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Treatment of Al-Dewaniya hospital wastewater by electrocoagulation method using SS/Fe electrodes

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ABSTRACT

The present study focused on the treatment of hospital wastewater generated from Al-Diwaniya Hospital located at Al-Diwaniya City/ southern Iraq via an Electrocoagulation (EC) process with SS/Fe electrodes. Response Surface Methodology (RSM) was used to evaluate the main effects of parameters, their simultaneous interactions, and the quadratic effect to achieve the optimum condition for the EC process. Chemical Oxygen Demand (COD) was observed and measured for each experiment as it can be used as a good indicator of the quality of wastewater. The impacts of three factors such as current density (5-25mA/cm^2), pH (4-10), and addition of NaCl(0-3g/l) were evaluated. The obtained experimental data were fitted to a second-order polynomial equation with analysis by variance analysis (ANOVA). The results show that current density has a major impact on the efficiency of COD removal followed by the addition of NaCl while pH has a lower effect on the COD removal under the studied range of pH. ANOVA results showed that the determination coefficient of the models was R^2 98.18% confirming that the quadratic model was significant with a good fitting between the experimental and predicted results. The optimized operating parameters were a current density of 25 mA/cm^2, pH of 7.8, and NaCl addition of 3 g/l in which COD removal efficiency of 97.14% was achieved with a specific energy consumption of (30.914) kWh/kgCOD.

1. Introduction

Hospitals release significant quantities of contaminants into their wastewater. Medical waste management is a significant issue for countries all around the world, particularly because of the dangers it presents to the environment. Ö. Gökkuş et al. [1]. Hospitals are general offices that offer health services to people from all walks of life and serve as medical offices for health and research education. Hospitals may host a wide range of activities, from nonmedical to medical, all of which create solid, liquid, and gas wastes. These wastes will have an influence on the land, water, and air. Purwanto et al.[2]. Hospital Waste Waters (HWWs) consider the most harmful kinds of pollutants in nature. HWWs contain pathogens such as bacteria, parasites, and viruses, as well as radioactive isotopes and hazardous chemical compounds. Dehghani et al. [3]. Untreated medical waste can have disastrous consequences not only for those working in clinical facilities but also for those in the surrounding community. El-Haggar et al.[4]. As a result, this kind of pollutant cannot be discharged into the sewerage system as untreated effluent. Ö. Gökkuş et al.[1] . In the case of treated hospital wastewater, they can be reused for agricultural purposes. Beier et al. [5].

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Nomenclature

Adj. MS	The adjusted mean of the square	*HWWs*	Hospital wastewaters
adj. R^2	The adjusted coefficient of multiple correlations	*I*	Current applied (A)
Adj. SS	The adjusted sum of the square	*NTU*	Turbidity
a_i	The first-class(linear) major effect	*pred.* R^2	Predicted multiple correlation coefficient
a_{ji}	Second-class major effect	*RE%*	Removal Efficiency (%)
a_{ijj}	The interaction effect	R^2	Linear regression coefficient
ANOVA	Analysis of variance	*RSM*	Response surface methodology
Al	Aluminium	*SEC*	specific energy consumption (kWh/kg COD)
BBC	Box–Behnken Design	*SeqSS*	The sum of the square
CE	Current Efficiency (%)	*SS*	Stainless steel
CI	Confidence interval	*t*	Time
Cr. %	Percentage contribution for each parameter	*TOC*	Total organic compound
COD	Chemical Oxidation demand	*TDS*	Total dissolved solids
Contr.	Percentage contribution for each parameter, %	*V*	The volume of electrolyte, cm^3
DC	Direct current power supply	x_1	Coded value of Current density
D_F	The desirability function	*X1*	Current density
DOF	Degree of freedom	x_2	Coded value of pH
U	The voltage of the cell, Volt	*X2*	pH
EC	Electrocoagulation	x_3	Coded value of NaCl addition
F	Faraday constant (96500) , A s mol^{-1}	*X3*	NaCl addition
Fe	Iron	*Y*	Represents the dependent variable (RE, %)

The treatment of HWWs is mostly quite complex since each effluent has its own characteristics that may be different from others hence posing specific troubles for treatment.

Tekin et al.[6] The conventional methods for treating HWWs are basically biological and physiochemical processes [7],[8]Notwithstanding, these strategies have shown restricted accomplishment for the treatment of HWWs because of the nature and structure of these effluents. a lot of sludge was produced when using chemicals in the sewage processing system, meanwhile, the microbiological processing system involves using a large area of land and a long processing time, therefore the conventional processing methods have been less applied in this field. Murdani et al.[9]. Another alternative approach that can be tried to cover the shortage of conventional technology is called the electrocoagulation process. Kermet et al.[8]

Electrocoagulation (EC) is an electrochemical technique that uses soluble electrodes such as aluminum or iron. When voltage is applied to the soluble electrodes, active cation species or "mediators" are generated, which can react with the target pollutant. Hydrogen generated at the cathode also aids in the separation of formed flocs. Mission et al.[10] The employment of electrons rather than chemical reagents or microbes to assist electrochemical treatment in the elimination of hazardous organic contaminants is extremely intriguing. Carlesi Jara et al. [11].

The electrochemical process has many advantages over traditional ones because of its unique properties, such as less chemical addition, simple design, reduced sludge generation, a little area required for setup, and very quick sedimentation due to the development of flocs. Bracher et al.[12]. EC systems have high efficiency, a rapid reaction rate, cost-effectiveness, and compact size (enabling decentralized treatment), easy automation. Palahouane B et al.[13] and no hazardous material creation. They created minimal TDS and secondary pollutants, and have the ability to remove the smallest size of colloidal particles as well. M. Yoosefian et al.[14] .EC is an electrochemical management approach that produces active coagulants using sacrificial anodes. Many mechanisms are used in this procedure to remove contaminants from aqueous effluents. As an anodic reaction, the dissolution of Fe and production of adsorbents (hydrated iron hydroxides) occurred simultaneously with the evolution of hydrogen gas as a cathodic reaction, resulting in absorbent flotation. As a result, either gas flotation or sedimentation can be used to remove the produced flocs. All reactions that happened at the surface of the anode and cathode, as well as in the solution, during electrocoagulation, are represented by equations (1, 2, and 3) [8],[15]:

Anodic reaction:

$$Fe \rightarrow Fe^{3+} + 3e^- \quad (1)$$

Cathodic reaction:

$$3H_2O + 3e^- \rightarrow 3/2H_2(g) + 3OH^- \quad (2)$$

In the solution:

$$Fe^{3+}{}_{(aq)} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+{}_{(aq)} \quad (3)$$

