



Treatment of ointment pharmaceutical wastewater by electrocoagulation process

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ABSTRACT

In the present study, electrocoagulation process was investigated for the removal of chemical oxygen demand (COD) and turbidity from ointment pharmaceutical wastewater in batch operation under different conditions. The effects of solution temperature, type of electrode pair, current density, conductivity and initial COD concentration on the removal efficiency of COD and turbidity were investigated. Experimental results indicated that the removal efficiency of COD and turbidity are 95% and 98%, respectively, found with the use of Fe/Al as electrode pair and the specific energy consumption was 0.48 kWh/kg COD after 20 min of electrolysis time. The optimum temperature, current density, conductivity and initial COD concentration were found to be 298 K, 15.56 mA/cm², 3.20 mS/cm and 5,000 mg/L, respectively.

Keywords: Ointment pharmaceutical wastewater; Electrocoagulation; Aluminum; Iron

1. Introduction

Different pharmaceutical products are used vastly to protect human and animal life, and drug consumption increases with the increase of diseases. When released into the environment, the pharmaceutical products become dangerous and toxic. Many researchers have reported the presence of pharmaceutical products in superficial water [1], in surface water [2] and underground water [3]. The toxicity of pharmaceutical wastewater is localized in the apparition of other bacteria in nature [4] and the feminization of fish, which was the first source of the alimentary chain of humans [5].

Pharmaceutical wastewater was treated first by a biological treatment, but it is not efficient for the treatment of wastewater with chemical oxygen demand (COD) higher than 4,000 mg/L [6]. The reason for the low efficiency in inhibition of microbial growth could be the presence of

the antibiotic aromatic pollutant in the wastewater [7]. The pharmaceutical wastewater was also treated by a physicochemical treatment [8] and chemical coagulation [9]; these methods need the use of reagents. Other methods are used to treat the pharmaceutical wastewater like activated carbon [10], chlorination [11], ozonation [12], perozonation [13] and photo-Fenton [14], but these methods are expensive.

Electrocoagulation is the method of treatment based on the formation of coagulant in situ with the use of electrode consumable and current electric. It is occurring via several steps such as: electrolytic reactions at electrode surfaces, the formation of coagulants in the aqueous phase and adsorption of soluble or colloidal pollutants on coagulants, which are removed by sedimentation or flotation.

Electrocoagulation has been reported to successfully treat wastewater of different kinds, cutting oil emulsions [15], 4-nitrophenol [16], textile wastewater [17,18], arsenic [19], polyvinyl alcohol [20,21], olive mill wastewater [22], dye [23–25], laundry wastewater [26], fluoride [27,28],

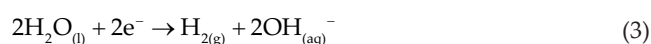
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nitrites [29], petroleum hydrocarbons [30], chromium ions [31], phosphate [32] and other wastewaters. The most widely used electrode materials in electrocoagulation (EC) process are aluminum and iron. They give after oxidation trivalent cation species, which provide better results compared with the divalent cations [33].

Anode:



Cathode:



The amorphous $\text{Al}(\text{OH})_3$ and $\text{Fe}(\text{OH})_3$ formed from the combination of Eqs. (2) and (3), and (1) and (3), respectively, has large surface areas, which are beneficial for a rapid adsorption of pollutants in the solution. Finally, these flocs are removed from the solution by sedimentation or flotation by the hydrogen and oxygen bubbles formed at the level of cathode and anode, respectively.

2. Materials and methods

2.1. Materials

The ointment pharmaceutical wastewater was supplied by Soidal pharmaceutical factory from Algeria. The characteristics of the wastewater are summarized in Table 1.

2.2. Experimental apparatus

The experimental setup is shown in Fig. 1. The electrochemical cell was made of Plexiglas with the dimensions of $14.0 \times 7.0 \times 15.5$ cm. Two vertical electrode plates of aluminum or iron with dimensions of 2 mm (thick) $\times 60$ mm (width) $\times 120$ mm (height) were used: one anode and one cathode. The total effective electrode area was 16.06 cm^2 . A magnetic stirrer has been used for mixing at the rate of 300 rpm in all stages of the study.

