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Experimental and Theoretical Investigation of Chemical Oxygen Demand Reduction in Industrial Petroleum Wastewater Using Electrocoagulation

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Abstract

Industrial petroleum wastewater often contains high levels of turbidity and chemical oxygen demand (COD), making treatment challenging and costly. In this study, electrocoagulation (EC) was investigated as an effective and potentially cost-efficient treatment method for oily wastewater. The process was optimized using Response Surface Methodology (RSM) based on a Central Composite Design (CCD). Three independent variables—current density (20–50 mA/cm²), pH (2–9), and treatment time (10–120 min)—were evaluated, with COD and turbidity removal efficiencies as the response variables. Analysis of variance (ANOVA) was used to assess the significance and adequacy of the developed models. Experimental results showed that COD and turbidity removal efficiencies reached 93% and 96.5%, respectively, within the investigated operating range. The results indicate that current density, pH, and treatment time substantially affect EC performance, with coagulation, adsorption, coprecipitation, and flotation contributing to pollutant removal. Overall, the optimized EC process demonstrated strong potential for reducing COD and turbidity in industrial petroleum wastewater and improving the feasibility of wastewater treatment.

Keywords: COD, Turbidity, Electrocoagulation, Response Surface Methodology

1. Introduction

Electrocoagulation (EC) has attracted considerable attention for industrial wastewater treatment because it can generate coagulants in situ and requires relatively simple equipment and operation [1, 2]. Recent studies have demonstrated the effectiveness of EC for reducing COD, suspended solids, color, and heavy metals in different industrial wastewaters [3–8]. Unlike conventional chemical coagulation, EC produces metal ions electrochemically from sacrificial electrodes, which can promote charge neutralization, adsorption, coprecipitation, and flotation. These mechanisms can be particularly useful for treating petroleum and refinery wastewaters containing emulsified oil and suspended matter. Treatment performance is strongly influenced by operating parameters such as current density, pH, electrode material, electrolysis time, and inter-electrode distance [9–12]. Machine-learning studies have also shown that electrolysis time, inter-electrode distance, and current

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density can be among the most influential variables controlling COD removal [13]. For example, an aluminum-electrode EC reactor achieved high COD and color removal from highly contaminated greywater under optimized conditions [14]. Despite these advances, further experimental and thermodynamic analysis is needed to clarify the relationships among operating conditions, aluminum speciation, and pollutant-removal mechanisms in real petroleum wastewater. Therefore, the present study investigates COD and turbidity removal from industrial petroleum wastewater using aluminum and stainless-steel electrodes and optimizes the effects of current density, pH, and treatment time using RSM and CCD.

1.1. Electrocoagulation principles

Consider two reactants, A and B, that undergo a reaction involving the transfer of n electrons, as represented by Eq. (1) [15–17].



The parameters a , b , c , and d represent the stoichiometric coefficients in Eq. (1). The Gibbs free-energy change for the reaction is given by Eq. (2).

$$\Delta G = \Delta G^0 + RT \ln([A]^a [B]^b [H_2O]^c [H^+]^d) = \Delta G^0 + RT \ln K \quad (2)$$

In these equations, ΔG is the Gibbs free-energy change; ΔG^0 is the standard Gibbs free-energy change at $T = 298.15$ K and atmospheric pressure; R is the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$); T is the absolute temperature (K); and K is the equilibrium constant.

For a spontaneous reaction, $\Delta G < 0$ and $K > 1$. The cell potential, E , is related to the Gibbs free-energy change per unit charge transferred in an electrochemical reaction.

$$\Delta G = -nFE \quad (3)$$

- n : number of electrons transferred in the electrochemical reaction.
- F : Faraday constant.
- $F = 96,485 \text{ C mol}^{-1}$; E denotes the cell potential.

