numerous techniques have been employed to treat phenolic wastewater, including biological treatment [8], adsorption [9, 10], extraction [11], electro-Fenton technique [12], electrocoagulation [13], ion exchange [14], photo-decomposition [15], and advanced oxidation process (AOP) [9, 10]. However, very few treatment technologies are considered effective in removing phenol.

Electrochemical methods of organic compound removal from petroleum refinery effluent have received a lot of interest because they have numerous distinct benefits over other methods of removal such as environmental compatibility, adaptability, energy efficiency, selectivity, safety, automation flexibility, and cost-effectiveness [16]. Because electrons are a flexible, efficient, clean reagent that is easy to automate and has greater pollutant degradation efficiency than standard methods, electrochemical technologies offer an alternate solution to many environmental problems in the process industry, so organic materials can be removed from wastewater by electrochemical oxidation methods by using direct or indirect oxidation strategies on the anode [17].

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AOP techniques like chemical, electrochemical, photocatalytic, and sonochemical oxidation have a lot of interest. Each approach has advantages of its own, but depending on the situation, it may not always be possible for an oxidation process to meet all the needs for treating wastewater. Nonetheless, combining multiple oxidation processes has become a viable choice for current investigation. Recently, sonoelectrochemistry combined ultrasound with an electrochemical reaction [18, 19].

The combination of electrochemistry and ultrasonic irradiation has garnered significant interest as a means of eliminating dissolved contaminants from water. This curiosity is sparked by the possibility that sonoelectrochemical methods could fully mineralize dissolved contaminants, transforming them into innocuous mineral species like carbon dioxide and water [20].

The cavitation (bubble creation and deflation) that is caused by intense ultrasound can have both chemical and physical effects producing oxidizing species such as $OH^{\bullet}$ radicals and H_2O_2, disruption of the diffusion layer, improved ion mass transfer over the double layer, periodic cleaning and activation of the electrode's surfaces. It can destroy toxic pollutants in a wide concentration range without the need for high temperatures or pressures, and it is environmentally friendly because it does not require the use of chemicals or gas emissions. It is the reason for the destruction of contaminants or the amplification of other oxidation processes in the US [21, 22].

The performance and efficiency of electrochemical processes are affected by several factors, including electrode potential and electrode materials. The best degradation of phenol can be obtained when the electrode material has high oxygen overpotential (OER), excellent electrical conductivity, and high stability [23]. Boron-doped diamond (BDD), Ti/PbO_2, and Ti/SnO_2 electrodes all have quite high OER potential [24].

Choosing the right electrode material is crucial for maximizing process efficiency and selectivity. The following characteristics must be present in electrode material features: high physical and chemical stability, resistance to corrosion and erosion, low cost-to-life ratio, selectivity and activity of the catalyst, excellent electrical conductivity, and inexpensive and dependable electrode materials [25- 28].

In this work, a comparison between sonoelectrochemical processes over different anodes (graphite, stainless steel, and titanium) would be accomplished and the Taguchi design approach was employed to investigate the impact of three variables (CD, time, and phenol concentrations) on the effectiveness of phenol removal by using the best anode in the sonoelectrochemical indirect oxidation process.

2- Experimental work

2.1. Chemicals

All compounds used in this investigation were of reagent grade and no more purification was required. These chemicals were: H_2SO_4 (Alpha Chemika, purity of 98.0%), phenol (LOBA Chemie, purity of 99.5%), NaCl (Central Drug House (P) Ltd-CDH, purity of 99.9 %), the aqueous solutions were prepared with distilled water.

2.2. Sonoelectrochemical setup

The electrochemical cell in the present study was a glass beaker with a 500 ml volume that was immersed in an ultrasonic bath (Ultrasonic; model: 031S) at a frequency of 40 kHz as displayed in Fig. 1. Sonication was produced parallel to the liquid surface and electrodes was positioned vertically in the solution. Graphite- graphite, or stainless steel-stainless steel, or titanium-titanium plates were used as the working and auxiliary electrodes (cathode & anode) and the distance between the cathode and anode was kept at 2.5 cm. A DC power supply was used to provide the required current, and the temperature of the electrolyte was maintained at about 30°C. The performance of sonoelectrochemical indirect oxidation process was examined by using it to remove phenol from aqueous solution with different phenol concentrations. Hence, indirect oxidation was utilized in the present study, and 3 g/l of NaCl was added to the solution. Just a few drops of H_2SO_4 were added to the electrolytic solution to reduce the pH of the aqueous solution to 3, which is the best value in electrochemical systems to remove phenol effectively [29, 30].

