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Original Article

Electrocoagulation Process Efficiency for Removing Effluent Pollution Caused by Drilling of Oil Rigs

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Abstract

Background: Electrocoagulation (EC) is a safe method for removing environmental pollutants without the need for additional chemical materials. This study investigates the performance of EC in removing chemical oxygen demand (COD), total organic carbon (TOC), total suspended solids (TSS), and turbidity from drilling waste generated by oil rigs.

Methods: An experimental study was performed on a pilot scale in an EC reactor provided from a cylindrical glass cell (height: 30 cm and inner diameter: 5 cm), a pair of aluminum and iron electrodes, a power supply, an aeration system. wastewater Samples were collected from one of the drilling rigs in Khuzestan. The effect of current density, operation time and pH parameters on removal of COD, TOC, TSS and turbidity were determined and the optimal values of the parameters were determined.

Results: It was found that system voltage, operation time and pH values on the removal efficiency of pollutants were statistically significant at the 0.01 level. The optimum values of pH, current density and operation time were obtained 7, 20 mA/cm2 and 60 minutes, respectively and the removal efficiencies of COD, TOC, TSS and turbidity under the optimum conditions were 72%, 79%, 67% and 63%, respectively. Also, the consumption of energy was estimated to be 8.4 kWh/m^3.

Conclusion: The results indicated that the EC process is a cost-effective method in removing pollutants caused by drilling of oil rigs and its efficiency can be improved by applying optimal conditions such as current density and pH.

Keywords: Electrocoagulation, Chemical oxygen demand, Environmental pollution, Wastewater

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Introduction

The activities in the oil, gas, and petrochemical industries have been extended from exploration to production, transportation, refining, and consumption. These activities lead to the production of two types of waste, including drilling fluids wastewater and drill cuttings. Many of the waste products produced during oil and gas well drilling activities contain toxic materials such as biocides, crude oil, brine, and drilling muds, which have adverse effects on the environment.[1,2] Drilling operations generate various types of waste, including drilling logs and drilling fluid residues. Most of these wastes contain toxic compounds and can pose significant hazards to the environment if not properly managed.[2,3] The primary environmental concerns associated with oil rig drilling relate to organic hydrocarbons, biocides, stimulation fluids (crude oil, brine), electrical conductivity (amount of chloride in the salt phase), and the presence of heavy metals. Due to the complex chemical composition and toxicity of these pollutants, they are classified as hazardous substances that must be prioritized for removal from the environment.[4,5]

There are various methods available to treat pollutants in wastewater, including precipitation, flocculation, ion exchange, adsorption, membrane filtration, and photocatalysis, each with its own set of disadvantages; these may include high cost, low efficiency, the addition of chemicals, and the generation of a large amount of sludge. Therefore, it is important to carefully evaluate the advantages and disadvantages of each method and select the most appropriate one based on the specific characteristics of the wastewater and the desired treatment goals.[6,7] In recent years, advanced oxidation processes such as electrochemical methods have been widely applied due to their rapid oxidation and lack of secondary pollution.

These methods are becoming increasingly popular as a means of treating pollutants in wastewater, as they offer several advantages over conventional treatment methods, including high efficiency, low operating costs, and minimal generation of sludge or other waste products.[8,9] Generally, treatment of aqueous solution by electrochemical methods involves three types of mechanisms of electrocoagulation (EC), electro-flotation and electro-oxidation.[4] EC is an emerging and evolving technology in wastewater treatment that combines the benefits of coagulation, flotation, and electrochemistry. This process involves the use of electrodes to generate coagulants via electrical current, which can effectively remove various pollutants, including suspended solids, oil and grease, organic compounds, and heavy metals, from wastewater. EC has several advantages over conventional treatment methods, such as high efficiency, low operating costs, and minimal generation of sludge or other waste products. The soluble metal anode (e.g., Al, Fe) is oxidized and dissolves the metal ions when the power is applied from an external electric field.[10] The hydrogen ions reduce in the vicinity of the cathode and generate hydroxide flocs and bubbles.[11] Electrodes used in EC process can be arranged in monopolar or bipolar configurations.[9,12] Aluminum and iron cations dissolve from the anode, according to equations 1 and 2 and cathode according to equation 3. In this solution, positively charged species are attracted to the negatively charged hydroxide ions, resulting in the formation of ionic hydroxides. These hydroxides can then strongly attract dispersed particles, and the counter ions can cause coagulation. This process is known as EC and is a commonly used method for removing suspended particles, organic matter, and other contaminants from wastewater. [13,14]