The theoretical amount of aluminum dissolved from the anode can be estimated from Faraday's law using Eq. (4).

$$m = \phi \frac{ItM}{nF} \quad (4)$$

In Eq. (4), m is the mass of aluminum dissolved at the anode (g), I is the applied current (A), t is the electrolysis time (s), M is the molar mass of aluminum (g mol^{-1}), n is the number of electrons transferred, and F is the Faraday constant. The actual aluminum dissolution may differ from the theoretical value because of current efficiency, side reactions, electrode passivation, and mass-transfer effects. Electrical energy supplied to the cell can also be partly converted into heat through ohmic losses, causing a temperature increase. Supporting electrolyte addition, such as NaCl or Na_2SO_4 , can increase solution conductivity and reduce ohmic losses; however, the effect of chloride ions on possible chlorine formation should also be considered. Previous studies have reported deviations between experimental and theoretical electrode dissolution [18–20]. Pourbaix diagrams are useful for interpreting the thermodynamic stability of aluminum species as a function of potential and pH and for relating electrochemical equilibria to the Nernst equation.

$$E = E^0 - \frac{RT}{nF} \ln K \quad (5)$$

Here, E is the cell potential at a specified temperature, and E^0 is the standard cell potential at 298.15 K and atmospheric pressure.

At room temperature, the gas constant and temperature terms can be substituted into the Nernst equation to obtain Eq. (6).

$$E = E^0 - \frac{0.0591}{n} \log K \quad (6)$$

An important aspect of EC is the relationship between electrochemical reactions and the potential–pH (Pourbaix) diagram. The boundaries shown in a Pourbaix diagram represent equilibrium conditions for different electrochemical reactions and can be classified according to the dependence of the reaction on electron transfer and proton participation:

- Vertical lines: These lines denote reactions in which no electron transfer occurs ($n = 0$).
- Horizontal lines: These lines indicate reactions in which no hydrogen ions participate ($d = 0$).
- Inclined lines: These lines characterize reactions where both hydrogen ions are involved and electron transfer takes place.

For EC reactions in aqueous media, the operating conditions generally lie within or near the thermodynamic stability region of water, bounded by the oxygen- and hydrogen-evolution reactions. Reactions R8 and R9 in Table 1 define these boundaries in Fig. 1. Aluminum or iron sacrificial anodes release Al^{3+} or $\text{Fe}^{2+}/\text{Fe}^{3+}$ species, respectively, which undergo hydrolysis and complexation depending on pH. The resulting hydrolyzed and polymeric species can neutralize particle surface charge and promote coagulation [21].

Table1. Main anodic reactions of aluminum-containing species with relatively high abundance.

R #	Reaction	Eh -pH	ΔG^0
1	$Al^{3+} + H_2O \rightleftharpoons Al(OH)^{2+} + H^+$	$pH = \log \frac{[Al(OH)^{2+}]}{[Al^{3+}]} + 5.26$	29.9 79
2	$Al(OH)^{2+} + H_2O \rightleftharpoons Al(OH)_2^+ + H^+$	$pH = 5.66 + \log \frac{[Al(OH)_2^+][H^+]}{[Al(OH)^{2+}]}$ $\log[Al(OH)^{2+}] = 5.66 + \log[Al(OH)_2^+] - 2pH$	32.2 99
3	$Al(OH)_2^+ + 2H_2O \rightleftharpoons Al(OH)_4^- + 2H^+$	$pH = 0.5 \log \frac{[Al(OH)_4^-]}{[Al(OH)_2^+]} + 6.14$ $\log[Al(OH)_2^+] = 2(6.14 - pH + 0.5 \log[Al(OH)_4^-])$	69.9 28
4	$Al(OH)_4^- + 4H^+ \rightleftharpoons Al^{3+} + 4H_2O$	$pH = 5.8 - 0.25 \log \frac{[Al^{3+}]}{[Al(OH)_4^-]}$ $\log[Al(OH)_4^-] = 4(pH - 5.8 + 0.25 \log[Al^{3+}])$	- 132.21
5	$Al^{3+} + 3e^- \rightleftharpoons Al^0(s)$	$E = -1.69 + 0.02 \log[Al^{3+}]$	488. 69
6	$Al_2O_3(s) + 6H^+ + 6e^- \rightleftharpoons 2Al^0(s) + 3H_2O$	$E = -1.5 - 0.0591pH$	870. 81
7	$Al^{3+} + 3(OH)^- \rightleftharpoons Al(OH)_3(aq)$	$\log[Al(OH)_3(aq)] = \log[Al^{3+}] - 15.97 + 3pH$	-148.5
8	$2H^+ + 2e^- \rightleftharpoons H_2$	$E = -0.0591pH$	0
9	$1/2O_2 + 2H^+ + 2e^- \rightleftharpoons H_2O$	$E = 1.23 - 0.0591pH$	- 237.13

The derivations of the equations used to construct the Pourbaix diagram are summarized in Table 1. The listed reactions illustrate the three principal types of potential–pH boundaries.