The phenol content was measured by using a UV-visible spectrophotometer (MANAGEMENT CO. LTD, UV-9200, UK) at a wavelength (λ) of 265 nm. The phenol removal efficiency (PRE) is determined by Eq. 1 [31].

$$\text{PRE} = \frac{Ipc - Cpc}{Ipc} \times 100 \quad (1)$$

Where Ipc is the initial phenol concentration in mg/l, and Cpc is the final phenol concentration in mg/l.

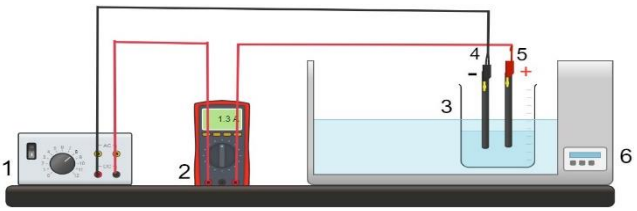

Fig. 1. Scheme Diagram of the Sonoelectrochemical Process, 1. Power supply, 2. Multimeter, 3. Beaker,4. Cathode, 5. Anode, and 6. Digital Ultrasonic Bath

2.3. Experimental design

The Taguchi design technique was utilized to examine the effects of three factors on the efficacy of phenol

removal by using a graphite anode as the working electrode and another graphite electrode as the auxiliary electrode. The experiment's data was analyzed using MINITAB 18 software. Three factors were considered in the sonochemical indirect oxidation which were; CD (25, 50, 75, and 100 mA/cm^2), time (1, 2, 3, 4 h), and phenol concentration (100, and 200 mg/l), there were two levels of phenol concentration and four levels for each CD, and time as illustrated in Table 1.

Table 1. Control Parameters and Levels

Studied parameters	values
CD, mA/cm^2	25, 50, 75, 100
Time, h	1, 2, 3, 4
phenol concentration, mg\l	100, 200

The orthogonal array layout of L_{16} ($4^{\wedge 2}\times 2^{\wedge 1}$) was utilized in the present study as shown in Table 2. Where $4^{\wedge 2}$ denotes two of the factors had four distinct levels, and $2^{\wedge 1}$ signifies that one parameter had two distinct levels.

Table 2. Layout of L_{16} ($4^{\wedge 2}\times 2^{\wedge 1}$) Experimental Design

Exp. No.	CD, mA/cm^2	Time, h	phenol concentration, mg\l
1.	25	1	100
2.	25	2	100
3.	25	3	200
4.	25	4	200
5.	50	1	100
6.	50	2	100
7.	50	3	200
8.	50	4	200
9.	75	1	200
10.	75	2	200
11.	75	3	100
12.	75	4	100
13.	100	1	200
14.	100	2	200
15.	100	3	100
16.	100	4	100

3- Results and discussion

3.1. Comparison between sonoelectrochemical processes over different anodes

Comparison between sonoelectrochemical processes over different anodes (graphite, stainless steel, and titanium) was carried out at 45 mA/cm^2 CD, 40 kHz ultrasonic frequency, 150 ppm phenol concentration, and 30 °C and the results of removal were recorded after 4 h. Fig. 2 shows that phenol removal % was 63.45, 39.43, and 17.98 for graphite, stainless steel, and titanium anode, respectively. This means that the best phenol removal was obtained with graphite anode in the Sonoelectrochemical processes due to its greater capacity for oxidation, and significant conductivity, besides the effect of hypochlorous acid (HOCl) that is generated in acidic conditions which is a very strong oxidant in comparison with hypochlorite ion (OCl-). The findings of this investigation are consistent with the findings of [32] who demonstrated that the kind of anode material and electrochemical parameters had a major impact on the rate of pollution removal.