$$4Fe_{(s)} \rightarrow 4Fe^{+2}_{(aq)} + 8e^{-} \quad [1]$$

$$Al_{(s)} \rightarrow Al^{+3}_{(aq)} + 3e^{-} \quad [2]$$

$$2H_2O + 2e \rightarrow H_2 + 2OH^{-} \quad [3]$$

Major advantages of the EC compared to other traditional methods are higher efficiency in the removal of pollutants with lower electrical energy usage, simple experimental set-up, less operation time and no addition of chemicals.[10] Some of the most important effective factors on the efficiency of EC processes are initial pH of solution, type of electrodes, current density, operating time, and charge loading. During the EC process, both the anode and cathode can be involved in oxidation and reduction reactions. At the cathode, reduction occurs as the electrode gains electrons, which can facilitate the formation of hydrogen gas or hydroxide ions. These species can then help to neutralize the solution and promote flocculation, which aids in water purification.[12] Various studies have been conducted to investigate the efficiency of the EC process in order to remove the pollutants from water and wastewater and optimize the refined operating conditions. Zhao et al studied chemical oxygen demand (COD), turbidity and hardness removal from water by EC affected by pH, current density.[15] Also, Ighilahriz et al compared the effectiveness of EC and electrooxidation treatments for the leachate of oil-drilling mud.[4] Safari et al studied the diesel and COD removal from oily wastewater by EC. They investigated the effects of different parameters including pH, time, current density, supporting electrolyte, electrode material and initial diesel concentration on the efficiency of EC.[10] Moussavi et al studied the remediation of groundwater contaminated with hydrocarbons by the method of EC.[16] However, one of the problems with the EC technology is the high operation cost, especially when the operation time is long.[17] Therefore, energy consumption under optimum conditions has the major importance.[18] Considering the importance of removing the pollutants from the effluents caused by drilling oil rigs and the need to optimize the effective factors on the EC process for treating the wastewater, the aim of present study was to investigate the efficiency of the EC process in reducing pollution load and optimize some of its factors that are effective in removing pollutants from drilling rigs effluent.

Materials and Methods

Sampling

Samples of the wastewater were collected from the vicinity of one of the drilling rigs in Khuzestan. A centrifugal pump was used for sampling. The effluent samples were immediately transferred to the laboratory by a 20-L container made of polyethylene and stored in a refrigerator at 4 °C. Table 1 presents the characteristics of the initial tested wastewater. Concentrations of COD, total organic carbon (TOC), turbidity, total suspended solids (TSS), total Kjeldahl nitrogen (TKN), and total dissolved solids (TDS), and levels of pH and electrical conductivity were determined at all stages of the study according to Standard Methods for the Examination of Water and Wastewater.[19]

Experiment Set up

All chemicals and reagents used in this study were of analytical reagent grade (Merck, Germany). The batch experimental apparatus has been shown in Figure 1.