A- Vertical line (R#1)

$$Al^{3+} + H_2O \rightleftharpoons Al(OH)^{2+} + H^+ . \Delta G^0 = 29.979 \text{ kJ/Rx}$$

$$\Delta G^0 = -RT \ln K \Rightarrow \log K = \frac{29.979 \times 1000}{-8.314 \times 298 \ln 10} = -5.26$$

$$K = \frac{[Al(OH)^{2+}][H^+]}{[Al^{3+}]}$$

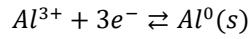
$$\log K = \log \frac{[\text{Al}(\text{OH})^{2+}]}{[\text{Al}^{3+}]} + \log [\text{H}^+] = -5.26$$

$$pH = 5.26 + \log \frac{[\text{Al}(\text{OH})^{2+}]}{[\text{Al}^{3+}]}$$

$$\log \frac{[\text{Al}(\text{OH})^{2+}]}{[\text{Al}^{3+}]} = -5.26 + pH$$

At $pH = 5.26$, the value of $[\text{Al}(\text{OH})^{2+}]$ is equal to that of $[\text{Al}^{3+}]$.

B- Horizontal line (R#5)



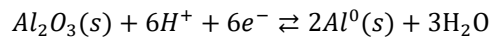
$$\Delta G^0 = 488.69 \text{ kJ/Rx} = -RT \ln K \Rightarrow \log K = \frac{488.69 \times 1000}{-8.314 \times 298 \ln 10} = -85.66$$

$$K = \frac{1}{[\text{Al}^{3+}]}$$

$$\Delta G^0 = -nFE^0 \Rightarrow E^0 = \frac{488.69 \times 1000}{-3 \times 96485} = -1.69 \text{ V}$$

$$E = E^0 - \frac{0.0591}{3} \log K = -1.69 + 0.02 \log [\text{Al}^{3+}]$$

C- Inclined line (R#6)



$$\Delta G^0 = 870.813 \text{ kJ/Rn} = -RT \ln K \Rightarrow \log K = \frac{870.813 \times 1000}{-8.314 \times 298 \ln 10} = -153.7$$

$$K = [\text{H}^+]^{-6}$$

$$\Delta G^0 = -nFE^0 \Rightarrow E^0 = \frac{870.813 \times 1000}{-6 \times 96485} = -1.5 \text{ V}$$

$$E = E^0 - \frac{0.0591}{6} \log K = -1.5 - \frac{0.0591}{6} \log [\text{H}^+]^{-6} = -1.5 - 0.0591 pH$$

The thermodynamic properties used to derive the equilibrium relationships and construct the Pourbaix diagram were obtained from the data summarized in Table 2 [22, 23].

Table 2. Compounds used in calculations and their standard Gibbs free energy at 298.15 K and atmospheric pressure [22, 23]

Compound	$\Delta G_0(kJ/mol)$
$H_2O(l)$	-237.129
$OH^-(aq)$	-157.269
$AlHO_2^*$	-207.50x4.18
$Al(OH)^{+2}$	-166.47 x4.18
$Al(OH)_2^+$	-215.47 x4.18
AlO_2^-	-198.70x4.18
Al^{3+}	-488.69
Gibbsite, $Al(OH)_3$	-1154.9±1.2
Gibbsite, $Al(OH)_3$	-1155.05
Diaspore, $AlO(OH)$	-922.0±5
Boehmite, $AlO(OH)$	-918.4±2.1
Boehmite, $AlO(OH)$	-918.15
Bayerite, $Al(OH)_3$	-1153±2
$[Al(OH)]_4^-$	-1305±1
$[Al(OH)]_2^+$	-914.2±2.1
Al^{3+}	-489.4±1.4
N-octane(l)	8.58x4.18
N-Heptane(l)	6.47x4.18
N-Hexane	4.42x4.18
$Al(OH)_3(aq)$	-265.3x4.18

