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### The Treatment of Hospital Wastewater Using Electrocoagulation Process – Analysis by Response Surface Methodology

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#### ABSTRACT

Electrocoagulation (EC) can be defined a method utilized to remove pollutants from wastewater by applying an electric current to sacrificial electrodes. Many experimental variables like NaCl content (0–4 g/l), current density (5–25 mA/cm<sup>2</sup>), time (30–90 mins), and pH (4–10) that influence the removal efficiency regarding COD were considered. In the presented research, three distinct configurations related to electrodes, i.e. Al-Al, Fe-Al, and Fe-Fe, have been utilized to determine which was the most effective. RSM depending on BBD was utilized for optimizing various operational parameters with regard to HWW by use of EC. Maximum COD removal (97.9%) was reached at Fe-Al electrodes, NaCl (3.2 g/l), current density (24.7 mA/cm<sup>2</sup>), time (81.7 mins), and pH (7.4). COD removal (91.3%) was achieved at the Al-Al electrodes, NaCl (3.8 g/l), current density(23.5 mA/cm<sup>2</sup>), time-86.3 min, and Ph (7.7). At the Fe-Fe electrodes, the removal of COD (89.5%) was obtained at NaCl (2.3 g/l), current density (24.6 mA/cm<sup>2</sup>), pH 8.5, and time (86.9 min). This indicates that EC could remove pollutants from different types of wastewaters under many operating parameters and with arrangements of electrodes.

Keywords: electrocoagulation; RSM, COD removal.

#### INTRODUCTION

Hospitals are public places that play an important role for all stages of community via providing health services and constituting centers for health and research education. Hospitals are significant consumers of water and generate a considerable amount of wastewaters containing various hazardous materials (Gökkuş and Yıldız, 2015).

Hospital wastewaters (HWWs) are those effluents that are generated from different hospital activities, such as surgery rooms, radiology rooms, nursery rooms, examination rooms, laundry rooms, laboratories, kitchens and canteens (Bajpai and Katoch, 2020a). Recently, HWWs have been identified as a serious issue that may have harmful effects on the human beings and the environment, either directly or indirectly. As a result, every time higher difficulties in the case when treating prevalent diseases (Hussein Ibrahim Abdel-Shafy et al., 2019; Tahreen, Jami, and Ali, 2020). Therefore, these effluents should be treated on-site to avoid the pollution of different sectors such as the locally sewage system, watercourses and rivers. In the case of a proper treatment of HWWs, they could be reused for agricultural purposes (Beier et al., 2011; Mahajan, Khandegar, and Saroha, 2013). The pollutants in hospital wastewater involve organic compounds at high concentrations, chemical substances, such as disinfectants and solvents, and pathogenic microorganisms that resulted in spreading disease to the neighboring societies (Ekawati and Nadira, 2018; Yánes, Pinedo-Hernández, and Marrugo-Negrete, 2021). Wastewater treatment has become an absolute necessity (Hussein I Abdel-Shafy and Mohamed-Mansour, 2013). The treatment of HWWs is typically fairly complex due to the fact that each effluent has its unique features and, as a result, faces unique treatment challenges (Tekin et al., 2006). Many conventional techniques of HWWs treatment are used, such as biological and physiochemical processes (Kermet-Said and Moulai-Mostefa, 2015; Ebba, Asaithambi, and Alemayehu, 2022). Nevertheless, these methods have not the ability to treatment HWWs perfectly because of the composition and nature of these effluents. Because of the negative impacts of contaminants on community of organisms that are utilized in the biological treatment process, it was discovered that the biological treatment process for HWWs was plagued with numerous issues (Martínez et al., 2018). Therefore, further technologies have been investigated aiming for reducing the concentration of pharmaceutical contaminants. These methods consist of the separation by membrane (Sheng et al., 2016), the technology of advanced oxidation (Ghanbari et al., 2021), and electrochemical methods, such as electrocoagulation, as well as electro-flotation (Jack et al., 2014). In comparison with the conventional treatment techniques, electrocoagulation (EC) has the ability to overcome many drawbacks existing in these conventional methods. Comparative research comparing chemical coagulation (CC) and EC revealed that the former takes 20 times more mass of reagent to treat the same volume of wastewater with the same degree of efficiency. In addition, reduction of acidification of wastewater and its salinity, coagulant with low doses, and the feasibility for automating the treatment system are other advantages observed by EC in comparison with CC (Chen et al., 2020). Recently, more attention was given to the electrocoagulation (EC) as an efficient wastewater treatment method due to many features, such as its simple operation and design, minimal space required for setup, low cost and energy consumption combined with high removal efficiency, less chemicals requirement, and environment al-friendliness, since it produces little sludge with good settling ability it that can be used in hilly areas, since it requires less area (Shahedi et al., 2020). This process also has the capacity to remove contaminants, like heavy metals. The EC method was confirmed as an innovative approach for color and suspended solids removal from various wastewaters (Nidheesh et al., 2020). EC is an electrochemical management technique used sacrificial anodes to produce active coagulants. In this process, removal of pollutants from the aqueous effluents is based on many mechanisms. Dissolution of Al and production of the adsorbents (i.e. hydrated aluminum hydroxides) as an anodic