Table 1. Initial Characteristics of the Wastewater Collected From Drilling Rig

Parameter (Unit)	Value
COD (mg/L)	410
TOC (mg/L)	330
Turbidity (NTU)	54
TSS (mg/L)	1568
TKN (mg/Lt)	219
TDS (mg/L)	564
EC (dS/m)	6
pH	7.7

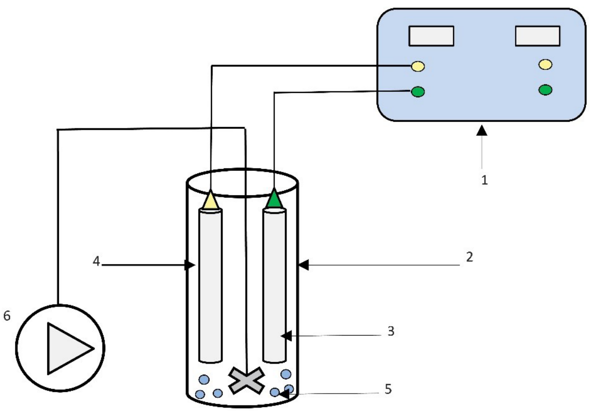

Figure 1. Schematic of the Experimental Equipment. 1- Power supply source. 2- Reactor. 3- Cathode. 4- Anode. 5-Bubble maker. 6- Air blower

The EC cell consisted mainly of a cylindrical glass cell with a height of 30 cm and an inner diameter of 5 cm, a direct power supply and carrier wires, and an aeration system equipped with a fine bubble diffuser. Aluminum and iron were used as anode and cathode electrodes as a thin rectangular sheet. The dimensions of the electrode plate were 0.2×2×10 cm. The electrodes were placed parallel to each other inside the cell at a fixed distance of 2 cm and 2 mm were used to measure the current density and voltage. The working volume of the electrochemical cell was maintained at a constant volume of 100 mL throughout the study. In this study, the closed process in a reactor was performed in the laboratory.

It was used 100 mL from the sampled wastewater in each run. The wastewater solution was mixed with the appropriate amount of sodium chloride as a supporting electrolyte. Sodium chloride was selected as supporting electrolyte due to its electrical conductivity, which subsequently affects the cell voltage and energy consumption in the electrolyte cell.[10] The DC generator was started and current density and operation time were adjusted after transferring the samples to the reactor. In this experiment, aeration at the rate of 0.5 L/min was used to mix and enter the oxygen required for the oxidation of divalent iron. In all the experiments, the speed of the magnetic stirrer was constant at 200 rpm. At the end of the operation time, the flow was stopped and the solution was passed through filter paper to remove the formed clots and then the necessary analyzes were performed on the liquid based on standard methods.

The pH was adjusted by the addition of NaOH (0.1 N) and/or HCl (0.1 N) solutions and all of the experiments were controlled by analyzing the pH. Direct current from the DC power supply was passed through the solution via the two electrodes during the 30–60 minutes of electrolysis run. Next, 10 mL of the solution was withdrawn at every 5 minutes interval for the first half hour and 10 minutes interval for the remaining time of the run.

All of the parameters were analyzed according to Standard Methods for the Examination of Water and Wastewater at all stages of the study.[19] Moreover, COD and TOC were measured by closed reflux titrimetric method and TOC Analyzer (ANATOCTM SERIES II, Australia), respectively. Turbidity and TSS were measured by a turbidimeter (AQUA LYTIC AL400T-WL, Germany) and a conductivity meter (PL-700PC, GONDO Company, Taiwan), respectively.[19]

Furthermore, removal efficiency was calculated with equation (4), in which C_0 and C_f are the initial concentration and concentration at time t, respectively, for the studied parameter (COD, TOC, TSS and Turbidity).[10,12]

$$E = \frac{C_o - C_f}{C_O} \times 100 \qquad [4]$$

Where E, C_o and C_f are the removal efficiency, content of the pollutant before the EC process, and content of the pollutant after the EC process, respectively.[3] The percentage of pollutant removal was applied under different conditions by varying the pH (3, 5, 7, 9 and 11), current density (2, 4, 6, 8, 10, 15 and 20 mA/cm^2), and operation time (1, 5, 10, 15, 20, 30, 60 and 90 min) parameters at three replications.