Table 1
Characterization of the ointment pharmaceutical wastewater

Parameter	
pH	7.89
Chemical oxygen demand (COD) (mg/L of O_2)	5,000
Nitrates (mg/L)	1.138
Phosphates (mg/L)	7.15
Turbidity (mg/L)	3,280
Color	White
Conductivity (mS/cm)	1.9

2.3. Experimental procedure

In this study, 800 cm^3 of a pharmaceutical wastewater was placed in the electrolytic cell. The current density was adjusted to the desired value (galvanostatic mode). The run was performed for 60 min. Samples were taken every 5 min interval from the electrocoagulator, filtered through $0.22 \mu\text{m}$ nylon syringe filter in order to measure COD concentrations using closed-reflux method and turbidity with turbidimeter. The electrode was weighed before and after the run.

2.4. Analytical method

The pH and conductivity were adjusted using NaOH or H_2SO_4 and NaCl, respectively. The calculation of removal efficiency (RE%) after EC was calculated using the following equation:

$$\text{RE}\% = \frac{(C_0 - C)}{C_0} \times 100 \quad (4)$$

where C_0 and C are the concentrations of COD before and after EC in mg/L, respectively.

The electrode consumption ($C_{\text{electrode}}$) having a unit of kg Al/ m^3 of wastewater treated is calculated from Faraday's law in the following relation:

$$C_{\text{electrode}} = \frac{I \times t \times M \times 10^{-3}}{z \times F \times v} \quad (5)$$

where I is the current intensity (A); t is the retention time (s); v is the volume of the treated wastewater (m^3); F is the Faraday's constant (96,487 C/mol); M is the mass of aluminum (26.98 g/mol) and mass of iron (55.847 g/mol); and z is the number of electron transfer ($z_{\text{Al}} = z_{\text{Fe}} = 3$).

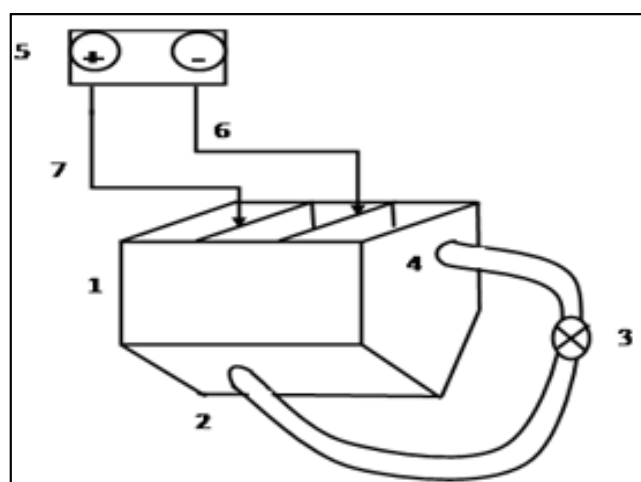


Fig. 1. Schematic diagram of the experimental setup.
Note: 1 – Electrolytic cell, 2 – outlet, 3 – pump, 4 – inlet, 5 – power supply, 6 – cathode and 7 – anode.

Specific energy consumptions (SEC) can be expressed as:

$$SEC = \frac{U \times I \times t}{(COD_0 - COD_t) \times V} \quad (6)$$

where SEC is the specific energy consumption (kWh/kg of COD removed); U is the applied voltage (V); I is the current intensity (A); t is the retention time (h); COD_0 is the chemical oxygen demand before treatment (g/L); COD_t is the chemical oxygen demand after treatment (g/L); and V is the volume of the treated wastewater (L).

3. Results and discussion

The effects of the solution temperature, type of electrode pair, current density, conductivity of the solution and initial concentration of COD are investigated on the removal efficiency of COD and turbidity. The energy consumption (kWh/kg COD_{eliminated}) is also determined by these factors.

3.1. Effect of solution temperature

Temperature plays an important role in the chemical and electrochemical relationship; in this study, it varied between 286.5, 298 and 303 K. The effect of the temperature on the COD and turbidity removal efficiency is presented in Fig. 2 with the use of electrode of aluminum as anode and cathode.