Figure 1 presents the Pourbaix diagram for reactions R1–R9 in Table 1. The diagram is divided into regions in which different aqueous or solid aluminum species are thermodynamically predominant. For example, along R1, the equilibrium activities of Al^{3+} and $Al(OH)^{2+}$ are equal, with the boundary occurring at approximately pH 5.26 under the specified conditions. Similar interpretations can be made for the other boundaries. These equilibrium predictions describe thermodynamic tendencies and should not be interpreted as direct measurements of the species concentrations during EC operation.

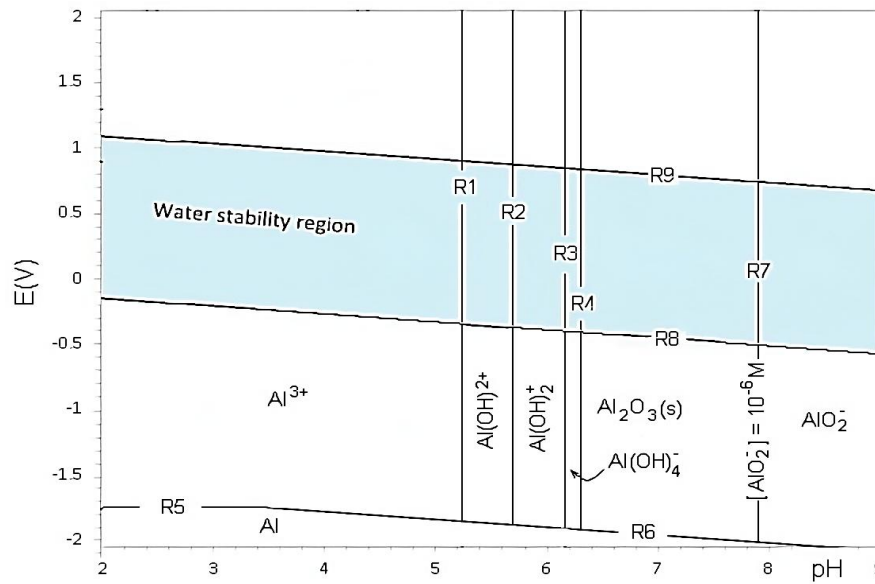


Fig. 1. EH-pH diagram for Al electrocoagulation chemical reactions

The relative abundance of Al(III) species varies strongly with pH, as illustrated in Figure 2. Figures 1 and 2 represent thermodynamic equilibrium relationships and therefore do not directly account for kinetic limitations, mass transfer, electrode passivation, or the hydrodynamics of the experimental reactor.

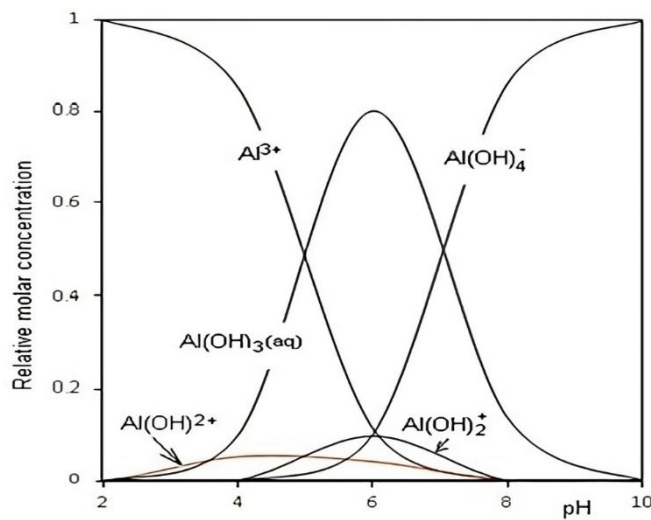


Fig. 2 Relative abundance of Al (III) species at equilibrium

1.2. Main chemical reactions in EC systems

An EC reactor typically consists of a sacrificial anode and a cathode connected to a direct-current power supply. Aluminum or iron is commonly used as the sacrificial anode, while stainless steel can be used as the cathode. Supporting electrolytes such as sodium chloride may be added to increase electrical conductivity [24]. At the

aluminum anode, aluminum is oxidized to Al^{3+} , whereas water reduction at the cathode generates hydrogen gas. Depending on the applied potential and electrolyte composition, oxygen evolution may also occur at the anode.