reaction occurred concurrently with development of hydrogen gas as cathodic reaction, which was responsible for the flotation of absorbents (Un, Koparal, and Ogutveren, 2009). Because of their large surface area, the produced metal hydroxides can rapidly absorb organic substances. Therefore, the generated flocs could be eliminated using gas flotation or sedimentation. When the dissolution regarding Fe and production of the adsorbents (i.e. the hydrated iron hydroxides) as an anodic reaction occurred concurrently with development of hydrogen gas as cathodic reaction, which is responsible for absorbent flotation. Throughout EC, the processes that took place at the cathode and anode surfaces and in solution have been represented by the eqs. (1, 2, 3, 4, 5, and 6) (Davarnejad, Mohammadi, and Ismail, 2014; Can, Gengec, and Kobya, 2019):

Cathode reaction when Al electrode used

$$3H_20 + 3e \rightarrow 3/2H_2(g) + 30H^-$$
 (1)

Anode reaction:

$$Al \to Al^{+3} + 3e^{-} \tag{2}$$

In the solution:

$$Al^{+3}_{(aq)} + 3H_2 0 \rightarrow Al(OH)_3 + 3H^{+}_{(aq)}$$
 (3)

Anodic reaction:

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

Cathodic reaction:

$$3 H_2 0 + 3e \rightarrow 3/2 H_2(g) + 30 H^2$$
 (5)

In solution:

$$Fe^{3+}_{(aq)} + 3H_2 0 \rightarrow Fe(OH)_3 + 3H^+_{(aq)}$$
 (6)

Two interaction mechanisms (precipitation and adsorption) occurred between contaminants and hydrolysis products during the electrocoagulation process. Adsorption takes place at high pH ranges (6.5), but flocculation is indicated as precipitation at low pH ranges (López-Maldonado et al., 2014). The EC effluent could be used for irrigation and industrial purposes (Fathy et al., 2020). EC is suited for a broad range of the wastewater treatment plants, which include the dairy (Yavuz and Ögütveren, 2018), textile (Şengil, 2006), laundry wastewater (Shivayogimath and Jahagirdar, 2013), sugar industry (Dimoglo et al., 2019), HWW (Ramcharan and Bissessur, 2017), and COD removal from petroleum refinery wastewater (Yan et al., 2014). The purpose of HWW treatment is preventing health problems and environmental

pollution. Yet, earlier research did not use Response Surface Methodology (RSM) to evaluate COD in HWW. RSM is chosen since it may optimize a process by examining the impact of variables on performance (Ghafari et al., 2009; Fahim and Abbar, 2020; Kumar, Singh, and Kumar, 2018; Mosayebi, 2021; Mohammed et al. 2020).

This study seeks to treat total suspended solids and improve them statistically through RSM utilizing BBD and Design Expert 13 software. BBD was not utilized to measure the optimum COD removal. Four significant variables (current density, contact time, NaCl concentration, and pH) are taken into account; COD removal is specified as a response utilizing an electrochemical cell with aluminum electrodes one time and iron electrodes the second time. Variables are altered in accordance with BBD and RSM. Modeling the influence of variables on COD performance employs the RSM and BBD. To find the optimum response, the variable interactions are investigated. The impacts of time and current density on Fe and Al anode materials in an electrochemical cell were evaluated individually. Under optimal conditions, the COD removal efficiency of both electrodes was greater than 99%, according to their findings.

#### **EXPERIMENTAL WORK**

Prior to mixing with the city's domestic wastewater, 20 l of HWW has been collected from the sewage system of Al-Dewaniya hospital

**Table 1.** The characteristics of the wastewater fromAl-Dewaniya hospital sewage system

Parameters	Values		
рН	6.5		
TSD (mg/l)	2500		
COD (mg/l) 745	710–745		
SO <sup>-2</sup> <sub>4</sub> (mg/l)	500–600		
Cl- (mg/l)	1.55		
Turbidity (NTU)	10		

(which is located in Al-Dewaniya city, Iraq) for this investigation. The properties of HWW are listed in Table 1. Throughout the experimental program, this HWW has been stored at 4 °C, and 1-liter samples were collected at the time of every one of the experiments.