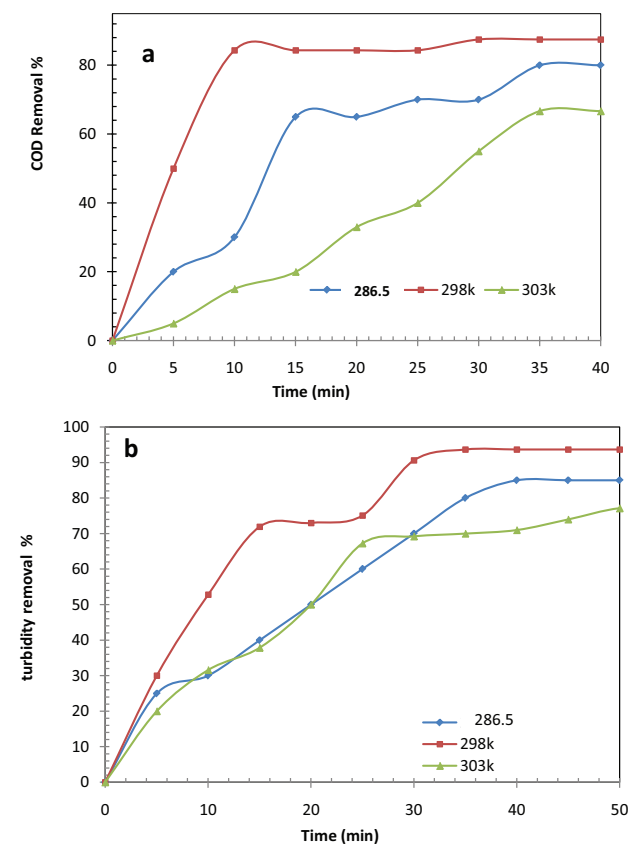


Fig. 2. The effect of the temperature on: (a) removal efficiency of COD and (b) removal efficiency of turbidity ($I = 15.56 \text{ mA/cm}^2$, $d = 2 \text{ cm}$, $\text{pH} = 7.89$ and conductivity = 4.89 mS/cm , Al/Al).

The results show that COD and turbidity removal efficiency increase with the increase of times of electrolysis in all temperatures used and the best removal of COD and turbidity was found with temperature 298 K. The best removal efficiency of COD was 84.37% attained after 10 min of electrolysis times, but the maximum removal efficiency of turbidity was 93.67% after 35 min of electrolysis times.

The effect of temperature on the removal efficiency of COD and turbidity after 20 min of treatment and the energy consumption is presented in Fig. 3.

The result shows that COD removal efficiency increases from 65% to 84.38% and turbidity removal efficiency increases from 50% to 73%, respectively, with the increase of the solution temperature from 286.5 to 298 K, which can be explained by the increase of mobility and collision of pollutants with hydroxyl polymers. The removal efficiency of COD decreases from 84.38% to 33% and turbidity removal efficiency from 73% to 50%, respectively, when the solution temperature increases from 298 to 303 K. The reason could be the increase of the solubility of precipitates formed [22]. The minimum energy consumption was 0.43 kWh/kg COD found in temperature 298 K because the electrical potential decreases with the increase of temperature of the solution.

3.2. Effect of type of electrodes

The type of electrode pair is a significant factor affecting the performance of the EC [16,20,21,34–36], and it depends on the type of pollutants. Four combinations of iron and aluminum plates were investigated in this study to determine the optimum electrode pair. Fig. 4 shows the effect of the electrode pair on the removal efficiency of COD and turbidity. As shown in Fig. 4(a), the Fe/Al had the highest removal efficiency of COD after 15 min of electrolysis time, followed by the Fe/Fe, Al/Al and Al/Fe anode/cathode pairs. The best removal efficiency of turbidity was found with the Fe/Al and Fe/Fe followed by the Al/Al and Al/Fe anode/cathode pairs as shown in Fig. 4(b).