At the cathode, water reduction produces hydrogen gas and hydroxide ions. These reactions increase the local pH and promote the hydrolysis and precipitation of aluminum hydroxide species.



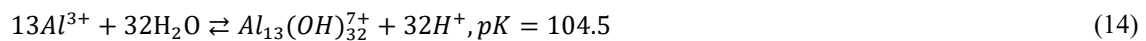
Although the bulk pH may remain relatively stable in some EC systems, local pH changes occur near the electrodes because of water electrolysis. During hydrolysis, dissolved aluminum species undergo a series of acid–base and complexation reactions that generate positively charged and neutral hydroxo species.



The EC reactor contains a distribution of Al(III) hydrolysis species that depends strongly on pH. $Al(OH)_3(aq)$ and related hydrolyzed species can subsequently form polymeric and precipitated aluminum hydroxides [25, 26].



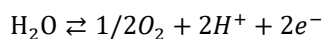
At moderately acidic pH, polymeric aluminum species can form and contribute strongly to charge neutralization and particle destabilization [27].



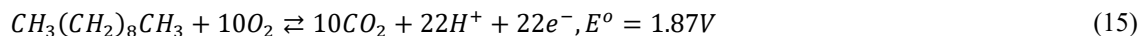
According to the Schulze–Hardy principle, multivalent counter-ions can destabilize colloidal particles at substantially lower concentrations than monovalent ions. In EC, the high specific surface area of freshly formed aluminum hydroxide flocs can also enhance adsorption of organic compounds and promote particle aggregation [25, 28].

1.3. COD removal mechanisms

After destabilization, adsorption, enmeshment, and aggregation contribute to the removal of suspended and dissolved contaminants by trapping them within or on aluminum hydroxide flocs. For petroleum wastewater, flotation by hydrogen bubbles can further enhance the separation of oil droplets and low-density particles.



The standard potential of the relevant oxygen-reduction/oxidation reaction should be interpreted together with the actual electrode potential and solution conditions. In petroleum wastewater, indirect oxidation may contribute to COD reduction; however, coagulation, adsorption, coprecipitation, and flotation are generally important EC removal mechanisms. Direct oxidation of individual hydrocarbons should therefore not be assumed to be the dominant pathway without specific chemical evidence.



$$\Delta G^0 = -10 \times 394.36 - 17.4 = -3961 kJ/Rx$$

$$\Delta G^0 = -RT \ln K \Rightarrow \log K = \frac{-3961 \times 1000}{-8.314 \times 298 \ln 10} = 694.3$$

$$E^o = \frac{3961 \times 1000}{22 \times 96485} = 1.87V$$

In chloride-containing electrolytes, chloride oxidation can produce chlorine and, subsequently, hypochlorous acid/hypochlorite depending on pH and electrode potential [29, 30]. These oxidizing species may contribute to indirect oxidation of organic contaminants, although their formation should be verified experimentally when mechanistic conclusions are required.



At sufficiently high anodic potentials, chloride-containing solutions may generate active chlorine species. These species can oxidize dissolved organic matter and thereby contribute to COD reduction. Consequently, COD removal in EC may result from a combination of coagulation/adsorption and electrochemical or indirect chemical oxidation rather than from a single mechanism.

1.4. Turbidity removal mechanisms

Suspended particles and colloids are often stabilized by negatively charged surfaces. In EC, positively charged aluminum hydrolysis species interact with these particles, compress the electrical double layer, and reduce the magnitude of the zeta potential. Adsorption and charge neutralization then destabilize the suspension, allowing particles to aggregate and settle. Hydrogen bubbles generated at the cathode can additionally promote electroflotation by attaching to oil droplets and suspended particles, thereby enhancing their separation. Previous studies have reported high turbidity-removal efficiencies for oil-in-water emulsions treated by EC [31–33].