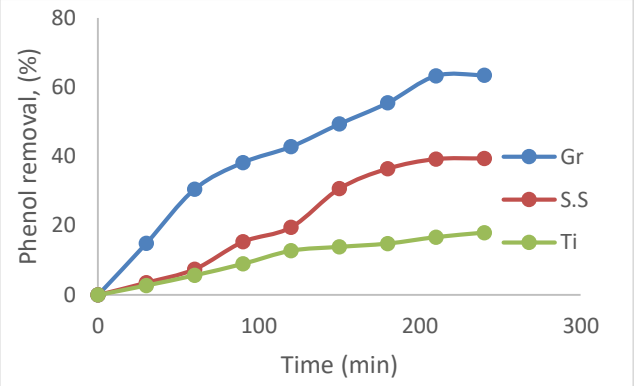

Fig. 2. Removal of Phenol by Sonoelectrochemical Processes Over Various Electrodes at 45 mA/cm^2 CD, 40 kHz Ultrasonic Frequency, 150 ppm Phenol Concentration, and 30 °C

3.2. Taguchi design analysis for phenol removal

a. Signal to noise (S\N) and multiple regression model

The Taguchi design is usually utilized in order to determine the optimum operating parameter levels that provide high removal efficiency. It determines which parameters had the greatest and fewest significant effects on the process's efficiency. Regression analysis was used to establish a mathematical relationship between the parameters and the output. The multiple regression equation (Eq. 2) which illustrated the relationship between the parameters and phenol removal efficiency is shown as follows:

$$\text{Phenol Removal \%} = 8.04 + 0.4546\,X1 + 18.12\,X2 - 0.1001\,X3 - 1.980\,X2^2 \quad (2)$$

Where X1 is the CD (mA/cm^2), X2 is time (h), and X3 is the phenol concentration (mg/l). The predicted equation had a correlation coefficient (R^2) of 98.02%.

Taguchi method implements the signal-to-noise (S/N) ratios to identify control parameter levels that minimize response variance resulting from noise factors. The S/N ratio refers to the desired value (mean of the output) to the standard deviation of the output value (undesired value) [33]. There are three categories of S/N ratio: the larger is the better (LTB), the nominal is the better (NTB), and the smaller is the better (STB). The type of S/N was chosen based on the experimental process's purpose and the intended process quality feature. To maximize phenol removal efficiency, the (LTB) of S/N was used [34]. The standard formula of The LTB response is presented in Eq. 3 [35].

$$S/N = -10 \log \left[\frac{1}{n}\left(\sum_{i=1}^{n}\frac{1}{yi2}\right)\right] \quad (3)$$

Where yi is the response of each experiment, and n is the number of repetitions of each experiment. Generally, the highest S/N value is preferred. Fig. 3 displays a plot of S/N versus CD, time, and phenol concentration.

Table 3 displays the experimented results of the PRE and the S/N ratios for each experiment's response that were determined using Eq. 3, and the predicted values of PRE based on Eq. 2.

Table 3. Experimental and Predicted Values of PRE and S/N Ratio Results of Taguchi Experimental Design

Exp. No.	CD, mA/cm^2	Time, h	phenol concentration, mg/l	Experimental PRE, %	S/N	Predicted PRE, %
1	25	1	100	26.78	28.5562	25.535
2	25	2	100	39.56	31.9451	37.715
3	25	3	200	38.00	31.5957	35.925
4	25	4	200	39.53	31.9385	40.185
5	50	1	100	33.50	30.5009	36.9
6	50	2	100	45.87	33.2306	49.08
7	50	3	200	43.56	32.7818	47.29
8	50	4	200	52.67	34.4313	51.55
9	75	1	200	38.54	31.7182	38.255
10	75	2	200	49.32	33.8605	50.435
11	75	3	100	73.50	37.3257	68.665
12	75	4	100	73.78	37.3588	72.925
13	100	1	200	52.66	34.4296	49.62
14	100	2	200	60.78	35.6752	61.8
15	100	3	100	80.34	38.0986	80.03
16	100	4	100	81.79	38.2540	84.29

The impact of every control parameter on the response is determined from the response for S/N (Table 4), which is graphically depicted in Fig. 3, and in addition to means (Table 5) which is graphically depicted in Fig. 4. These tables display ranks based on delta statistics. The delta statistic is calculated by subtracting the highest average for each variable from the lowest average for the same variable. Rank (1) is given to the highest delta value. Rank (2) is given to the second highest delta value, and so on [36]. The delta values in Table 4 showed that CD had the largest impact on phenol elimination, the second factor that affected phenol removal was time, with phenol concentration having the smallest impact.