Analysis the Energy Consumption

The content of energy consumption was evaluated using the equation 5:

$$\text{Energy Consumption (kWh/m}^3) = \frac{U.I.t}{V} \qquad [5]$$

Where U is the applied voltage (V), I is the applied current (A), t is the contact time (h), and V is the volume of the polluted solution (m^3).[10,11]

Data Analysis

Data were analyzed using the SPSS software version 16, and One-way ANOVA was performed to determine whether there was the significant effect on removal efficiency of pollutants in wastewater by EC process.

Results and Discussion

The results of the analysis of variance (ANOVA) method, which was used to descript the effect of operational variables (current density, operation time and pH values) on the efficiency of COD, TOC, TSS and turbidity removals, have been presented in Table 2. Also, the results of COD, TOC, TSS and turbidity removal by the EC process at different current densities (2, 4, 6, 8, 10, 15, 20 mA/cm^2), operation times (1, 5, 10, 15, 20, 30, 60, 90 minutes), and pH (3, 5, 7, 9, 11) have been presented in Figures 1, 2 and 3, respectively. The findings of ANOVA showed that the effect of the studied variables on the parameters (COD, TOC, TSS and turbidity) was significant at the 0.01 level.

Effect of pH on Removal Efficiency of COD, TOC, TSS and Turbidity

pH is one of the most important factors affecting the performance of the EC process. The pH value mostly

Table 2. Results of the ANOVA Test for COD Removal Efficiency

	Variable	Degree of Freedom	Average Squares	F Value
	Current density	6	69.125	6.68**
	Reaction time	7	64.84	8.65**
COD	pH	4	125.26	10.6**
	Repeat	2	6.247	0.116 ns
	Error	25	0.215	
	Current density	6	55.93	9.49**
	Reaction time	7	55.14	7.32**
TOC	pH	4	111.48	9.53**
	Repeat	2	5.68	0.07 ns
	Error	25	0.197	
	Current density	6	116.13	79.34**
	Reaction time	7	1162.58	18.65**
TSS	pH	4	2288.51	29.31**
	Repeat	2	12809.19	0.481 ns
	Error	25	4.58	
	Current density	6	28.82	3.67*
	Reaction time	7	30.94	3.94**
Turbidity	pH	4	20.81	3.34*
	Repeat	2	1.23	0.223 ns
	Error	25	0.028	

** Significant at 0.01, *Significant at 0.05, ns: not significant.

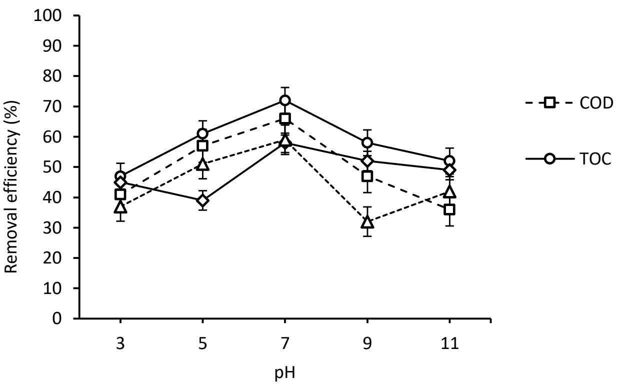

Figure 2. Effect of pH Value on Removal Efficiency of COD, TOC, TSS and Turbidity (operation time 60 min, and current density 20 mA/cm^2)

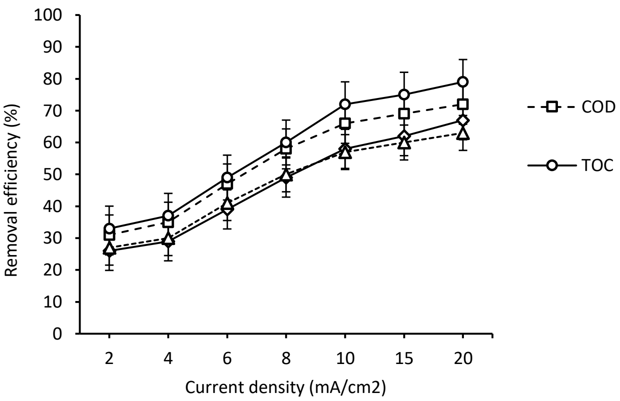

Figure 3. Effect of Current Density on Removal Efficiency of COD, TOC, TSS, and Turbidity (pH 7 and operation time 60 min)

influent by the cathodes. Increasing current density influences the pH as well.[7] pH is an effective factor in chemical and biochemical reactions. The effect of the initial pH of the environment varies greatly depending on the type of the used process and pollutant.[16]