1.5. Mechanisms responsible for the removal of other contaminants

Electrocoagulation can also reduce other contaminants in industrial wastewater through charge neutralization, adsorption, coprecipitation, flotation, and, when appropriate, electrochemical oxidation. The removal efficiency depends strongly on contaminant type, electrode material, current density, pH, treatment time, and wastewater composition.

2. Materials and Methods

All chemicals and standard solutions used for COD and turbidity measurements were of analytical grade [34, 35]. COD was determined using a Cary 100 Bio UV–Vis spectrophotometer in combination with a Hach DRB200 reactor. Turbidity was measured using a Hach 2100Q portable turbidity meter. Analytical procedures followed the applicable Standard Methods for the Examination of Water and Wastewater [36] and the other referenced analytical guidelines [37–39]. The cathode was a stainless-steel plate (SS316), and the sacrificial anode was an aluminum plate (Al 6061); both electrodes measured 12.5 cm × 4 cm × 0.3 mm. Direct current was supplied by a DAZHANG PS-302 power supply. Mixing was provided by a ZAG Chemical Co. magnetic stirrer. The EC reactor was a cylindrical Pyrex vessel 25 cm in height and 14 cm in diameter, with a working capacity of 1 L. The experimental setup is shown in Fig. 3.

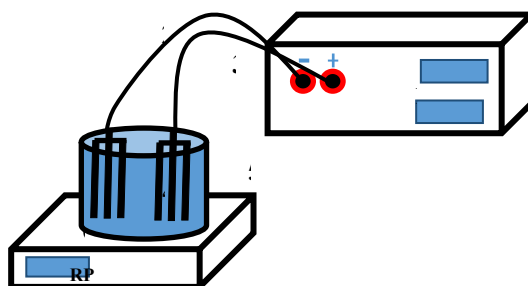


Fig. 3. Schematic diagram of the experimental setup.

3. Experimental Procedure

Wastewater samples were freshly collected from the refinery effluent collection point and centrifuged before EC treatment. The main characteristics of the wastewater are summarized in Table 3.

Table 3. Initial characteristics of the refinery wastewater.

Characteristic	Measurement Unit	Quantity
COD (without pre-treatment)	g/L	16.5
COD (with pre-treatment)	g/L	5.637
Turbidity	NTU	~231

Twenty experimental runs were conducted to evaluate the effects of current density (CD), pH, and treatment time on COD and turbidity removal. The investigated ranges were selected using the CCD experimental design and are presented, together with the measured responses, in Table 4.

Table 4. Experimental conditions and measured COD and turbidity removal efficiencies.

Run	Factors			Responses	
	pH	CD (mA/cm ²)	Treatment time (min)	COD removal (%)	Turbidity removal (%)
1	8	26	98	83.5	76.5
2	3	44	98	87.5	86.5

3	8	26	32	55	55
4	8	44	32	68	66
5	8	44	98	90	88
6	3	26	98	79	77
7	6	35	65	85	81.5
8	3	44	32	66	66
9	6	35	65	85.5	81
10	3	26	32	48	29
11	6	35	65	85.5	81.5
12	6	35	65	85.5	81.5
13	6	35	65	86	82
14	6	35	120	92	92.5
15	6	35	65	86	82
16	6	35	10	56	53.5
17	6	20	65	76	72
18	6	50	65	92	93
19	9	35	65	96.5	69
20	2	35	65	64	46

4. Results and Discussion

RSM based on CCD was used to evaluate the effects of pH, current density, and treatment time on COD and turbidity removal. ANOVA was used to assess the statistical significance and adequacy of the fitted models. The experimental results indicate that turbidity removal is strongly affected by pH and treatment time. Near-neutral conditions favor the formation of amorphous aluminum hydroxide species, which can enhance sweep flocculation and particle settling, whereas charge neutralization and soluble aluminum species become more important under acidic conditions.

