

Treatment of Tannery Wastewater by the Application of Electrocoagulation Process Using Iron and Aluminum Electrodes

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Abstract

Until relatively recently, little has been done of effective technique “zero effluent” to conserve energy and water. Tannery wastewater is known as complex characteristics. In this study batch electrocoagulation experiments were carried out to assess the removal of color and chemical oxygen demand (COD) from tannery wastewater using two types of electrode materials: aluminum and iron. The effects of current density, electrolysis time and initial pH were investigated for tannery wastewater. Therefore, the operating costs for each electrode have been calculated. Based on results, it can be concluded that iron is tremendous to aluminum as electrode material, from COD removal and energy consumption views. All the conclusions of the study revealed that treatment of tannery by EC can be applied as a step of a hybrid treatment.

Keywords

Tannery Wastewater, Chemical Oxygen Demand, Electrocoagulation, Operating Costs

1. Introduction

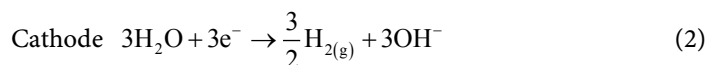
High consumption of water is one of the most important environmental concerns in tannery industry. Nowadays, zero effluent technologies are being developed to overcome this problem. The tannery industry is among the most polluting industries in terms of the volume and the complexity of treatment of its effluents discharge [1]. Tanneries produce wastewater in the range of 30 - 35 L/kg skin/hide processed with variable pH and high concentrations of suspended solids, BOD, COD, tannins including chromium as well as low biodegradability

[2] [3] [4] [5]. It should absolutely be searched a cleaner technology and economically as well as environmentally sustainable for treatment of complex characteristics of tannery wastewater.

However, wastewater treatment methods are broadly classified into physical, chemical and biological processes [6]. However, these methods don't always reach acceptable performance due to the complex of tannery wastewater which affects the bacterial activity. Therefore, traditional physical-chemical is comparatively expensive as well as may lead to secondary pollution due to requiring additional chemicals [7].

Nowadays, the selection of wastewater treatment process is based on several issues like efficiency, cost and environmental fitness as well as the wastewater characteristics [8]. Electrochemical treatment processes have extended such a state that they are not only comparable with other processes in terms of cost, but they are also more efficient and more effective [9]. One of these processes is electrocoagulation (EC) which has attractive characteristics as simple, reliable, and cost-effective operation for the treatment of water/wastewater [10]. In EC process, the coagulating ions are produced "*in situ*" and it involves six main processes: 1) electrophoresis and aggregation due to charge neutralization; 2) precipitation due to collective cation or hydroxyl ion with pollutant; 3) bridge coagulation resulting by interaction between metallic cation with OH⁻ to form a hydroxide, which has high adsorption properties, therefore bonding to the pollutant; 4) sweep coagulation when the hydroxides form larger lattice-like structures and sweep through the water; 5) oxidation of pollutants to less toxic species; 6) electro-flotation or sedimentation and adhesion to bubbles lead to removing the pollutants [11] [12].

In general, aluminum and iron are selected as a type of electrode material due to their characteristics such as low cost, ready availability, and fitness and effectiveness [13]. The driven force of EC is electron "green technology", so an electrical current is passed through a metal electrode; the anode material undergoes oxidation, while the cathode will be subjected to reduction or reductive deposition of elemental metals [14]. In the case of aluminum, main reactions can be given as [15]:

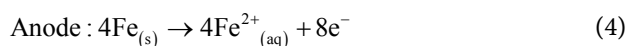


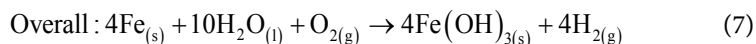
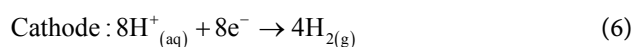
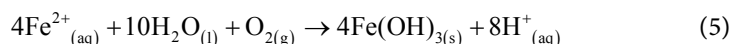
Al³⁺ and OH⁻ react with each other to form Al(OH)_{3(s)} according to complex precipitation kinetics.



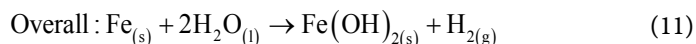
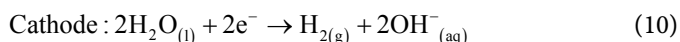
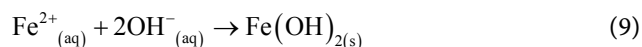
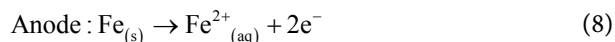
In the case of iron, two mechanisms for the reduction of metal hydroxide have been proposed [12].

Mechanism I





Mechanism II



EC has acceptable been operated decades to treat water/wastewater of oily wastewater [16], vegetable oil refinery [17], textile [18], toxic metal [19], arsenic [20], chemical oxygen demand (COD) [21], tannery [1] [5] [12] [22] [23], paper industry [24] [25], rose processing [26], domestic [27], etc.