At the electrocoagulation process, two interaction mechanisms between hydrolysis products and contaminant were observed namely adsorption and precipitation each of which is suggested for a different pH range. Flocculation at low pH levels is explained by precipitation, whereas at higher pH levels, it is explained by adsorption. Gökkuş et al.[1]. Electrocoagulation (EC) can remove toxic pollutants from wastewater, such as Cr(IV), dyes, olive mill pollutants, Vepsa¨la¨inen M et al.[16].COD from petroleum refinery effluent, Alkurdi et al.[17] and is a good choice for wash water treatment Wang et al. [18], industrially processed water, and medicinal water treatment. Bajpai et al.[15]. For a better experimental methodology, statistical methods were favored for finding the optimal combinations of parameters and their interactions. These methods offer benefits such as reducing time and study costs. Montgomery D.C et al.[19]. Response Surface Methodology (RSM) is a statistical approach for designing and optimizing trials in which the experimental responses are fitted to quadratic functions. Kalil S.J et al.[20].

The dependent variable value is estimated using the RSM regression modeling approach, which is dependent on the controlled values of

independent variables. This approach generates a large number of trial combinations in a short amount of time to improve laboratory testing results. In order to fit a quadratic model, BBD plans only work at three levels for one of each factor with the fewest runs. The estimated parameter may be used to compute the factor that contributes the most to the estimated value, allowing researchers to emphasize the parameters most important to accept the product. M. Otto et al.[21] .RSM has been used to model and optimize wastewater treatment systems with great success [22],[23].
This study aimed to evaluate the feasibility of using an electrocoagulation process using iron electrodes as a cheap material to treat wastewater generated by a local Iraqi hospital called the general hospital, which is located in Al-Diwaniya City. The impacts of operating parameters such as current density, pH, and NaCl addition on the efficiency of COD removal from hospital wastewater were studied, and the optimum conditions were determined by using RSM.

2. Experimental work

Hospital wastewater was collected from the Al-Dewaniya Hospital's sewage system (located in Al-Diwaniya City, Iraq) right before it was mixed with the campus's household wastewater. Table 1 shows the characteristics of hospital wastewater. During the experimental program, this hospital wastewater was stored at 4 ^{0}C, and the needed sample for each experiment (0.7 L) was taken at the time of each experiment.

Table 1. Physiochemical properties of effluents in Al-Dewaniya hospital sewage system.

parameter	Value
COD(mg/l)	735
pH	7.8
T.D.S(mg/l)	2480
Cl-(mg/l)	1.4
SO4-2(mg/l)	0.7
Turbidity(NTU)	9.5
Conductivity (mS/cm)	1.95

All experiments were carried out in a batch system using a cylindrical jacketed Perspex electrochemical cell having a length of 200 mm, a diameter of 100 mm with an internal thickness of 5 mm,(Fig.1). The cell has a working capacity of around 0.7 L. The cover of the cell has exterior dimensions of (130 mm outer diameter and a thickness of 10 mm) and has five slots for electrode inserting and holes for embedding pH and conductivity meter, as well as sample taking out. As cathode and anode electrodes, three stainless steel and two Iron plates with dimensions of 15 × 5 × 1 cm were used respectively. A 2.5 cm gap between the anode and cathode was fixed. To ensure homogeneity inside the reactor and reduce floc separation, stirring the mixture at 300 rpm was applied. Bajpai et al.[15]. By using a water circulator (Memmert, Germany, type: WNB22), all runs were achieved at a constant temperature of 25±2 ºC.
The tested Hospital wastewater was found to have a low conductivity of 1.95ms/cm, causing an increase in cell potential. In this case, a supported electrolyte should be used to raise the conductivity of the solution. Sodium sulfate (Na_2SO_4) was used as an electrolyte at a concentration of 0.05 M, resulting in a final conductivity of 12.9 mScm^{-1}, which is within the needed range for obtaining low cell potential. Souza et al.[24].
The pH of the solution was measured by a digital pH meter (HNNA Instrument Inc.PH211, Romania) and adjusted using 0.1 M NaOH or 0.1 M H_2SO_4. Conductivity and TDS were determined using (HM Digital Inc. model COM-100, Korea). To deliver the required electrical current, a DC power supply (UNI-T, UTP3315PF) with a maximum voltage of 30 V and a maximum current of 5 A was utilized. Anodes and cathodes were washed with ethanol and water before each cycle to eliminate contaminants. After filtering the samples of each run, the COD value was calculated to assess the process' performance. All of the trials were repeated three times and only the average values were taken.

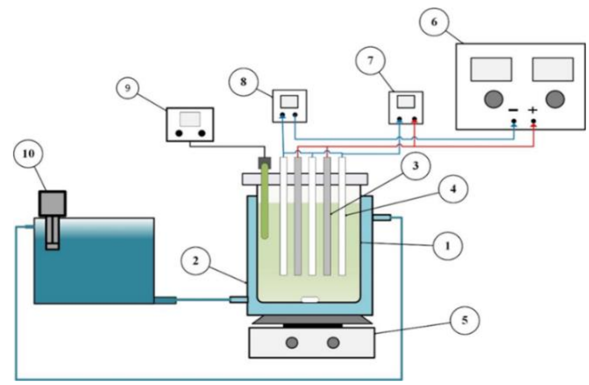


Figure 1. The electrochemical system: 1) cell body, 2) jacket, 3) iron anode, 4) stainless steel cathode, 5) magnetic stirrer, 6) power supply, 7) voltmeter 8) Ammeter,9) pH-meter,10) water bath circulator.

By using a COD thermos-reactor (RD125, Lovibond), a sample (2ml) of effluent was digested with $K_2Cr_2O_7$ at 150 °C for 120 minutes to determine its COD value after cooling it to room temperature, then analyzing it in spectrophotometer (MD200, Lovibond). The removal efficiency of COD was calculated using Eq. 4, Şengil et al.[25]:

$$RE\% = (Ci - Cf)/Ci \times 100 \quad (4)$$

Where C_i is the initial concentration (mg/l) and C_f is the final concentration (mg/l). Iron consumption was determined by weighing electrodes before and after each experiment, and the Fe consumption (kg^{-3}) was estimated using Eq. 5:

$$Fe\ (kgm^{-3}) = (\text{Initial weight} - \text{final weight}) / \text{volume of sample} \quad (5)$$

The quantity of energy required to digest one kilogram of COD is known as the specific energy consumption (SEC). Eq. 6 may be used to calculate SEC in (kWh/kg). Alkurdi et al.[17]:

$$SEC = \frac{U.I.t \times 1000}{(COD_i - COD_f)\,V} \quad (6)$$

Where SEC is the specific energy consumption (kWh/kg COD), I is the current (A), U is the applied cell voltage (Volt), t is the electrolysis time (h), V is the volume of effluent(L), and COD_i and COD_f are the initial and final values of COD (mg/l).