For EC treatment studies, a Perspex glass labscale batch electrochemical reactor with a working volume of 1.25 l (width of 120 mm × length of 120 mm × height of 120 mm) and a Perspex cover (width of 150 mm × length of 150 mm × thickness of 28 mm) have been utilized. The cover has slits for fixing electrodes and holes for the insertion of the pH-meter and conductivitymeter probes. Three plat cathodes (length of 130 mm × width of 80 mm × thickness of 3 mm) and two anodes (length of 130 mm × width of 80 mm × thickness of 3 mm) depending on mode running (Fe-Fe, Fe-Al, and Al-Al) have been used in an electrochemical reactor with a parallel plate layout. The inter-electrode gap was maintained at



**Figure 1.** The electrochemical system:1) electrochemical cell; 2) magnetic stirrer; 3) power supply; 4) ammeter; 5) voltmeter; 6) cathode; 7) anode; 8) pH-meter

15 mm. A digital power supply of direct current, Type (UNI-T, UTP3315PF), has been utilized so as to maintain a constant current during the process. The test solution was properly mixed using a magnetic stirrer before being placed in the cell. The experimental setup for EC is depicted in Figure 1, together with the supporting information. The pH of the electrolyte has been evaluated with a digital pH meter (HNNA Instrument Inc, PH-211, Romania), and acidity regarding electrolyte has been modified with NaOH or HCl to meet experimental requirements. Cathodes and anodes have been rinsed with water and ethanol for removing impurities before each run. At the conclusion of each run, samples have been filtered and their COD values were analyzed to determine the process performance.

To estimate COD, a sample (2 ml) related to effluent has been digested with  $K_2Cr_2O_7$  for 120 min at a temperature of 150 °C in COD thermosreactor (RD 125, Lovibond) before being cooled to room temperature and measured with a spectrophotometer (MD200, Lovibond). The COD removal efficiency has been estimated with the use of Equation 7 (Alkurdi and Abbar, 2020):

$$\text{RE\%} = \frac{Ci - Cf}{Ci} * 100 \tag{7}$$

where:  $C_f$  represents the final COD (mg/l);  $C_i$  represents the initial COD (mg/l).

Specific Energy Consumption (SEC) can be defined as quantity of energy that is required for digesting a kg of COD. SEC in (kWh/kg) could be obtained using Eq. 8 (Fahim and Abbar, 2020):

$$EC = \frac{U.I.t * 1000}{(Ci - Cf)V}$$
(8)

where: *EC* – represents the energy consumption (kWh/kg COD);

U-represents applied cell voltage (Volts), t - represents electrolysis time (hours), I-represents current (A),

 $C_f$  and  $C_i$  – represents final and initial COD values (mg/l),

*V* – represent effluent volume (L).

#### **Experimental design**

BBD was used to create the design matrix having three levels (-1, 0, and 1) to observe the influence of four factors (time, pH, current density, and NaCl) on the efficiency of COD removal via EC. The suggested design provides 25 experimental combinations upon which ANOVA modeling and statistical analysis were carried out. In the experiment, the current density has been maintained between 5 and 25 mA/cm<sup>2</sup>, the pH range was maintained between 4 and 10, the electrolysis time was adjusted between 30 and 90 minutes, and the addition of NaCl to the solution was maintained between 0 and 4 g/l. Table 2 displays the values of chosen variables with their set EC process levels. As stated in Equation 9, the acquired experimental results are consistent with the quadratic model (2nd order polynomial).

 $Y = \beta 0 + \sum \beta i xi + \sum \beta ii xi 2 + \sum \beta ijxixj$ (9)

where: RE% – indicated as Y;

*j* and *i* – denotes patterns index numbers;  $\beta 0$  – represents intercept term, *x*1, *x*2 ...; *xk* – have been coded form regarding process variables;  $\beta i$  – denotes the 1<sup>st</sup>-order (i.e. linear)

 $\beta i$  – denotes the 1<sup>st</sup>-order (i.e. linear) main effect;

 $\beta ii$  – denotes the 2<sup>nd</sup>-order main effect,

 $\beta ij$  – denotes the interaction effect.

After doing ANOVA, the regression coefficient (R<sup>2</sup>) has been determined in order to assess the model's goodness of fit. A CCD has been done for 5 independent variables, and DoE has been utilized in order to reduce the number of the required runs and in order to aggregate the individual independent variables. The selected parameters are NaCl concentration (B), current density (A), pH (C) and time (D),. The coded and actual values of variables are shown in Table 3, whereas Tables 2 and 3 disclose an experimental design matrix with 45 coded conditions regarding Al–Al electrode combination. In Table 2, the real values represent the initial values assigned to various factors, and the coded values are also provided by default or can be modified for the levels of factors. Coded and actual factors are variables and A, B, C, and D, in this instance.

**Table 2.** Actual and coded values of variables of the design of the experiments for the EC

Variables	Linit	Codoo	Levels			
Valiables	Unit	Codes	+1	0	-1	
Current density	mA/cm <sup>2</sup>	А	5	15	25	
NaCl <sub>conc.</sub>	g/l	В	0	2	4	
рН	-	С	4	7	10	
Time	min	D	30	60	90	

#### **RESULTS AND DISCUSSION**

#### Removal efficiency of COD and energy consumption

A total of 25 batch runs have been carried out at different process variable combinations to examine the combined effects of independent factors on COD removal efficiency. The experimental results are shown in Table 3 as Specific Energy Consumption (SEC) and COD Removal Efficiency (RE%). As a result, when it is operating in the Fe-Al mode, the COD removal efficiency ranges from 71.3 to 97.67% as could be seen, and the energy consumption range is (4.1-33.1)KWh/kg COD. However, in the case when operating in Al-Al mode, the efficiency of the COD removal ranges from (39.7 to 89.3%), whereas energy consumption (39.3–2.2). In the case when the third scenario is operating in the mode (Fe-Fe), COD removal efficiency ranges between

(63.4 and 93.6%). The energy consumption range is (1.9–29.4) kWh/kg COD.