The optimal conditions for phenol removal, according to the main effect plot of the S/N ratio, are CD of 100 mA/cm^2, time of 4 h, and phenol concentration of 100 mg/l. These optimum conditions also agree with the conditions of run (16) where the efficiency of the phenol removal was 81.79%.

Fig. 5 shows the interaction plot for phenol removal. It can be predicted that there is a small interaction between parameters which can be not considered and this agrees with the multiple regression equation.

Table 4. Response Table for S/N

Level	CD	Time	Phenol concentration
1	31.01	31.30	34.41
2	32.74	33.68	33.30
3	35.07	34.95	-
4	36.61	35.50	-
Delta	5.61	4.19	1.10
Rank	1	2	3

Table 5. Response Table for Means

Level	CD	Time	Phenol concentration
1	35.97	37.87	56.89
2	43.90	48.88	46.88
3	58.79	58.85	-
4	68.89	61.94	-
Delta	32.92	24.07	10.01
Rank	1	2	3

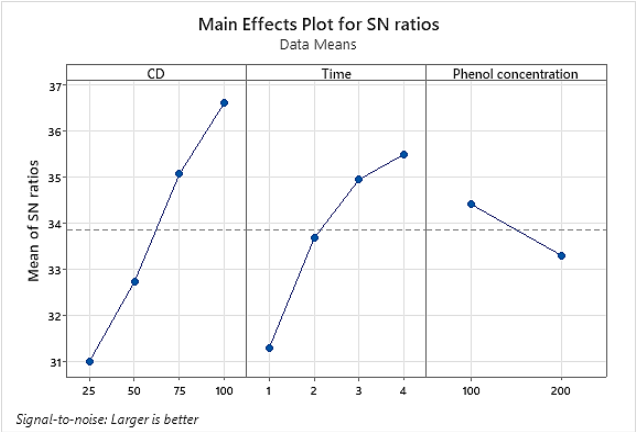

Fig. 3. Main Effects Plot of S/N Ratio

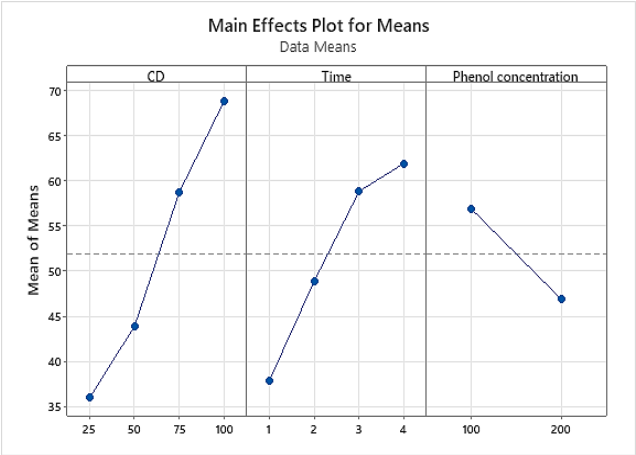

Fig. 4. Main Effects Plot of Means

b. ANOVA results

ANOVA is used to find out which parameters have the most impact on the process and find the parameter's percentage contribution, since it is feasible to determine the source of the variation during the process, ANOVA makes it simple to determine the order of significance of the process variables [35]. The ANOVA has been

established based on the degree of freedom (DF), sequential sum of the square (seq SS), adjusted sum of square (Adj SS), adjusted mean of square (Adj MS), F-value, and p-value. These elements demonstrated the significance of every parameter for process performance. [37].