2.1. Experimental design

Model fitting and determining the optimum operating conditions for a response can be achieved by using a collection of statistical and mathematical techniques formulated by Minitab-17 Software. In Minitab-17 Software, There are a variety of techniques for optimization of the response, but in this study, the Box Bhenken design was used to optimize and determine the influence of factors like current density, pH, and electrolyte (adding NaCl) on COD elimination effectiveness by electrocoagulation. Current density (5-25mA/cm^2), pH (4–10), and NaCl addition (0-2g/l) were the range of operational factors. The chosen values of operational factors were designed based on reviewing some literature [6], [15],[26],[27]. The factors were labeled as X1, X2, and X3 in Table 2. All factors were divided into three categories, with -1, 0, and +1 representing low, moderate, and high values, respectively. Before starting the experimental runs, a preliminary run was achieved to determine the suitable electrolysis time. The selected operating conditions were current density (25mA/cm^2), pH (7), and NaCl addition (1.5 g/l). The results of COD decreasing with time is shown in Table 3. Based on the results of Table 3, it was found that an electrolysis time of 90 min is suitable for achieving the experimental design to give significant results of RSM since the removal efficiency of COD is greater than 80%. Using higher time may be not giving a clear picture for the effects of parameters.

Table 2. Process parameters and their levels.

Process parameters	range in Box–Behnken design		
Coded levels	Low(-1)	Middle(0)	High (+1)
X1- Current density (mA/cm^2)	5	15	25
X2- Ph	4	7	10
X3-NaCl (g/l)	0	1	2

Table 3. Selecting the best electrolysis time based on decreasing COD with time.

Time (min)	0	20	40	60	70	80	90	100	120
COD(mg/l)	759	714	620	541	400	236	130	60	20

Based on Minitab-17 Software using Box Bhenken design, 15 runs should be performed in a trial design with three repetitions of the center point. Repetition is useful for evaluating pure errors from the sum of squares. Table 4 illustrates the BBD proposed for the present research.

Table 4. Box- Behnken experimental design.

Run	Blocks	Coded value			Real value		
		x1	x2	x3	Current density	PH X2	NaCl (g/l) X3
1	1	0	0	0	15	7	1.5
2	1	1	-1	0	25	4	1.5
3	1	0	1	1	15	10	3
4	1	0	0	0	15	7	1.5
5	1	-1	-1	0	5	4	1.5
6	1	0	-1	1	15	4	3
7	1	-1	0	-1	5	7	0
8	1	1	0	-1	25	7	0
9	1	0	0	0	15	7	1.5
10	1	0	1	-1	15	10	0
11	1	1	0	1	25	7	3
12	1	0	-1	-1	15	4	0
13	1	1	1	0	25	10	1.5
14	1	-1	0	1	5	7	3
15	1	-1	1	0	5	10	1.5

For the assessment of results, BBD provides correlation in which the data are set in a 2nd-order polynomial equation as follows [17]:

$$Y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_i^2 + \sum \beta_{ij} x_i x_j \quad (7)$$

Where Y represents the response (RE%), i and j are patterns index numbers, β_0 is the intercept term, x_1, x_2 … x_k are coded forms of process variables. β_i is the first-order(linear) main effect, β_ii second-order main effect and β_ij is the interaction effect. ANOVA has applied then the regression coefficient (R2) was calculated to check the model fit goodness.

3. Results and discussion

3.1. Statistical analysis

To investigate the combined impact of the independent variables on COD removal efficiency, fifteen batch runs were conducted at various process variable combinations. Table 5 shows the experimental findings obtained after 90 minutes of electrolysis time, including COD Removal Efficiency (RE%), Fe consumption, and Specific Energy Consumption (SEC).

Table 5. Experimental results of Box–Behnken design for COD removal.

Run-Order	Pt-type	Blocks	X1	X2	X3	RE%		Iron cons	EC
						Actual	Predicte		
1	0	1	15	7	1.5	93.40	93.2133	2.23	12.276
2	2	1	25	4	1.5	92.50	92.4500	3.8	27.966
3	2	1	15	10	3	95.57	95.9000	2.25	11.113
4	0	1	15	7	1.5	93.85	93.2133	2.51	12.296
5	2	1	5	4	1.5	84.90	85.5150	1.21	2.455
6	2	1	15	4	3	93.30	93.0650	2.3	10.892
7	2	1	5	7	0	88.90	88.6150	0.7	2.665
8	2	1	25	7	0	95.18	95.5600	4.1	30.233
9	0	1	15	7	1.5	92.39	93.2133	2.21	11.990
10	2	1	15	10	0	91.97	92.2050	2.21	14.266
11	2	1	25	7	3	97.96	98.2450	3.72	26.260
12	2	1	15	4	0	90.00	89.6700	3.17	14.156
13	2	1	25	10	1.5	94.90	94.2850	3.52	30.124
14	2	1	5	7	3	93.40	93.0200	2.72	1.988
15	2	1	5	10	1.5	89.00	89.0500	1.23	2.534

COD removal efficiency ranges from 84.90 to 97.96%, as can be observed. The iron consumption ranges from (0.7-4.1) kg/m3. (1.988-30.233)Kwh/kg COD is the energy consumption range. The variation between the design's center points is less than 2%, indicating high outcomes repeatability. Based on Minitab-17 software, a quadratic model in terms of real units of process variables was obtained which relates COD Removal Efficiency (RE%) with process variables as shown in Eq.8:

RE% = 61.20 + 1.351 X1 + 2.722 X2 + 9.87 X3 - 0.02042 (X1) - 0.1200 (X2)^2 - 1.297 (X3)^2 - 0.0072 X1X2 - 0.1005 X1X3- 0.233 X2X3 (8)