In Table 3, which depends on the case study involving the combination of Al-Al electrodes, Fe-Al electrodes, and Fe-Fe electrodes together with their respective predicted values from RSM, the removal efficiency regarding COD with energy consumption was displayed. Table 3, column 1, 2, 3, 4, and 5 demonstrate that a number of the runs or the tests, which were carried out in a laboratory, specify current density (A), NaCl (B), pH (C), and time (D), respectively. In order to make the COD removal from HWW easier, NaCl has been employed as electrolytic concentration. The remaining columns show the actual COD% and energy consumption (kWhr/m<sup>3</sup>) from the lab, as well as the RSM-predicted value. Table 3 shows that variables like current density, pH, reaction time, and NaCl which were taken into account for three different electrode combinations. Comparably, all aspects were taken into account to

Table 3. Energy consumption and removal percentage with predicted vs. and actual values

	Α	В	С	D	COD r	COD removal, % (Fe-Al)		COD removal, % (Al-Al)			COD removal, % (Fe-Fe)		
Run	$\frac{mA}{cm^2}$	g/l	-	Min	Act. value	Predicted value	E.C.	Act. value	Predicted value	E.C.	Act. value	Predicted value	E.C.
1	15	0	4	60	75.32	76.18	14.56	39.78	39.92	27.57	63.42	64.00	17.30
2	15	2	7	60	87.66	87.66	12.51	69.15	70.40	15.86	84.56	84.65	12.97
3	5	0	7	60	75.55	75.33	4.31	62.56	62.93	5.21	80.89	81.15	4.03
4	15	2	4	90	81.32	83.14	20.23	80.12	81.60	20.54	82.59	82.36	19.92
5	15	2	10	30	76.25	75.06	7.19	54.02	55.23	10.15	73.058	72.36	7.51
6	25	0	7	60	89.55	88.91	24.12	63.35	64.48	34.09	73.23	72.57	29.49
7	15	0	7	90	87.03	86.09	18.90	77.75	78.24	21.16	85.22	86.23	19.31
8	25	2	10	60	90.88	90.35	23.76	82.63	83.04	26.14	90.32	91.37	23.91
9	15	4	7	30	79.99	79.61	6.86	43.44	42.86	12.62	66.78	66.41	8.21
10	15	4	4	60	83.12	82.05	13.20	68.89	67.77	15.92	75.67	75.89	14.50
11	15	2	10	90	89.5	90.19	18.38	64.56	63.97	25.48	81.88	81.29	20.09
12	25	2	7	90	97.67	97.33	33.17	89.35	88.22	36.26	93.68	93.68	34.58
13	15	0	7	30	76.89	76.05	7.13	54.37	53.42	10.09	76.06	75.69	7.21
14	5	2	4	60	74.23	73.44	4.39	76.78	76.06	4.25	80.59	80.34	4.04
15	25	2	7	30	84.36	85.79	12.80	66.96	65.98	16.13	75.69	75.58	14.27
16	5	4	7	60	79	80.27	4.13	78.31	77.55	4.16	85.78	85.79	3.80
17	5	2	7	90	84.35	83.62	5.80	50.78	51.05	9.63	73.21	73.42	6.68
18	5	2	7	30	71.3	72.34	2.29	72.98	72.68	2.23	83.98	84.09	1.94
19	25	2	4	60	88.02	87.26	24.54	54.73	55.12	39.46	75.93	75.84	28.44
20	25	4	7	60	93	93.85	23.22	82.85	82.66	26.07	87.21	87.02	24.76
21	5	2	10	60	77.57	77.01	4.20	51.48	51.38	6.33	66.74	67.39	4.88
22	15	0	10	60	78.68	80.45	13.94	65.56	65.01	16.73	72.56	73.42	15.12
23	15	4	10	60	84.61	84.44	12.96	68.04	68.68	16.12	83.17	82.33	13.19
24	15	2	4	30	75.5	75.44	7.26	89.08	89.26	6.16	91.78	91.15	5.98
25	15	4	7	90	92.89	92.4	17.71	71.05	71.05	23.16	82.02	82.03	20.06