The removal efficiency of COD and turbidity and energy consumption after 20 min of electrolysis time are presented in Fig. 5. The result shows that the minimum

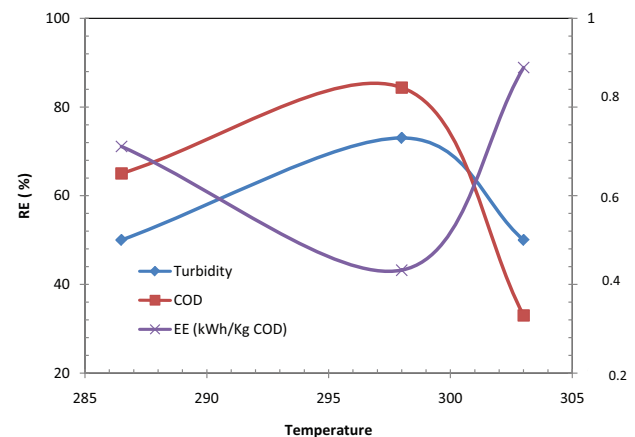


Fig. 3. Effect of solution temperature on COD and turbidity removal efficiency and on energy consumption ($t = 20 \text{ min}$, $I = 15.56 \text{ mA/cm}^2$, $d = 2 \text{ cm}$, $\text{pH} = 7.89$ and conductivity = 4.89 mS/cm).

energy consumption was 0.41 kWh/kg COD found with the use of Fe/Al and Al/Fe but the best removal efficiency of COD. The effect of the type of electrode pair on electrode consumption is shown in Fig. 6. The results show that the mass consumption of anode experimental is higher than

the mass anode theoretical consumption with every type of electrode used. But the difference between them increases with the use of iron as anode compared with the use of aluminum as an anode. That can explain the good removal efficiency of COD and turbidity found with the use of Fe as an anode, and turbidity was found with the Fe/Al pairs of the electrode. The coagulant formed with electrode iron can adsorb more pollutant compared with the coagulant formed by the use of electrode of aluminum. The mass consumption of cathode experimental is lower than the mass consumption of anode experimental. But it is smaller with the use of iron as a cathode.

3.3. Effect of the conductivity of the solution

Conductivity is an important parameter in the electrochemical process; it influences the quantity of the current; and it will be increased by the addition of salt. In this study, we used the sodium chloride, NaCl; it is varied between 0.5 and 2 g/L.

The conductivity growth with the increase of the concentration electrolyte supporting NaCl, contrarily to the potential it decreases as seen in Fig. 7. The effects of the conductivity on the removal efficiency of COD and turbidity are shown in Fig. 8. The result shows that the removal efficiency of COD and turbidity increases with the increase of conductivity of the solution from 1.90 to 3.20 ms/cm, but they stay unchanged when the conductivity increases from 3.20 to 4.79 ms/cm but SEC decreases from 0.44 to 0.41 kWh/kg COD (Fig. 9).

3.4. Effect of the current density

The current density is the most important parameter affecting the EC performance; in this study, it is varied between 7.78 and 28 mA/cm². Fig. 10 shows the effect of the current density on the removal efficiency of COD and turbidity for various electrolysis times. The results show that