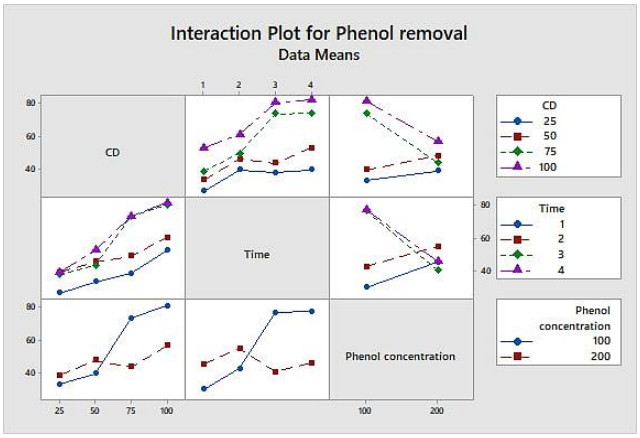

Fig. 5. Interaction Plots for Phenol Removal

The ANOVA results are listed in Table 6. The contributions of the three parameters; CD, time, and phenol concentration were 58.30, 31.66, and 8.93%, respectively, the significance of the elements succeeded in the following order of importance: CD > time > phenol concentration.

The F value for each analyzed parameter is statistically defined as the ratio of the mean value of squared deviations to the mean value of squared error. In general, when the F value is more than 4, a change in the examined parameter has a significant impact on process performance. Since the F values are more than 4, it is obvious that all parameters, including CD, time, and phenol concentration, have a considerable impact on phenol elimination by the sonoelectrochemical oxidation process. The P-value indicates the relevance of each factor in response. When the factor's p-value is less than 0.05, it is considered significant. The study's P-values of < 0.05 show that every model term is significant [38].

Table 6. Analysis of Variance Results for Phenol Removal

Source	DF	Seq SS	Contribution	Adj SS	Adj MS	F-Value	P-Value
CD	3	2615.97	58.30 %	2615.97	871.989	140.27	0.000
Time	3	1420.40	31.66 %	1420.40	473.466	76.16	0.000
phenol concentration	1	400.60	8.93 %	400.60	400.600	64.44	0.000
Error	8	49.73	1.11 %	49.73	6.217		
Total	15	4486.70	100.00 %				
General Linear Model	S	R-sq	R-sq(adj.)	PRESS	R-sq(pred.)		
	2.49334	98.89 %	97.92 %	198.935	95.57 %		

c. Confirmation experiment

Two experiments were conducted to confirm the optimal parameters and Table 7 shows the results. After 4 h of electrolysis by using graphite-graphite electrodes, the average phenol removal efficiency by the sonochemical indirect oxidation system was 80.99%.

Table 7. Confirmation Experiments of Phenol Removal

Run	CD, mA/cm^2	Time, h	phenol concentration, mg/l	Experimental PRE, %	Average
1	100	4	100	80.95	80.99
2	100	4	100	81.03	

3.3. Factors controlling the sonoelectrochemical advanced oxidation processes

a. The effect of current density

The sonoelectrochemical process was carried out at different CD (25, 50, 75, and 100) mA/cm^2, initial phenol concentrations of 100 mg/l, pH of 3, and ultrasonic frequency of 40 kHz, temperature of 30°C and NaCl of 3 g/l. Fig. 6 (2D plot (A) and surface plot (B)) shows that the phenol removal percentages were 50.195, 61.56, 72.925, and 84.29% at 25, 50, 75, and 100 mA/cm^2, respectively, after 4h of electrolysis. It means that phenol degradation was enhanced as a result of the rise in CD because of the increase in the removal efficiency of phenol proportionate to the number of charges developed in electrochemical techniques with improving CD [39]. The CD is an important component in influencing the production of highly oxidizing compounds (HOCl), because an increase in CD enhances the synthesis of effective oxidizing chemicals over a defined period of time, it typically leads to a faster breakdown of organic pollutants [40, 41, 42, 43]. Furthermore, it improves phenol removal efficiency and reduces reaction time by promoting the hydroxyl radical production rate at the anode interface [44, 45].

b. The effect of initial phenol concentration

Sonoelectrochemical oxidation studies were investigated to assess the treatment efficiency with various initial phenol concentrations (100, 150, and 200 mg/l), CD of 100 mA/cm^2, Temperature of 30°C, pH of 3, NaCl concentration of 3 g/l, ultrasonic frequency: 40 kHz. Fig. 7 (A and B) displays the experimental results, which demonstrate that the phenol removal was 84.29, 79.285, and 74.28 % in 4 hours at 100, 150, and 200 mg/l initial phenol concentrations, respectively. Therefore, lower initial concentrations of phenol were associated with higher phenol removal, because the initial concentration of phenol pollutants affects the degrading efficiency of all electrochemical advanced oxidation procedures [46].