Where RE% is the response, and X1, X2, and X3 are current density, pH, and addition of NaCl respectively. Whereas X1X2, X1X3, and X2X3 represent the interaction effect of parameters. (X1)2, (X2)2, and (X3)2 represent a measure of the main effect of parameters current density, pH, and NaCl addition respectively. The effects of individual parameters (linear and quadratic) or double interactions on COD removal efficiency are shown in Eq.(8), where COD removal efficiency increases with increasing factors whose coefficients have positive values, whereas COD removal efficiency decreases with increasing factors whose coefficients have negative values. Current density, pH, and the addition of NaCl all have a favorable impact on COD removal efficiency, but all interactions have a negative impact. The predicted COD removal efficiency values were calculated using Equation 8 and reported in Table 5. The ANOVA of the response surface model is shown in Table 6. In this table, DF, SeqSS, Adj SS, Adj MS, and Contr .%, represent the degree of freedom, sum of the square, adjusted sum of the square, adjusted mean of the square, and the contribution for each parameter respectively. Fisher F-test and P-test are denoted by F-value and P-value respectively. The model's acceptance was tested using F-value, and P-value. When the F-value of the regression equation is big, it can fit most of the variation in the answer. The P-value is used to determine if F has a large enough value to acknowledge the model's statistical significance. At a P-value less than 5%, 95 % of the model's variability could be explained. Segurola et al.[28]

Table 6. Analysis of variance for COD removal of hospital wastewater treatment.

Source.	DOF	Seq. SS	Contr.(%)	Adj. SS	Adj. MS	F-value	P-value
Model.	9	143.110	98.18	143.11	15.9011	29.90	0.01
Linear	3	113.607	77.94	113.61	37.8690	71.21	0.00
(X1)	1	74.054	50.80	74.054	74.0544	139.25	0.00
(X2)	1	14.418	09.89	14.418	14.4184	27.11	0.03
(X3)	1	25.134	17.24	25.134	25.1340	47.26	0.01
Square	3	28.019	19.22	28.019	9.3395	17.56	0.04
X1*X1	1	2.592	01.78	2.7890	2.7894	5.25	0.07
X2*X2	1	16.943	11.62	15.054	15.0537	28.31	0.03
X3*X3	1	8.484	05.82	08.484	8.4840	15.95	0.01
2-Way Inter	3	1.485	01.02	01.485	0.4949	0.93	0.49
X1*X2	1	0.722	00.50	00.722	0.7225	1.36	0.29
X1*X3	1	0.740	00.51	00.740	0.7396	1.39	0.29
X2*X3	1	0.022	00.02	00.022	0.0225	0.04	0.84
Error	5	2.659	01.82	02.659	0.5318	----	----
Lack of Fit	3	1.541	01.06	01.541	0.5137	0.92	0.55
Pure-Error	2	1.118	00.77	01.118	0.5590	----	----
Total	14	145.769	100.0	----	----	----	----
Model-summary		S.	R^2.	R^2(adj)	PRESS	R-sg(pred)	
		0.729248	98.18 %	94.89 %	27.1709	81.36%	

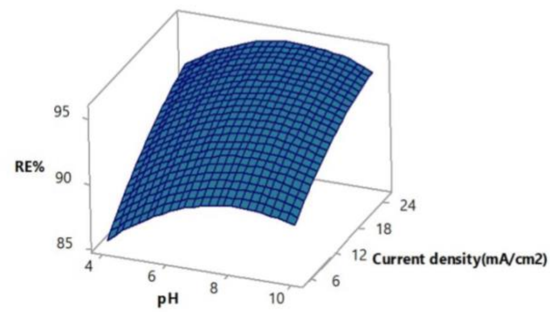
Table 6 shows that the quadratic model is significant, with a 95 percent confidence level and an F-value of 29.90. The COD elimination model's p-value was estimated to be 0.001, indicating that the created model is significant. Furthermore, in comparison with pure error, the lack of fit is not significant (p-value=0.559> 0.05), demonstrating that the model is effective, appropriate, and significant in describing the EC process's pollution removal [29],[30]. The three most essential correlation coefficients in the statistical summary model are correlation coefficient (R2), adjusted correlation coefficient (Adj. R2), and predicted correlation coefficient (pred. R2). To provide a high degree of fitting between observation and estimate value, the correlation coefficient (R2) should be near 1. Zhao et al.[31]. Based on the value of Adj. R2, which does not always rise with the addition of variables, the sample size and number of terms in the models might be adjusted. As a result, the Adj. The R2 value should be extremely near to the R2 value. Furthermore, the difference between Adj. R2 and pred. R2 should be less than 0.2 for the experimental and model-predicted values to be in excellent agreement. Zhao et al.[31]. In the present work, values of R2, Adj. R2, and pred. R2 was found to be 0.9818, 0.9489, and 0.8136 respectively. This demonstrates that the experimental and model-predicted values are compatible. Furthermore, the difference between Adj. R2 and pred. R2 was 0.1353, indicating the model's high significance.Table 6 shows that current density (X1) was the most important factor influence COD removal, accounting for a proportion of the total contribution (50.80%). The effect of adding NaCl (X3) is the second most important, accounting for 17.24% of the total, indicating the significance of chloride ions in the breakdown of organic compounds during electrocoagulation. The pH factor (X2) has a lower influence on COD removal with a percentage contribution (9.89%) when the pH range (4-10) was used, demonstrating that the highest pollutant removal efficiency may be achieved in this pH range Bajpai et al.[15]. Different works [32],[33] found similar outcomes.

Furthermore, interaction effects are not significant. The quadratic effects on COD elimination were found to contribute 19.22%, with the quadratic impact of current density (X1) being non-significant in comparison to pH and NaCl addition.