In this work, the efficiency of electrocoagulation in removing color and COD from tannery wastewater was investigated using aluminum and iron electrodes. The effect of the initial pH and operational variables, current density, electrode material, metal consumption and treatment time, on the removal efficiency is explored and discussed to determine the optimum operational conditions, as well as the process economies, especially operational costs, are energy and electrode consumptions.

Novelty of this work brings the inclusive treatment facility to this kind of recalcitrant wastewater. The experimental conditions and cost analysis found in this study could help and be guidance on simple robust and economical treatment process for tannery wastewater.

2. Experimental

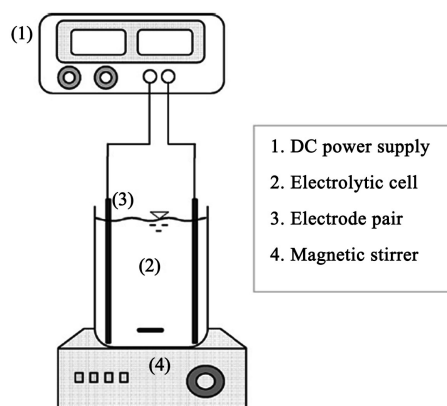
Tannery wastewater was collected from different outflow wastewaters of the Organized Tannery Industrial Region (OTIR) which is located in the Tuzla quarter of Istanbul, Turkey. The composition of the effluent wastewater is presented in **Table 1**. The tannery wastewater was first filtered using a screen filter to eliminate large suspended solids before it was used for the experiments. The experimental setup is shown in **Figure 1**.

The electro-coagulator reactor was fabricated of Plexiglas with the dimensions $5 \times 10 \times 20$ cm at constant stirring speed (200 rpm). There are four and six monopolar electrodes, two anodes, and two cathodes of the same dimensions. The dimensions of electrodes in 2.0 mm thickness were $5.0 \text{ cm} \times 15.0 \text{ cm}$. The total effective electrode area is calculated to be $(9 \text{ cm} \times 5.4 \text{ cm}) 48.0 \text{ cm}^2$. Both aluminum (99.53%) or iron (99.50%) cathodes and anodes were made from plates and the spacing between electrodes was 20 mm. The electrodes were connected in monopolar parallel mode to a DC power supply ((GW Instek, GPS 3030 DD, 0 - 30.0V, 0.0 - 3.0 A)).

Table 1. Characteristics of raw tannery wastewater used in this study.

Parameter	Value
pH	4.0 at 6.5°C
Conductivity (mS/cm)	11.71
COD mg·L ⁻¹	3500 - 3800
Color	824

Note: Color ADMI (10) Pt-Co.

**Figure 1.** Experimental set up.

All the runs were achieved at constant temperature of 25°C. In each run, 400 cm³ of the tannery wastewater solutions was placed into the electrolytic cell. The current density was adjusted to a desired value and the coagulation was started. At the end of electrocoagulation, the solution was filtered and then was analyzed. Before each run, electrodes were washed with acetone to remove surface grease, and the impurities on the aluminum or iron electrode surfaces were removed by dipping for 5 min in a solution freshly prepared by mixing 100 cm³ HCl solution (35%) and 200 cm³ of Hexamethylenetetramine aqueous solution (2.80%).

The wastewater analyses were carried out in accordance with the Standard Methods for Examination of Water and Wastewater [28]. “pH, conductivity, COD, and color were determined with (A Jenway 3040 brand, HACH HQ40d, closed reflux titrimetric method 5220C, and HACH LANGE GmbH DR 5000 (spectrophotometer), respectively)”. Merck analytical quality chemicals were used in the preparation of reagents.

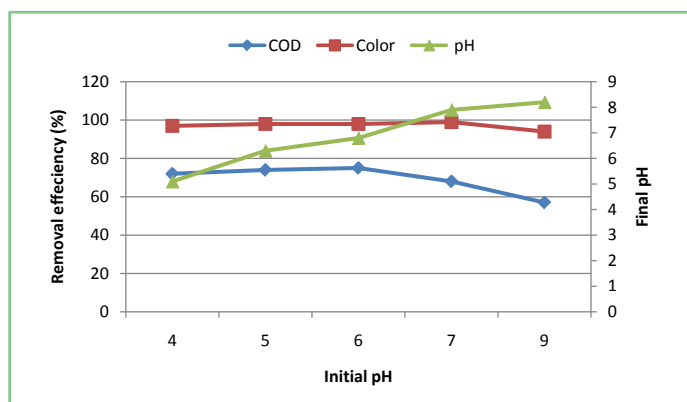
3. Results and Discussion

3.1. Effect of Initial pH

It is well known that pH is considered a vital parameter in any chemical or electrochemical separation process. Four different initial pH values were studied: original (natural) pH of 4.10, 5.0, 6.0, 7.0 and 9 to examine their effects on COD and color removal efficiency. H₂SO₄ and NaOH solutions were used to adjust the pH to desired initial value. **Figure 2(a)** & **Figure 2(b)** shows the removal efficiencies of COD and color as a function of the initial pH. As the characteristic of

EC process, the pH of the medium tended to increase during the process due to the type of electrode material and initial pH. For aluminum, the final pH is higher for initial pH < 7, and above this point the final pH is lower as it can be seen in **Figure 2(a)**. At alkaline medium suggest that electrocoagulation exhibits pH buffering capacity causing a pH decrease. On the other hand, for iron, the final pH is continuously higher than initial pH as it can be noticed in **Figure 2(b)**. At low pH, CO₂ is over saturated in wastewater and can be released during H₂ evolution, causing a pH increase.