The maximum removal efficiency of COD at constant value of current density (20 mA/cm^2) and operation time (60 minutes) occurred at the pH of 7 (66%) and the minimum removal efficiency (only 36%) was occurred at the pH value of 11, according to Figure 1. Also, the results indicated that the removal efficiency at neutral pH was higher than that at acidic and alkaline pH and the maximum removal efficiency occurred at pH 7 and the removal efficiencies of COD, TOC, TSS and turbidity decreased to less than 50%, with increasing and decreasing pH from 7. Therefore, the optimum pH in the removal efficiency of contaminants in this method was determined 7.

Although pH is a very important and key factor in the EC process that affects the zeta potential, the conductivity of the solution, and the dissolution of the electrode, it is very difficult to determine a relationship between the pH value and the removal efficiency of the EC process due to the continuous changes in the pH of the treated water during the EC process. Therefore, the initial pH of the solution was usually determined as the basis for the study.

Aluminum and iron anodes behaved in a different manner during EC; when aluminum was used as the anode, it underwent electrolysis to form trivalent aluminum ions, which was followed by spontaneous hydrolysis to give various species depending on the pH of the solution. The anodic reactions make the vicinity of the anode slightly acidic, which is opposed, by the cathode vicinity being slightly alkaline due to hydrogen evolution and production of OH^- ions. As a general observation, when the initial pH of the solution is highly acidic ($pH < 3$) or highly alkaline ($pH > 11$), there is no considerable change in the initial pH. However, when the initial pH is acidic, pH is expected to rise throughout the EC process and, when the initial pH is alkaline, pH is expected to decrease along the EC process. Hence, EC using aluminum anode is considered a pH-neutralizer. The effect of pH on EC is affected by the solubility of metal hydroxides. Generally, aluminum and iron electrodes are more suitable for the EC method and for the Al electrode generally an acidic medium is used ($pH < 6$), whereas, an alkaline and neutral condition is preferred for the Fe electrode. Unlike aluminum anodes, iron can dissolve as divalent or trivalent cations, which are then hydrolyzed to form insoluble iron compounds depending on the pH of the solution and the cell potential.[9,17] Bener et al evaluated the relationship between the pH and the removal efficiency of TOC and observed the highest removal efficiency of 65% at pH 5. They showed that the higher pH than 5 resulted in a reduced efficiency which could be due to the formation of hydrogen gas at the cathode.[20] The results of Bayar et al study on the effect of initial pH on the treatment of poultry slaughterhouse wastewater by EC indicated that the suitable pH range was

3–5 for COD and turbidity removals.[21]

Effect of Current Density on Removal Efficiency of COD, TOC, TSS and Turbidity

Another very important factor in the EC process, which plays a significant role in the removal of contaminants, is the effect of current density changes. This parameter affects the rate of EC reactions by affecting the amount of metal ions removed from the electrode surface.[18] In order to evaluate the optimal current density in the removal efficiency of COD, the effect of current density at 2, 4, 6, 8, 10, 15 and 20 mA/cm^2 was investigated and the results have been presented in Figure 3.

According to Figure 3, the maximum removal efficiency of COD at constant value of pH (7) and operation time (60 minutes) was obtained at the 20 mA/cm^2 (72%); while the minimum removal efficiency (31%) was occurred at 2 mA/cm^2. Based on Figure 3, the removal efficiency of contaminants increased with increasing current density of the system; hence, the highest percentage of removal efficiency of contaminants was related to 20 mA/cm^2.