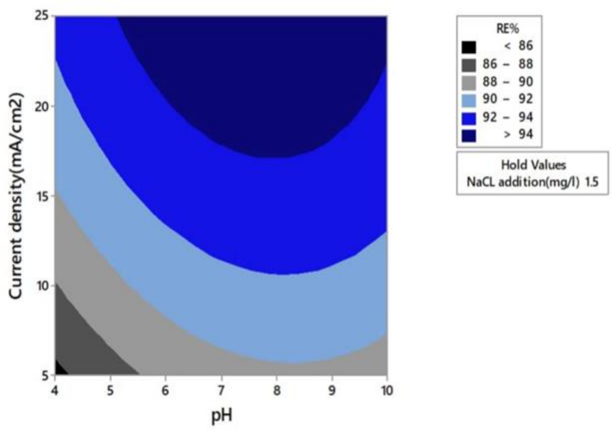
3.2. Effect of process variables on the COD removal efficiency

The influence of process parameters and their combinations on COD removal efficiency was studied using graphical demonstrations of statistical optimization based on RSM. Fig (2-a, 2-b) shows the combined effects of current density and solution pH on COD removal efficiency when NaCl (1.5g/l) is added at a constant rate. The response surface plot is shown in Fig 2-a, while the contour plot is shown in Fig 2-b. Fig 2-a shows that increasing current density increases COD removal effectiveness over the pH range (4-10). For example, increasing the current density from 5 to 25mA/cm^2 results in a significant increase in COD removal from 84.90% to 92.50% at pH=4 (Table 6, Exp.2 and 5). Besides, approximately the same increase in COD removal efficiency occurred at pH=10 from 89.00% to 94.90% (Exp.13 and 15, Table 6). When the EC procedure was carried out with an iron electrode, it was discovered that current density had the greatest impact on COD elimination efficiency. The rationale for these findings may be explained using Faraday's rule, which states that rising current density causes the dissolving rate of the Fe anode to rise, resulting in an increase in the formation of coagulants (Fe(OH)3 particles) at the anode. Liu et al.[34]. Furthermore, the size and rate of formation of hydrogen gas bubbles play an important role in the removal of pollutants by floatation, with an increase in current density leading to an increase in production rate and a decrease in bubble size Elazzouzi et al.[35]. Furthermore, when the number of bubbles produced at the cathode rises, the mass transfer rate and floc production increase. Yoosefian et al.[36]. Previous research [36],[37],[38] had shown similar findings.

As shown in Fig. 2, raising the pH enhances COD removal effectiveness; for example, increasing the pH from 4 to 10 at a current density of 5mA/cm2 leads in an increase in COD removal from 84.90% to 89.00% (Table 6, exp.5 and 15). At larger current densities, however, this increase in COD removal efficiency became less noticeable. Clearly, the influence of pH on COD removal efficiency is greater at pH values of 4–7 than at pH values of 7–10. besides at pH 7-10, Fig.2.b showed that pH starts to decrease when increasing pH beyond 8. However, this decrease is relatively low in comparison with increases in pH observed in the pH range of 4-7. This behavior may be interpreted as follows: The decline in efficiency at acidic pH might be due to a lack of hydroxyl ions as well as very low $Fe(OH)_3$ production. Furthermore, iron hydroxide particles are soluble at low pH (less than 7) and so do not have the potential to absorb contaminants. At pH 7, the insoluble $Fe(OH)_3$, $Fe(OH)^{2+}$, $Fe_2(OH)_2^{4+}$, and $Fe_{13}(OH)_{32}^{7+}$ are the dominant compounds and have the ability to adsorb the pollutants. At high pH values, $Fe(OH)_4^-$ is formed which is soluble in water, decreasing the removal effectiveness, especially at pH greater than 10[35,40]. Fig. 3. Predominance-zone diagrams for Fe^{+3}, it was observed that $Fe(OH)_3$ is the predominant species in the pH range(6-9) conferring the effect of pH on COD removal. Barrera-Diaz et al.[39].



(a)



(b)

Figure 2. The combined effects of current density and solution pH on the COD removal efficiency at constant addition of NaCl (1.5 g/l): (a) 3D surface plot,(b) contour plot.

Different literatures [40],[41],[42] have shown similar results. Based on the contour plot findings, it is evident that a COD removal efficiency of ≥94% could be accomplished within a certain pH range (6-10) and a current density range of 18-25 mA/cm^2.

At constant solution pH=7, the combined effects of current density and the addition of NaCl on COD removal efficiency are shown in Fig (4-a, 4-b). Fig 4-a denotes the response surface plot while Fig 4-b demonstrates the equivalent contour plot. It can be shown in Fig 4-a, that increasing current density increases COD removal efficiency across the whole range of NaCl addition. The elimination effectiveness of COD improves with increasing NaCl addition, as seen in Fig.4, As NaCL was added up to 3g/l at a current density of 5mA/cm^2, COD removal efficiency rose from 88.90% to 93.40% (Table 6, exp.7 and 14) when compared with no addition.

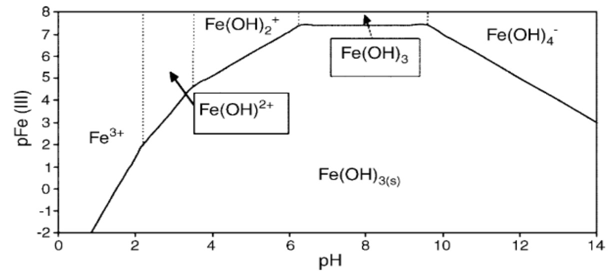
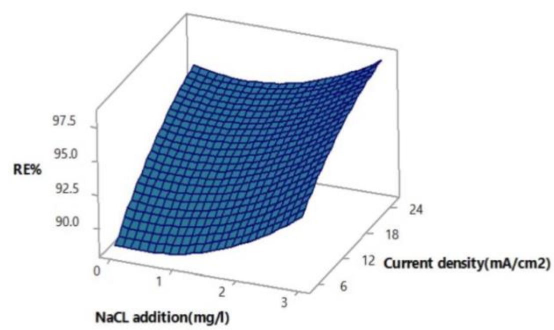
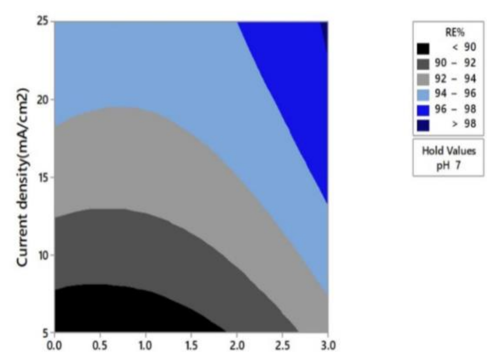


Figure 3. Predominance-zone diagrams for Fe+3 chemical species in aqueous solution [41].

According to literature reviews, adding NaCl to the EC process can improve efficiency by lowering the cell voltage resulting in a lowering of the process energy consumption. Furthermore, with the presence of NaCl electrolyte, the opposing effects of anions such as CO32-, HCO3 −, and SO4 2−, can be avoided. The presence of such anions causes Ca2+ or Mg2+ cations to precipitate as an insulating layer on the cathode surface, hence increasing the EC cell's ohmic resistance. Ahmadzadeh et al.[43]. Adding NaCl to the electrochemical process also causes the following reactions:



(a)



(b)

Figure 4. The combined effects of current density and addition of NaCl on the COD removal efficiency at constant solution pH=7(a) 3D surface plot,(b) contour plot.