Well, the effect of initial pH on the COD and color removal efficiencies is presented in **Figure 2(a)** & **Figure 2(b)**. For aluminum electrode, as seen in **Figure 2(a)**, for pH of 6 and pH of 6.8 for the initial and final values, respectively, the COD and color removal were 75% and 99% respectively. Color and COD removals drop dramatically at pH > 6. In the case of iron electrode, it can be seen in **Figure 2(b)** that COD and color removal is 81% and 98% as a maximum in accordance with a pH of 7 and pH of 7.8 for the initial and final values, respectively. And also, it can be seen that at pH of 6 and pH of 7.5 for the initial and final values respectively, iron electrode has achieved COD removal of 75% as shown in **Figure 2(b)**.



(a)

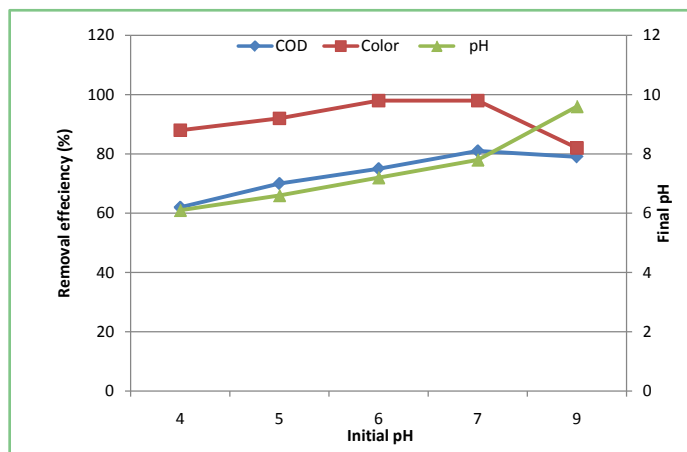


Figure 2. (a) Effect of initial pH on COD and color removal by aluminum electrodes; (b) Effect of initial pH on COD and color removal by iron electrodes.

For both type of electrode materials, it is clear that COD and color removals indicate the same trend. The highest removal efficiencies have been attained with aluminum and iron at initial pH of 6 and 7 respectively. Thus, it can be pointed out that in acidic medium, higher removal efficiencies are obtained with aluminum, while in neutral and weakly alkaline medium iron is more efficient. And it is clear from this figure that initial pH has no significant effect on color removal.

Figure 3 depicts the specific energy demand in relation for aluminum and iron electrodes during the electrocoagulation, calculated in kWh consumed per kg COD removed. It can be concluded that iron electrodes are more energetically efficient than aluminum. However, at $\text{pH} > 7$ the energy consumption is almost constant at 2.1 kWh/kg COD, for the iron case. And for aluminum at pH of 6 the energy consumption is at 2.35 kWh/kg COD.

Figure 4 shows the electrode consumption per kg of COD removed, in relation to initial pH in electrocoagulation. Electrode consumption is an important economic parameter of electrocoagulation process especially with iron electrode. However, it can be concluded that the electrode consumption in the case of aluminum is at 0.096 kg Al/kg COD at pH of 6. But in the case of iron it is at 0.31 kg/kg COD at pH of 7. This result indicates that iron is more efficient than aluminum, for COD removal. This may result from the differences in the mechanisms of COD removal for the iron and aluminum electrodes.

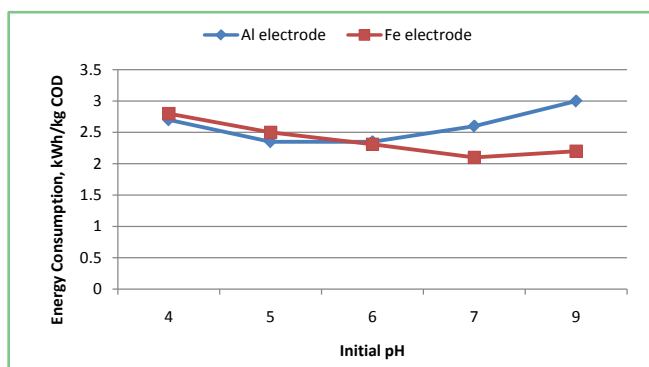


Figure 3. Effect of initial pH on energy consumption.