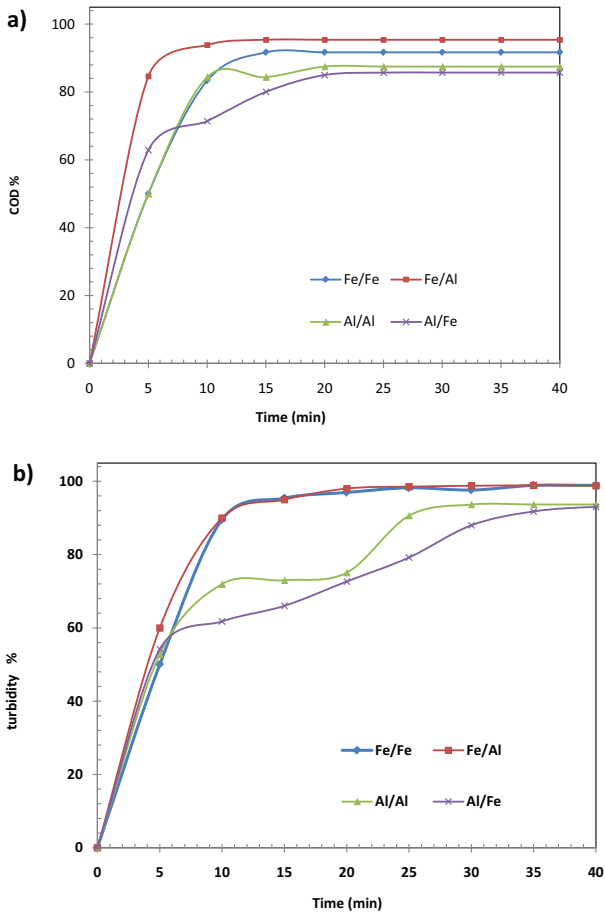


Fig. 4. Effect of the type of electrode pair on: (a) removal efficiency of COD and (b) removal efficiency of turbidity ($I = 15.56 \text{ mA/cm}^2$, $d = 2 \text{ cm}$, $T = 298 \text{ K}$, $\text{pH} = 7.89$ and conductivity = 4.89 mS/cm).

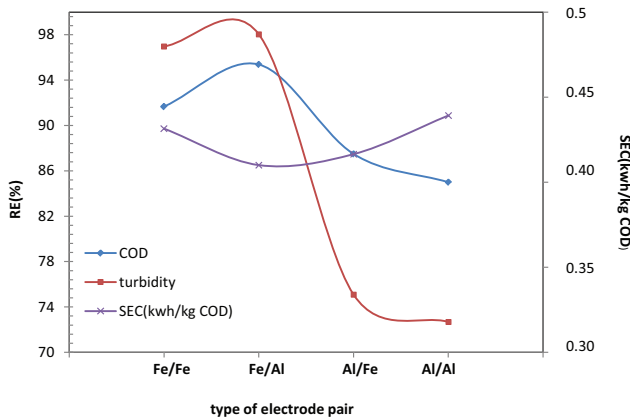


Fig. 5. Effect of electrode materials on COD and turbidity removal efficiency and on energy consumption ($t = 20 \text{ min}$, $I = 15.56 \text{ mA/cm}^2$, $d = 2 \text{ cm}$, $T = 298 \text{ K}$, $\text{pH} = 7.89$, conductivity = 4.89 mS/cm).

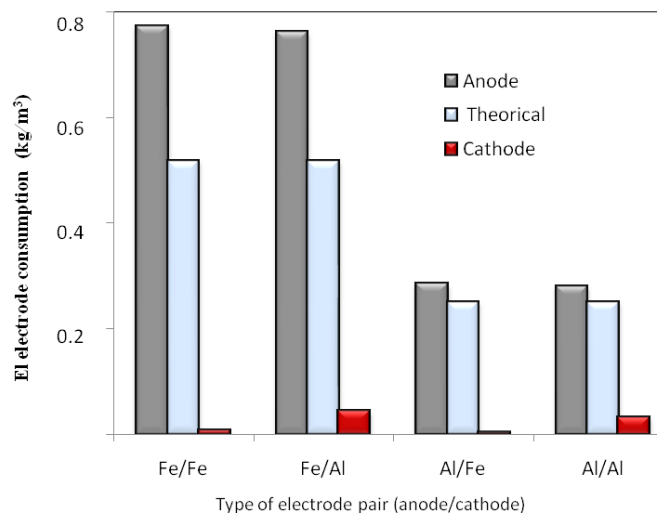


Fig. 6. Effect of the type of electrode pair on electrode consumption ($t = 60 \text{ min}$, $I = 15.56 \text{ mA/cm}^2$, $d = 2 \text{ cm}$, $T = 298 \text{ K}$, $\text{pH} = 7.89$ and conductivity = 4.89 mS/cm).