$2Cl^- \rightarrow Cl_2 + 2e^-$ (9)

$Cl_2 + H_2O \rightarrow HOCl + Cl^- + H^+$ (10)

$HOCl \rightarrow OCl^- + H^+$ (11)

According to reactions 9 and 10, Cl- may oxidize to Cl2, a powerful oxidant that may aid in the oxidation of dissolved organic compounds, or it may lead to the production of HOCl, a strong oxidizer that may result in additional COD elimination in addition to electrocoagulation. Singh et al.[43] . Similar findings were reported in other studies[26],[43],[44]. Based on the contour plot findings, it is evident that a COD removal efficiency of ≥96% could be achieved within a range of NaCl addition (2-3g/l) and current density (12-25 mA/cm^2).

3.3. The optimization and confirmation test

For optimizing the system using Minitab-17 Software, many criteria should be considered to get the desired objective by making the desirability function (DF) maximum as possible as via adjusting the weight or importance. Five options should be considered as a target namely maximize, objective, minimize, within the range, and none. The aim of optimization is to get higher removal efficiency of COD therefore COD removal was selected to be the maximum with a corresponding weight of 1.0. The parameters studied in this research were identified within the range of the designed levels (Table 2). The lower and upper values of COD removal efficiency were allocated at 84.9% and 97.96% respectively. Optimization has been achieved using the response optimizer of Minitab-17 Software based on these constraints and settings. Results of optimization are illustrated in Table 7 with the desirability function of (1).

Table 7. Optimization of process factors for maximizing COD removal efficiency (RE%).

Response	Aim	Lower%	Target %		Upper %	Weight	Important
RE (%)	Maximum	84.9	97.9		97.9	1	1
Solution: Parameters :			Results				
Current density (mA/cm2)	pH	NaCl addition (g/l)	RE (%) Fit	**D_f**	SE. Fit	95% CI	95% PI
25	7.8	3	98.367	1.0	0.642	(96.717, 100.017)	(95.869, 100.864)

Table 8. The optimum COD removal efficiency confirmation.

NO	Current density mA/cm2	Ph	NaCl (g/l)	U Volt	COD (ppm) In	COD (ppm) Out	RE % Actual	RE % Average	EC. Kwh/kg COD
1	25	7.8	3	4.64	735	21	97.14	97.335	30.914
2	25	7.8	3	4.64	728	18	97.53		31.09
3	25	7	3	4.58	750	38	94.93		30.601

For confirming the optimization results, two experiments were performed based on the optimized parameters as shown in Table 8. After 90 min of the electrolysis, 97.335% COD removal efficiency (average value) was accomplished which is compatible with the range of the optimum value obtained from optimization results (Table 7). Therefore, Box–Behnken design combined with the desirability function can be applied as a successful and effective method for optimizing COD removal using the EC process. Further experiments were conducted in which a pH value of 7 was considered maintaining other parameters at their optimal values and its results are tabulated in Table 8. Results showed the possibility of using pH=7 with approximately good COD removal efficiency (94.93%) with the same energy consumption. A comparison between the characteristics of treated effluent based on the present work with the characteristics of effluent without treatment is shown in Table 9. It was clear that treated effluent has enhanced characteristics with a COD removal efficiency of 97.14%.

Table 9. Comparison between the wastewater effluent and the treated effluent.

Parameter / Effluent	COD (ppm)	pH	Turbidity (NTU)	EC (mS/cm)	Cl^- (g/l)	SO_4^{-2} (g/l)
Raw effluent	735	7.9	9.66	1.89	1.56	0.6
Treated effluent	21	7.8	2.36	14.6	1.7	2.5

Table 10. Comparison of hospital wastewater treatments by electrocoagulation process using various types of electrodes under several conditions.

Type of wastewater	Characterization of wastewater	Optimum conditions	Efficiency	Reference
Textile	pH: 11.6, COD: 800mg/L, Color: 401 mg/L, Turbidity: 105 NTU,Fe electrodes	C.D.= 8 mAcm^{-2} pH= 7.1, T: 15 min	Color%=86% 82% turbidity COD%= 59%	46
Hospital	COD: 768 mg/L, pH:7.85, Fe electrodes	C=0.5A,NaCl= 1.5 g/l, pH = 8.0,T = 120 min	COD%=92.3 %	47
Hospital	Cefazolin: 42.3 pbb, COD: 528 mg/L, Turbidity: 269 NTU, pH: 7, Al electrodes	Voltage(15 V), T: 30 min	COD%=85% turbidity=94 % cefazolin=94 %	39
Hospital	COD:398mg/L, Turbidity: 186NTU, pH= 6.2-8.3, Fe electrodes	pH 3 , 30 V,60 min	COD%= 87%	3
Hospital	COD: 807mg /l pH=8.1 Fe electrodes	pH=7.56, C.D.=4.87mA/cm^2	COD%=99.11 %	28
Hospital	COD:735 ppm, Turbidity:9.5NTU pH=7.8, Fe electrodes	Initial pH: 7.8, C.D: 25 mA/cm^2, NaCl Addition: 3 g/l. T=90 min	COD%=97.14 % SEC=(*) kWh/kgCOD)	Present work

The optimum conditions showed that the electrocoagulation process can be applied successfully for treating Al-Diwaniya hospital wastewater using iron electrodes. By starting from an initial COD (735 ppm), a COD removal efficiency of 97.14% could be accomplished at 90 min electrolysis time. In this case, a specific energy consumption of not more than 0.914 kWh/kg COD should be provided. In Table 10, a comparison between the results of the present work with the others related to hospital wastewater degradation by electrocoagulation process using iron electrode under various conditions have been achieved. Based on this Table, the EC process is efficient and suitable for treating hospital wastewater requesting only 90 min to remove approximately most of COD starting from COD of 735 mg/l with a suitable energy consumption related to previous works.

4. Conclusions

In the present study, the electrocoagulation process as a green technology was applied for treating hospital wastewater generated from Al-Dewaniya Hospital located in Al-Diwaniya city/south of Iraq. The experimental design was performed based on BBD with Response Surface Methodology. RSM was used to assess the impacts of process factors and their interactions to achieve their optimum conditions.