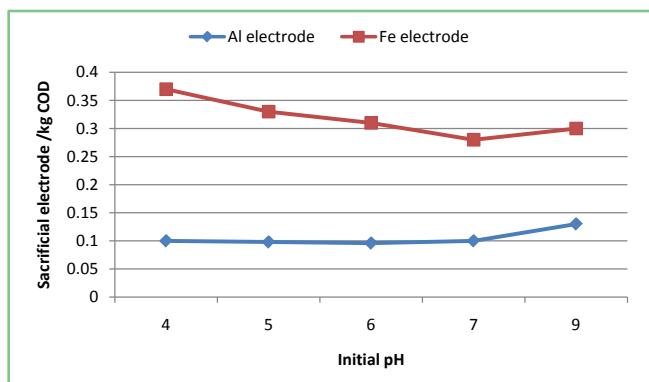
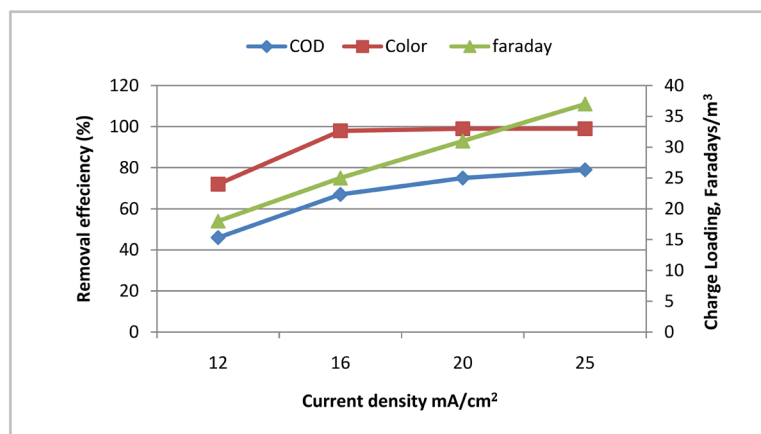


Figure 4. Effect of initial pH on electrode consumption.

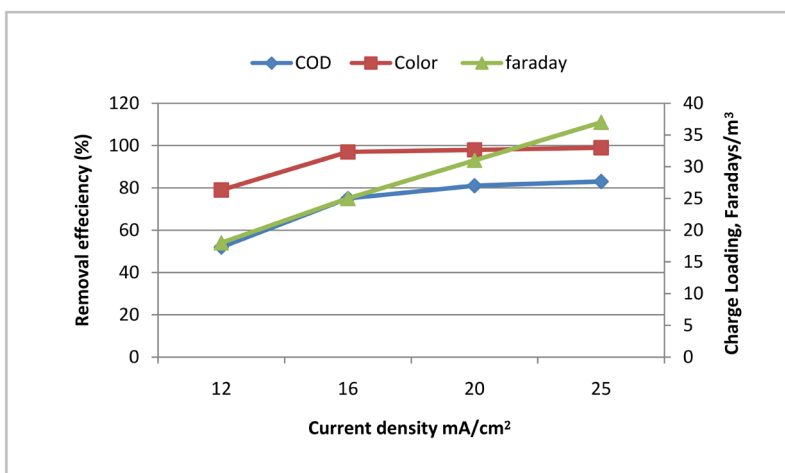
3.2. Effect of Current Density

The effect of current density on the removal of COD and color from TWW was investigated at 25°C using two different electrode materials, namely aluminum (Al) and iron (Fe). Each electrode was examined separately. Different current densities in the range of 12 - 25 mA/cm² were applied to the electrochemical reactor to inspect its effect. **Figure 5(a)** and **Figure 5(b)** depicts the effect of current density on COD and color removal efficiencies, for aluminum and iron electrode materials with operating time constant at 20 min as well as at pH of 6 and 7 respectively. However, variations of COD and color removal efficiency (%) and energy consumption (kWh/kg COD) for different current densities have shown in **Figure 6(a)** and **Figure 6(b)**. And also, electrode consumption per COD removed as kg/kg has included in **Figure 7**.

Anyway, as seen in **Figure 5(a)** and **Figure 5(b)**, for both types of electrode material (aluminum and iron) minimum 20 mA/cm² is essential for good efficiencies, with a charge loading approximately equal to 31 F/m³. On the other hand, in the case of aluminum, COD and color removal efficiency has been 75% and 99% respectively at the current density of 20 mA/cm² at 20 min with the energy consumption value of 2.35 kWh/kg COD removed as shown in **Figure 6(a)**. For iron, as seen in **Figure 6(b)** COD and color removals have maximum removal rates (81% and 98% for COD and color, respectively) at the current density of 20 mA/cm² with the energy value of 2.1 kWh/kg COD removed. Here, it can be pointed out aluminum electrode consume more energy than iron electrode. Finally, the electrode material consumption is given in **Figure 7**. When current density was increased from 12 to 25 mA/cm², electrode consumption per COD removed has weakly changed from 0.9 to 0.11 kg AL/kg COD, but for iron, it has increased from 0.26 to 0.32 kg Fe/kg COD. These efficiencies and consumption values show the superior performance of iron over aluminum as electrode material. Therefore 20 mA/cm² was selected as the operational current density to keep the energy consumption and electrode consumption as the removal efficiency is higher for the wastewater studied in this paper.