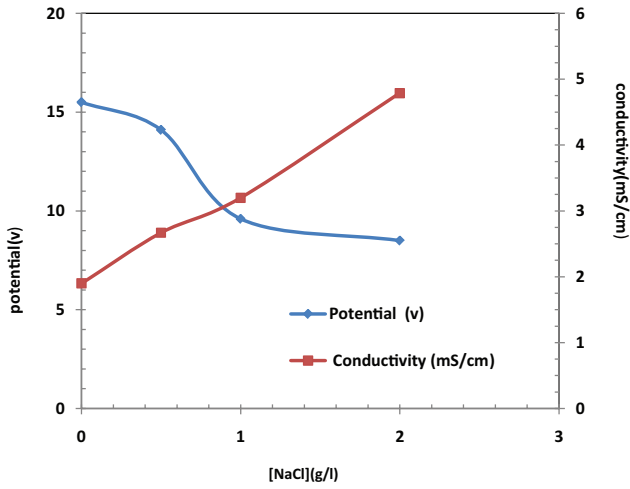


Fig. 7. Effect of concentration electrolyte supporting NaCl on the variation of potential and conductivity of the solution (Fe/Al ; $I = 15.56 \text{ mA/cm}^2$, $d = 2 \text{ cm}$, $T = 298 \text{ K}$ and $pH = 7.89$).

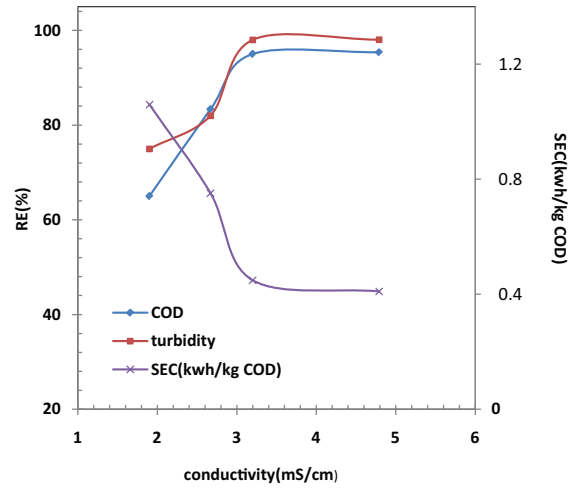


Fig. 9. Effect of the conductivity on the specific energy consumption and COD and turbidity removal efficiency ($I = 15.56 \text{ mA/cm}^2$, Fe/Al , $d = 2 \text{ cm}$, $T = 298 \text{ K}$, $pH = 7.89$, $t = 20 \text{ min}$).

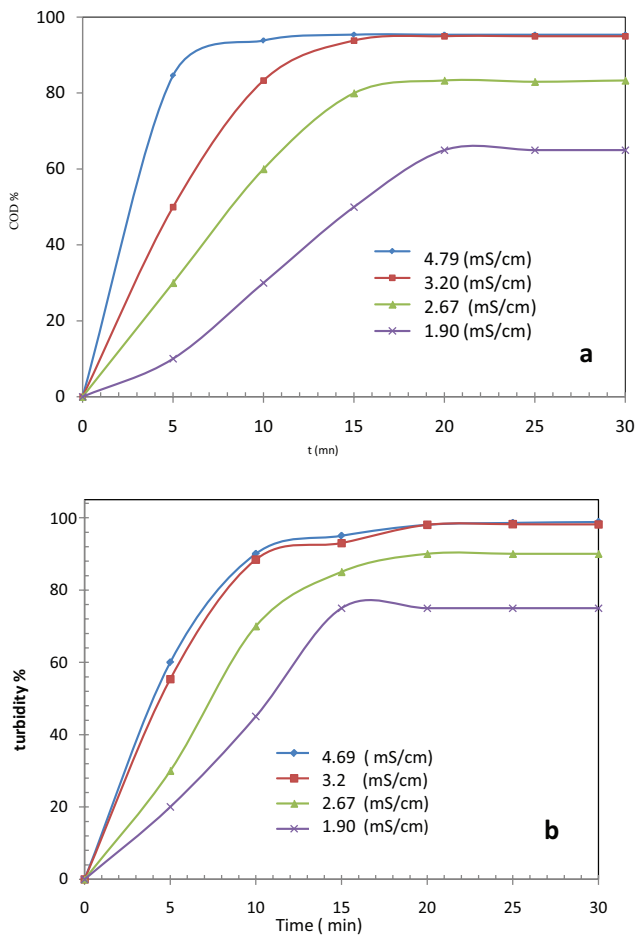


Fig. 8. Effect of the conductivity of the solution: (a) on the removal efficiency of COD and (b) on the removal efficiency of turbidity ($I = 15.56 \text{ MA/cm}^2$, Fe/Al , $d = 2 \text{ cm}$, $T = 298 \text{ K}$ and $pH = 7.89$).