Under the optimum operating conditions involving a current density of 25 A/cm2, pH of 7.8, and NaCl addition of 3 g/l, the COD removal efficiency was found to be 97.14%. The results showed that a sensible operating cost of 30.914 kWh/kg COD was needed as energy consumption for COD removal from hospital wastewater. The proposed quadratic model was found to be fitted very well with the experimental data with R^2 (98.18%). Besides, results obtained in the present work confirmed the technical feasibility of the electrocoagulation method as a dependable approach for the removal of COD from hospital wastewater when the iron is used as anode material. Because of the high efficiency of EC, it could be used as an effective and economical method for the treatment of different kinds of hospital wastewater.

Authors' contribution

All authors contributed equally to the preparation of this article.

Declaration of competing interest

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References

[1] Ö. Gökkuş and Y. Ş. Yildiz, "Application of electrocoagulation for treatment of medical waste sterilization plant wastewater and optimization of the experimental conditions," *Clean Technol. Environ. Policy*, vol. 17, no. 6, pp. 1717–1725, 2015, doi: 10.1007/s10098-014-0897-2.

[2] S. A. Nurdijanto, Purwanto, and S. B. Sasongko, "PENGOLAHAN LIMBAH CAIR RUMAH SAKIT (Studi Kasus Rumah Sakit Kristen Tayu , Pati)," *J. Ilmu Lingkung.*, vol. 9, no. 1, 2011.

[3] M. Dehghani, S. Seresht, and H. Hashemi, "Treatment of hospital wastewater by electrocoagulation using aluminum and iron electrodes," *Int. J. Environ. Health Eng.*, vol. 3, no. 1, p. 15, 2014, doi: 10.4103/2277-9183.132687.

[4] E.-H. DSM, "Chapter 9—sustainability of clinical solid waste management. In: El-Haggar DSM (ed) Sustainable industrial design and waste management," *Acad. Press. Oxford*, pp. 293–306, 2007.

[5] J. Beier, S., Cramer, C., Köster, S., Mauer, C., Palmowski, L., Schröder, H. F., & Pinnekamp, "Full scale membrane bioreactor treatment of hospital wastewater as forerunner for hot-spot wastewater treatment solutions in high density urban areas," *Water Sci. Technol.*, vol. 63, no. 1, pp. 66–71, 2011.

[6] U. Tekin, H., Bilkay, O., Ataberk, S., Balta, T., Ceribasi, I., Sanin, F., Yetis, "Use of Fenton oxidation to improve the biodegradability of a pharmaceutical wastewater," *J. Hazard. Mater.*, vol. 136, no. 2, pp. 258–26, 2006.

[7] H. Kermet-Said and N. Moulai-Mostefa, "Optimization of turbidity and COD removal from pharmaceutical wastewater by electrocoagulation. Isotherm modeling and cost analysis," *Polish J. Environ. Stud.*, vol. 24, no. 3, pp. 1049–1061, 2015, doi: 10.15244/pjoes/32334.

[8] N. Kermet-Said, H., & Moulai-Mostefa, "Optimization of Turbidity and COD Removal from Pharmaceutical Wastewater by Electrocoagulation," *Isotherm Model. Cost Anal. Polish J. Environ. Stud.*, vol. 24, pp. 1049–1061, 2015.

[9] and D. Murdani, Jakfar, D. Ekawati, R. Nadira, "Application of Response Surface Methodology (RSM) for wastewater of hospital by using electrocoagulation," *IOP Conf. Ser. Mater. Sci. Eng*, vol. 345, no. 1, 2018.

[10] and G. P. T. C. E. G. Mission, P. D. Gaspillo, L. P. Belo, "Effect of Electrode Assembly on the Performance of a Compact Electrocoagulation Reactor System Applied to Pharmaceutical Residue Ibuprofen," *3rd Reg. Conf. Chem. Eng.*, pp. 1–8, 2011.

[11] E. G. Mission, P. D. Gaspillo, L. P. Belo, and G. P. T. Cruz, "Effect of Electrode Assembly on the Performance of a Compact Electrocoagulation Reactor System Applied to Pharmaceutical Residue Ibuprofen," *3rd Reg. Conf. Chem. Eng.*, pp. 1–8, 2011.

[12] M. Bajpai and S. S. Katoch, *T*echno-economical optimization using Box–Behnken (BB) design for chemical oxygen demand and chloride reduction from hospital wastewater by *electro-coagulation*, vol. 92, no. 12. 2020.

[13] B. K. Palahouane B, Drouiche N, Aoudj S, "Costeffective electrocoagulation process for the remediation of fluoride from pretreated photovoltaic wastewater," *J Ind Eng Chem*, 2014.

[14] M. Yoosefian, M., Ahmadzadeh, S., Aghasi, M., & Dolatabadi, "Optimization of electrocoagulation process for efficient removal of ciprofloxacin antibiotic using iron electrode; kinetic and isotherm studies of adsorption," *J. Mol. Liq.*, vol. 225, pp. 544–553, 2017.

[15] S. S. Bajpai, M., & Katoch, "Techno-economical optimization using Box-Behnken (BB) design for COD and Chloride reduction from Hospital wastewater by electro-coagulation," *Water Environ. Res.*, 2020.

[16] S. M. Vepsa¨la¨inen M, Kivisaari H, Pulliainen M, Oikari A, "Removal of toxic pollutants from pulp mill effluents by electrocoagulation," *Sep Purif Technol*, vol. 81, pp. 141–150, 2011.

[17] A. H. Alkurdi, S. S., & Abbar, "Removal of COD from Petroleum refinery Wastewater by Electro-Coagulation Process Using SS/Al electrodes," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 870, no. 012052, 2020.

[18] Y.-M. Wang, C.-T., Chou, W.-L., & Kuo, "Removal of COD from laundry wastewater by electrocoagulation/electroflotation," *J. Hazard. Mater.*, vol. 164, no. 1, pp. 81–86, 2009.

[19] M. D.C, "Design and Analysis of Experiments," *John Wiley Sons, New York*, 2008.

[20] R. M. . KALIL S.J., MAUGERI F., "Response surface analysis and simulation as a tool for bioprocess design and optimization," *Process Biochem*, vol. 35, no. 539, 2000.

[21] C. M. Otto, Wiley-VCH, *Chemometrics — Statistics and Computer Application in Analytical Chemistry*. 1999.

[22] S. R. . DESAI K.M., SURVASE S.A., SAUDAGAR P.S., LELE S.S., "Comparison of artificial neural network (ANN) and response surface methodology (RSM) in fermentation media optimization: Case study of fermentative production of scleroglucan," *Biochem. Eng*, vol. 41, no. 266, 2008.