Organic compounds will take more time to break down electrochemically at higher concentrations, and complete mineralization of organics will be inadequate, resulting in ineffective phenol removal [47]. This is due to an insufficient amount of hydroxyl radicals to break down additional organic substances [48].

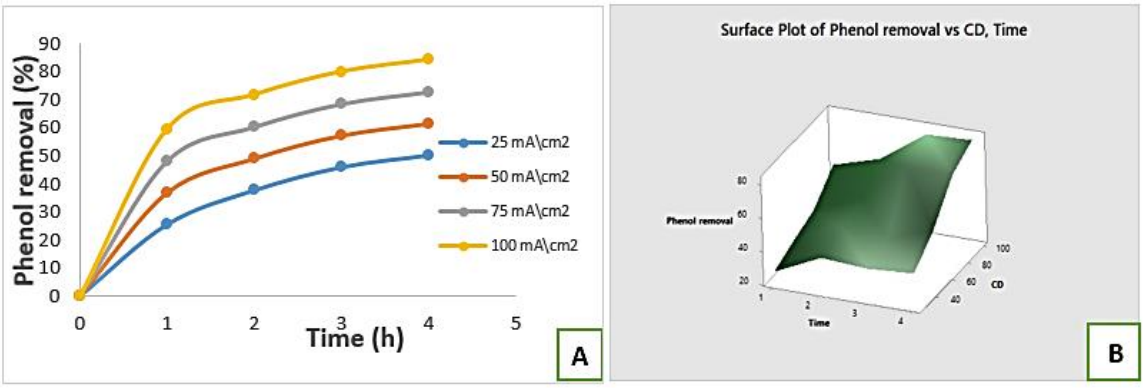

Fig. 6. The Effect of CD on Phenol Removal (%), at Initial Phenol Concentrations = 100 mg/l, NaCl conc. = 3 g/l, pH = 3, Temp. = 30 °C, and Ultrasonic Frequency = 40 kHz, (A) 2D Plot (B) Surface Plot

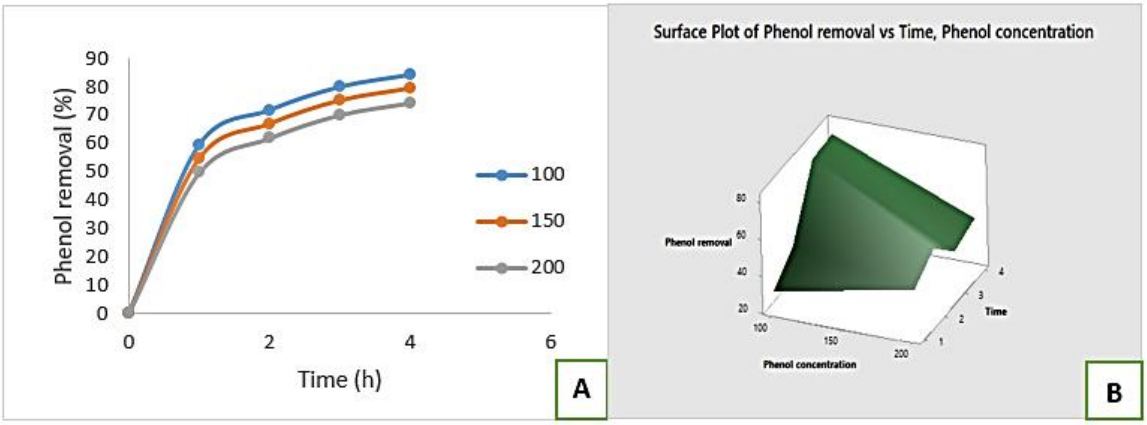

Fig. 7. The Effect of Initial Phenol Concentrations on Phenol Removal (%), at CD = 100 mA/cm^2, NaCl conc. = 3 g/l, pH = 3, Temp. = 30 °C, and Ultrasonic Frequency = 40 kHz, (A) 2D plot, (B) Surface Plot