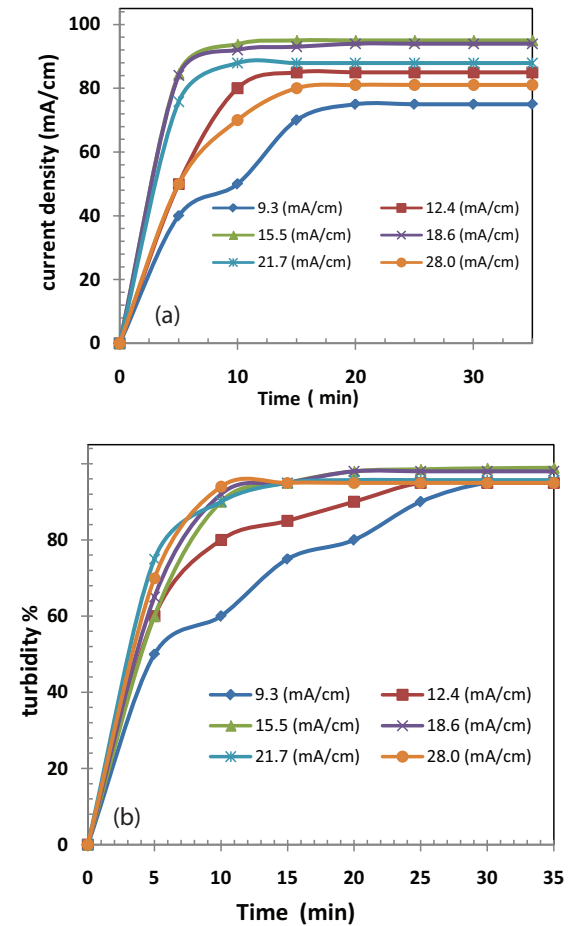


Fig. 10. Effect of current density on COD (a) and turbidity (b) removal in the pharmaceutical wastewater ($d = 2 \text{ cm}$, Fe/Al , $T = 298 \text{ K}$, $pH = 7.89$ and conductivity = 3.20 ms/cm).

for all current densities the removal efficiency of COD and turbidity increased with the increase of the electrolysis time. The COD and turbidity removal efficiency also increased when the current density was increased.

At high current densities, the anodic dissolution of iron increases, which leads to an increase in the amount of iron metal hydroxides and results in a better COD and turbidity removal. As the current density increased, the required times for the EC process decreased; as seen with the 15.56 mA/cm² of current density, electrolysis time was 15 min for a high removal efficiency of COD, and turbidity was 95.38% and 95%, respectively.

When there is enough current in the solution, the metal ions generated by the dissolution of the sacrificial electrode are hydrolyzed and form a series of metal hydroxides. The effects of current density on the removal efficiency of COD and turbidity after 20 min of electrolysis time are shown in Fig. 12. The results show that an increase in the current density from 9.3 to 15.56 mA/cm² led to increasing the removal efficiency of COD from 75% to 95.38% and turbidity from 80% to 98.05%, but when the current density increases

to 28 mA/cm², the removal efficiency of turbidity decreases to 95%, and removal efficiency of COD decreases to 81%. With the increase of current density, the concentration of ions Fe³⁺ increases in solution as presented in Fig. 11. That can explain that when the current density increases the formation of bubbles of hydrogen and oxygen increase in solution and the ions Fe³⁺ was floated to the surface of the water, he had not the time to form the coagulant in the solution or the formation of others complexes between iron and the chlorides.

The effect of current density on electrode consumption was presented in Fig. 12. The result shows that the mass of anode consumption experimental is higher than the mass of anode consumption theoretical and the difference between them increase with the increase of the current density. That can be explained by the attack of the electrodes corrosion pitting by chlorides ions, which were present in solution.

3.5. Effect of initial concentration of COD

The turbidity of the ointment pharmaceutical wastewater increases with the increase of the COD of this wastewater as presented in Table 2.

The effect of initial concentrations of COD on the removal efficiency of COD and turbidity after 20 min of times of electrolysis is represented in Fig. 13. The results show that the removal efficiency of COD and turbidity decreased when the initial concentration of COD had increased that can cause by less than the required number of coagulant, Fe(OH)_n(s), species generated from the anode electrode for applied current density. The SEC increases when the initial concentration of COD increases that can be to the increase of the potential and the increase of the resistivity of the solution.