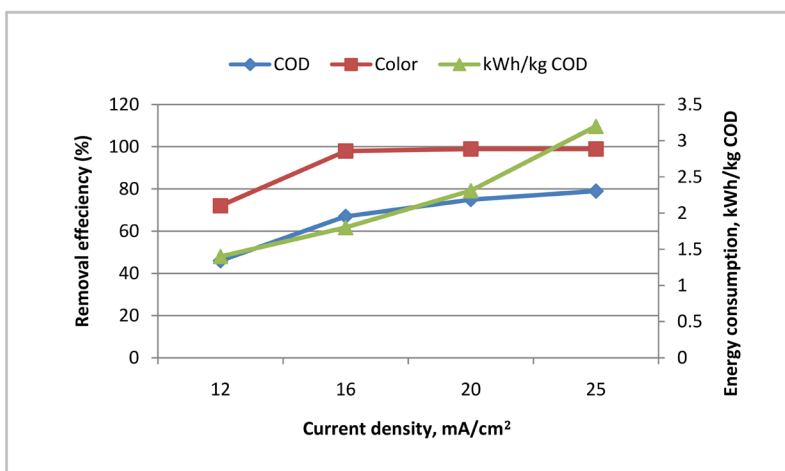


(a)

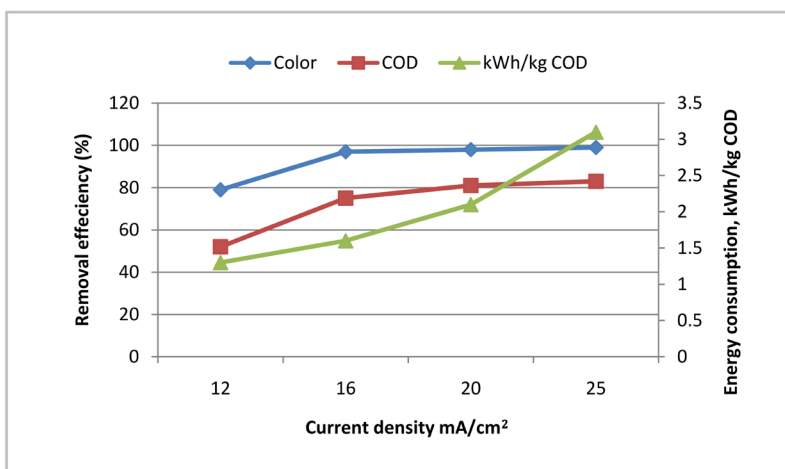


(b)

Figure 5. (a) Effect of current density on COD and color removal by aluminum electrodes; (b) Effect of current density on COD and turbidity removal by iron electrodes.



(a)



(b)

Figure 6. (a) Effect of current density on energy consumption by aluminum electrodes; (b) Effect of current density on energy consumption by iron electrodes.

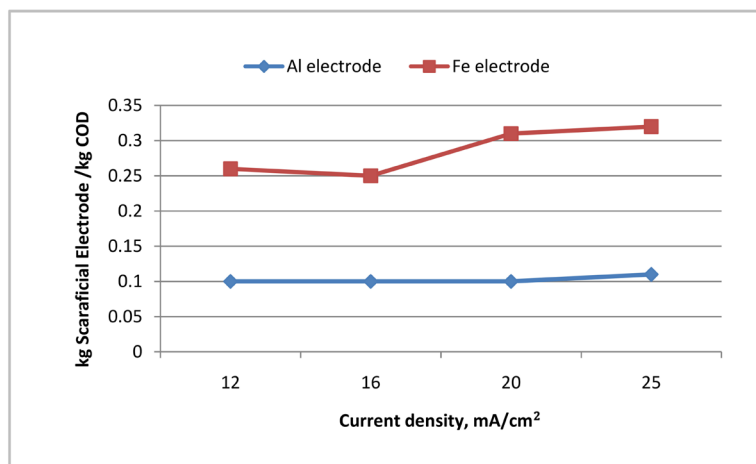


Figure 7. Effect of current density on electrode consumption.