4- Conclusions

The present study aimed to eliminate phenol from wastewater by using sonoelectrochemical- oxidation process with different electrodes: graphite, stainless steel, and titanium. The best phenol removal was obtained with graphite anode. Consequently, the graphite electrode was used as the main electrode in the Taguchi experimental design, where the L16 orthogonal array was accomplished. The study investigated the impact of three parameters (CD, time, and initial phenol concentration) on phenol elimination. The optimum conditions were acquired by using a linear model analysis for the means and S/N ratios. The optimal conditions for removing phenol were 100 mA/cm^2, 100 mg/l phenol concentration, and 4 hours of electrolysis. Under optimal operating conditions, phenol was removed at 80.99 % efficiency. The CD had the greatest impact on phenol reduction efficiency, whereas the phenol concentration had the least effect.

Nomenclature

Symbols	Description	Units
CD	Current density	mA/ cm^2
t	Time	
T	Temperature	°C
PRE	Phenol removal	%

Symbol	Description
Adj MS	Adjusted mean of square
Adj SS	Adjusted sum of square
AOP	advanced oxidation process
H_2O_2	Hydrogen Peroxide
HO$^{\bullet}$	hydroxyl Radical
Seq SS	Sequential sums of squares
UV	Ultraviolet

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ازالة الفينول بوساطة الاكسدة الكهروكيمياوية مع الموجات فوق الصوتية باستخدام اقطاب الكرافيت والفولاذ المقاوم للصدأ والتيتانيوم: إيجاد الظروف المثلى باستخدام طريقة تاكوتشي

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الخلاصة

يعد الفينول أحد أسوأ الملوثات العضوية ضررًا، وينتج مجموعة متنوعة من المواد العضوية الوسيطة السامة جدًا، لذلك كان من المهم إيجاد طرق فعالة لإزالته. إحدى التقنيات الواعدة هي المعالجة عن طريق الاكسدة الكهروكيمياوية مع الموجات فوق الصوتية، واختيار الأقطاب الكهربائية ذات الكفاءة العالية والسعر المعقول هو التحدي الأكبر. الهدف الرئيسي من هذه الدراسة هو دراسة إزالة الفينول عن طريق الاكسدة الكهروكيمياوية مع الموجات فوق الصوتية باستخدام أقطاب كهربائية مختلفة مثل الكرافيت والفولاذ المقاوم للصدأ والتيتانيوم. تم اختبار أفضل قطب كهربائي باستخدام طريقة تصميم تاكوتشي حيث تم دراسة ثلاث متغيرات كثافة التيار(٢٥ و٥٠ و ٧٥ و١٠٠ ملي أمبير/سم٢) والوقت (١و ٢ و ٣ و٤ ساعة) وتركيز الفينول الأولي (١٠٠ و ٢٠٠ ملغم/لتر) على ألاكسدة الكهروكيمياوية مع الموجات فوق الصوتية للفينول . تم استخدام الإشارة إلى الضوضاء (S/N) وتحليل ANOVA لفحص تأثير كل عامل. وكانت الظروف المثلى لإزالة الفينول هي ١٠٠ ملي أمبير/سم^{٢}، وتركيز الفينول ١٠٠ ملجم/لتر، و٤ ساعات من التحليل الكهربائي. وفي ظل ظروف التشغيل المثالية، تم التخلص من الفينول بكفاءة تصل إلى ٨٠,٩٩ %. وكان كثافة التيار هو العامل الأكثر تأثيرا على فعالية إزالة الفينول، في حين كان تركيز الفينول هو الأقل تأثيرا.

الكلمات الدالة: الملوثات العضوية، الأكسدة غير المباشرة، مياه الصرف الصحي، الموجات فوق الصوتية، الإزالة.