establish the removal efficiency for COD and energy use. EC is well acknowledged to be highly dependent upon the value of the pH of the starting wastewater. The initial pH regarding wastewater has an impact on the performance of the EC as well as the pH related to aqueous solution affects the formation of metallic hydroxides (Bener et al., 2019; Dalvand et al., 2011). The removals efficiency was enhanced when the initial HWW's pH was raised, as shown in Table 3. The current intensity has a great impact on EC. Due to anodic dissolution based on Faraday's law, the efficiency of the removal increased both at increasing current levels and as the current intensity rose (Khan et al., 2019). Since the rate at which EC and gas bubbles are generated significantly affect the rate at which flocs develop, the effect of applied current on studied processes is particularly crucial (AlJaberi, Ahmed, and Makki, 2020). It must be taken into account in any EC approach for wastewater treatment, since it controls the amount of Fe and Al ions discharged from the electrodes, along with the release of gas bubbles and the formation of flocs (Asaithambi and Govindarajan, 2021). The removal of COD for 3 cases rose as the electric current was raised from 5 to 25 mA/ cm<sup>2</sup>, as indicated in Table 3. The low cost and availability of sodium chloride led to its selection as a supporting electrolytic. Pollutant removal efficiency for the process of wastewater treatment is significantly impacted by the electrolytic concentration regarding waste-water in an electro-chemical process (Abbasi et al., 2020). Using highly conducting solution and supporting electrolyte for treating strong wastewater has a number of benefits, including preventing migration effects, reducing electrode resistance, boosting solution conductivity, improving process effectiveness, and reducing energy consumption (Abbasi et al., 2022; Tak et al., 2015). The protective layer of double coagulant, kinetic electro-dissolution regarding sacrificial anodes, and the shape of flocs are all significantly influenced by the electrolytic concentration (Camcioglu, Ozyurt, and Hapoglu, 2017). Table 3 demonstrates that as the electrolytic concentration has been raised, the efficiency related to COD removal from HWW increased. The ability of electrodes to produce sufficient amounts of different ions, like Al(OH), at the case of the electrodes that have been made of the aluminum, which are necessary for the production of adsorbents. Electrolysis time affects the release of gas bubbles from the two electrodes,

which are effectively given greater aid to move the destabilized contaminants toward the solution's surface(Bajpai and Katoch, 2020b). Electrolysis time has a direct impact on the amount of Fe and Al that had been released from the electrodes, which in turn affects the amount of Al and Fe released from the anode and determines the efficiency of COD removal (Deveci et al., 2019). Ions will have a better chance of colliding and creating OH° (Shokoohi et al., 2020). As Table 3 indicates, it shows that even though the electrolysis time was increased from 30 to 90 minutes, the removal efficiency regarding COD from the two electrode combinations was high. Additionally, the distinctions between the electrodes allow for the recognition of COD removal. Moreover, 2 cm was the ideal distance between two electrodes (Liu et al., 2019). The impact of process factors as well as their combination on efficiency of COD removal has been investigated with the use of graphical demonstrations that are related to the statistical optimization depending upon RSM for case one. The combined effects of the NaCl concentration and current density on COD removal efficiency at a constant pH and time are shown in Figures (2-a). Figure 2-a shows that over the whole NaCl concentration range (0-4 g/l), an increase in current density leads to an increase in COD removal efficiency. Accordingly, the increase of current density from 5 mA/cm<sup>2</sup> to 25 mA/cm<sup>2</sup> causes a large increase in COD removal from 75.55% to 89.55% at NaCl  $_{\text{conc.}} = 0$  (Table 3, Exp. 3 and 6), whereas the rise in COD removal efficiency remained almost the same at NaCl \_\_\_\_\_= 4 from 80.27 to 93.85%. (Exp.16 and 20, Table 3). In the case where the EC process has been carried out by the use of an aluminum electrode, it has been found that the current density had a maximum impact on COD removal efficiency. On the basis of Faraday's law (El-Khateeb et al., 2017), the formation of coagulants  $(Al(OH)_{2})$ particles) at the anode increases along with the dissolution rate of the aluminum anode. This is how the results may be explained. A rise in current density causes the generation rate of hydrogen gas bubbles to increase while the size of the bubbles decreases, which has a positive influence on the removal of pollutants via floatation (Elazzouzi, Haboubi, and Elyoubi, 2019). As can be seen in Fig. 2-a, the COD removal efficiency increases along with the NaCl concentration. As an illustration, raising the NaCl concentration from 0 to 4 at a 5 mA/cm<sup>2</sup> current density led to an

increase in the COD removal of 75.55% to 79% (Table 3, exp. 3 and 16). At higher values of the current density, however, such increase in the efficiency of COD removal had been decreased considerably. Literature reviews indicate that lowering the cell voltage through adding NaCl to EC might increase efficiency by reducing process energy usage. Additionally, the opposite effects of anions like HCO<sub>3</sub>,  $CO_3^{2-}$ , and  $SO_4^{2-}$  may be prevented when there is a NaCl electrolyte present. These anions raise the ohmic resistance regarding EC cell by precipitating Mg<sup>2+</sup> or Ca<sup>2+</sup> cations as an insulating layer on cathode surface. The next reactions also occur when NaCl is added to the electrochemical process:

$$2 \operatorname{Cl}^{-} \rightarrow \operatorname{Cl}_{2} + 2 \operatorname{e}^{-}$$
(10)

$$Cl_2 + H_2O \rightarrow HOCl + Cl^- + H^+$$
(11)

$$HOCI \to OCI^- + H^+ \tag{12}$$

According to reactions 10 and 11, Cl<sup>-</sup> could oxidize to  $Cl_2$ , a potent oxidant which might help in oxidation regarding dissolved organic compounds, or it might result in creation of the HOCl, a potent oxidizer which could lead to extra COD removal along with EC. Other research revealed comparable results (Najeeb and Abbar 2022; Ebba, Asaithambi, and Alemayehu 2021).

At constant current density and NaCl concentration, Figure (2-b) illustrates the combined impacts of pH and time on COD removal efficiency. In the case when the time from 30 to 90 minutes at pH 7 is increased, it is seen that the COD removal efficiency will rise from 71.3% to 84.35% at constant current density =  $5 \text{ mA/cm}^2$ , among other observations. COD removal efficiency rises with increasing pH, as seen in Fig. 2-b. As an illustration, increasing pH from 4 to 10 at a 5 mA/ cm<sup>2</sup> current density causes COD removal to increase from 74.2% to 77.5% (Table 3, exp. 14 and 21). At larger current densities, however, this increase in the efficiency of COD removal has been decreased considerably. It is obvious that the pH values between 4 and 7 have a greater impact on COD removal efficiency than pH values between 7 and 10. The following could be done to stop this behavior. When the pH is acidic, there are not enough hydroxyl ions present, which can



**Figure 2.** Response surface plot for COD removal (Fe-Al): a) the interaction between  $\text{NaCl}_{\text{conc.}}$  and current density, b) the interaction between pH and time, c) interaction between pH and  $\text{NaCl}_{\text{conc.}}$ , d) the interaction between time and current density

Source	Summation of the squares	Df	Mean square	F-Value	P-Value		
Model	1152.57	14	82.33	40.02	< 0.0001		
A-current density	553.25	1	553.25	268.94	< 0.0001		
B-NaCl conc.	72.96	1	72.96	35.47	0.0001		
C-pH	33.27	1	33.27	16.17	0.0024		
D-time	390.68	1	390.68	189.91	< 0.0001		
AB	0	1	0	0	1		
AC	0.0576	1	0.0576	0.028	0.8704		
AD	0.0169	1	0.0169	0.0082	0.9296		
BC	0.8742	1	0.8742	0.425	0.5292		
BD	1.9	1	1.9	0.9257	0.3587		
CD	13.8	1	13.8	6.71	0.0269		
A <sup>2</sup>	2.39	1	2.39	1.16	0.3068		
B <sup>2</sup>	13.06	1	13.06	6.35	0.0304		
C <sup>2</sup>	63.15	1	63.15	30.7	0.0002		
D <sup>2</sup>	10.99	1	10.99	5.34	0.0434		
Residual	20.57	10	2.06				
Cor total	1173.14	24					
Model Summary Statistics							
R²	Adjusted R <sup>2</sup>	Standard deviation	Coefficient of variation%	Adeq precision	Mean		
0.9825	0.9579	1.43	1.73	22.495	82.97		

Table 4. ANOVA of quadratic model and Summary Statistics for % COD removal (Fe-Al)

lead to very low  $Al(OH)_3$  production, which can lead to a loss in efficiency. Additionally, aluminum hydroxide particles are soluble at low levels of the pH (< 7), which prevents them from having the capacity to absorb pollutants. At pH 7, the major compounds that can adsorb the pollutants are insoluble  $Al(OH)^{2+}$ ,  $(OH)_3$ ,  $Al_3(OH)_3^{7+}$ , and  $Al_2(OH)_2^{4+}$ .  $Al(OH)^{4-}$ , which is soluble in water and forms at high pH values, reduces the removal efficiency, especially at pH values > 10.



Figure 3. Predicted vs. actual values for % COD removal (Fe-Al)

In the second case, where Al-electrodes cathode and anode were used, the efficiency if COD removal was in the range (39.7-89.3%) compared with the case when that efficiency of COD removal was (71.3–97.6%); this results take same parameters but different between in a range, In the first case, more parameters effect not only operation parameters the rate for  $Al(OH)_3$ ,  $Fe^{+3}$ ,  $Al_2(OH)_2^{4+}$ ,  $Al(OH)^{2+}$  and  $Al_{13}(OH)_{3}^{27+}$  all the can be effect on efficiency that it will improved the efficiency, so its share above. Its mean can get on efficiency removal at required range when the parameters working at a high value and some middle value for it. When take third case when Fe-Fe running can show in Table 3 can observe the range from (63.4–93.6%) and more Fe loss, additionally for all parameters effect on COD efficiency removal the chemical potential energy.