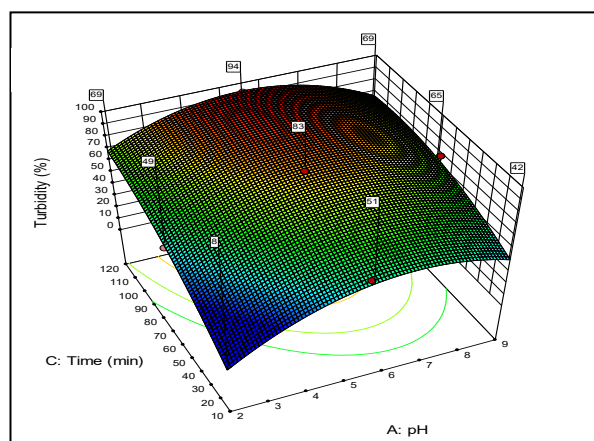


Fig. 4. Effect of pH and treatment time on turbidity removal.

The experimental results show that increasing current density generally improves turbidity removal within the investigated range because a higher current increases the rate of aluminum dissolution and floc generation. However, higher current densities also increase electrical energy consumption and may promote excessive electrode consumption. Therefore, an optimum current density should balance pollutant removal and energy/electrode costs.

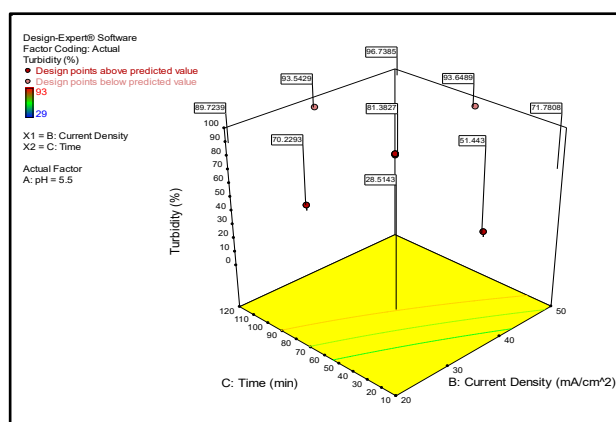


Fig. 5. Effect of current density on turbidity removal.

COD removal was strongly influenced by pH and treatment time. The highest COD-removal values were observed under conditions that favored the formation of aluminum hydroxide flocs and effective destabilization of emulsified and suspended organic matter. The response surface in Fig. 6 indicates that treatment performance changes substantially across the investigated pH range, with the most favorable region occurring around neutral to mildly acidic conditions.

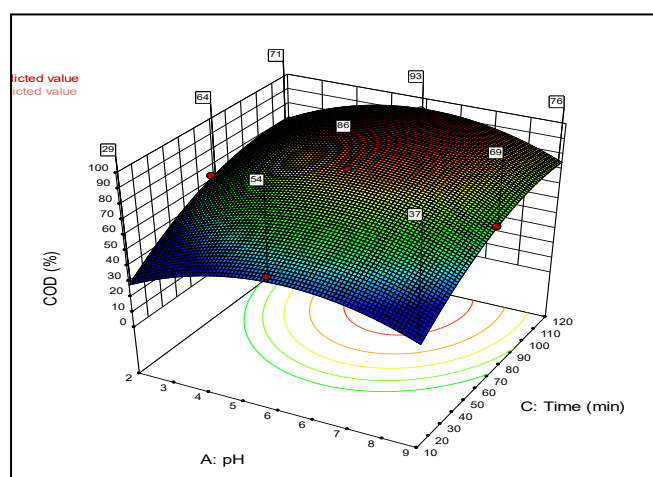


Fig. 6. Effect of pH and treatment time on COD removal.

Increasing current density generally increased COD removal within the investigated range because higher current promotes faster generation of aluminum coagulant and hydrogen bubbles. Nevertheless, increasing current density indefinitely is not necessarily beneficial because it increases energy consumption and may alter floc characteristics. The results therefore support the existence of an operating range in which COD removal is balanced against process cost.