3.3. Effect of Operating Time

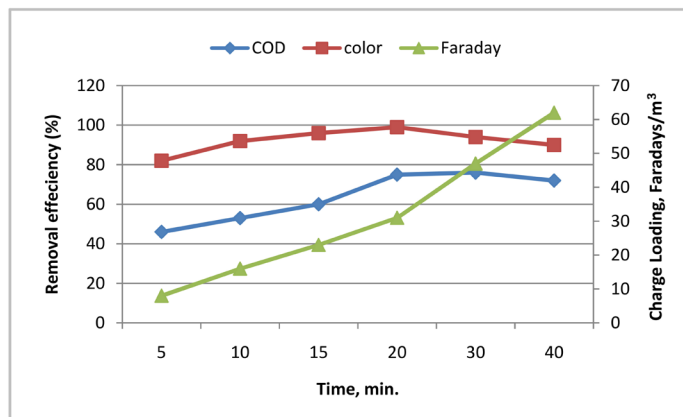
Operating time experiments were carried out at pH 6 for Al electrode, at pH 7 for Fe electrode at 20 mA/cm². As given in **Figure 8(a)** and **Figure 8(b)** both electrode materials (Al & Fe) require 20 min. for good removal efficiencies. With regarding energy and electrode material consumptions are given comparatively for two materials, in **Figure 9** and **Figure 10**, respectively. As seen in **Figure 9** longer operating times may not be used with both electrode materials, to obtain higher wastewater treatment efficiencies using less electric energy. Finally, it can be seen in **Figure 10** that electrode consumption values are higher for Fe electrode than aluminum electrode. Ultimately, based on our results in this paper, it is concluded that the effect of current density and operating time on performance criteria are very similar, this leads that two variables may be joined as a single variable, as well as Faradays may greatly use for the process optimization studies.

3.4. Temperature and Conductivity

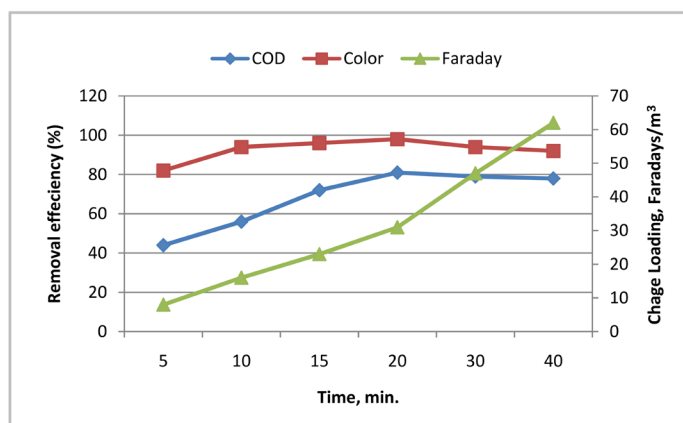
Temperature is considered always significant parameter in any chemical or electrochemical separation process. During the experiments, temperature was monitored in the reactor. It was clear that increasing in temperature as a result of reactions. This increase in temperature refers to electrolytic reactions depending on contact time, electrode type and applied electrical power is given in **Figure 11**. From **Figure 11**, when current density was 20 mA/cm², temperature changed from 23.7°C to 25.1°C with Al-electrode and from 23.7°C to 26.3°C with Fe-electrode. The temperature tends to increase as a result of electrolytic reactions.

On the other hand, the Change of conductivity depending on electrode type and applied current densities with respect to time is monitored. As seen in **Figure 12**. It is concluded that conductivity decreases as a result of electrochemical treatment. When current density was 25 mA/cm², conductivity with Al-electrode

was decreased from 11.72 to 9.50 mS, and was from 11.71 to 9.80 mS with Fe-electrode. When current density was 20 mA/m², conductivity was decreased from 11.80 to 10.50 mS with Al-electrode, and was from 11.80 to 10.75 mS with Fe electrode as shown in **Figure 12**.



(a)



(b)

Figure 8. (a) Effect of electrocoagulation time on COD and turbidity removal by aluminum electrodes; (b) Effect of electrocoagulation time on COD and turbidity removal by iron electrodes.

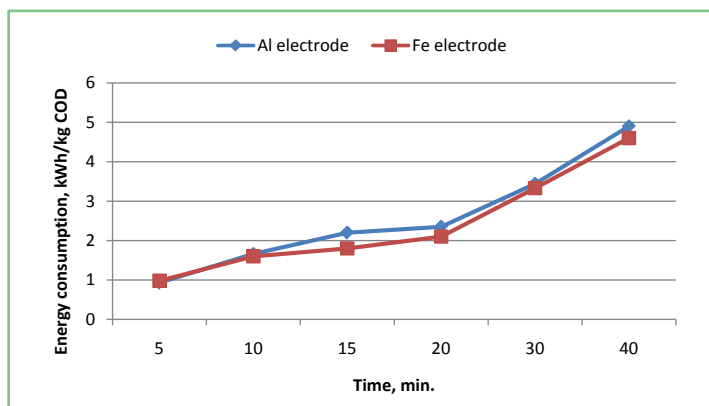


Figure 9. Effect of electrocoagulation time on energy consumption.

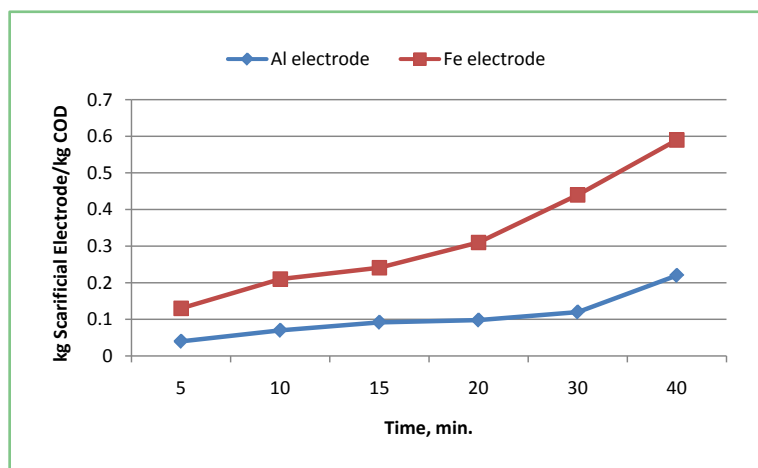


Figure 10. Effect of electrocoagulation time on electrode consumption.