The combined effects of NaCl concentration and current density on COD removal efficiency

in the case when time and pH are constant are shown in a third instance can be seen in Figure 9. The response surface plot demonstrates that for NaCl concentrations between 0 and 4 g/l, increasing the level of the current density enhances COD removal efficiency. Thus, we observe when increasing the level of the current density from 5 mA/cm<sup>2</sup> to 25 mA/cm<sup>2</sup> increase in COD removal from 75.3–88.9% at NaCl = 0 (exp.3&6), and a same increase approximately in COD removal efficiency from 79 to 93% was occurred at NaCl = 4 g/l (exp.16 & 20). We show the increasing in NaCl<sub>cone</sub> that effect on COD removal increased from 75.55 to 79% (exp.3&6) (Table 6, exp. 7 and 14) compared to no additions.

#### Validity of the model

RSM is a specific set of statistical and mathematical techniques which can be used for model



**Figure 4.** Response surface plot for COD removal (Al-Al): a) the interaction between NaCl<sub>conc</sub> and current density, b) interaction between time and pH, c) interaction between pH and NaCl<sub>conc</sub>, d) interaction between time and current density

Source	Summation of squares	Df	Mean square	F <sub>Value</sub>	$P_{_{Value}}$			
Model	4359.98	14	311.43	216.97	< 0.0001			
A-current density	1812.78	1	1812.78	1262.97	< 0.0001			
B-NaCl	877.74	1	877.74	611.53	< 0.0001			
С-рН	148.05	1	148.05	103.15	< 0.0001			
D-time	1295.22	1	1295.22	902.39	< 0.0001			
AB	34.87	1	34.87	24.29	0.0006			
AC	8.76	1	8.76	6.1	0.0331			
AD	0.1089	1	0.1089	0.0759	0.7886			
BC	30.58	1	30.58	21.31	0.001			
BD	12.11	1	12.11	8.44	0.0157			
CD	4.95	1	4.95	3.45	0.0929			
A <sup>2</sup>	73.62	1	73.62	51.29	< 0.0001			
B²	14.06	1	14.06	9.79	0.0107			
C²	0.9053	1	0.9053	0.6308	0.4455			
D²	0.1576	1	0.1576	0.1098	0.7472			
Residual	14.35	10	1.44					
Cor total	4374.33	24						
Model Summary Statistics								
R <sup>2</sup>	Adjusted R <sup>2</sup>	Standard deviation	Coefficient of variation%	Adeq precision	Mean			
0.9967	0.9921	1.2	1.78	53.1759	67.14			

Table 5. ANOVA of the quadratic model and Summary Statistics for COD removal (Al-Al)

validation, model fitting, and optimization (Bezerra et al., 2008). The RSM seeks to maximize the interest response, which is affected by a number of factors (Hill and Hunter, 1966). The RSM can be defined as a practical statistical approach for optimizing chemical and/or industrial processes, and it is frequently utilized for experimental design. Using this approach, response surface, which is influenced by process parameters (Dean, Voss, and Draguljić, 2017), is optimized. The quadratic model's ANOVA



Figure 5. Predicted vs. actual values for COD removal (Al-Al)

Sources	Summation of squares	DF	Mean square	F <sub>Value</sub>	$P_{_{Value}}$		
Model	1483.2	14	105.94	160.15	< 0.0001		
A-current density	358.39	1	358.39	541.79	< 0.0001		
B-NaCl	165.44	1	165.44	250.1	< 0.0001		
С-рН	21.43	1	21.43	32.39	0.0002		
D-time	800.53	1	800.53	1210.17	< 0.0001		
AB	94.48	1	94.48	142.82	< 0.0001		
AC	0.065	1	0.065	0.0983	0.7603		
AD	2.12	1	2.12	3.2	0.1039		
BC	7.71	1	7.71	11.65	0.0066		
BD	1.95	1	1.95	2.94	0.1171		
CD	6.07	1	6.07	9.18	0.0127		
A²	7.42	1	7.42	11.22	0.0074		
B²	15.82	1	15.82	23.91	0.0006		
C²	0.2729	1	0.2729	0.4126	0.5351		
D <sup>2</sup>	3.33	1	3.33	5.03	0.0488		
Residual	6.62	10	0.6615				
Cor total	1489.82	24					
Model summary statistics							
R <sup>2</sup>	Adjusted R <sup>2</sup>	Standard deviation	Coefficient of variation%	Adeq precision	Mean		
0.9956	0.9893	0.8133	1.02	47.1104	79.44		

Table 6. ANOVA of quadratic model and summary statistics for COD removal (Fe-Fe)



**Figure 6.** Response surface plot for COD removal (Fe-Fe): a) the interaction between NaCl<sub>cone.</sub> and current density, b) interaction between time and pH, c) interaction between pH and NaCl<sub>cone.</sub>, d) interaction between time and current density