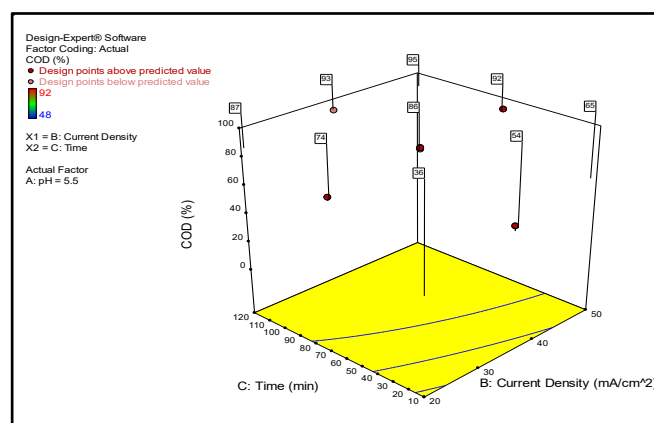


Fig. 7. Effect of current density and treatment time on COD removal.

4.1. Comparison of COD and turbidity removal using other chemical coagulants

Poly(aluminum chloride) (PAC) was also evaluated as a conventional chemical coagulant. PAC achieved approximately 75% turbidity removal at a concentration of 20 mg/L, whereas aluminum sulfate and lime showed lower performance under the tested conditions and were not considered further (Fig. 8).

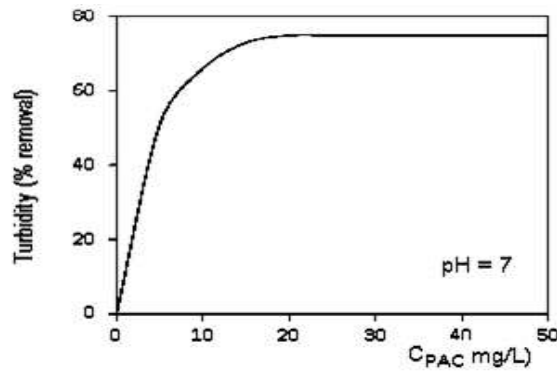


Fig. 8. Effect of PAC concentration on wastewater turbidity removal.

The addition of 15 mg/L PAC to the EC system at a current density of 50 mA/cm² produced only a modest additional improvement in turbidity removal under the tested conditions (Fig. 9). This comparison suggests that EC can reduce the required dose of conventional chemical coagulant; however, a rigorous economic comparison would require measurements of electricity consumption, electrode consumption, chemical dosage, and sludge production.

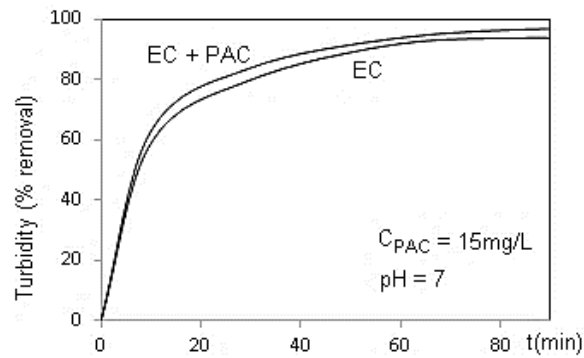


Fig. 9. Turbidity removal using PAC and combined EC–PAC treatment.

5. Conclusions

This study demonstrated that electrocoagulation can effectively reduce COD and turbidity in industrial petroleum wastewater under controlled pH, current-density, and treatment-time conditions. The experimental data showed maximum observed removal efficiencies of approximately 93% for COD and 96.5% for turbidity within the investigated design space. The results indicate that aluminum hydroxide formation, charge neutralization, adsorption, coprecipitation, enmeshment, and electroflotation collectively contribute to pollutant removal. RSM and CCD provided a systematic framework for evaluating the effects of the operating variables. Because higher current densities can increase both pollutant removal and electrical/electrode consumption, process optimization should consider treatment performance together with energy and material requirements. Electrode passivation remains an important practical limitation and should be addressed in future studies through electrode cleaning, polarity reversal, or alternative electrode configurations. Overall, the results support EC as a promising treatment

option for petroleum wastewater, while further work should include energy consumption, sludge characterization, long-term electrode stability, and validation with larger-scale continuous reactors.

6. Funding declaration

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7. Data availability

All data analysed during this study are included in this published article.

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