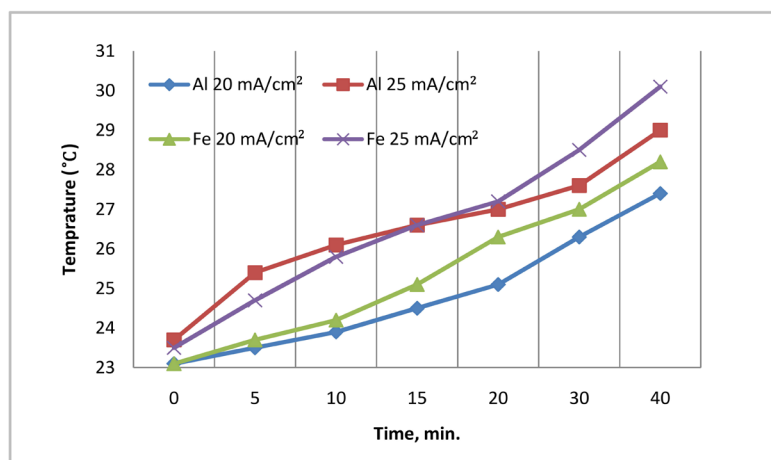


Figure 11. Change of temperature depending on electrode type and applied current densities with respect to time.

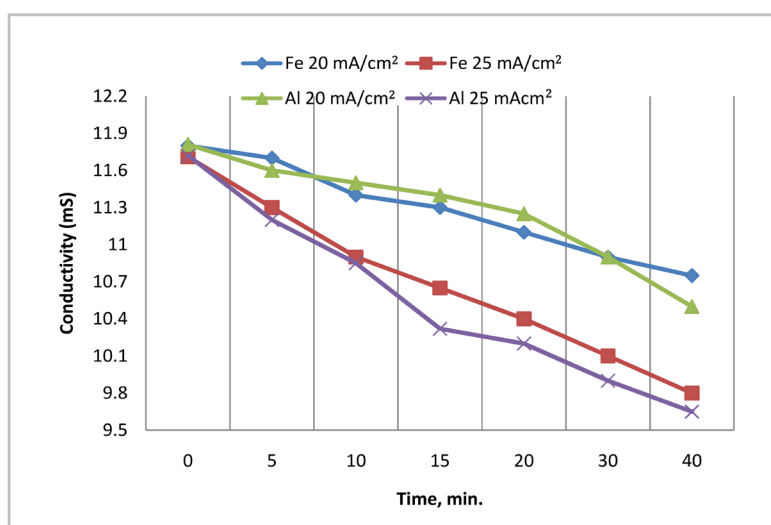


Figure 12. Change of conductivity depending on electrode type and applied current densities with respect to time.

3.5. Operating Costs

The EC process required an operating cost which included material (electrodes and electrical energy). The operating cost could be calculated as displays:

$$\text{Operating cost} = aC_{\text{energy}} + bC_{\text{electrode}} \quad (12)$$

where (a) the energy cost: 0.07 \$/kW (Turkish price); (b) the aluminum cost: 1.5 \$/kg or the iron cost: 0.07 \$/kg (Turkish price). C_{energy} (kW h/m³) and $C_{\text{electrode}}$ (kg Al/m³ or kg Fe/m³).

Cost of electrical energy (kW h/m³) is calculated as [29]:

$$C_{\text{energy}} \left(\text{kW} \cdot \text{h/m}^3 \right) = \frac{UIt}{V} \quad (13)$$

where U is the voltage cell (V), I is the current (A), t is the time of electrolysis (h) and V is the volume (m³) of wastewater treated.

Cost of electrode (kg Al or Fe/m³) is calculated by the following equation according to Faraday's Law [29]:

$$C_{\text{electrode}} \left(\text{kg/m}^3 \right) = \frac{ItM}{nFV} \quad (14)$$

where I is current (A), t is time of electrolysis (s), M is molecular mass of aluminum or Fe (26.98 or 56 g/mol) respectively, z is number of electrons transferred ($z = 3$ for Al); or ($z = 2$ for Fe), F is Faraday's constant (96,500 C/mol) and V is volume (m³).

In this work, essential removal efficiency was obtained in first 20 min reaction time at current density of 20 mA/cm² (**Figure 8(a)** & **Figure 8(b)**); therefore, consumption calculations were selected as 20 min and as 20 mA/cm².