There is a direct relationship between current density and the removal efficiency of COD by the EC process.[11] Increasing current density increases the density of the bubbles formed and decreases their size. This increase in current density also causes the formation of iron hydroxide deposits and clots in the process. Increasing the amount of these two substances has a direct effect by increasing the efficiency of the electrochemical process. Also, according to Faraday's law, at a constant operation time, the rate of electron transfer, and consequently the rate of oxidation and reduction at the anode and cathode, increases with increasing current density through the electrode surface.[17] Therefore, with increasing current density, the amount of metal hydroxide production and destabilization of colloidal materials and consequently the removal efficiency by the EC process increases. The reason for the increase in process efficiency by increasing current density could be attributed to an increase in electricity flow through the solution, which leads to further decomposition of electrodes and the production of metal hydroxides and gelatinous suspensions.[18] Bener et al studied the relationship between current density and TOC removal. They achieved the maximum removal efficiency of pollutant at the current density of 100 mA/cm^2, while, the lowest was observed at 12.5 mA/cm^2. The researchers showed that, when the current density was raised from 50 to 100 mA/cm^2, the removal efficiency improved from 28.5 to 34.42%.[20] Maha Lakshmi and Sivashanmugam investigated the effects of current density on the treatment of oil tanning wastewater and reported an incensement of COD removal efficiency from 87% to 90% with an increase in current density from 10 to 20 mA/cm^2.[22]

Effect of Operation Time on Removal Efficiency of COD, TOC, TSS and Turbidity

Operation time is one of the influential factors in performing EC processes. During the EC reaction, the operation time is the duration of the application of the desired electrical potential in the cell, which causes the current of the electrical circuit to intensify. Increasing the electrolysis time, according to Faraday law, increases the amount of metal ions and hydroxide produced at the anode and cathode, thus increasing the concentration of metal hydroxide and increasing the removal efficiency.[20,21] According to Figure 4, the maximum removal efficiency of COD at the optimum values of current density (20 mA/cm^2) and pH value (7) was at 60 minutes (72%); moreover, the minimum removal efficiency (only 12%) was at 1 minute. The removal efficiency of all contaminants increased with an increase in operation time; the incremental process lasted up to 60 minutes, according to Figure 4.

The relationship between operation time and pollutant removal efficiency depends on the reaction between the coagulant and the pollutant. As the contact time between the coagulant and pollutant increases, the amount of iron oxide and production of $Fe(OH)_3$ deposits and clots also increase, resulting in improved pollutant removal. Also, according to Faraday's law, there is a direct relationship between the amount of coagulation and the contact time of the coagulant and pollutant and the amount of coagulation and the removal value increases with time.[17] Ghorbanian et al also reported that increasing the operation time increased the production of aluminum cations at the anode and hydroxyl anions at the cathode and provided ample opportunity for the formation of aluminum hydroxide precipitate. As a result, with increasing the amount of aluminum hydroxide as a coagulant, the removal efficiency also increased.[23] At the current study, although further increasing the time resulted in slight increase in removal efficiency of pollutant removal, the optimal time should be selected according to the high energy consumption. Therefore, based on the obtained results obtained in the present study, the operating time of 60 minutes was selected as the optimal time. The removal efficiency is consistent with the study by Maha Lakshmi and Sivashanmugam study, which evaluated the effects of operation time on COD removal from oil tanning effluent

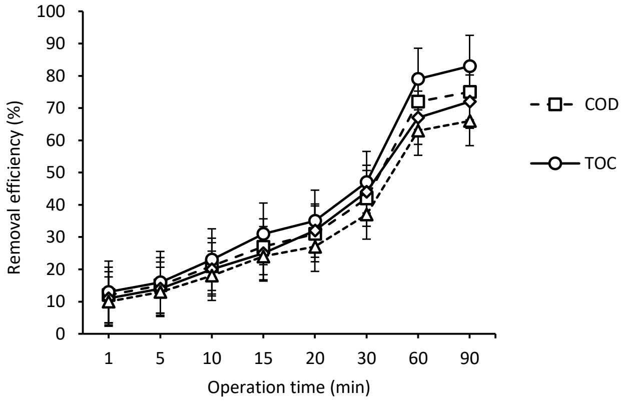

Figure 4. Effect of Operation Time on Removal Efficiency of COD, TOC, TSS and Turbidity (pH 7 and current density 20 mA/cm^2)