[23] G. H. . IMANDI S.B., BANDARU V.V.R., SOMALANKA S.R., BANDARU S.R., "Application of statistical experimental designs for the optimization of medium constituents for the production of citric acid from pineapple waste," *Bioresour. Technol*, vol. 99, no. 4445, 2008.

[24] L. A. M. Souza, R. B. A., & Ruotolo, "Electrochemical treatment of oil refinery effluent using boron-doped diamond anodes," *J. Environ. Chem. Eng.*, vol. 1, no. 3, pp. 544–551, 2013.

[25] M. Şengil, İ. A., & özacar, "Treatment of dairy wastewaters by electrocoagulation using mild steel electrodes," *J. Hazard. Mater.*, vol. 137, no. 2, pp. 1197–1205, 2006.

[26] Murdani, Jakfar, D. Ekawati, R. Nadira, and Darmadi, "Application of Response Surface Methodology (RSM) for wastewater of hospital by using electrocoagulation," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 345, no. 1, 2018, doi: 10.1088/1757-899X/345/1/012011.

[27] D. Veli, S., Arslan, A., & Bingöl, "Application of Response Surface

Methodology to Electrocoagulation Treatment of Hospital Wastewater," *CLEAN - Soil, Air, Water*, vol. 44, no. 11, pp. 1516–1522, 2016.

[28] A. Segurola, J., Allen, N. S., Edge, M., & Mc Mahon, "Design of eutectic photoinitiator blends for UV/visible curable acrylated printing inks and coatings," *Prog. Org. Coatings*, vol. 37, no. 1–2, pp. 23–37, 1999.

[29] M. Z. Mohamed, W. S. E., Hamad, M. T. M. H., & Kamel, "Application of statistical response surface methodology for optimization of fluoride removal efficiency by Padina sp. Alga," *Water Environ. Res.*, 2020.

[30] A. Samarghandi, M. R., Nemattollahi, D., Asgari, G., Shokoohi, R., Ansari, A., & Dargahi, "Electrochemical process for 2,4-D herbicide removal from aqueous solutions using stainless steel 316 and graphite Anodes: optimization using response surface methodology," *Sep. Sci. Technol.*, pp. 1–16, 2018.

[31] Y. Zhao, Q., Kennedy, J. F., Wang, X., Yuan, X., Zhao, B., Peng, Y., & Huang, "Optimization of ultrasonic circulating extraction of polysaccharides from Asparagus officinalis using response surface methodology," *Int. J. Biol. Macromol.*, vol. 49, no. 2, pp. 181–187, 2011.

[32] L. Khataee, A. R., Zarei, M., & Moradkhannejhad, "Application of response surface methodology for optimization of azo dye removal by oxalate catalyzed photoelectro-Fenton process using carbon nanotube-PTFE cathode," *Desalination*, vol. 258, no. 1–3, pp. 112–119, 2010.

[33] G. Molla Mahmoudi, M., Khaghani, R., Dargahi, A., & Monazami Tehrani, "Electrochemical degradation of diazinon from aqueous media using graphite anode: Effect of parameters, mineralisation, reaction kinetic, degradation pathway and optimisation using central composite design," *Int. J. Environ. Anal. Chem.*, pp. 1–26, 2020.

[34] M. Yoosefian, S. Ahmadzadeh, M. Aghasi, and M. Dolatabadi, "Optimization of electrocoagulation process for efficient removal of ciprofloxacin antibiotic using iron electrode; kinetic and isotherm studies of adsorption," *J. Mol. Liq.*, vol. 225, pp. 544–553, 2017, doi: 10.1016/j.molliq.2016.11.093.

[35] M. S. Elazzouzi, M., Haboubi, K., & Elyoubi, "Enhancement of electrocoagulation-flotation process for urban wastewater treatment using Al and Fe electrodes: techno-economic study," *Mater. Today Proc.*, vol. 13, pp. 549–555, 2019.

[36] and M. D. M. Yoosefian, S. Ahmadzadeh, M. Aghasi, "Optimization of electrocoagulation process for efficient removal of ciprofloxacin antibiotic using iron electrode; kinetic and isotherm studies of adsorption," *J. Mol. Liq*, vol. 225, pp. 544–553, 2017.

[37] N. Atousa, G. K., & Fallah, "Comparison of Electrocoagulation and Photocatalytic Process for Treatment of Industrial Dyeing Wastewater: Energy Consumption Analysis," *Environ. Prog. Sustain. Energy*, vol. 13288, 2019.

[38] F. G. Esfandyari, Y., Saeb, K., Tavana, A., Rahnavard, A., & Fahimi, "Effective removal of cefazolin from hospital wastewater by electrocoagulation process," *Water Sci. Technol.*, 2020.

[39] M. R.-R. C. Barrera-Diaz, F. Urena-Nunez, E. Campos, M. Palomar-Pardave, "A combined electrochemical-irradiation treatment of highly colored and polluted industrial wastewater," *Radiat. Phys. Chem*, vol. 67, pp. 657–663, 2003.

[40] S. Liu, F., Zhang, Z., Wang, Z., Li, X., Dai, X., Wang, L., Wang, "Experimental study on treatment of tertiary oil recovery wastewater by electrocoagulation," *Chem. Eng. Process. - Process Intensif.*, p. 107640, 2019.

[41] M. KOBYA, M., DEMIRBAS, E., CAN, O., & BAYRAMOGLU, "Treatment of levafix orange textile dye solution by electrocoagulation," *J. Hazard. Mater.*, vol. 132, no. 2–3, pp. 183–188, 2006.

[42] A. K. Verma, "Treatment of textile wastewaters by electrocoagulation employing Fe-Al composite electrode," *J. Water Process Eng.*, vol. 20, pp. 168–172, 2017.

[43] M. Ahmadzadeh, S., Asadipour, A., Pournamdari, M., Behnam, B., Rahimi, H. R., & Dolatabadi, "Removal of ciprofloxacin from hospital wastewater using electrocoagulation technique by aluminum electrode: Optimization and modelling through response surface methodology," *Process Saf. Environ. Prot.*, vol. 109, pp. 538–547, 2017.

[44] C. Núñez, J., Yeber, M., Cisternas, N., Thibaut, R., Medina, P&Carrasco, "Application of electrocoagulation for the efficient pollutants removal to reuse the treated wastewater in the dyeing process of the textile industry," *J. Hazard. Mater.*, vol. 371, pp. 705–711, 2019.