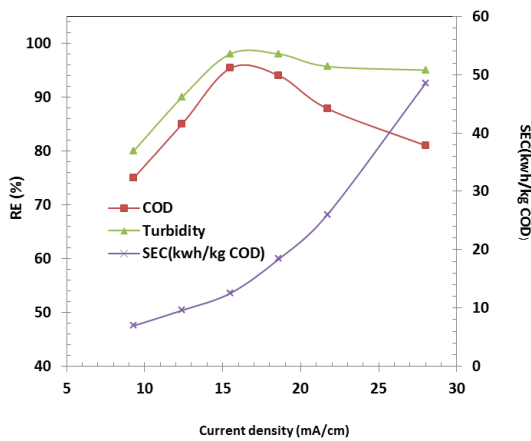


Fig. 11. Effect of current density on COD and turbidity removal efficiency and energy consumption ($d = 2$ cm, Fe/Al, $T = 298$ K, pH = 7.89, conductivity = 3.20 mS/cm and $t = 20$ min).

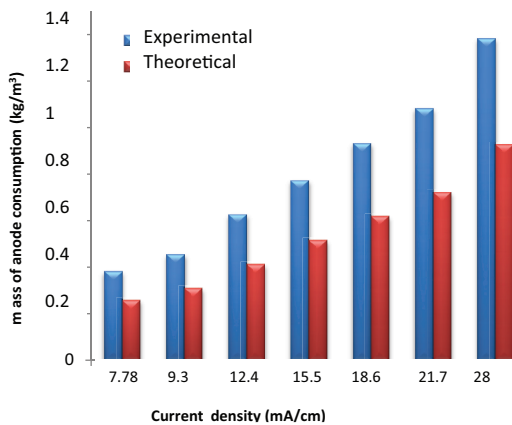


Fig. 12. Effect of current density on the mass of anode consumption ($d = 2$ cm, Fe/Al, $T = 298$ K, pH = 7.89 and conductivity = 3.20 mS/cm).

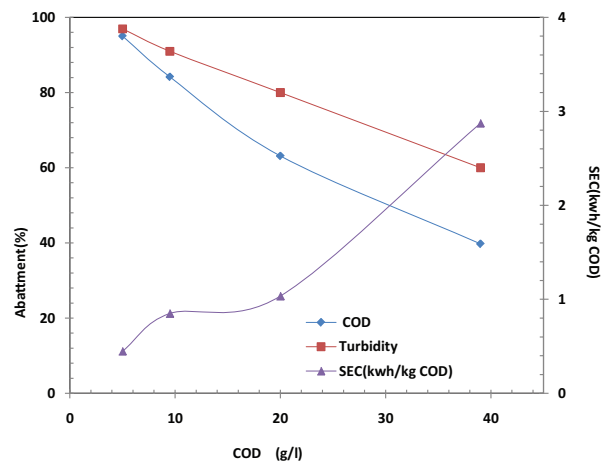


Fig. 13. Effect of initial concentration on COD on removal efficiency of COD and turbidity and on energy consumption ($d = 2$ cm, Fe/Al, $T = 298$ K, pH = 7.89, conductivity = 3.20 mS/cm and $t = 20$ min).

Table 2
Variation of the pharmaceutical wastewater turbidity over COD

COD (g/L)	5	9.5	20	39
Turbidity (NTU)	3,280	4,624	5,500	6,800

4. Conclusion

In this research, the performance of an EC in the batch was investigated with experimental operating parameters such as the solution temperature, type of electrode pairs, current density, conductivity and initial COD concentration to determine the best removal efficiency of COD and turbidity from the ointment pharmaceutical wastewater. The results showed that an Fe/Al electrode pair was the most efficient choice for the best removal efficiency of COD 95% and turbidity 98.05% after 20 min of electrolysis times with 0.48 kWh/kg COD SEC. The optimum temperature, current density, conductivity and COD were found to be 298 K, 15.56 mA/cm², and 5,000 mg O₂/L, respectively.

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