Figure 7. Actual versus Predicted values for COD removal (Fe-Fe)

results and summary statistics for the percent of COD are shown in Tables 4, 5 and 6. This model has been significant for the process of COD removal because the value of p < 0.005 that indicates that the model has been significant at a 95% probability level. The model's output shows that for COD removal (Fe-Al), 0.9967 and 0.9921, 0.9967 and 0.9921 for COD removal (A-Al), and 0.9956 and 0.9893 for COD removal (Fe-Fe), respectively, the coefficient of determination (R<sup>2</sup>) and adjusted determination coefficients (R<sup>2</sup>). The interaction of NaCl concentration, current density, electrolysis time, and pH impacts on COD removal in the three cases, according to ANO-VA results. The coefficient of determination  $(R^2)$ expresses the quality of the fit, and the p-value and f-value are key metrics which show how appropriate and significant the models are [65]. Table 3 displays experimental (i.e. actual) as well as the predicted values for COD and energy usage. Figures 1, 2, and 3 show the experimental data with all points close to the diagonal line and model predictions that agreed with these values. The quadratic models could be utilized for predicting the percent of COD removal because they were demonstrated to be significant (P less than 0.005) in ANOVA analysis.

## Experiment performance analysis utilizing DoE

The operating variables NaCl concentration (B), current density (A), time (D), and pH (C) are

used to express the percent removal efficiency of COD. In Eqs. (9), (10), and (11) for COD removal (Fe-Al), COD removal (Fe-Al), and COD removal (Fe-Fe), the DoE offered the quadratic model regression.

$$RE\% (Fe-Al) = 87.66 + 6.79A + 2.47B + + 1.67C + 5.71D + 0.0AB - 0.12AC + + 0.065AD - 0.467BC + 0.69BD + + 1.86CD - 0.919A2 - 2.15B2 - - 4.73C2 - 1.97D2$$
(13)

RE% (Al-Al) = 71.05 + 12.29A + 8.55B +  
+ 
$$3.51C + 10.39D - 2.95AB + 1.48AC -$$
  
-  $0.165AD - 2.76BC + 1.74BD -$  (14)  
-  $1.11CD - 5.11A^2 - 2.23B^2 -$   
-  $0.566C^2 - 0.236D^2$ 

$$RE\% (Fe-Fe) = 82.03 + 5.47A + 3.71B + + 1.34C + 8.17D - 4.86AB + 0.127AC + + 0.727AD + 1.39BC + 0.697BD + (15) + 1.23CD - 1.62A2 - 2.37B2 - - 0.312C2 - 1.09D2$$

#### **Optimization with RSM**

Obtaining optimum conditions based on laboratory experiments is one of the most significant advantages and the most important reason for going to RSM analysis methodology, specifically BBD. On the basis of the BBD and by using the regression equation, the results



Figure 8. Optimization of experimental parameters based COD removal (Fe-Al)



Figure 9. Optimization of experimental parameters based COD removal (Al-Al)



Figure 10. Optimization of experimental parameters based COD removal (Fe-Fe)

are improved. The Design-Expert-13 software searches the design space in an effort to improve the process while taking into account a number of limitations. Multiple randomly selected starting points are picked in order to find the genuine maxima or minima. Each of the process variables and response variables needs to have a target set in advance. The answer options (Anderson and Whitcomb 2016) are minimize, maximize, within range, target, and none. Additionally, factors may be set to a precise value. The responses, like COD removal efficiency, have been maximized when current density (A) was optimized along with pH (C), NaCl concentration (B), and time (D).

Depending on these operating parameters, a slight variance was observed in the optimal values for the three cases, where it was at NaCl concentration -3.2 g/l, 7.4 pH, current density -24.7 mA/cm<sup>2</sup>, and time -81.7 min for Fe-Al electrodes, current Density -23.5 mA/cm<sup>2</sup>, NaCl concentration -3.8 g/l, 7.7 pH, and time -86.3 min for Al-Al electrodes, and current Density -24.6 mA/cm<sup>2</sup>, NaCl concentration -2.3 g/l, 8.5 pH, and time -86.9 min for Fe-Fe electrodes.

#### CONCLUSIONS

Large amounts of water are provided by the hospital for all activities, and therefore, wastewater is produced. With no treatment, which affects the state of natural environment, water is subsequently consumed and released into the environment as waste. With the use of various types of electrodes, an EC is one of the efficient technologies utilized for treating HWW. The outcomes demonstrated that utilizing Al-Al, Fe-Al, or Fe-Fe electrodes and varying conditions like concentration of NaCl (0-4 g/l), current density (5-25 mA/cm<sup>2</sup>), electrolysis time (30-90 mins), and pH (4-10), it is effective to remove COD from HWW. The research found that greater pollution removal percentages may be attained with less energy use. RSM was used to provide the best value through maximizing COD removal efficiency. Additionally, RSM examines the statistical modeling regarding an experiment and displays the predicted value depending on actual value acquired through laboratory analysis. Lastly, the findings of this research indicated that the EC process could be a successful and effective treatment to treat industrial and wastewater effluent.

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