Table 3. The Summary of the optimum conditions and energy consumption of EC based studies to remove the pollutants from wastewater compared to the current study

Type of Wastewater	Current density (mA/cm^2)	pH	Time (min)	Consumption of Energy (kWh/m^3)	Removal Efficiency (%)	References
Wastewater from drilling oil rigs	20	7	60	8.4	72% (COD) 79% (TOC) 67% (TSS) 63% (Turbidity)	Current study
Wastewater from distillery industries	5	6	400	5.7	100% (COD)	8
Baker's yeast wastewater	120	5	20	14.9	77.64% (COD) 97.5% (Turbidity)	11
Leachate of oil drilling mud	28	7.5	60	11.3	95% (COD)	16
Wastewater from textile industries	6	6	120	7.41	90% (COD) 98% (TSS) 98% (Turbidity)	27
Petroleum refinery wastewater	13	5	60	0.3	60% (COD)	28
Oil tanning wastewater	200	6.5	15	6	90% (COD)	29

and observed a significant decrease at the COD removal percentage after 15 min.[22]

Shahriari and Saeb have obtained 87%, 91% and 97% removal efficiencies of COD, TSS, and dye in 120 minutes, pH 7 and voltage 30 V, but in the present study, lower removal rate in less time and less current density were obtained.[24] Esfandyari et al have achieved more than 92% of the removal efficiency in 50 min by using EC in cefazolin and COD removal from hospital wastewater.[25] Safari et al reported the highest removal efficiency of COD (99.1%) from oily wastewater by EC under the optimal conditions: of pH 7, operating time 40 min, 10.5 V, NaCl concentration 0.5 g/L and energy consumption 6.47 kWh/m^3.[10] Moussavi et al showed a removal efficiency of total petroleum hydrocarbons 95% at a pH close to 7 at a current density of 181 A/cm^2.[16] Other parameters including the type and configuration of electrodes, the volume and concentration of wastewater and the type of background electrolyte also affect the efficiency and costs of the EC method, which are necessary to be considered in future studies.[26] The Summary of the optimum conditions and energy consumption of EC based studies to remove the pollutants from wastewater compared to the current study have been presented in Table 3.

Conclusion

In the present study, the application of EC process to remove COD, TOC, TSS and turbidity from oil rig effluents was studied. Also, the effect of pH, current density and the operation time were considered. The results demonstrated that the EC process is capable of removing the COD, TOC, TSS and turbidity of the wastewater from drilling oil rigs. According to the results, pH, current density and the operation time had the significant effect on the COD, TOC, TSS and turbidity removal. In this study, the optimum condition for removal COD, TOC, TSS and turbidity was obtained at current density 20 mA/cm^2, pH 7 and operation time 60 minutes; The results at the optimum condition showed the EC process removed 72%, 79%, 67% and 63% of COD, TOC, TSS and turbidity, respectively. Also, the amount of energy consumption was 8.4 kWh/m^3. Therefore, it can be stated that the EC process can be applied as an environmentally friendly and cost-effective method to remove the effluent caused by drilling of oil rigs.

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Data curation: Ehsan Derikvand.
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Supervision: Ehsan Derikvand.
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Visualization: Ehsan Derikvand.
Writing–original draft: Seyed Mehdi Ale-Tayeb, Ehsan Derikvand, Mohsen Solimani Babarsad, Majid Razaz, Hossein Eslami.
Writing–review & editing: Ehsan Derikvand.

Competing Interests

The authors declared no conflict of interest.

Ethical Approval

There was no ethical consideration to be considered in this research.

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