The electrode and energy consumptions in relation to current density are given in **Figure 13**. Electrode and energy consumption was calculated using Equations (13) and (14). Thus, **Figure 14** shows the relationship between operation cost and current density.

For aluminum electrode, the energy and electrode consumptions were calculated as 6.6 kWh/m³ and 0.28 kg/m³, respectively. Meanwhile, the costs for electrode and energy used were attained as \$0.46 and \$0.42 per m³ treated wastewater, respectively. For iron electrode, the energy and electrode consumptions were calculated as 6.6 kWh/m³ and 0.87 kg/m³, respectively. Ultimately, the costs for electrode and energy used were attained as \$0.46 and \$0.52 per m³ treated wastewater, respectively. As expected electrode and energy consumptions increase with increasing of current density.

Ultimately, optimum conditions from the experimental results are summarized in **Table 2**. It can be pointed out that there is no difference between Al or Fe electrodes type material regards to view operating cost point in this paper.

4. Conclusions

Electrocoagulation was evaluated as a potential technique for the reduction of color and COD concentration in tannery wastewater, which was treated in a

batch electrochemical reactor using two types of electrode materials; aluminum and iron. The experimental results showed that electrocoagulation with iron electrode can achieve percentage removal of up to 98% and 81% for color and COD, respectively. However, electrocoagulation with aluminum electrode can realize percentage removal up to 98% and 75% for color and COD respectively.

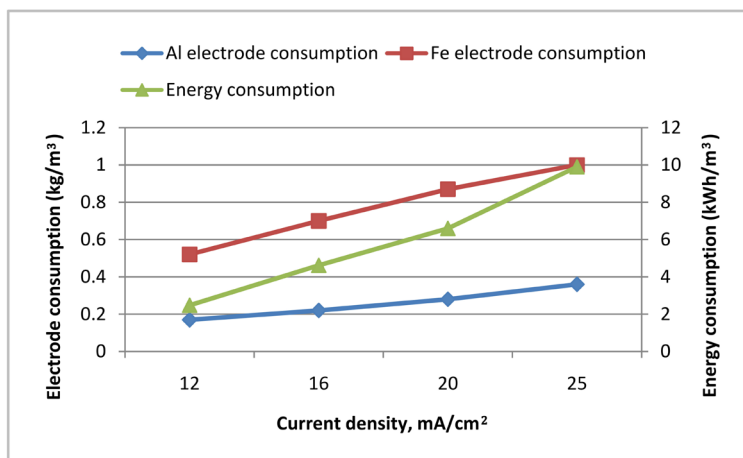


Figure 13. Effect of current density on the energy and electrode consumptions.

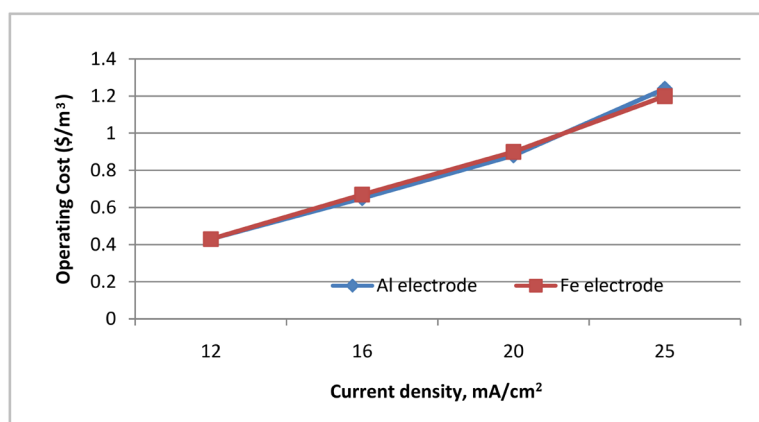


Figure 14. Effect of EC current density on operating cost.

Table 2. Optimum operating conditions and operating cost for types of electrode material for EC.

	Al-Electrode	Fe-Electrode
Current density (mA/cm²)	20	20
Operating time (min)	20	20
Initial pH	6	7
Final pH	6.8	7.8
COD removal (%)	75	81
Color removal (%)	98	98
Operating cost (\$/m³)	0.88	0.90

The performance of the electrochemical reactor was found to be highly influenced by the pH of the wastewater and the current density. According to the results at pH of 6, color and COD removal efficiencies of aluminum than those of iron, while at pH of 7 iron was preferable. On the other hand, for the same color or COD removal efficiencies aluminum and iron require the same current density of 30 mA/cm² for an operating 20 min. Therefore, operating time and current density display similar effects on the process performances, on electrical energy and electrode consumption values. Based on results, it can be established that charge loading of two process variables, may be carried out more efficient in process design and optimization missions.

Finally, electrocoagulation with iron confirms that the energy consumption kWh per kg COD removed is lower, on the contrary the electrode consumption per kg COD removed is lower generally with aluminum. Energy and electrode consumption is vital for operating costs which will powerfully influence the decision about the type of sacrificial electrode material for given wastewater COD and color removal levels forced by environmental restrictions about process effluents.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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